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Partitioning and transmutation (P&T) 1997

Status report

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Summary

Long-lived radioactive nuclides which are radiotoxic and which must be isolated from the biosphere for a very long period of time are formed in nuclear reactors. However, the long-term radiotoxicity of the waste can be reduced nowadays by converting these long-lived nuclides into short-lived or stable nuclides through neutron irradiation. This conversion process is called transmutation. By reducing the radiotoxicity of the waste, the size and cost of a future repository can be reduced at the same time that the fuel is used more efficiently. During the actual transmutation process, energy is released which can be converted into electrical energy in the same way as in a conventional nuclear reactor. To be able to transmute the long-lived radioactive atoms in the waste it is necessary to separate them from other types of atoms. The reason for this is that otherwise, the fission products absorb the neutrons which are necessary to incinerate the long-lived atoms and this results in an inefficient transmutation process. Furthermore, additional quantities of certain long-lived radioactive nuclides can be formed in connection with the irradiation of fission products.

The possibility of carrying out partitioning and transmutation, as a stage in the management of spent nuclear fuel and high-level waste, can nowadays largely be considered to be scientifically proven. However, it is still too early to determine whether this type of waste treatment has cost- or safety-related advantages in the short term, compared with current plans for the management of spent nuclear fuel and high-level waste. Neutrons from both thermal reactors and fast reactors as well as accelerator-driven subcritical reactors can be used for the transmutation of long-lived radioactive nuclides. It should be emphasized that thermal reactors and fast reactors have been constructed and operated and adequate experience has been obtained over a long period of time, while accelerator-driven reactors are only at the drawing board stage. Each of these neutron sources has advantages and disadvantages for transmutation and they are probably each best suited for certain types of nuclides. Current consensus with respect to comparisons is that transmutation in reactor types with high-energy neutrons is advantageous with respect to the reduction of the total quantity of higher actinides coupled with a relatively efficient energy production, while reactors with low-energy neutrons are primarily advantageous if the aim is to reduce the quantity of certain radiotoxic atoms and generate low-cost energy. One possible development is to use hybrid systems where each type of neutron source has a determined function. In such systems, the special properties of an accelerator-driven system are used to achieve the efficient transmutation of neptunium, americium as well as possibly curium and other types of atoms with small fission cross-sections. In such cases, the accelerator-driven system serves to supplement more conventional nuclear reactors rather than to replace them.

What is characteristic for all transmutation processes is that they must work together with a suitable chemical partitioning process where non-transmuted material is recovered from irradiated material with a high degree of efficiency. The rest becomes radioactive waste. When the effect of a combined partitioning and transmutation process is evaluated, there is a strong link between the degree of recovery in the partitioning process, the efficiency of the transmutation process and the losses of non-transmuted material to various waste streams. Both aqueous liquid-liquid extraction and

various pyrochemical partitioning methods have been proposed for the partitioning process. There is considerable knowledge of aqueous liquid-liquid extraction and this is a result of over 40 years of operating experience on an industrial scale. This means that there is considerable awareness of the advantages and disadvantages of this technology. On the other hand, pyrochemical partitioning methods for the treatment of radioactive substances must still be considered to be at the laboratory stage and still require the extensive development of methods as well as new kinds of equipment. This means that there is still a lack of adequate knowledge of the advantages and disadvantages of these methods in connection with the industrial-scale treatment of radioactive substances. Therefore, it still seems to be too early to attempt to compare aqueous chemistry with pyrochemistry on the basis of equal knowledge. Currently, the USA and Russia show the greatest interest in pyrochemistry while the largest member states of the European Union and Japan mainly continue to focus their development work on the improvement and further development of aqueous methods. If partitioning and transmutation is to be a realistic alternative to the current fuel cycles, significant long-term R&D work on partitioning techniques will be necessary.

It is quite clear today that partitioning and transmutation cannot eliminate the need for a high-level waste repository. In a short-term perspective, the radioactivity of the waste will even be higher than is classically the case due to the fact that long-lived nuclides, to a large extent, will have been converted to considerably more short-lived nuclides. On the other hand, the shorter half-lifes will mean that the waste will become less hazardous relatively quickly with time. In spite of the use of an efficient partitioning and transmutation technology, the waste will contain small quantities of very long-lived radioactive nuclides.

This conditioning of the fuel also makes it possible, in principle, to gain access to weapons-grade material in a relatively pure form. However, on the other hand, there is a somewhat greater possibility of designing fuel, target or entire accelerator-driven systems to make them more secure in terms of safeguards than is the case with conventional reactors and fast reactors.

Even if there is a broad scientific consensus that current types of fuel cycles in combination with planned waste management and geological final disposal will provide a satisfactory protection for humanity in the foreseeable future, there is still a considerable interest in investigating whether an additional reduction in the future potential hazard of the waste can be achieved through partitioning and transmutation and in investigating the cost at which this can be done. One of the weaknesses of the repositories, which are currently being planned, is the difficulty of predicting all possible future events which could affect the performance of the repository. The strength of a partitioning and transmutation process could be to reduce any possible future consequences of such unanticipated events. On the other hand, is it the case that the development of partitioning and transmutation processes will hide the fact that the future risks associated with a well-designed repository are already considered to be very small?

In previous reports, we have emphasized that the introduction of an economically viable partitioning and transmutation technology is most probable in an expanding nuclear power programme. In this context, it is still being debated whether an increased use of nuclear power along with partitioning and transmutation processes should be considered

to be a possible objective or whether partitioning and transmutation technology can be an objective, regardless of the future development of conventional nuclear power. A relatively rapid global expansion of nuclear power will certainly result in an increased price of fresh nuclear fuel which may make fuel management based on reprocessing and recycling more economical than it is today. Then fast reactors and accelerator-driven reactors may become attractive in economic terms and from the standpoint of the management of natural resources, since they can utilize the energy stored in natural uranium and thorium a hundred times more efficiently than today's light water reactors. It is therefore possible that the spent nuclear fuel generated today may, instead of being waste, become an important raw material for the further generation of nuclear energy.

Research on and the evaluation of partitioning and transmutation are currently in progress in many industrial countries and within several international organizations due to its potential as a long-term, sustainable energy source with low environmental impact and due to its ability to destroy many long-lived nuclides. This can lead to radioactive waste repositories which are easier to accept. The cost of the research and development work on partitioning and transmutation is considered to be so great that international co-operation is required. In this context, it may be worth observing that the justification for the development of partitioning and transmutation technology in Europe, Russia and Japan seems to be different from that of the USA.

With respect to Sweden, we recommend a balanced research work on both partitioning and transmutation technology. Sweden must conduct its own research to gain insight into and to build confidence in evaluations of international development work within the area of partitioning and transmutation. Since the further education of highly qualified personnel is required for as long as Sweden still has nuclear facilities, research and development within the areas of partitioning and transmutation technology is a good basis for attracting new and competent students to subjects related to nuclear power.

Within the area of partitioning, it is above all a question of locating new reagents which can be used to simplify the necessary partitioning processes and minimize the losses. The requirements with respect to high selectivity and minor losses will be significantly higher in a recirculating system based on transmutation than in the reprocessing facilities of today where only uranium and plutonium are recovered. Since, in terms of experience, today's losses are intimately connected to the generation of secondary waste from which the substances in question can only be recovered with great difficulty, new reagents are required which both generate the smallest amount of secondary waste possible and which generate the type of secondary waste which allows for efficient and simple recovery of the long-lived radionuclides. If the utilized reagents can be easily destroyed, by dry or wet incineration and conversion into non-complex gaseous chemical compounds, this will open up good opportunities for the recovery of the radionuclides. Suitable reagents may, for example, be based on hydrogen, carbon, oxygen and nitrogen atoms alone so that they can be completely incinerated. By applying modern molecular modelling and quantum mechanical calculations, the design of suitable molecules can be facilitated. However, experimental studies of their partitioning properties are still necessary. The limited work which is currently being conducted in Sweden should be continued in a long-term research work.

From a purely technical standpoint, it would seem that a combination of different types of reactor systems would give the best possible transmutation efficiency. While existing light water reactors can be utilized for increased plutonium incineration, there is currently consensus about the view that reactors with high-energy neutrons are necessary to achieve a sufficiently high transmutation efficiency for neptunium, americium, curium and certain fission products. Sodium-cooled breeder reactors can be used to transmute neptunium and americium with a high degree of efficiency while the transmutation of curium and long-lived fission products requires different types of coolants, such as liquid heavy metals (lead, lead/bismuth). By allowing an accelerator-based neutron source to drive a subcritical heavy metal-cooled reactor, the potential for transmutation of fission products is increased, at the same time that satisfactory safety margins are achieved for certain fuel types with a low share of delayed neutrons and a high heat conductivity.

Regardless of what types of systems are ultimately applied, as a rule, it can be stated that 3–4 reactors are necessary with a high-energy neutron spectrum to transmute waste from the Swedish light water reactor park. Thus, the Swedish research programme on transmutation systems, should perhaps focus on studies of fast reactor systems for transmutation, especially heavy metal-cooled systems with or without connected neutron-producing accelerators. Studies of fuel types which are suitable for transmutation, such as oxides and mononitrides are of particular interest. At the same time, calculations of the efficiency of the fission product transmutation in accelerator-driven systems should be carried out. Measurements of less well-known cross-sections for reactions which are of importance for the transmutation of certain actinides are also required as a basis for such work.

Contents

1 Introduction

Nuclear reactions can, in principle, be utilized to convert most of the long-lived radioactive nuclides in radioactive waste into short-lived or stable nuclides. This is of interest, since the presence of long-lived radioactive nuclides in the waste means that this can be perceived to be a potential environmental hazard for a very long period of time. The most attractive nuclear reactions, from the standpoint of transmutation, are based on the bombardment of selected elements by neutrons. Irradiation is then followed by the partitioning and recycling of non-reacted material.

The use of a combination of partitioning and transmutation to minimize the quantity of long-lived radioactive atoms in waste from nuclear energy generation was proposed as early as 1964 by Steinberg et al. [Ste64]. The development of separation and transmutation until the end of the 1980's has been described by Croff in a report published in 1990 [Cro90].

Transmutation through the fission of heavy radioactive nuclides generates a considerable quantity of energy. A transmutation system can therefore also function as a facility for energy production. In a self-sustaining system (critical reactor), thermal energy produced can be converted into electricity and largely be transferred from the facility. On the other hand, in a non-self sustaining system (a combination of an accelerator, target and subcritical reactor) a part of the electricity generated will be consumed by the facility to operate the accelerator which, via the target, supplies the system with extra neutrons.

Interest in partitioning and transmutation gained new momentum through the development of technology during the end of the 1980's and this was accelerated by an international conference in Saltsjöbaden in 1991 [Jam91] and through political decisions in several countries. The development within the area during the first half of the 1990's has been summarized in a few previously published SKB reports. The high intensity of research activities within the area has continued and, in certain cases, it has intensified. This report describes developments up to the end of 1997. Unlike the situation at the time of the publication of previous status reports on partitioning and transmutation within SKB's technical report series [Skå92, Gud93, Skå95], several new international status reports now exist (for example [IEA97, Gud97:2]) and a comprehensive report from the OECD/NEA is currently being prepared.

This report, which was originally published in Swedish and has been translated into English, provides a basic technical description of transmutation and related partitioning processes, summarizes work in progress in different countries as well as describes the most probable future developments within the area from the current perspective.

Due to the interest which currently exists in accelerator-driven subcritical reactors, a more detailed description of such systems is provided in Appendix 1. We hope that this will be helpful to laymen. For those who are interested in the mathematical and process-related aspects, a few essential relationships between efficiencies and transmutation efficiency are briefly described in Appendix 2. Since a large number of specialist terms

and abbreviations have been used in this report, a short glossary with explanations is provided in Appendix 3.

2 Description of the basic Partitioning & Transmutation processes (P&T)

Nowadays, nuclear power production is based on the fission of heavy atomic nuclei, mainly uranium isotopes. During the fission process, thermal energy is released and new neutrons and fission products are formed. The fission products are a mixture of various elements from copper to holmium. In order for the fuel to withstand the high temperatures which arise, uranium dioxide, UO_2, is used which is a ceramic with a melting point of about 2 860°C. A side effect of the fission of uranium atoms is the simultaneous and slow formation of radioactive elements with higher atomic numbers than uranium. Several of the newly formed heavy elements contain atoms with long half-lifes.

The conversion of one element into one or more elements is called transmutation. The elements which are newly formed from uranium, in particular plutonium, contribute significantly, through fission, to the production of energy during reactor operation. The increasing concentration of fission products and the decreasing concentration of fissile atoms results in the decrease in power production with time.

Many of the newly formed elements compete for available neutrons with the atomic nuclei of heavy elements which can undergo fission during the production of new neutrons. Mainly for these reasons, nuclear fuel can only be utilized for a certain total energy production before it must be replaced by fresh fuel. The fuel is consumed in a process known as fuel burnup. During burnup, the fuel produces thermal energy which is usually specified in MWd/kg (megawatt days per kilogram of original heavy metal content, i.e. most often per kg uranium in the fresh fuel). One MWd/kg corresponds to 24 000 kWh/kg in the form of heat. Typical burnups for the reactor types which currently exist in Sweden are between 40 and 60 MWd/kg, more than twice the level usual in the early days of nuclear power.

When fuel is removed from the reactor, almost all of the original uranium is still present (about 97%). However, there is a change in the isotopic composition. Fission products and a large part of the heavy elements formed are also present in the fuel (a significant amount of the heavier elements, such as plutonium, have also undergone fission).

At this point, there are two courses of action. Either all of the spent fuel is considered to be hazardous waste, which must be stored and safely disposed of, or the unutilized uranium and plutonium are recovered from the fuel through chemical partitioning, known as reprocessing. In the latter case, fresh nuclear fuel can be produced from the recovered material. Since this fuel, already from the start, contains both uranium and plutonium dioxide, it is called MOX fuel (MOX = Mixed OXide). The separated fission products and other heavy elements comprise the high-level reprocessing waste. In most cases, this high-level waste is immobilized in a glass matrix by heating and adding vitrification substances. The vitrified waste is then cast in stainless metal containers and the aim is to dispose of this waste in a similar manner to unprocessed spent nuclear fuel.

Plutonium from spent MOX fuel consists of a mixture of various plutonium isotopes which is somewhat worse as fuel in conventional nuclear reactors than the plutonium which is obtained in connection with the reprocessing of the uranium-based fuel. This deterioration of the plutonium quality is accelerated when plutonium is re-used time after time and limits the number of times that it is economically viable to reprocess the fuel to three. In a similar way, the quality of recycled uranium deteriorates. However, this can partially be compensated for if the uranium is recycled to an enrichment plant where it replaces a small quantity of natural uranium. This means that, in the case of reprocessing and recycling, after a few cycles, it is no longer worth reprocessing and converting the residual spent MOX fuel into fresh fuel for thermal reactors. However, such plutonium can still be utilized as fuel in fast critical reactors or in accelerator-driven reactors.

The partitioning of spent nuclear fuel into suitably selected groups of elements makes it possible, in principle to utilize suitable nuclear reactions to convert most of the long-lived nuclides into short-lived or stable nuclides, often to elements other than the original elements. Through this process, it may be possible to reduce the long-term hazard of the waste. Transmutation has also become a synonym for the conversion of long-lived radioactive nuclides in radioactive waste to more short-lived nuclides. Through a transmutation process, it may be possible to reduce nuclear waste management to a safe management of relatively short-lived radioactive nuclides as well as relatively small quantities of remaining long-lived radioactive nuclides. Separation and transmutation are related processes since partitioning is a prerequisite for a meaningful transmutation. The process requirements are also intimately linked, see Appendix 2. On the other hand, the processes do not have to be located on the same site or even in the same country.

Since most chemical partitioning processes are normally not capable of separating isotopes of one element (but only different elements), the irradiation of elements with the existing isotope compositions will be the normal alternative for the transmutation process. Isotope separation is a technically feasible but expensive process. This means certain limitations concerning what can be achieved by industrial scale partitioning and transmutation within a given cost frame.

As has been mentioned, the most important nuclear reactions for the transmutation processes are achieved through irradiation with neutrons which results in fission or neutron capture in the irradiated atoms. In order to achieve a tangible reduction in the long-term hazard, the reaction products should either be stable nuclides or short-lived nuclides which rapidly decay to stable nuclides. A high neutron fluence (flux times time) is required for adequate conversion. The desire to achieve a fast conversion means that a very high neutron flux is required. For this reason, high flux reactors or accelerator-driven subcritical reactors are of interest. However, conventional thermal heavy and light water reactors may be of some use.

Since the reaction cross-sections (probability of reaction) are relatively small for many of the long-lived radioactive nuclides which are of interest, possible neutron fluxes are limited in size and many reaction products must be removed sooner or later in order not to result in undesirable by-products. The irradiated atoms must be cleaned and recycled many times to achieve a high level of destruction. This places considerable demands on the selectivity as well as the efficiency of the chemical partitioning process which is used in connection with the cleaning of the irradiated material. Several types of chemical processes are considered to be feasible. Of these, aqueous-based partitioning is currently the only technique where there is considerable industrial-scale experience of high level purification of radioactive nuclides with very high efficiency demands.

The number of cycles which are required is determined by the destruction level desired for individual long-lived radioactive nuclides, the destruction level during an irradiation and the efficiency of the separation process. The mathematical relationships are described in greater detail in Appendix 2. If, for example, the destruction level is low during an irradiation, the efficiency of the separation process must be very high in order to achieve the desired reduction of the waste inventory of long-lived nuclides, and vice versa. Since the chemical separation process plays a key role, at least as many resources must be invested in the development of this as in the development of a suitable irradiation system and in radiation target. Figure 1 shows a flow chart of a partitioning and transmutation process.

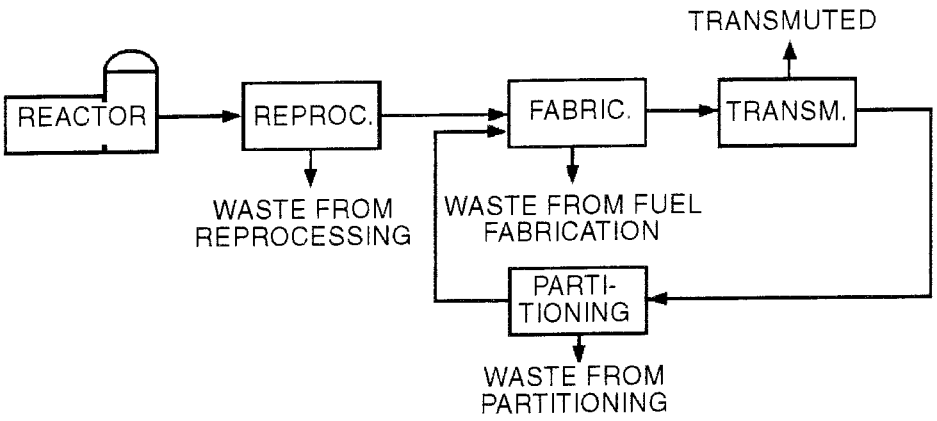

Figure 1. *Flow chart showing a partitioning and transmutation process.*

2.1 Radioactive nuclides suitable for P&T

For the destruction of a radioactive nuclide through a combination of partitioning and transmutation with neutrons, the following is required:

- The nuclide must be long-lived or lead to a decay chain which contains at least one long-lived radioactive nuclide.

- The reaction cross-section for neutrons must be sufficiently high.

- The reaction product (or products) must be stable or must have a much shorter half-life than the original nuclide.

- The reaction product must, even if it is short-lived, not lead to a decay chain which contains a long-lived radioactive nuclide.

- The nuclide shall occur in such a large concentration in radioactive waste and should be so radiotoxic that a reasonable decrease in its concentration in the waste will result in a tangible reduction in the potential hazard of the waste, in a long-term perspective.

- The nuclide must preferably belong to an element whose other isotopes in the mixture in question do not result in a tangible new production of long-lived radioactive nuclides through irradiation.

- The nuclide shall belong to an element which can be recovered chemically in pure form, and with a high degree of efficiency, from radioactive waste.

- Finally, it is advantageous if the nuclide is fissile and can thereby generate energy, the sale of which can contribute to the financing of the partitioning and transmutation processes.

It is mainly only the heavy elements in the actinide series which, to a reasonable extent, fulfill all of the requirements. Table 1 summarizes the long-lived nuclides which are possible candidates for a partitioning and transmutation process. Other isotopes of each element which normally occurs in spent fuel from conventional nuclear power plants are also shown in the table.

Table 1. Nuclides which are suitable candidates for a partitioning and transmutation process.

Nuclide	Half-life	Decay type	ALI (Bq)	Other isotopes in high-level waste[+]
Actinides				
^{228}Th	1.913 years	α	2×10^{5}	
^{229}Th	7340 years	α	2×10^{4}	
^{230}Th	7.7×10^{4} years	α	1×10^{5}	
^{232}Th	1.41×10^{10} years	α	3×10^{4}	
^{231}Pa	3.28×10^{4} years	α	7×10^{3}	
^{232}U	68.9 years	α	8×10^{4}	
^{233}U	1.59×10^{5} years	α	4×10^{5}	
^{234}U	2.45×10^{5} years	α	4×10^{5}	
^{235}U	7.04×10^{8} years	α	5×10^{5}	
^{236}U	2.34×10^{7} years	α	5×10^{5}	
^{238}U	4.47×10^{9} years	α	5×10^{5}	
^{237}Np	2.14×10^{6} years	α	3×10^{3}	
^{238}Pu	87.7 years	α	3×10^{5}	
^{239}Pu	2.41×10^{4} years	α	2×10^{5}	
^{240}Pu	6570 years	α	2×10^{5}	
^{241}Pu	14.4 years	α	1×10^{7}	
^{242}Pu	3.76×10^{5} years	α	3×10^{5}	
^{244}Pu	8.26×10^{7} years	α	3×10^{5}	
^{241}Am	432.1 years	α	5×10^{4}	
^{242m}Am	152 years	γ	5×10^{4}	
^{243}Am	7380 years	α	5×10^{4}	
^{242}Cm	162.8 d	α	2×10^{6}	
^{243}Cm	28.5 years	α	7×10^{4}	
^{244}Cm	18.11 years	α	9×10^{4}	
^{245}Cm	8500 years	α	5×10^{4}	
^{246}Cm	4730 years	α	5×10^{4}	
^{247}Cm	1.56×10^{7} years	α	5×10^{4}	
^{248}Cm	3.39×10^{5} years	α	1×10^{4}	
^{250}Cm	9000 years	SF	n/a	
Very long-lived fission products				
^{87}Rb	4.8×10^{10} years	β^-	4×10^{7}	85(s)
^{99}Tc	2.13×10^{5} years	β^-	1×10^{8}	none
^{107}Pd	6.5×10^{6} years	β^-	1×10^{9}	104(s), 105(s), 106(s), 108(s), 110(s)
^{129}I	1.57×10^{7} years	β^-	2×10^{5}	127(s)
^{135}Cs	2.3×10^{6} years	β^-	3×10^{7}	133(s), 134(k), 137(k)
Moderately long-lived fission products				
^{90}Sr	28.8 years	β^-	1×10^{6}	86(s), 87(s), 88(s)
^{137}Cs	30.1 years	β^-	4×10^{6}	133(s), 134(k), 135(l)
Long-lived activation products				
^{93}Zr	1.5×10^{6} years	β^-	5×10^{7}	90(s), 91(s), 92(s), 94(s), 96(s)

[+] Text in brackets, s = stable, k = relatively short-lived, l = long-lived

2.2 Transmutation

In principle, the transmutation of a nuclide into one or several others, or in a more limited sense, one isotope of an element to another isotope of the same element, can occur through irradiation by neutrons, high-energy gamma quanta or charged particles, which will lead to a change in the composition of the atomic nucleus.

Since neutron irradiation is currently considered to be most favourable, the discussion will focus on this type of particle. The nucleus captures a neutron and thereby becomes heavier. In many cases, the newly formed nucleus is unstable and undergoes radioactive decay while emitting a high-energy electron, a helium nucleus or undergoes fission if from the beginning, it is so large that fission will result in liberation of energy. The transmutation of uranium and plutonium to fission products through neutron irradiation occurs routinely in nuclear power plants.

2.2.1 Basic physics

The properties of an atomic nucleus are determined by its composition with respect to the number of protons (defines the element) and the number of neutrons. The sum of these particles is the weight of the atom. This is always close to an integer of units of atomic weight (unit of atomic weight = 1/12 of the atomic mass of ^{12}C). This integer is called the mass number and has the same numerical value as the total number of protons and neutrons in the atomic nucleus. Since there may be somewhat different numbers of neutrons for a given number of protons (an element), there are atoms of one element which have different mass numbers. Such atoms are called isotopes of one element. One or several of the element's isotopes are often stable while others undergo radioactive decay with a half-life which is characteristic of the composition of the nucleus. Through the radioactive decay, the nucleus is converted into another more stable composition. At the same time, the element is often changed by an increase or decrease in the number of protons. Sooner or later, these types of changes lead to a stable atomic nucleus. When the radioactive changes occur through the emission of high-energy electrons, β particles, the mass number remains constant. Such atoms are said to belong to an isobaric chain.

The reaction rate, dN/dt, expressed in terms of reacting (disintegrated) atomic nuclei per unit of time in a thin target is described by the following equation

$$dN/dt = -(\phi\,\sigma_0 + \lambda_0)\,N_0\,e^{-(\phi\,\sigma_0 + \lambda_0)\,t} \qquad (1)$$

where ϕ is the particle flux per unit area and unit of time (unit $m^{-2}s^{-1}$), σ_0 is the total reaction cross-section of the nucleus in a target atom (m^2), λ_0 is the decay constant (s^{-1}) ($\lambda_0 = \ln(2)/t_{½}$ where $t_{½}$ is the half-life in seconds for the atom in question if this is radioactive, for stable atoms $\lambda_0 = 0$), N_0 is the original number of target atoms which existed in the particle flux at the beginning of the irradiation and t is the irradiation time (s). The remaining number of atoms, N, at time t is expressed by the relationship

$$N = N_0\,e^{-(\phi\,\sigma_0 + \lambda_0)\,t} \qquad (2)$$

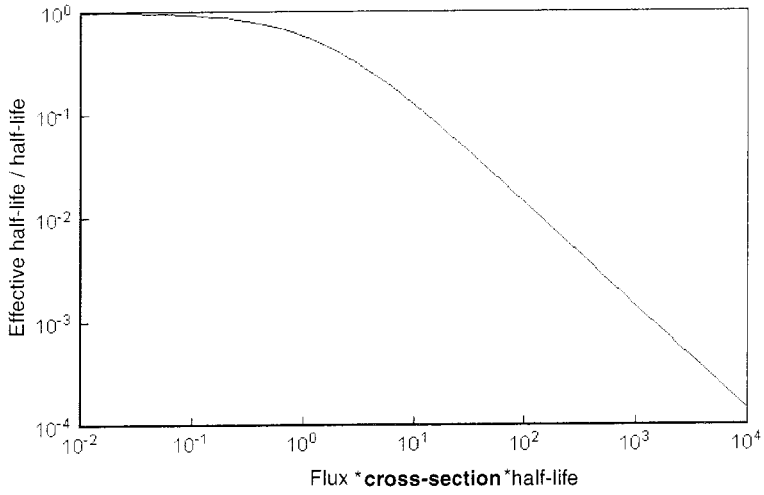

Figure 2. *Effect of flux, cross-section and half-life on the relationship between the effective and normal half-life.*

The reaction cross-sections are very small. However, this can be compensated for by a high particle flux value and a long irradiation time. Neutron fluxes of the order of magnitude of 10^{17} $m^{-2}s^{-1}$ are common in power reactors and time can be on the order of years. Note that time in equation 2 must be expressed in seconds. Equation 2 can be perceived as though the irradiated atom has an effective half-life in the particle flux which is $\ln(2)/(\phi\ \sigma_0 + \lambda_0)$ seconds. In order for the irradiation to be able to accelerate the natural decay, the product $\phi\ \sigma_0$ must be of the same order of magnitude or greater than λ_0. If $\phi\ \sigma_0 \ll \lambda_0$ there will be no significant transmutation through irradiation. Practically all atoms undergo radioactive decay regardless of whether they are irradiated or not. This is shown in Figure 2 where the relationship between the effective half-life during irradiation and the normal half-life is specified for various values of the particle flux product ($m^{-2}\,s^{-1}$), the total reaction cross-section (m^2) and the half-life (s).

Since the atoms of the reaction product will be irradiated with the particle flux in question, as long as the product is present in the flux and can, thereby, continue to react with the projectiles, the following expression for the number of product atoms N_p, applies after the irradiation time t.

$$N_p = \{(\phi\ \sigma_{0r} N_0)/[\phi\,(\sigma_p - \sigma_0) + \lambda_p - \lambda_0]\}\{e^{-(\phi\ \sigma_0 + \lambda_0)t} - e^{-(\phi\ \sigma_p + \lambda_p)t}\} \qquad (3)$$

where the index p specifies data for the product atoms, the index 0 specifies data for the original atom and σ_{0r} is the cross-section for the transmutation reactions in the original atom which leads to the formation of the product. If the product is formed through neutron capture, σ_{0r} is the cross-section for this process while σ_0 and σ_p are the total cross-sections for all reactions with each atom. If the product is formed through fission of the original atom, σ_{0r} is the cross-section for fission times the probability of the formation of the fission product.

Charged particles and gamma quanta normally have small reaction cross-sections, often of the order of magnitude of 10^{-31} m^2. Consequently, irradiation using these is less attractive for transmutation purposes since extremely high particle fluxes and high particle energies would be required for a reasonable reaction rate.

The cross-section for reactions with slow neutrons is relatively high and this then decreases with an increase in velocity. At low velocities, the cross-section is normally in reverse proportion to the speed of the neutrons. In the range with medium-high neutron speeds, the reaction cross-section shows a complex dependency on the speed with alternate high and low values. However, at very high neutron velocities, the cross-section is usually almost constant, of the order of magnitude of 10^{-28} m^2.

When an atom has captured a relatively low-energy neutron, a new nucleus is formed which has excess energy. For most atomic nuclei, the new nucleus is permanently or temporarily stabilized through the emission of excess energy in the form of electromagnetic radiation, a gamma quantum. For elements with higher atomic numbers than lead, the new nucleus can also split into two fragments of somewhat different sizes, nuclear fission. Gamma emission and nuclear fission are two competing processes which occur here. This is described by using two reaction cross-sections, one for gamma emission ($\sigma_{n,\gamma}$) and one for fission ($\sigma_{n,f}$). The total reaction cross-section then becomes the sum of these and the cross-sections for scattering etc.

Actinide atoms with an odd number of neutrons in the nucleus have large reaction cross-sections for fission with slow neutrons (order of magnitude of 10^{-25} m^2) while actinide atoms with an even number of neutrons in the nucleus have small cross-sections for fission with slow neutrons. Both types of atoms have a moderately large reaction cross-section for capturing such a neutron followed by energy emission in the form of gamma radiation (order of magnitude 10^{-26} m^2). Fission and capture are two competing processes. This means that slow neutrons often split actinide nuclei with an odd number of neutrons while actinide nuclei with an even number of neutrons instead capture the neutron, thereby increasing their weight by one unit of mass. If the new nucleus has captured too many neutrons, it will be converted, as it emits a high-energy electron, into an atomic nucleus of the next highest element. This can be illustrated by the following reactions which occur during the operation of a power reactor.

odd n-number $^{235}U + n \langle \begin{matrix} \text{2 fission products} + 3\ n \\ ^{236}U + \gamma \end{matrix}$

even n-number $^{238}U + n \rightarrow {}^{239}U + \gamma \rightarrow {}^{239}Np + \beta^- \rightarrow {}^{239}Pu + \beta^-$

In this example, ^{239}U, ^{239}Np and ^{239}Pu are an isobaric chain. The radioactive conversion from ^{239}U to ^{239}Pu is fast since ^{239}U and ^{239}Np are short-lived. ^{235}U, ^{236}U, ^{238}U and ^{239}Pu are all very long-lived and are slowly converted while emitting a helium atom (α-particle). Since ^{239}Pu has an odd number of neutrons, it can react in a similar way as ^{235}U, that is either undergo fission or form ^{240}Pu.

Elements with a somewhat lower atomic number than the actinides cannot be split by neutrons of normal energies. Instead, they can only capture the neutrons and this can result in isobaric chains. The reaction cross-sections are of the order of magnitude of

$10^{-29} - 10^{-27}$ m^2. This step by step building of higher elements is also considered to occur at certain stages in the development of stars and is considered to be one of the main reasons why elements with higher atomic numbers than iron are found on Earth.

In most cases, neutron capture within the nucleus of a lighter element leads to the new nucleus having a shorter half-life than the original nucleus. However, there are many exceptions. This phenomenon can be used to convert many unstable atomic nuclei with long half-lives to more short-lived nuclei, which are thereby spontaneously converted into stable nuclides at a faster rate.

2.2.2 Products from neutron irradiation

Equation (2) describes how the quantity of an atom is changed through neutron irradiation during a given period of time. The reaction cross-section which must be used is the sum of the reaction cross-section for capture, $\sigma_{n,\gamma}$, and for fission $\sigma_{n,f}$. Data are provided in Tables 2 and 3 for a number of selected atoms and a few typical neutron flux types. The result of an irradiation will therefore be a mixture of fission products and different actinide atoms (old and new). The share which will be split can be estimated from the ratio $\sigma_{n,f}/(\sigma_{n,f} + \sigma_{n,\gamma})$ and the share which will be captured by neutrons from $\sigma_{n,\gamma}/(\sigma_{n,f} + \sigma_{n,\gamma})$, see Figure 3.

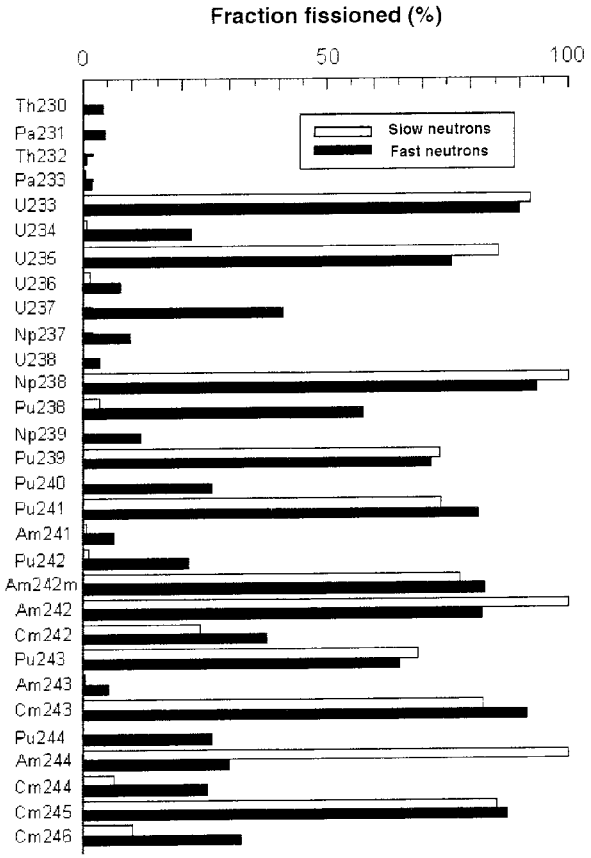

Figure 3. *The fraction of fission in all reactions during neutron irradiation of a few important actinide atoms.*

Table 2. Reaction cross sections with thermal neutrons for different types of actinide atoms.

	$t_{1/2}$	$\sigma_{n,\gamma}$ (10^{-28} m^2)	$\sigma_{n,\gamma m}$ (10^{-28} m^2)	$\sigma_{n,f}$ (10^{-28} m^2)
^{227}Ac	21.8 a	890		2.9×10^{-4}
^{227}Th	18.72 d			200
^{228}Th	1.913 a	123		0.3
^{229}Th	7340 a	54		30.5
^{230}Th	77000 a	23.2		0.0012
^{231}Pa	32500 a	210		0.010
^{232}Th	1.405×10^{10} a	7.32		2.5×10^{-6}
^{232}Pa	1.310 d	464		700
^{232}U	68.90 a	74.9		77
^{233}Th	22.30 m	1500		15
^{233}Pa	26.97 d	19	20	0.1
^{233}U	159200 a	45.5		529.1
^{234}Th	24.1 d		1.8	0.01
^{234m}Pa	1.170 m			500
^{234}Pa	6.700 h			5000
^{234}U	245500 a	99.8		0.6
^{235}U	7.038×10^{8} a	98.3		582.6
^{236}U	23420000 a	5.11		0.07
^{237}U	6.750 d	443		0.35
^{237}Np	2140000 a	176		0.0215
^{238}U	4.468×10^{9} a	2.68		4.0×10^{-6}
^{238}Np	2.117 d			2088
^{238}Pu	87.7 a	540		17.9
^{239}U	23.45 m	22		14
^{239}Np	2.357 d	36	32	
^{239}Pu	24110 a	269		748.1
^{240}Pu	6563 a	289.5		0.06
^{241}Pu	14.35 a	358		1010
^{241}Am	432.2 a	533	54	3.2
^{242}Pu	373300 a	18.5		0.2
^{242m}Am	141 a	2000		6950
^{242}Am	16.02 h			2100
^{242}Cm	162.8 d	16		5
^{243}Pu	4.956 h	87		196
^{243}Am	7370 a	75.1	3.8	0.184
^{243}Cm	29.1 a	130		617
^{244}Pu	80800000 a	1.7		
^{244m}Am	26.0 m			1600
^{244}Am	10.1 h			2300
^{244}Cm	18.1 a	15.2		1.04
^{245}Pu	10.5 h	150		
^{245}Cm	8500 a	369		2145
^{246}Cm	4730 a	1.22		0.14
^{247}Cm	15600000 a	57		82
^{248}Cm	340000 a	2.6		0.37
^{249}Cm	64.15 m	1.6		
^{249}Bk	320.0 d	746		
^{249}Cf	351.0 a	497		1642
^{250}Bk	3.217 h	350		960
^{250}Cf	13.08 a	2034		
^{251}Cf	898 a	2850		4895
^{252}Cf	2.645 a	20.4		32
^{253}Cf	17.81 d	17.6		1300
^{253}Es	20.47 d	5.8	178	
^{254m}Es	39.3 h			1826
^{254}Es	275.7 d	28.3		1966
^{254}Fm	3.240 h	76		1324
^{255}Es	39.8 d	55		
^{255}Fm	20.07 h	26		3360
^{256}Fm	157.6 m	45		
^{257}Fm	100.5 d			2950

Table 3. Reaction cross-sections for different actinide atoms with 60 keV neutrons in a metal-cooled acclerator-driven reactor.

	$t_{1/2}$	$\sigma_{n,\gamma}$ (10^{-28} m^2)	$\sigma_{n,\gamma m}$ (10^{-28} m^2)	$\sigma_{n,f}$ (10^{-28} m^2)
^{230}Th	77000 a	0.391		0.017
^{231}Pa	32500 a	3.953		0.184
^{232}Th	1.405×10^{10} a	0.578		0.004
^{233}Pa	26.97 d	1.630		0.029
^{233}U	159200 a	0.365		3.304
^{234}U	245500 a	0.913		0.256
^{235}U	7.038×10^{8} a	0.739		2.355
^{236}U	23420000 a	0.786		0.065
^{237}U	6.750 d	0.850		0.588
^{237}Np	2140000 a	2.210		0.235
^{238}U	4.468×10^{9} a	0.531		0.019
^{238}Np	2.117 d	0.275		4.021
^{238}Pu	87.7 a	0.824		1.126
^{239}Np	2.357 d	2.774		0.371
^{239}Pu	24110 a	0.820		2.085
^{240}Pu	6563 a	0.847		0.305
^{241}Pu	14.35 a	0.701		3.103
^{241}Am	432.2 a		2.570	0.178
^{242}Pu	373300 a	0.698		0.192
^{242m}Am	141 a	0.813		3.941
^{242}Am	16.02 h			
^{242}Cm	162.8 d	0.801		0.484
^{243}Pu	4.956 h	0.578		1.086
^{243}Am	7370 a	2.381		0.130
^{243}Cm	29.1 a	0.355		3.920
^{244}Pu	80800000 a	0.433		0.154
^{241}Am	10.1 h	0.770		0.330
^{245}Cm	8500 a	0.468		3.295
^{246}Cm	4730 a	0.368		0.176

Since fission and neutron capture in actinide atoms compete, both fission products and heavy atoms are formed. It is relatively complex to calculate the quantities and types of fission products which are formed. This is due to the fact that each kind of atom which are split by a neutron of a certain energy results in a unique distribution of product atoms. This is usually described by the following equation:

$$y_{A,Z} = y_A \, s^{-1} \, (2\pi)^{-1/2} \, e^{-(Z-Z_p)^2/(2s^2)} \qquad (4)$$

In this instance $y_{A,Z}$ is the fraction of fissions which leads to atoms of the element with the atomic number of Z and the atomic mass of A, y_A is the fraction of all fissions which result in atoms with a mass of A, which is also called the chain fission yield, s is a breadth parameter which specifies how large the distribution is with respect to Z at a particular A and Z_p is the most probable atomic number for the mass A. Z_p has values which almost exclusively lead to atoms that are richer in neutrons than those that are stable at the same mass. Consequently, most of the fission products will be radioactive and will emit β radiation when they are converted into more stable atoms. The three parameters, y_A, s and Z_p are calculated from measurements of the quantities of the products during fission under well-defined conditions, and are normally stated in tabular form for different A and a given split atom. The distribution of the total quantity of products at a given atomic mass, y_A, as a function of A often shows two maximum values during the fission of the actinides which are most interesting for transmutation. Some typical examples are provided in Figure 4. As shown in the figure, different quantities of each mass are obtained when different actinide atoms are split. The

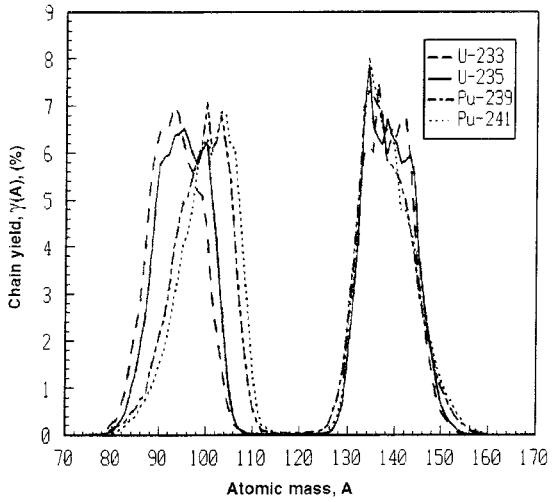

Figure 4. *The distribution of different masses of products from the fission of a few actinide isotopes with thermal neutrons.*

greatest differences are observed for the lightest fission products. Since the split actinide atom always results in two fission product atoms, all of the chain fission yields add up to 200% since the number of atoms is doubled. The figure also shows that there are distinct differences in the quantity of certain light masses. As an example, the fission of ^{241}Pu results in half of the production of the mass of 90 compared with the fission of ^{235}U. Since the chain with the mass of 90 results in the relatively long-lived ^{90}Sr the produced quantity of this element will be dependent on which actinide atoms are split. The curves in Figure 4 apply to fission with thermal neutrons. As more and more high-energetic neutrons are used, the appearance of the curves becomes somewhat different. This primarily occurs through an increase in the values in the minimum range around the 120 mass at the cost of other high values. At the same time a minor broadening of the figure occurs through a downward respectively upward displacement in the mass of the lower and upper boundaries. Figure 5 illustrates this relationship by showing the chain reactions at a high neutron energy (14 MeV) for the same uranium

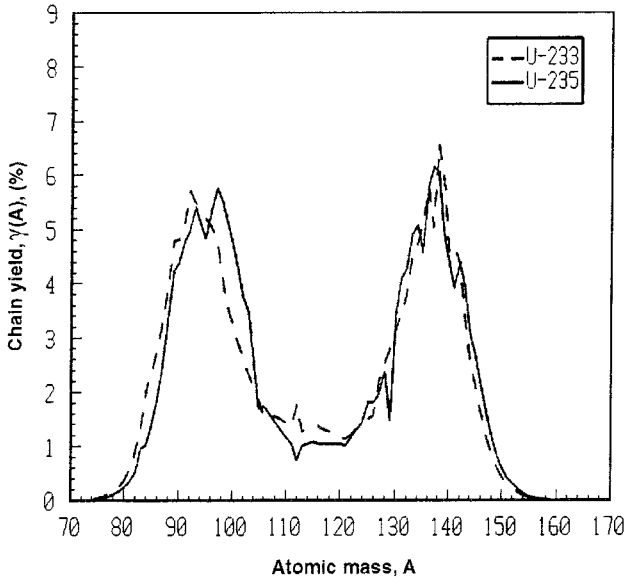

Figure 5. *The distribution of different masses of products from the fission of ^{233}U and ^{235}U with 14 MeV neutrons.*

isotopes as in Figure 4. Consequently, the composition of the fission product mixture is dependent on which actinide atoms that occur in the irradiated material, their relative occurrence in the material and the energy of the neutrons used.

Neutron capture leads to atoms with higher masses. When a sufficiently high mass is achieved, the newly formed atom of the particular element in question will decay to the next highest element while emitting β radiation. One example of this process has already been provided previously where ^{238}U, through neutron capture forms the short-lived ^{239}U which then decays while emitting β radiation to ^{239}Pu via an interim stage as ^{239}Np.

The quantity of an initial neutron capture product after a given irradiation time, N_p can be calculated using equation (3).

The formation of heavier atoms does not only occur among the actinides. It also occurs in the fission products which are formed during the irradiation of heavy elements. This results in a slow change in the composition of the mixture of fission products. In certain cases, new radioactive atoms are thereby formed from the stable atoms which originate from the radioactive decay of short-lived fission products in a mass chain. A typical example is the moderately long-lived cesium isotope ^{134}Cs ($t_{½}$ 2.06 years, $\sigma_{n,\gamma}$ 1.4×10^{-26} m^2). Primarily formed fission products in the 134 mass chain lead relatively rapidly (via β decay) to the stable xenon isotope ^{134}Xe. Therefore, nothing leads to ^{134}Cs which is formed at a later stage in this mass chain. On the other hand, radioactive decay in the 133 mass chain leads relatively rapidly to the formation of the stable isotope ^{133}Cs. Neutron capture in ^{133}Cs ($\sigma_{n,\gamma}$ 2.9×10^{-27} m^2) then leads to the formation of the radioactive ^{134}Cs. A very short bombardment of an actinide by a large number of neutrons, such as ^{233}U, therefore leads to the formation of almost no ^{134}Cs while a long irradiation time with the same quantity of neutrons results in tangible quantities of ^{134}Cs. Further irradiation of ^{134}Cs then leads to the formation of the very long-lived ^{135}Cs ($t_{½}$ 2×10^{6} years, $\sigma_{n,\gamma}$ 8.7×10^{-28} m^2). In other cases, a very slow conversion occurs of relatively long-lived radioactive fission products into short-lived products which rapidly decay into stable atoms. One example of this is ^{90}Sr ($t_{½}$ 28.5 years, $\sigma_{n,\gamma}$ 9×10^{-29} m^2), which is formed during the decay of other short-lived predecessors in the 90 mass chain. During neutron capture in ^{90}Sr, the short-lived ^{91}Sr ($t_{½}$ 9.5 hours) is formed which decays via ^{91}Y ($t_{½}$ 58.5 days) to stable ^{91}Zr. The short half-lifes for the interim products means that these occur in very small quantities if the neutron flux is not very high (ϕ in equation 2). A minor quantity leads, in turn, to a very small quantity of the next neutron capture product.

Even if certain simple, rough calculations can be manually performed, the scope of the problem means that calculations of the composition after neutron irradiation is normally performed by computer codes using extensive data bases of the properties of different atoms.

Usually, one code package is used to calculate the neutron spectrum and effective transmutation cross-section and another is used for burnup calculations. In recent years, the need for a specially adapted code for transmutation calculations has led to the development of several new computer codes for this purpose.

Through the occurrence of various competing processes, an optimum product mixture is often obtained if the irradiation of the material which is to be transmuted occurs during a short time and with a relatively low flux of only high-energy neutrons. This is then followed by a chemical partitioning of products (waste) and non-reacted material, after which the non-reacted material is recycled for an additional irradiation. Unfortunately, this leads to a relatively large loss of non-transmuted material to the waste (see Appendix 2). Furthermore, the time required for a reasonable degree of transmutation is very long, which results in a large inventory of non-transmuted material circulating in the system. These conditions mean that a compromise must be made between the neutron energy used, the transmutation rate, the product composition, the irradiation time between the partitioning and the loss of non-transmuted material to the waste. This is discussed in greater detail in the next section.

2.2.3 Target and fuel

The material which is to be irradiated in order to reduce its volume and/or half-life is, per definition, radioactive, see Table 1. For this reason, the waste must be well-contained in order to prevent its dispersion to the environment. Radioactive substances always generate energy which is converted into thermal energy. Furthermore, the nuclear reactions during irradiation also lead to the generation of heat, which is why effective cooling of the irradiated material is necessary. In general, only physical and chemical properties will be discussed in this section. The nuclear properties such as neutron multiplication, criticality, etc., will be dealt with in a subsequent section.

Irradiation is normally carried out using a solid or liquid target. Since the design of the irradiation system, containment and management of the irradiated material will be different, depending on whether the target is liquid or solid, these alternatives will be dealt with individually.

Liquid targets. Liquid targets can be a purely liquid substance, such as molten metal or a solution of the substance to be irradiated in a suitable solvent, such as a molten salt. One advantage of liquid targets is that the target can also act as a coolant, thereby rendering a special coolant for the target unnecessary. A general disadvantage of liquid targets is that a rupture or leakage in the surrounding tank and pipe system can easily lead to the release of high-level radioactive substances in the facility.

Pure molten metals have the advantage of having a large number of atoms per unit of volume, which results in large numbers of irradiated atoms per unit of volume, a large N_0 per unit of volume in the equation for the reaction rate. The largest possible transmutation rate is thereby obtained for a given volume. Molten metals are also physically resistant to the effects of particle flux and emitted radiation since the absorbed energy is rapidly and fully converted into thermal energy. The degree of conversion through nuclear reactions which can be tolerated without necessitating the cleaning of the molten metal is largely determined by the solubility of the reaction products which are formed and by the handling possibilities bearing in mind the resulting radiation levels. One general advantage of molten metals is their high heat conductivity. The molten metal must be contained in a leak-tight system of tanks and pipes to prevent the release of radioactive substances. The construction material of the

system must therefore be able to withstand the particle fluxes, radiation levels, temperatures as well as the molten metal or reaction products in the target.

A solution containing the substance which is to be irradiated will lead to a lower number of irradiated target atoms per unit of volume and, thereby, to a lower reaction rate per unit of volume. A solvent, in the form of molecular liquid, such as water, can be used. However, this is less suitable than metals and molten salts due to the fact that the absorbed radiation energy will cause some decomposition of molecular liquids. In the case of water, hydrogen, oxygen, hydrogen peroxide free radicals etc. will be formed. When molten metals and ions are used, the absorbed radiation energy is mainly converted into heat. The advantage of a solution is that the number of atoms per unit of volume of the substance which is to undergo the intended nuclear reactions can easily be affected. In this way, the energy produced per unit of volume can be controlled, the concentration of reaction products can be kept at a reasonable level and the solubility of the products can be sufficiently high if a suitable solvent is used. Other properties such as melting point, viscosity and heat conductivity can also be affected through the choice of solvent and concentration. In this way, a relatively large degree of transmutation of the target atoms can be tolerated, in principle. The choice of solvent is also determined by its corrosion properties in relation to the material which can be used in the tanks and pipes in the system containing the target matrix.

A liquid target allows for the continuous purification and supply of new material to the system. This ensures that the target composition remains constant, thereby facilitating the dimensioning of the system from the standpoint of neutron physics. However, as is shown in Appendix 2, a continuous purification of the target leads to larger relative losses of non-reacted material than discontinuous purification.

Solid targets. When solid substances are used, the target must normally be divided into discrete units. The division is necessary in order to allow for cooling via a surrounding coolant. Since the units will contain large quantities of radioactive substances, they should be enclosed in a suitable material. The cladding is provided in order to prevent any interaction between the target and the surrounding coolant as well as to prevent the irradiated target and reaction products from being released to the coolant. As the composition of the solid target changes when reaction products are formed, form and volume changes occur. Such changes create a mechanical load on the cladding which limits the quantity which can be allowed to react. The cladding is also irradiated by the particles (usually neutrons) which are used to drive the nuclear reactions in the target. The composition of the cladding material thereby changes slowly at the same time as its mechanical properties change. This often leads to a minor increase in volume and considerable embrittlement. Finally, large concentrations of transmutation products can cause chemical reactions with the cladding.

The most common geometrical form of the encapsulated solid target is the long, thin cylinder. However, prisms and pellets also occur. The actual target must have a chemical form that is stable during irradiation, a high melting point and a reasonable heat conductivity. Furthermore, the chemical form of the target must be such that it does not cause an undesirable chemical or physical interaction with the cladding. Common chemical forms of encapsulated solid targets are oxides, nitrides and carbides. In older reactors, metallic fuel was also used. In the same way that liquid targets can be dissolved in an inert matrix, solid targets can consist of a mixture of the target and an

inert matrix. Through dilution using an inert material, the energy generated per unit of volume can be reduced and the heat conductivity can be changed. In turn, the cladding must tolerate a large dose of the particles in question and must remain inert to the surrounding coolant.

The target is removed from the system after a suitable degree of transmutation. In order to separate the reaction products from the non-reacted target material, the cladding must often be removed . The removal of the cladding can either be carried out chemically or mechanically. The latter is most common since mechanical removal normally results in less radioactive cladding waste. Relationships between the degree of transmutation during irradiation, the degree of chemical recovery of non-reacted material, losses etc., and the quantity of irradiated material which will become radioactive waste are provided in Appendix 2.

2.2.4 Technical system solutions

As described above, neutrons are the most attractive particles for the transmutation of atoms through particle irradiation. In general, there are five possible system solutions in addition to explosive systems. A short description of all of the alternatives is provided below.

Thermal reactors. Thermal nuclear reactors are critical, self-sustaining systems. This means that the production and consumption of neutrons balance each other exactly during operation with a constant energy release. The reactors are called thermal reactors because they have a low-energy neutron spectrum. This is achieved by slowing down the speed of high-energy neutrons from nuclear fission using a moderator. This results in a relatively soft neutron spectrum, that is, most of the neutrons have energies corresponding to the thermal equilibrium of their surroundings. For this reason, the reaction cross-sections of fissile actinide atoms are of the order of magnitude of 10^{-26}–10^{-25} m^2, see Table 2. This means that both fission rates and capture rates can be relatively large at a reasonable neutron flux. The reaction cross-section for fission of those of the fuel's atoms which have an odd number of neutrons is considerably greater in this case than the reaction cross-section for capture. The exposure of a target for a period of one year in a typical thermal reactor corresponds to a fluence of about 10^{25} neutrons/m^2.

Light water reactors have a more energy-rich neutron spectrum than corresponding heavy water or graphite-cooled reactors. Regardless of type, these reactors are most suitable for the transmutation of uranium and plutonium.

Modern reactor fuels often contain burnable absorbers in the form of gadolinium or erbium. A burnable absorber is a highly neutron-absorbing substance which is removed through transmutation during reactor operation and hereby compensates for the burnup of fissile material. In principle, it is possible to replace some or all of these substances by a suitable mixture of non-fissile actinides in an inert matrix, such as ^{237}Np and ^{241}Am in magnesium oxide. Such a measure would lead to a slow but significant transmutation of these atoms. Contrary to what was previously considered, transmutation of the quantities of neptunium and americium which are normally produced in the same

reactor is perceived as possible, even if the inventory of these elements will be quite high before production and destruction balance each other [And78, Cam77, Sow77].

Some of the practical and economical disadvantages of transmutation in thermal reactors are:

- Loss of reactivity which means that the interaction of the target with neptunium and americium must be compensated for by a higher reactor fuel enrichment which leads to higher fuel costs.

- A special forced cooling when the reactor is shut down for servicing will be required for those rods which contain actinides during transmutation.

- Neutron radiation from higher actinides will require extra neutron shielding in connection with the transport of fresh and spent fuel as well as in connection with refuelling.

Fast reactors. Fast reactors are self-sustaining critical systems which work directly with high-energy neutrons and, therefore, do not need a moderator to reduce the neutron energy.

Due to greater neutron speed, the neutron flux in a fast reactor is several orders of magnitude greater than in a thermal reactor with the same neutron density (number of n per unit of volume). This makes it possible to compensate for the fact that the cross-sections for the capture and fission of heavy nuclei with an odd number of neutrons are 100–1 000 times less than for slower neutrons.

In an ideal fast reactor, the neutron spectrum would be the same as for the neutrons which are released during fission. In a self-sustaining reactor, the neutrons are produced through nuclear reactions in the fissile atoms. A high neutron flux therefore requires that many neutrons are produced per unit of volume. This can be achieved through a large quantity of fissile material per unit of volume. Since a fast reactor therefore requires a very high neutron flux in a limited volume of fuel, the energy generated in individual fuel rods is very high and special measures are required to remove the heat. Cooling using a metal coolant is most often required, although the use of helium has also been discussed. However, a metallic coolant – most often liquid sodium or a molten lead-bismuth mixture has been used – slows down the neutrons to some extent. Sodium can slow down neutrons to an extent which corresponds to about 1% of that of graphite. However, the ability of lead to reduce the speed of neutrons is considerably lower. The reduced neutron energies mean that a sodium-cooled reactor has somewhat less adequate transmutation capabilities than an ideal or lead-cooled fast reactor. On the other hand, most sodium compounds are relatively or completely non-toxic whereas lead and lead compounds are toxic. It is possible to achieve annual doses of 10^{27}–10^{28} neutrons/m^2.

The advantages from the standpoint of transmutation with a fast reactor are that all actinide atoms are fissile with high-energy neutrons, regardless of whether they have an even or odd number of neutrons. Furthermore, the ratio between the cross-sections for fission and absorption are higher in a fast neutron spectrum. The number of released neutrons per fission is also higher in a fast spectrum. Altogether, about 0.2–0.4 extra

neutrons are obtained per actinide fission to transmute fission products, compared with a slow spectrum. The high neutron flux also results in a fast transmutation rate, even with relatively moderate reaction cross-sections.

Fast reactors could have a special function in a system with light water reactors which re-use uranium and plutonium from reprocessing in the form of MOX fuel. After a few cycles, the quality of the plutonium will be significantly degraded as light water reactor fuel since it will contain a higher concentration of non-thermal fissile plutonium isotopes with an even number of neutrons. If plutonium with such a composition is used as fuel in the core in a sodium-cooled fast reactor, the quality of the plutonium from the reprocessed core and blanket will be significantly greater because the concentration of plutonium isotopes with an even number of neutrons will have decreased considerably. This is due to the fact that all plutonium isotopes are split with comparable speed in the reactor core whereas fresh plutonium, with mainly an odd number of neutrons, is formed in the blanket. Recycled plutonium is therefore suitable for re-use as raw material for MOX fuel for thermal reactors. It is jokingly said that a fast reactor can act as a "washing machine" for plutonium.

Thermal accelerator-driven subcritical reactors . If a suitable accelerator is used to irradiate a thick target comprising a relatively heavy element with high-energy protons, large quantities of neutrons will be obtained, largely through spallation ("smashing up") of the atomic nuclei in the target. Typically, several tens of neutrons will be produced from each proton colliding with the target. This means that a reasonable beam of protons (for example 20 mA at 1 GeV of proton energy) can produce a large number of neutrons per unit of time. If the spallation target is placed in the center of a reactor core, the latter can act as a neutron multiplier even if it would not otherwise be self-sustaining. This is due to the fact that losses of neutrons can be compensated for through the supply of new neutrons from the spallation target. Through the fissions that occur in the core during neutron multiplication, more energy can be generated than is consumed to produce the proton beam. This therefore results in another type of self-sustaining system. The conversion of heat from the core into electricity in the conventional manner, via steam generators, turbines and generators, produces electrical energy which is more than sufficient to operate the accelerator. In turn, this produces the protons which, after conversion into neutrons in the target, sustain the production of energy in the core.

The new neutrons from both the target and the fuel in the reactor core originally have high energies. By introducing a moderator, the energy level can be reduced in the same way as in a thermal reactor. The advantage of this is that most reaction cross-sections are greater at low neutron energies than at high energies. Thus, less fissile material is needed for a given reaction rate at low neutron energies than at high neutron energies, that is for a given energy. In principle, considerably higher neutron fluxes can be achieved in this type of system than in a thermal self-sustaining reactor.

Water and graphite normally require encapsulated solid fuel and are therefore less suitable as moderators in accelerator-driven systems due to the large gradients in power density etc. Consequently, "thermal" molten salts, where actinides are dissolved in different types of fluoride salts have been considered to be a better combination of fuel and moderator. However, a slower reduction in the speed of the neutrons is obtained in these systems than in self-sustaining water-moderated reactors. Consequently, the cross-

sections for fission are somewhat lower than for systems where the neutron energies are reduced faster. Furthermore, the homogenization of the fuel means that a substantial neutron flux is obtained close to the target, with a high transmutation rate, while most of the core has a considerably lower neutron flux. This can not be compensated for by increasing the supply from the accelerator-driven target, since material damage on the accelerator window, above all on the wall between the target and core, will be unacceptable at high proton and neutron fluxes [Tak97].

Fast accelerator-driven subcritical reactor. Since the supply of neutrons from the target can be high, it is relatively much easier to design a suitable neutron multiplying core which is driven by high-energy neutrons from the target than if the reactor had to be self-sustaining.

In the same way as with a thermal, accelerator-driven system, the reaction heat can be used to produce electrical energy. Some of this heat is used up to drive the accelerator. In a fast accelerator-driven system, this share is typically on the order of 4–5%, and comparable to the energy which is used for secondary needs in a self-sustaining reactor.

Combinated system. A concept to combine the incineration of plutonium in MOX form in thermal critical reactors with fast critical reactors and accelerator-driven systems for the incineration of transuranic elements has been developed in Japan and further refined in France (see Figure 6). In this combined system ("Double Strata") existing reactors in the transmutation cycle are used in an efficient manner, the isotope composition of plutonium in the fast reactor is "improved" as described above, at the same time that the accumulation of neptunium, americium and curium in the fuel for the critical systems can be kept to a minimum. These are instead incinerated in a subcritical accelerator-driven system together with excess uranium. The plutonium formed is returned to the thermal reactors. The fuel forms are assumed to be oxides in the critical reactors, while nitrides and/or chlorides should be used in the subcritical system in order to provide the best possible neutron economy which, at the same time results in simultaneous transmutation of some fission products. Table 4 provides an estimate of the quantity of actinides at different stages of the combined fuel cycle.

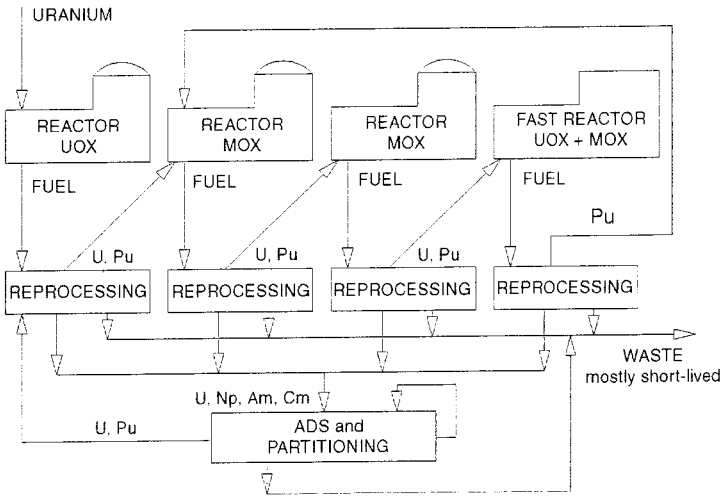

Figure 6. *Possible combination system with thermal, fast and accelerator-driven reactors.*

Table 4. Quantities of important actinides in a combination cycle.

PWR series 1 UOX:	10 PWR / fast reactor		
		In (kg/years)	Out (kg/years)
	Uranium	1.77×10^{5}	1.67×10^{5}
	Neptunium	0	95.4
	Plutonium	0	1.81×10^{3}
	Americium	0	54.7
	Curium	0	4.75
PWR series 2 MOX:	5.25 PWR per fast reactor		
		In (kg/years)	Out (kg/years)
	Uranium	8.67×10^{4}	8.47×10^{4}
	Neptunium	0	52.2
	Plutonium	4.56×10^{3}	2.18×10^{3}
	Americium	0	298
	Curium	0	65.4
PWR series 3 MOX:	2.5 PWR per fast reactor		
		In (kg/years)	Out (kg/years)
	Uranium	4.13×10^{4}	3.96×10^{4}
	Neptunium	0	39.8
	Plutonium	2.18×10^{3}	1.50×10^{3}
	Americium	0	269
	Curium	0	160
Fast breeder UOX+MOX:	1 FR		
		In (kg/years)	Out (kg/years)
	Uranium	1.62×10^{4}	1.45×10^{4}
	Neptunium	0	12.7
	Plutonium	1.50×10^{3}	1.97×10^{3}
	Americium	0	58.3
	Curium	0	8.07
Accelerator-driven fast reactor: 1 ADS (data for first batch of fuel)			
		In (kg/years)	Out (kg/years)
	Uranium	4.66×10^{3}	3.70×10^{3}
	Neptunium	200	78
	Plutonium	0	822
	Americium	680	196
	Curium	238	409

Explosive systems. Even if explosive systems can probably never be meaningfully used for transmutation, it should be emphasized that all of the actinide isotopes, even ^{231}Pa, have such cross-sections that suitably designed and sufficiently large masses can explode, in principle. For example, in the case of ^{237}Np, the unreflected critical mass is as low as 51.18 kg [Sri89]. The quantity of neutrons produced during a nuclear explosion is very large and can correspond to one hundred years of irradiation in a power reactor. At the same time, the neutron flux is extremely high near to the centre.

2.2.5 Degree of transmutation

The degree of transmutation achievable during irradiation depends on several different factors.

For liquid targets, this limit is dependent on how high concentrations of fission products can be accepted before the solubility of a particular product is exceeded. Corrosive reactions between the wall material in connecting tanks and pipe systems can also limit the permissible concentration of reaction products. Since the fission products comprise almost 70 different elements, extensive chemical investigations are required to ensure that precipitation does not occur due to exceeded solubility at a given concentration. For many liquids, often molten metals or salts, comprehensive solubility data for all elements concerned (and chemical compounds between these) at different temperatures are unavailable. Consequently, the upper boundary for the concentration of fission products is relatively uncertain. Corrosion and precipitation were observed, for example during the Molten Salt Breeder Reactor Experiment but could be satisfactorily controlled through different measures, such as cooling traps where some of the products that were formed were cooled out of the molten salt [Fer67].

In the case of solid targets, the limit depends on the volume changes that arise, gas pressure at particular working temperatures, chemical reactions between fission products and target atoms and, last but not least, possible corrosive reactions between the cladding and the target material containing fission products and neutron capture products. In general, the acceptable concentration of fission products appears to be considerably lower than for liquid targets. The dilution of target atoms with an inert matrix can, in principle, allow for a higher degree of transmutation than would be achieved without dilution.

The type of transmutation system is also important to the transmutation rate and the composition of the transmutation products. As was previously pointed out, this is due to the fact that the reaction cross-section varies with neutron energy and that the distribution of fission products among different atoms is also dependent on the neutron energy. Thus, it would be appropriate to discuss the neutron energy in thermal systems and the neutron energy in fast systems separately. In most cases, whether the neutrons are produced in a critical or subcritical system is not important. However, for fuel types which result in small negative temperature coefficients (such as nitrides of transuranic elements), it may be advantageous from the standpoint of safety to use subcritical accelerator-driven reactors.

Thermal systems. The advantage of thermal neutrons is that most of the reaction cross-sections are one or a few orders of magnitude higher than for high-energy neutrons. This applies to the capture cross-section in particular and to the fission cross section of heavy atoms with an odd number of neutrons, see Table 2.

Since the fission cross-section for heavy nuclei with an even number of neutrons is very small, in general, two neutrons are required to transmute these to fission products. The neutron economy then only in exceptional cases allows for the transmutation of long-lived fission products.

Fast systems. Normally, the neutron flux in fast breeder systems is over 100 times higher than in thermal systems. At the same time, the reaction cross-section is correspondingly lower for many atoms. The greatest difference exists for the actinides with an even number of neutrons. These actinides have fission cross-sections with fast neutrons which are of the same order of magnitude as the capture cross sections, see Table 3, in spite of the fact that corresponding cross-sections for thermal neutrons are small. This means that the irradiation of actinides by neutrons of higher energy generally leads to a greater share of fission and a smaller share of neutron capture. For example, for ^{237}Np, this leads to the fission of about 20% and the formation of^{238}Np by about 80%, while the corresponding figures for a thermal neutron flux are <1% and >99%.

2.2.6 Energy production

The release of energy during the fission of actinide atoms increases marginally within the actinide series and, for each element, it increases somewhat with the mass number. For simple estimates of the instantaneous released energy at fission, 200 MeV can be used for all split atoms without a particularly large error [Cho95]. This means that energy production will be practically proportional to the fission rate regardless of which atoms are split.

The rate at which thermal energy is produced, the power, is limited as in a conventional reactor, by the highest permissible temperature in the core. In turn, this is dependent upon the fuel form and construction material. In a configuration with solid fuel, melting must be avoided, which limits the maximum power density to about 200 MW/m^3 of heat for water-cooled thermal reactors. The maximum power density can normally not be used in the entire core, since the neutron flux decreases in the interface with the surrounding reflector. In a self-sustaining reactor, an attempt is made to compensate for this by utilizing burnable absorbers such as gadolinium and erbium in order to initially reduce the flux and power density in central parts of the core. Furthermore, different grades of enrichment are used in different parts of the core. In a self-sustaining transmutation reactor, it is possible to use heavy nuclei with an even number of neutrons as burnable absorbers. In a subcritical reactor, there are even greater problems with differences in power density and inefficient use of peripheral core parts if the fuel composition is homogeneous. In such a case, it is difficult to reach total outputs which are greater than 1 000 MW of thermal energy, regardless of the strength of the external neutron source. However, there is a better possibility of using the concentration of fissile material as a variable in the design of the core since a subcritical system is not dependent on delayed neutrons as a safety mechanism. By placing the fission products in the inner part of the core, and by gradually increasing the concentration of transuranic elements in the outer parts, total outputs can be achieved which are of the same order of magnitude as in light water reactors.

2.3 Reprocessing and partitioning

2.3.1 Partitioning

In order to transmute the long-lived radioactive nuclides in the waste, it is necessary to separate them from other nuclides. This is because the fission products in the waste will

otherwise absorb the neutrons which are necessary to "burn" the actinides and the transmutation process will be inefficient. A long and intensive neutron irradiation of fission products also leads to the formation of some undesirable long-lived radioactive nuclides. There are a number of partitioning methods which may be of interest for a future transmutation facility. A more detailed description of these methods is provided in a recent OECD/NEA report [NEA97].

2.3.2 Liquid-liquid extraction

Aqueous-based partitioning is the method which is most developed which means that there is considerable knowledge of both chemical and technical problems. Liquid-liquid extraction is a partitioning method which is based on two immiscible liquid phases, e.g. an aqueous phase and an organic phase [Ryd92]. In the organic phase, an extracting reagent with certain specific properties is dissolved and in the aqueous phase the different elements which are to be separated are dissolved. By contacting the two phases with each other under suitable conditions (such as pH, ligand concentration) and then allowing them to separate, selected elements can be enriched in the organic phase. This partitioning method has long been used in the reprocessing industry where uranium and plutonium have been recovered from spent nuclear fuel [Mus92]. Furthermore, in a future partitioning process in a transmutation facility it must be possible to separate the other actinides (mainly neptunium, americium and curium). In the traditional reprocessing (PUREX process), TBP is used (Tri Butyl Phosphate) to extract uranium and plutonium. TBP contains phosphorous which means that, at the end of the process, waste will be generated with a large concentration of phosphorous which cannot be completely burnt. Other phosphorous-based reagents, such as CMPO (Carbamoyl Methyl Phosphine Oxide) and DIDPA (Di-IsoDecyl Phosphoric Acid) can separate the heavy actinides in a future separation process. However, there will be a problem with secondary waste. One condition for achieving a very high degree of recovery in these processes is to minimize the quantity of secondary waste. The most interesting reagents will probably be based on carbon (C), hydrogen (H), oxygen (O) and nitrogen (N) alone. They follow the CHON principle (from the chemical symbols of the component elements) and are, thereby, completely incinerable [Mad98]. The possibility of replacing TBP by a suitable monoamide which fulfils the CHON principle has also been considered.

2.3.3 Pyrometallurgy

Pyrometallurgy is a general term which is usually used to designate chemical processes which occur at high temperatures. The term is often used to mean some form of partitioning of metals. Two different processes are currently of interest in the nuclear field; i) metallurgy with the addition of slag-forming agents and the partitioning of metal from slag, ii) two-phase extraction through the mixture of molten metal with a molten salt followed by the partitioning of the metal and salt.

During slag formation, partitioning is achieved by ensuring that the components in the molten metal have different propensities to react with the slag-forming agent. The slag product must be not easily soluble in the molten metal and must be of another density, possibly gaseous. This technique is used on a large industrial scale, such as in connection with steel and copper manufacturing, although it has a significant impact on

the environment. Previously, it was also used on a relatively large scale within the nuclear field, such as for oxidizing slagging for reprocessing fuel from the now decommissioned EBR-II fast breeder reactor [Hes63].

Separation through two-phase extraction is often based on the fact that the substances dissolved in the metal or salt phase have different solubilities and/or redox properties. By adding suitable amounts of reducing/oxidizing substances, such as metallic lithium or zirconium, to the metal phase or molten salt, substances present in the molten salt, such as ions, can be reduced to metal and pass from the molten salt to molten metal phase or the opposite. This technique has been used for the cleaning of weapons-grade plutonium from americium which is formed during the decay of the relatively short-lived ^{241}Pu which occurs as an undesirable compound in weapons-grade plutonium [Kni81].

Another possibility would be to use electrolytic reduction or oxidation to achieve a change in the distribution between molten metal and salt. This technique has a large-scale industrial use, such as in the production of aluminium through molten salt electrolysis. The reprocessing of nuclear fuel through electrolysis in molten salt with molten metal electrodes has been studied on the laboratory scale [Lai94, Lem97].

In both cases, substantial quantities of extra waste are generated, such as off-gas filters. Furthermore, the separated waste often contains an unacceptable quantity of fuel or target material and, therefore, further purification must be carried out, probably using aqueous-based methods [Kni81]. Another difficulty lies in designing equipment so that the processes are continuous instead of batch wise.

Pyrometallurgy may provide a simpler and less expensive means of reprocessing metallic fuel, and perhaps also oxide fuel [Kor96], since the transition from solid material to a solution does not have to be made prior to partitioning and the transition from solution to solid material does not have to be made after partitioning. Since there are no severe radiolysis-related effects nor neutron-moderated light elements, processes with high concentrations of fissile material can be achieved. This means that the facility would be smaller in size for a given annual production. However, these advantages are normally obtained at the cost of the degree of recovery since the quantity of material which is unintentionally lost with different types of waste is probably larger than for aqueous-based processes. Within industry, pyrometallurgical processes have in many cases been replaced by aqueous-based chemistry, such as in the production of copper and nickel from ore. Thus, a certain reduction in energy consumption is obtained, but above all, this has led to a substantial reduction in environmentally hazardous substances to the air and water.

2.4 Recycling and losses

The recovery and re-use of fissile material from spent nuclear fuel should be a "green" objective since this can lead to nuclear energy becoming a clean and highly sustainable source of energy with a relatively limited consumption of natural resources in the form of uranium ore.

Table 5. Estimated inventory of heavy elements in fuel stored at CLAB in 1997.

Element	Quantity (tonnes)
Uranium	2650
Neptunium	2.2
Plutonium	24
Americium	0.9
Curium	0.2

The quantity of spent nuclear fuel which is now being stored at the central interim storage facility for spent nuclear fuel (CLAB) corresponds to about 2 800 tonnes of uranium, counted as unused spent nuclear fuel. Taking into account the changes which occur during reactor operation, this corresponds to the quantities of different actinides shown in Table 5.

The recycling of the material which is now stored at CLAB in a hybrid system comprising light water reactors, fast breeder reactors and accelerator-driven subcritical reactors, in accordance with Figure 6, will result in an estimated electricity production in the range of 2 000–16 000 TWh. Where within the range the exact figure will lie depends on the number of PWRs per fast breeder reactor and on the number and size of the accelerator-driven facilities. The lower electricity production corresponds to a system with the material streams and facilities as described in Table 4. The maximum possible quantity of electricity corresponds to Sweden's current electricity production from nuclear power, about 80 TWh/year, for more than 200 years. By ensuring that the system contains reprocessing and partitioning facilities, fast reactors and accelerator-driven systems, the cost of electricity produced by such a system will, in all likelihood, be considerably higher than that of today's system which only consists of light water reactors and the planned final disposal of spent nuclear fuel.

As described in the examples presented in Appendix 2, a combination of the highest possible flux, the greatest possible fission cross-section, the longest possible irradiation time and batchwise partitioning with a high total chemical efficiency is required to achieve the smallest possible quantity of radiotoxic long-lived actinides in the waste which is to be disposed of. An optimization which only takes into account these factors will certainly lead to a relatively complex hybrid system where different types of elements are separated and transmuted in special facilities which will provide maximum destruction of the elements and minimum loss of radioactive waste. The recovered actinides and special fission products which will not be irradiated again in the facility where they were produced will be transported between these facilities. If other factors are also taken into account, i.e. economy and safety, the design of the system can change considerably. Unfortunately, no reliable data are available concerning the performance and cost of many of the components in possible hybrid systems since these only exist in the form of ideas, at the drawing board stage, on a laboratory scale or, in the best case, as prototypes. Consequently, it is very difficult or impossible to propose optimum systems taking into account all factors.

2.5 Safeguards

Like conventional reactors, accelerator-driven systems must be subjected to control in accordance with the Non-proliferation Treaty. However, certain new issues arise:

In principle, the reprocessing of fuel, which is a necessary stage of the transmutation process, makes it possible to gain access to weapons material in a relatively pure form. Even if thorium, uranium and plutonium are already present on the IAEA's and Euratom's inspection list, a review of procedures, to adapt the inspection procedures to the new systems, will probably be necessary. On the other hand, there is a somewhat greater possibility of designing fuel, targets or entire accelerator-driven systems so that they are more proliferation resistant than is the case with conventional reactors.

Accelerators and their possible use in transmutation systems also contribute to the need for more stringent control. Just like reactors, accelerators have, from the very beginning been used to produce or to try to produce weapons material. It was with the help of an accelerator that Glenn Seaborg produced plutonium for the first time. Even a medium-sized accelerator has the potential of producing a few kilograms of weapons material per year.

A safeguards concept for accelerator-driven systems can be based on the following elements:

(i) control (verification) of design (to avoid simple methods for process changes),
(ii) control of fuel design, components and their transport,
(iii) on site control of the inventory,
(iv) on site ad hoc inspection (as at reactor facilities, with photography or video technique, computers can improve the inspection),
(v) control of the accounting system for potential weapons material, as well as
(vi) monitoring of important operational parameters (accelerator power, core power etc., for accelerator systems and reprocessing facilities.

3 Swedish activities

3.1 P&T from a Swedish perspective

Extensive studies of partitioning and transmutation were carried out in several countries during the 1960's, 1970's and at the beginning of the 1980's. A broad knowledge of certain areas within the field was thereby acquired. In Sweden, research was conducted at the Department of Nuclear Chemistry at Chalmers University of Technology, financed by the National council for radioactive waste management (PRAV) which existed at that time. The research activities culminated when a newly developed partitioning process was successfully demonstrated on a high-level waste solution in co-operation with the Institutt for Energiteknik, Lillestrom, Norway. Interest in this activity declined as less work was focussed on breeder reactors and the interest in reprocessing and plutonium recovery declined in Sweden.

The development of accelerator technique meant that at the beginning of the 1990's, interest was awakened in investigating the possibility of using accelerator-driven systems for transmutation. The previously mentioned conference in Saltsjöbaden in the summer of 1991 was an important event for Sweden. In RD&D-Programme 92, SKB proposed that a deep repository for spent nuclear fuel should be constructed in stages. A first stage would comprise depositing a small quantity of spent nuclear fuel and would be followed by a careful evaluation of results from this stage and of parallel ongoing research and development on alternative methods in Sweden and abroad. In order to provide a basis for this future evaluation, SKB considered that it was important to support and, to a certain extent, initiate Swedish research work on alternative methods, especially the transmutation of long-lived radioactive nuclides. In the programme, areas of particular interest were considered to be studying the partitioning of long-lived nuclides, the technically feasible efficiency for transmutation, materials-related problems and the reliability and safety of the processes.

SKB therefore initiated co-operation with the Department of Nuclear Chemistry at Chalmers University of Technology (CTH) in Gothenburg and with the Center for Safety Research and the Department for Nuclear and Reactor Physics at the Royal Institute of Technology in Stockholm. Over the years, this co-operation has been expanded somewhat.

Within the government energy research programme, Studsvik has for a long-time been allocated the task of following international developments within new nuclear technologies.

3.2 Activities at CTH

Activities on the development of selective partitioning processes with high efficiencies started in 1974 at the Department of Nuclear Chemistry at Chalmers University of Technology (CTH), in Gothenburg, with financial support from the National council for radioactive waste management (PRAV) as well as from AB Atomenergi in the initial

stages. The project led to pilot-scale experiments during the early 1980's using 16 litres of concentrated high-level waste solution from the old Norwegian-Swedish reprocessing facility in Kjeller, Norway. The chemistry as well as the equipment were thereby verified. The process performed largely as expected and resulted in a very good level of partitioning at an extremely high efficiency, for example, total efficiencies were measured of >99.8% in the case of neptunium, >99.99% in the case of plutonium and >99.83 in the case of americium. The fact that these values are lower boundaries is mainly due to the difficulty of the measurement technique in detecting extremely small remaining concentrations of the particular elements in the cleaned radioactive waste. The cleaned high-level waste contained less than 1/100000 of original α activity after the liquid-liquid extraction-based process. Most of the results have been published [Lil80, Lil81:1, Lil81:2, Lil82, Lil84, Lil91, Per80, Per83, Per84:1, Per84:2, Sva79:1, Sva79:2, Sva80, Sva82, Sva84, Win84].

Since 1991, the Department of Nuclear Chemistry at Chalmers University of Technology has been conducting an SKB-financed project within the partitioning and transmutation field and, since May 1996, the Department has also been participating in the EU programme "Nuclear Fission Safety" within the NEWPART (New Partitioning Techniques) Project. Besides Sweden, Commisariat à l'Energie Atomique (CEA, France), University of Reading (England), European Transuranium Institute (EU), Forschungszentrum Karlsruhe (FzK, Germany) Forschungsanlage Jülich (Fa, Germany) and Ente per le Nuove tecnologie, l'Energia e l'Ambiente (ENEA, Italy) are participating in the NEWPART Project. The Department also has a more informal co-operation with Los Alamos National Laboratory (LANL, USA) and Japan Atomic Energy Research Institute (JAERI, Japan).

Activities at the Department comprise the development of new aqueous-based partitioning processes and, to reduce the volume of waste from future advanced partitioning processes, the extractants only contain carbon, hydrogen, oxygen and nitrogen (called the CHON principle after the chemical symbols for the component elements) which means that the reagents are completely incinerable and do not contribute to the secondary waste. Five different types of reagent are being studied, see Figure 7.

Aliquat-336 is a quaternary ammonium salt which acts as an anion exchange reagent. The reagent was proposed by LANL to be used in a partitioning process where Aliquat-336 is dissolved in an aromatic diluent, di-iso propylbenzene. In the first stage, Pu, Np, Tc and I are extracted from a 2 M nitric acid solution. By then lowering respectively raising the nitric acid concentration in the aqueous phase, Pu and Np respectively Tc and I can be returned to the water phase.

Malonamides are a group of reagents which are proposed for use in the DIAMEX process (France) to extract both trivalent actinides and lanthanides from the remainder of the fission products [Mad98]. In order to then separate the trivalent actinides from the lanthanides, it has been found that nitrogen-donor reagents can separate the two groups. Trivalent actinides and lanthanides are difficult to separate from each other since they have similar chemical properties, the same charge and ionic radius. The intention is to use nitrogen-donor reagents in the SANEX process for the partitioning of trivalent actinides from lanthanides recovered using the DIAMEX process.

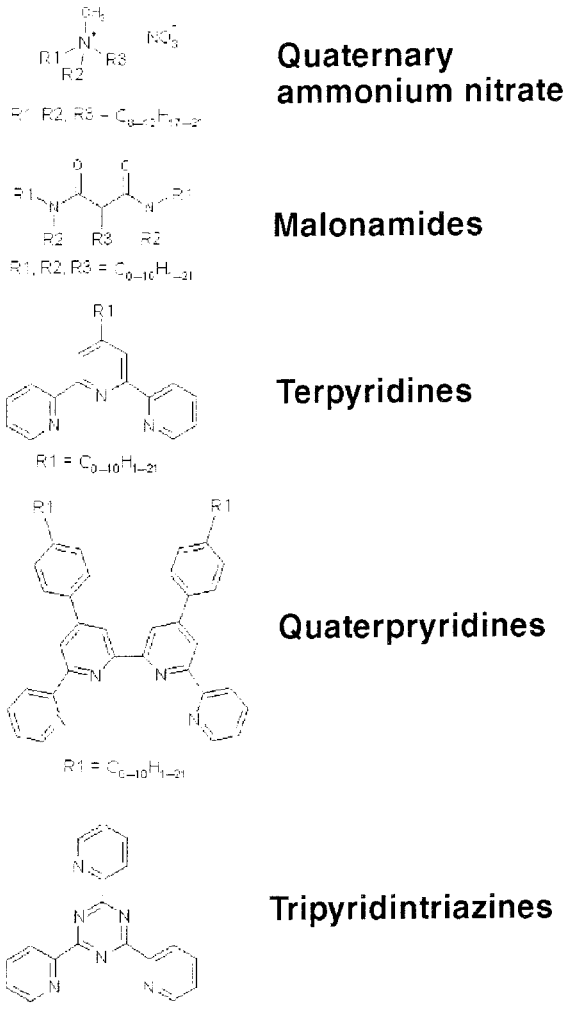

Figure 7. *Some CHON reagents which can be used for the partitioning of actinides and lanthanides.*

3.3 Activities at KTH

Research in the area of transmutation at the Royal Institute of Technology, Stockholm mainly focuses on accelerator-driven systems (ADS). This activity has grown and has developed considerably over the past two years. Five researchers are currently working in the Department of Nuclear and Reactor Physics on ADS, mainly financed by SKB.

Simulations of transmutation processes as a function of the neutron spectrum show that using liquid heavy metals as a moderator makes it possible to maintain constant transmutation rates for many atoms with large concentration variations [Möl97]. The low moderating capability of heavy metals also allows for the transmutation of the ^{99}Tc and ^{129}I fission products if these are placed in the reflector.

An analysis of transient neutron flux in connection to the spallation target has been carried out and has shown that the flux to essential parts is determined by transient components.

A Monte Carlo code for the calculation of the neutron flux and transmutation rates for a fixed fuel composition has been connected to a burnup code as a part of a project to create a complete tool for the calculation of accelerator-driven systems. Adjustments of cross-section databases and conversions for different core temperatures have been carried out. The results of an initial series of calculations have been published [Tuc97].

The simulation of the spallation process and thermohydraulics in a liquid lead/bismuth target has been carried out using the FLUKA [Car96, Tal97] and Flow3D [Car98] codes. The modelling of the thermohydraulics is being carried out in co-operation with the ISPRA research centre in Italy.

A study of accelerator behaviour in linear accelerators with the aim of creating a database to be used in probability-based safety analyses and reliability estimates has been initiated in co-operation with Los Alamos.

The Department has formulated, assigned and is conducting the certification process for ISTC's 559 Project. A liquid lead/bismuth target is being manufactured by IPPE (Institute of Physics and Power Engineering) at Obninsk, with the aim of irradiating the target at the Los Alamos accelerator for a total of six months in the year 2000-2001. KTH is one of the managers of the project and the results will be essential to any future work on designing a demonstration facility for ADS.

A member of the Department has been the editor-in-chief for the IAEA's status report on accelerator-driven systems [Gud97:2] and the Department is co-ordinating the EU's IABAT Project (Impact of Accelerator-Based Technologies on nuclear fission safety).

3.4 Other activities in Sweden

3.4.1 University of Uppsala

Accelerator-driven systems include the production of neutrons with energies up to the GeV range, which is much higher than the neutron energies which occur in self-sustaining reactors. In order to be able to simulate and optimize the target and core of such a system, knowledge of neutron-induced nuclear reactions for these energy regions is required. At high energies, a considerably larger quantity of reaction channels are open. The experiments which are on the OECD's list of high priority measurements will therefore take decades to carry out using existing resources. The cross-section for elastic scattering, which is about half of the total cross-section, is of particular importance. Detailed data for differential elastic cross-sections in a limited number of nuclei can be used to determine parameters in optical model potentials at relevant energies. With a good optical model, elastic, total and reactive cross-sections can be predicted.

At Uppsala University's Department of Neutron Physics, a project was started in 1997 with the aim of measuring differential elastic cross-sections at 100 MeV. Measurements will be conducted at the unique quasimonoenergetic neutron beam facility at The Svedberg Laboratory in Uppsala. A prototype of SCANDAL (SCAttered Neutron Detection AssembLy) has been constructed and installed in connection to the neutron beam. The rig consists of two identical arms, with scattering angles of between 10 and 50 degrees as well as between 30 and 70 degrees. Each arm consists of a thin veto

scintillator for rapid discrimination of charged particles, a plastic scintillator for triggering, two operating chambers for proton followup and a series of cesium-iodine detectors for determining energy. The veto scintillator mainly reacts to charged particles and the signal from this can therefore be used to block other signals which occur when a charged particle comes from outside. The equipment is therefore insensitive to incoming charged particles and mainly reacts to phenomena which are triggered by incoming neutrons. The rig is designed for an energy of about 2.5 MeV, which is sufficient to isolate the ground state from the initial excited state in most of the nuclei of interest, i.e.

^{12}C (4.4 MeV), ^{16}O (6.1 MeV), ^{40}Ca (3.3 MeV), ^{90}Zr (1.8 MeV), and ^{208}Pb (2.6 MeV).

Support from the Swedish Nuclear Power Inspectorate, the nuclear power plants and SKB means that it is now possible to add new equipment and to employ PhD students. Full-scale data collection will be carried out during 1998.

Experiments to establish the distribution of fission products during fast fission of ^{232}Th and ^{233}U as well as the expansion of transport codes and cross-section data libraries to the energy range of 20-150 MeV are being carried out at the OSIRIS facility at Studsvik.

At the newly founded Centre for Astronomy and Physics in Uppsala (CAP) an industry-related research school (AIM) will also be started during 1998. A couple of PhD degree projects within the transmutation area should be performed at this research school.

3.4.2 Studsvik

For a long time, Studsvik AB was given the task by the Swedish Government of following and analyzing the international development of new nuclear technology and reporting on the status on an annual basis. As a part of this work, developments in partitioning and transmutation technology were also followed. However, Studsvik did not conduct any of its own research in the area. This activity has recently been terminated.

4 Foreign P&T programmes in progress

Since the end of the 1980's, research on the transmutation of long-lived and biologically hazardous radioactive nuclides has increased all over the world. In particular, interest in accelerator-driven systems for the transmutation of nuclear waste and energy production has increased over the past two years. In France and Japan where, for a decade, active research programmes on transmutation have existed, new political problems concerning fast breeder reactors have stimulated greater interest in accelerator-driven systems as a solution to the waste management problem and in both countries, about 100 researchers are currently involved in tasks relating to the partitioning and transmutation of nuclear waste. In Russia, there are currently about 500 former weapons researchers involved in transmutation research, financed by the USA, the EU and Sweden through ISTC. The USA has a military programme for the development of high intensity accelerators which is only classified to a minor extent. Consequently, it will be possible to use results from this programme in the evaluation and possible design of an accelerator for a subcritical system.

Countries such as Italy and Spain have recently started research programmes while other countries have allocated additional funds to existing programmes (Belgium, India, Sweden, South Korea, Czech Republic).

While research was previously characterized by efforts to establish system parameters, with sometimes diametrically opposed views being held by different factions, systematic studies in recent years have led to a more uniform view with respect to several important points, including:

- Transmutation of nuclear waste cannot replace geological disposal but could be a complement which will lead to a considerable reduction in the quantity of long-lived substances which must be deposited in such a repository. Estimates of possible reduction factors vary between 90 and 99 per cent of the original waste volume.

- Proton targets in accelerator-driven systems should consist of a liquid metal; molten lead/bismuth, or lead. In high power facilities the use of wolfram or actinides as a target would lead to problems related to contamination as well as cooling.

- The neutron spectrum should be fast, in order to achieve good neutron economy as well as the best possible transmutation of actinides and individual fission products.

- Liquid metal (lead/bismuth or lead) should therefore be used as a coolant. Sodium is still part of the picture but is considered to be less attractive after the accident at the Monju reactor in Japan and the decision to close down the large multinational fast breeder reactor, Super Phenix, in France.

- Research concerning pyrometallurgical methods for partitioning should be conducted to determine whether known problems involving secondary waste streams can be overcome. Pyrometallurgical methods may have certain cost-related

advantages and may allow for partitioning on site without any need for fissile material to be transported elsewhere.

Transmutation experiments are carried out at CERN, in France, Russia and other countries. Accelerator components are being developed in the USA, Japan and at CERN. Simulation models have been improved. E.g. the EET group at CERN has developed a complete code which is specially optimized for accelerator-driven systems. New cross-section data for nuclear reactions have emerged, especially in the energy range of 20-150 MeV.

4.1 Belgium

In Belgium [IAE97, Gud97:2] research and development work within partitioning and transmutation is carried out by Belgonucléaire. Knowledge exists within areas such as techniques for the partitioning of actinides and fission products, radioanalytical procedures for measurements of spent nuclear fuel as well as actinide production. In the future, the intention is to separate and transmute ^{237}Np, ^{241}Am, ^{243}Am and curium as well as some of the long-lived fission products ^{129}I and ^{99}Tc. The partitioning of ^{237}Np can be achieved through a minor modification of the existing PUREX process, whereas the partitioning of Am and Cm from the lanthanides (Ln) is considered to be one of the most important problems to be resolved. However, the main thrust is still that a deep geological repository will be designed for most of the waste produced in Belgium. Since 1972, investigations concerning such a repository have been in progress i.a. in an underground laboratory (SCK-CEN). Furthermore, the Myrrha Project, focussing on the production of isotopes for medical purposes, has been further developed. Research groups at SCK-CEN and IBA (Ion Beam Applications) have shown that two small cyclotrons of a type that is already being produced on a commercial basis by IBA (150 MeV, 2mA) applied in an accelerator-driven system should be sufficient to cover the global need for ^{99m}Mo, for example. The cost of the system, based on a target of liquid lead/bismuth as well as a blanket of enriched uranium plate, would probably be lower than the cost of special reactors for the same purpose.

4.2 France

Within the French nuclear programme, efforts were made at an early stage to recover plutonium from spent nuclear fuel using aqueous-based partitioning methods [Skå95]. MOX fuel is then fabricated from the recovered plutonium while recovered uranium is converted to uranium hexafluoride and re-enriched. The reprocessing rate is planned to correspond to the recycling of plutonium as MOX fuel. In this way, the accumulation of large quantities of unused plutonium can be avoided. It is still expected that future advanced partitioning methods will be based on aqueous-based systems which can be integrated with current partitioning processes.

An Act has entered into force in France, as of December 1991, which defines the political requirements regarding research within nuclear waste up to the year 2006. The alternatives which should primarily be studied are (i) partitioning in combination with transmutation, (ii) partitioning in combination with element-adapted waste forms (iii) safe geological final disposal of vitrified high-level waste and (iv) safe direct geological

final disposal of spent nuclear fuel. Therefore, the CEA has launched long-term research programmes in order to attain the level of knowledge specified in the Act. The programme which deals with the first alternative is called SPIN (short for Separation et Incineration). The research programme for partitioning technology is currently divided into two sub-programmes; PURETEX, which aims at improving the existing PUREX process by reducing the volume of process waste to one-third at the same that neptunium is also recovered quantitatively, as well as ACTINEX, which deals with the more long-term waste problems with heavier actinides and long-lived fission products and where the possibility of recovering these atoms from the high-level waste stream is being investigated. The intention is then to, in the first instance, transmute these atoms using conventional nuclear reactors or subcritical accelerator-based systems, and in the second instance, to develop and produce improved chemical end products for storage. The aim is to reduce the volume and the radiotoxicity of the waste in a deep geological repository [IAE97].

Experiments in the PHENIX reactor (the SUPERFACT-1 experiment) have shown that U-Pu-dioxide and uranium dioxide can be used to transmute the heavier actinides (Np and Am). The results from the SUPERFACT-1 experiment are provided in Table 6. A second experiment (SUPERFACT-2) is planned for the PHENIX reactor where a higher burnup is used than in SUPERFACT-1. Irradiation of Tc in the PHENIX reactor is also planned. Basic irradiation studies have also been planned for the OSIRIS thermal reactor (the ACTINEAU experiment).

Table 6. Results from the SUPERFACT-1 experiment.

Fuel composition	Burnup (atom%)	Effective half-life Nuclide	(years)
74% U, 24% Pu, 2% Np	6.4	^{237}Np	2.05
60% U, 20% Am, 20% Np	4.1	^{237}Np	1.8
		^{241}Am	2.2
65% U, 45% Np	4.5	^{237}Np	2.2
74% U, 24% Pu, 2% Am	6.4	^{241}Am	2.2

The realization of accelerator-driven systems requires research and development within several areas which, in France, come under the GEDEON programme where the research organizations, CEA and CNRS, as well as the nuclear industry (EDF), are participating [Sal97, Gud97:2].

Within the CEA's ISAAC programme, reactor physics-related phenomena in subcritical accelerator-driven systems are being investigated. In the MASURCA facility (CEA/Cadarache), experiments have been carried out using source-driven subcritical reactor cores during the period of 1995-1997 (MUSE 1, 2 and 3). A series of measurements have thereby been conducted to verify models and system parameters. In December 1997, CEA/Cadarache's proposed prototype for an accelerator-driven system (HADRON) was given the same status as a national programme.

4.3 India

The Indian nuclear programme [IAE97] comprises three parts, of which the first step is to construct a number of heavy water reactors (PHWR). The plutonium which is produced in these reactors is recovered and used in a second stage in fast breeder reactors to produce additional plutonium fuel from uranium and/or ^{233}U fuel from thorium. The third stage which is in progress in parallel with the second consists of reactors which are operated on fuel with ^{233}U. India has developed techniques that covers the entire cycle from uranium mining to reprocessing. Research and development for the improvement of existing reprocessing processes is in progress, which includes actinide partitioning and the partitioning of long-lived fission products. Improvements of the PUREX process to reduce the plutonium losses to the waste as well as to recover ^{237}Np and possibly ^{99}Tc and ^{129}I are being considered. The TRUEX process with the extractant CMPO (Carbamoyl Methyl Phosphine Oxide) is investigated as a possible method of separating actinides (Np, Am and Cm) from high-level PUREX waste. The research is being conducted at the Bhabha Atomic Research Centre.

4.4 Italy

Italian research centres are participating in the testing of the DIAMEX process within the framework of an ongoing EU project (NEWPART). There is no commercial nuclear power in Italy at present. However, radioactive waste exists from previous nuclear programmes in the form of spent nuclear fuel and in the form of high-level waste from small-scale reprocessing. In order to reinforce the competitive strength of Italian research and industry within the area, the Ministry of Science and Technology has granted financial support to a research project on partitioning and accelerator-driven systems.

4.5 Japan

In October 1988, the AEC (Atomic Energy Commission), which is responsible for the use and development of nuclear power in Japan, started the OMEGA programme. OMEGA stands for "Options for Making Extra Gain of Actinides and fission products generated in the nuclear fuel cycle" and includes research on both partitioning and transmutation. JAERI (Japan Atomic Energy Research Institute), PNC (Power Reactor and Nuclear Fuel Development Corporation) and CRIEPI (Central Research Institute of Electric Power Industry) are the three largest research institutes in Japan which are involved in the OMEGA programme. The first phase (1988-1996) concentrated on basic studies and investigations into the possibility of developing partitioning and transmutation technology [Skå95].

The focus of the research varies from institute to institute. JAERI has proposed a two-way nuclear cycle where a conventional nuclear fuel cycle with uranium and plutonium recovery is combined with a partitioning and transmutation cycle for actinides. There is also a proposal concerning a partitioning process where a number of actinides are recovered and these are recycled as fresh fuel for light water reactors. PNC's proposed partitioning process is based on an improved PUREX process with a subsequent TRUEX-like process where the remaining actinides are separated. The intention is then

to incinerate the actinides together with MOX fuel in a fast breeder reactor (FBR). On the other hand, CRIEPI is investigating the possibility of using pyrometallurgical partitioning methods to recover the actinides and then recycle them as metallic fuel in an FBR.

In 1995, a reorganization was carried out and the OMEGA programme was integrated with a national neutron research programme, where the development of a supraconducting high-intensity accelerator with a proton energy of 1.5 GeV and a 5.3 mA current is in progress. The accelerator is expected to be taken into operation in around the year 2005, and to be supplemented by an experimental transmutation facility after the year 2008.

Since the Japanese fast reactor programme has been shelved as of 1998, it is possible that the focus of research may shift towards accelerator-driven systems in the next few years.

4.6 China

High-level waste from Chinese reprocessing plants is currently being stored in tanks pending the construction of a vitrification facility some time during the period of 1998-2000 [IAE97]. Research is being conducted on how to recover the actinides and valuable metals prior to final disposal. One of the world's largest neutron data libraries, CENDL-2, was released by CNDC (Chinese Nuclear Data Center) in 1992 and CNDC has also developed an even larger library, CLIB-2, which is based on CENDL-2 but is more suitable for transmutation calculations. With data from these libraries, calculations have been performed to investigate the possibility of the transmutation of heavier actinides in the CEFR (China Experimental Fast Reactor) and in fusion-fission-hybrid reactors. For actinide partitioning, three different partitioning processes have been studied, the reverse TALSPEAK process [Ryd92, Sva79:2, Sva82, Sva84], the DHDECMP process [Sch73, Hug90, Ron93] and the TRPO process [Cho92]. Chinese researchers have shown that highly pure Cyanex 301 has a large capacity to selectively extract the trivalent actinides Am and Cm from a mixture of lanthanides [Jin96, Zhu96]. This reagent is being tested in co-operation with German researchers at the Forschungsanlage Jülich within the NEWPART EU project.

4.7 The Netherlands

Since 1991, the ECN in Petten has had a transmutation research programme which is referred to as RAS. The aim is to contribute to international research on transmutation and fuel recycling and to show whether it is possible to introduce such a process in an acceptable manner in the waste management system. After an initial review of the project, the Government has approved a continuation (1994–1997) of the project which will aim at evaluating the technical possibilities and risks of partitioning and transmutation and at proposing a possible strategy for waste management.

4.8 The Russian Federation

The political aspects of nuclear waste in the Russian Federation are complicated by the after effects of the Chernobyl accident, the fact that large areas of land are contaminated with radioactive nuclides from military nuclear activity, the border problem due to the division of the Soviet Union into several states, safety and proliferation problems in connection with nuclear disarmament as well as major economic problems. In spite of this, a R&D programme is in progress which covers many different areas and with the involvement of many different research institutes. The main line of the Russian nuclear power programme is the same as before the Chernobyl accident, i.e. thermal reactors operated using uranium fuel and sodium-cooled fast reactors using recycled fuel. In the Russian Federation, there are currently four different types of material containing actinides which are temporarily being stored: (i) Leftover reactor waste with low burnup from which U and Pu have been separated (^{237}Np is the main actinide component in the waste). (ii) A complex mixture of heavy actinides which have remained after the reprocessing of spent nuclear fuel from commercial nuclear reactors (half of the mass comprises ^{237}Np and the rest of Am and Cm). (iii) Spent fuel elements, i.e. the waste product from an open nuclear fuel cycle. (iv) Weapons-grade plutonium in the form of dismantled warheads. Actinides are considered to be suitable for transmutation due to the fact that they are highly radiotoxic and that they are fissile with neutrons and therefore can be used as components in nuclear fuel. Two different concepts for actinide transmutation are being considered, namely, accelerator-driven subcritical systems and fast reactors cooled with liquid metal (Pb, Bi).

Since 1994, several Russian research institutes have been involved in research on accelerator-driven systems as a possible means of eliminating weapons-grade plutonium and nuclear waste. The studies have comprised the realization of a 100 MW linear proton accelerator to drive a specially designed subcritical transmutation core as well as reprocessing processes which can be applied in an integrated transmutation facility. The work has largely been financed by ISTC (International Centre of Science and Technology). The transmutation facility which was studied comprised a target of liquid lead/bismuth as well as a subcritical core with two zones. Important results which have emerged include the recommendation of using lighter material such as titanium and graphite as window material for the proton beam as well as the conclusion that problems with the accumulation of ^{210}Po in lead/bismuth-cooled systems are less important than previously believed.

In Russia, there is a unique experience (a total of about 70 operating years) of lead/bismuth as a coolant for reactors in submarines. Within the ISTC's project #559 which was financed and granted permission to start in 1996, a liquid lead/bismuth target will be designed in Obninsk to be irradiated in Los Alamos prior to the end of 1999. This may be an experiment which will be decisive for the development of a demonstration facility. IPPE (Obninsk), LANL, CEA/Cadarache and KTH are participating in the experiment.

4.9 Spain

Spanish universities and Spanish industry have recently (1996-1997) launched a research programme concerning accelerator-driven systems in co-operation with CERN.

The co-operation is in the form of a company founded through the LAESA consortium, whose first task will be to convince the local population and the Spanish Government of the need to construct a prototype of CERN's energy amplifier on Spanish soil in Zaragosa. However, the current level of funding is low.

4.10 South Korea

Since 1992, a long-term research project has been in progress at KAERI (Korean Atomic Energy Research Institute) with the aim of developing a method for reducing the radiotoxicity of high-level waste [IAE97]. This programme comprises the evaluation of data, the study of the possibility of transmuting heavy actinides in PWRs, the development of codes for the calculation of transmutation rates and the design of transmutation systems. Conventional reactors, fast reactors and hybrid systems consisting of a subcritical reactor and an accelerator are being studied. In 1997, the programme was reviewed and a decision was made that research on accelerator-driven systems will be one of KAERI's main areas of work up to the year 2007 (for the period of 1997–2000, the allocated funds correspond to SEK 30 million per year). Within partitioning, a four-group process is currently being investigated where heavy actinides and lanthanides are precipitated with oxalic acid. They are then separated by extraction with HDEHP and the heavy actinides further purified.

4.11 Czech Republic

In the Czech Republic, a research project is in progress within partitioning and transmutation where the first step has been to establish a national centre for the documentation of research within the area [Hro97]. The Czech Republic has considerable experience of molten salt and pyrometallurgical methods and therefore accelerator driven-systems can be considered to be a possible solution to the problem of nuclear waste management in a densely populated country. The research institutes and the industry (SKODA) have formed a consortium to co-operate on the development of techniques for molten salts and are planning a complex experiment called LA-0 to test a number of parameters in a fluoride-based subcritical reactor and related chemical processes. Furthermore, the Czech Republic will arrange a conference on accelerator-driven transmutation technology and applications in 1999.

4.12 USA

Several different strategies for the transmutation of nuclear waste have been presented in the USA. The purpose of R&D within the area has not been clearly defined. However, three targets have been specified, namely (i) the development of a system that is secure from the standpoint of nonproliferation, (ii) fast destruction of weapons-grade plutonium and (iii) an energy system without long-lived radioactive waste. Some projects focus on accelerator-driven systems and one of these (APT) is investigating the possibility of producing tritium for nuclear weapons and other applications. In 1995, the National Research Council of the National Academy of Science have presented a report on partitioning and transmutation in the USA and presented recommendations for future research in the area [Ras96]. Six different partitioning and transmutation concepts were

discussed based on light water reactors (LWR), advanced liquid metal reactors (ALMR), accelerator-driven subcritical reactors (ATW), pepple-bed reactors as well as a mixture of fast and thermal reactors (CURE). The report presents the conclusion that partitioning and transmutation are not adequate reasons to delay the development of the first repository for spent nuclear fuel. However, the deposited waste in the repository should be accessible over the next 100 years since partitioning and transmutation systems may be improved during this time. Furthermore, the opinion is that it may be possible to use three of the evaluated concepts for the transmutation of transuranic elements. Finally, it is established that, over the next ten-year period, the USA will conduct further research and development within the area, with an emphasis on improved partitioning processes for the reprocessing of light water reactor and transmutation fuels, i.e. the partitioning of additional actinides and certain selected fission products with greater efficiencies.

During 1996 and 1997, research on P&T was actively conducted mainly in Los Alamos where experiments with pyrometallurgical methods and lead/bismuth techniques are being set up [Ven98]. Within the framework of the APT project (Accelerator-driven Production of Tritium), a high-power accelerator is being developed. However, it does not seem very likely that this accelerator will be constructed.

Besides the APT project, there is a lack of funding for research on accelerator-driven systems. However, in January 1998, an independent evaluation of LANL's ADS project was conducted to provide a basis for a proposal to the senate concerning funds of about SEK 175 million per year.

4.13 Belorussia

In Belorussia, an interesting experiment is currently being conducted at the Institute of Radiation Physics and Chemistry in Sosny. A powerful 14 MeV DT neutron generator has been built and is being used to measure transmutation rates for fission products and actinides in subcritical cores with different neutron spectra. This experiment will be significant in the validation of code systems and cross-section databases.

4.14 European Union (EU)

During 1997, the European Union's Science and Technology Committee (STC) presented its opinion to the European Commission on an energy amplifier system for electricity generation [EU97:1]. STC considers that it is not realistic to immediately develop an overall system since new technique must be developed in several areas, such as new reactor systems, new accelerators, new fuels, new fuel fabrication, new reprocessing systems and new waste management methods. It would be very expensive to develop an entire system. This development cost is estimated at several tens of billion ECU. An energy amplification system would, therefore, not be sufficiently competitive, in economic terms, compared with the new improved light water reactors that are being developed, such as the European pressurized water reactor. STC therefore recommends the European Commission to focus on the development and evaluation of thorium-based fuels as well as, in addition, to continue to support research on subcritical, fast neutron multipliers intended for actinide incineration. Since 1990, the European Union

has contributed research funds to different partitioning and transmutation projects (Radioactive waste 1990–1994), including funds to the CEA (France), which has investigated the possibilities and the costs of P&T of long-lived waste. Siemens (Germany) has analyzed the use and safety of reactors intended for transmutation and the ECN (the Netherlands) has developed a database for transmutation and data collection has been initiated. Belgonucléaire (Belgium) and AEA Technology (England) have also received research grants. The CERN experiment TARC (Transmutation through Adiabatic Resonance Crossing) is also funded by the EU. In parallel with these programmes, research on transmutation in fast reactors has been carried out at the Transuranium Institute (TUI) in Karlsruhe, which is an EU laboratory. At TUI, the partitioning of some actinides using different extraction reagents (CMPO, TRPO, DIDPA) has been studied. The new EU programme "Nuclear Fission Safety 1994–1998" has a total budget of 170.5 million ECU and includes an P&T programme. Sweden is participating in two of the projects in the programme [EU97:2].

The research project "Impact of Accelerator Based Technologies on nuclear fission safety" (IABAT) is being funded by the European Union and was started in 1996 with 11 participating institutes/universities from six countries. The project is being co-ordinated by KTH and is divided into four parts:

- System studies of accelerator-driven hybrid systems.
- Evaluation of accelerator technology as well as the cost of circular accelerators.
- Basic cross-section and materials data.
- Studies of fuel cycles for accelerator-driven systems.

During the first year, different types of systems have been analyzed from the standpoint of fuel cycle, neutron physics and toxicity, through the development of a number of code systems. Comparisons with transmutation in light water reactors have been made. Experiments for the determination of the distribution of fission products during fast fission of ^{232}Th and ^{233}U have been carried out at the OSIRIS facility in Sweden. Work on expanding transport codes and cross-section data libraries to the energy range of 20–150 MeV has started.

4.15 IAEA, OECD/NEA and CERN

4.15.1 IAEA

The IAEA has arranged a number of conferences on P&T and is focusing on compiling work from countries conducting research within the area. The "Status report on actinide and fission product transmutation studies" (IAEA-TECDOC-948), published in June 1997, is one such compilation. The IAEA supports research on accelerator-driven systems within a general programme for nuclear systems for energy production and transmutation. The aim is to create a global forum for discussion and technical information on development within the area. In recent years, the following projects have been carried out.

- The preparation and publication of a status report on accelerator-driven systems. The report presents an overview of ongoing research activities all over the world, trends in development as well as an evaluation of the potential of accelerator-driven

systems applied to energy production, the incineration of weapons-grade plutonium and the transmutation of radioactive waste.

- The preparation and publication of a status report concerning thorium-based fuel cycles. The report presents an overview of experience and new research as well as highlights areas where greater knowledge is required.

- A co-ordinated research programme concerning thorium-based fuel cycles in accelerator-driven systems where about ten research groups from all over the world are participating in a comparative study of calculation methods for neutron physics, burnup and toxicity, with the aim of obtaining well-defined evaluation methods for simulation codes for accelerator-driven systems.

4.15.2 OECD/NEA

In 1988, the OECD's Nuclear Energy Agency (NEA) started a programme called "International Information Exchange Programme" which deals with physical and chemical properties of elements in the nuclear fuel cycle, partitioning and transmutation. The NEA's standpoint on the issue can be summarized in four points: (i) P&T of actinides has the potential to contribute to resolving the problem of the radioactive waste by reducing the quantity of long-lived radioactive atoms. However, P&T should not be considered as an alternative to deep geological disposal and should not be presented as such. (ii) The value of P&T must be verified by the evaluation of strategy, science, technology, cost and safety. (iii) P&T is an area which is suitable for international co-operation where the NEA should participate within various areas such as waste management, scientific and strategic studies, technology and safety. (iv) In order to achieve an efficient co-ordination of work, an efficient exchange of information between the different NEA committees is required.

4.15.3 CERN

Since 1995, Rubbias' EET team (Emerging Energy Technology) has worked on an accelerator- driven system with liquid lead as a target and coolant as well as a thorium-based fuel cycle. The aim has gradually changed from "safe" energy production with a "pure" $^{232}Th/^{233}U$ cycle, to the transmutation of higher actinides with the thorium cycle as a basis. Experimental studies in co-operation with French CRNS and southern European universities have improved knowledge of the spallation process and neutron physics in a light water cooled ^{238}U target (FEAT – First Energy Amplifier Test) and solid lead target (TARC – Transmutation through Adiabatic Resonance Crossing). CERN has also initiated the founding of the LAESA consortium (Laboratorio del Amplificador de Energia), see 4.9 above.

Significant work has been carried out to develop the EET's Monte Carlo code for the simulation of an accelerator-driven system. The entire chain of events from proton-induced spallation and high-energy cascade to the change in the composition in time during burnup is described in a well-integrated code package.

CERN is also working on accelerator design and has emphasized the advantages of using a high-intensity cyclotron as a proton source. The concept mainly appears to be of

interest for the design of a compact prototype, where proton energies of 400–800 MeV and currents of 2–5 mA should be sufficient. For the effects which are estimated to be necessary in full-scale facilities (> 10 MW), either several cyclotron complexes or linear accelerators are necessary.

5 Conclusions from a Swedish perspective

The possibility of carrying out partitioning and transmutation, as a stage in the management of spent nuclear fuel and high-level waste, can nowadays largely be considered to be scientifically proven. However, it is still too early to determine whether this type of waste treatment has cost- or safety-related advantages in the short term, compared with current plans for the management of spent nuclear fuel and high-level waste. Neutrons from both thermal reactors and fast reactors as well as accelerator-driven subcritical reactors can be used for the transmutation of long-lived radioactive nuclides. It should be emphasized that thermal reactors and fast reactors have been constructed and operated and adequate experience has been obtained over a long period of time, while accelerator-driven reactors are only at the drawing board stage. Each of these neutron sources has advantages and disadvantages for transmutation and they are probably each best suited for certain types of nuclides. Current consensus with respect to comparisons is that transmutation in reactor types with high-energy neutrons is advantageous with respect to the reduction of the total quantity of higher actinides coupled with a relatively efficient energy production, while reactors with low-energy neutrons are primarily advantageous if the aim is to reduce the quantity of certain radiotoxic atoms and generate low-cost energy. One possible development is to use hybrid systems where each type of neutron source has a determined function. In such systems, the special properties of an accelerator-driven system are used to achieve the efficient transmutation of neptunium, americium as well as possibly curium and other types of atoms with small fission cross-sections. In such cases, the accelerator-driven system serves to supplement more conventional nuclear reactors rather than to replace them.

What is characteristic for all transmutation processes is that they must work together with a suitable chemical partitioning process where non-transmuted material is recovered from irradiated material with a high degree of efficiency. The rest becomes radioactive waste. When the effect of a combined partitioning and transmutation process is evaluated, there is a strong link between the degree of recovery in the partitioning process, the efficiency of the transmutation process and the losses of non-transmuted material to various waste streams. Both aqueous liquid-liquid extraction and various pyrochemical partitioning methods have been proposed for the partitioning process. There is considerable knowledge of aqueous liquid-liquid extraction and this is a result of over 40 years of operating experience on an industrial scale. This means that there is considerable awareness of the advantages and disadvantages of this technology. On the other hand, pyrochemical partitioning methods for the treatment of radioactive substances must still be considered to be at the laboratory stage and still require the extensive development of methods as well as new kinds of equipment. This means that there is still a lack of adequate knowledge of the advantages and disadvantages of these methods in connection with the industrial-scale treatment of radioactive substances. Therefore, it still seems to be too early to attempt to compare aqueous chemistry with pyrochemistry on the basis of equal knowledge. Currently, the USA and Russia show the greatest interest in pyrochemistry while the largest member states of the European Union and Japan mainly continue to focus their development work on the improvement and further development of aqueous methods. If partitioning and transmutation is to be

a realistic alternative to the current fuel cycles, significant long-term R&D work on partitioning techniques will be necessary.

It is quite clear today that partitioning and transmutation cannot eliminate the need for a high-level waste repository. In a short-term perspective, the radioactivity of the waste will even be higher than is classically the case due to the fact that long-lived nuclides, to a large extent, will have been converted to considerably more short-lived nuclides. On the other hand, the shorter half-lifes will mean that the waste will become less hazardous relatively quickly with time. In spite of the use of an efficient partitioning and transmutation technology, the waste will contain small quantities of very long-lived radioactive nuclides.

This conditioning of the fuel also makes it possible, in principle, to gain access to weapons-grade material in a relatively pure form. However, on the other hand, there is a somewhat greater possibility of designing fuel, target or entire accelerator-driven systems to make them more secure in terms of safeguards than is the case with conventional reactors and fast reactors.

Even if there is a broad scientific consensus that current types of fuel cycles in combination with planned waste management and geological final disposal will provide a satisfactory protection for humanity in the foreseeable future, there is still a considerable interest in investigating whether an additional reduction in the future potential hazard of the waste can be achieved through partitioning and transmutation and in investigating the cost at which this can be done. One of the weaknesses of the repositories, which are currently being planned, is the difficulty of predicting all possible future events which could affect the performance of the repository. The strength of a partitioning and transmutation process could be to reduce any possible future consequences of such unanticipated events. On the other hand, is it the case that the development of partitioning and transmutation processes will hide the fact that the future risks associated with a well-designed repository are already considered to be very small?

In previous reports, we have emphasized that the introduction of an economically viable partitioning and transmutation technology is most probable in an expanding nuclear power programme. In this context, it is still being debated whether an increased use of nuclear power along with partitioning and transmutation processes should be considered to be a possible objective or whether partitioning and transmutation technology can be an objective, regardless of the future development of conventional nuclear power. A relatively rapid global expansion of nuclear power will certainly result in an increased price of fresh nuclear fuel which may make fuel management based on reprocessing and recycling more economical than it is today. Then fast reactors and accelerator-driven reactors may become attractive in economic terms and from the standpoint of the management of natural resources, since they can utilize the energy stored in natural uranium and thorium a hundred times more efficiently than today's light water reactors. It is therefore possible that the spent nuclear fuel generated today may, instead of being waste, become an important raw material for the further generation of nuclear energy.

Research on and the evaluation of partitioning and transmutation are currently in progress in many industrial countries and within several international organizations due to its potential as a long-term, sustainable energy source with low environmental impact

and due to its ability to destroy many long-lived nuclides. This can lead to radioactive waste repositories which are easier to accept. The cost of the research and development work on partitioning and transmutation is considered to be so great that international co-operation is required. In this context, it may be worth observing that the justification for the development of partitioning and transmutation technology in Europe, Russia and Japan seems to be different from that of the USA.

With respect to partitioning, Sweden participated, at an early stage, in the development of the aqueous partitioning systems which nowadays comprise the basis for reprocessing technology. Furthermore, Sweden has previously been involved, through its research, in the development of the more advanced partitioning processes described in this report. Therefore, Sweden has always had considerable competence and experience of aqueous partitioning technology. In the past decades, the emphasis has been on work carried out at the Department of Nuclear Chemistry at CTH and with the current research which is being conducted within partitioning and transmutation, competence within the area can be maintained to a certain extent. Swedish experience is very limited with respect to other partitioning methods involving high temperatures (metallurgy, electrolytic partitioning) within nuclear applications. On the other hand, considerable industrial experience of these methods has been gained within the Swedish metal industry. However, the technology, with respect to waste production, releases to the environment, industrial safety and other safety-related aspects cannot in any way be compared with the nuclear activities.

Neutron physics and reactor technique are also areas with a very strong competence base in Sweden. This is illustrated by the fact that 9 of Sweden's 12 reactors are of Swedish original design. From the international standpoint, subsequent Swedish proposals from ABB Atom (formerly Asea-atom) concerning new reactor technology (e.g. PIUS) have attracted considerable interest abroad, even if in retrospect, it can be said that these proposals for new safe reactor designs were ahead of time and have not yet been accepted internationally. This has represented a considerable setback in terms of maintaining the broad competence that existed within the area, even if the knowledge is not yet lost. Research activities within reactor technology and neutron physics are being conducted at KTH, CTH and Uppsala University. KTH's research has a clear bias towards accelerator-driven transmutation, which is described in this report.

In the area of accelerator technology, Sweden has maintained a leading position within R&D and has also been able to hold its own with respect to the commercial manufacturing of accelerators. With close ties to universities and institutes of technology (Uppsala University and Manne Siegbahn Institute, MSI), the development of components for accelerators is currently in progress and this is gaining international recognition.

Research concerning the safety-related aspects of nuclear work started at an early stage in Sweden which has resulted in internationally accepted methods for risk analyses. This activity is now being conducted at the Centre for Safety Research at KTH.

To summarize, we find that Sweden has a good possibility of maintaining a relatively high level of competence within areas which are directly related to partitioning and transmutation on condition that funds are available to conduct research within these areas.

Since the further training of highly qualified personnel is required for as long as Sweden still has nuclear facilities, research and development within the areas of partitioning and transmutation technology is a good basis for attracting new, competent students to subjects related to nuclear power. Even if a partitioning and transmutation facility never will be realized in Sweden, there is a further need for individuals with qualified knowledge within the nuclear areas in the forseeable future.

We therefore recommend that SKB continues to support Swedish R&D within the area, with the aim of carrying out the activity in the form of broad international co-operation. We also recommend that SKB (possibly in co-operation with a government authority such as SKI and SSI) participate in the co-ordination of Swedish work within the area. Merely following international developments within partitioning and transmutation will not be sufficient to maintain an adequate level of competence within the area. Sweden must conduct its own research in order to gain insight and, above all, to build confidence in evaluations of international developments within the areas of partitioning and transmutation.

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Appendix 1

Accelerator-driven subcritical systems

During the past decade, an old concept within nuclear technology has stimulated new interest, namely the source-driven subcritical system or "hybrid system". The idea is that a powerful neutron source can drive a subcritical core. Even more neutrons and large quantities of heat will be produced in the subcritical core through the fission of fissile material. Due to technical progress, high-power accelerators can now be manufactured which make hybrid systems practically feasible. The spallation (tearing out) of neutrons in targets for accelerators is the most powerful available source of neutrons for these systems.

Particle accelerators and reactor technology have, for several decades, developed along parallel lines, where a common goal has been the ability to produce a large number of neutrons via nuclear fission (reactors) or spallation (accelerators). Progress in particle accelerator technology coupled with political obstacles to the construction of new reactors has led to the proposal of several large-scale applications of accelerator-driven systems. These applications are based on the better neutron balance in hybrid systems and on their greater flexibility during operation, compared with conventional reactors. Spallation neutron sources, accelerator-driven production of special isotopes for medical and military purposes, accelerator-driven transmutation of nuclear waste and accelerator-driven energy production belong to these.

Accelerators and targets

For physicists, particle accelerators have, for a long time been important research tools. In order to reach high energies, charged particles are collected into bundles and accelerated using electromagnetic fields. All charged particles can be accelerated but usually protons are used for neutron production through collision with a target.

Both linear accelerators and cyclotrons can accelerate uninterrupted streams of radiation, but they have different properties. In a cyclotron, the beam circulates around the middle of the path many times with a radius which varies between several decimeters up to several meters. However, since the distance between one path and the next is very small when the particles approach the speed of light, there may be problems in removing the beam. Cyclotrons are thereby most effective in the case of low energies where the energies of the particles are non-relativistic. Cyclotrons currently produce an average current of somewhat more than 1 mA and can accelerate protons up to 700 MeV.

Linear accelerators are larger than cyclotrons, often several hundred meters long (about 1 m per MeV of proton energy) and are more expensive to build. However, linear accelerators are capable of reaching an average current of about 100 mA with a maximum particle energy which is only limited by the length of the accelerator and are

therefore the best choice for applications which require powerful beams with energies which are greater than 1 GeV.

Until recently, copper cavities were used for particle acceleration in most accelerators as well as iron magnets with copper coils to bend or focus the beams. However, high-intensity systems require large quantities of electricity and in both copper cavities and solenoids there are problems with resistive losses and heating. In the last few decades, successful attempts have been made with superconducting cavities of niobium and solenoids of niobium-titanium alloys, which allow for very high gradients in the acceleration and reduce the resistive losses to a minimum. Superconducting cavities are now being used in several accelerators.

The highest capacity of a linear accelerator which is planned to be built is that of the APT project (Accelerator-driven Production of Tritium) in the USA which is expected to reach a current of 100 mA at 1.7 GeV. The main aim of the project is to secure the supply of tritium for nuclear weapons. Development work on linear accelerators is also being conducted within the Japanese OMEGA project.

Work on designing high-intensity cyclotrons is being conducted at CERN and PSI (Paul Scherrer Institute) in Switzerland. The proton cyclotron at PSI was recently upgraded to a current of 1.5 mA at 650 MeV. An additional increase in capacity to ~5MA is feasible and is planned to be carried out within the next few years.

All proposed accelerator-driven systems have certain main components in common. In addition to an accelerator, there is also a spallation target whcih is used to bombard high-energy charged particles (usually protons with energies from 0.5-2GeV) and emit neutrons and other particles, a multiplication area, where high-energy particles from the target induce secondary production of neutrons and a blanket where the neutrons from the target are used for different purposes, as well as a region which acts as a radiation shield.

The initial collision between the incoming particle and a nucleus in the target leads to an "intranuclear cascade", where neutrons, protons or lighter fragments of nuclei are pulled out from the nucleus in an excited state. "Evaporation" of nucleons from the excited nucleus subsequently occurs when it relaxes to its ground state. Most of these nucleons are neutrons, most of which leave the nucleus during the evaporation phase which is isotropic. In heavy materials such as lead and wolfram, high-energy fission can also occur. If actinides are used, such as uranium and torium, low-energy neutrons can also induce fission.

The highest number of neutrons per incoming proton will be obtained for a target of actinides, even if the contribution from secondary fission is excluded. However, for proton beams of high intensity, the cooling of the target is a problem that is difficult to resolve due to the heat generated from the fissions as well as the large quantity of fission products. Therefore, actinides are currently not used as target components. The most common materials proposed are wolfram, lead and a liquid eutectic lead-bismuth mixture. Lead has a significantly lower cross-section for neutron absorption (0.17 barn compared with 18.2 for wolfram, at thermal energies). Furthermore, lead is assumed to be in the liquid phase while the system is in operation, while wolfram is solid.

Actinides occur in the multiplier and blanket areas. These areas may contain sufficient quantities of fissile material to reach significant subcritical multiplication factors. With a sufficiently powerful neutron source (lead target and 1 GeV of protons with a 10-100 mA current), it is possible, in principle to run a subcritical core (k $\sim$0.95, with a neutron multiplication of $\sim$20) with a generation of fission heat which is fully comparable to that of a large nuclear power plant.

Subcritical fuel cycles

An important question is what advantage an accelerator-driven system (ADS) has compared to a critical reactor with respect to actinide fission. There are a couple of technical justifications.

- For certain fuel types with high heat conductivity and a low share of delayed neutrons (e.g. nitrides of Am and Cm), greater safety margins can be obtained at (unintentional) increases of reactivity.

- In an accelerator-driven system with a high-energy neutron spectrum, individual fission products can also transmute and in greater quantities than with critical fast reactors.

- The possibility of a high burnup (10–20% of the fuel material) will lead to less losses in the partitioning process in an ADS. This is also the case with self-sustaining fast reactors.

Appendix 2

Mathematical relationships for system efficiencies

A short description is provided here, for readers interested in mathematics and processes, of how the performance of the transmutation system, the efficiency of the partitioning process and the system's mode of operation interact to achieve a total efficiency for the removal of long-lived products from the waste which is to be disposed of. Since this is of greatest interest when evaluating the suitability of a system for a particular purpose, we have estimated the fraction of the original existing long-lived substance which is finally left in the waste which must be disposed of. The aim of a combined transmutation and partitioning process is to reach values of this fraction, F, which, for example, are less than one-hundredth, i.e. $< 1\%$ of the original quantity may be allowed to remain in some form of waste.

From the outset, two main scenarios can be distinguished. In the first scenario, the partitioning occurs continuously online to the transmutation system, which requires a gaseous, liquid, powdered or pellet-shaped fuel. In this scenario, the transmutation and partitioning facilities must be located next to each other. In the second scenario, the partitioning occurs discontinuously offline in the same way as during reactor operation with the reprocessing of spent nuclear fuel. In this scenario, the physical form of the fuel is much less significant. The material is transported, when necessary, between the facilities, which can thereby be located independently of each other. In both scenarios, it is assumed that the transmutation rate is independent of the irradiation time, which is a rough estimate. Since the calculations are performed in different ways we will examine each scenario separately.

A2.1 Continuous transmutation and partitioning

During continuous operation of a transmutation process with directly connected continuous partitioning, the aim is probably to keep the concentration of fuel atoms constant in the irradiation system. In order to achieve this, a flow chart, as shown in Figure A2-1 will apply. According to this flow chart, as many new fuel atoms will be

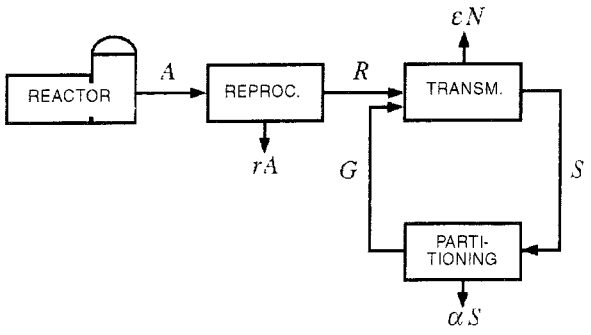

Figure A2-1. *Flow chart showing the continuous transmutation and partitioning but discontinuous reprocessing.*

supplied per unit of time to the system as those that disappear per unit of time through transmutation and losses in the partitioning process. The following material balance can then be established for the entire process:

$$(1\text{-}r)\,A = R = \epsilon\,N + \alpha\,S \qquad \text{(A2.1)}$$

where N is the total number of fuel atoms which are being irradiated, ϵ is the transmutation rate constant, R is the number of fuel atoms which is supplied per unit of time, S is the number of fuel atoms per unit of time which pass into the partitioning process and α is the fraction of S which is lost to the waste from the partitioning process. Let us now assume that the irradiation system has a given volume V and that the volume flow rate v to the partitioning facility is a fix fraction k of the irradiation volume V. The following series of relationships is obtained:

$$C = N / V$$
$$v = k\,V$$
$$S = v\,C$$

If we designate the total loss to the waste from the entire process as L, the following is obtained:

$$L = r\,A + \alpha\,S$$

where A designates the quantity of material (number of atoms) per unit of time which pass into the facility for reprocessing. Since we are interested in the size of the fraction, F, of A which finally ends up in the waste, we can express this as follows:

$$F = L / A$$

If we now insert our different relationships into the equation (A2.1) and simplify the result, the following is obtained:

$$F = r + (1\text{-}r)/[1+\{\epsilon/(k\,\alpha)\}] \qquad \text{(A2.2)}$$

As explained by this equation, we aim to achieve the greatest possible value for $\epsilon/k\alpha$ in order to obtain the smallest possible loss at a given loss during reprocessing. According to the equation (1) we can, for relatively short irradiation times, t, express the transmutation rate $\epsilon\,N$ ($= -dN/dt$) as the product $\phi_{\text{average}}\,\sigma_{\text{average}}N$. If we insert this relationship into equation (A2.2) the following is obtained:

$$F = r + (1\text{-}r)/[1+\{\phi_{\text{average}}\,\sigma_{\text{average}}/(k\,\alpha)\}] \qquad \text{(A2.3)}$$

At reasonable cross-sections and neutron fluxes, the product $\phi_{\text{average}}\,\sigma_{\text{average}}$ will always have a very low value, $< 10^{-8}$. This means that the product $k\,\alpha$ must be $\ll 10^{8}$ in order for the losses to the waste to be reasonable. On the other hand, if we decide that the process should be carried out at very small k values, one of the advantages of a continuous partitioning will disappear; the possibility of minimizing the quantity of fission products in the irradiation system.

To illustrate this point, let us examine the transmutation of plutonium in a metal-cooled accelerator-driven system. The fission cross-section is assumed to be about 2×10^{-28} m^2 and the neutron flux 10^{19} n m^{-2} s^{-1}. During reprocessing, the loss to the waste is 0.2% (r = 0.002), and during the continuous "on-line" partitioning, it is assumed that the loss is as great, i.e. 0.2% (α = 0.001). The flow through the partitioning process is set up so that the entire irradiated volume on average passes through the partitioning facility once per day ($k = 1.16\times10^{-5}$ s^{-1}). If these numerical values are inserted into the equation (A2.3), we obtain a calculated total loss, F, of plutonium to different types of waste which is about 92%, which means that the partitioning and transmutation process is very inefficient under these conditions.

A2.2 Discontinuous transmutation and partitioning

During discontinuous operation of a transmutation process and the corresponding partitioning process with the flow chart according to Figure A2-2, we can imagine a given incoming volume of material A, circulating around in the system, cycle after cycle, until it is destroyed, i.e. nothing is left which can be circulated. After a few simple mathematical operations, this will lead us to a relationship for the total lost fraction, F, of the original quantity of the atoms which has been subject to transmutation.

The process starts after removing a certain quantity of spent nuclear fuel which contains the quantity A, of the substance which is to be destroyed through transmutation. In the figure, values are specified for the first cycle through the process. Spent nuclear fuel is reprocessed in the usual manner, and the fraction r of the original quantity of the fuel is lost as waste. The recovered material $(1-r)A$, then goes on to a facility for the manufacturing of targets for the transmutation process. In this manufacturing process, the fraction δ is lost as waste. In the irradiation facility, the fraction ϵ of the remaining quantity of the substance is then transmuted. Let us imagine that this fraction is completely removed from the process as it goes through the partitioning process. After irradiation, the target goes on to the partitioning process where non-transmuted material is recovered. The fraction α is thereby lost to different waste streams while the final production of new material in a suitable form for transmutation once again will mean

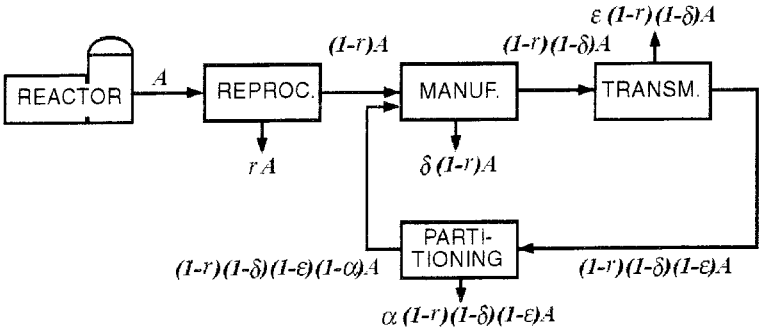

Figure A2-2. *Flow chart showing the transmutation of material from discontinuous reprocessing and transmutation.*

that δ will become waste. The process is then repeated. This can be expressed in mathematical terms by the following equation for the total loss, F, as a fraction of A.

$$F = r + (1-r)\{\delta\sum[(1-\delta)^n(1-\epsilon)^n(1-\alpha)^n] + \alpha\sum[(1-\delta)^{n+1}(1-\epsilon)^{n+1}(1-\alpha)^n\}] \qquad \text{(A2.4)}$$

The summation sign means that the entire expression after each summation sign must be calculated and totalled for all integers of n from zero to infinity. Using the theory of infinite geometrical series, the equation (A2.4) can be converted into the following expression:

$$F = r + (1-r)\{\delta + \alpha(1-\delta)(1-\epsilon)\}/\{1-(1-\alpha)(1-\delta)(1-\epsilon)\} \qquad \text{(A2.5)}$$

An efficient transmutation process requires that r as well as α and δ are small («1).This will lead to a simplification of the equation (A2.5) with the help of calculation rules for mixtures of small and large numbers. The following approximate relationship is then obtained, if also ϵ is relatively small:

$$(\alpha + \delta) \approx \epsilon\,(F-r)/(1-F) \qquad \text{(A2.6)}$$

This relationship allows an estimate to be made of the necessary total efficiency in the partitioning and fuel fabrication processes, i.e. the value of $\{1-(\alpha+\delta)\}$, for known values of the loss from reprocessing (r), the transmuted fraction during one irradiation cycle (ϵ), and the permitted total loss (F) of the input material to all the waste streams. Note that F is always greater than r but less than 1.

Let us once again consider plutonium on the assumption that we can irradiate it using an average flux which is the same as in the previous example, 10^{19} n/m^2 s, for a period of 3 years and with an average fission cross-section of 2×10^{-28} m^2. With these values, ϵ can be calculated to 0.1725. Furthermore, let us assume the same chemical efficiency for both the reprocessing process and the partitioning process, $r = \alpha = 0.2\%$. Experience from the fabrication of MOX fuel shows that an efficiency of 99.99% can probably be reached in the fabrication of fuel. This corresponds to $\delta = 0.0001$. From these values we can now calculate the total loss of plutonium to different waste streams, F, using the equation (A2.5). We will then find that about 1.2% of the plutonium which was originally in the spent nuclear fuel will end up in the radioactive waste while the rest will transmute into fission products or into higher actinides.

The two examples of losses during continuous partitioning and transmutation as well as discontinuous partitioning and transmutation clearly indicate the advantages of discontinuous chemical partitioning over directly connected continuous chemical partitioning.

Appendix 3

Glossary with synonyms and explanations

ACTINEAU	French research programme for the improvement of current reprocessing technology.
ACTINEX	ACTINide EXtraction. French research project for the development of new improved technique for the reprocessing of nuclear fuel with the recovery of not only uranium and plutonium but also neptunium, americium and curium. The project is run by CEA in co-operation with COGEMA.
Actinide	Element belonging to the actinide series, i.e. elements of atomic numbers (= nuclear charge) 89 through 103.
AEA	Atomic Energy Authority. UK nuclear organization.
AEC	United States Atomic Energy Commission – earlier federal US organization for military and civil research, development and use of nuclear energy.
ALI	Annual Limit on Intake. Limit on the annual intake of a radioactive nuclide for personnel involved in work with radioactive substances, Bq. In this report, the intake is assumed to occur through the mouth.
Aliquat-336	Quaternary organic ammonium salt, extractant manufactured by the Henkel Corporation.
ALMR	Advanced Liquid Metal Reactor. US reactor type which has been developed in order to work together with a simplified pyrochemical reprocessing process. Exists only at the drawing board stage.
APT	Accelerator-driven Production of Tritium. Accelerator-driven tritium production, R&D project at LANL.
Atomic number	The atomic number of an element designates its place in the periodic table of elements. The atomic number is the same as the elements nuclear charge (number of protons in the atomic nucleus).
ATW	Accelerator-driven Transmutation of Waste. R&D project at LANL.
BWR	Boiling Water Reactor.

CEA	Commissariat á l'Énergie Atomique. French state-owned organization responsible for research and development within civil and military nuclear activities.
CEFR	China Experimental Fast Reactor. Chinese experimental fast reactor.
CENDL-2	Chinese Evaluated Nuclear Data Library. Chinese neutron data library.
CERN	Conseil Européen pour la Recherche Nucléaire. European research organization whose activities mainly focus on elementary particle physics.
CHON principle	Molecules which only contain carbon (C), hydrogen (H), oxygen (O).
CLIB-2	Chinese LIBrary. Chinese neutron database library.
CMPO	The extractant, Carbamoyl Methylene Phosphine Oxide.
CNDC	Chinese Nuclear Data Center.
CNRS	Centre National de la Recherche Scientifique. French, state-owned centre for basic scientific research.
CRIEPI	Central Research Institute of the Electric Power Industry. Japanese research institute.
CURE	Clean Use of Reactor Energy. US reactor project.
Cyanex-301	The extractant, bis(2,4,4-trimethylpentyl)ditiophosphinic acid.
Depleted	An element whose concentration of a certain isotope is reduced through isotope separation. For example, depleted uranium; the concentration of ^{235}U (and ^{234}U) is reduced in relation to natural uranium.
DHDECMP	The extractant, Di Hexyl-N,N-Di Ethylene Carbamoyl Methylene Phosphonate.
DIAMEX	DIAMide EXtraction. French liquid extraction process based on diamide.
DIDPA	The extractant, Di-IsoDecyl Phosphoric Acid.
ECN	Energieonderzoek Centrum Nederland. Dutch energy research centre.

EDF	Electricité de France. French state-owned power utility.
EET	Emerging Energy Technology. Research team at CERN working on the thorium-based fuel cycle in accelerator-driven systems. Led by Carlo Rubbia.
ENEA	Ente per le Nuove tecnologie, l'Energia e l'Ambiente. Italian energy research organization.
Enriched	An element whose concentration of a certain isotope is raised through isotope separation. An example: enriched uranium; the concentration of the uranium isotope ^{235}U (and ^{234}U) is raised in relation to natural uranium.
Enrichment	An increase in the concentration of an isotope of an element (see also isotope separation). The term is used in a limited sense also to refer to the concentration of ^{235}U in enriched uranium intended as reactor fuel.
FBR	Fast Breeder Reactor.
FEAT	First Energy Amplifier Test. Subcritical experiment at CERN using ^{238}U as the target.
Fission	Normally refers to the splitting of an atomic nucleus into two lighter nuclei (fission products). The number of atoms is doubled when atomic nuclei are split.
FLUKA	FLUctuating KAscade. Computer code for calculating particle transport at high energies.
Half-life	The time taken for the original number of a radioactive nuclide to decay to half of the number. Since radioactive decay is a process which (with few exceptions) cannot be affected, the half-life is a unique unit for each type of radioactive nuclide.
HDEHP	The extractant, Di-2-Ethylene Hexyl Phosphoric Acid.
IABAT	Impact of the Accelerator-Based Technologies on Nuclear Fission Safety. EU project which evaluates accelerator-driven systems.
IAEA	International Atomic Energy Agency. Independent part of the United Nations organization which aims at promoting the peaceful use of nuclear energy.
IBA	Ion Beam Applications. Belgian company designing and manufacturing accelerators on a commercial basis.

IPPE	Institute of Physics and Power Engineering. Federal Russian research centre for nuclear technology, located in Obninsk.
ISAAC	Investigations on Subcritical Systems Driven by Accelerators. CEA/Cadarache's research programme on accelerator-driven systems.
Isotope	An atom of an element which has a unique atomic mass. This is due to the fact that the number of neutrons in the atomic nucleus can be different for atoms with the same nuclear charge, i.e. for atoms of one and the same element. Since the atomic mass mainly comprises the mass of protons and neutrons in the atomic nucleus, the composition of the nucleus determines the weight of the atom in practice.
Isotope separation	Change in the natural isotope composition of an element so that one (or several) of the element's isotopes is concentrated in a product (enriched) while the residual product (depleted) has a reduced concentration of the corresponding isotope (or isotopes).
ISTC	International Centre of Science and Technology. Agency with its headquarters in Moscow and founded to act as an intermediary for research grants from developed countries to former weapons research institutes in the former Soviet Union.
JAERI	Japan Atomic Energy Research Institute. Japanese state-owned nuclear research institute.
KAERI	Korea Atomic Energy Research Institute. South Korean nuclear research institute.
Nuclear charge	The positive charge of the atomic nucleus, i.e. the number of protons in the nucleus (= atomic number).
LAESA	Laboratorio del Amplificador de Energia. Spanish-Italian consortium initiated by the EET to obtain funds for the construction of an accelerator-driven prototype facility in Spain.
LANL	Los Alamos National Laboratory. Federal research laboratory in the USA.
Lanthanide	Element belonging to the lanthanide series, i.e. elements with a number (= nuclear charge) of 57–71. Also known as a "rare earth element" in the older literature.
LWR	Light Water Reactor. Light-water moderated and cooled thermal reactor.

MASURCA	Research reactor in Cadarache, France, where a number of subcritical experiments are carried out.
MOX	Mixed OXide. Nuclear fuel containing a mixture of uranium dioxide and plutonium dioxide.
Myrrha	R&D project at SCK-CEN on accelerator-driven systems.
NEA	Nuclear Energy Agency. OECD's nuclear energy agency.
NEWPART	NEW PARtitioning Techniques. EU-funded research project within the EU Nuclear Fission Safety Programme. The NEWPART project is developing new reagents and partitioning methods for actinides.
Nuclide	General term designating atoms of a given type, i.e. an isotope of an element.
OMEGA	Options for Making Extra Gain of Actinides and fission products generated in the nuclear fuel cycle. Japanese research programme on partitioning and transmutation.
OSIRIS	On-line System for Isotope separation at the Reactor In Studsvik. Research equipment at the R0-reactor at Studsvik, Sweden, which is used to separate and study short-lived fission products.
Partitioning	Separation. Recovery of one (or sometimes several) elements in pure form from a mixture of elements (see also isotope separation).
PBR	Pebble Bed Reactor. Originally a German gas-cooled reactor design with graphite encapsulated fuel in pellet form.
PHENIX	Name of a small French fast breeder reactor located at Marcoule in France (250 MWe). The reactor was started up in 1972 and is operated jointly by EDF and CEA.
PHOENIX	Name of a subcritical accelerator-driven fast reactor design proposed by BNL (Brookhaven Nat. Lab.), USA.
PHWR	Pressurized Heavy Water Reactor. Heavy water-moderated pressurized water reactor.
PNC	Power reactor and Nuclear fuel development Corporation. Japanese company.

PRAV	National council for radioactive waste management. Earlier Swedish state-owned research agency for R&D on the treatment and handling of radioactive waste.
PUREX	Plutonium Uranium Redox EXtraction. Name of the most common chemical process for the reprocessing of spent nuclear fuel.
PURETEX	French research project which aims at improving the existing PUREX process.
PWR	Pressurized Water Reactor.
Pyrometallurgy	Pyroprocessing. A general term which refers to inorganic chemical processes carried out at high temperatures. These processes are most common within the metallurgical industry (production and refining of iron, steel, alloys, copper, lead, zinc, etc.) And for the production of certain metals through molten salt electrolysis (e.g. aluminium and magnesium).
RAS	Transmutation research programme at the ECN.
Radioactivity	Radioactive decay rate. Specified in Becquerel (Bq), i.e. the number of atoms decaying per second.
Radionuclide	General term referring to atoms undergoing radioactive decay. Some type of radiation is emitted during decay.
Reprocessing	Treatment of spent nuclear fuel for the recovery of unused uranium and plutonium. This is usually achieved through the PUREX process. Other processes have been developed and tested, but these are no longer used for large-scale industrial applications.
SANEX	Selective Actinide Extraction. Process for the selective recovery of actinides.
SCK-CEN	Studie Centrum voor Kernenergie – Centre d'Étude de l'Énergie Nucléaire. Belgian nuclear research centre.
SKODA	Czech company which has participated in the design of Russian pressurized water reactors.
SPIN	Separation and Incineration in Reactors. French research programme for the development of new partitioning and transmutation technology.

STC	(Science and Technology Committee) Committee within the EU for evaluations and advice concerning technology.
TALSPEAK	Trivalent Actinide Lanthanide Separation by Phosphorous-reagent Extraction of Aqueous Complexes. Name of a chemical process for the separation of actinides and lanthanides through liquid extraction with HDEHP in the presence of DTPA's sodium salt.
TARC	Transmutation through Adiabatic Resonance Crossing. CERN Experiment financed by the EU where measurements of cross-sections for particle scattering in lead are being carried out.
TBP	The extractant, Tri Butyl Phosphate. An extractant which is used in the PUREX and other processes.
Transmutation	Conversion of an element into one or several elements. This usually occurs through the capture of a neutron followed by radioactive β decay to the next higher element. For heavy elements (high atomic numbers) an isotope of the element can instead be split after neutron capture causing two lighter elements to form (fission products). Neutron capture and fission are two competing processes in the latter case.
TRPO process	TRialkyl Phosphine Oxide. Process intended to separate actinides.
TRUEX	TransUraniumEXtraktion. Name of an old liquid extraction process for the separation of trivalent actinides and lanthanides which is based on aminextraction from concentrated chloride solutions.
TUI	Trans Uranium Institute. The EU research centre in Karlsruhe, Germany.

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Itasca Geomekanik AB, Stockholm, Sweden

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Johan Andersson[1], Karl-Erik Almén[2], Lars O Ericsson[3], Anders Fredriksson[4], Fred Karlsson[3], Roy Stanfors[5], Anders Ström[3]

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C Juhlin[1], T Wallroth[2], J Smellie[3], T Eliasson[4], C Ljunggren[5], B Leijon[3], J Beswick[6]

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[3] Conterra AB

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[5] Vattenfall Hydropower AB

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I Gurban[1], M Laaksoharju[1], E Ledoux[2], B Made[2], A L Salignac[2],

[1] Intera KB, Stockholm, Sweden

[2] Ecole des Mines, Paris, France

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Claes-Göran Andersson

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