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Partitioning and transmutation. A review of the current state of the art

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ABSTRACT

The recent development in the field of partitioning and transmutation (P-T) of long-lived radioactive waste nuclides from nuclear power production is reviewed and evaluated. Current national and international R&D plans are summarized. It is concluded that P-T is technically feasible but much R&D remains to be done before it is technically mature. At present there seems to be no economic gain from P-T as compared to direct disposal of spent nuclear fuel. There seems only to be an insignificant reduction in future radiation doses by P-T when compared to current disposal plans. However, future long term research may perhaps change these conclusions. Therefore the further development in this area should be followed. Some areas where a limited research by swedish scientists could be worth while are indicated.

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1 INTRODUCTION

The concept of partitioning and transmutation "P-T" has reemerged as an alternative for handling of radioactive waste during late 1980ies. The goal of P-T has been presented as a way to reduce the long term hazard of such a waste, which is supposed to make geological disposal more acceptable to the public. A few proposals have even suggested that P-T would eliminate the need for geological deposition.

1.1 THE BASIC CONCEPTS FOR PARTITIONING AND TRANSMUTATION

Nuclear processes may be used to destroy some of the radionuclides in nuclear wastes. This has obvious attractions, since otherwise long lived nuclides remain as a potential hazard for very long periods of time.

The nuclear reactions considered are induced by neutrons, causing either fission or neutron capture. The reaction products should be either stable, or short lived with a decay to stable, to reduce the long term radiological hazard. To have an acceptable transmutation rate, high neutron fluxes are needed and the transmutation devices with the greatest potential at present are high flux reactors or accelerator driven neutron sources.

Since the considered transmutation cross sections are relatively low and the highest achievable neutron flux is limited, one have to recycle the radionuclides many times to burn them up sufficiently. This demands for separation procedures with high efficiency. One can consider several different chemical separation processes, but most experiences are at present available in liquid-liquid extraction from aqueous solutions.

The reduction factors one want to achieve for individual radionuclides in the nuclear waste will determine the numbers of recycling and the required chemical separation efficiency. For example if the transmutation efficiency in one cycle is low, the separation efficiency must be high to sustain the overall wanted reduction factor after several cycles. This means that one have to put as much research efforts to the chemical separations as to the construction of a transmutation device.

The main benefits of partitioning and transmutation is that the residual waste, depleted from long lived radionuclides, has in the ideal case lost its radiotoxicity after that the dominating fission products, ^{90}Sr and ^{137}Cs, have decayed to a low level, which takes about 1000 years. It should be mentioned that transmutation of ^{90}Sr and ^{137}Cs have been considered in some studies.

1.2 TERMINOLOGY

1.2.1 Partitioning

Chemical treatment of spent nuclear fuel in order to recover uranium and plutonium is called reprocessing. Further separation of residual isotopes in the high active waste (HAW) was previously called partitioning. During the last years all chemical processes discussed in connection with transmutation, including modification of existing reprocessing schemes and new separation procedures, have been assigned partitioning. In the context of this report, the term partitioning is used for all chemical processes connected with transmutation.

1.2.2 Transmutation

Nuclear transmutation is the transformation of a nuclide into another either naturally by radioactive decay or artificially by nuclear reactions. The term transmutation occurs historically from alchemy when gold was to be produced by transmutation from other substances.

The term nuclear incineration have also been used as an analogue for nuclear transmutation, in the context of "burning" radioactive waste products in nuclear reactors. In this report, only the term transmutation is used, with the meaning nuclear transmutation.

1.3 ABBREVIATIONS

ABR	Actinide Burning Reactor
ALI	Annual Limit of Intake
ALMR	Advanced Liquid Metal Reactor
ATW	Accelerator Transmutation of Waste
BWR	Boiling Water Reactor
CEA	Commissariat a l'Energie Atomique, France
CEC	Commission of the European Communities
CMPO	Carbamoylmetylphosphine oxide
CTH	Chalmers University of Technology, Sweden
CURE	Clean Use of Reactor Energy
DFR	Dounreay Fast Reactor
DHDECMP	Dihexyl-N,N-diethyl-carbamylmethylene-phosphonate
DTPA	Diethylene-triamine-pentaacetic acid
CRIEPI	Central Research Institute of Electric Power Industry
FBR	Fast Breeder Reactor
HAW	High Active Waste
HLLW	High Level Liquid Waste
HLW	High Level Waste
IAEA	International Atomic Energy Agency

ICRP	International Commission on Radiation Protection
IFR	Integral Fast Reactor
INEL	Idaho Nuclear Engineering Laboratory, USA
LLW	Low Level Waste
JAERI	Japan Atomic Energy Research Institute
LMFBR	Liquid Metal Fast Breeder Reactor
LWR	Light Water Reactor
MA	Minor Actinides
MOX-fuel	Mixed OXide fuel
NUCEF	Nuclear Fuel Cycle Safety Engineering Research Facility, Japan
OECD/NEA	Organization for Economic Cooperation and Development/Nuclear Energy Agency
OMEGA	Options Making Extra Gain from Actinides
ORNL	Oak Ridge National Laboratory, USA
PNC	Power Reactor and Nuclear Fuel Development Corporation, Japan
PRISM	Power Reactor, Innovative, Small Module
PUREX	Plutonium Uranium Redox by EXtraction
P-T	Partitioning-Transmutation
SPIN	Separation - Transmutation
SREX	Strontium extraction process
TALSPEAK	Trivalent Actinide Lanthanide Separation by Phosphorous reagent Extraction from Aqueous Complexes
TBP	Tri-n-butylphosphate
TRUEX	TRansUranium element EXtraction process
TRUMP-S	TRansUranic Management by Pyroprocessing - Separation

1.4 RADIONUCLIDES CONSIDERED FOR P-T.

There is a worldwide scientific consensus that the present reference fuel cycle scenario, including final storage of high level waste, gives adequate protection to mankind, but there is a strong interest to see whether further reduction of the long termed potential hazard can be achieved and at what cost.

Earlier P-T programs, (see 2.1) were only concerned with transmutation of actinides in nuclear reactors. The idea was to have a total recycling of all the actinides, with the exception of natural Th and U isotopes, and possibly also the very long lived ^{236}U isotope [IAEA 82]. Fission products like ^{99}Tc and ^{129}I are also contributors to the long term potential hazard. But these nuclides were not considered to be transmuted in any type of nuclear reactor.

Most of the new proponents concerning P-T agree that research and development should concentrate on the radionuclides which are of major

concern because of their expected contribution to the dose to mankind. The nuclides which are of major concern in this context may depend on the nature and location of the waste repository. Today one agrees that plutonium can be recycled to Light Water Reactors (LWR's) as Mixed Oxide (MOX) fuel or as fuel to fast reactors. There is also an agreement in dose calculations that the most dominating long term hazardous waste nuclides, that not recycled as MOX fuel, are ^{237}Np (and its precursors ^{245}Cm, ^{241}Am and ^{241}Pu) and the fission products ^{129}I and ^{99}Tc. Besides, ^{135}Cs, ^{93}Zr and ^{14}C may be of significant interest because of their long term radiotoxicity. In repositories containing reactor wastes as well as reprocessing wastes, ^{36}Cl may also be of concern, as have been shown in safety analyses in e.g. the UK [Web 91]. A chart of the generic relationship between the dominating actinides using the uranium fuel cycle is presented in figure 1.1.. This shows the main nuclear processes involved, both in normal thermal reactor operation and when actinides are recycled/transmuted. SF and FISS in the figure shows nuclides with spontaneous fission decay and nuclides with high cross-sections for fission with thermal neutrons, respectively .

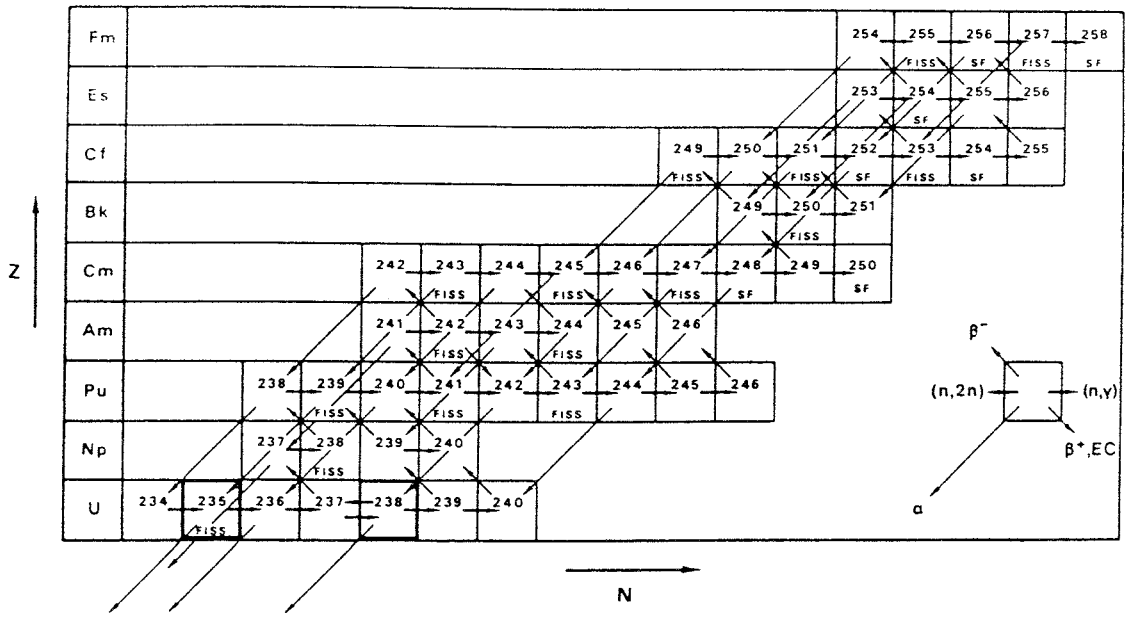

Fig. 1.1. **Major nuclear processes involved in build-up and destruction of actinides in nuclear reactors. Possible transmutation pathways end with FISS and SF marked nuclides [Per 83].**

In the Japanese proposals [AEC 88], transmutation of ^{90}Sr and ^{137}Cs are also considered. This seems not, however, to be realistic to achieve with present technology. Spent nuclear fuel contain significant quantities of valuable metals including the platinum group elements (ruthenium, rhodium, palladium) which are very rare in nature. In the Japanese proposals the incentive to recover these elements is discussed [Kub 89].

Some of the important properties of the nuclides intended to transmute are summarized in table 1.1. It should be remembered that chemical

partitioning normally only gives a mixture of the different isotopes of an element present in the waste. It could therefore be necessary to make an isotope separation of a specific nuclide before it can be transmuted.

Tab 1.1. Some properties of nuclides intended for P-T.

Nuclide	Half life	Decay mode	Other isotopes present in HLW[1]
Actinides			
^{238}Pu	87,7 y	α	see other Pu isotopes
^{239}Pu	2,4 10^4 y	α	see other Pu isotopes
^{240}Pu	6,6 10^3 y	α	see other Pu isotopes
^{241}Pu	14,4 y	β^-	see other Pu isotopes
^{242}Pu	3,8 10^5 y	α	see other Pu isotopes
^{237}Np	2,1 10^6 y	α	
^{245}Cm	8,5 10^3 y	α	^{243}Cm(sh), ^{244}Cm(sh), ^{246}Cm(l), ^{247}Cm(l), ^{248}Cm(l)
^{241}Am	433 y	α	^{242m}Am(l), ^{243}Am(l)
Long lived fission and activation products			
^{129}I	1,6 10^7 y	β^-	^{127}I(s)
^{99}Tc	2,1 10^5 y	β^-	
^{135}Cs	2 10^6 y	β^-	^{133}Cs(s), ^{134}Cs(sh), ^{137}Cs(sh)
^{93}Zr	1,5 10^6 y	β^-	^{90}Zr(s), ^{91}Zr(s), ^{92}Zr(s), ^{94}Zr(s), ^{96}Zr(s)
^{14}C	5,7 10^3 y	β^-	^{12}C(s), ^{13}C(s)
^{36}Cl	3,0 10^5 y	β^-	^{35}Cl(s), ^{37}Cl(s)
Relatively short lived fission products			
^{90}Sr	28,5 y	β^-	^{86}Sr(s), ^{88}Sr(s)
^{137}Cs	30,2 y	β^-	^{133}Cs(s), ^{134}Cs(sh), ^{135}Cs(l)
Valuable metals			
Ru	stable		^{99}Ru(s), ^{100}Ru(s), ^{101}Ru(s), ^{102}Ru(s), ^{104}Ru(s)
Rh	stable		^{103}Rh(s)
Pd	(stable)	(β^-)	^{104}Pd(s), ^{105}Pd(s), ^{106}Pd(s), ^{107}Pd(l)[2], ^{108}Pd(s), ^{110}Pd(s)

[1] The letters within brackets means: (s)=stable (l)=long lived >100 y, (sh)=short lived ≤100 y

[2] ^{107}Pd ($t_{½}$ = 6,5 10^6 y), β^-

1.5 ASSESSMENT CRITERIA

1.5.1 Reduction of the long term radiological hazard by P-T

A properly designed underground repository is expected to lead to insignificant doses on the surface and insignificant concentrations of radionuclides in surface waters, soil, plants and animals for a foreseeable future and long beyond that. Hence, we can safely assume that relatively short lived radioisotopes, without long lived parents, will never present any undue hazard to the environment. However, the predicted behavior of a repository becomes by necessity increasingly uncertain with time. An absolutely certain way to reduce risks from a repository in a far future is to reduce its content of very long lived radionuclides not normally present in nature.

Assume that a, today unrecognized or regarded as very unlikely, process in a far future will release a substantial fraction of the inventory of surviving radionuclides. The relative importance of these nuclides will mainly depend on their amount, specific activity of the corresponding element, transfer probability to living organisms, and radiotoxicity. The first two factors can be estimated from available data. However, the last two factors are almost impossible to estimate for a far future as they involve predictions of the future types, properties, and food chains of living organisms. In this situation it is usually assumed that these factors remain forever as they are today. That assumption permits us to compute the relative potential hazard of an instantaneous release to the environment of each nuclide, for the amount present in the repository or in part of the repository, as a function of time. Such data, normalized in some way, are usually referred to as potential hazard indices. Graphs of a typical hazard index for unreprocessed fuel and for high-level waste from reprocessing of the same fuel are shown in figure 1.2.

Assuming figure 1.2 to be valid, it is obvious that in the time span 1000 years to above 10 million years the potential hazard from the actinides in a repository is very much higher than the potential hazard from the long lived fission products. The conclusion is the same whether the spent fuel is reprocessed or not. The ratio between the potential hazard indices for actinides and fission products is almost always large regardless of the different assumptions made when computing such indices. This permits us to, with some confidence, rank the various components in a repository with regard to their potential hazard.

Highest potential hazard: Actinides, perhaps with exception of uranium.
Lower potential hazard: Fission and activation products.

This conclusion is important for the possible reduction in potential hazard by transmutation because:

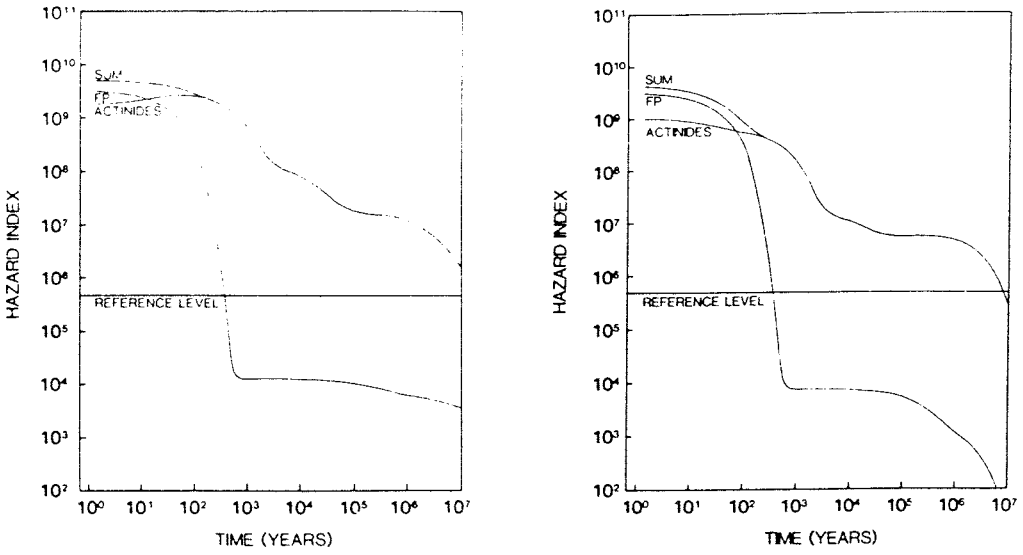

Fig. 1.2. **Example of a potential hazard index base on ALI values. The left diagram is for unreprocesses fuel and the right diagram is for reprocessing HLLW [Per83].**

a) All of the actinides are fissionable with high energy neutrons (fissible) and many also with thermal neutrons (fissile). Fission of actinides always releases about the same amount of energy (≈ 200 MeV/atom) and several neutrons. When fissionable by either slow or fast neutrons, the fission cross sections are normally higher for fission by slow than by fast neutrons.

b) Conversion of an actinide atom to two fission product atoms reduces the total potential hazard index after about 1000 years storage.

c) Long lived fission products are not fissionable with neutrons. They can only be transmuted to stable or shorter lived nuclides by changing the mass number and/or the nuclear charge. In general, the long lived fission products have small reaction cross sections for the processes of interest, i.e. (n,γ), (n,2n), (n,p), (n,α) and perhaps (γ,n). The energy released by such reactions is also small (and may even be negative) compared to that for fission of actinides. A further complication is the mixture of stable and radioactive isotopes produced in a fission reactor. Chemical separation of an element (from Bi and below with two exceptions, Pm and Tc) having a long lived isotope produces a mixture of stable and radioactive isotopes of the element and it has no sense to transmute the already stable part of this isotope mixture.

2. BACKGROUND

2.1 HISTORY OF P-T STUDIES

The idea of P-T is not new. Several studies were initiated after a report presented by Claiborne in 1972 [Cla 72]. A detailed review of the history of P-T concepts is given by Croff [Cro 90], who pointed out that the first documented suggestion of transmutation as a radioactive waste management option was made by Steiberg et al. [Ste 64] in 1964. The history of P-T can be divided into (1) pre-1982 studies and (2) current events. Most of the previous studies, until 1982, were concerned with transmutation in fission reactor systems, but fusion reactors were also considered, whereas only a few papers dealt with the possibility to use accelerator-driven systems. It is also evident that most of the work performed during 1976-1982 concerned specific technical details in connection with P-T, and not so much the overall assessment of P-T, this is reflected in the literature survey (c.f. 2.2).

2.1.1 Pre-1982 P-T Studies

Before 1976, studies were primary initiated by individuals in universities or national laboratories. The main concern was separation of long lived actinides from high level waste (HLW).

In 1976 several larger P-T programs were initiated including the European studies [Sch 83] and the Oak Ridge National Laboratory (ORNL) P-T study [Cro 80]. Several international meetings and conferences were arranged during this period to discuss P-T and related problems. Some of the major meetings were:

- *The Management of Radioactive Waste: Waste Partitioning as an Alternative, US Nuclear Regulatory Commission, Battelle-Seattle Research Center, Seattle, Washington, USA, 8-10 June 1976 [NRC 76].*

- *First Technical Meeting on the Nuclear Transmutation of Actinides, Nuclear Science and Technology, Commission of the European Communities, Ispra, Italy, 16-18 April 1977 [Hag 77].*

- *Second Technical Meeting on the Nuclear Transmutation of Actinides, Commission of the European Communities (CEC), Ispra Italy, 21-24 April 1980 [Hag 80].*

- *International Conference on Nuclear Waste Transmutation, The University of Texas, Austin, Texas, USA, 22-24 July 1980 [Aus 81].*

In 1976 the International Atomic Energy Agency (IAEA) started a coordinated research program on the "Environmental Evaluation and Hazard Assessment of the Separation of Actinides from Nuclear Wastes followed by either Transmutation or Separate Disposal". In 1982 the work were summarized in a report [IAEA 82]. In the overall assessment of P-T some of the conclusions were:

* *"It is considered that with some reservations the technical feasibility of P-T can be regraded as established."*

* *"Technical feasibility should not be taken to imply that routine operation under industrial conditions is assured."*

* *"The magnitude of the long-term hazard depends on the disposal mode and conditions. Estimates of such hazards are still at preliminary stage and are in general conservative based. They indicate, nevertheless, that actinide containing wastes can be disposed of with acceptable safety without introduction of P-T"*

* *"Since the long-term hazards are already low, there is little incentive to reduce them further by P-T. Indeed, the incremental costs of introducing P-T appear to be unduly high in relation to the prospective benefits."*

* *"Implementation of P-T would require a very large effort, and could hardly become general until well into the next century. The principle nuclear reactors will then probably be FBR's, so these would have to be used for P-T. They appear, in any case, to be technically somewhat superior to LWR's for the purpose."*

* *"All in all, the implementation of P-T would be an immense undertaking, involving a large proportion of a country's nuclear power programme, but providing at best a rather small reduction in potential long-term radiological hazards."*

These conclusions diminished the P-T research for a few years as national and international agencies withheld fundings, which also is reflected in the decreasing number of publications (see 2.2). Work that continued appears to have been performed on a small scale. The story is not completely true, because the decrease in publications after 1982 is also based on political decisions. In the USA it was decided that all work concerning reprocessing should be classified. This decreased the American contribution and it seems like other countries became more restrictive with their contributions after some years.

2.1.2 Background to current P-T studies.

Advances have been made during since 1982 in demonstrating some of the engineering practicality and economical feasibility to transmute actinides in nuclear reactors. In the French nuclear program, recycling of plutonium in LWR's as MOX-fuel started on regular basis 1990. Liquid Metal Fast Breeder Reactors (LMFBR's) has been operated in Russia for some time and operating experiences has also been obtained by France, UK and USA. An experiment called SUPERFACT, using both homogeneous and heterogeneous fuels of Minor Actinides (MA) in the French PHENIX reactor is in progress, see 4.3. These experiences with both LWR's and LMFBR's have confirmed the technical feasibility of transmutation of actinides in nuclear reactors.

In 1987 the Atomic Energy Commission in Japan initiated the program "Long Term Program for Research and Development on Nuclide Partitioning and Transmutation Technology" [AEC 88]. In the program, also called OMEGA (Options Making Extra Gain from Actinides), the transmutation device is considered to be either an accelerator or a nuclear reactor. In the USA several different strategies for transmutation of nuclear wastes were presented around 1990. Among them, ATW (Accelerator Transmutation of Waste) was introduced by the Los Alamos National Laboratory. The idea with the new P-T proposals is to use an accelerator to produce neutrons.

Several international organizations are involved in P-T research work. The Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA) initiated a series of information exchange workshops at the request of the Japanese OMEGA programme and promoted coordination among OECD member states. The Commission of the European Communities (CEC) have initiated international and community programmes on the technical and scientific aspects of P-T within the community. In 1991 IAEA was recommended by an advisory group to coordinate the P-T efforts between countries outside international organizations like OECD/NEA and CEC.

The reemerged international interest in P-T has been large, which is reflected in the numerous meetings that have been arranged during the last years, as listed below:

- *Workshop on Partitioning and Transmutation of Minor Actinides, Nuclear Science and Technology, Commission of the European Communities (CEC), Karlsruhe, Germany, 16-18 October 1989, [Koc 91b].*

- *Information Exchange Meeting on Actinide and Fission Product Separation and Transmutation, OECD/NEA-Japan Atomic Energy Research Institute (JAERI), Mito, Japan, 6-8 November 1990, [OECD 91].*

- *Specialist Meeting on Accelerator-Driven Transmutation Technology for Radwaste and other Applications, Swedish National Board for Spent Nuclear Fuel (SKN) - Los Alamos National Laboratory (LANL), Saltsjöbaden, Stockholm, Sweden, 24-28 June 1991, [SKN 91].*

- *Workshop on Nuclear Transmutation of Long-Lived Nuclear Power Radiowastes, Institute of Nuclear Power Engineering - Obninsk, Institute of Theoretical and Experimental Physics - Moscow, Obninsk, Russia, 1-5 July 1991, [INPE 91].*

- *IAEA Advisory Group Meeting on Partitioning and Transmutation of Actinides and Selected Fission Products from HLW, IAEA, Vienna, Austria, 21-24 October 1991, [IAEA 91].*

- *OECD/NEA Workshop on Partitioning of Actinides and Fission Products, OECD/NEA-JAERI, Mito and the Tokai Research Establishment, Japan, 19-21 November 1991, [OECD 91b].*

- *Symposium on Separation Technology and Transmutation Systems (STATS), National Research Council, Washington, USA, 13-14 January 1992, [NRC 92].*

- *OECD/NEA Specialist's Meeting on Accelerator-Based Transmutation, OECD/NEA-Paul-Scherrer Institute (PSI), Würenlingen/Villingen, Switzerland, 24-26 March 1992, [OECD 92].*

- *Partitioning and Transmutation of Minor Actinides and Fission products: 2nd Working group meeting on Targets and Fuels, Commission of the European Communities (CEC), Karlsruhe, Germany, 23-24 june 1992.*

2.1.3 Literature survey of P-T.

A literature survey in the field of P-T has been performed. The bibliography is presented as a separate report [Skå 92]. The survey was performed by using the Chemical Abstract (CA) data base and known conference proceedings. The material that was presented during meetings from 1990 to 1992 is not always published in proceedings. However, the available literature is included in the publication list. The bibliography is divided year by year and cover the time from 1967 to april 1992. The publications have been coarsely divided by their content in five different groups, namely; (1) partitioning, (2) transmutation in nuclear reactors, (3) transmutation by accelerator driven devices, (4) transmutation by fusion sources and (5) general aspects of P-T, for example radiological hazard.

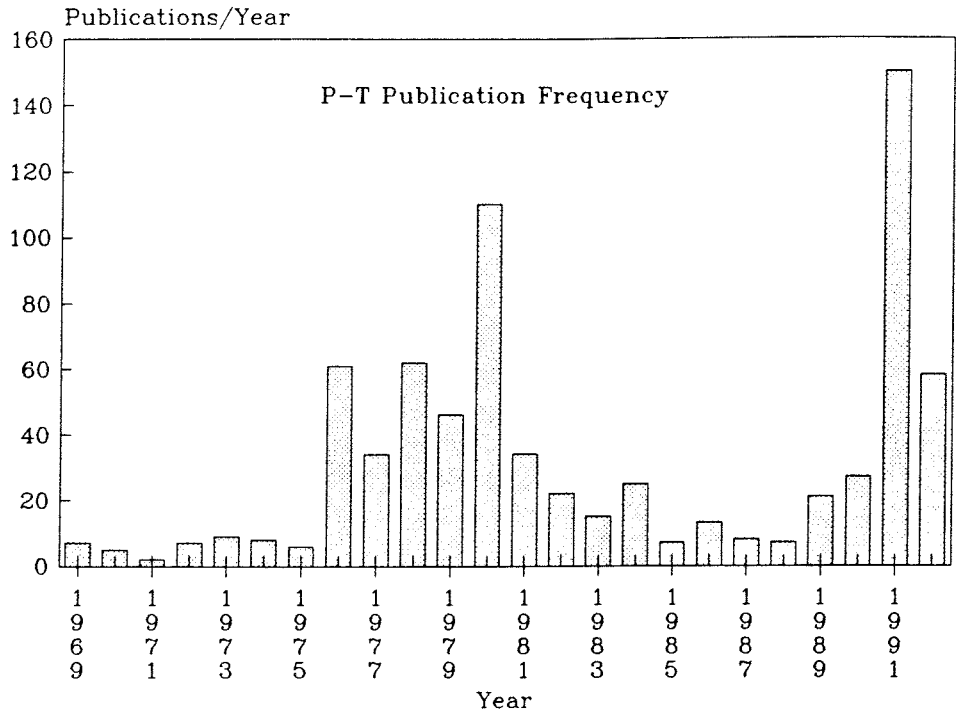

Fig. 2.1. The annual distribution of publications on P-T from 1968 to 1992.

In total the bibliography includes about 800 publications. The annually distribution of the publications is shown in figure 2.1.

The intense period of research from 1976 to 1982 is well reflected in the number of publications. The sharp decrease in publications after 1982 is due to the unfavorable conclusions about P-T in 1982 and some political decisions, as mentioned before (see 2.1.1). The number of publication from 1991 and 1992 is not complete. There are still proceedings not published and P-T will be topics at several meetings and conferences during the rest of 1992.

2.2 WHAT HAS CHANGED SINCE EARLIER ASSESSMENTS?

The earlier P-T studies in the late 1970s and early 1980s were concerned with transmutation of actinides in nuclear reactors. The general conclusion was then, "Since the long-term hazards are already low, there is little incentive to reduce them further by P-T". The reemerged interest in P-T has been motivated by different individuals and organizations who claim that changes of factors involved in the earlier conclusions, combined with some entirely new factors, justify reexamination of the P-T option. Some of these changes are:

- The radiotoxicity of actinides has been re-considered according to the new guidelines from the International Commission on Radiation Protection (ICRP). This may increase the incentive for transmutation of actinides.

- By using accelerator based spallation neutron sources instead of fission reactors it is possible to have high neutron fluxes purely for transmutation purposes and a low formation of new long lived radionuclides. This have several advantages, one of which can be exemplified by the transmutation of ^{237}Np that can be achieved with a net gain of neutrons, due to the high fission cross section of the short lived ^{238}Np.

- Not only actinides but also fission products that can cause a long-term potential hazard are considered for transmutation in the new P-T proposals.

- New technological developments of reprocessing technology and advances in robotics can also increase the effectiveness in partitioning.

3 GENERAL CONSIDERATIONS

Transmutation of minor actinides and long lived fission products is only feasible after separation from the bulk constituents of the spent nuclear fuel, i.e. uranium, plutonium and most of the short lived or stable fission products. Hence, reprocessing of the spent fuel must be the first step in any P-T strategy. Two options exist: i) conventional PUREX reprocessing followed by an add-on chemical process to recover those elements that

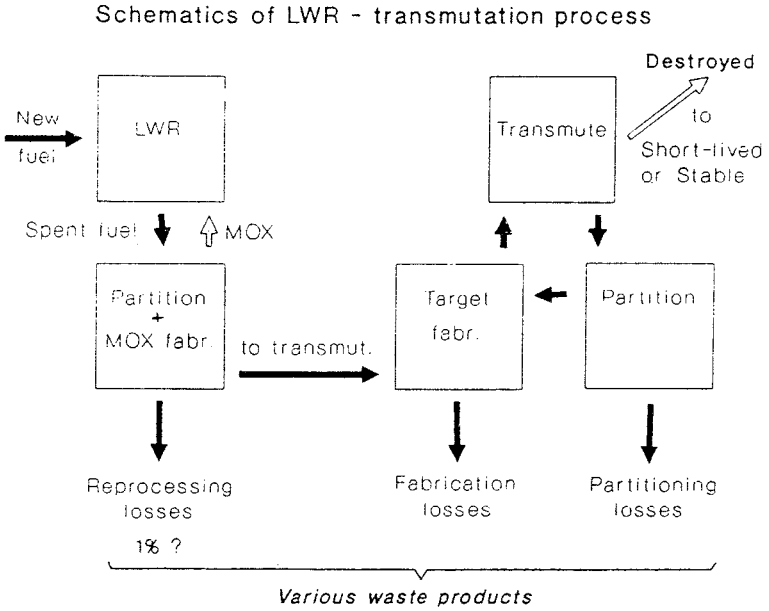

Fig. 3.1. Schematic of LWR fuel cycle with transmutation of selected waste products.

contain isotopes which should be transmuted from the HLLW and other waste streams, ii) an upgraded process where separation of uranium and plutonium is combined with recovery of elements to be transmuted. Recycle of recovered uranium and plutonium as reactor fuel has to be considered as a normal part of any P-T strategy, see figure 3.1.

3.1 TRANSMUTATION

Due to the large difference in potential hazard index between actinides and fission products, the primary goal of a P-T process must be to transmute, i.e. fission, the actinides, including U and Pu. Only when this can be accomplished with very high efficiency (probably ≥ 99.99 % for the transuranium elements) will the remaining long lived fission products have any significance for the potential hazard of the radioactive waste after 1000 years storage. Then it may be of interest to reduce the remaining potential hazard further by also transmuting long lived fission and activation products.

The easiest way to fission actinides is by irradiation with neutrons. Thermal neutrons have high reaction cross sections with the fissile actinide isotopes (mostly odd neutron numbers), but do not fission the other actinide isotopes (mostly even neutron numbers). The latter capture neutrons by the (n,γ)-reaction, increasing their mass number by one unit (n-capture will continue until the isotope formed is either fissioned or undergoes β^--decay in which case an isotope of the next higher element is formed, the so called s-process). Energetic neutrons will fission all of the actinide isotopes, but the reaction cross sections are much smaller than for thermal neutron fission of the fissile isotopes. The build-up of higher actinides by the s-process is thus much smaller with energetic neutrons than with thermal neutrons.

Fission of the heavier nuclides by irradiation with energetic γ-rays is possible (photo-fission), but the reaction cross sections are normally much smaller than for neutrons. Irradiation with γ-rays of suitable energy can also induce (γ,n), (γ,p) and (γ,2n) reactions. Of these, the (γ,n) reaction usually has the largest cross section, in the range 100-300 mbarn.

Finally, fission is also possible to induce by irradiation with high energy ions. However, particle energies of about 10 MeV/u is needed to penetrate the coulomb barrier and the total reaction cross sections will at most be a few barn. The use of an U-target, instead of the normally discussed Pb- or Bi-targets, in accelerator driven transmuter systems would lead to fission or spallation of the uranium by high energy p^+-bombardment.

In principle most of the above mentioned reactions, except fission, will also work on isotopes of the lighter elements. Transmutation of activation and fission products can be achieved by irradiation with slow neutrons leading to the s-process. The reaction cross sections are mostly in the range 0.1-10 barn for the long lived fission product isotopes. This is complicated by the presence of stable isotopes of the same element as such isotopes also will capture neutrons as part of the s-process. The products formed are either stable or have short to moderate half lives in most cases.

3.1.1 Irradiation facilities

As nuclear transmutation is accomplished by irradiation with neutrons, γ-rays or charged particles, devices for production of these in sufficient quantities and at useful rates are needed. Neutron spectra for possible transmutation devices are shown in figure 3.2. Current and planned development work is described in section 4.

3.1.1.1 Thermal reactors

Thermal fission reactors are characterized by a comparatively soft n-spectrum, see figure 3.2. This means that fissile isotopes will have high

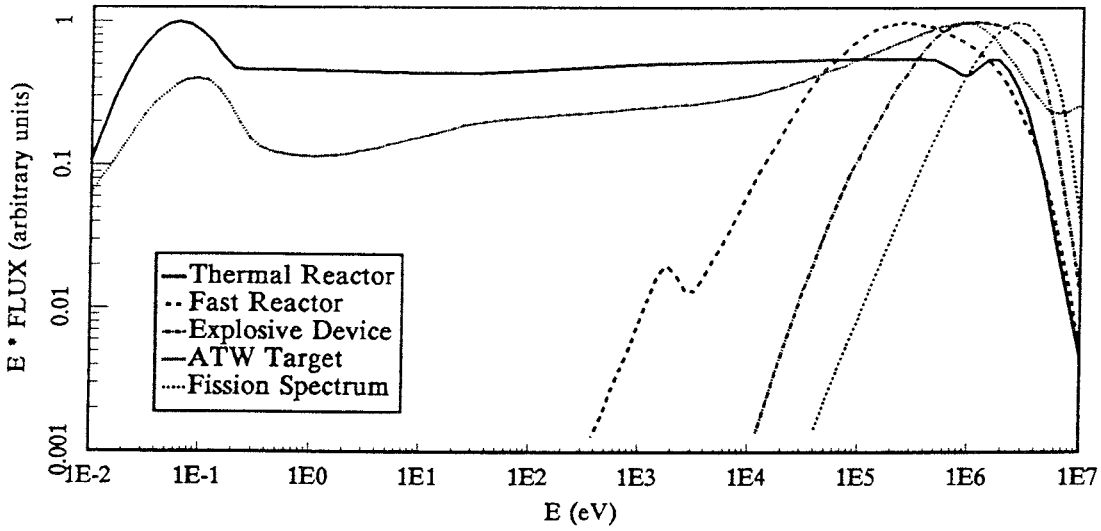

Fig. 3.2. **Neutron spectra for som potential transmutation devices. The abscissa shows n-flux times energy on an arbitrary scale.**

reaction cross sections, in the order of 100-1000 barn. At the same time the build-up of higher actinides by the s-process is large. One year of operation may yield an exposure of about 10^{21} n/cm^2. Light-water moderated power reactors have harder n-spectra than e.g. heavy-water or graphite moderated reactors. Regardless of type these reactors are mostly useful for transmutation of uranium and plutonium. However, replacement of burnable absorbers, e.g. Gd, by Np+Am in a suitable diluting matrix, e.g. MgO, will lead to a slow but significant burn-up of these elements. Contrary to what was once assumed, transmutation of self-generated minor actinides in thermal power reactors is possible although the halving time will be rather long, see e.g. [Cam 77, Sow 77, And 78]. In spite of this the total inventory of minor actinides in an equilibrium cycle will not be very large compared to the total amount of transuranium elements normally present in the fuel [Cam 77].

The envisaged major inconveniences in using thermal power reactors as transmutation devices have been summarized by Hage and Schmidt [Hag 77b]. They give the following negative points:

- The reactivity loss of the reactor due to actinide target elements must be compensated for by a small increase in fuel enrichment.

- Additional forced cooling of target rods will be needed after reactor shutdown and during any fuel element manipulation.

- The high build-up of spontaneous fission neutron sources in the target rods will require additional shielding and special measures during refueling operations and also increases the shut-down n-flux in the reactor.

3.1.1.2 Fast reactors

Fast reactors have by definition a much harder n-spectrum than thermal reactors and from the transmutation point of view, the ideal fast reactor should have a nearly unmodified fission n-spectrum, see figure 3.2. The LMFBR type has, however, a considerable fraction of the n-flux at lower energy due to moderation by the sodium coolant, which has a slowing down power of about 1% of that of graphite. This makes that type of breeder reactor somewhat less suitable as a minor-actinide burner. Possible yearly doses for transmutation purposes are of the order of 10^{22} n/cm^2. The ideal fast reactor for transmutation purposes should have as hard a n-spectrum as possible and a very high n-flux, somewhat contradictory requirements.

Fast reactors are more efficient than thermal reactors as far as the long term hazard reduction is concerned and can be considered as transmutation devices in a mixed LWR-LMFBR power system [Hag 77b].

3.1.1.3 Nuclear explosive devices

From a pure neutronics point of view, a nuclear explosive fission device is the ideal transmuter with its extremely high n-flux (maximum $\approx 10^{31}$ n cm^{-2} s^{-1}) and high average n-energy, see figure 3.2. Material present near the origin will react by fission (if fissible) or by multiple n-capture, the r-process. The achievable neutron dose is extremely high, $> 10^{25}$ n/cm^2 near the center. This corresponds to > 100 years of exposure in a high flux nuclear reactor and > 1000 years in a nuclear power reactor. In practice, these favorable conditions are completely offset by the problems arising from the explosive nature of such an event and by the large amount of unconsumed fissionable material remaining after the explosion. Efficient containment of the explosion permitting high yield recovery of untransmuted target material and unused fissile material for recycle presents a formidable engineering problem. Perhaps future development of almost pure fusion devices and a practical containment system could make them useful for some special transmutation purposes.

3.1.1.4 Accelerators

The development of high current, high energy proton accelerators has opened the possibility to use them as high flux n-sources. Currents in the order of hundreds of mA of protons at GeV energy makes it possible to generate immense amounts of neutrons in a spallation target, in some tentative designs more than 0.1 gram neutrons per second, see section 4.4.2. The calculated shape of the n-spectrum on the surface of the

proposed ATW target container according to [Ire 91] is shown in figure 3.2. A combination of a subcritical transmutation system and accelerator driven neutron source may become self-supporting with electrical energy, or even produce excess electrical energy, see section 4 for current proposals.

Accelerators can also be used to produce energetic γ-rays for transmutation of actinides. However, today this seems to be much less attractive than n-production.

3.1.1.5 Fusions reactors

If and when fusion reactors could be designed, built and operated they may offer large n-fluxes at high n-energy (e.g. 14 MeV) and could be useful for nuclear transmutation.

3.1.2 Transmutation reactions

The basis for transmutation are nuclear reactions induced by a projectile, usually a neutron, that change the irradiated nuclide in a desirable way. The nuclear mass and charge of light elements (below bismuth) can only be changed in small steps for relatively small projectile energies. Heavy elements like the actinides can fission as well.

Even in the case of high energy projectiles, e.g. 800 MeV p^+, most of the nuclear reactions in a very thick target will be induced by low energy secondary particles, mainly neutrons, see figure 3.3. The reason for this is twofold. First of all each nuclear reaction induced by the primary high energy projectile will lead to the formation of many secondary particles, most of which are neutrons of much lower energy than the primary projectile. Secondly, the total reaction cross section normally increases when the projectile energy decreases. Hence, low energy neutrons have a higher probability to react than high energy neutrons. There is no lower energy limit for n-induced exothermal reactions. Charged particles must have energies at least near the coulomb barrier in order to react with target nuclei. For this reason low energy charged particles cannot effectively react further, they have a cut-off energy.

Irradiation with high energy γ-rays is rather inefficient because of the low cross sections (100-300 mb) which implies that an efficient use of the incident γ-radiation requires a very large amount of target material. Hence, the inventory of target material must always be comparatively large. The nuclear reaction which is most interesting is photo-fission.
Regardless of the projectile and reaction used, only a limited amount of material can be transmuted in a single irradiation because; i) the concentration of target nuclei decreases with time and ii) the concentration of product nuclei increases with time. Hence, more and more of the projectiles will be used to transmute the reaction products instead of the target. As long as only a minor part of the particles are

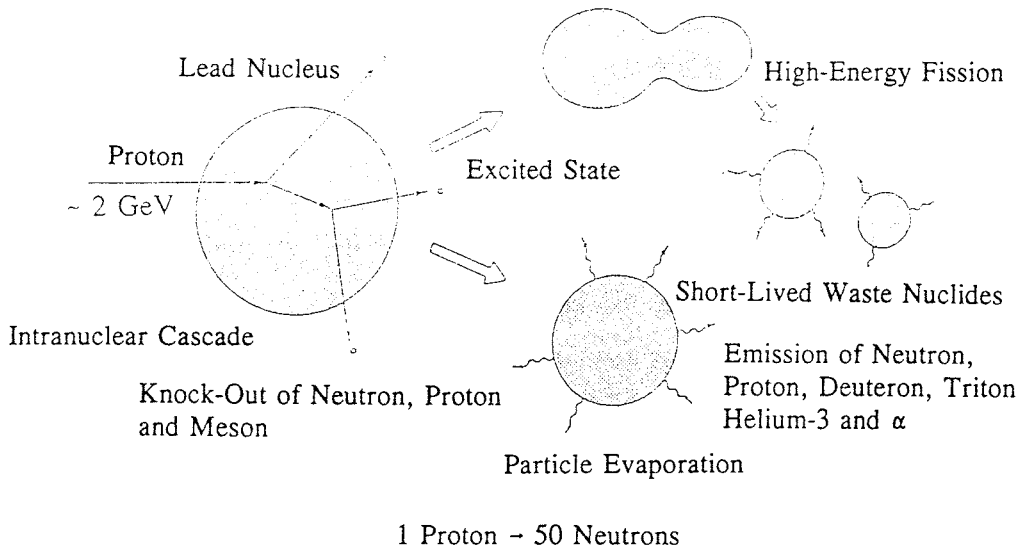

Fig. 3.3. Nuclear reactions induced by high energy protons.

consumed in the target, the amount of target nuclei transmuted per unit time by irradiation with photons, neutrons, charged particles or by radioactive decay, dN/dt atoms/s, is given by

$$-dN/dt = (\phi\ \sigma + \lambda)\ N_0\ e^{-(\phi\sigma+\lambda)t} \qquad (3.1)$$

where N_0, ϕ, σ and λ are the initial number of target nuclei, the projectile flux, the effective reaction cross section, and decay constant, respectively. As can be seen, the reaction rate decreases with time, t. The remaining amount of substance, N, is given by

$$N = N_0\ e^{-(\phi\sigma+\lambda)t} \qquad (3.2)$$

The untransmuted substance can thus be regarded as having an effective half life of $\ln(2)/(\phi\sigma+\lambda)$ seconds. To effectively shorten the radioactive decay, the product $\phi\sigma$ must be much larger than the decay constant, λ. An equation similar to 3.2 describes the amount of product present, N_p.

$$N_p = \{(\phi\sigma+\lambda)N_0/[(\phi\sigma_p+\lambda_p)-(\phi\sigma+\lambda)]\}\{e^{-(\phi\sigma+\lambda)t}-e^{-(\phi\sigma_p+\lambda_p)t}\} \qquad (3.3)$$

where subscript p denotes data for the transmutation product.

3.1.2.1 Reactions induced by photons

Reactions between low- and medium-energy photons (γ-rays) and atomic nuclei are dominated by a so called giant resonance, a few MeV wide. The energy of this resonance peak maximum varies smoothly with the mass number, approximately according to equation 3.4

$$E_{peak} \approx (48 + 0.139\ A) / A^{1/3} \quad \text{(MeV)} \tag{3.4}$$

The integrated cross section below the peak, $\int\sigma(E)\,dE$, is given to a good approximation by

$$\int\sigma(E)\,dE = 0.06\ N\,Z / A \ \text{(MeV barn)} \tag{3.5}$$

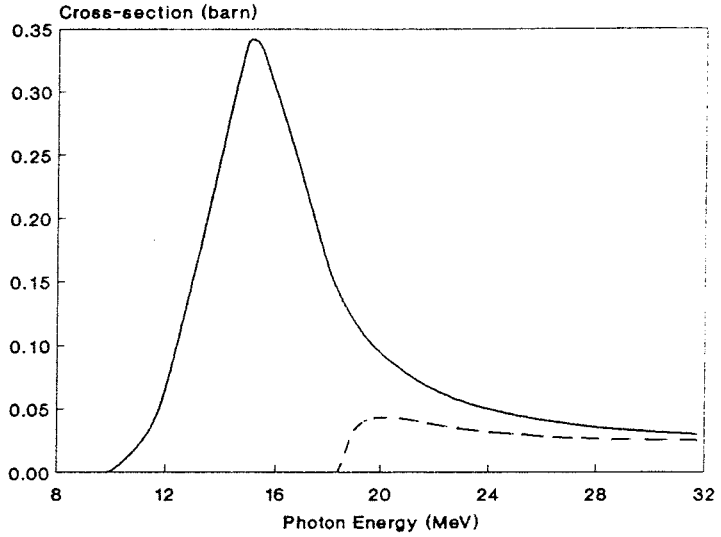

Fig. 3.4. Photo cross section for ^{141}Pr leading to emission of 1 (–) or 2 (--) neutrons [Lie 69].

Typical reaction cross sections for γ-induced emission of neutrons are shown in figure 3.4. The reactions induced are usually, in order of decreasing importance, (γ,n), (γ,p), (γ,2n), (γ,pn), etc and for the heavy elements also (γ,f) [Fri 81].

3.1.2.2 Reactions induced by neutrons

Reactions between neutrons and all except the lightest elements are (n,γ), (n,2n) and for the heaviest elements also (n,f). At low neutron energies, the cross sections for the (n,γ) and (n,f) reactions are inversely proportional to the square root of the neutron energy. For the low energy range and using a neutron energy, E, in eV we can express the cross section at energy E by

$$\sigma(E) \approx \sigma_{2200}\ (E/0.025)^{-1/2} \tag{3.6}$$

where σ_{2200} is the cross section for neutrons with a velocity of 2200 m/s.

At intermediate neutron energies most nuclei exhibit a complex set of resonances. Finally, at the highest n-energies there is often a small increase in reaction cross sections, see figure 3.5 for an example. The complicated behavior of the reaction cross section with neutron energy

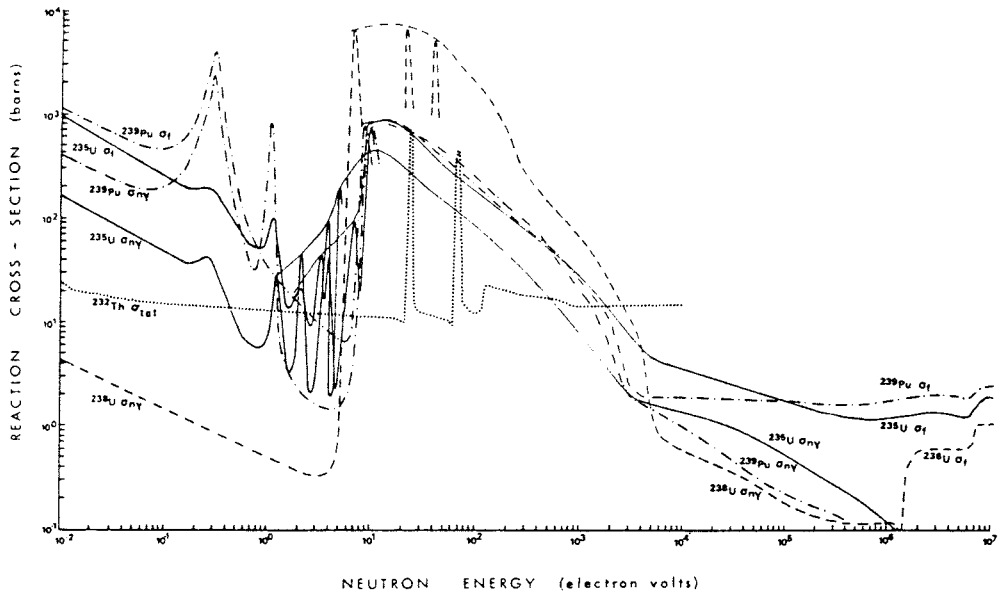

Fig. 3.5. **Reaction cross sections for scattering, neutron capture, fission and total reaction as function of n-energy. Only envelopes of peaks in the 1-5000 eV region are given [Cho 80].**

makes the total cross section for any specific reaction very dependent on the actual n-spectrum. Even for simple calculations (e.g. using equations 3.1 to 3.3) it is necessary to use properly n-spectrum averaged reaction cross sections according to equation 3.7.

$$\langle\sigma\rangle = \int\sigma(E)\,\phi(E)\,dE \,/\, \int\phi(E)\,dE \qquad (3.7)$$

Tab. 3.1. **Some actinide cross sections for 2200 m/s neutrons and avaraged for a typical fast reactor neutron spectrum**

Material	2200 m/s			Typical Fast Reactor Spectrum		
	$\langle\sigma_{n,f}\rangle$	$\langle\sigma_{n,\gamma}\rangle$	$\bar{\nu}$	$\langle\sigma_{n,f}\rangle$	$\langle\sigma_{n,\gamma}\rangle$	$\bar{\nu}$
^{238}U	0.0	2.73	0.0	0.047	0.27	2.76
^{237}Np	0.0	172	0.0	0.34	1.87	2.87
^{239}Pu	742	271	2.88	1.82	0.51	2.94
^{241}Am	3.80	832	3.22	0.45	1.49	3.29
^{242m}Am	6600	700	3.26	3.33	0.10	3.32
^{243}Am	0.0	159	-	0.19	1.66	3.33
^{242}Cm	?	16	3.32	1.26	0.50	3.38
^{243}Cm	600	225	3.43	3.14	0.10	3.49
^{244}Cm	1.2	13.9	3.49	0.45	0.57	3.40
^{245}Cm	2018	340	3.83	2.92	0.09	3.89

Some data typical for fast reactors are given in table 3.1 together with the 2200 m/s values.

3.1.2.3 Reactions induced by charged particles

The energetics of reactions with charged particles differ from those of photons and neutrons because the mutual repulsion of like electrical charges, the coulomb barrier. This may lead to a deflection of the projectile before it hits the target nucleus if the initial kinetic energy is too small or the impact not central enough, see figure 3.6.

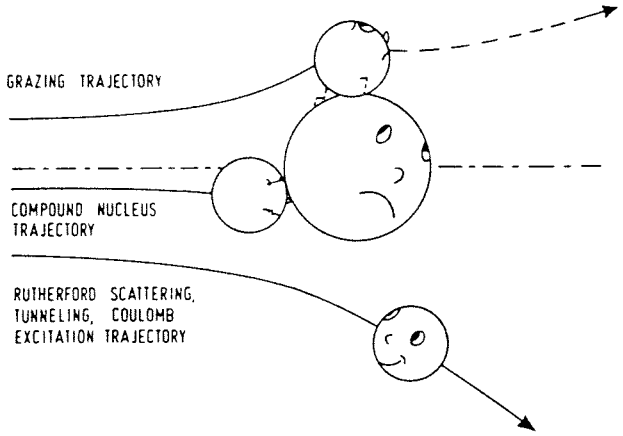

Fig. 3.6. Schematic representation of three different types of charged ion/nucleus interaction for different impact parameters and kinetic energies above the barrier [Kau 60].

The minimum projectile energy, E_{CB}, for different projectile - target combinations can be computed from equation 3.8.

$$E_{CB} = 1.109\ [(A_p + A_t)/A_t]\ Z_p\ Z_t\ /\ (A_p^{1/3} + A_t^{1/3})\ (\text{MeV}) \qquad (3.8)$$

where subscripts p and t refer to projectile and target, respectively. In practice a higher energy is needed to achieve useful cross sections by bringing non-central collisions into play.

At low energies (X,n) reactions dominate for all except the heaviest elements, but when the energy increases further above the barrier reactions with release of an increasing number of neutrons become dominant and even (X,p) and (X,α) reactions become important, see figure 3.7. Heavy elements will also fission, the (X,f) reaction.

3.2 PARTITIONING

Because the transmutation rate decreases exponentially with irradiation time, a 100% transmutation in a single irradiation is impossible, see eqn. 3.1. Furthermore, if not very short lived, the transmutation product will increase in amount with time and also transmute, usually an undesirable effect. Untransmuted material must thus be separated from transmutation

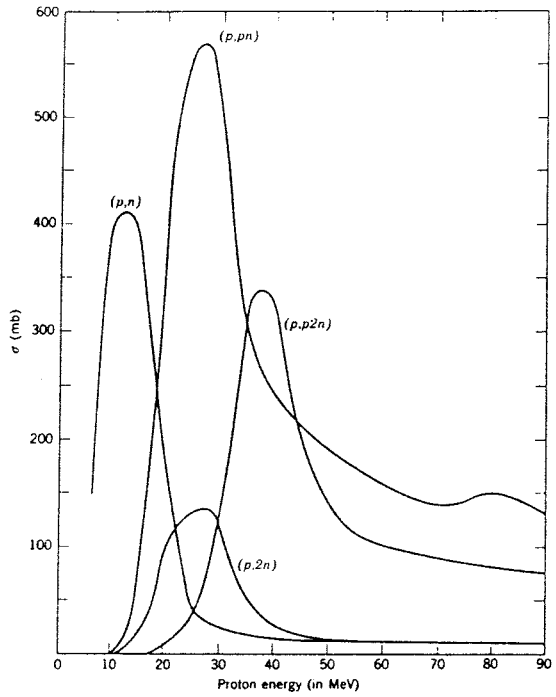

Fig. 3.7. Partial reaction cross sections for proton-induced reactions with ^{63}Cu [Mea 53].

products and recycled for further irradiation.

This offers special problems in case long lived activation or fission products are transmuted by (n,γ)-reactions to stable isotopes of the same element, e.g. $^{79}Se(n,\gamma)^{80}Se$(stable), or to radioactive isotopes with appreciable half lives, e.g. $^{94}Nb(n,\gamma)^{95}Nb(t_{1/2}$ 35d) because normal chemical separation procedures will only separate different chemical elements. In the first case, no simple chemical separation procedure exists which can remove the product from untransmuted target material. In the second case, irradiated material must be stored before the chemical separation until most of the active product has decayed, i.e. formed another element. This will give a large inventory of the target material at the transmutation facility.

Transmutation of actinides by (n,f)-reactions makes a chemical removal of the transmutation products fairly easy as the products have a chemistry which differs from that of the target element(s). However, also in this case it may be favorable to include a pre-separation intermediate storage in order to let most of the short lived fission products decay as this would lessen the radiolytic effects during separation.

3.2.1 Aqueous processing

Aqueous separation technology is by far more developed than other methods. This means that a considerable knowledge exists about

chemical and engineering difficulties. This should not be taken as an excuse to turn to other, less well known, methods, e.g. pyroprocessing, without a serious consideration of the pros and cons for aqueous systems. The term aqueous separation may be slightly misleading as many of the separations classified as aqueous use in fact both aqueous and organic solutions and/or organic reagents. The usually preferred technique is solvent extraction because it offers high separation factors, continuous operation, and avoids the handling of radioactive solids. The only aqueous separation process operated on a large industrial scale today is the PUREX process (with some variants) used for reprocessing spent nuclear fuel. As this, or a somewhat similar, process must be the initial stage of any transmutation scheme it will be discussed shortly before the special process(es) needed in the transmutation cycle itself.

3.2.1.1 Reprocessing of spent nuclear fuel

The PUREX process (Plutonium Uranium Redox EXtraction) was designed to recover uranium and plutonium from irradiated nuclear fuel. It has replaced all earlier separation processes in this field. This process uses an organic solvent consisting of a solution of tri-n-butyl phosphate, TBP, (normally about 30% v/v) in an aliphatic diluent. At least three variants of the PUREX process have been developed and used on an industrial scale, see figure 3.8. Of these, only two are currently in industrial use; i) early split for reprocessing of highly enriched uranium from military reactors in the U.S. (at INEL), where the plutonium formed is negligible because the fuel contains very little ^{238}U, ii)late split 2 is the variant preferred today for reprocessing unenriched or slightly enriched uranium from power reactors.

Currently the emphasis in reprocessing operations is on production of very pure uranium and plutonium. This leads to operation of the first separation at the highest possible uranium loading of the organic solvent in order to suppress the extraction of some fission products. As a consequence, there is a loss of a small amount of uranium (usually < 0.5% of the amount in the feed) and some plutonium (also usually < 0.5%) to the HLLW stream. Further losses are caused by incomplete dissolution of the chopped fuel and by solvent clean-up operations from which concentrated waste may be added to the HLLW stream.

In the late split 2 variant of the PUREX process practically all of the trans-plutonium elements end up in the HLLW stream. Neptunium poses a special problem as it has a tendency to split between many process streams, see e.g. [Mil 91b]. However, special measures can be taken in the future to force almost all neptunium into a single process stream from which it may be recovered.

Regular reprocessing of core-fuel from breeder reactors will in the future require an improved recovery of plutonium as else the total amount lost to waste streams will be unacceptable. Hence, research aiming at an

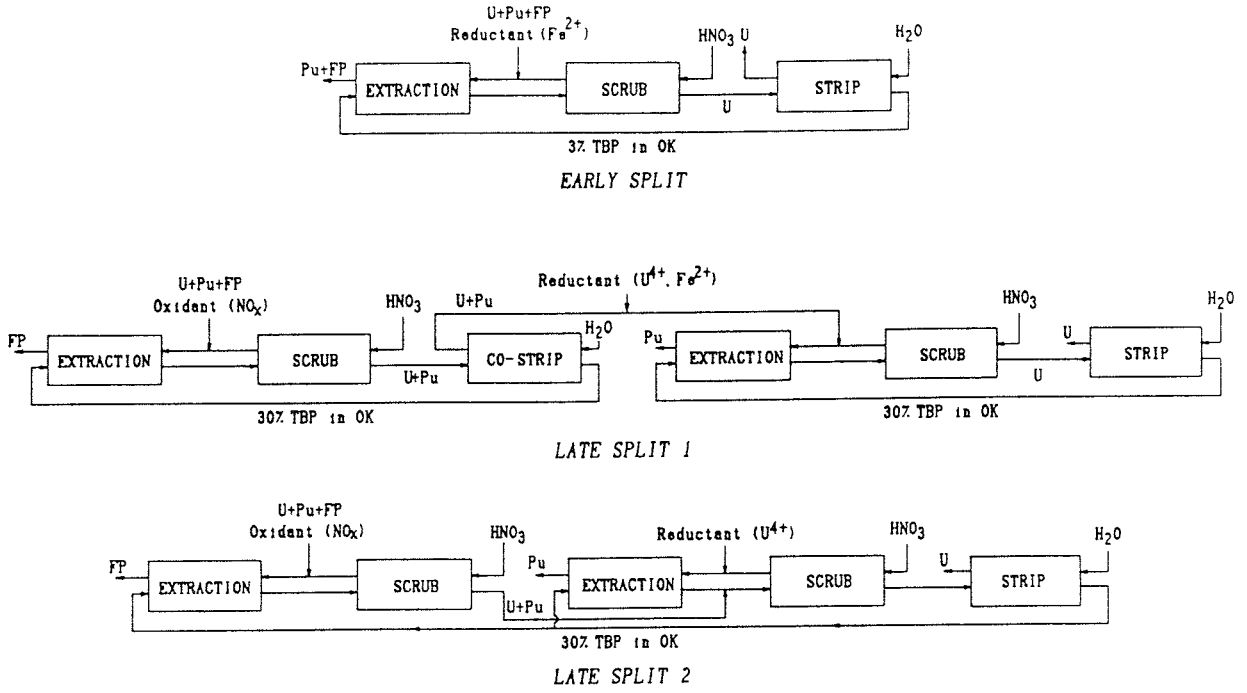

Fig. 3.8. Main PUREX process variants for reprocessing of nuclear fuels.

improved recovery of plutonium is well under way. At the same time, options permitting a good recovery of neptunium and the trans-plutonium elements are being investigated.

Hence, two possible alternatives exist for recovery of actinides in the reprocessing operation; i) add-on processes for recovery of neptunium from one of the process streams and for recovery of neptunium and the transplutonium elements from the normal HLLW stream, ii) a new process flow-sheet, probably using another reagent than TBP, which permits a high recovery of all actinides. Transmutation of the minor actinides (Np, Am, Cm, Bk and Cf) is only meaningful if they are recovered in high yield during fuel reprocessing. One possible add-on process, the CTH-process, was developed and tested on old HLLW during the late 1970ies and early 1980ies [Per 83], [Sva 84], [Lil 91], [Lil 92], see figure 3.9.

Transmutation of long lived fission products would require additional separation processes to recover the proper elements, containing the long lived isotopes, from the HLLW stream and probably also from other waste streams. Such processes have been developed and used in the U.S. for recovery of strontium, technetium and cesium from old military waste, mainly to produce heat- and radiation-sources containing ^{90}Sr or ^{137}Cs. However, a considerable research and development work is needed until industrial processes are available that can recover all long lived material in high yield from reprocessing wastes.

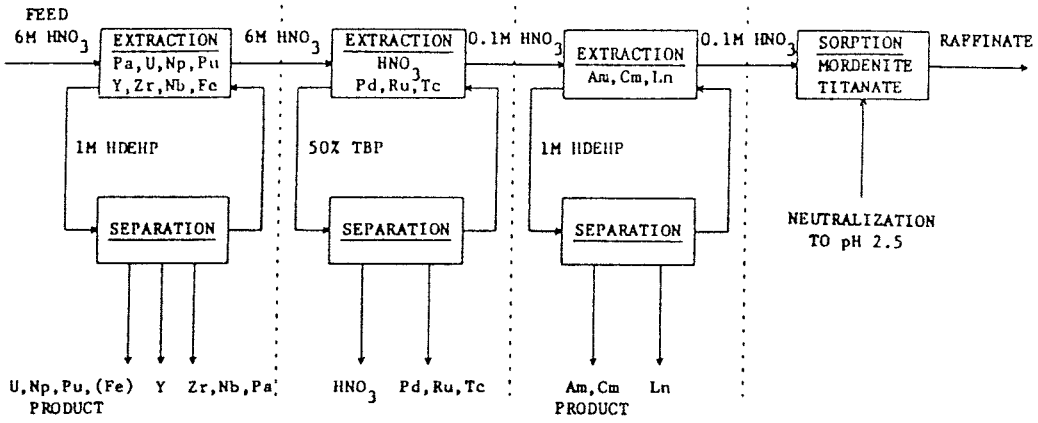

Fig. 3.9. **General flow-sheet of the CTH-process for recovery of actinides and technetium from PUREX HLLW.**

3.2.1.2 Processing of transmutation targets

Several different separation processes will be needed to recover untransmuted material in high yield from irradiated transmutation targets. Because the target material has to be recycled many times to achieve a high degree of transmutation the tolerable losses are very small, see 3.2.3. One of the problems that have to be solved is the development of sufficiently sensitive measurement methods for routine supervision of losses to all waste streams.

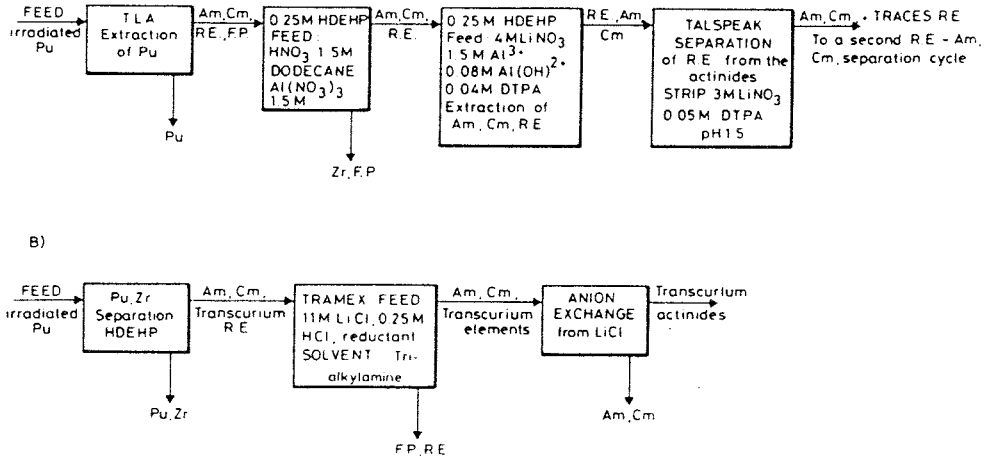

Fig. 3.10. **Examples of processes used for recovery of trans-plutonium elements from irradiated Pu-targets [Ryd 92].**

Actinides can probably be separated in high yield from fission products comparatively easy. Figure 3.10 shows two process variants used for recovery of actinides from irradiated plutonium targets. Several other

separation processes for actinides have been suggested, but will probably need further development and testing before they can be applied on an industrial scale in a transmutation flow-sheet, see e.g. references [Sea 90] and [Ryd 92] for further details.

High yield separation of transmutation products from targets containing long lived activation- and fission-products is probably more complicated, especially if the transmutation yields stable isotopes, or radioactive isotopes with intermediate half lives, of the element to which the isotope to be transmuted belongs. In the first case, isotope separation will be necessary. In the second case, pre-separation intermediate storage may be necessary. Processes for high yield recovery of some elements containing long lived isotopes, e.g. technetium or iodine, are available and may only need some further development. In most other cases, however, new processes have to be developed.

Finally, processes for conversion of separated material to suitable targets are needed. In case of liquid targets this would present only minor problems. However, when solid targets are used this involves production and handling of radioactive solids with accompanying scrap and dust generation giving losses to waste streams. If such losses are too high, additional recovery processes will be needed.

3.2.2 Pyroprocessing

Pyroprocessing involves two different types of processes; 1) metallurgy with addition of slag forming material and separation of metal from slag, 2) two-phase extraction of undesirable elements by contacting molten

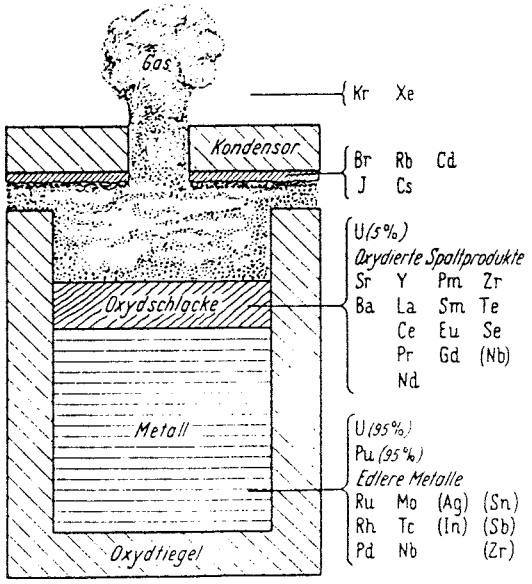

Fig. 3.11. Example of a reprocessing operation using a pyrochemical method [Car 59].

metal with a molten salt followed by separation of unreacted metal. The first kind of process has been used for reprocessing of metallic fuel from the EBR-II reactor, see figure 3.10 [Lin 61]. The latter kind of process has been used for separation of ingrown ^{241}Am from plutonium scrap metal [Kni 81]. In both cases, the separated waste products normally contains an unacceptable amount of target material as impurity (often in the % range). Hence, it is very likely that it will have to undergo further purification by aqueous methods. The major driving force towards pyroprocessing has been that it offers a cheaper reprocessing route because it avoids normal conversion from solution to solid and permits simpler and cheaper fuel fabrication. However, this is usually achieved at the expense of high separation yields, see e.g. yield of U in figure 3.11.

3.2.3 Recycling and losses

The unavoidable incomplete transmutation makes separation and recycling important in any transmutation scheme. The less material that can be transmuted in a single irradiation, the more cycles are needed to destroy a given amount. Each time the material passes through one cycle there will be some loss of untransmuted material to various waste streams.

Assume a flow-sheet according to figure 3.1 and consider the reprocessing and transmutation cycles. If the lost fraction in reprocessing operations is designated by r, the transmuted fraction per irradiation by ε, the lost fraction in a single pass through the separation process by α and the lost fraction in the target fabrication step by δ we can obtain the following equation for the total lost fraction, F, which is valid for an infinite number of recycles

$$F = r + (1 - r)(\alpha + \delta - \alpha\delta)/\{1 - (1 - \alpha)(1 - \delta)(1 - \varepsilon)\} \qquad (3.9)$$

For the small values of α and δ ($\ll 1$) required by an efficient transmutation of target material equation 3.9 can be simplified to

$$(\alpha + \delta) \approx \varepsilon/\{\varepsilon + (1 - r)/(F - r)\} \qquad (3.9a)$$

This form permits a direct estimation of the tolerable combined losses in target material recycling for a given single cycle transmuted fraction, ε, known fractional loss in fuel reprocessing, r, and given total fraction of initial target material lost to various wastes, F.

If we assume a loss to wastes during the initial reprocessing operation of 0.5% (r=0.005) and a single cycle transmuted fraction of 5% (ε=0.05), equation 3.9a shows that the yields in target recovery and fabrication operations have to be high ($\alpha+\delta$=0.00475, i.e. combined yield=99.525%) in order to achieve even the moderate reduction of the total amount disposed of as wastes by a factor of 10 (F=0.1), compared to a direct disposal operation.

Because of the inevitable limit on the achievable combined recycling yield, reprocessing losses should be decreased as far as possible and target single cycle transmutation be as high as practicable in order to obtain an efficient transmutation operation.

3.2.4 P-T Inventory

A knowledge of the expected total inventory of material in the transmutation cycle is important for an assessment of the hazards involved in a transmutation scenario.

Using the same symbols as in equation 3.9 and denoting the amount of a specific substance fed to the transmutation cycle per unit time by m, the cycle time for transmutation by Δt, the total equilibrium amount of that substance present in the transmutation cycle, M, is given by

$$M = m\Delta t/\{1-(1-\alpha)(1-\delta)(1-\varepsilon)\} \qquad (3.10)$$

where it was assumed that α and δ are <1 and that the cycle time is dominated by the irradiation time. As can be seen from equation 3.10, the total amount in the process of any specific substance is approximately inversely proportional to its transmutation rate, $\varepsilon/\Delta t$.

With the same data as used in the example in 3.2.3, the total equilibrium inventory would be $\sim 18.3\, m\Delta t$.

3.3 QUANTITIES OF WASTE RADIONUCLIDES

The total future inventory of various nuclides in a swedish repository for spent nuclear fuel is not known very accurately today, see 3.3.1 below. The inventory is among other things also dependent on the future of nuclear power in Sweden, development of more efficient nuclear fuel designs. As pointed out in section 3.2.4, the inventory of nuclides in a recycle - transmutation scenario is dependent on which nuclides will be recycled for transmutation and the single pass transmuted fraction for each recycled nuclide. It is therefore also very difficult to make any reliable estimate of that inventory. These uncertainties should be remembered when comparing predicted inventories with and without P-T.

3.3.1 Comparison of radionuclide inventories

The composition of spent fuel from swedish PWR:s and BWR:s at normal burnup has been estimated several times using different computer codes and data libraries, see e.g. [Kje 78] and [Edl 83]. However, such data have normally not been used to estimate the total inventory, with its variation interval, in a swedish repository with due consideration of the actual fuel mix, expected variation in burnup, uranium recycle, enrichment, initial and final fuel batches, and reactor shut-down date etc. The only attempt we are aware of was made by Liljenzin [Lil 90].

It may be illustrative to make a comparison between the inventories of some selected nuclides without P-T and with P-T using a fast reactor similar to DFR (n-flux 1.35×10^{16} n cm^{-2} s^{-1}). Computed effective half-lives and yearly transmuted fractions for a few important minor actinides are given in table 3.2.

Tab. 3.2. Effective half-lives and yearly transmuted fractions for some actinides in the DFR core-center position estimated from data in [Kee 75].

Nuclide fraction/year	Effective half-life (y)	Transmuted
^{237}Np	1.15	0.45
^{241}Am	1.32	0.41
^{243}Am	1.56	0.36

In these favorable cases the total inventories will be rather small compared to a direct disposal of spent fuel. Assuming a three year irradiation cycle, the equilibrium inventories of these actinides will be from 3.6 to 4.1 times the yearly amount fed to the P-T cycle. Thus after 5 years the P-T cycle will have a smaller inventory of these specific nuclides than what would otherwise be accumulated as spent fuel. This, for the P-T option very positive, result is due to the combination of a high n-flux with a hard n-spectrum that was characteristic for DFR.

Another favorable condition for transmutation of ^{237}Np is the combination of very high n-flux and a thermal n-spectrum predicted for accelerator driven transmuters.

Transmutation of ^{99}Tc seems also feasible in a thermal n-flux as its n,γ cross section for thermal neutrons is about 18 barns. This should lead to a reasonably small inventory. The product formed, short lived ^{100}Tc ($t_{1/2}$ 15.8 s), will decay continuously to ^{100}Ru (stable). Further n-capture in the transmutation product will form ^{101}Ru (stable). Hence, separation of the untransmuted ^{99}Tc from ruthenium is needed at regular intervals.

On the other hand, ^{90}Sr and ^{137}Cs have rather small n,γ cross sections for thermal neutrons, in both cases < 1 barn. For ^{90}Sr, long irradiation periods in a high n-flux will lead to a small production of long lived ^{93}Zr. Furthermore n-irradiation of separated cesium, which also contains the stable isotope ^{133}Cs (n,γ cross section 26.5 b), will also produce ^{134}Cs ($t_{1/2}$ 2.06 y) and some ^{135}Cs ($t_{1/2}$ 2×10^6 y). Low transmuted fractions and many recycles can be expected for these isotopes resulting in rather large P-T inventories.

3.4 RADIOLOGICAL HAZARD OF WASTE RADIONUCLIDES

Figure 3.12 shows the number of ALI:s present per kg of spent BWR fuel after 100 years decay as function of the corresponding half life. At this moment in time, the potential hazard is dominated by ^{241}Am. The potential hazards of ^{90}Sr, ^{238}Pu, are about one order of magnitude lower. However,

after 1000 years further into the future the situation has changed somewhat. The potential hazard of isotopes with half lives less than 100 years will be lower by a factor of more than 1000. However, ^{241}Am will still be dominant. After 500.000 years ^{237}Np will dominate the potential hazard as shown in figure 3.13 and ultimately the potential hazard will be dominated by the naturally occurring isotope ^{238}U and its daughter products.

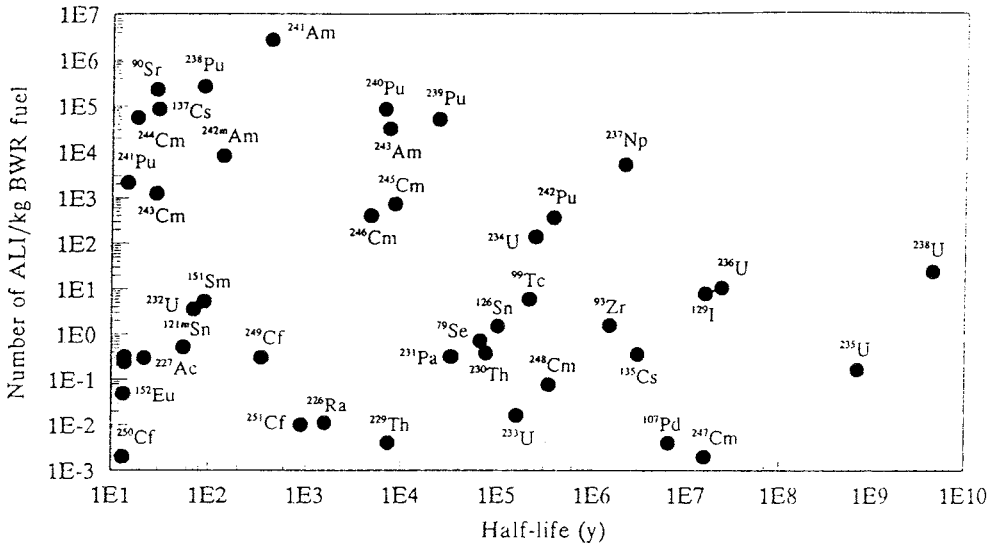

Fig. 3.12. Illustration of the potential hazard of spent BWR fuel after about 100 years storage as function of isotopes and their half lives.

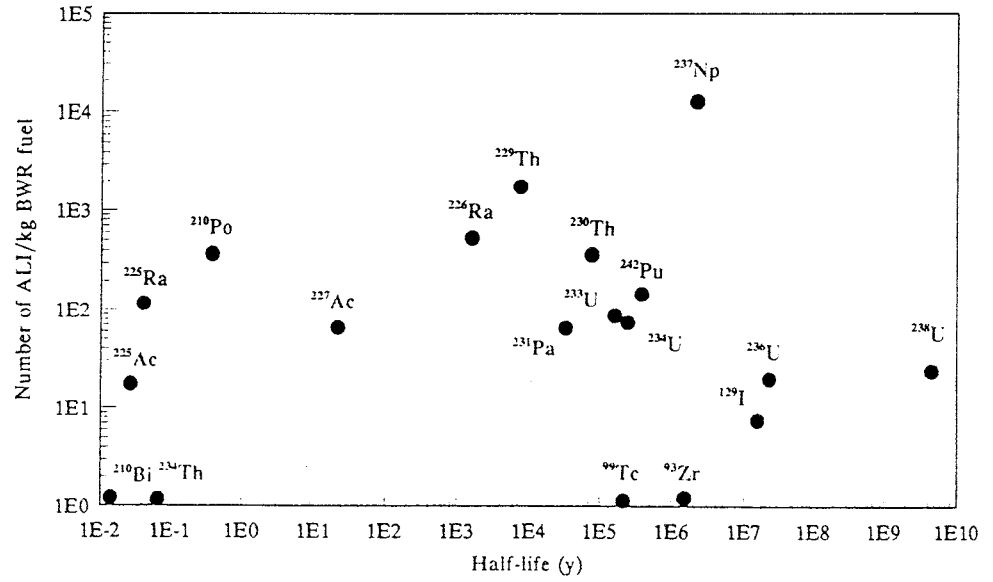

Fig. 3.13. Illustration of the potential hazard of spent BWR fuel after 500.000 years storage as function of isotopes and their half lives.

3.5 OVERALL TECHNICAL FEASIBILITY OF P-T

There are no firm indications today that P-T should not be technically feasible. However, even if many individual system components have been developed, and sometimes even tested, many still remain in the conceptual stage and considerable technical research, development and testing will be needed before the overall technical feasibility of an integrated P-T system can be determined. Current long-range research programs will help in this respect.

It seems that P-T systems based on nuclear reactors in some respects are in a more advanced stage than accelerator-driven systems. However, much R&D work needs to be done on improvement of reprocessing technology, on chemical processes for target material recovery and purification, on target development and on reactor operation with P-T targets present.

3.6 ECONOMICAL CONSIDERATIONS

As we are discussing complete P-T systems that only exists in the conceptual stage any estimate of the overall cost will be extremely uncertain. A typical P-T system would consist of an improved fuel reprocessing facility, a target fabrication plant, one or more irradiation facilities, a chemical separation plant for target material recovery and purification and various waste handling facilities including a final repository. The costs of some of the parts can be, or have been, estimated.

3.6.1 General costs

The cost of an additional separation plant for minor-actinide recovery co-located with a PUREX reprocessing plant has been estimated by several authors, see table 3.3. No cost estimates are currently available in the literature for an improved reprocessing plant with intrinsic recovery of the minor-actinides.

Tab. 3.3. Estimates of the incremental cost, in % of reference fuel reprocessing cost with Pu recycling, for some operations needed in a minor-actinide P-T system.

Source	JRC, Ispra [Sch 80]	JRC, Ispra [Hun 80, Dwo 81]	ORNL [Cro 80]	CTH [Per 83]
Chemical process	51	13-19	49	45
Fuel fabrication	34	34	29	≤24
Transportation	13	13	16	14
Waste storage & disposal	-	-	1	5
Total	98	60-66	95	≤88

The cost of operating the transmutation device depends on if this unit will be a net producer or a consumer of electricity. If transmutation of some part of the minor-actinides is done in normal LWR:s by replacement of ^{235}U with Pu and/or by replacement of gadolinium with a Np-Am mixture, this part of the operation may even have a positive economy. Further work on the benefits and drawbacks of such actinide recycle and burning in LWR:s may be needed before its cost can be assessed. Other possible transmutation devices, like LMFBR:s, ALMR:s, IFR:s or accelerator driven systems are discussed in section 4.

3.6.2 Cost in relation to other waste handling methods

It is difficult to find any reports covering a general comparison of the economics of P-T and other fuel cycle scenarios. The following discussion will be based on some reports concerning the economics of the present nuclear fuel cycle. The cost numbers will be given in the Swedish currency SKR and öre (1 öre= 0,01 SKR) and with the assumption that 1 U.S. $ = 6 SKR. OECD-NEA presented in 1985 a report on the economics of the nuclear fuel cycle [OECD 85]. It was concluded that the reference cost of the once-through cycle was 4.7 öre/kWh and 5.2 öre/kWh for the reprocessing cycle. The overall costs were compared using the constant money cost method referred to the U.S. dollar in January 1984 and a 5 % discount rate. The OECD-NEA calculations were based on the assumption of a uranium price of 608 SKR/kg U_3O_8 over the reactor life time (25 years) and a reprocessing cost of about 4500 SKR/kg HM (heavy metal). Since the OECD-NEA report has an overall fuel cycle cost that is highly sensitive for uranium prices, a low price of uranium will decrease the once-through cost significantly relative to the reprocessing cost. Looking to the uranium prices in 1989, the weighted average spot price in the U.S. was about 130 SKR/kg U_3O_8 [DOE 90], and the long term contracts had somewhat higher prices, in the U.S. it was about 260 SKR/kg U_3O_8 and in Europe about 390 SKR/kg U_3O_8 [NUE 91]. The present low uranium prices will make the reprocessing cycle even more non-competitive to the once-through cycle than indicated in the OECD-NEA report. There is a lot of disagreement on the reprocessing costs, ranging from 600 SKR/kg HM to 12,000 SKR/kg HM as presented in ref. [Ram 92b]. The British and French current commercial price of reprocessing is about 6000 SKR/kg HM. This means that the uranium price would have to increase to about 1900 SKR/kg U_3O_8, which is a 5-15 times increase of the current uranium price, or/and there have to be a significant decrease of the reprocessing cost, to make the reprocessing cycle competitive to the once-through cycle.

The OECD-NEA [OECD 85] calculated that reprocessing would account for 25 % of the cost of the reprocessing cycle corresponding to 1.3 öre/kWh, and the additional cost of disposal of reprocessing waste would be 0.048 öre/kWh. The corresponding disposal cost in the once-through cycle would account for 2 % of the overall fuel cycle cost, which corresponds to 0.11 öre/kWh. This means that even if P-T would greatly

reduce the disposal costs, it would however be a marginal cost reduction in the overall fuel cycle.

Looking to the whole chain of waste handling involving transportation, storage, conditioning and disposal of spent fuel, the costs will increase significantly. OECD-NEA reports a cost of 0,6 öre/kWh, which corresponds to about 13 % of the once-through cycle. In the Swedish back-end policy the cost of facilities for handling spent nuclear fuel and reactor components has been calculated to be about 1.9 öre/kWh [SKB 92] (Swedish currency january 1992), which at present corresponds to about 10-13 % of the net generating cost. If one furthermore include the costs for low and medium level waste disposal, operation of the facilities and research, one can expect that the total cost of radioactive waste disposal in Sweden will be about 2.7 öre/kWh. One should, however, not assume that these costs can be eliminated by introduction of P-T. Most of the costs will still be included in a fuel cycle including P-T.

The presented CURE concept (see 4.4.4) include some cost comparisons [Bin 90]. It is estimated that the net cost increase is about 2,6 öre/kWh even if the saving of 0.6 öre/kWh for the spent fuel repository is subtracted from the total cost. It is furthermore assumed that the recovered uranium can be sold at a price of about 260 SKR/kg U_3O_8 and that ^{137}Cs and ^{90}Sr can be sold as by products. The largest increase in the cost comes from partitioning of spent LWR fuel, 1.9 öre/kWh, and development of the fast transmutation reactor, 1.6 öre/kWh.

3.6.3 Economical feasibility

At present the reprocessing fuel cycle is more expensive than a once-through LWR fuel cycle. The reprocessing costs would have to decrease and/or uranium prices have to increase to make reprocessing competitive to the once-through fuel cycle. Thus, it appears that there is no economic incentive for introducing P-T at present.

3.7 TIME SCALE AND EFFORTS REQUIRED

It is quite clear that the development and implementation of a P-T system is a long-term activity. Hence, it should not interfere with current plans for direct disposal of spent reactor fuel. The complexity of a P-T system probably makes its development too costly and resource consuming for any country except may be the US or Japan. If such a system should be developed, considerable international cooperation would be needed.

4 P-T PROGRAMS IN PROGRESS

4.1 THE JAPANESE OMEGA PROGRAM

The Japanese national policy on high level radioactive waste management is disposal of a stable solidified waste form into a deep geological repository. Japan has, however, a national "Long-Term Programme for Development and Utilization of Nuclear Energy" including a relatively large program on P-T. The long-term program was approved by the Atomic Energy Commission of Japan in 1987. The P-T program was formalized under the name OMEGA (Options Making Extra Gain of Actinides and fission products generated in nuclear fuel cycle) and presented in 1988 [AEC 88]. The goal of the P-T program is to develop technology for two primary subjects:

i. *Separating groups of nuclides from reprocessing HLW according to half-life and potential value and usefulness.*

ii. *Transmutation or transforming long-lived nuclides into short-lived or non-radioactive ones.*

The R&D is carried out by the Japan Atomic Energy Research Institute (JAERI), the Power Reactor and Nuclear Fuel Development Corporation (PNC) and the Central Research Institute of Electric Power Industry (CRIEPI). In this field JAERI and PNC had a budget of about 90 MSkr in 1991 for equipment and maintenance, not including personnel and facility operation costs [Hos 91]. The personnel efforts was in 1991 estimated to be 40-50 man-years [IAEA 91b].

In 1989 the OMEGA project was presented to OECD/NEA [OECD 89], who initiated a coordinated information exchange program, see 4.6.1, which so far has resulted in three information exchange meetings [OECD 91, OECD 91b, OECD 92].

The Japanese P-T research program should be conceived as a long-term research effort to pursue benefits for future generations, and not regarded as a short-term alternative to planned back-end policies. The program consists of three major objects to be evaluated until the year 2000:

i. *R&D on nuclide partitioning technology.*

ii. *R&D on transmutation technology using nuclear reactors.*

iii. *R&D on transmutation technology using accelerators.*

Reviews of the Japanese project have been presented by Naito and Mukaiyama [Nai 91] and by Kawata et al. [Kaw 92].

4.1.1 R&D on nuclide partitioning technology

The present efforts in nuclide partitioning are devoted to the development of technology for separation of reprocessing high-level liquid waste (HLLW) into four groups: i) transuranium elements, ii) Sr and Cs, iii) Tc and noble metals (Ru, Rh, Pd) and iv) other elements (residual). In addition to the group separations, techniques for single element separation and purification are under development. Technology for utilization of the separated elements is also included in the partitioning program. Most of the research is directed towards aqueous chemistry but separation of the transuranium element group using molten salt is also considered.

The research on a three group separation procedure started in 1973 and was extended in 1984 to a four group separation procedure. The present partitioning program is divided into three phases. Phase 1, to be evaluated in 1996, includes basic research and separation technology development. The experiments will be performed with simulated HLLW and actual insoluble residues containing Tc and noble metals. In phase 2 (1997-2000) actual HLLW will be used for engineering tests of partitioning procedures. Phase 3, after the year 2000 a pilot plant test of nuclide partitioning is planned, including purification and fabrication of useful elements.

4.1.1.1 Partitioning research at JAERI

The recent achievements in partitioning at JAERI were presented by Kubota [Kub 89, Kub 91, Kub 91b], Morita [Mor 91] and Maeda et al. [Mae 91]. A procedure for counter-current extraction of Np(V) with DIDPA (Di-isodecyl phosphoric acid) from a simulated HLW solution has been tested yielding a high recovery of Np. Separation of three-valent actinides from lanthanides using the extractant DIDPA in a TALSPEAK-type separation has been tested. Am and Cm could be back-extracted from the DIDPA using a DTPA solution. Separation of Tc and noble metals has been studied. A quantitative recovery of Tc has been demonstrated, using a charcoal column and elution with a thiocyanate solution. Analytical techniques for on-line measurement of different radionuclides are under development. The advantages of a PUREX-integrated partitioning process have also been considered.

A research facility containing equipments for flow-sheet studies with actual spent nuclear fuel is under construction. The NUCEF (Nuclear Fuel Cycle Safety Engineering Research Facility) is expected to be in operation 1993-1994. The research to be performed includes i) criticality safety, ii) advanced reprocessing processes and iii) waste management of transuranium elements.

4.1.1.2 Partitioning research at PNC

Some of the recent partitioning achievements at PNC have been presented by Horie and Yoneya [Hor 91], Kawata et al. [Kaw 91], Nemoto [Nem 91], Ozawa et al. [Oza 91], Yamana [Yam 91b] and Wada et al. [Wad 91, Wad 91b]. One of the works performed at PNC is improvement of the TRUEX process, using CMPO (Carbamoyl methyl phosphine oxide) and TBP (Tributyl phosphate) as extractants. A first reprocessing experiment with fuel from a FBR (Fast Breeder Reactor) has been performed in a hot-cell line at the CPF (Chemical Processing Facility), to identify major R&D items and get some evidence on the feasibility. A schematic block diagram of the experimental flow-sheet is shown in figure 4.1.

Other research activities at PNC are improvements of the PUREX process to recover Np as well as Tc and noble metals, and basic research on new selective extractants that are completely burnable.

The behavior of some long-lived nuclides in the PUREX process has been studied. Several path-ways for Np have been considered; i) through HAW, followed by an external process for Np separation, ii) through the

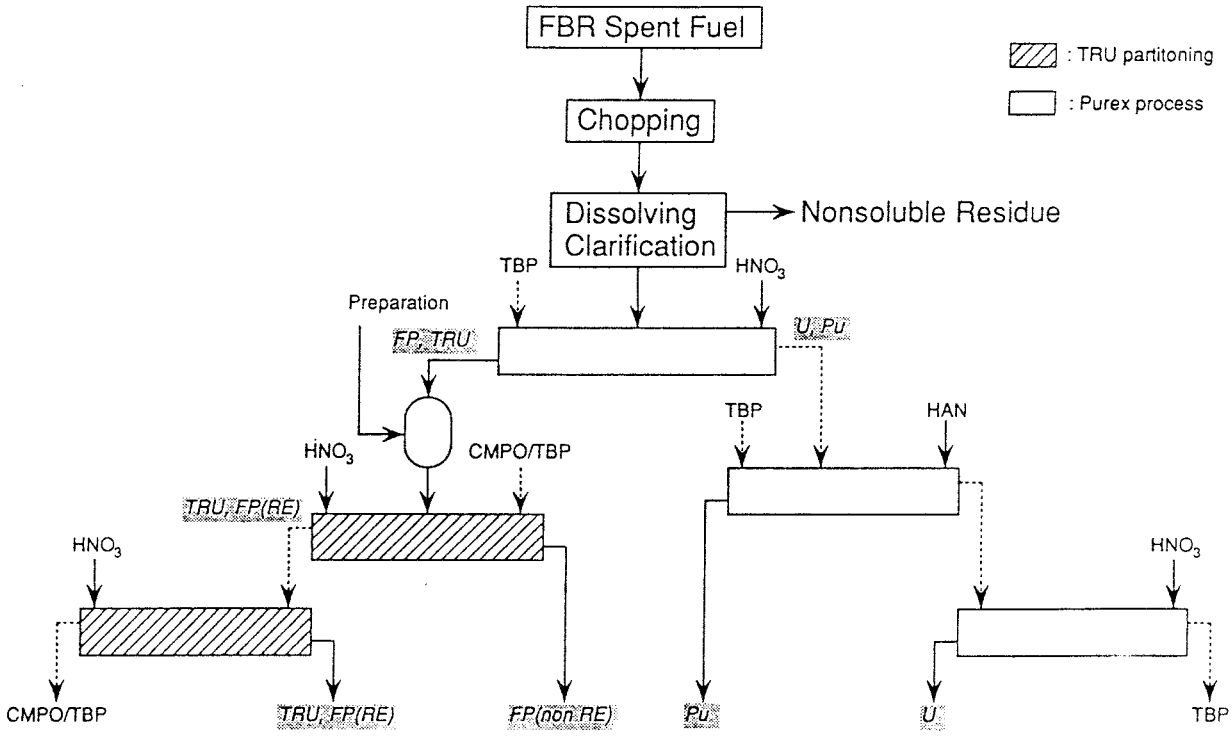

Fig.4.1. Schematic block diagram of a reprocessing experiment with FBR fuel performed at CPF [Oza 91].

Pu product and iii) through a midpoint in the process. It is assumed that the most realistic path-way is along with the Pu product. This will,

however, require a more effective process for rapid oxidation of Np(V) to Np(VI), which will be further studied. Another research item is an overall high Tc recovery in the PUREX process. Purification of both the uranium and plutonium product streams would then be required. The final separation of Tc from the HLLW may be achieved by either anion exchange, adsorption on charcoal or by high temperature sublimation.

4.1.1.3 Partitioning research at CRIEPI

The presented partitioning work at CRIEPI concerns dry processes for transuranium separation [Miy 91, Sak 91, Sak 91b, Roy 91, Ino 91b]. The project, called TRUMP-S (TRansUranic Management by Pyroprocessing - Separation), has as a goal to design a process that removes 99% of each actinide from PUREX HLLW and produces an actinide product that contains less than 10 % rare earths. The present efforts are focused on basic research, including measurements of activity coefficients and distribution coefficients of actinides and lanthanides in the system molten KCl-LiCl eutectic salt and liquid Cd. To illustrate the principles of pyro-processing a schematic drawing of a pyrometallurgical partitioning process is shown in figure 4.2.

4.1.2 R&D on transmutation technology using nuclear reactors

A conceptual design study of waste actinide burning fast reactors (ABR's) that can achieve criticality with pure transuranium element fuel is in progress. Two reactor concepts have been studied i) a Na-cooled actinide alloy fuel fast reactor and ii) a He-cooled pebble-bed fuel fast reactor. Technical details are given by Mukaiyama et al. [Muk 89, Muk 89b, Muk 91, Muk 91b]. The reactors should have a high flux of fast neutrons, to achieve a high fission to capture ratio, since most of the heavier actinides are fissible with a fission threshold in the hundred keV range.

The Na-cooled alloy fuel reactor will give a hard neutron spectrum because the low content of light metals. The fuel will be based on two alloy systems consisting of Np-(Pu)-Zr to be used in the inner part of the core and Am-Cm-(Pu)-Y in the outer part. The fuel is to be arranged in pin-bundles with an initial loading of 666 kg transuranium elements consisting of 38 % Np, 30 % Am and Cm and 32 % Pu. The reactor will have an thermal output of 170 MW_{th} and the net minor actinide (such as Np, Am and Cm) burn-up rate is 49 kg/y.

The He-cooled particle bed fast reactor will have a fuel consisting of micro-spheres of actinide nitride coated with a refractory material such as TiN. The particles will have a diameter of ~1,5 mm. The initial loading of 2065 kg transuranium elements contains 37 % Np, 29 % Am and Cm and 34 % Pu. The thermal output will be 1200 MW and the net minor actinide burn-up rate 355 kg/y.

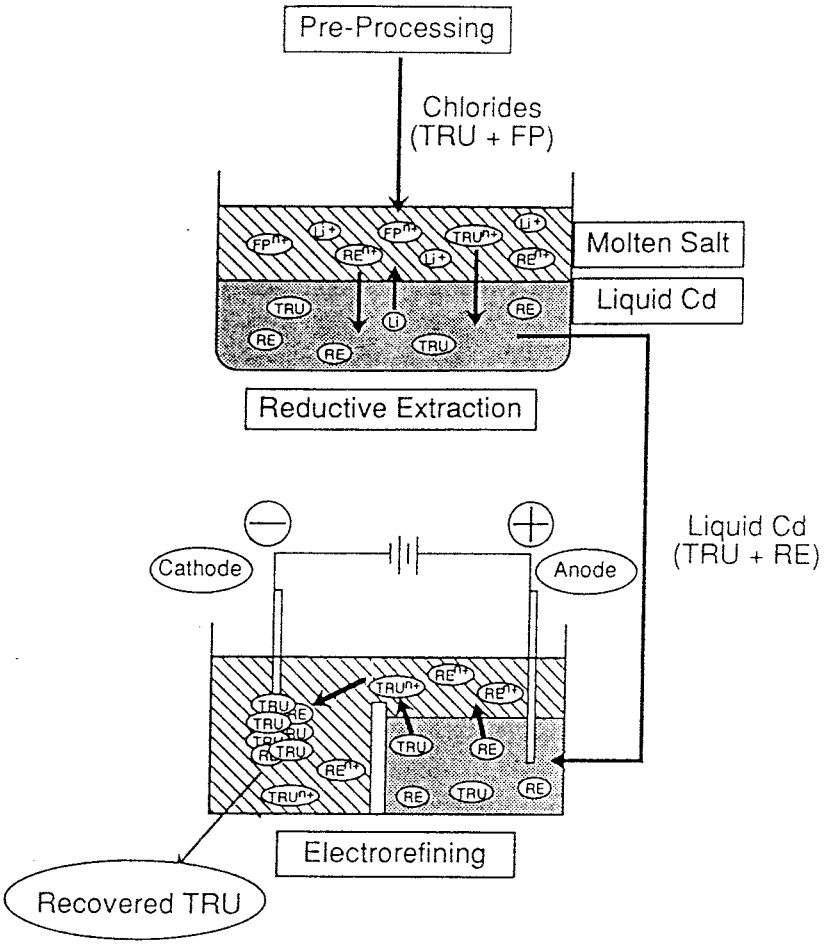

Fig. 4.2. Schematic illustration of a pyrometallurgical partitioning process [Sak 91b].

The annual amount of minor actinides (Np, Am, Cm) that is generated in a 3 GW_{th} PWR corresponds to about 25 kg. This can be compared to the net minor actinide burnup rate that can be achieved in an actinide burning reactor. It is assumed that one 1 GW_{th} actinide burning reactor can serve as a transmutation facility for all minor actinides that are produced in 10 LWR's with 3 GW_{th} power each. A schematic drawing of the fuel cycle considered is shown in figure 4.3.

The possibility to transmute minor actinides in LWR's and FBR's has also been considered in the Japanese P-T program [AEC 88, CRI 89, Hat 89, Ino 91, Muk 89, Muk 89b, Muk 91, Tak 91, Wak 89, Yam 91]. It has been found that recycling of minor actinides to a well moderated LWR as MOX-fuel can improve the reactor performance by utilizing the minor actinides as burnable poisons [Tak 91]. The initial excess reactivity and burn-up reactivity swing will be significantly reduced and the initial conversion ratio will increase. It is, however, noted that the coolant void reactivity coefficient may become positive by including a large amount of minor

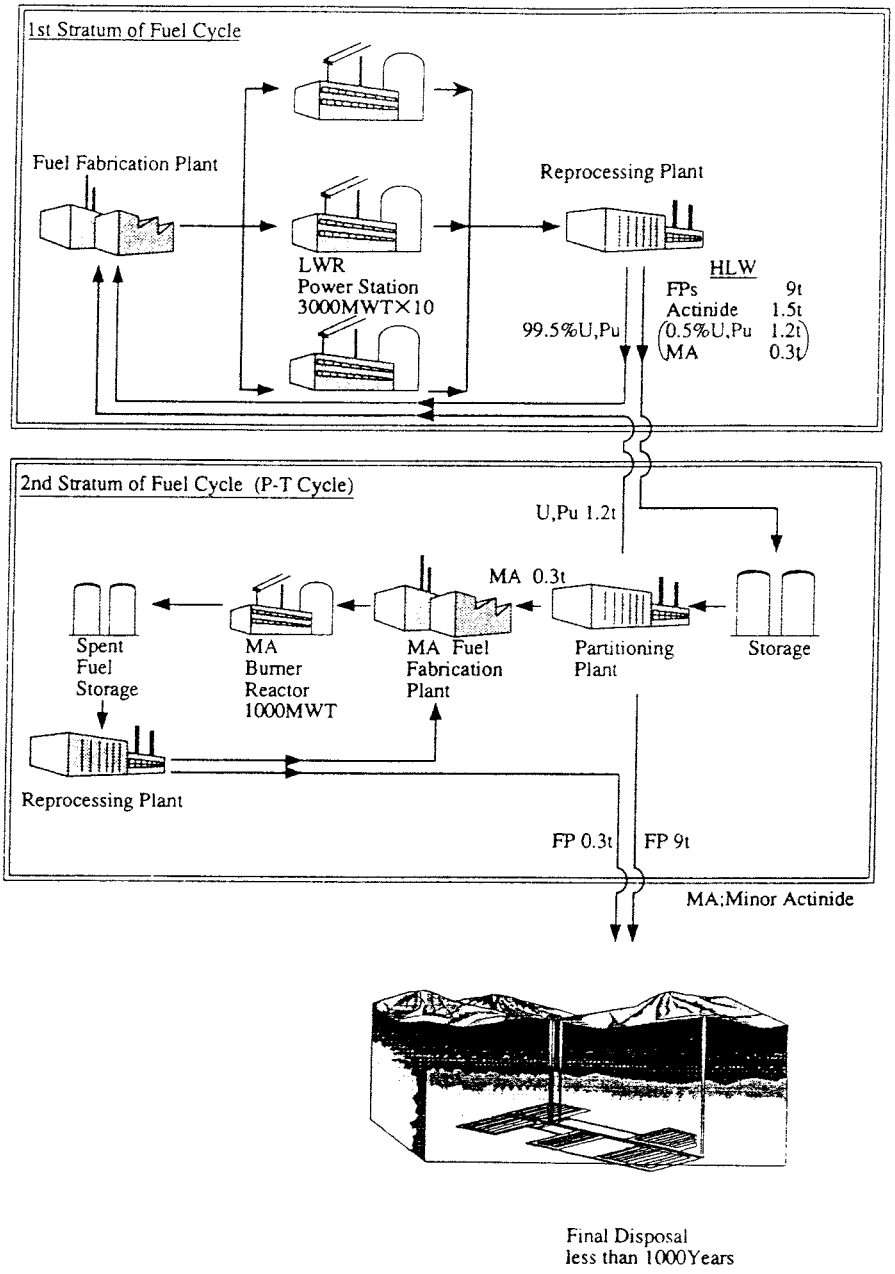

Fig. 4.3. **Fuel cycle and waste flow with partitioning and transmutation of minor actinides, such as Np, Am and Cm [Muk 91].**

actinides. The core should be designed so that a high burn-up is possible, which reduces the number of minor actinide recyclings, but this would require a high enrichment of ^{235}U or fissile Pu in the fuel. Fast breeder reactors (FBR's) have also been considered for transmutation of minor actinides due to their hard neutron spectrum. It has been shown that adding large amount of these actinides, will increase the positive sodium void coefficient and thereby reduce the inherent safety [Tak 91]. To overcome the problem with a positive void coefficient, an advanced fast breeder reactor consisting of a two region core, has been proposed [Tak 91]. The technology of an actinide-burning fast reactor has much in

common with that of a conventional FBR, and the conceptual design study will be continued in parallel with the conventional FBR R&D program. Comparison of homogeneous or heterogeneous recycling of transuranium elements in a LMFBR has been performed [Yam 91]. It was found that homogeneous recycling is preferable, because heterogeneous loading of transuranium elements can cause a critical swing in the power peaking near to the assemblies.

Concerning the transmutation efficiency it was shown that one 3 GW_{th} LWR designed for minor actinide transmutation can annually transmute the minor actinide quantities that are generated in 6-13 existing UO_2-fuel LWR's with a power of 3 GW_{th}. It was also shown that a FBR designed for transmutation of minor actinides can have an about 4 times higher transmutation rate than the corresponding transmutation-LWR [Tak 91]. In other studies [Yam 91, Ino 91], transmutation in commercial LMFBR's has been considered. With a 5 % loading of transuranium elements it could be possible to transmute the transuranium elements produced by 6 LWR's.

The future Japanese research on transmutation in LWR's and FBR's includes i) feasibility studies of the whole fuel cycle including partitioning (decontamination factors), ii) cost benefit analysis, material properties, iii) nuclear data for minor actinides, iv) optimization of core design and fuel handling systems. It is planned that the technical feasibility should be evaluated by the year 2000, whereafter the whole system should be demonstrated with an experimental reactor. There are several experimental facilities available i Japan that can be used for development of transmutation technology in FBR:s. The fast critical assembly (FCA) will be used for determination of nuclear data, and the fast experimental reactor "Joyo" will be used to evaluate the burning chain and transmutation efficiency of transuranium elements. One additional fast reactor "Monju" will soon be in operation and other fast reactors are planned.

4.1.3 R&D on transmutation technology using accelerators

Transmutation of nuclear wastes by use of accelerators is defined as one of the important tasks in the OMEGA program. Both proton and electron accelerators are considered. A review of the accelerator program will be performed in the near future to determine whether either or both a proton or/and an electron accelerator is feasible for transmutation [AEC 88]. Several reports concerning the proton accelerator program have been presented [Kan 91, Nis 89, Nis 89b, Nis 91, Miz 91, Miz 91b, Tak 91d,

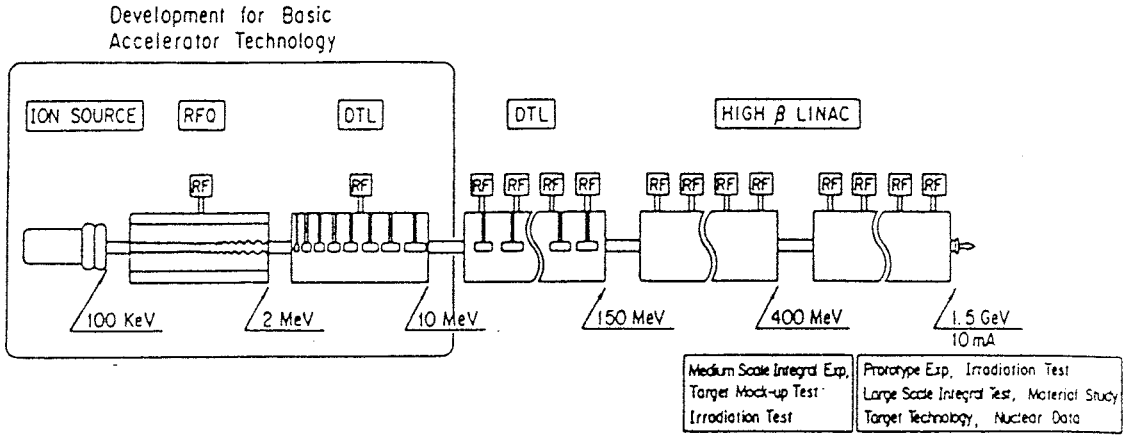

Fig. 4.4. Conceptual layout of the Engineering Test Accelerator (ETA) [Kan 91].

Toy 91], and most of the work is performed at JAERI. The possibility to transmute ^{90}Sr and ^{137}Cs in accelerator systems is also evaluated [Kas 91], but at present it does not seem to be realistic. The accelerator program is divided in several phases. At first, a Basic Technology Accelerator (BTA) with a beam energy of 10 MeV and a current of 10 mA will be constructed. The accelerator is planned to be in operation 1996 and will provide experience to build a larger accelerator. The second accelerator, engineering test accelerator (ETA), is planned to the end of the 1990's. It will provide a proton beam of 1.5 GeV at a current of 10 mA. This accelerator will be used for transmutation studies. The conceptual layout of the Engineering Test Accelerator is shown in figure 4.4.

The last phase of the accelerator program includes a demonstration plant with a linac, providing a proton-beam at 1.5 GeV and >10 mA current. A conceptual design of a sodium cooled subcritical (k_{eff} ~0.9) transmutation plant has been performed by Takizuka et al. [Tak 91d, Tak 92c], as schematically shown in figure 4.5. The plant will produce 820 MW_{th} power by using a 1.5 GeV proton-beam with a current of 39 mA. The electric power generated is 246 MW_e, which should be sufficient to power the accelerator and still deliver 100 MW_e to the external grid.

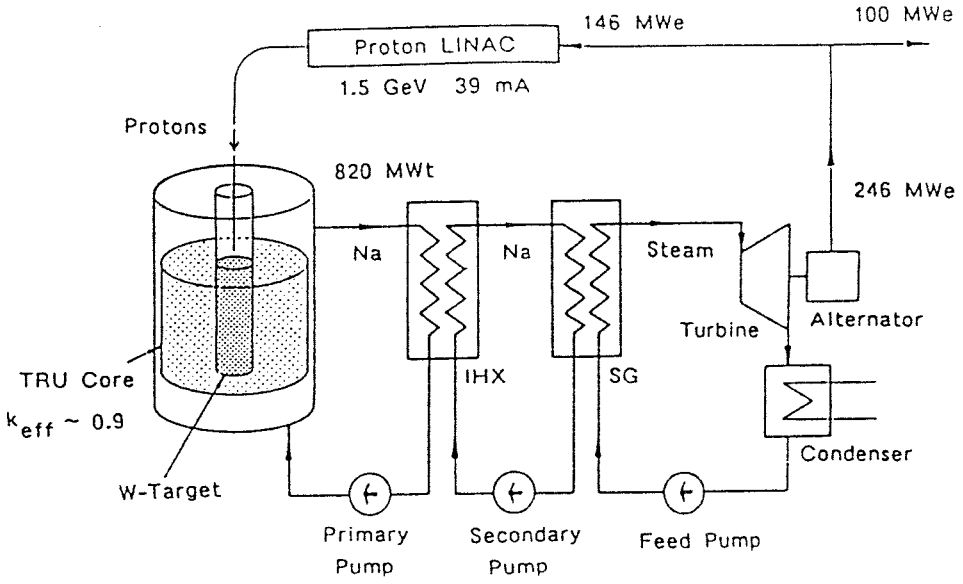

Fig. 4.5. **Conceptual flow diagram of an accelerator-driven actinide transmutation plant [Tak 92c].**

The core consists of two regions, the tungsten target acting as a spallation neutron source, and the actinide fuel. The cylindrical target is installed in the central of the core and surrounded by the actinide region, arranged as fuel pin assemblies with Np-Pu-Zr and Am-Cm-Pu-Y alloys. The actinide region is furthermore surrounded by reflectors of stainless steel. The target-core-reactor configuration is shown in figure 4.6. This system would transmute about 250 kg of actinides annually, corresponding to the amount of actinides yearly produced in about 10 LWR's. A molten chloride salt target system is also evaluated [Kat 92]. In this arrangement, the 1.5 GeV proton beam with a current of 25 mA will be directly focused into the molten salt container with an actinide inventory of 5400 kg. The transmutation rate is estimated to be about 250 kg of actinides per year.

4.2 THE FRENCH SPIN PROGRAM

France was earlier involved in the European P-T program (1975-80), mostly concerning a separation procedure for α-emitting nuclides from HLLW. The recently initiated SPIN (SP: Separation, IN: Incineration) program aims to reduce the volume and activity of the radioactive waste potentially intended for geological disposal. Some of the general objectives with the French P-T program are summarized in refs. [Com 90, Lef 91, Lef 91b, Lef 92]. The SPIN program consists of two projects of which the first project, PURTEX, has as goal to improve the separation of Pu and Np and to decrease the volume of category B waste (low heat-generating with long-lived radionuclides) originating from existing industrial reprocessing plants. The second project, ACTINEX, aims to improve the

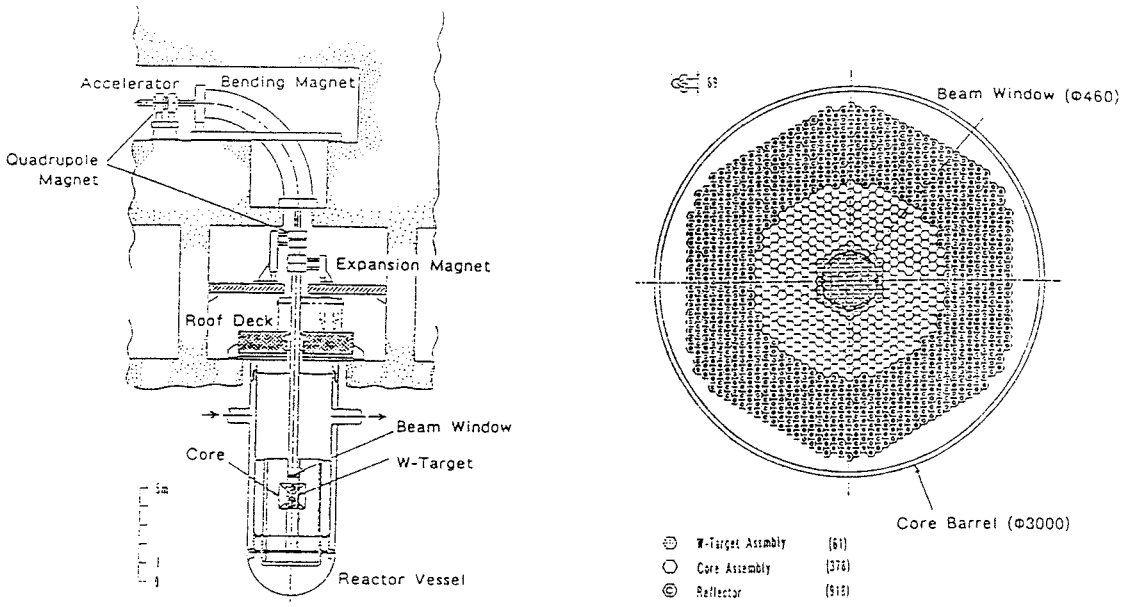

Fig. 4.6. Core and reactor configuration of an accelerator-driven actinide transmutation plant [Tak 92c].

separation of Pu, Np, Am, Cm as well as separating the long-lived fission products and furthermore to evaluate the feasibility of transmutation of all plutonium isotopes and minor actinides in thermal or fast reactors. ACTINEX is a long-term project (30-40 years), which also includes construction of facilities to reduce the volume of wastes and to develop technology for actinide fuel and target fabrication. The R&D program will be carried out by CEA (Commissariat a l'Energie Atomique), and the personnel effort was 20 man-years during 1991, which is expected to be expanded to 40-50 man-years within a 5 year plan [IAEA 91b]. The French P-T program had in 1991 a budget of about 10 MSkr for partitioning and 5 MSkr for transmutation, but it is expected to be significantly increased in 1992 [Bau 91]. Furthermore, there are some French projects involved in the CEC P-T program, see 4.6.2.

4.2.1 French partitioning research

In the French P-T program, partitioning has the highest priority because one wants to prevent accumulation of large amount of wastes containing actinides. It is expected that 20-30 years will be necessary to develop an industrial partitioning process. In the mean-time handling of the separated actinides, by either transmutation or separate storage, will be evaluated.

Development of separation procedures within the PURETEX program will be based on the present La Hague technology. The separation process will be designed to improve the Pu separation and a first separation of minor actinides (primarily ^{237}Np). Furthermore, the volume of category B waste should be reduced. At present time, large amounts of low level

waste containing α-emitters are generated at the reprocessing plants. Radionuclides are separated from different effluents by co-precipitation. The precipitates are embedded in bitumen and placed in drums. The UP3 and UP2-800 reprocessing facilities generates annually about 4000 drums of low level waste that contains more than 3.7 GBq α-emitters which cannot be disposed of by shallow land burial [Bau 91]. At present, 2.3 % of the initial Np goes with the refined U and Pu products or in the LLW, whereas 97.7% is present in the high activity effluent, which finally will be vitrified. There are several ideas of how to separate Np in the UP3 reprocessing plant, including Np separation in the acid recovery steps. It is also possible to separate a large part of the Np (about 80 %) with the Pu product, which could be useful for transmutation purposes [Bou 91, Com 90].

The ACTINEX program will define other separation processes which have a more complete separation of actinides as well as of long-lived fission products. Both solvent extraction and dry processes (pyrometallurgy) will be evaluated. R&D on aqueous based partitioning is in progress. Musikas et al. [Mus 91] presented two possible ways to achieve separation of minor actinides from effluents in the PUREX process or from HLLW: i) extraction of actinides and lanthanides in their stable oxidation state and thereafter separation of the trivalent elements by specific extraction. ii) selective extraction of actinides in an higher oxidation state by use of specific extractants. The use of diamides instead of polyfunctional organophosphorus extractants for actinides is being evaluated. One of the advantages of diamides is that they are nearly completely burnable. The actinide/lanthanide separation can be performed by selective back-extraction of actinides or lanthanides from a diamide solvent by using TPTZ (tri-pyridyl triazine) or thiocyanate. The separation of actinides in an oxidation state higher than three will also be further explored. Np will be oxidized to its hexavalent state and americium to oxidation state IV or VI before separation. A possible separation process for actinides from HLLW is proposed.

4.2.2 French transmutation research

Transmutation of minor actinides in nuclear reactors seems to be the most important issue in the French SPIN program. The transmutation program includes several topics as: i) basic studies, ii) reactor studies, iii) fuel studies and iv) fuel cycle strategies as presented in refs. [Sal 91, Sal 91b, Sal 91c, Sal 91d]. Both thermal and fast reactors are considered in these studies. Basic studies, including integral experiments to validate e.g. cross-sections, self-shielding, delayed neutrons etc., will be performed using three types of neutron spectra (LMFBR, PWR and high conversion LWR). The reactor studies will evaluate the feasibility of using LMFBR and LWR as minor actinide burners (primary ^{237}Np). Several questions are addressed to the possibility of homogeneous or heterogeneous recycling of minor actinides. In the fuel studies, fuel fabrication and material issues will be evaluated. In the SUPERFACT

experiment both homogeneous and heterogeneous oxide fuel containing minor actinides have been irradiated in the fast PHENIX reactor. Experiments with metallic fuels and minor actinides mixed with different inert matrixes (MgO, ZrO_2, Al_2O_3) are also planned. Furthermore, experiments with UO_2 fuel containing minor actinides in a thermal experimental reactor (OSIRIS) is planned.

The SUPERFACT experiment was performed in order to verify that minor actinides can be transmuted to a significant degree in a single irradiation in an existing fast reactor (PHENIX) and that the fuel elements of Np and Am could be fabricated in the same way as standard Pu fuel. The experiment was performed by CEA in collaboration with CEC, see 4.6.2, and some of the first results were presented by Prunier et al. [Pru 91]. Two types of fuel elements, homogeneous (2 % minor actinides and $U(Pu)O_2$ matrix) and heterogeneous (40-45 % minor actinides and UO_2 matrix), were fabricated according to a method used for $U(Pu)O_2$-fuel. The fuel pins were arranged according to figure 4.7 and irradiated in the PHENIX reactor for a period of 383 equivalent full power days, corresponding to a 8.5 % burn-up. The SUPERFACT experiment proved the ability to fabricate minor actinide fuel and the first analyses of the irradiated fuel pins reveal no special problems concerning the behavior during irradiation, compared to the standard mixed oxide fuel. A considerable helium release could, however, be observed in the fuel pins containing Am, which could affect the mechanical strength of the cladding during a long-term irradiation. The evaluation of the SUPERFACT experiment is not finished and further results will be published.

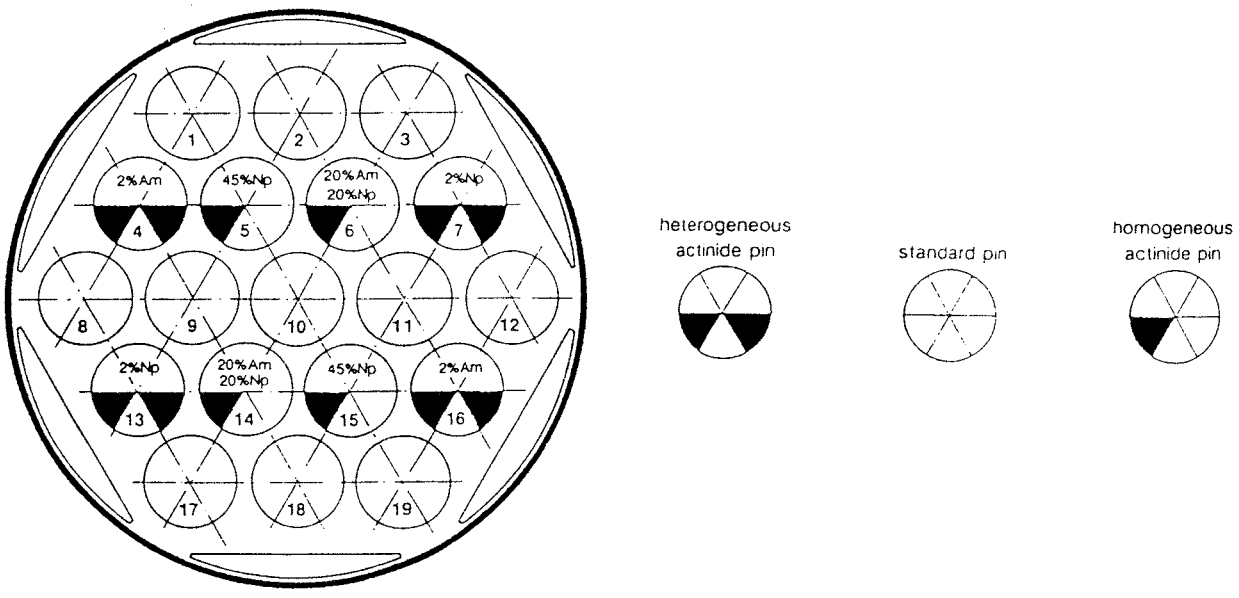

Fig. 4.7. Fuel pin arrangement of the 19-pin PHENIX standard capsule used in the SUPERFACT experiment [Pru 91].

4.3 THE CIS P-T PROGRAM

The current CIS technology for radioactive waste management includes geological repositories. A long-term R&D program will, however, evaluate waste management alternatives, such as: i) deposition of radioactive wastes in space, ii) self-burying of radioactive waste by self-heating through the earth-crust and iii) transmutation of long-lived radionuclides [Chu 92]. The P-T research program in the former USSR was presented in refs. [Kud 91, Rom 91] at the IAEA Advisory Group Meeting on P-T [IAEA 91]. The work is coordinated by the Ministry of Atomic Power and Industry (MAPI) and several research institutes are involved, including e.g. the Institute for Theoretical and Experimental Physics (ITEP) in Moscow, the Moscow Radiotechnical Institute (MRTI), the All-Union Scientific Research Institute of Inorganic Materials (AUSRIIM), the Moscow Physical Engineering Institute (MPEI), the Institute of Nuclear Power Engineering (INPE) in Obninsk, the Physical-Energy Institute (FEI) in Obninsk and the Khlopin Radium Institute in St. Petersburg. The interest is focused on transmutation in fast reactors and heavy water reactors, but the use of proton and deuteron accelerators is also considered. Some new reprocessing technologies have either been commercially realized or tested in pilot tests and the development of partitioning techniques will therefore rely on experience obtained during the R&D on reprocessing. The personnel effort that is put into P-T is estimated to be about 100 man-years [IAEA 91b].

Several papers concerning the CIS P-T program was presented at the "Workshop on Nuclear Transmutation of Long-Lived Nuclear Power Radiowastes" arranged by INPE and ITEP in July 1991 [INPE 91].

4.3.1 CIS partitioning research

Most of the recent CIS P-T reports concerns transmutation in reactors or accelerators and there are only a few reports concerning partitioning technology. It has been mentioned that the Khlopin Radium Institute and the AUSRIIM have technological solutions for reprocessing spent nuclear fuel and waste management [Kud 91]. A separation process for Cs, Sr, lanthanides and transplutonium elements from HLLW, based on extraction with cobalt dicarbolyde, has been developed and tested in pilot scale [Gal 88, Rom 91]. The radionuclides that have been suggested to be transmuted are ^{237}Np, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{243}Am, ^{129}I, ^{14}C, ^{151}Sm, ^{180}Sn, ^{79}Se, ^{93}Zr. Transmutation of ^{90}Sr and ^{137}Cs is also considered, but a special storage will be evaluated as an alternative [Rom 91]. Separation of C, I, Tc and Zr could also be made according to procedures developed at the Khlopin Radium Institute [Rom 91]. Tc could be controlled in the reprocessing process by quantitative (99.5 %) extraction into the raffinate at the stage of Pu purification.

4.3.2 CIS transmutation research

The R&D efforts will evaluate the feasibility of transmutation in i) thermal nuclear reactors, ii) fast reactors, iii) accelerators (p^+, d^+) and iv) fusion installations [Chu 92].

Studies have shown that minor actinide transmutation in thermal reactors is feasible [Chu 91]. But this would require changes of the reactor operation due to the radiation and neutron characteristics of the secondary fuel [Ser 91]. It has been shown that ^{99}Tc can be successfully transmuted in some thermal reactors [Rom 91].

Transmutation of actinides in fast reactors has also been studied [Nik 91, Ser 91, Shm 91, Shm 91b]. Calculations show that a fast reactor of the BN-800 type, can burn 100 kg actinides in one year and still maintain full-scale power production and safety. Burning 100 kg/y of actinides correspond to the annual actinide production from three commercial reactors of the VVER-1000 type. As an alternative design, a heavy water reactor, providing an epithermal neutron spectrum, has also been considered for transmutation. Furthermore, it has been shown that the annual actinide transmutation rate in fast reactors (11 %) is higher than in thermal reactors (~ 5%) [Ser 91].

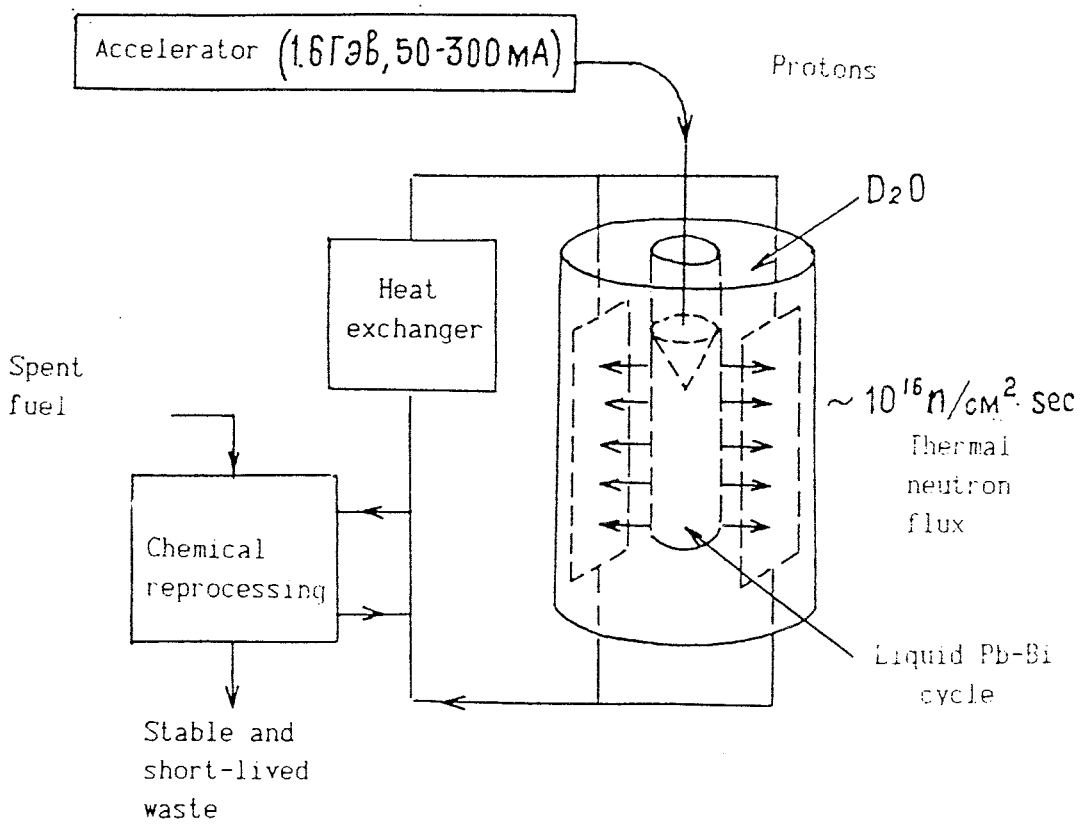

Fig. 4.8. Schematic design of an accelerator-driven transmutation plant [Kaz 92].

Transmutation by using particle accelerators has been of special concern during the last two years, see e.g. refs. [Ado 91, Ale 91, And 91, Avd 91, Bel 92, Bla 91b, Byc 91, Chi 91, Chu 92, Her 91, Kap 91, Kaz 91, Kaz 91b, Kaz 92, Kaz 92b, Kis 92, Mur 91, Nik 92, Shv 92, Smi 91, Toc 92]. These reports contain different approaches for accelerator-, target- and blanket-design. But in the recent reports it can be found that the ITEP R&D efforts will be focused on a medium energy high intensity proton accelerator, a spallation target and a heavy water based blanket, see figure 4.8. The design of the transmutation plant will be similar to the earlier proposed Los Alamos ATW concept, see 4.4.2. ITEP has also proposed to the Ministry of Atomic Power and Industry to build a prototype of a proton accelerator having an energy of 100 MeV and 30-50 mA current, in order to develop accelerator technology. The final accelerator should, however, have an energy in the GeV range and a current of several hundred mA.

4.4 THE USA P-T PROJECTS

There exists several technical proposals for P-T in the USA. But the primary US government program will evaluate the potential of using an Advanced Liquid Metal Reactor (ALMR) for transmutation of long-lived toxic components in HLW originating from civilian nuclear power, see 4.4.1 [Gol 91]. The considered benefits of an ALMR system includes i) potential reduction of major long-lived toxic actinides in HLW, ii) having a process with the ability to remove heat- producing radionuclides that can affect the design of the repository and iii) having a system which can make better use of the energy content in uranium fuel. The US Department of Energy (DOE) has initiated a study of the technical and economical feasibility of ALMR with regard to waste management and long-term energy production. The first part of this study will be evaluated in 1995, whereafter decisions about further work will be taken [Gol 91].

Several of the US P-T programs were presented at the "Symposium on Separations Technology and Transmutation System (STATS)" arranged by the US National Research Council [NRC 92]

4.4.1 The ALMR program

The Advanced Liquid Metal Reactor (ALMR) program is coordinated and sponsored by the US Department of Energy (DOE). The program will evaluate two possible advanced reactor concepts, namely the IFR (Integral Fast Reactor) and the PRISM (Power Reactor, Innovative, Small Module). Furthermore, the ALMR program includes development of reprocessing technology and studies of fuel cycle scenarios which also include P-T [Gol 91, Web 91].

4.4.1.1 The IFR program

R&D on the IFR (Integral Fast Reactor) concept is performed by the Argonne National Laboratory (ANL). The IFR consists of a Na-cooled, pool-type reactor [Til 89]. A schematic drawing of the IFR fuel cycle is shown in figure 4.9. The core consists of metallic fuel elements and the IFR has an integral fuel cycle including reprocessing and fuel fabrication on-site. A key feature of the IFR concept is the pyrometallurgical reprocessing process, which employs electro refining at 500 °C, using a cadmium anode and a molten salt electrolyte (LiCl-KCl-U/$PuCl_3$). Minor actinides will accompany the plutonium product stream and therefore, actinide recycling occurs naturally. The IFR fuel cycle and the pyro-metallurgical processing were presented in some recent reports [Cha 92, McP 91]. It is possible to use the IFR either as a net actinide burner or in an actinide self-sustaining mode (i.e. breeding). The potential of using an IFR/LWR synergetic fuel cycle has been considered and a R&D program has been initiated to develop a pyrochemical process that can extract actinides from spent LWR fuel. If the pyro-processing of spent LWR fuel is feasible it will be of real interest for the IFR concept. It is possible to consume 425 kg actinides/GW_e per year with an initial core inventory of 7400 kg actinides/GW_e, by using IFR as a net actinide burner.

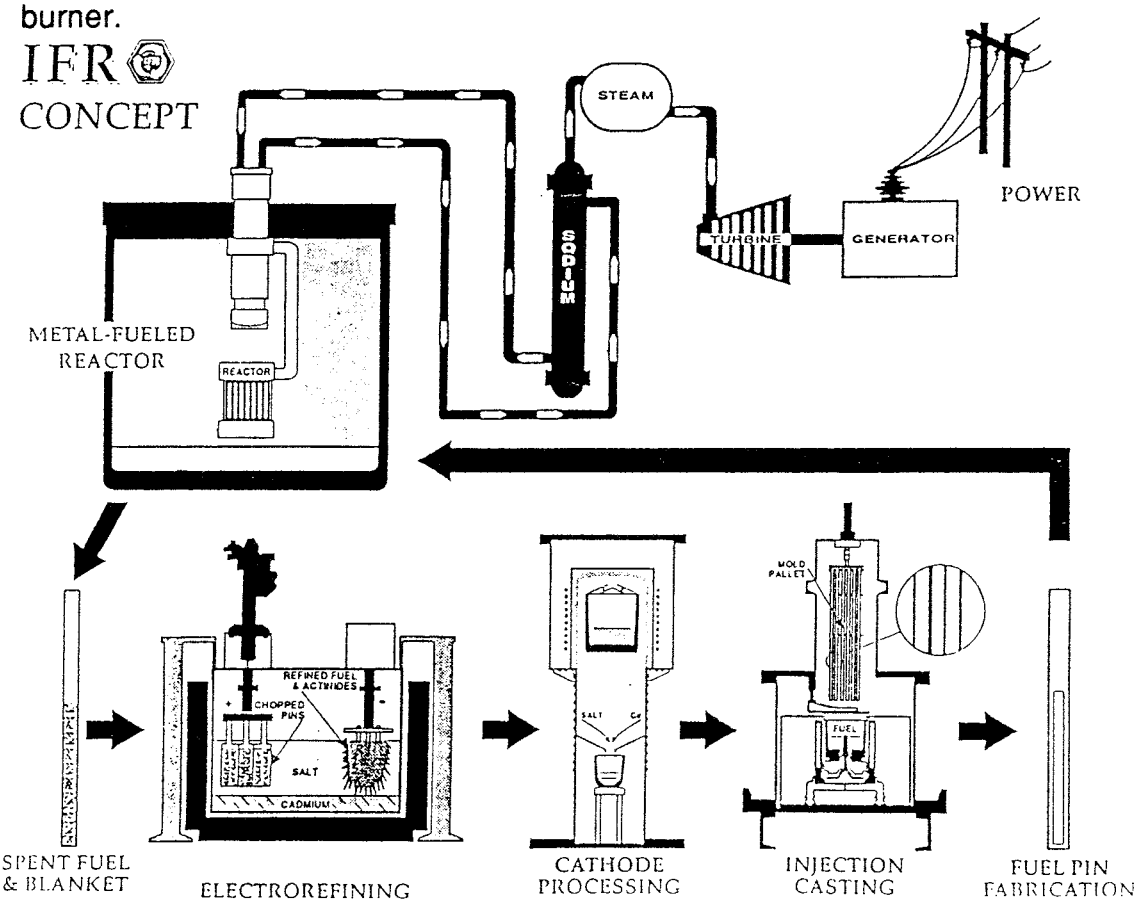

Fig. 4.9. A schematic picture of the IFR concept [Cha 92].

4.4.1.2 The PRISM program

The PRISM (Power Reactor, Innovative, Small Module) modular reactor concept was developed by General Electric Co. [Tho 92]. PRISM consist of a relatively small and compact Na-cooled reactor module, sized to enable factory fabrication and easy shipment. Each plant consists of three reactor modules of 471 MW_{th}, providing a net electrical output of 465 MW_e. The metallic fuel and fuel cycle are based on the IFR concept, see 4.4.1.1. PRISM was initially designed as a breeder reactor, to start with plutonium from LWR spent fuel, but is now evaluated as a net actinide burner for all minor actinides. The core layout of a PRISM module used as an actinide burner is shown in figure 4.10. The core consists of low and high enriched fuel. As an example, it has been shown that the transuranium element consumption rate can be about 35 kg/year in a PRISM module having a total actinide inventory of 1818 kg and a Pu enrichment of 14% and 20% in the low and high enriched fuel, respectively.

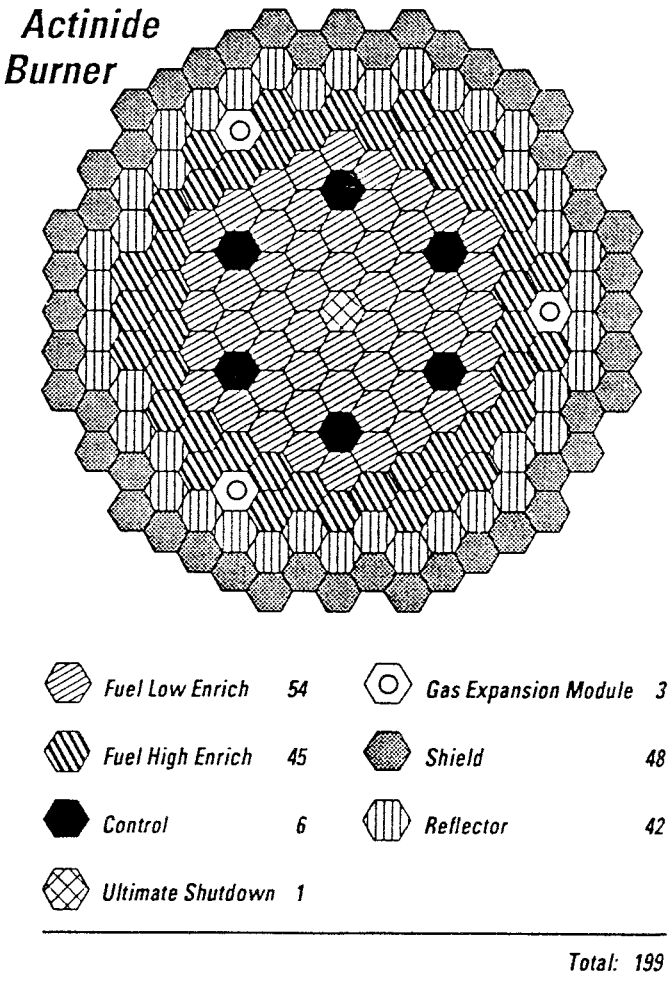

Fig. 4.10. PRISM core module layout in actinide burning mode [Tho 92].

4.4.2 The ATW concept

The Los Alamos Accelerator Transmutation of Nuclear Waste (ATW) concept, has got a lot of attention during the last two years and several reports have been presented [Art 91, Art 91b, Art 92, Att 92, Bow 90, Bow 91, Bow 91b, Ire 92, Ire 92b, Kra 92, Los 90, Sch 91c]. A schematic drawing of the ATW system is shown in figure 4.11. The ATW consists of an advanced, high current, medium energy (< 2 GeV) linear accelerator which delivers a proton beam to a heavy metal spallation target. A tank containing heavy water surrounds the target and works as a moderator to get thermal neutrons. The resulting thermal neutron flux is >2-3×10^{15} n cm^{-2} s^{-1} in a large volume. Due to the high neutron flux and the large thermal cross sections the system will have a low inventory of radionuclides to be transmuted. The blanket containing the radionuclides consists of several loops whose content will be circulated in closed cycle through the moderator tank. Several options, aqueous media, oxide slurries and molten salt have been considered as carrier materials to be used in the loops.

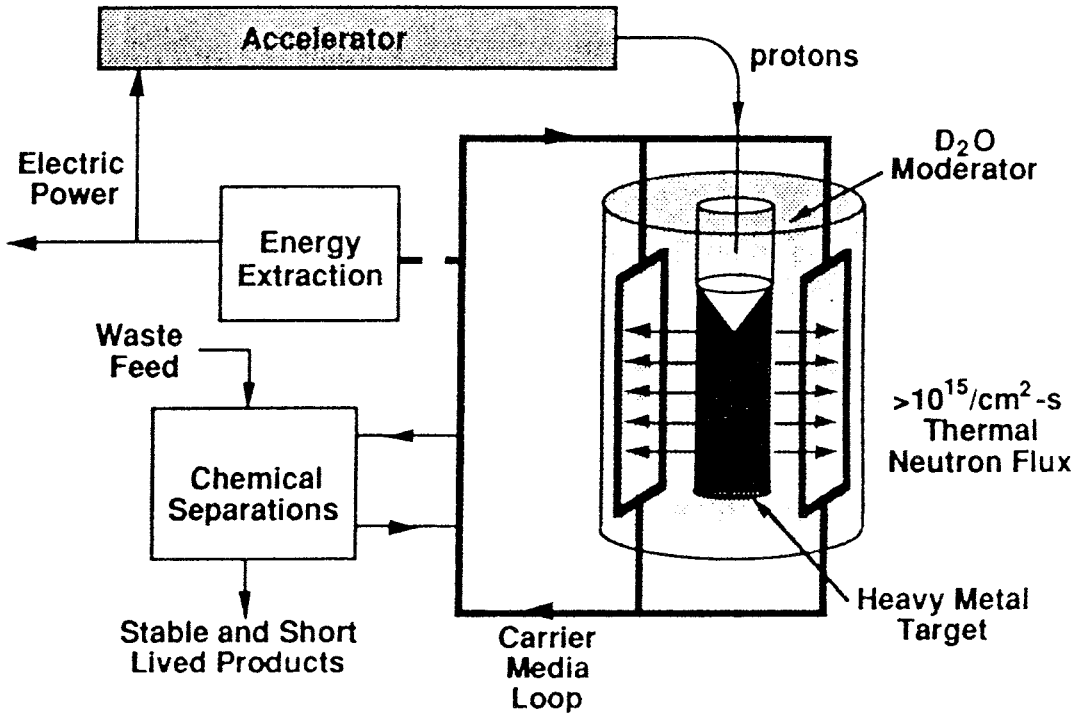

Fig. 4.11. Schematic design of the ATW system [Art 92].

The linear accelerator will generate a 1.6 GeV proton beam with a mean current of 250 mA. Preliminary design concepts for the accelerator are discussed in refs. [Law 91, McM 91, Sch 91b]. The proposed accelerator configuration is shown in figure 4.12. It consists of a 700 MHz coupled-cavity linac (CCL), which is fed by two 125 mA linear accelerators consisting of radio frequency quadrupole linacs (RFQ's) and drift-tube

linacs (DTL's). It is expected that one accelerator can support four separate target/blanket modules, producing 1.5 GW_{th} each. The accelerator will require 900 MW of electric power and with a thermal to electric conversion efficiency of 30% it is possible to produce 1 GW_{el} to the external grid. The calculated transmutation efficiency shows that the system will annually transmute the actinide waste from ~7.5 LWR's, corresponding to 2450 kg/y. The excess neutrons will also provide a possibility to transmute ^{99}Tc and ^{129}I from 7.5 LWR's in each blanket.

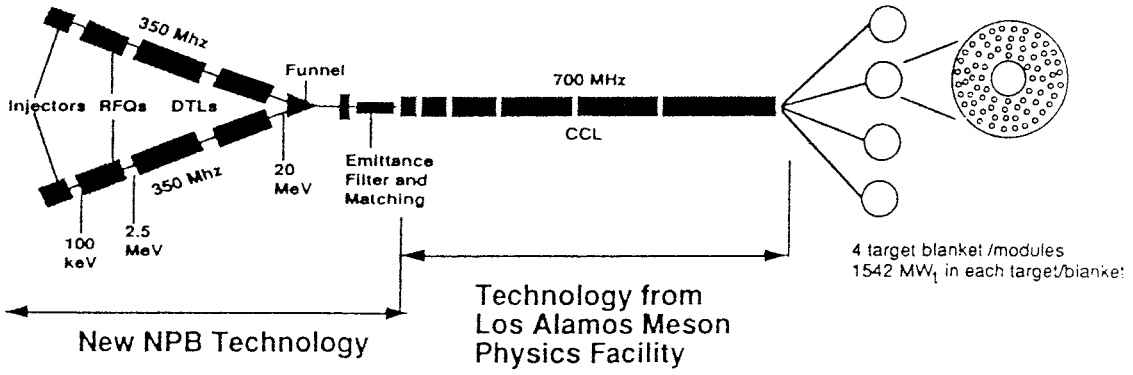

Fig. 4.12. The linear accelerator for ATW [Art 92, Cap 92].

The chemical separation procedures, target/blanket and material issues are discussed in refs. [Abn 92, Cap 91, Cap 92, Cap 92b, Jar 92, Jen 91, Jen 91b, Wal 92, Wal 92b]. Several high atomic number elements can be used for the spallation target including Pb, Bi, W, Ta and U. Both a solid W-target and a liquid Pb-(Bi) target are considered. The blanket region technology is based on existing heavy-water reactor technique employed in the CANDU reactors. The actinides will be circulated in double-walled pressure tubes in the form of a D_2O slurry, whereas the long-lived fission products will be dissolved in heavy water. The on-line chemical treatment of the actinide slurry will be performed by dissolving the slurry in nitric acid, whereafter Np, Pu, Tc and Pd are extracted by Aliquat 336. Further separation of trivalent actinides will be performed by a reversed-TALSPEAK process. Separation of Ru from the $^{99}TcO_4^-$ solution is proposed to be made by using ozone to volatilize RuO_4.

One of the driving objectives behind the Los Alamos concept is transmutation of radioactive defence waste. It has been suggested that an accelerator-driven intense thermal neutron source could, within 30 years, transmute all the ^{99}Tc and ^{129}I that has been accumulated at the Hanford site (~2000 kg) [Los 90]. A secondary objective could be a large scale production of ^3T for the national defence program.

4.4.3 The PHOENIX concept

The Brookhaven National Laboratory PHOENIX concept uses a large proton accelerator to drive one or more subcritical lattices of minor actinides in order to transmute the long-lived radionuclides from LWR's. The PHOENIX concept have been presented in refs. [Tak 92, Van 91, Van 92], and a schematic figure of the concept is shown in figure 4.13.

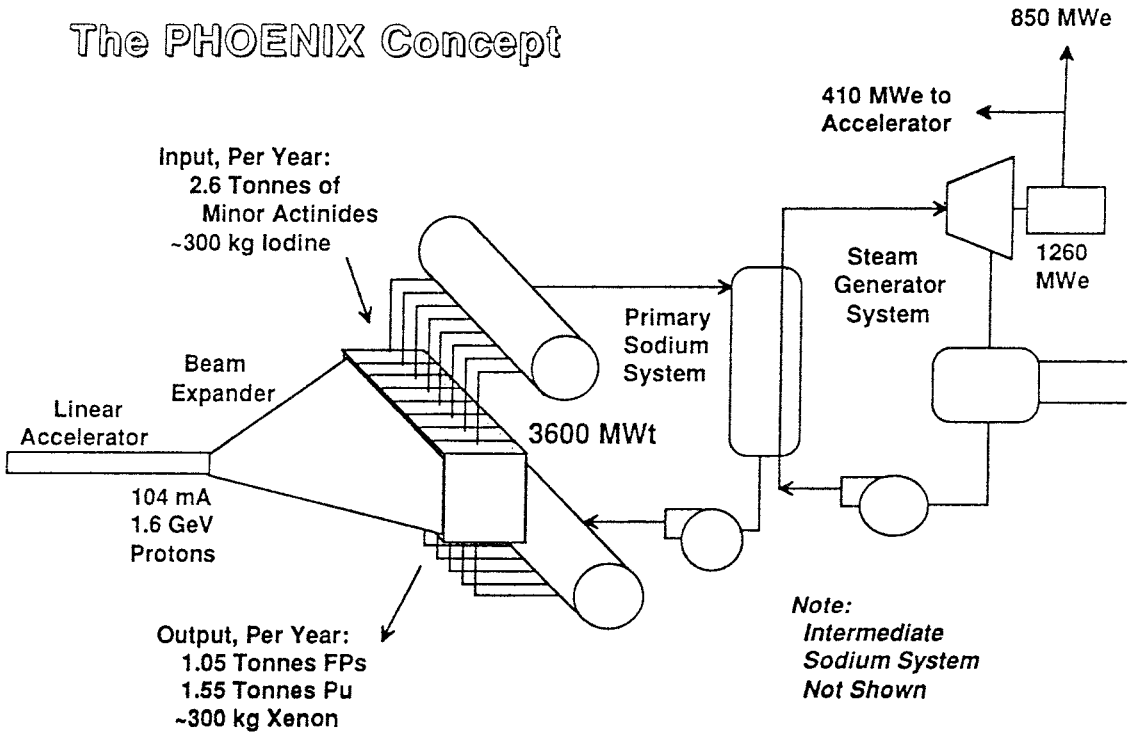

Fig. 4.13. The PHOENIX concept [Van 92].

The accelerator provides a 1.6 GeV proton beam with a current of 104 mA. The beam irradiates 1 to 8 target modules consisting of a Na-cooled cores, resembling the core of the Westinghouse Fast Flux Test Facility (FFTF). Each target module, of 450 MW_{th}, has the size of 0.75×0.82×1.88 m, and the modules are designed to be removed independently for fuel reloading. The fuel consists of oxide fuel rods and the target modules have a k_{eff} in the range of 0.9-0.95. This means that each proton will trigger about 170-350 fission events, assuming that 50 neutrons are released via the spallation and evaporation processes as the proton passes into the target lattice. Each fuel cycle would be limited to 2 years, to be able to remove plutonium while it largely consists of ^{239}Pu. The estimated annual ^{239}Pu production will be ~1500 kg. One 3600 MW_{th} machine can transmute the Np, Am, Cm and most of the ^{129}I produced in ~75 LWR's, and it will generate about 850 MW_{el} to the external grid.

The reprocessing technology to be applied in the PHOENIX concept is based on the proposed CURE concept, see 4.4.4.

4.4.4 The CURE concept

The Westinghouse-Hanford CURE (Clean Use of Reactor Energy) concept [Bin 90] is not focused on a specific technology. It is rather a multidisciplinary work to evaluate the possibility of partitioning and transmutation, with the goal of reducing the need of several large geologic repositories. Some ideas behind the concept have been presented by Rawlins [Raw 92]. An aqueous partitioning procedure based on solvent extraction (PUREX-TRUEX) will be used for separation of actinides and technetium. Some of the experiences with the chemical processing have been presented by Boomer [Boo 92] and by Swanson and Lumetta [Swa 91]. Aqueous based separation procedures for long-lived fission products eg. Cs, Sr(SREX) will also be used. Most of the long-lived radionuclides are considered to be transmuted. Only Sr, Cs and some activation products will not be transmuted. At first, a Na-cooled fast reactor was considered as a transmutation facility, and it was assumed that one fast reactor should be used for transmutation of actinides from four LWR's. With this ratio of fast to light water reactors, some plutonium and recycled uranium would have to be returned to the LWR's.

It was concluded that aqueous processing technology can meet the separation factors required. However, deep geologic disposal would still be required for the resulting waste. The R&D efforts, required for the CURE concept, are estimated to cost a billion dollars prior to the pilot plant demonstration phase, and would take one or two decades to perform. The dominant cost would be investigation of transmutation schemes, including reactor or accelerator driven concepts.

4.5 OTHER P-T PROJECTS

4.5.1 Accelerator driven reactors

An accelerator driven sub-critical reactor utilizing the Th-fuel cycle has been proposed by Furukawa et al. [Fur 91]. The concept called THORIUM-NES (Thorium Molten-Salt Nuclear Energy Synergetic) is a development of the ORNL Molten Salt Breeder Reactor (MSBR). One of the proposed reactors is the Accelerator Molten-Salt Breeder (AMSB), also having the ability to transmute fission products and using minor actinides as fuel. The AMSB is driven by a 1 GeV, 300 mA proton accelerator.

Accelerator driven sub-critical systems have also been proposed by Takahashi et al. [Tak 91c, Tak 92, Tak 92b]. Operation of sub-critical (k_{eff} 0.99) metal-fuel reactors by 1 GeV, 15-20 mA proton accelerators is proposed. By supplying external neutrons produced by the spallation and high-energy reactions, the reactor can supposedly be operated in safer

conditions than a reactor operated in critical conditions. The slightly sub-critical condition makes the power distribution more flat, compared to cores with lower k_{eff}. Because of a lower required beam current in this facility, the possibility to use a multistage cyclotron arrangement instead of a linear accelerator is also pointed out. This type of accelerator-driven sub-critical reactor can be used as a breeder and transmutation facility.

Another accelerator driven sub-critical (k_{eff} 0.28) breeder reactor was presented by Kugeler [Kug 91]. This reactor is based on the Th-fuel cycle and was developed in the beginning of the 1980'ies. The target consists of spherical Th-fuel elements placed in a He-cooled pebble bed and irradiated by 1 GeV, 300 mA protons. The ^{233}U production rate was estimated to be about 800 kg/y, which should be sufficient to produce fuel for 9 high temperature reactors. Several technical solutions to the accelerator driven reactor system was developed during this project.

4.5.2 Fusion-Fission hybrids

The possibility of using fusion-fission hybrid systems for transmutation purposes has been discussed. Two different fusion approaches have been considered: i) heavy-ion inertial fusion [Mar 91, Mar 91b] and ii) muon catalyst fusion [Chu 92, Har 92, Pet 92]. The 14 MeV neutron spectrum from fusion reactions has the unique ability to transmute long-lived fission products by the (n,2n) reaction. Furthermore, plutonium- and tritium-breeding in a muon catalyst fusion reactor have been studied by Kalcheva et al. [Kal 91]. The technical feasibility of fusion must, however, be realized before any fusion-fission hybrid can be considered.

4.5.3 Other national P-T efforts

4.5.3.1 Belgium

R&D in the field of P-T is performed by Belgonucleaire, and recent work is presented in ref. [Ren 91]. There exist experience in separation technology for actinides and fission products, from radioanalytical procedures developed for measurements of irradiated fuels and from projects directed to the production of actinides. A special attention is given to the impact of separation and transmutation of ^{237}Np, ^{129}I and ^{99}Tc on waste disposal strategies [Bae 91]. The introduction of Np, Am and Cm in fuels for fast reactors or PWR's has also been evaluated [Ren 91, Pil 91]. It has been shown that insertion of 2.3% Np in the conceptually designed European Fast Reactor (EFR) can have some advantages, the required Pu-enrichment of the fuel is reduced, the burn-up reactivity losses decreases and the Pu-breeding increases to some extent. Calculations showed that U-fuelled LWR and FBR can burn 17-27% of an initial loading of ^{237}Np and ^{241}Am during a standard assembly residence time, but there are several advantages of using a FBR as mentioned above. The problem of the intense γ-emitter ^{208}Tl resulting from the decay chain fed by the (n,2n) reaction in ^{237}Np is also considered.

4.5.3.2 Canada

In Canada the Atomic Energy of Canada Ltd. (AECL) is involved in nuclear technology research. AECL does not have any specific P-T program, but several of the research programs do have relevance to the international P-T efforts, including i) development of high-current, high-power accelerators, ii) basic nuclear physics research, and iii) chemical research, as been presented in ref. [Har 91]. The development of high-current continuous wave (cw) linacs has generated a lot of interest in the design of accelerator-driven transmutation facilities, e.g. the ATW concept, see 4.4.2. The accelerator program includes development of a high-current ion source, a 75 mA cw proton radio frequency quadrupole (RFQ), and high duty-factor graded-β and β=1 coupled-cavity linacs (CCL) [McM 91]. The basic nuclear research include studies of the neutron yields from spallation targets [Lon 91b] and determination of neutron cross sections to improve existing nuclear data bases on fission products and actinides. The thermal neutron capture cross section of ^{90}Sr was determined to be 7±4 mb in a recent work presented by Lone and Edwards [Lon 91].

4.5.3.3 China

The dominating interest of P-T in China is in the field of partitioning. At present the HLW from chinese reprocessing is accumulated and stored in underground tanks and a solidification facility is planned to be in operation in 1998-2000. There is some interest to separate actinides and valuable metals from the HLW before vitrification. During the last decade, three different separation flow-sheets has mainly been studied, i) the reversed-TALSPEAK process, ii) the DHDECMP process and iii) the TRPO process [Cri 91]. Some of the results with the TRPO process (Trialkyl Phosphine Oxide) was presented by Zhu [Zhu 91], showing that the process have a potential to be industrially applicable for removal of actinides from HLW. A hot test experiment with high-level defence waste, using the TRPO process, is planned to 1995. There is also an interest to develop processes for separation of noble metals, like Ru, Rh and Pd from HLW.

4.5.3.4 Germany

The German activities in the field of radioactive waste handling were reported by Ache [Ach 91]. At present time there is no German P-T program, but small groups of scientists will closely follow the international P-T efforts. It is estimated that 10-12 people will be involved for some years. At the Nuclear Research Centre in Karlsruhe (KfK) a two year coordinated R&D program between the Institute for Neutron Physics and the Institute for Hot chemistry started in 1991. The program will evaluate different strategies for actinide transmutation [Sch 91]. The program is connected to the CEC P-T program, see 4.6.2. The proposed partitioning program will concentrate on aqueous processes with the objective to

separate residual Pu, U, Np, Am, Tc and in some cases also Cm (MOX-fuel) from light water reactor high active waste. The partitioning will be considered as part of the one-cycle PUREX process, the so called IMPUREX (IMproved PUREX) process. The choice of transmutation strategy will be further evaluated and the first feasibility study will be reported in 1992. In the case of actinide transmutation in thermal reactors it has been shown that Pu burning obviously can be more effective when replacing U by Ce in MOX fuel assemblies.

4.5.3.5 India

Significant quantities of actinides have been accumulated in the spent nuclear fuel in India. Calculations showed that there is a significant value of these actinides as fuel for both fast and thermal reactors [Cri 91]. Some of the recent efforts to separate actinides and fission products from PUREX HLLW was presented by Mathur [Mat 91]. Two organophosphorus extractants, CMP (dihexyl-N,N-diethyl carbamoylmethyl phosphate) and CMPO (octyl(phenyl)-N,N-diisobutyl carbamoyl methyl phosphine oxide) have been tested together with TBP. Experiments with both simulated and actual HLLW have been performed.

4.5.3.6 Italy

In 1991 the Italian organization ENEA was converted from the "Italian State Commission for R&D on Nuclear and Alternative Energy Sources" to the new "Italian State Organization for New Technology, Energy and Environment" and a drastic reduction of activities in the nuclear field is planned. The new ENEA R&D program on nuclear energy includes development of reactor safety and radioactive waste management, and only a few scientists will be studying P-T. There is not any transmutation research program in Italy, but a lot of experience was collected in the earlier P-T studies in the period 1974-1982, which has resulted in some conclusions about actinide transmutation in fission reactors [Buc 91, Buc 92]. One of the conclusions was that LWR's are not able to effectively solve the actinide transmutation problem. Concerning partitioning there is an interest to develop a process for separation of long-lived radionuclides from HLW [Gro 91, Gro 91b]. Some of the interest is focused on separation techniques that can be utilized for HLW stored at the ENEA experimental facilities. A five year, more basic research, partitioning program was initiated in 1991, to evaluate the chemical processes and design of an optimized flow-sheet for separation of actinides Tc, Sr and Cs from HLW. This program is partly financed by the CEC, see 4.6.2.

4.5.3.7 Korea

The current Korean nuclear waste management policy is direct disposal of spent nuclear fuel as was presented in refs [Cri 91, Yoo 91]. The spent nuclear fuel is at the moment temporarily stored in water pools at the nuclear power plants but an intermediate storage is planned to be built in

1997. It is difficult to find a site location for the intermediate storage facility as well as the planned final storage facility, due to the public concern against underground storage of nuclear waste. Even though Korea do not reprocess their spent fuel, it is a strong interest to evaluate the possibility of P-T. A long-term R&D program on P-T has been initiated by the Korean Atomic Energy Research Institute (KAERI), which includes development of partitioning technology, establishment of nuclear data bases and experimental studies of transmutation. Some experiments to evaluate the possibility to transmute long-lived radionuclides will be performed in the Korean Multi-Purpose Reactor (KMPR), which will be in operation in 1993.

4.5.3.8 Netherlands

The Netherlands Energy Research Foundation (ECN) is engaged in a research program on transmutation of actinides and fission products, with the intention to contribute to the international P-T efforts [Abr 91, Abr 92]. The program includes i) strategy and scenario studies, ii) physical research and iii) demonstration the possibility of ^{99}Tc and ^{129}I transmutation. Calculations show that LWR's can consume their own ^{99}Tc production in case some surplus technetium is added to the core. If technetium is homogeneously mixed with the fuel, it would, however, be difficult to reprocess such fuel with current reprocessing technology. If technetium is heterogeneously loaded to a core, self-shielding effect would require even higher initial surplus loading. The best option for technetium transmutation would be a high flux thermal neutron field obtained in for example an accelerator driven facility. An experimental program using the Petten High Flux Reactor (HFR) have been initiated together with CEC to study the transmutation of ^{129}I, see 4.6.2..

4.5.3.9 Sweden

In Sweden the nuclear waste management policy is final disposal of the spent nuclear fuel in a deep geological repository. It is, however, stated by the Government that Sweden is not committed to this deposition method, and alternative methods must be evaluated [Ryd 91]. There is a strong interest in the scientific community to evaluate the feasibility of P-T, and a coordination group between different universities has been formed. At present only few scientists are involved with questions related to P-T. Swedish scientists were involved in the earlier P-T projects (1974-1982) and the partitioning research resulted in the CTH-process for separation of actinides and some fission products from HLLW [Lil 91, Lil 92]. Theoretical investigations of transmutation in nuclear reactors were also performed at Studsvik during this time.

4.5.3.10 Switzerland

The nuclear waste management policy in Switzerland includes deposition of HLW in a geologic repository. Swiss spent nuclear fuel is reprocessed

according to foreign contracts and there is an interest in how to handle this waste, when it is returned. The Paul Scherrer Institute (PSI) has announced a strong interest to participate in the international P-T efforts within several topics, including i) theoretical physics, ii) accelerator physics iii) construction of a spallation neutron source, iv) handling of actinides [Str 91]. A continuous spallation neutron source (SINQ) is planned to be in operation 1995 at PSI [Atc 92], which can be used for basic spallation research. Some of the recent work related to accelerator-based transmutation is presented in refs. [Wen 92, Wyd 91, Wyd 92].

4.5.3.11 United Kingdom

In the UK there is no interest in P-T, one wants to maintain the current fuel cycle policy, which is based on reprocessing technology [Bus 91, Tan 91, Tan 92, Web 91]. The waste management and disposal technology will be developed to fulfil the required safety guidelines within the short- and long-term. Plutonium recycled from reprocessing will be used in fast reactors. The once through fuel cycle and P-T are regarded as threats to the UK policy and a development program for cost reduction of reprocessing will be performed in order to make this policy competitive to the direct disposal option. Furthermore, development of the fast reactor and its fuel cycle will continue.

4.6 INTERNATIONAL P-T PROJECTS

4.6.1 The OECD/NEA program

The OECD-Nuclear Energy Agency (NEA) "International Information Exchange Programme" was initiated in 1988 by a suggestion from the Japanese Government, who suggested a program related to separation and use of actinides. The OECD/NEA program has been presented by Stevens [Ste 92]. Formally this exchange program is intended to run for five years and the subjects covered includes i) physical and chemical properties of elements generated in the nuclear fuel cycle, ii) nuclide partitioning technology and iii) nuclide transmutation. The program is limited to the members of the NEA together with the CEC. Three information exchange meetings have been arranged, see 2.1.2. [OECD 91, OECD 91b, OECD 92], and some investigations by consultants have been founded by this program.

Other activities within OECD/NEA can also be of importance for P-T studies. The NEA Data bank will be used for nuclear and chemical data and for comparing different codes used in transmutation calculations [Bla 91, Mil 91, Nor 91]

4.6.2 The CEC program

The CEC activities in the field of P-T of long-lived radionuclides have been presented in refs. [Cec 91, Koc 92]. CEC was involved in an earlier

P-T program during the second half of the 1970'ies. Results from this program were presented at two international symposia in Ispra [Hag 77, Hag 80]. Because of the rather pessimistic conclusions from the earlier P-T efforts, see 2.1.1, the direction of research was changed in the 1980'ies towards breeder reactor experiments and studies of more efficient extractants for some actinide liquid wastes. New fuels containing minor actinides were developed and tested in fast reactors. The reemerged interest in P-T in the late 1980'ies has influenced the CEC research activities in several ways. The Joint Research Centre (JRC) in Karlsruhe is reviewing the ongoing research activities and new research programs are now carried out. Furthermore, the CEC research program (1990-1994) on "Radioactive Waste Management" includes work on transmutation strategies, which was requested by the European Parliament. A new evaluation of the feasibility of P-T will be done, and an experimental program on partitioning is in progress, as will be discussed below.

The CEC work is performed in its own laboratories (JRC's) and in cooperation with other European laboratories on the basis of cost-sharing contracts. Some of the projects with relevance to transmutation are:

* *Development of a second SUPERFACT fuel containing minor actinides which will be irradiated in the PHENIX reactor at a higher burn-up than the first experiment (see. 4.2.2), (JRC Karlsruhe, CEA France).*
* *Development of ^{99}Tc and ^{129}I targets for transmutation studies in Petten HFR, see 4.5.3.8, (JRC Karlsruhe, JRC Petten, ECN Netherlands)*

Within the CEC Radioactive Waste Management program an experimental research program on actinide partitioning from high-level waste has been initiated. Four tentative separation flow sheets have been identified for experimental investigations, including the extractants TBP, diphosphine oxides, CMPO, diamide compounds and branched tripyridyl-triazine. Some of the projects within this program are:

* *Determination of the extraction behavior of actinides and possibly also technetium from strongly acidic HLLW (CEA France, KfK Germany, ENEA Italy)*
* *Synthesis of new extractants (University of Reading UK)*
* *Flow-sheet design for the separation of trivalent actinides from lanthanides (CEA France, KfK Germany, ENEA Italy)*
* *Determination of the irradiation stability of new extractants (JRC Karlsruhe)*

4.6.3 The IAEA program

During the mid 1970'ies IAEA conducted a coordinated research program on the "Environmental Evaluation on and Hazard Assessment of the Separation of Actinides from Nuclear Wastes followed by either Transmutation or separate Disposal". The results and conclusions were published in 1982 [IAEA 82]. During the 1980'ies there has been some

activities on the partitioning of specific elements from HLW as presented in ref. [IAEA 89]. The strong interest of P-T in the late 1980'ies resulted in an "Advisory Group Meeting on Partitioning and Transmutation of Actinides and Selected Fission Products from HLW" at IAEA [IAEA 91]. In the final document from this meeting, IAEA was recommended to coordinate the efforts of P-T in countries that are not members in other international organizations with efforts in the member countries of OECD and CEC [IAEA 91b]. Some of the recent IAEA P-T activities are presented in ref. [Cri 91].

5 R & D ISSUES

Development and implementation of a P-T system would require a long-term research effort and should not be regarded as a short-term alternative to the planned back-end policies. It is obvious that the debate concerning P-T reflects different national and international policies. In the UK, P-T is regarded as a threat that could compete with their reprocessing fuel cycle. In the USA, P-T is evaluated and compared with their planned back-end policy [Ram 92b], whereas Japan has a more long-term program with different R&D issues to be evaluated before any decisions about P-T will be taken. As has been mentioned before, one can expect that development of P-T may be too costly and resource consuming for a national development program, and an international cooperation would therefore be needed.

Beside the economical feasibility there are several technical issues that have to be evaluated, see 5.1.

5.1 SOME TECHNICAL R&D ISSUES.

5.1.1 Nuclear data

There are important needs for a large number of special nuclear data for transmutation purposes as was pointed out at the OECD/NEA meeting at the Paul Scherrer Institute [Cie 92, Cie 92b]. There are different cross sections needed depending on the transmutation concept, e.g. medium energy protons, light-ions and fast and thermal neutrons. Even if a large amount of data already is available from previous fission and fusion research and other basic research fields, there is still a lot of data needed, especially for high energy reactions. Several key-data are urgently needed in some transmutation concepts, including i) the total neutron yield, surface spectra and angular distribution of neutrons obtained when bombarding a thick spallation targets by medium-energy protons (< 2 MeV), ii) cross sections to determine the isotope production rates (inventories) for both proton and neutron induced reactions, iii) cross sections for atom displacement and gas production in construction materials.

5.1.2 Chemistry

Partitioning will be one of the key-issues concerning the feasibility of P-T. It is important to have a separation process for radionuclides that can meet the requested separation factors without producing secondary radioactive wastes. Both aqueous- and pyro-processing are considered, as presented in chapter 4. The proposed processes are different depending on the transmutation concept and the considered transmutation strategies. Concerning R&D in partitioning it is important to consider the overall requested reduction of the amount of a radionuclide

by P-T, as discussed in 3.2.3. The unavoidable incomplete transmutation in a single irradiation will have a significant influence on the requested separation and recycling efficiency. The less material that is transmuted in a single irradiation, the higher partitioning efficiency would be required to meet the overall reduction goal. To design an optimal partitioning process a lot of basic research is needed initially. From basic chemical data it is possible to identify different separation systems and have a flow-sheet design. In a later phase it is important to have large-scale demonstrations of the partitioning processes, to show the adequacy of equipment and to verify the separation efficiencies and losses. The proposed partitioning processes include several new extractants and new technology, and one can therefore assume that a large research effort is needed before these can be technically mature.

5.1.3 Accelerator and reactor technology

In several of the P-T proposals, a large linear proton accelerator will be utilized. Some parts of the accelerator have been developed and experimentally tested, whereas the whole accelerator never has been built. A large and costly R&D issue will therefore be to build and demonstrate operation of a large accelerator, made up of the proposed individual components. The requested linear accelerator should continuously produce a proton beam of 1-2 GeV at a current of 100-300 mA, which can be compared with one of the largest linear proton accelerators available today, at the Los Alamo National Laboratory, producing 800 MeV protons with a mean current of < 1 mA.

The possibility of transmutation in existing nuclear reactors, especially fast reactors, has been proven and there are also experience with production of reactor fuels containing minor actinides. To increase the capacity of transmutation, several different reactor concepts have been proposed. Development of these reactors will rely on current reactor technology, but still a significant R&D program would be needed. Design of an actinide burning reactor will demand better cross section measurements for minor actinides and some fission products.

5.1.4 Construction materials

Construction materials to be used in accelerator-driven transmutation facilities will be exposed to extremely high radiation doses and heat. It is therefore important to evaluate the displacement and gas-production cross sections, see 5.1.1, to evaluate the life time of e.g. target, target container and other nearby structural materials. Some experience can be obtained from material studies in connection with fusion research, but still particle irradiation and high energy reactions have to be evaluated.

5.1.5 Process control

The proposed P-T facilities will be technically complicated and a lot of R&D efforts would be needed for process control. The technical reliability must be high and adverse effects caused by human factors must be minimized. One of the major tasks in controlling a partitioning process will be analytical techniques for measurement of low levels of radionuclides within the process and in waste streams.

5.1.6 Radiological hazard evaluation

There are several arguments for implementation of P-T, including total reduction of long-lived radionuclides, safer containment of radionuclides in a repository and easier licensing of a repository. Regarding an international P-T effort there is a need for internationally agreed radiological hazard criteria, which can be evaluated in the framework of some international organisations, e.g. ICRP and IAEA. The radiological risk should be evaluated for both short- and long-term.

5.1.7 Technical hazard evaluation

A technical hazard evaluation has to be performed for each P-T facility. The possibility of technical and human faults and their consequences must be evaluated in e.g. fault tree analysis. Large safety studies would certainly be required in licensing of a P-T facilities.

5.1.8 Proliferation

All nuclear fuel cycles entail some proliferation risks. It is therefore important to evaluate possible proliferation risks with the proposed P-T options. There are several proliferation risks involved in current P-T programs including, i) that most of the partitioning processes will separate plutonium from the minor actinides ii) that there could be a significant tritium production in transmutation facilities using heavy water as moderator. In an international P-T effort, a more proliferation-resistant fuel cycle would be desirable.

5.2 POSSIBLE SWEDISH CONTRIBUTION TO INTERNATIONAL P-T EFFORTS

As been mentioned before, see 4.5.3.9, there is not any P-T program in Sweden, but there is a strong interest within the scientific community to participate in international P-T efforts. Only a few scientists are at present involved with questions related to other P-T programs, but some further small-scale activities are planned. Some of the research topics identified that can have a Swedish participation are:

* *Development of partitioning processes based on aqueous processing with both theoretical and experimental work.*

* *Theoretical physics and perhaps also experimental basic research.*
* *Technical safety analysis.*
* *Fuel cycle scenario studies.*

6 NUCLEAR ENERGY DEVELOPMENT THROUGH P-T

Implementation of P-T is more likely in case of continuation and expansion of nuclear power than otherwise. There is an ongoing debate if expansion of nuclear power with P-T should be a valid goal, or if P-T and nuclear power expansion should be evaluated independently. We believe that there are so small incentives to introduce P-T in the current dominating once through nuclear fuel cycle, so that the eventual introduction of P-T should be part of a long term decision about using new nuclear power technology as a future energy source.

The new nuclear technology development can be directed to more advanced systems including P-T. Fast reactors that make more efficient use of the nuclear fuel are already in an advanced stage of development. These reactors can efficiently be used for transmutation of "waste" actinides. There exist also several conceptual ideas of accelerator driven facilities that can be used for transmutation of actinides and long lived fission products and still have a net energy production. Such facilities have an advantage compared with nuclear reactors, consisting of an inherent safety because they are subcritical when the accelerator is turned off. The overall gain with the more advanced systems is a more efficient use of the nuclear fuel compared to current light water reactors (LWR:s) and a decreased production of long lived nuclear waste. Furthermore, all depleted uranium that remains from fabrication of nuclear fuel to LWR:s and the spent nuclear fuel itself can be used as fuel in these facilities. Implementation of new nuclear technology would, however, require a very large concentration of resources both economically and scientifically, which could look too high in the short time perspective.

6.1 ENVIRONMENTAL IMPACT OF P-T

Any P-T scheme must be designed with the goal to keep releases of radioactive substances to the environment extremely low as otherwise the benefits of the P-T operations would be lost. This would probably also imply a reduction of current releases to the environment of radioactive material from reprocessing and reactor operation.

If the transmutation operations have a net electricity production, a P-T operation could somewhat lessen the load on uranium resources and hence have a small positive effect on the environment near uranium mines.

When spent fuel is deposited in a well designed underground repository in a geologically stable formation predicted releases of radioactive elements to the environment are very small. In comparison the possible reduction in such releases by use of a P-T system would be insignificant from an environmental point of view. However, in case today unforseen

phenomena or processes would lead to releases to the environment in the far future there might be some difference in favor of the P-T approach.

7 CONCLUSIONS AND OUTLOOK

7.1 GENERAL CONCLUSIONS

It can be considered that, with some reservations, the technical feasibility of partitioning and transmutation, P-T, can be regarded as established, whereas it is difficult to see any cost or major safety incentives for P-T with the present nuclear fuel cycle scenario.

Insurance against the unknown is the most powerful argument for P-T. By removing long lived isotopes from nuclear waste one will have an unchallengeable ability to predict future effects due to the simple fact that if radionuclides do not exist, they can not cause any effect.

Introduction of P-T will not eliminate the need for radioactive waste disposal. But reducing the long term potential risks can make it easier to have decisions about site location, site licensing and how to build a geologic repository. However, new problems will arise regarding siting and safety of the necessary reprocessing and transmutation facilities. It should also be remembered that long lived radiotoxic nuclides in practice cannot be quantitatively destroyed by transmutation. There will always be some fraction of these nuclides present in the high active waste remaining after partitioning and in secondary waste streams. It is therefore important to consider the global short and long term distribution of long lived nuclides as a result of P-T, so that it would not exceed the most pessimistic predictions about the long term distribution from direct disposal of spent nuclear fuel or disposal of vitrified HLW.

Several countries have P-T research programmes. Japan, France and CIS have presented national programmes. In the USA several groups are involved with large activities and a national program seems to be in progress. A number of countries, including Sweden, have small or medium sized activities of relevance to the global P-T option.

At present, transmutation using reactor neutrons and accelerator based spallation neutron sources is considered. Of these neutron sources the reactors are, by far, in a more advanced stage of development, while accelerator driven neutron sources of adequate type only are in a R&D stage. The general demand on a neutron source is a high neutron flux available for transmutation. Due to limited neutron fluxes, secondary neutron capture reactions in transmutation products and relatively low cross sections for some transmutation reactions it is impossible to achieve a high transmutation efficiency in a one cycle irradiation. It would therefore be necessary to make several irradiations of the radionuclides intended for transmutation. This can be achieved by sequential irradiations interleaved with chemical separations or by a continuous separation of the transmutation products from a solution that is irradiated on-line.

R&D on partitioning processes should concentrate on the radionuclides which are of major concern because their long half-lives and radiotoxicity which make them to a potential hazard for a very long time. In this connection it is important not only to calculate the dose from the actual radionuclide, but also consider the specific activity by including stable isotopes. This can decrease the expected dose to man for some nuclides, due to limited uptake. When defining quantitative goals for a partitioning process it must be remembered that there is a strong coupling between the required separation efficiency, transmutation efficiency and the tolerable losses to secondary waste streams. For example, if the amount transmuted in a single irradiation is low, it will require many recyclings including partitioning. The demands of the partitioning efficiency and the limit of losses to secondary wastes will therefore be extremely high to meet the overall reduction goal for a specific radionuclide. Both aqueous processing and pyroprocessing have been considered for partitioning. The current knowledge of aqueous based processing is large, due to more than 40 years of experience, whereas pyroprocessing should be regarded to be in a R&D stage. It is therefore too early too make any final conclusions about partitioning procedures. In the presented P-T programmes one can generally say that the research on partitioning has been included but much larger resources must be allocated to this field in the future in order to make P-T more realistic.

7.2 WHAT HAS CHANGED SINCE EARLIER P-T ASSESSMENTS?

The earlier P-T studies in the late 1970s and early 1980s were concerned with transmutation of actinides in nuclear reactors. The general conclusion was then, "Since the long term hazards are already low, there is little incentive to reduce them further by P-T". The reemerged interest in P-T has been motivated by different individuals and organizations who claim that changes of factors involved in the earlier conclusions, technical development and some entirely new factors, in combination would justify reexamination of the P-T option. Some of these changes are:

- The radiotoxicity of actinides has been reconsidered according to the new guidelines from the International Commission on Radiation Protection (ICRP). This makes transmutation of actinides more favorable than earlier.

- By using accelerator based spallation neutron sources instead of fission reactors it is possible to have high neutron fluxes purely for transmutation purposes and a low formation of new long lived radionuclides. This have several advantages, one of which can be exemplified by the transmutation of ^{237}Np that can be achieved with a net gain of neutrons, due to the high fission cross section of the short lived ^{238}Np.

- Not only actinides but also fission products that can cause a long term potential hazard are considered to be transmuted in the new P-T proposals.

- New technological developments of reprocessing technology and advances in robotics can also increase the applicability of P-T.

7.3 THE IMPACT OF P-T ON GEOLOGIC REPOSITORY

There is a worldwide scientific consensus that the present reference nuclear fuel cycle scenario, including geologic repository, gives adequate protection to mankind, but there is a strong interest to see whether further reduction of the long term potential hazard can be achieved and at what cost. The most serious criticism that can be put upon a geologic repository program is the limited ability to predict future effects, which can be exemplified by possible changes of chemical properties in the repository, caused by human activities on the ground surface.

It is important to decide in what time perspective the long term risks from a geologic repository should be evaluated. In some countries it is concluded that the long term risks are associated not with actinides, but with long lived, soluble and weakly sorbing fission products. This is true if the time perspective is limited to one million years, but after that time a substantial contribution will be due to the actinide ^{237}Np.

When evaluating the long term risks of a geologic repository it is important not only to look for radiotoxicity and radioactive inventory, because these factors can not be used alone as valid measures of risk. The risk has to be evaluated from all site specific parameters that have an influence on the intended function of a repository. The risk reduction benefits that P-T might offer depend on the release scenario involved. This can be exemplified with some actinides. If the release is solubility limited, P-T does not substantially reduce the risk of actinides, in a limited time perspective, due to the low solubility which prevents them from being problem in the first place. On the other hand if the release is not solubility limited or looking for very long time perspectives, P-T could significantly reduce the risk. This could be the case for some of the long lived soluble fission products. It should, however, be evaluated if the risk reduction possible by P-T could be achieved by better immobilization of these fission products at a lower cost than P-T.

Introduction of P-T can directly affect a geologic repository in several other ways. Some of the issues that have been discussed are benefits of heat removal and reduced risk of human intrusion into a repository. Concerning the heat removal, P-T of actinides could increase the capacity of a repository. Separation of cesium and strontium for a separate short term repository could increase the capacity even further. Possible problems with heat removal from a repository could, however, partly be

solved by having a longer cooling time before direct storage or by a different design of the repository. Regarding the scenario of human intrusion into a repository it seems more likely that the risk of intrusion will increase. This will be due to the lower safety requirements on such a repository because of a lower release risk.

Several other questions can be addressed to the impact of P-T on a geologic repository, but it could be worthwhile to ask if promotion of P-T will hide the fact that the absolute risk of radioactivity release, from the considered types of geologic repositories, is already very low?

7.4 NUCLEAR TECHNOLOGY DEVELOPMENT TRHROUGH P-T

Implementation of P-T is more likely in case of continuation and expansion of nuclear power than otherwise. There is an ongoing debate if expansion of nuclear power with P-T should be a valid goal, or if P-T and nuclear power expansion should be evaluated independently. We believe that there are so small incentives to introduce P-T in the current dominating once through nuclear fuel cycle, so that P-T should be introduced in a more long term decision about using new nuclear power technology as a future energy source.

Environmental aspects can be a reason to increase the efforts of nuclear power developments. To achieve a global decrease of the carbon dioxide emission in order to decrease the possible greenhouse effect, one has to change the energy sources. A recent German study showed that a doubling of current nuclear power in Germany would yield a positive net economic gain for the society and still meet the goal of a 30% reduction in the carbon dioxide discharge.

An increased interest in nuclear power would certainly increase the costs of the current fuel cycle which could make other fuel cycle scenarios more economically competitive.

New nuclear technology development can be directed to more advanced systems including P-T. Fast reactors that make more efficient use of the nuclear fuel are already in an advanced stage of development. These reactors can efficiently be used for transmutation of "waste" actinides. There exist also several conceptual ideas of accelerator driven facilities that can be used for transmutation of actinides and long lived fission products and still have a net energy production.

7.5 THE APPLICABILITY OF P-T TO SWEDISH CONDITIONS

In the short time perspective we can not see any reasons to delay the Swedish nuclear fuel cycle including final deposition of radioactive waste in a geologic repository. There are, at present time, no cost and no clear safety incentives for P-T in connection with waste management of LWR fuel that has been utilized in the once through fuel cycle. In a longer time

perspective there could be some incentives to use the spent nuclear fuel, instead of dispose of it in a geologic repository. This would, however, require a change in the Swedish moratorium on nuclear power.

Considering the energy content in uranium, only about 0.5 % is utilized in LWR:s, whereas as much as 70 % can be used in fast breeder reactors with current technology. This value could possibly be further increased with a fuel cycle including P-T, which speaks in favor of having an intermediate storage of the spent fuel until P-T has been thoroughly evaluated. If one assumes that introduction of P-T takes 20 years, the present available spent LWR fuel has cooled down to a level where its lower activity makes reprocessing easier at the time when P-T is practicable.

It is clear that P-T will be further evaluated in several countries and by international organizations, due to its potential as a long term energy resource combined with its potential for destruction of long lived radioactive waste. It is difficult to see that P-T could be developed on a national basis, due to the large efforts required. International cooperations are therefore essential and we believe that Sweden should join these common projects, at least on a small scale.

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Ann-Chatrin Nilsson[3]
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Scope of activities and main results

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W Dershowitz[1], K Redus[1], P Wallmann[1], P LaPointe[1], C-L Axelsson[2]
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