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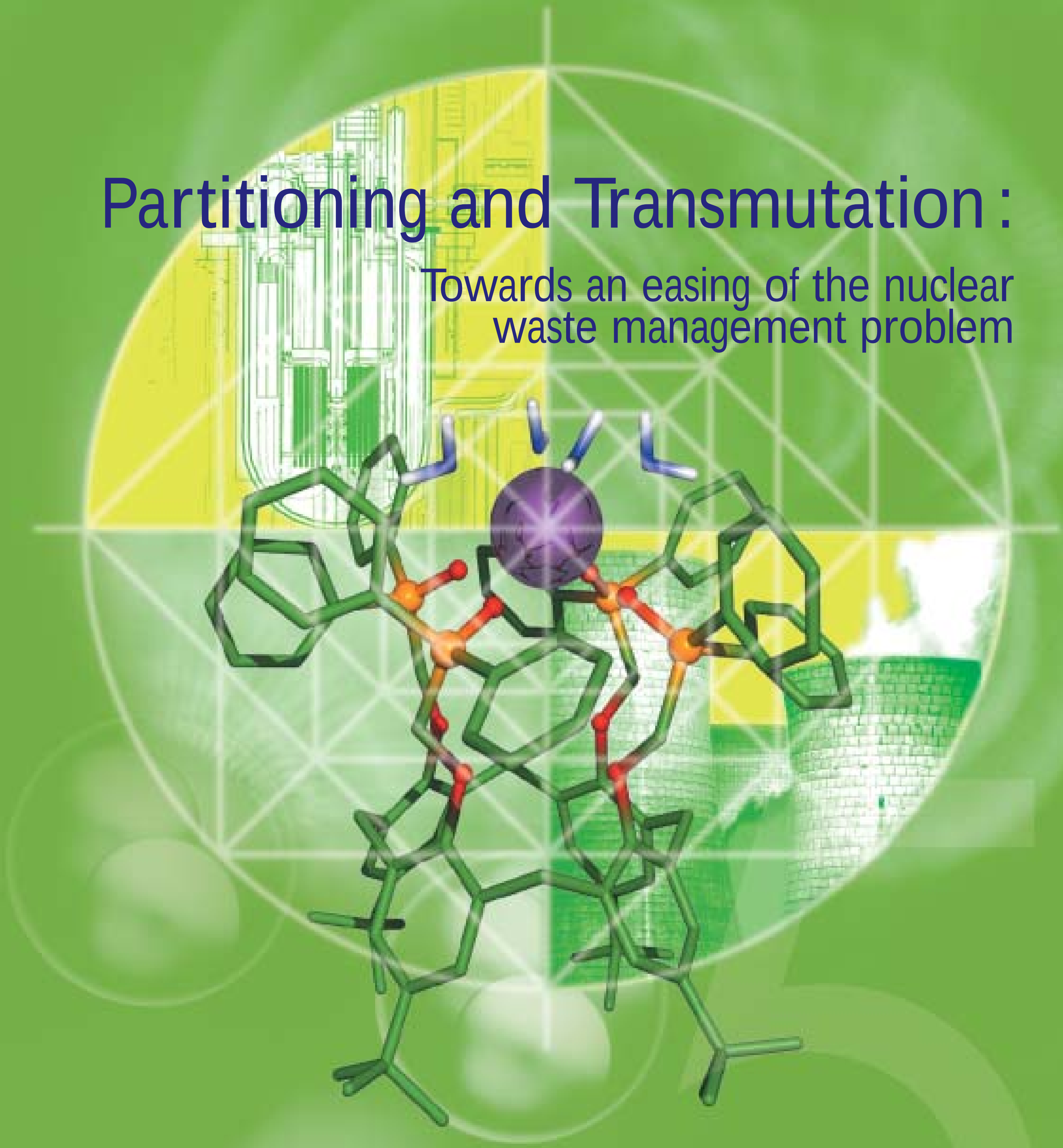
Partitioning and Transmutation :

Towards an easing of the nuclear waste management problem



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Calixarenes molecule, Courtesy CEA, Cadarache (F) and ADS/EA Demo design, Ansaldo Nucleare, Genova (I).

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1. Overview

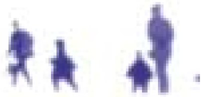
Nuclear power is a secure large-scale baseload supplier of electricity and provides about 35% of the European Union's electrical power. As nuclear energy processes do not emit greenhouse gases, its potential is increasingly underlined by mounting demands for cleaner air and growing disquiet about carbon pollution. Nuclear energy enables the EU to reduce its greenhouse gas emissions by 7%—the equivalent of the CO_2 emissions produced by some 75 million cars.

Nuclear fission energy gives rise to waste as a by-product. This waste is mainly contained in the spent fuel, which is highly radioactive for a long time. If inadequately managed, the nuclear waste could constitute a potential risk to be borne by future generations.

At present, the nuclear power industry safely contains all fuel waste near reactor sites in separate dedicated water-pool storage facilities. With the continued supply of nuclear energy, this waste is accumulating and a more permanent waste-disposal solution has to be found.

The main concern in the disposal of radioactive waste is related to long-lived radionuclides—some of them will remain hazardous for tens of thousands of years. A sizeable effort to resolve this problem is already well under way—the main approach being the disposal of nuclear waste in deep geological formations.

By partitioning (chemical separation) and transmutation (radionuclide conversion), it might be possible to reduce the long-lived component of the radioactive waste, thus easing the waste management problem. The objective of the EU research and development effort on partitioning and transmutation is to study its practicability on an industrial scale.



2. Nuclear waste management

The safe long-term management of radioactive waste concerns us all—scientists and engineers, politicians and the general public. Techniques for the safe disposal of the least noxious forms of waste are in hand; some countries, such as Finland, France, Spain, Sweden and the UK, are already implementing systems for safe disposal of low-level and intermediate-level waste. Until now, however, no disposal facility has been built for the most noxious waste—the high-level radioactive waste from a civilian nuclear power plant. This needs to be contained and isolated for a very long period of time—tens of thousands of years or more.

In most countries with nuclear power programmes, research is conducted to find suitable underground geological sites for high-level waste disposal. Good progress has been made in this endeavour. After a long study in the USA, one geological disposal facility has recently started operation—the Waste Isolation Pilot Plant (WIPP), a facility for long-lived transuranic waste of military origin. However, regulatory and public approval for geological disposal of long-lived and high-level waste (HLW) has been hard to come by in the EU. A law passed by the French government in December 1991 requires a fifteen-year programme of work on alternatives to geological disposal of HLW. Other countries, including Japan and the USA, have also been pursuing alternatives in parallel with geological disposal.

Courtesy of EIG EURDICE, Mol (B)

Underground geological disposal:

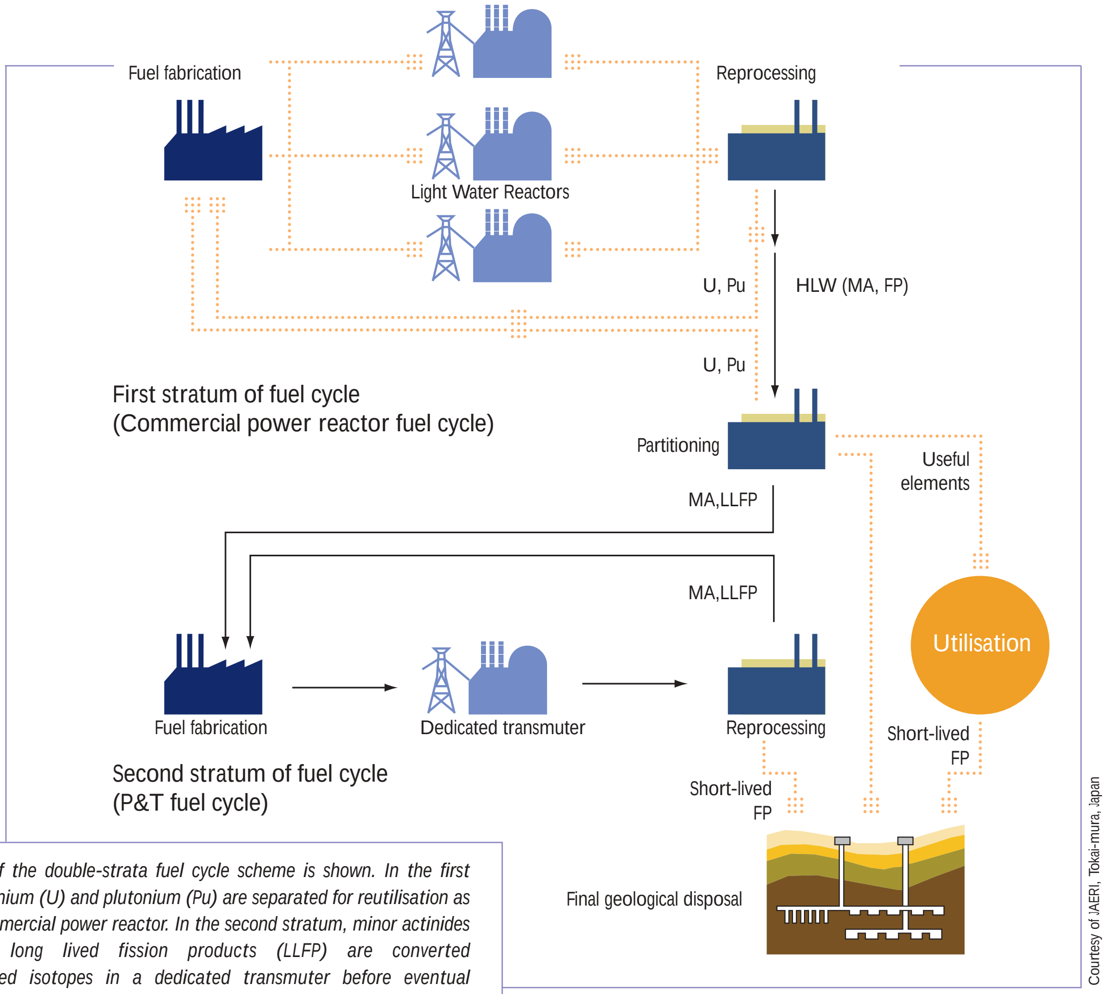
In principle, spent nuclear fuel and other high-level radioactive waste can be disposed of safely in deep geological formations, where, it is believed, it will require little or no continuing human involvement and the likelihood of future human intrusion, unless accidental, is minimal. Scientists and engineers have put forward technically sound proposals for such an underground geological repository.

Nevertheless, since the waste becomes harmless only after tens of thousands of years or more, studies have to be carried out on the radiological hazard in the case of accidental human intrusion and radiological dose release via groundwater pathways. One such study showed that if the human intrusion took place within 10,000 years or so, the radioactive doses caused by direct human exposure to waste would be much higher than those caused by exposure via groundwater pathways.

3. Partitioning and transmutation (P&T)

The small risk associated with the disposal of HLW in well evaluated geological sites can be further reduced by techniques known as partitioning (chemical separation) and transmutation (radionuclide conversion), which are carried out on the spent fuel before disposal.

The main aim of P&T is to reduce the long-term (>300 years) radio-toxic inventory of the waste, making its disposal safer and alleviating the uncertainties that go with the long time scales. This is achieved by separating and transmuting appropriate long-lived



An outline of the double-strata fuel cycle scheme is shown. In the first stratum, uranium (U) and plutonium (Pu) are separated for reutilisation as fuel in a commercial power reactor. In the second stratum, minor actinides (MA) and long lived fission products (LLFP) are converted to short lived isotopes in a dedicated transmuter before eventual disposal in a geological repository.

radioactive isotopes to form short-lived or even stable ones. P&T also aims to reduce the volume of waste requiring deep geological disposal and hence the associated space requirements and costs.

It is recognised that P&T is a long-term basic R&D effort for the benefit of future generations. It is likely to require several tens of years to come to fruition. In the short term, it can make only minor contributions to established and planned strategies for waste storage and disposal. Nevertheless, P&T technologies are already capable of reducing radio-toxicity while producing energy through the utilisation and burning of civil and military plutonium.

Partitioning (Chemical separation)

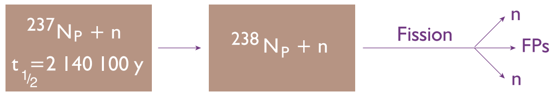
Current reprocessing technology is based on the aqueous PUREX process in which the spent fuel is dissolved in nitric acid. Using an organic extractant, uranium and plutonium are recovered and can be used for new reactor-fuel fabrication. However, the remaining liquid contains the minor actinides (americium, neptunium and curium) and fission products of which some are long-lived (such as isotopes of technetium, caesium and iodine). Together, these form the high-level waste to be disposed of.

To reduce the long-term radio-toxicity of the waste, a small number of the hazardous isotopes (such as americium, neptunium, caesium, technetium and iodine) need to be further separated out (partitioned) before the remaining waste is vitrified—that is embeded in glass blocks—for geological disposal.

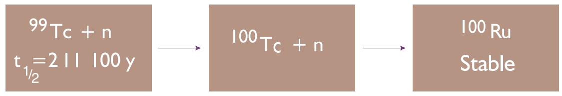
The above-mentioned separated isotopes could then be converted to lesser radio-toxic (or even stable) isotopes by nuclear transmutation in fast neutron reactors or accelerator-driven systems (ADS).

Transmutation

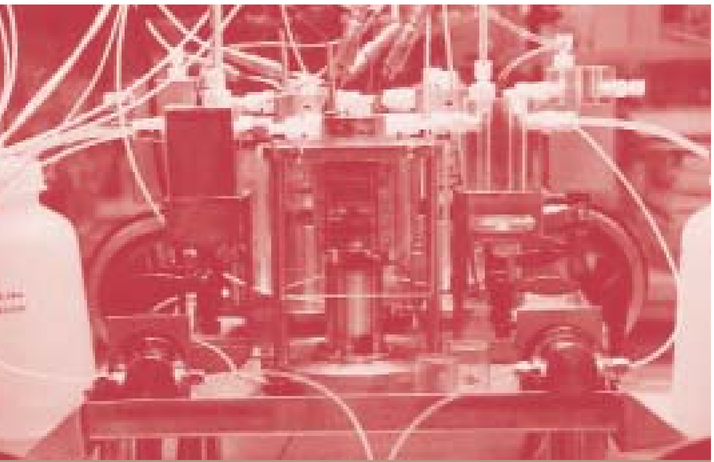
The troublesome components of high-level waste are a small number of isotopes that are radioactive for tens of thousands of years. If bombarded with neutrons after separation, these isotopes can be transmuted to others that are radioactive for only hundreds or even tens of years, reducing the duration required for waste disposal. In some cases, we end up directly with stable, that is non-radioactive, isotopes.



Transmutation by fission of minor actinide neptunium (Np) by external neutrons leading to fission products (FPs). The half-life ($t_{1/2}$) is given in years (y).



Transmutation of fission product technetium (Tc) by external neutrons leading to the stable isotope of Ruthenium (Ru).



Courtesy of CEA, Paris (F)

Advanced partitioning Processes

At present most of the advanced partitioning processes require further R&D before they can be operated on an industrial scale. Alternative 'dry' processes (in contrast to 'wet' or aqueous ones) are also being studied—processes in which actinides are separated out using pyro-chemical processes in molten salts. These dry processes could potentially be integrated into reactor operation. Such on-site separation would avoid the risks associated with the transport of spent fuel to reprocessing plants.

Energy amplifier and waste burner

In the 'Energy Amplifier' concept proposed by the Nobel laureate Carlo Rubbia, the objective is to produce energy and burn actinides and fission products within the thorium fuel cycle, which produces few transuranic elements. This concept is presently the subject of further studies and evaluation.

A fast sub-critical reactor coupled to a particle accelerator is a concept that would allow large quantities of minor actinide waste to be burned efficiently. Present-day reactor systems operate at criticality in which neutrons that are necessary for chain reaction are generated in the fission process itself. In a sub-critical reactor, on the other hand, additional neutrons are supplied by an external source—from spallation reactions, for instance, in which an energetic proton beam from a particle accelerator impinges on a heavy metal such as lead. Subject to more detailed studies, an ADS seems likely to be safer, as the neutron chain reaction can, in principle, be stopped when desired by switching off the additional supply of neutrons (the accelerator). This, however, still leaves the task of removing the decay heat.

4. Problems to be resolved in P&T

There are three key milestones that have to be passed before P&T can make a significant contribution to easing the waste management problem:

1. the development of a high-efficiency, industrial-scale partitioning technology for separating the minor actinides and long-lived fission products from the spent fuel;

2. the development of dedicated fuels and targets to host minor actinides and long-lived fission products for irradiation in transmutation reactors; and

3. the engineering design, licensing and regulatory approval, construction and testing of transmutation devices such as ADS including prototypes and demonstration devices.

These tasks will require several decades of dedicated skilled manpower and financial resources.

5. Progress in P&T achieved so far by Fourth Framework Programme (FP4) projects

In the specific programme on Nuclear Fission Safety in the Euratom Fourth Framework Programme (1994-1998), there were three areas of research activities on P&T: (1) Strategy studies, (2) Partitioning and (3) Transmutation. The strategy studies suggest that to enhance the efficiency of transmutation, accelerator-driven systems (ADS) should be thoroughly investigated and evaluated. The results from partitioning projects predict that a direct extraction of minor actinides from the high-level liquid waste will be possible in a single cycle in the near future. The studies on transmutation reveal that an ADS would be an efficient waste burner.

Strategy studies

The P&T strategy studies reveal that the fuel-cycle management policy of balancing production and consumption of waste will require several decades of work before it can be fully effective. This period will itself have to be preceded by the significant amount of time and effort that will be required to implement the partitioning and transmutation of waste on an industrial scale. It is proposed that to enhance the efficiency of transmutation, accelerator-driven sub-critical reactor systems should be thoroughly investigated and evaluated. Also, R&D on fuels for transmutation of minor actinides and long-lived fission products should be carried out.

Partitioning studies

In the NEWPART (NEW PARTitioning Techniques) project, a major breakthrough has been achieved in the partitioning of minor actinides by aqueous processes. With some additional effort, a one-cycle process for extracting minor actinides directly from liquid high-level waste could soon be demonstrated at the pilot-plant scale. Also, significant progress has been achieved in the extraction of caesium, strontium and actinides using new "Calixarene" derivatives.

Courtesy of NRG, Petten (NL)

Transmutation studies

(1) A preliminary assessment of ADS was carried out by the project entitled "Impact of Accelerator-Based Technologies on Nuclear Fission Safety" (IABAT). Neutronic studies indicate that the fast-spectrum and liquid-lead (or Pb-Bi eutectic) cooled, solid-fuel ADS allows efficient waste transmutation. Accurate neutron cross-section data at intermediate and high energy (20-200MeV) are required for the design of an ADS system. A project 'Lead for Accelerator Transmutation Devices' evaluated the present availability of the neutron cross-section data for lead as a material for an ADS in just this energy range.

(2) Transmutation experiments were carried out on americium-241 in the High Flux Reactor at Petten (NL) in a project designed to study the Experimental Feasibility of Targets for Transmutation (EFTTRA). It was found that after irradiation for 350 days, 96% of the americium was transmuted. However, the target pellets were swollen (by about 18%) due to helium release. This dictates modifications in the design of pellet fabrication.

(3) The experiment carried out at European Nuclear Research Centre (CERN) in the TARC project directly verified the concept of adiabatic resonance crossing for significantly enhancing the neutron-capture efficiency for long-lived fission products. In lead, neutrons lose their energy in small steps and slowly reach the capture resonance energy of an element for which the transmutation rate is maximum. Large amounts of technetium-99, iodine-129 or other long-lived fission products could be destroyed on the periphery of the core where the neutrons have the right energy. The process could also be used for the production of radio-isotopes for medical applications.

6. P&T activities in the Fifth Framework Programme (FP5)

The objective of the research being carried out on Partitioning and Transmutation under FP5 is to provide a basis for evaluating the practicability of reducing on an industrial scale the amount of long-lived radionuclides to be disposed of. Projects in the field of P&T have been subdivided into four groups: (1) Partitioning, (2) Transmutation – Fuel, (3) Transmutation – Basic Studies and (4) Transmutation – Technological Support.

The aim of the projects in the area of partitioning is to develop detailed flow-sheets for the separation of desired long-lived isotopes using wet or dry chemical processes.

The projects on transmutation are focused on the study of important aspects of a sub-critical ADS system. These include characterisation and fabrication of minor actinide fuels for transmutation. A project on preliminary design studies of an ADS is also planned. It is supported by basic studies such as sub-critical neutronic behaviour of an ADS and neutron cross-section data in a wide energy range for the desired isotopes for waste

Courtesy of CEA (Ph. Brault), Paris (F)

Partitioning

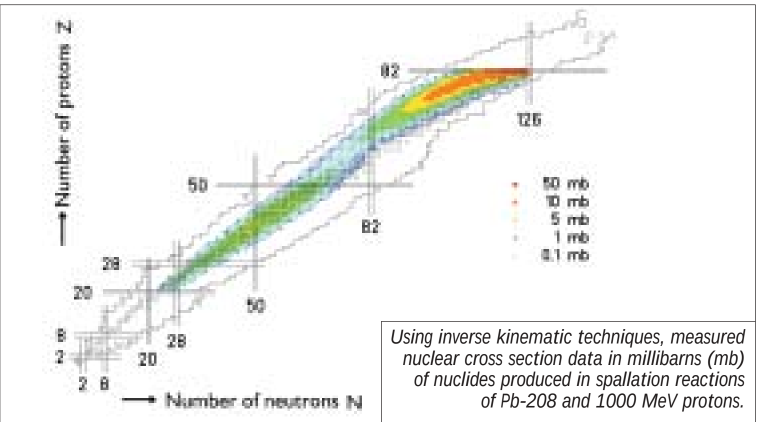
The first group, Partitioning, has three projects: PYROREP, PARTNEW and CALIXPART. The PYROREP project will assess flow sheets for pyrochemical processing of spent fuel where the salt/metal extraction and electro-refining methods will be investigated for separation of actinides from lanthanides. The development of processes for solvent extraction of minor actinides (americium and curium) from HLW will be investigated in the PARTNEW project. The minor actinides are extracted from HLW in two steps: they are first coextracted with the lanthanides and then separated further from the lanthanides. Synthesis of more innovative organic extractants will be studied by the CALIXPART project with a view to directly extracting minor actinides from HLW.

Courtesy of NRG, Petten (NL)

Transmutation – Fuel

The second group, Transmutation – Fuel, has two projects on uranium-free fuels: CONFIRM and THORIUM CYCLE. The main advantage of uranium-free fuels is that when bombarded with neutrons, they do not produce other long-lived actinides. Fabrication, characterisation and safety evaluation of plutonium and americium nitride fuels for ADS will be carried out in the CONFIRM project. Plutonium nitride will be irradiated at high power in the Studsvik (S) reactor. The THORIUM CYCLE project will investigate the irradiation behaviour of thorium/plutonium fuel at high burn-up and will carry out full-core calculations for thorium-based fuel with a view to supplying key data on the burning of plutonium and minor actinides. Irradiation of thorium/plutonium oxides will be carried out in the HFR reactor at Petten (NL) and the KWO reactor at Obrigheim (D). Another project is planned to study the feasibility of fabrication and characterisation of uranium-free americium oxide fuel.

transmutation and for ADS-related materials. These projects are accompanied by technological support studies on spallation liquid-metal target materials and coolants, for example lead or lead-bismuth eutectic as well as the liquid metal supporting structure materials such as martensitic steels. Results from this ensemble of projects would form the basis of future engineering design activity of an ADS.

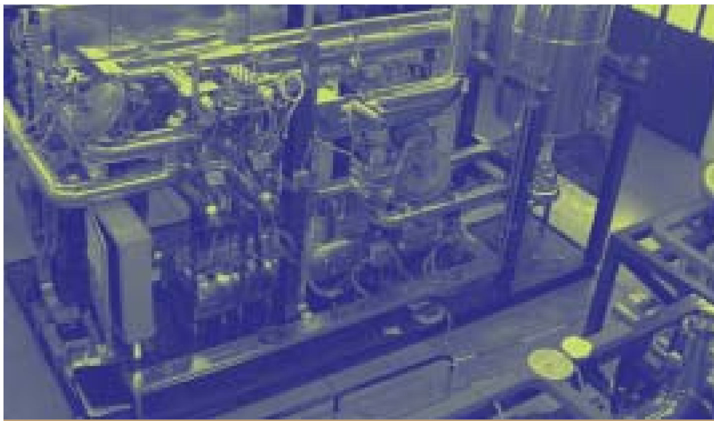


Using inverse kinematic techniques, measured nuclear cross section data in millibarns (mb) of nuclides produced in spallation reactions of Pb-208 and 1000 MeV protons.

Courtesy of GSI, Darmstadt, (D)

Transmutation – Basic Studies

The third group, Transmutation – Basic Studies, has three projects: MUSE, n_TOF_ND_ADS and HINDAS. The MUSE project aims to provide validated analytical tools for sub-critical neutronics calculations for ADS study. The corresponding experiments will be carried out in the MASURCA sub-critical facility at Cadarache (F) driven by a D-T neutron source. The other two projects will provide accurate nuclear data for a number of relevant isotopes for the design of an ADS. The n_TOF_ND_ADS project at CERN aims to produce, evaluate and disseminate neutron cross-section data in the energy range from 1 eV up to 250 MeV for most of the isotopes among the actinides and long-lived fission products that are being considered for transmutation. HINDAS will provide high quality experimental data on proton- and neutron-induced reactions above 20 MeV for iron, lead and uranium elements and will develop validated nuclear models in the energy range of 20-200 MeV and beyond.



Courtesy of ENEA, Brasimone (I)

Transmutation – Technological Support

The fourth group, Transmutation – Technological Support, has two projects that will study spallation liquid metal target materials and coolants as well as the liquid metal supporting structure materials. The TECLA project will assess the use of lead alloys both as spallation liquid metal targets and as coolants for ADS. The corrosion of structural materials by lead alloys, the protection of structural materials and the physiochemistry and thermo-hydraulics of liquid lead alloys will also be studied. An assessment of the combined effects of proton/neutron irradiation and liquid metal corrosion will be made. The SPIRE project addresses the irradiation effects on the supporting structure of a liquid metal spallation target. The effects of spallation products on the mechanical properties and microstructure of selected structural steels (e.g. martensitic steels) will be investigated by ion-beam and neutron irradiations in several reactors. These studies will be supplemented by experimental operation of a liquid metal spallation target irradiated by a proton beam at a power level of about 1MW at the SINQ facility at PSI in Villigen (CH). Preliminary design studies of an ADS transmutation device are also planned.

7. Outlook

The transmutation of minor actinides and long-lived fission products in present-day thermal fission reactors is unlikely to be practicable or economically viable. However, a partial transmutation of plutonium is possible in thermal reactors as has been demonstrated by the growing use of MOX fuel. The transmutation of minor actinides in thermal reactors requires new modes of operation and gives rise to higher actinides. Fast reactors would be more effective but their operation is not without problems for minor actinide-rich fuels.

The future for transmutation of long-lived fission products and minor actinides is likely to be tied up with accelerator driven systems (ADS). Conceptually, ADS have many advantages but a major R&D effort over several decades is required before they will be suitable for transmutation on an industrial scale. Some of the problems to be resolved relate to the reliable continuous operation of accelerators at high power, the continuous operation of targets, and problems relating to radioactivity and impurity build-up in the liquid metal target. Other problems to be solved are reliability of the window separating the accelerator and target, the development of materials with good resistance to corrosion and radiation damage, the development of safe actinide fuels, and adequate safety and control of the coupled accelerator and the sub-critical reactor system.

Transmutation can ease the waste-management problem only if adequate and highly efficient partitioning (chemical separation) techniques can be developed on an industrial scale. However, R&D on partitioning and transmutation can proceed in parallel. Nevertheless, the need for a geological repository will not go away completely as in practice neither partitioning nor transmutation can be implemented with the ideal 100% efficiency.

8. Conclusions

Partitioning and transmutation techniques offer the possibility of minimising the by-products from spent nuclear fuel that contribute to the long-term risks of disposal. Chemical separation and transmutation of the most hazardous elements contained in the waste (such as americium, neptunium, caesium, technetium and iodine) in accelerator-driven sub-critical systems to less harmful products with shorter half-lives will go a long way towards resolving the waste-management problem. Successful implementation of such a P&T system would significantly reduce the long-term technical demands placed upon sites for geological disposal of radioactive waste by reducing long-term toxicity and waste volume.

We note, however, that the fuel-cycle management policy of balancing the production and consumption of waste via P&T will be fully effective only if it is pursued consistently for several decades. Partitioning and transmutation of long-lived radionuclides will render them less hazardous, though not without significant financial investment. Any possible additional risk to individual operators must be carefully minimized. Overall, partitioning and transmutation promises to open a path towards an easing of the nuclear waste management problem.

9. Glossary of terms

Actinides:
These are the fifteen elements from actinium (atomic number 89) to lawrencium (atomic number 103) in the periodic table.

Americium-241:
An isotope of the minor actinide americium with mass number 241.

Calixarenes:
These are macrocycles made up of phenolic units linked by methylene bridges and possessing basket-shaped cavities. The calixarenes are useful starting building blocks to synthesise molecular receptors with a desired property. Their easy functionalisation leads to a large variety of macrocyclic architectures involved, in particular, in selective extraction of cations. Association of basic calixarenes with chosen binding units leads to receptors which can be tailored for a target cation.

Fission products:
They are the main fragments from nuclear fission or products of their decay. Fission of U-235 leads to fission fragments with isotopes in the range of mass numbers 72 to 161.

Geological repository:
A deep underground facility in which hazardous nuclear waste may be placed for long-term disposal.

High-level waste (HLW):
Either spent fuel or waste from spent fuel after reprocessing. More than 99% of the radioactivity in HLW at the time of discharge from the reactor is contained in the fission products.

Intermediate-level waste (ILW):
Contains high amounts of radioactivity and requires shielding. Sources of ILW arise from nuclear fuel cladding and structures of the reactor core.

Lanthanides:
These are the fifteen elements from lanthanum (atomic number 57) to lutecium (atomic number 71) in the periodic table. They are also known as the rare earth elements.

Low-level waste (LLW):
Contains only a small amount of radioactivity with negligible amounts of long-lived activity. Disposal sites (shallow-earth burial sites or underground silos) for LLW are operated in many countries.

MeV:
Million electron Volts

Minor actinides:
Neptunium, americium and curium elements formed directly or indirectly in irradiated nuclear fuel in a fission reactor.

MOX fuel:
Mixed OXide uranium and plutonium fuel used in standard light water reactors.

Pyrochemical processes:
These are liquid-liquid extraction processes using immiscible molten metal, molten salt phases or electrometallurgical processes in non-aqueous media.

Spallation reactions:
When a heavy metal such as lead is bombarded by a high energy proton beam from an accelerator, protons dislodge one or two nucleons with high energy from a nucleus through direct impact and transfer a significant amount of energy to the atom. This energy is released by the atom in “evaporating” high energy neutrons—this constitutes a useful source of neutrons. These reactions are known as spallation reactions.

Transuranic elements:
Those elements that lie beyond uranium (atomic number 92) in the periodic table. They are essentially produced artificially although traces of plutonium (atomic number 94) are found in nature.

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One of the important items of Research and Training in the field of Nuclear Energy in the EURATOM Fifth Framework Programme (1998-2002) is partitioning and transmutation within the implementation of the work on safety of the fuel cycle which deals with nuclear waste and spent fuel management.

The main concern in the disposal of radioactive waste is related to long-lived radionuclides—some of them will remain hazardous for tens of thousands of years. A sizeable effort to that end is already well under way, the main approach being the disposal of nuclear waste in deep geological formations. By partitioning (chemical separation) and transmutation (radionuclide conversion), it might be possible to reduce the amount of long-lived component of the radioactive waste, thus easing the waste management problem. The objective of EU research and development effort on partitioning and transmutation is to study its practicability on an industrial scale.

The purpose of the present brochure on partitioning and transmutation is to give a wider dissemination of research activities supported by the European Community in this area to the non-specialist scientific community including the interested politicians.

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