

**Partitioning and transmutation
(P&T) 1995****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) is reviewed and evaluated. Current national and international R&D efforts are summarized. It is concluded that P&T is scientifically feasible and the recent R&D in the field of P&T makes it more technical mature than a few years ago. Nuclear transmutation with energy production is feasible in nuclear reactors where fast and thermal breeder reactors are the most efficient for transmutation purposes. The operation of subcritical nuclear reactors by high current proton accelerators that generate neutrons in a spallation target is also an interesting option for transmutation and energy production, that has to be more carefully evaluated. These accelerator-driven systems (ADS) are probably the only solution for the transmutation of long-lived fission products with small neutron capture cross sections and actinide isotopes with rather small fission cross sections.

The requirements on the separation chemistry in the partitioning process depends on the transmutation strategy chosen. Recent developments in aqueous based separation chemistry opens some interesting possibilities to meet some of the requirements, such as separation of different actinides and some fission products and reduction of secondary waste streams. In the advanced accelerator-driven transmutation systems proposed, liquid fuels such as molten salt are considered. The partitioning processes that can be used for these types of fuels will, however, require a long term research program. The possibility to use centrifuge separation is an interesting partitioning option that recently has been proposed.

P&T is a complex issue with regard to environmental, safety, technological, economical, political and public acceptance aspects. In some countries (Japan, France and the Russian Federation) it has, however, received political or institutional backing as a complementary future nuclear fuel strategy. At present there seems to be no economical gain and only an insignificant reduction in future radiation doses from P&T as compared to the reference concept of the closed fuel cycle and direct disposal of spent nuclear fuel. However, some of these conclusions can be changed by future long-term research and/or by a change in the economical situation.

SAMMANFATTNING

Den nationella och internationella forskningen på transmutations- och separationsområdet sedan 1992 sammanfattas och utvärderas. Slutsatsen är att möjligheten till transmutation och separation (P&T) nu är vetenskapligt säkerställd samt att utvecklingen på området gjort detta mer tekniskt moget än tidigare. Nukleär förbränning och transmutation med energiproduktion är möjlig i kärnreaktorer. Mest effektiva är termiska och snabba bldreaktorer. En annan intressant möjlighet som bör studeras närmare är användning av acceleratordrivna underkritiska kärnreaktorer för såväl energiproduktion som transmutation av vissa långlivade avfallsnuklider. Sådana acceleratordrivna system är förmodligen den enda lösningen när det gäller transmutation av långlivade fissionsprodukter med små infångningstvårsnitt och av aktinidisotoper med små fissionstvårsnitt.

Kraven på separationsprocessens kemi beror på vilken strategi som väljs. Nyare utveckling på vattenbaserade separationssystem ger intressanta möjligheter att lösa några av de aktuella kraven, t. ex. separation av olika aktinider och klyvningsprodukter samt reduktion av mängden sekundärt avfall. Flytande bränsle i form av t. ex. saltsmältor föreslås i flera av de föreslagna acceleratordrivna transmutationsreaktor systemen. För denna typ av bränsle krävs betydande och långvariga FoU-insatser för utveckling av praktiskt/tekniskt användbara separationssystem. Användning av ultracentrifugering för detta ändamål har nyligen föreslagits.

Transmutation och separation innehåller komplexa problem i fråga om miljömässig, säkerhetsmässig, teknisk, ekonomisk, politisk och allmänhetens acceptans. I några länder (Japan, Frankrike och Ryssland) har tekniken fått politiskt och institutionellt stöd som en extra framtida kärnbränslestrategi. För närvarande tycks emellertid tekniken inte leda till någon ekonomisk vinst eller till signifikant framtida dosreduktion i jämförelse med normala bränslecykler, med eller utan direktdeponering av använt kärnbränsle. Några av dessa slutsatser kan emellertid ändras genom framtida forskningsresultat och/eller förändringar av den ekonomiska situationen.

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1 BACKGROUND

The reemerged interest for P&T started during the late 1980ies and one can say that the starting point for the large interest was the Specialist Meeting on Accelerator-Driven Transmutation Technology for Radwaste and other Applications held in Saltsjöbaden, Stockholm, Sweden, 24-28 June 1991 [1]. A review of the different P&T projects was presented in some reports during 1992-1993 [2-4]. This report reviews the developments in P&T from 1992 to 1995.

2 INTRODUCTION

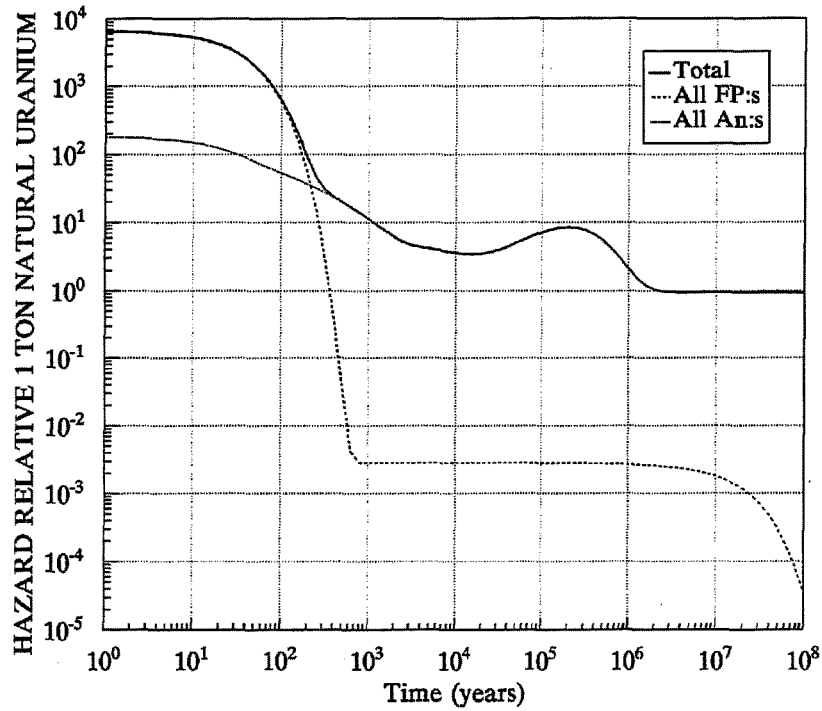
Many difficult problems of nuclear power technology are related to the neutron economy of the fission chain reaction. The excess of neutrons - if available - may be used for converting non fissile materials into nuclear fuel as well as for transmutation of some long-lived radioactive isotopes into short-lived or even stable ones. An accelerator-driven subcritical reactor can on the other hand be designed so that we no longer are bound by the neutron economy. Besides transmutation of long-lived fission products and actinide isotopes, it is also possible to breed plutonium from uranium and uranium from thorium in fast as well as thermal accelerator-driven systems (ADS). A review of ADS development up to the end of 1994 has been made by Condé et al. [5].

3 GENERAL CONSIDERATIONS REGARDING P&T

The transmutation process can take place in normal critical reactors or in accelerator-driven subcritical reactors (ADS). The major advantage of an ADS system is that it can be operated at a higher neutron flux and with less fuel than a normal critical reactor. It is often argued that ADS systems are inherently safer than critical reactors. To some degree that may be true, but the differences are not very large. i) Shut-down requires in both cases some action from an active or passive safety system. Failure to shut-down will have similar consequences in both cases. ii) For equal thermal power, the inventory of shortlived radionuclides is very similar. Thus, the decay power will be the same fraction of full power for critical reactors as for ADS. In both cases an emergency cooling system is required. However, the power level will decrease more rapidly in an ADS system in case it contains a much smaller inventory of longlived radionuclides (probably only true for an ADS with liquid fuel and on-line separation).

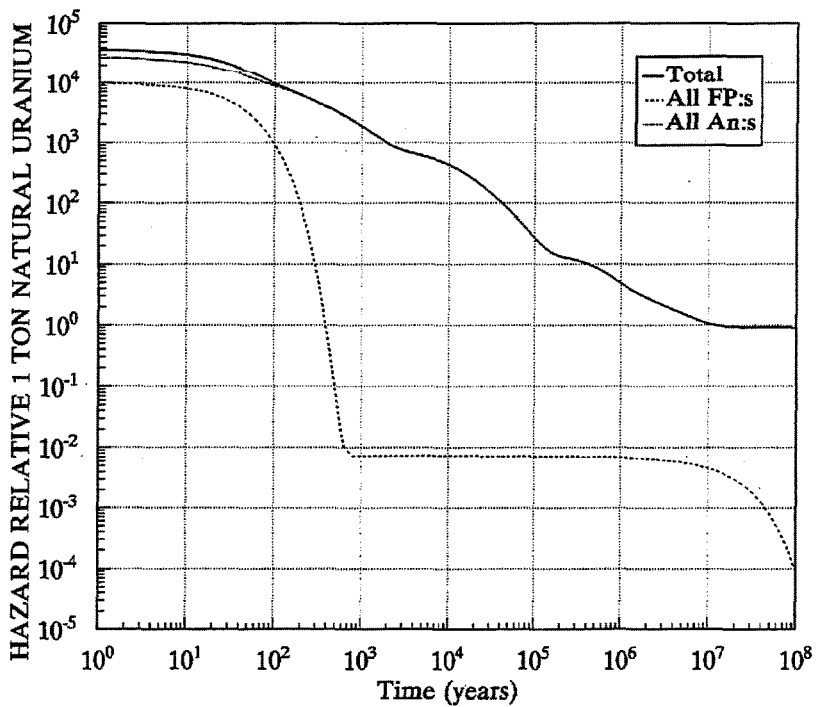
3.1 Radionuclides intended for P&T

It can be emphasized from the point of potential hazard, without taking into account the geological barriers in a repository, that the actinides are the most important



Figur 1 Relative hazard index of spent BWR fuel based on the same weight of natural uranium and MPC_w from ICRP publication-2 and -6.

radionuclides that should be investigated in a P&T concept. There are, however, some long-lived fission products that may play a role over a long period of time. A



Figur 2 Relative hazard index of spent BWR fuel based on the same weight of natural uranium and current ALI-data from ICRP.

comparison of the radiotoxicity for actinides and some long-lived fission products in spent nuclear fuel is summarized in Table 1.

The potential hazard of radioactive nuclides has often been expressed in terms of the dilution factor needed to reach the activity limit for drinking water (MPC_w , Bq/m³) or, more recently, in terms of the number of annual limits of oral intake (ALI, Bq) or in terms of the specific dose resulting from ingestion (D, Sv). In order to facilitate comparisons of risks the calculated values are often divided by the corresponding value for various amounts of natural uranium (e.g. for spent fuel, the hazard value for the same amount of natural uranium in equilibrium with its daughters) resulting in various relative hazard indices.

The data from which potential hazard is calculated are in principle nuclide amounts, half-lives and dose-factors, the latter given in various publications of ICRP. However, new results on the metabolism of elements and on the biological effects of radiation have over time lead to several changes in the dose-factors. The newest data from ICRP give an increased relative importance of the actinides present in spent fuel and radioactive waste, see Figures 1 and 2. This is due to a combination of reduced dose-factors for some uranium daughters and for some fission products and increased dose-factors for several actinide isotopes.

Table 1. Comparison of the radiotoxicity for actinides and some long-lived fission products in spent nuclear fuel after 100 years cooling time.

Nuclide	$t_{1/2}$ (years)	Abundance ¹ (g/tIHM)	Spec. act. ¹ (Bq/tIHM)	Dose factor ² (Sv/Bq)	Spec. dose (Sv/tIHM)
²³⁷ Np	2.1×10^6	689.5	1.8×10^{10}	1.7×10^{-6}	3.0×10^4
²⁴¹ Am	4.3×10^2	972.7	1.2×10^{14}	1.7×10^{-6}	2.1×10^8
²⁴³ Am	7.4×10^3	145.5	1.1×10^{12}	1.7×10^{-6}	1.8×10^6
²⁴⁵ Cm	8.5×10^3	6.9	4.4×10^{10}	1.7×10^{-6}	7.3×10^4
²³⁸ Pu	8.8×10^1	142.8	9.1×10^{13}	1.3×10^{-6}	1.1×10^8
²³⁹ Pu	2.0×10^6	5641.	1.3×10^{13}	1.3×10^{-6}	1.6×10^7
²⁴⁰ Pu	6.6×10^3	1611.	1.4×10^{13}	1.3×10^{-6}	1.7×10^7
²⁴¹ Pu	1.4×10^1	9.5	3.6×10^{13}	2.5×10^{-8}	9.0×10^5
²⁴² Pu	3.8×10^5	439.5	6.4×10^{10}	1.3×10^{-6}	8.0×10^4
⁷⁹ Se	6.5×10^4	5.2	1.4×10^{10}	5.0×10^{-9}	68.
⁹³ Zr	1.5×10^6	770.4	7.2×10^{10}	7.1×10^{-10}	51.
⁹⁹ Tc	2.1×10^5	849.	5.3×10^{11}	1.7×10^{-9}	883.
¹⁰⁷ Pd	6.5×10^6	181.1	3.4×10^9	1.7×10^{-10}	0.6
¹²⁶ Sn	1.0×10^5	11.8	1.2×10^{10}	1.7×10^{-8}	206.
¹²⁹ I	1.6×10^7	215.1	1.4×10^9	2.5×10^{-7}	345.
¹³⁵ Cs	2.3×10^6	262.7	8.6×10^9	5.0×10^{-9}	43.

¹Reference: SKI Technical Report 90:18, J. O. Liljenzin, April 1990 (tIHM = ton initial heavy metal)

²The dose factor is the ratio between the yearly maximum dose for workers (50 mSv) and ALI.

It can be shown for some long-lived fission products that if the normal chemical concentration of the element in the human body is totally exchanged by the same element having the isotopic composition in spent nuclear fuel one obtains an insignificant increased effective dose equivalent, corresponding to activities far below ALI (Annual Limit of Intake). As an example, when ^{134}Cs and ^{137}Cs have decayed away the normal body content of cesium would give a 50 year dose of about 1.2 mSv if all that cesium was replaced by old cesium from spent fuel. This is due to a combination of long half-life and dilution by stable isotopes already in the waste. Assuming further that the element which contains radioisotopes has escaped from a deep geologic repository its relative concentration of radioisotopes will be reduced by dilution if stable isotopes of the element exists in nature. This should also be taken into consideration when deciding which of the fission products that should be transmuted.

3.2 Transmutation

The most important nuclear reactions considered for transmutation are induced by neutrons, causing either fission or neutron capture. To have an acceptable transmutation rate and efficiency, high neutron fluxes are needed and the transmutation devices with the greatest potential at present are high flux reactors or accelerator-driven subcritical reactors. An overall reduction of the radiological

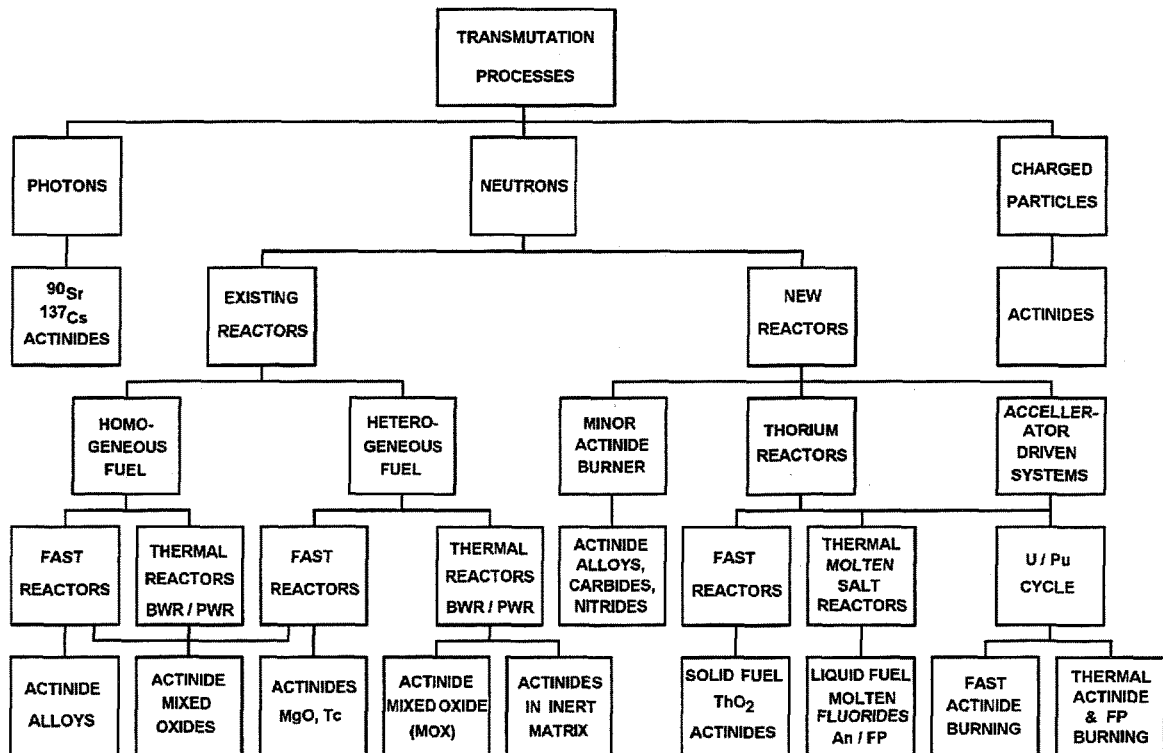


Figure 3 Proposed transmutation schemes

hazard requires development of new technology, e.g. an accelerator driven transmutation system. In order to reach this expected goal a major breakthrough in technology is required. Since there are several proposals of how P&T should be managed it has brought forward a variety of proposals of technological solutions, which are summarized in Figure 3.

The basic question "What is the objective of transmutation?" is not yet clearly answered in different P&T proposals. Several answers are given, e.g;

- (i) destruction of active waste from conventional nuclear reactors,
- (ii) production of nuclear energy without conventional nuclear reactors,
- (iii) destruction/denaturation of surplus nuclear weapons materials, i.e. ^{239}Pu , or
- (iv) rapid production of weapons grade material, i.e. ^3H , ^{233}U and ^{239}Pu .

In order to have an international collaboration in the field of P&T it is therefore essential that one can agree on the objective of transmutation. Since there is a worldwide scientific consensus that the current radioactive waste management concept provides adequate protection to the population and the environment by sufficient confinement of radionuclides, an overall goal could be reduction of the potential long-term radiotoxicity of actinides and some long-lived fission products. By this, the effects of unforeseen radionuclide releasing phenomena might be reduced.

3.3 Reprocessing and Partitioning

Some form of reprocessing of spent fuel is a prerequisite for transmutation of long-lived hazardous nuclides. By reprocessing we understand any method by which remaining uranium, and usually also plutonium, is recovered from the spent fuel. These elements typically constitute ~96% of the decladded spent fuel (incineration of uranium is usually not considered in any P&T concept). Industrial scale reprocessing is today based on the PUREX process. Improvements of the operating conditions and reduced generation of secondary wastes has over time led to decreasing losses of uranium and plutonium. Whereas losses of about 0.5% was assumed feasible to achieve 10 years ago, current information from the LaHague plant show that losses today have decreased to around 0.2% [6].

Further reductions of the losses from reprocessing are anticipated if the PUREX solvent TBP (tributyl phosphate) is replaced by a monoamide based solvent in existing plants. Monoamides only contain C, H, O and N atoms and are thus completely burnable whereas incineration or other means of destruction of TBP produces phosphoric acid or inorganic phosphates, which ultimately end-up as radioactive waste.

At present neptunium is not recovered during reprocessing of spent fuel. However, it is known that careful adjustment of the nitrous acid concentration in the primary separation step can force almost all neptunium to follow uranium [7]. By adding a uranium - neptunium separation step in the uranium purification circuit it is then possible to recover both elements in pure form.

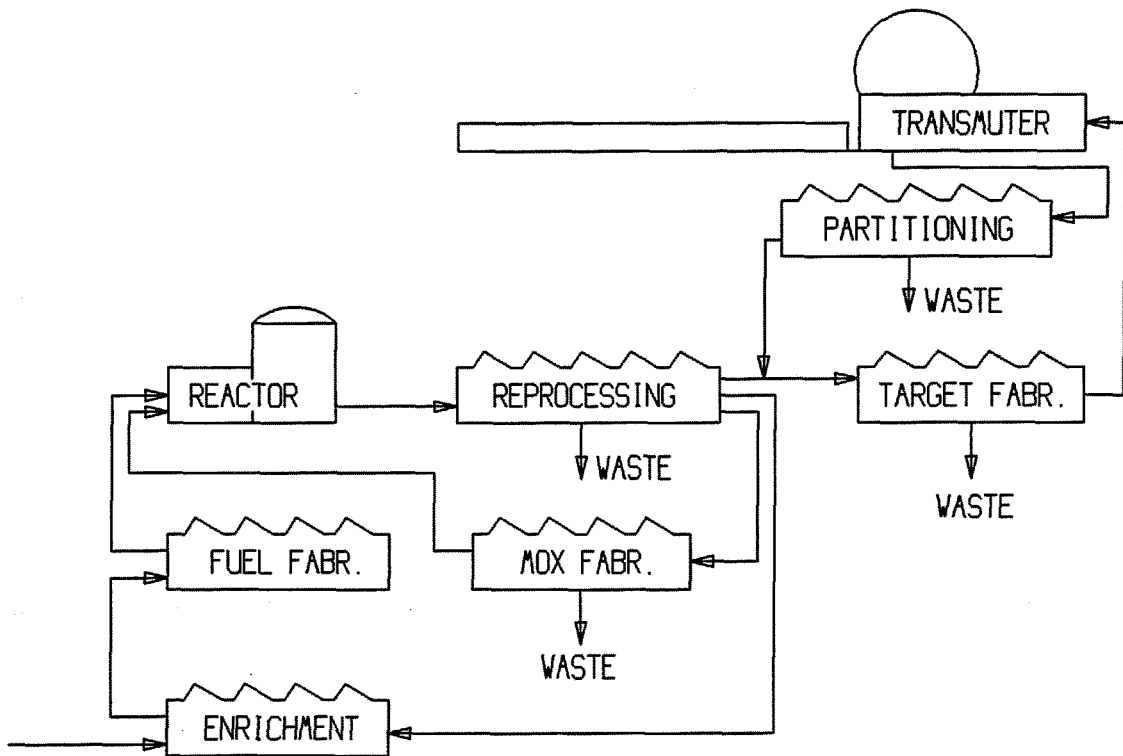


Figure 4 Fuel cycle with heterogeneous transmutation/partitioning operations.

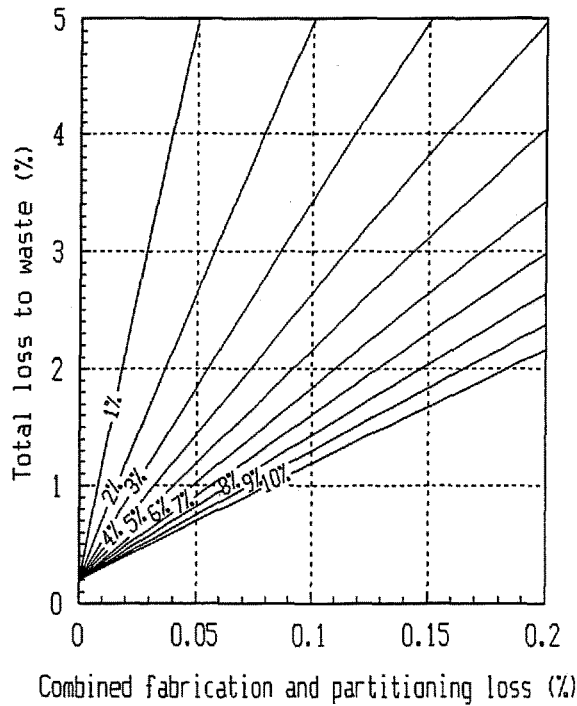


Figure 5 Relation between total loss to wastes and single pass losses in transmutation with separate partitioning for some single pass target burn-out values (assumed reprocessing loss 0.2%)

All americium, curium and heavier actinides together with neptunium and traces of uranium and plutonium currently follow the high-level liquid waste (HLLW) and end-up as vitrified radioactive waste. Current research in Europe (partly funded by EU) and in Japan aims at a complete recovery of all actinides from the HLLW stream before vitrification. As mentioned before, almost all neptunium can be recovered by a suitable adjustment of operating conditions in existing PUREX-based plants, but an almost complete recovery of all actinides requires the addition of further treatment of the HLLW stream. To simplify the HLLW treatment the development of suitable new reagents is necessary. Primary candidates are various diamides and TPTZ-derivatives (TPTZ = tri-pyridyltriazine). A suitable diamide based solvent would coextract all actinides and lanthanides (but no other fission products) from HLLW without acidity reduction. Separation of actinides from lanthanides could then be made using a TPTZ-derivative. Both types of reagents are completely incinerable.

Bowman et al. at Los Alamos National Laboratory have suggested the use of ultra centrifugation of molten metals, molten salts or aqueous solutions as an alternative to normal chemical separation of fission products from actinides [8]. They also propose this method for separation of the Cs-isotopes prior to transmutation of ^{135}Cs or ^{137}Cs as the separation effect only depends on the difference in mass. Without such a separation, neutron irradiation of the isotopic mixture would lead to the production of radioactive ^{134}Cs and ^{135}Cs by neutron capture in stable ^{133}Cs as well, thus counteracting the purpose. However, development of the necessary centrifuge technology is a major undertaking of comparable size to the original development of aqueous reprocessing technology. Many additional issues regarding the transformation of wastes into acceptable final forms, the arising and minimization of secondary wastes etc. must also be researched and solved. Bowman et al. offer two arguments for development of centrifuge based separation, (i) it should be more proliferation safe than conventional chemistry and (ii) an almost absence of chemistry would make it more attractive (to those who distrust chemistry?). Both arguments are probably irrelevant, but it would nevertheless be interesting to test this technology. The main drawback could be the possibility it may offer to separate uranium and plutonium isotopes secretly for military use.

3.4 Recycling and losses

The reduction factors one wants to achieve for individual radionuclides in the nuclear waste will determine the number of cycles and the required chemical separation efficiency. 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. A complete fuel cycle including reprocessing, transmutation, separation and recycling of untransmuted target material is shown schematically in Figure 4. For each type of irradiation target, either a single element or a group of elements, only a part can be transmuted in one irradiation. The target must then be processed, remaining material recovered, converted to new target material and recycled. The

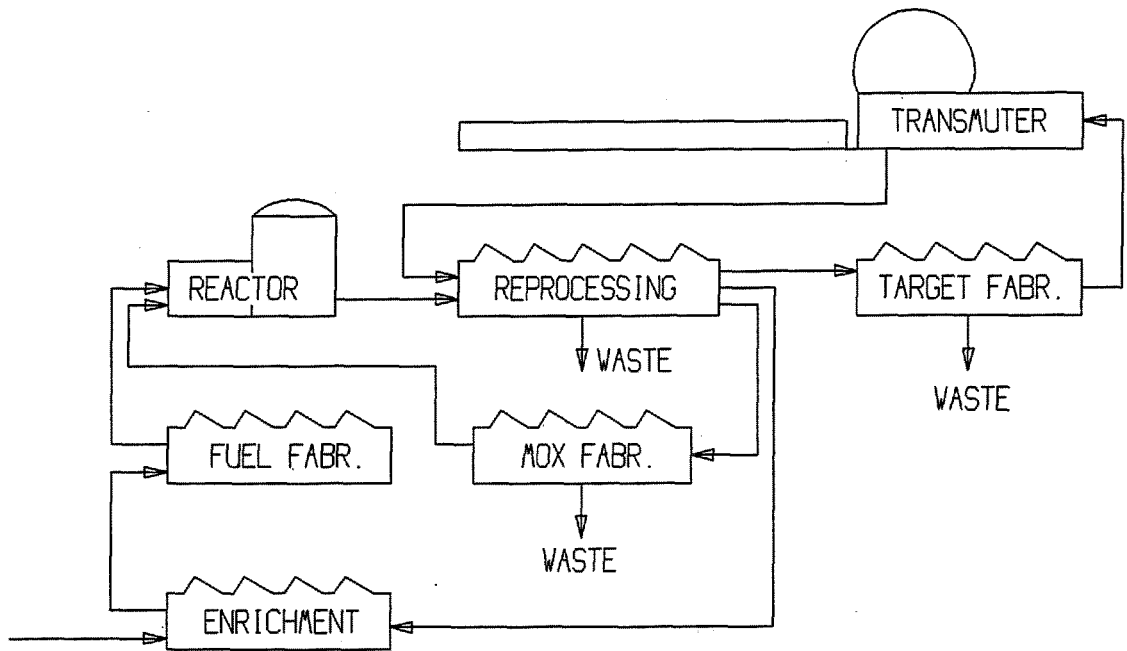


Figure 6 Fuel cycle with homogeneous reprocessing of spent fuel and transmutation targets.

total loss to waste as a function of the combined fabrication and partitioning losses for various single pass transmutation efficiencies is shown in Figure 5.

An alternative recycling scheme involves homogeneous separation of untransmuted target material and unprocessed fuel in a common reprocessing plant with integral recovery of the minor actinides as shown in Figure 6. The total loss to waste as a

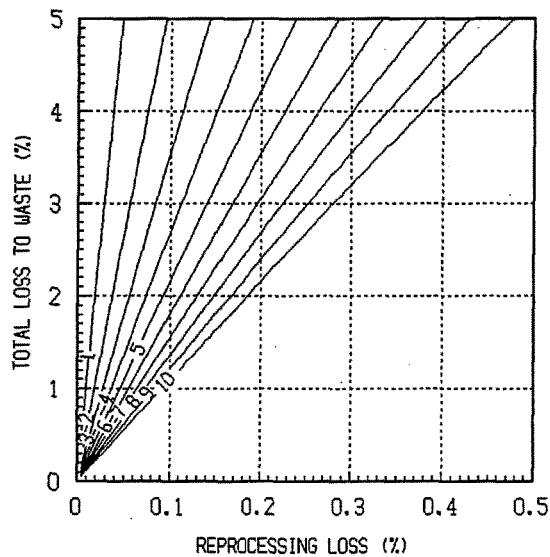


Figure 7 Relation between total loss to wastes and single pass losses in reprocessing with homogeneous reprocessing for some target burn-outs (%) and a fabrication loss of 10% of the reprocessing loss.

function of partitioning losses for various single pass transmutation efficiencies is shown in Figure 7.

In the calculations for Figures 5 and 7 it is assumed that the single pass burnup value is independent of the number of irradiation cycles. This will not be exactly the real case since the isotopic composition of the elements will change with irradiation time. However, these two fuel cycle scenarios clearly shows that the losses in the partitioning process in the homogeneous fuel cycle and the combined losses in target fabrication and partitioning process in the heterogeneous fuel cycle must be extremely low in order to minimize the total loss to waste. Assume that we operate with a special target reprocessing plant (heterogeneous fuel cycle, c.f. Fig. 4) and have a goal to destroy ≥ 99 % of the amount of a single actinide in spent nuclear fuel. It is further assumed that a reasonable loss of this element in the reprocessing step is 0.5 % and a that the burn-out per cycle is 5 %. Then the combined losses in target fabrication and partitioning steps must be ≤ 0.025 %, which is ≥ 20 times smaller than the initial loss in the reprocessing operation. It is not clear that an industrial scale process can be designed and operated with such very low losses.

4 P&T PROGRAMMES IN PROGRESS

Several countries have P&T research programmes in progress. In France, Japan and the Russian Federation P&T has received political or institutional backing as a complementary future nuclear energy strategy. In the USA several groups are involved with some activities. Furthermore, a number of countries have presented small or medium sized activities of relevance to different P&T options.

4.1 *The French SPIN and ISAAC programs*

In the French nuclear program, recycling of plutonium in LWR's as MOX-fuel started on regular basis 1990.

The reprocessing plants in France uses aqueous based separation methods to recover plutonium and uranium. Development of separation techniques will therefore rely on solvent extraction since they will fit into the technology used. The extractants are also required to contain only carbon, hydrogen, oxygen and nitrogen to assure a completely incinerability.

P&T of long-lived elements is one of the objectives in a French law which was stated at the end of 1991. CEA (Commissariat à l'Énergie Atomique) therefore started the SPIN (Separation-Incineration) program to meet these requirements. The aim of the SPIN program is to reduce the volume and the radioactivity of the wastes that is intended for a geological repository. The SPIN program is divided into two sub-programmes; the short and medium term program PURETEX and the more long-term program ACTINEX [9].

PURETEX is concerned with the improvement of the traditional PUREX process. Plutonium and neptunium are to be more efficiently separated by using a monoamide which is a completely incinerable and does not contribute to the secondary waste like TBP does.

The ACTINEX program concerns selective and specific separation of all actinides and long-lived fission products. The separated actinides are intended to be transmuted in PWR:s or FR:s. There are three main separation routes within the ACTINEX program;

- * Coextraction of actinides and lanthanides with a diamide in a first step and then separation of the actinides from the lanthanides in a second stage using e.g. TPTZ.
- * Development of new extractants, e.g. picolinamides, which extracts the actinides selectively leaving the lanthanides in the aqueous phase. This route is favorable since the large amounts of lanthanides do not have to be extracted.
- * Separation of Am at an oxidation state higher than three

The technical feasibility of the use of U-Pu and U-oxide for the recycling and transmutation of the minor actinides Np and Am has been proved in the SUPERFACT irradiation experiment [10]. Some results are given in Table 2. Further experiments are planned in the SUPERFACT project.

The ISAAC program, which stands for Investigations on Subcritical Systems Driven (in French : Alimentés) by Accelerators, has been setup at the Nuclear Reactor Directorate of CEA, with participation of other CEA units, in order to investigate the physics of subcritical accelerator-driven systems, in particular in the following areas:

- a) high intensity accelerators
- b) spallation in different target types: methods, data and validation
- c) technology of the targets
- d) subcritical, source-driven, multiplying systems
- e) system studies for different applications.

Table 2. Some results from the SUPERFACT irradiation experiment in PHENIX [10].

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

In area a) and b), the CEA is already active with experimental programs. A significant program is underway to validate the calculation tools used against data from experiments performed at SATURNE.

For point d), an exploratory experimental program is envisaged at the MASURCA facility at CADARACHE, where high accuracy measurement techniques are available for subcritical systems with k_{eff} in the range 0.995 - 0.900. That facility offers unique flexibility and can accommodate any type of external source-driven system. Some preliminary experiments could be performed early 1995.

For point e) some works have been already performed and published.

For point c) - no program has been defined yet but it is under evaluation and some significant experimental program will be defined.

The coordinator of the ISAAC-program is dr. M. Salvatores, the financing of the program corresponds to approximately 10 manyears/year for some years.

4.2 *The Japanese OMEGA-program*

In 1987 the Atomic Energy Commission in Japan approved the program "Long Term Program for Research and Development on Nuclide Partitioning and Transmutation Technology". 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.

Most of the R&D within the Omega program is today performed at JAERI, PNC and CRIEPI. The first phase of the program is expected to end during 1996. Phase-II is intended to be executed in the period 1997 - 2000 and will include engineering tests and demonstration of concepts selected on basis of the results from phase-I [11].

At JAERI, a conceptual study of an accelerator-driven transmutation systems is under way [12]. Two subcritical accelerator-driven reactor systems are studied: i) a system containing about 3.2 tons of actinides as a solid metal alloy, ii) a system containing 5.4 tons of actinides as a chloride melt. Both studies are based on the use of a linear 1.5 GeV proton accelerator with about 40 mA beam current. Incineration capacity would be about 250 kg/year of minor actinides for either system. However, much of the Japanese effort seems to be concentrated on the use of existing or new (actinide burner) reactors as transmutation devices [11].

The four group partitioning process developed at JAERI during phase-I and based on DIDPA (diisodecylphosphoric acid) and DTPA (diethylenetriaminepentaacetic acid) will be tested with actual HLLW in the NUCEF facility at the Tokai research center. Dissolution of enriched uranium for criticality tests began in the fall of 1994. Hot operation is expected to start during 1995. Studies of amide extractants have also been started at JAERI.

4.3 *Russian Federation*

The Russian Federation has a P&T program. Several research centers directed by the Ministry of the Russian Federation for Atomic Energy (Minatom) are involved in P&T research [13]. The radionuclides intended for transmutation are the minor actinides and the fission products ^{99}Tc and ^{129}I . Fast reactors are the main tool considered for actinide burning, but accelerator-driven systems are also under investigation. Transmutation in both specially designed and commercial fast reactors are investigated. Preliminary results show that sodium-cooled BN-type reactors could effectively be used as minor actinide and plutonium burners. For the nuclear safety, the concentration of minor actinides in the MOX fuel have to be less than 5 %. Under these conditions one BN-800 reactor can transmute up to 100 kg of minor actinides per year, which amounts to the minor actinides from 3 VVER-1000 type reactors. Production and test irradiation of MOX and Pu-inert matrix fuels has been made [14]. Reprocessing of these test fuels have also been examined on a laboratory scale. A preliminary investigation has shown that the reactor based P&T option is twice as expensive as direct disposal of solidified wastes and it is thought that "construction and operation of an accelerator-driven P&T facility is unpredictably expensive" [13].

4.4 *USA*

In the USA several different strategies for transmutation of nuclear waste have been presented. The objectives of the R&D on P&T in the US is not clear. However, three reasons have often been given; (i) development of a proliferation safe nuclear energy system, (ii) rapid 'destruction' of surplus weapons grade plutonium, and (iii) a nuclear energy system without long-lived radioactive waste. Some research on accelerator-driven systems in the US also focuses on the production of tritium for weapons use (APT) [15]

The National Research Council of the National Academy of Science has recently finished a report on the status of partitioning and transmutation in the US and made recommendations for future research in this area [16]. Six US P&T schemes are discussed in the report. The schemes are based on the use of: i) a LWR, ii) an advanced liquid-metal reactor (ALMR), iii) an accelerator-driven subcritical reactor (ATW), iv) a particle bed reactor (PBR), v) the PHOENIX accelerator-driven reactor, and vi) the Clean Use of Reactor Energy (CURE) concept consisting of a mix of fast and thermal reactors. The report concludes that only the first three of the proposed schemes have reached a sufficient degree of detail to be evaluated and that the sixth is fairly independent of the P&T system chosen. As a result of the evaluation the committee found no evidence that advanced P&T would have sufficient benefit for the US to delay development of the first repository for spent commercial reactor fuel. However, the fuel deposited in any final repository should be retrievable within the next 100 years because a future P&T system might be implemented within that time frame. Furthermore, the committee concludes that all

three of the evaluated P&T schemes could be effective in the transmutation of transuranium elements. Both the LWR and ATW based schemes have potential to transmute ^{99}Tc and ^{129}I , but the ATW might have an advantage due to its higher flux. Finally, the committee recommends that "over the next decade the US should undertake a sustained but modest and carefully focused research and development program on selected separation and transmutation technologies, with emphasis on improved separation processes for separating LWR and transmuter fuels beyond the existing plutonium and uranium (PUREX) extraction process and for fuels containing more actinide elements and selected fission products".

- * The ATW project has mainly abandoned its initial concept of a combination of CANDU and slurry fuel technology for transmutation by thermal neutrons. The current concept is based on a subcritical thorium burner with the LWR actinide waste serving as a fissile starter. Table 3 shows estimated power consumption for various extent of fission product transmutation. Three targets have been tested at the LAMPF facility: i) a stopping length target of Pb for ATW, ii) a distributed $\text{W}/\text{D}_2\text{O}/^3\text{He}$ stopping length target for APT, and iii) a 99% enriched ^7Li target with a multiplying lead blanket. The nuclear safety of the ATW system has been reviewed by Gudowski et al. [3].

4.5 *South Korea*

A series of transmutation systems are under investigation at KAERI (Korean Atomic Energy Research Institute) with the objective to define the optimal transmutation device [17]. The basic study involves three stages: i) selection of nuclides to be transmuted, ii) investigation of the physical characteristics of each nuclide, and iii) study the most favorable neutron energy environment for the transmutation. The studies performed indicate that the existing LWR and LMFBR cores would not be satisfactory with regard to the transmutation rate.

4.6 *China*

Studies have been made at the China Institute of Atomic Energy, Beijing, regarding the possibility to transmute minor actinides in the core of CEFR (China Experimental Fast Reactor, 65 MWth/25 MWe, sodium-cooled reactor) [18]. Calculations indicate that a transmutation rate of 9.4 kg/year of minor actinides can be achieved in this reactor. Future plans include irradiation tests with special fuel pins containing various amounts of minor actinides.

4.7 *The European Community*

The Commission of the European Communities placed research contracts on a strategy study of P&T with three European research institutions at the end of 1991. These contracts have been extended until the end of 1994. CEA (France) has been

Accelerator Power Required for Transmutation in % of electrical power produced	Long-lived fission products		
	Nuclide	Half-life (y)	PWR discharge (kg/y and GWe)
10%	⁹⁹ Tc	2.1x10 ⁵	10.0
	¹²⁹ I	1.6x10 ⁷	1.8
	⁷⁹ Se	<6.5x10 ⁴	0.1
	¹²⁶ Sn	~1x10 ⁵	0.4
	¹³⁵ Cs	3.0x10 ⁶	3.0
16%	⁹⁰ Sr	28.5	6.0
	¹³⁷ Cs	30.0	9.0
26%	⁹³ Zr	1.5x10 ⁶	10.0
	¹⁰⁷ Pd	6.5x10 ⁶	2.7
	⁸⁵ Kr	10.7	0.2

Table 3. Fission product burning capability as function of accelerator power consumption.

investigating the potential and cost of P&T of long-lived radionuclides. Siemens (Germany) has analyzed the use of advanced converter reactors for transmutation and their safety. ECN (The Netherlands) has been preparing and assessing nuclear data libraries for transmutation studies.

The new European program 'Nuclear Fission Safety' includes several areas of research on P&T. It is not yet known how much funds will really be allocated to P&T areas and how many applications regarding P&T issues that have been approved. Swedish research groups participate in at least two applications which have been approved by the EU.

4.8 *Other international organizations*

Several other international organizations are also involved in P&T research work. The Organization for Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA) coordinates a series of information exchange workshops and promotes coordination among OECD member states. IAEA organized a Technical Committee Meeting on the safety and environmental aspects of P&T near the end of 1993 [19]. The almost total absence of participants from the US (one only) at the IAEA-meeting is remarkable. IAEA also organized a Technical Committee Meeting on Advanced Fuels with Reduced Actinide Generation in November 1995. There was no participant from the US at this meeting. The proceedings will be published during 1996.

At CERN, a group headed by C. Rubbia is working on the conceptual design of a so called "Energy Amplifier". In principle a subcritical assembly of thorium and ^{233}U driven by a beam of high energy protons from a small three stage cyclotron which provides a proton beam of 10-15 mA at an energy of about 1 GeV. The originally suggested design used a moderated system [20], but after re-evaluation that design has been abandoned in favor of an unmoderated assembly cooled by liquid lead. The present conceptual design - a modular unit - has a nominal thermal power of 1500 MW, $k=0.98$ and uses a mixed oxide fuel [21]. This system was also proposed as an efficient burner for destruction of weapons grade plutonium [22] (see appendix).

5 CONCLUSIONS

It can be considered that, with some reservations, the scientific feasibility of partitioning and transmutation, P&T, can be regarded as established, whereas it is difficult to see any cost or major short term safety incentives for P&T compared with the present nuclear fuel cycle scenarios. Transmutation using either reactor neutrons or accelerator based spallation neutron sources is still 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 are still in an early R&D stage. However, accelerator driven transmuters may be necessary as a final stage to destroy long-lived actinides with small fission cross sections remaining after many incineration cycles in fast and thermal reactors. Thus they should be seen as a complement to nuclear reactors rather than a replacement.

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. Aqueous processing, molten-salt 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 to make any final conclusions about partitioning procedures. However, current R&D within the European Community is mainly focused on extensions and improvements of existing aqueous separation technology. One can generally say that the research on partitioning has been included to a larger extent in current and new research programmes than before. However, much larger resources must be allocated to this field in the future as part of an continued effort in order to make P&T a realistic alternative to current fuel cycles.

It is clear that introduction of P&T will not eliminate the need for radioactive waste disposal because long-lived radiotoxic nuclides cannot be quantitatively destroyed by transmutation without some losses to waste. 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. Even if there is a worldwide scientific consensus that the present reference nuclear fuel cycle scenarios, including geologic repository, gives adequate protection to mankind, 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, but it is important 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 and waste forms, is already very low?

As already pointed out in our previous report [2], implementation of P&T is more likely in case of continuation and expansion of nuclear power programmes 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. 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.

P&T is still under further evaluation 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 cooperation is therefore essential. However, European (and Japanese) motives for research on P&T seems to be different from those expressed by the US.

6 RECOMMENDATIONS

Based on the information presented in this report the following general recommendations are given:

- * P&T should be regarded as an alternative treatment method for spent nuclear fuel in Sweden.
- * SKB is recommended to support research, international cooperation and coordination of different research efforts in the field of P&T.
- * Development of P&T is a field in nuclear technology which is interesting for new students. A supported research in P&T at Swedish universities is, therefore, essential for the recruitment of good students and to keep the competence in nuclear technology. Even if P&T does not become a reality in Sweden there will be a strong need of good competence in nuclear technology.

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APPENDIX

ACCELERATOR-DRIVEN SYSTEMS - CONCEPTS, PHYSICS AND OTHER ASPECTS

A1 INTRODUCTION

In the last few years hybrid systems have been proposed for different purposes. Accelerator-Driven systems (ADS) on fast neutrons for the incineration of higher actinides was proposed at Brookhaven National Laboratory (PHOENIX-project) and now is carried out in Japan as a part of OMEGA-programme. Los Alamos National Laboratory has developed several ideas to use a hybrid system on thermal neutrons with a linear accelerator for incineration of plutonium and higher actinides as well as for transmutation of some fission products in order to reduce the long-term radioactivity of nuclear waste. Three years ago Carlo Rubbia and his European group at CERN proposed a cyclotron based hybrid system to produce nuclear energy based on the thorium fuel cycle.

The technology to accelerate charged particles to a high energy has been well demonstrated in the last years, as well as the necessary target technology. Extension of this technology to high current beam acceleration is required. One of the reasons why many scientists are not attracted to this concept is their unfamiliarity with the technologies involved. Another important reason is that the spallation reaction is not exothermic, hence the number of neutrons produced from this reaction may not be sufficient to make the system economically viable; this system cannot compete with a fast breeder or fusion hybrid reactor.

A2 SPALLATION, ACCELERATOR, TARGET, BLANKET

Spallation refers to nuclear reactions that occur when energetic particles (e.g. protons, deuterons, neutrons, pions, muons, etc.) interact with atomic nucleus - the target nucleus. In this context "energetic" means kinetic energies larger than about 100 MeV per nucleon. At such energies the nuclear reactions do not proceed through the formation of a compound nucleus. The initial collision between the incident projectile and the target nucleus leads to a series of direct reactions whereby individual nucleons or small groups of nucleons are ejected. At energies above a few GeV per nucleon, fragmentation of the nucleus can also occur. After the initial phase, the nucleus is left in an excited state and subsequently relaxes to its ground state by "evaporating" nucleons, mostly neutrons [A1].

The spallation process is illustrated in Figure 2-1, showing two stages of the process (intranuclear cascade and evaporation). For thick targets, high energy (> 20 MeV) secondary particles (and their progeny) can cause further spallation reactions in the target. For some target materials, low energy (< 20 MeV) spallation neutrons can enhance neutron production through (n,xn) reactions. For heavier nuclei, high

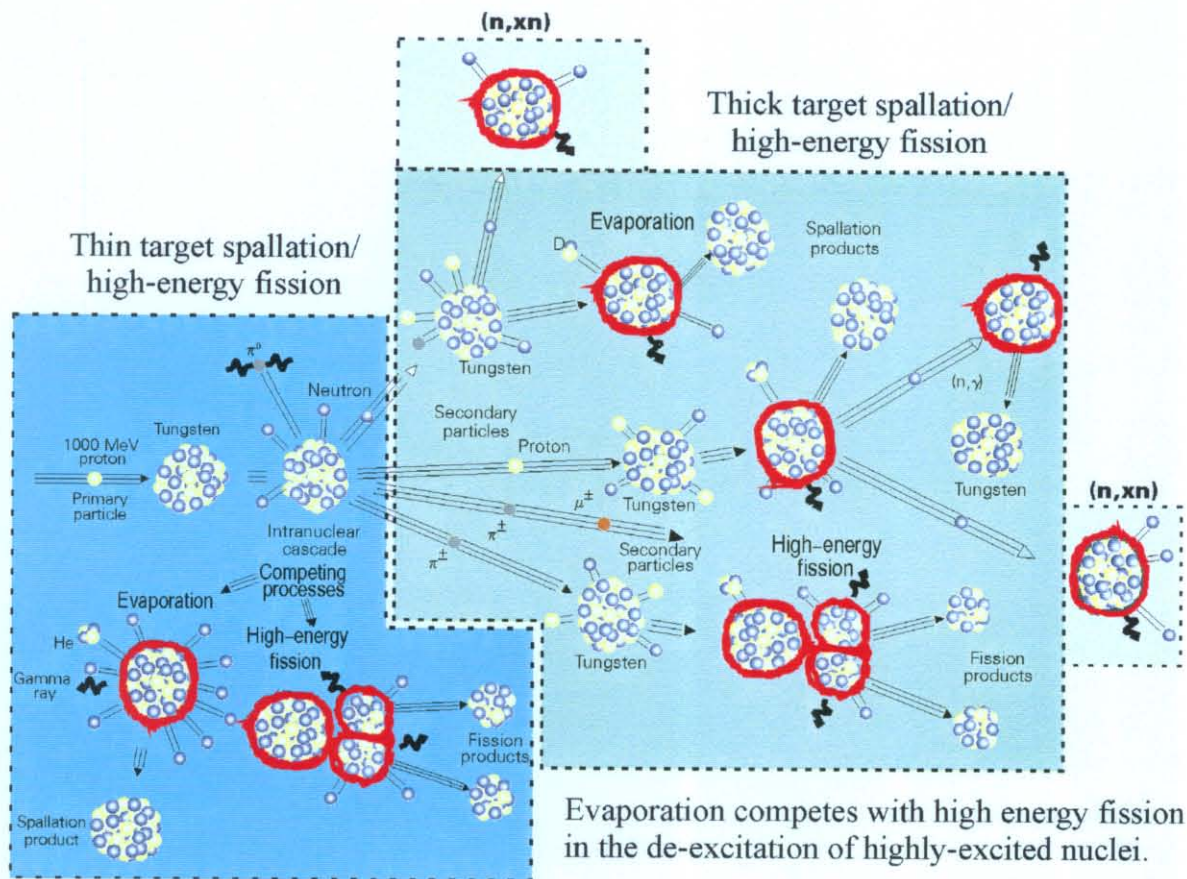


Figure 2-1. Illustration of high-energy-spallation processes in a thick target.

energy fission will compete with particle evaporation from highly-excited nuclei, see Figure 2-1. Tantalum, tungsten, lead and bismuth are examples of potential targets. Some target materials, such as ^{232}Th and ^{238}U , can also be fissioned by lower energy (~ 1 MeV to ~ 20 MeV) neutrons. Spallation, high energy fission and low energy fission produce different nuclear debris (spallation and fission products). Deuterons and tritons produce more neutrons than protons in the energy range below 1-2 GeV; thus, the efficiency of neutron production can be increased using them. However, the high yield of neutrons also from low energy deuterons and tritons can easily activate the low energy part of the accelerator if these particles are spilled from the beam; this may prevent hand-maintenance of the accelerator and increase operating costs.

The high power accelerator technology required for ADS has been under continuous development for the past decades. Linear accelerators, linacs, have been developed into highly reliable and efficient research (and military) tools. There is confidence in that a high power (200 mA, 1.6 GeV), continuous wave (CW) accelerator can be built at a reasonable cost [A2] (In October 1995 the US DOE committed to the demonstration of the accelerator technology for application to tritium production). On the other hand the technology of the circular proton accelerators, such as the segmented cyclotron or synchrotron recently improved so that a proton beam of 10-15 mA is achievable [A3]. The cyclotron does not require a large physical area

and has some other benefits compared to linac. In a recent evaluation it was found that the most efficient operation current for a cyclotron-type machine would be ~ 10 mA and for a linear accelerator ~ 100 mA [A4].

To provide external neutrons for a subcritical reactor, the proton beam must be stable, and the beam current well controllable so that there would be minimal power fluctuations in the subcritical assembly. Persistent power fluctuations may damage the fuel elements, especially in case of metal fuel. The cyclotron accelerator used as the PSI neutron source currently has a 5% fluctuation in beam power due to instability in the high current beam transport.

The function of the target in the ADS is to convert the incident high energy particle beam to low energy neutrons. These requirements can be summarized as [A5]:

1. Compact size to enable good coupling to the surrounding blanket,
2. High power operation, on the order of 10 to 100 MW,
3. High neutron production efficiency,
4. Reliable and low maintenance operation,
5. Safe and low hazard operation,
6. Small contribution to the waste stream.

It is believed today that molten lead or molten lead-bismuth eutectic (LBE) are the best target choices, meeting most of the requirements above. A problem with LBE, however, is the production of radioactive and highly mobile polonium isotopes, especially ^{210}Po , from high energy proton and neutron reactions on lead and bismuth. This will be a concern because the polonium in the LBE will be rapidly released at operating temperatures. Pure lead, on the other hand, has a lower polonium production, but higher operating temperatures. Further assessments are needed in order to make a choice between these target materials.

The thermal cross-sections for transmuting MA and FP is larger than the fast neutron cross-sections. However, the thermal neutron cross-sections of the transmutation products are also large. It is therefore desirable to remove the products in order to reduce unproductive losses of neutrons. The capture of fast neutrons by the fission products and by the structural material is smaller than for thermal neutrons and, from the point of neutron economy, a fast reactor is better than a thermal reactor. Also, one would like to take advantage of the high η -values for ^{239}Pu and the other actinides to produce extra neutrons by high-energy fission for use in transmutation of the long-lived fission products.

As mentioned above the thorium-uranium fuel cycle is very attractive for future ADS. It has two advantages over the traditional uranium-plutonium cycle used in most nuclear reactors:

1. The thorium-uranium cycle produces a smaller amount of higher actinides than the uranium-plutonium cycle, because of the small capture to fission ratio in ^{233}U and because of the presence of two other fissionable isotopes of uranium (^{235}U and ^{237}U) in the chain leading to plutonium and the other heavier actinides
2. The thorium-uranium cycle is perceived as safer than the uranium-plutonium cycle from a nuclear weapons proliferation standpoint, because of the

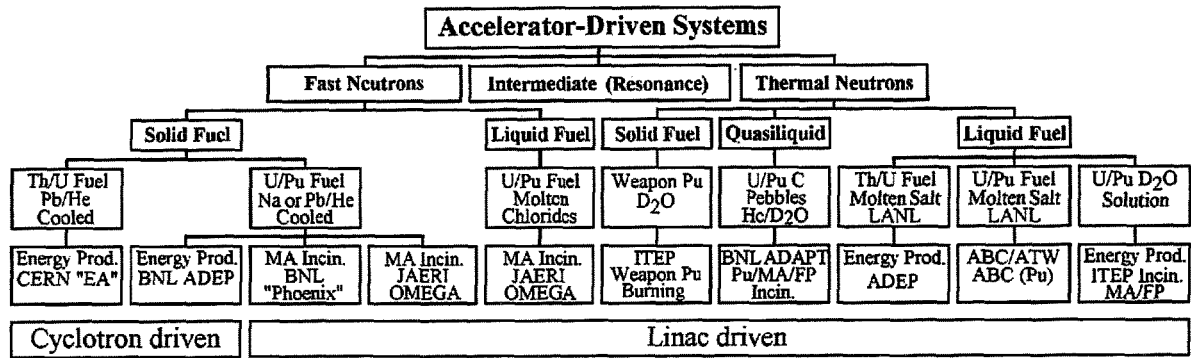


Figure 3-1. Classification of existing ADS concepts.

presence of ^{232}U (the mother of hard-gamma emitters) as a minor product of the cycle, and because it is imagined that an isotopic dilution of ^{233}U with depleted or natural uranium in the feed, or in the start-up fuel, would make ^{233}U difficult to extract in pure form.

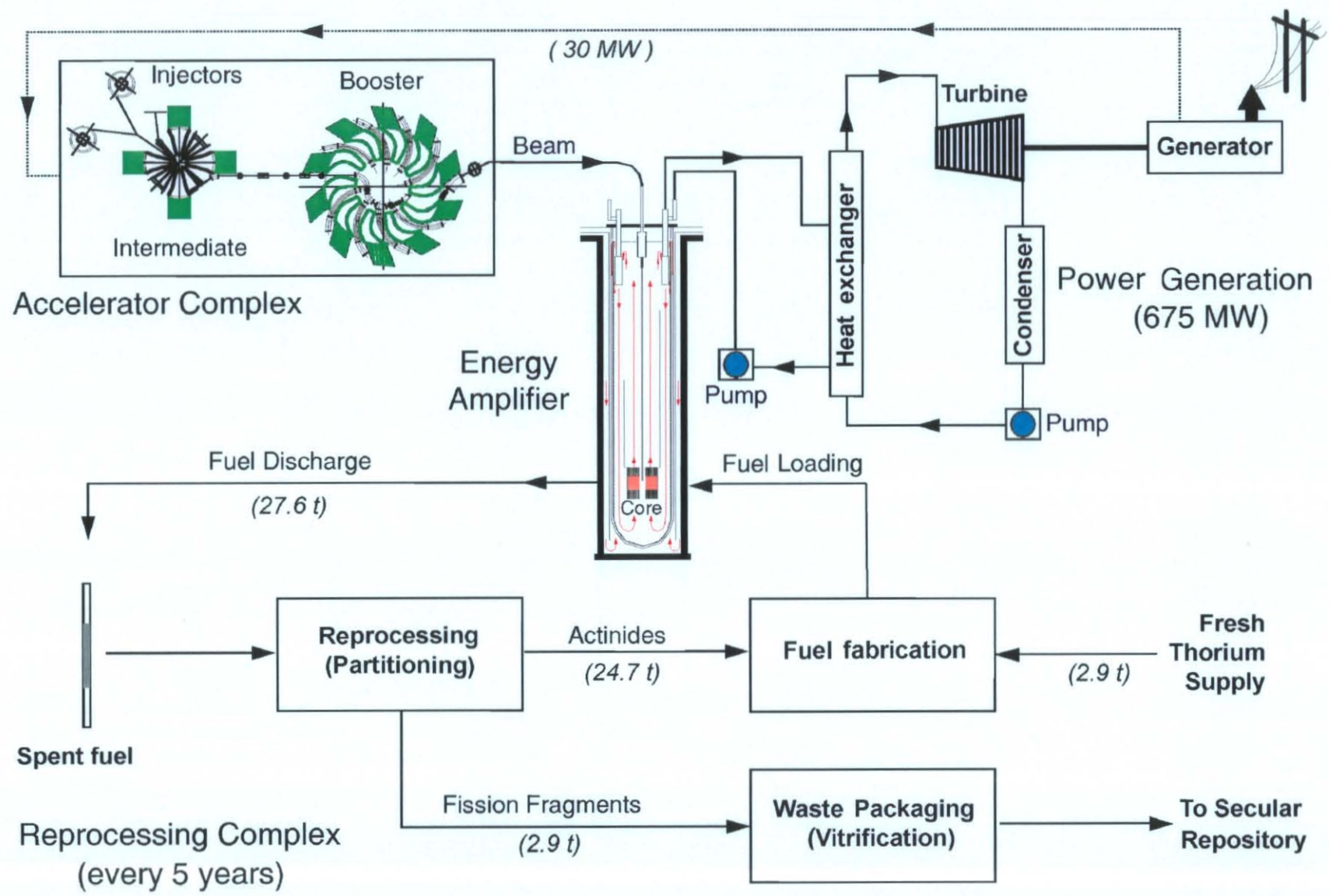
A3 DESCRIPTION OF THE EXISTING PROJECTS

Figure 3-1 shows a classification of existing ADS concepts according to their physical features and final objectives. The classification is based on neutron energy spectrum, fuel form (solid, liquid), fuel cycle and coolant/moderator type, and objectives for the system. ADS systems - like reactors - can be designed to work in two different neutron spectrum modes - on fast or on thermal neutrons. The CERN-group headed by C. Rubbia investigates the possibilities (CERN application to EC, 1995 [A6]) to design a system which will exploit the neutron cross-section resonances in what could be classified as a "resonance neutron" mode. Both, fast and thermal systems are considered for solid and liquid fuels. Even quasi-liquid fuel has been proposed based on the particle fuel (pebble bed) concept developed by BNL [A7]. The objective for some nuclear transmutation systems is to transmute existing nuclear wastes from light water reactors, mainly Pu and minor actinides, with or without concurrent energy production. These projects can be classified as an attempt to close the LWR-fuel cycle. Other systems are designed to take advantage of the thorium fuel cycle for energy production. As can be seen in Figure 3-1 most concepts are based on linear accelerators. However, the CERN-group and BNL propose to use a proton cyclotron.

A3.1 CERN

C. Rubbia developed his early ideas to design the accelerator-driven system (so called Energy Amplifier - EA) based on the thorium fuel cycle with solid fuel assemblies [A8]. However, it was later realized that a subcritical system with solid fuel and thermal neutrons moderated in light water would be impossible to construct except for very low power units. The problem with power peaking due to the very

Figure 3-2. Energy amplifier system according to Rubbia [A8-A12].



short neutron diffusion length in light water would require very sophisticated solutions like a distributed target, permanent fuel shuffling, many accelerator beams and so on. As the result CERN-group is now investigating a system based on a fast neutron spectrum.

An Energy Amplifier (EA) module consists of a 1500 MW_{th} unit with its dedicated 1 GeV proton accelerator of 12.5 mA, see Figure 3-2. A compact, reliable and modular cyclotron has been designed. A plant may consist of several such modules. The EA is a large, passive device into which a proton beam is dumped and the heat generated by nuclear cascades is extracted, without other major elements of variability. The delivered power is controlled exclusively by the current of the accelerator.

Rubbia estimates that after ≈ 700 years the radioactivity left is about 20 000 times smaller than in the spent fuel from an ordinary Pressurized Water Reactor (PWR) for the same energy production. Geological storage (10^6 years) can be virtually eliminated or at least strongly reduced ($\leq 600 \mu\text{Bq/J}_e$ after 1000 years). It could be further reduced ($< 40 \mu\text{Bq/J}_e$) by "incinerating" some of the nuclides [A9, A10]. The liquid lead cooled system is partially based on Russian work and experiences with liquid lead/liquid and lead-bismuth eutectic cooled reactors (e.g. Russian submarine reactors). This system has very good safety feature ensuring large safety margins and very good neutronics. However problems connected to chemistry - especially corrosive properties of high temperature liquid lead and of the Pb/Bi eutectic - put significant constraints on this concept.

The CERN-group already performed a demonstration experiment proving to a certain degree the feasibility of the Energy Amplifier concept [A11]. The experiment consisted of heat measurements in 3.6 tons of natural uranium irradiated by a high energy proton beam (1 - 3 GeV). The most interesting result of this experiment is the optimal proton energy for n-production by spallation processes in the thick target.

The activity of the CERN-group is based on the regular CERN-budget (the salaries) and concerted cooperation with different groups. The group is planning further experiments like studies of the phenomenology of spallation neutrons in a large lead block [A12] and seeking funding from various sources (EC, private funds, Italian government etc). EC has so far granted about 200 keCU to C. Rubbia.

A3.2 BROOKHAVEN NATIONAL LABORATORY, "PHOENIX"-PROJECT

The concept of a large-scale accelerator incinerator was studied by enlarging the early BNL-JRC(Euratom)-CERN concept using state-of-the-art technology. The Phoenix concept consists of modules of accelerator-driven subcritical lattices containing minor actinide fuel, see Figure 3-3. Conventional fast reactor technology is assumed, such as oxide fuel elements and sodium cooling. Each module resembles the core of the Fast Flux Test Facility (FFTF) with a $k_{\text{eff}} = 0.9$; however, from 1 to 8 target modules are aligned in front of a 104 mA beam of 1.6 GeV protons. The entire bank of reactor modules serve as a target for a proton beam that is expanded before entering the core region. With these parameters, eight modules would generate 3600 MW_{th}.

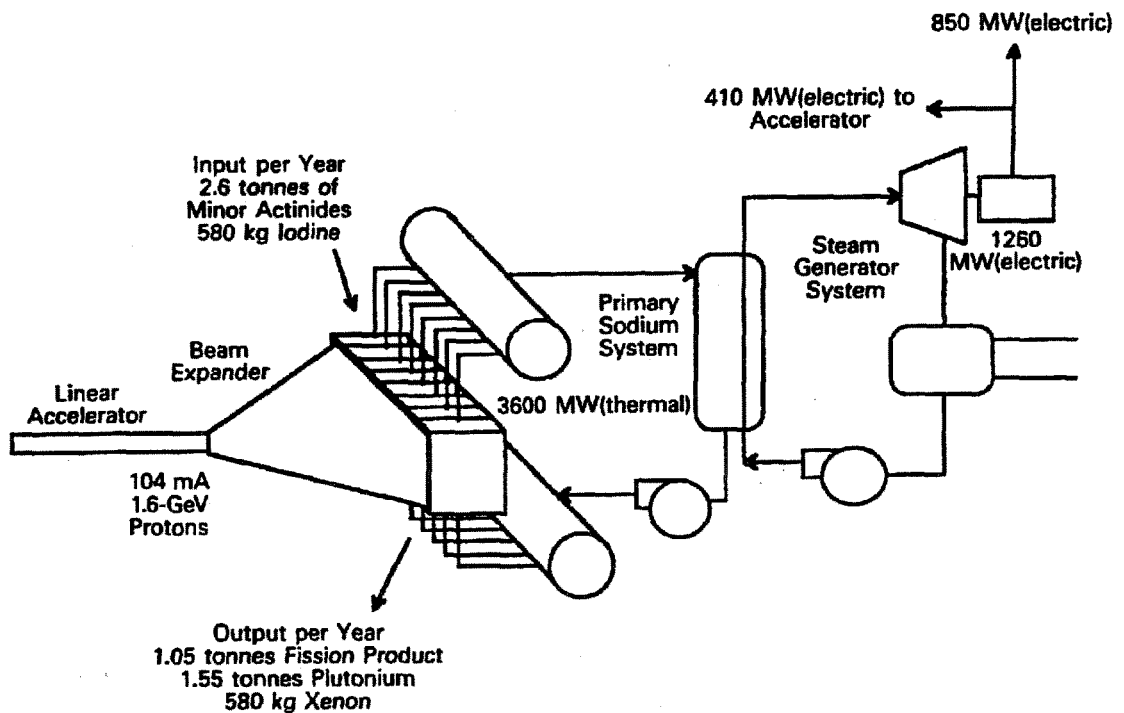


Figure 3-3. The PHOENIX concept.

The study showed that after 2 years of operation at 75% capacity, the average burn-up is 8.6%, with an additional 12.7% converted to plutonium. Using a 2-year cycle assures that most of the plutonium (> 85%) is ^{238}Pu , which is valuable as a long-term remote source of power, and also reduces concerns about degradation in the fuel or in the structural steels. During the 2-year reprocessing step, the plutonium and the fission products are replaced by new minor actinides from the LWRs. Plutonium not used for isotopic applications could be blended with plutonium from the LWR waste stream, so that the resulting mixture would contain enough Pu-238 and Pu-236 to make it difficult to use in weapons.

A3.3 JAERI OMEGA CONCEPTS

In Japan, a national program called OMEGA was started 1988 for research and development of new technologies for P-T of nuclear waste. Under the OMEGA program, the Japan Atomic Energy Research Institute (JAERI) is carrying out research and development for proton accelerator-driven transmutation, together with transmutation based on a fast reactor burner and an advanced partitioning technology.

The project on proton accelerator-driven transmutation at JAERI includes a conceptual design study of transmutation systems and the development of an intense proton accelerator. Two types of accelerator-driven transmutation system are proposed: a solid system and a molten-salt system. Both are specifically designed for nuclear waste transmutation purposes. In either system, an actinide loaded subcritical blanket is driven by a proton accelerator and utilizes the hard neutron spectrum to

burn actinides.

JAERI R&D includes the conceptual design study of accelerator-driven transmutation plant, the development of a spallation simulation code system [A13, A14] and spallation integral experiments [A15].

The small delayed neutron fraction from actinide fuel and a short neutron life-time coupled with a small Doppler coefficient tend to make the consequences severe of a reactivity-initiated transient in fast reactors. An accelerator-driven subcritical system with an external spallation neutron source can mitigate this safety problem and provide increased flexibility in design and operation. The power generation in the subcritical blanket can easily be controlled by adjustment of the incident proton beam intensity.

The goal of the design study is to develop technically feasible concepts for an accelerator-driven transmutation system. The proposed plant is designed as a dedicated system that would transmute about 250 kg of MAs per year, which corresponds to the actinide production rate from about 10 light water reactors of $\sim 3000 \text{ MW}_{\text{th}}$ each. The tentative capacity for the transmutation plant was based on the results from a preliminary strategic study of partitioning and transmutation systems. The subcritical blanket is required to operate at a multiplication factor of ≥ 0.9 to reduce the scale of proton accelerator and improve the energy balance of the system.

The budget of OMEGA program is about $3 \cdot 10^9$ yen/year, excluding salaries. Next year, the peer review of the OMEGA project is planned to be followed by a redistribution of the funding.

A3.4 LOS ALAMOS NATIONAL LABORATORY ADTT-PROGRAM

Nuclear systems under study in the Los Alamos Accelerator-Driven Transmutation Technology program (ADTT) have multiple objectives: the transmutation of nuclear spent fuel and weapons-return plutonium, as well as the production of nuclear energy from the thorium cycle, without a long-lived radioactive waste stream. The group at LANL claims that ADTT can phaseout production and eliminate world inventory of reactor and excess-weapons plutonium in 30 years [A16] and can provide an alternative to conventional nuclear reactors and allow the complete utilization of fertile fuel (thorium) without excess breeding or the use of enriched material at any time in the cycle [A17]. Finally ADTT systems can reduce the requirements for long-term storage of radioactive waste [A18]. LANL uses general ADTT nuclear design in all three basic applications of the concept. ADEP (Accelerator-Driven Energy Producer) is a thorium based energy producer, ATW (Accelerator Transmutation of Waste) feeds on spent fuel, ABC (Accelerator Based Conversion) burns weapons-return plutonium. All these systems produce electricity and short-lived or stable fission product isotopes only (Figure 3-4).

The subcritical systems proposed by the ADTT program represent a departure from traditional nuclear concepts (reactors), yet they strive to keep the best that the technology developed over the years, within a sensible conservative design envelope. The main elements and function of a Los Alamos ADTT system are illustrated in Figure 3-5 for a system which generates nuclear energy from thorium and destroys

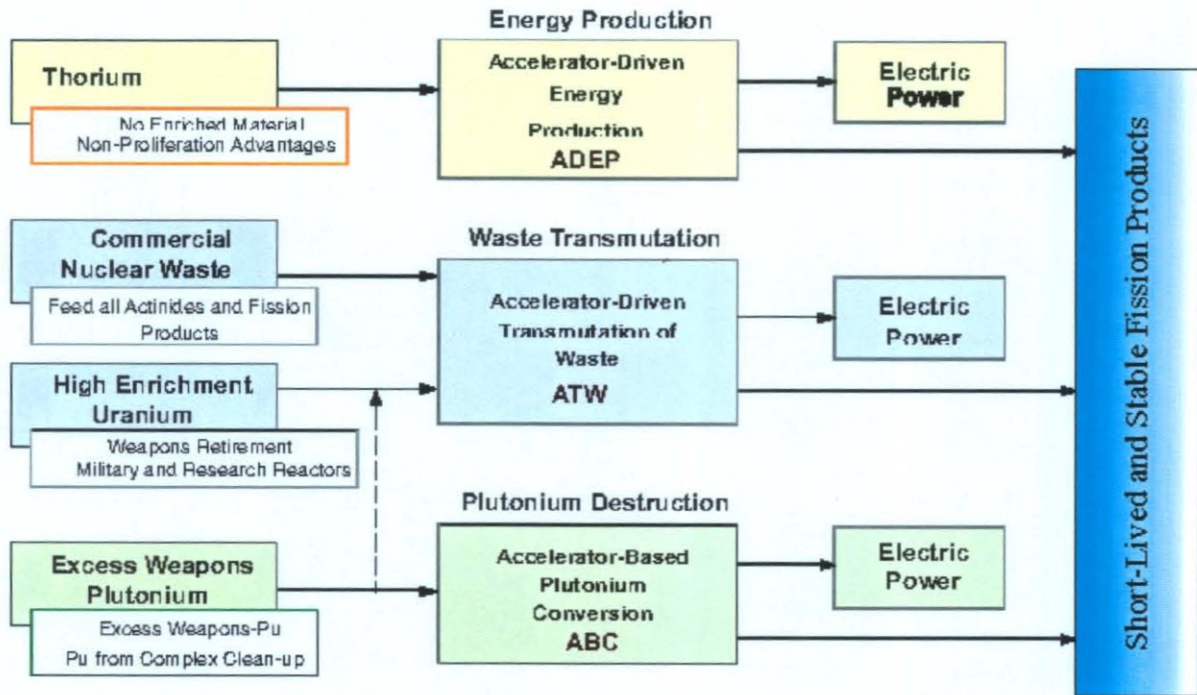


Figure 3-4. Overview of the different ADTT systems.

its long-lived high-level fission product waste. The system starts with ^{232}Th and converts it by neutron absorption into the fissile fuel ^{233}U from which energy is produced. The system consists of a reactor referred to in the figure as the target-blanket which contains the fissile material and the waste to be transmuted. For a reactor each fission on average produces enough neutrons after losses to cause another fission so that the chain of fissions is continuous. For all ADTT systems, the losses are made somewhat larger by the loss of neutrons for waste transmutation so

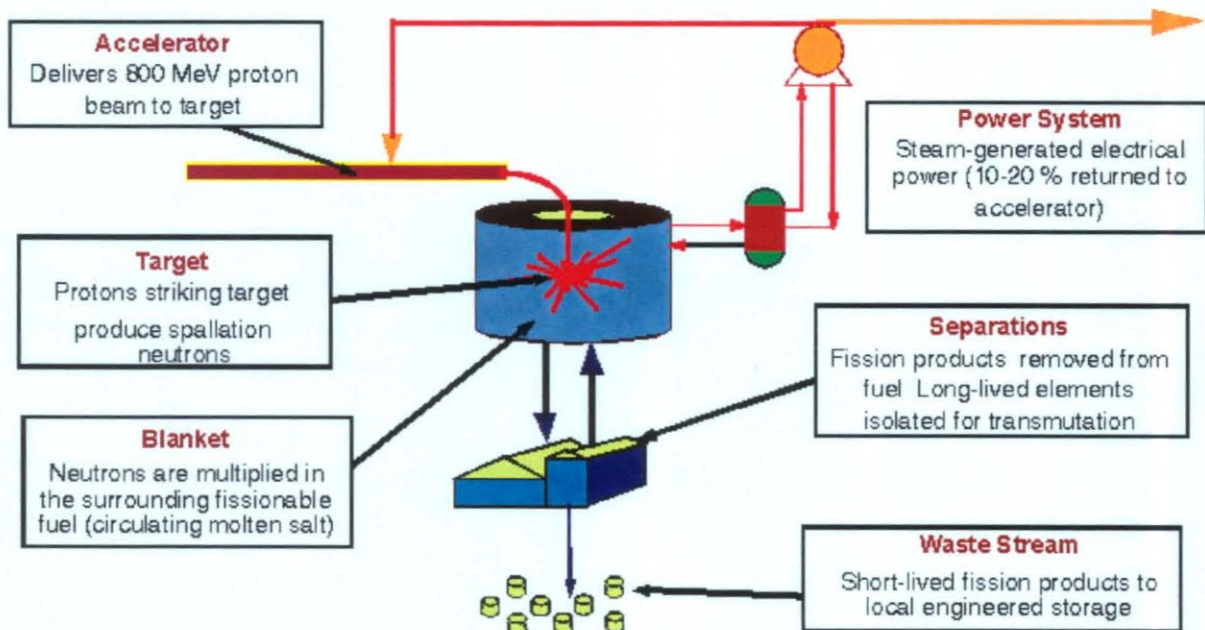


Figure 3-5. The main elements and functions of a Los Alamos ADTT system.

that there is no self-sustained chain reaction. Therefore by itself the system is passive and inoperative. However, by making up for the 5-10 % loss of neutrons from an external neutron source, the system would function effectively even though the chain reaction would not be self-sustaining.

A3.4.1 ABC

The Los Alamos National Laboratory has proposed an accelerator-driven subcritical system in which fission product poisons are allowed to build up until not only sufficiently to consume the excess fission neutrons from ^{239}Pu fission, but also the supplemental neutrons from the accelerator [A19]. The system achieves very high burn up without fuel reprocessing or fuel fabrication and refabrication. Also no fission product removal is required. The General Atomics Corporation has proposed a program with a similar objective. Its helium-cooled graphite-moderated reactor with ^{239}Pu fuel particles suspended in the graphite has been proposed as the first stage of ^{239}Pu destruction. After the Pu has been burned sufficiently that it will not sustain criticality, the fuel is transferred to an accelerator-driven assembly which continues to destroy the plutonium using accelerator-generated neutrons until k_{eff} of the system has dropped to about 0.6. The burn-up of the Los Alamos and the General Atomics systems are similar and are the highest of any of the proposed Pu-burning systems.

A3.4.2 ADEP

Perhaps the most important element of the ADTT project over the long term is Accelerator-Driven Energy Production (ADEP) which uses thorium as a nuclear fuel. The system is based on the Th-U cycle in which ^{232}Th is converted by neutron capture to fissile ^{233}U . This cycle has been studied extensively for use in commercial nuclear reactor power generation [A20]. The primary objective of the molten salt reactor experiment was to show that an effective breeder reactor could be built on this cycle which produced more ^{233}U than it consumed. This reactor technology lost out to the sodium-cooled fast breeder based on the U-Pu cycle because its breeding ratio was barely larger than unity even when fission products were promptly removed from the fuel. The U-Pu cycle showed much higher breeding ratios. The Th-U cycle development program was also focused on a molten salt liquid fuel program with on-line removal of fission products, and the operation of a liquid fuel reactor was demonstrated with the several-year Molten Salt Reactor Experiment (MSRE) at the Oak Ridge National Laboratory. Fission products could be continuously removed and the liquid fuel allowed the reactor to be continuously refueled. MSRE still holds the world record for the longest continuous chain reaction (impossible to beat because today's more stringent safety regulations which would require about one shut-down and inspection period per year). A great deal of research was done on the materials to contain the salt and the ADTT projects rely heavily on the materials work done for the MSRE. While the MSRE had no actinide waste stream (MAs and Pu remained in the waste), it had the usual fission product

waste and its neutron economy did not allow it to breed as much ^{233}U as it burned and still have excess neutrons left for transmutation of its fission products. The conclusions of studies at Oak Ridge were that critical thermal breeder reactors could not breed fissile material in this cycle, unless the fuel could be kept painstakingly clean from fission products and unless the ^{233}Pa formed could be promptly removed from the neutron flux and allowed to decay into ^{233}U . The introduction of the accelerator in ADEP largely reduces both problems.

The accelerator-driven spallation source supplements the number of available neutrons over that which can be achieved in a critical reactor by converting some of the electric power generated into neutrons. This neutron increase allows ADEP designs to produce energy using natural thorium as feed, without highly enriched fuels and even without chemical extraction of the protactinium precursor to ^{233}U while at the same time transmuted the internally generated radioactive wastes. To do the equivalent functions, a critical reactor would require the use of highly enriched fuels.

The fission products ^{90}Sr and ^{137}Cs are important contributors to the radio-toxicity of spent nuclear fuel for some hundred years after its removal from the reactor. ADEP cannot transmute these fission products in an effective way because of their small neutron cross-sections. Some of the other more long-lived fission products should, however, be possible to transmute in an ADEP-system. Examples are ^{99}Tc and ^{129}I as well as ^{79}Se , ^{126}Sn and ^{135}Cs if it is deemed justifiable to develop the necessary technology.

The ADEP plant consists of various systems, or facilities, categorized as:

- 1 The target-blanket system, in which source neutrons generated by the interaction of a ion beam with the molten salt target are thermalized and multiplied; the fission heat generated in the fuel salt in its passage through a graphite moderated region is removed in primary heat exchangers
- 2 A coolant-salt circulating system, steam generators, and a turbine-generator plant for converting the thermal energy into electric power
- 3 An off-gas system for purging the fuel salt of fission product gases and gas-borne particulates and an associated electrolytic system for plating out noble and semi-noble metals.
- 4 A separation processing facility for fuel salt cleanup and recycle; this facility would continuously process a slip stream of molten salt.
- 5 Auxiliary salt handling equipment
- 6 General facilities and equipment, including controls and instrumentation, maintenance tools, auxiliary power equipment, waste management, storage and disposal systems, condensing water systems, electrical switchyard, stacks, etc.

A3.4.3 ATW

The objective of the Accelerator Transmutation of Waste (ATW) sub-project of ADTT is to transmute the actinide and long-lived fission product waste from commercial nuclear reactor spent fuel. If the separations can be done sufficiently well, the amount of long-lived radionuclides in the remnant waste will be substantially reduced compared to LWR-wastes.

The ATW system also aims to treat waste from commercial light water reactors. To many this would appear to require the separation of plutonium and other components of the waste before feeding them into the system. But because of the excess neutrons provided by the accelerator, the needed front end reprocessing is removal of the zirconium cladding and the uranium. All of the other actinides and fission products can probably be fed to the blanket, because the capabilities for removal of the fission products already exist in the back-end separation system.

The front-end system has not been selected but there are at least two options under consideration. One would involve the crushing of the spent fuel assemblies which contain mostly UO_2 and the oxidation of this to U_3O_8 . Another means of removing the cladding might be to burn the spent fuel assemblies in a hydrogen chloride atmosphere thereby converting the zirconium to volatile ZrCl_4 . Reinhard (see section 2.31 in [A21]) discusses hydro-chlorination and presents data, together with many references to experimental work, on cladding removal by this method, e.g. results from pilot plant decanning of simulated spent PWR fuel.

In contrast to the standard aqueous reprocessing system developed over many years and now in common use, the processes described do not produce a pure stream of "naked" plutonium. The plutonium is never separated from the highly radioactive components of the spent fuel, but only from zirconium and uranium. The front-end separation required for the ATW therefore produces a stream which is mostly highly radioactive fission product and separation of the plutonium from this fission product and from the other actinides would be required before it could be used in weapons. Commercial nuclear power plants are typically sized at $3000 \text{ MW}_{\text{th}}$ and leave about 300 kg of plutonium and other higher actinide per year in the spent fuel while fissioning 1200 kg of fissile material per year. Therefore an ATW system operating at the same fission power level of the LWRs could burn the waste from four LWRs if its operating life were the same as the LWRs. Transmuting the LWR waste arising from all LWRs in Sweden using ATW systems would require the deployment of about three $3000 \text{ MW}_{\text{th}}$ ATW systems if the waste were to be transmuted in about 30 years. Unless the income from electric power sales were sufficient to offset the capital and operating costs of the ATW system, the cost of treating the waste by this means could be prohibitive. The economic picture for the ATW system will be less favorable than for the thorium-based energy producer (ADEP) system because the ADEP system need only treats its own waste and only a modest accelerator is required for the modest neutron supplement. However the ATW system must treat not only its own waste but also that from the three-four LWRs. Substantially more accelerator-produced neutrons are required with greater capital cost for a larger accelerator and for the additional power to the accelerator.

However, the growing concern about the increasing stock of the reactor grade plutonium in spent fuel - see Figure 3-6 - might change our perspective on this issue

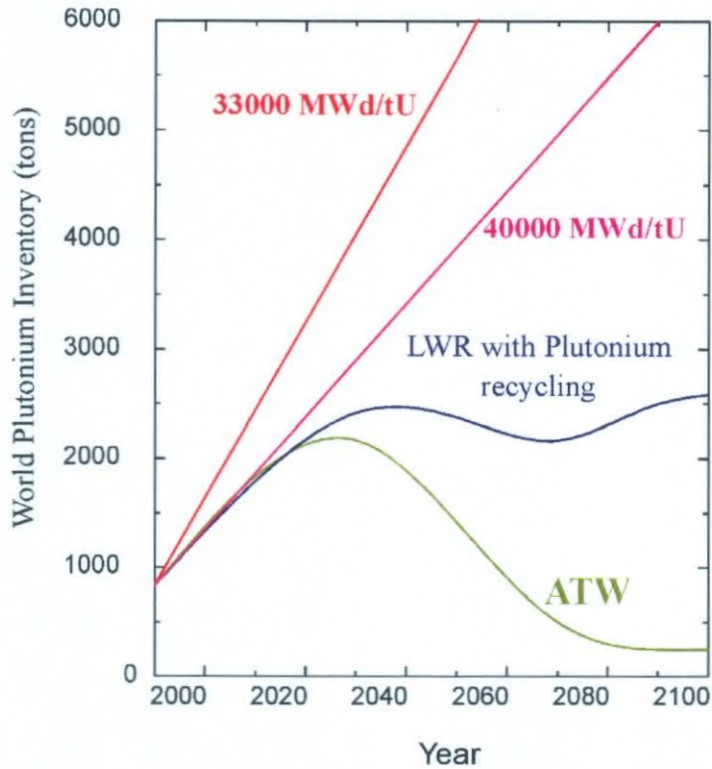


Figure 3-6. Variation of the amount of plutonium with time for some scenarios assuming a constant number of nuclear reactors and, in case of ATW, 2 units/3 years taken into operation.

in the future. The amount of the reactor grade Pu will reach 2 000 tons by the year 2010 assuming no new reactor constructions in the world. ATW-systems have a potential to change this trend, but the number of facilities needed would be about 15% of the number of existing LWRs - taking into account a time span of 100 years it does not look unreasonable. (About 400 existing power reactors were constructed under say 30 years).

A3.5 RESEARCH ACTIVITIES IN THE RUSSIAN FEDERATION

In the frame of the International Science and Technology Center (ISTC - organization established by USA, Japan, EC and Russia to fund projects leading to conversion of military research activities to civil research) 500 Russian scientists are employed in the project "Feasibility Study of Technologies for Accelerator Based Conversion of Military Plutonium". This project is the biggest single project financed by ISTC and its budget exceeds 3 M\$ over the 2 year period. The research program of the project is very broad covering several conceptual designs of accelerator-driven incineration of weapon-grade plutonium like: heavy water based systems, molten salt thermal systems, and even liquid lead based fast neutron systems. The spallation target design, molten salt based separation chemistry and basic nuclear data measurements are also under intensive investigation in the frame of this project. In the last few months of 1995 the Russian Ministry of Atomic Energy expressed its strong interest in this field and is also financing research on

ADS [A22, A23].

The project "Design and Construction of the Molten Lead Spallation Target for Accelerator-Driven Systems" prepared in the cooperation Russia-Sweden-USA is under consideration to be financed by the Swedish contribution into ISTC.

The ADS-research activities in the Russian Republic will be described a separate report.

A3.6 RESEARCH ACTIVITIES IN SWEDEN

The research interest in Sweden is driven by the strong expectation that the accelerator-driven transmutation technology offers a possibility to produce cleaner fission energy over an indefinite time (comparable to that achievable by fast breeder reactors). The research problems connected with the utilization of fusion energy are still numerous. Today it is difficult to predict when the basic problems in this field will be solved.

With the twofold aim to find methods for treating the high level nuclear wastes which could be more easily accepted by the public than a direct geological deposition of spent fuel, and at the same time recruit students to the nuclear energy field, a national collaboration has been initiated on the research of accelerator driven transmutation technologies. The incentive to start research in this field was positively influenced by the Specialists' Meeting on "Accelerator-Driven Transmutation Technology for Radwaste and other Applications" which was held at Saltsjöbaden, Sweden on 24-28 June 1991.

A "Group for Spallator Research" conducts some concerted research at CTH, KTH, the Manne Siegbahn Laboratory-Stockholm University and the Uppsala University. The group also has a pending application to "Stiftelsen för strategisk forskning" for about 11 MSEK/year for 5 years.

The main task of this group is to stimulate and to coordinate research and development projects in the accelerator driven transmutation technologies. These projects are aimed to:

- 1 Practical solutions for accelerator driven transmutation of longlived radioactive material (e.g. plutonium, minor actinides, fission products) into shortlived or stable elements.
- 2 Investigation of new options for nuclear energy production with inherently safe systems, either with uranium or thorium fuel and with reduced longlived radioactive waste production. If successful, it will result in a new, environmental friendly, safe, cheap and virtually unlimited source of energy.
- 3 Opening of new, exciting research and occupation possibilities for students and young specialists, which will ensure the proper level of competence needed for nuclear power utilities, governmental agencies etc.

The research activities of the Group for Spallator Research has been primarily devoted to system and feasibility studies together with participating in some

international efforts mainly in US (Los Alamos), Russia (Moscow, Obninsk), France (Saturne) and in future possibly Japan (PNC-JAERI), CERN and Switzerland (PSI).

A3.7 SOME EUROPEAN ACTIVITIES (involving Swedish participants):

Some institutes and research centers from different European countries (Sweden: KTH, CTH, UU; Germany: KFK, KFA; France: CEA, Italy: ENEA, Politecnica Torino; Netherlands: ECN, Great Britain: AEA and European Institute of Transuranium Elements, Karlsruhe) applied to EC for a shared-cost project funding in 4th Framework Programme of the European Community 1995-1998. The application has been accepted by CEC and contract negotiations are under way.

The overall objective is to make a European assessment of the possibilities of accelerator-driven hybrid reactor systems from the point of view of safe energy production, minimum waste production and transmutation capabilities. In particular:

- a. to perform system studies on accelerator driven hybrid systems
- b. to assess the accelerator technology
- c. to study the radiotoxicity of the fuel cycle for ADS and its nonproliferation aspects
- d. to provide basic nuclear and material data for ADS by means of evaluation and experiments

The total budget of this project is 1 130 kECU with an EC funding of 450 kECU.

A4 OVERALL TECHNICAL FEASIBILITY OF ACCELERATORS

A4.1 High-current Linac

The high-power accelerator technology required for ADTT has been under continuous development for the past three decades at Los Alamos. Accelerators up to five times larger than that required for the ADTT system described here have been proposed for the production of tritium for defense applications. These accelerators were reviewed by the Energy Research Advisory Board of the U.S. Department of Energy, subsequently by the JASONS, a scientific body which reviews proposed high technology programs of the U.S. Department of Defense, and were also evaluated by the General Accounting Office of the U.S. Congress. The result of these reviews was a general endorsement of the proposed accelerator technology with the provision that appropriate pilot and demonstration steps be made along the way towards the construction of a full scale facility [A24].

The average power needed for the very largest of systems proposed at LANL ($6 \times 500 \text{ MW}_{\text{th}}$), requires an accelerator average power of some tens of megawatts (100 mA beam). This can only be achieved with a linear accelerator. However, the highest power operational linear accelerator anywhere, (LAMPF), operates at around one megawatt (1 mA at 800 MeV). At first this seems like a very large extrapolation, but it is not nearly so bad as it appears. Firstly, in LAMPF, only

every fourth bucket is filled with beam and so filling every bucket (if we use funneling) or every other bucket (if un-funnelled) immediately gives a factor of four in average power respectively. Also LAMPF being a pulsed machine operating at 10% duty factor, going to 100% duty factor gives a factor of 10. The charge in each micro-bunch can be increased by about a factor of four and still stay well within the stable space charge regime. Therefore an improvement by a factor of 160 ($4 \times 10 \times 4$) is possible by simple extension of proven technologies, so that up to 150 mA 1 GeV LINACs can be built based on current technology.

Primary issues for the ADTT accelerator are the efficiency, reliability and maintainability appropriate to an industrial setting. Experience with the use of an accelerator in an ADTT application can be gained using the existing accelerator at Los Alamos National Laboratory (LAMPF). As it is, LAMPF is capable of reliably providing up to 1 mA of 800 MeV proton beam to a scaled down target-blanket. LAMPF could thus drive an experimental deeply subcritical facility with a multiplication of 5 to a fission power level of about 5 MW_{th} , a sufficient level for a testing of the ADTT concept

A4.2 High-current Circular Machines

For circular machines the situation is a little different. The highest average power machine at the Paul Scherrer Institute (PSI), a 600 MeV cyclotron, operates at about 0.5 mA. PSI believes that its machine can increase beam current to about 1-2 mA without major changes, but 10 mA would require a new design, which could be the Intermediate Separated Sector Cyclotron (ISSC) or Three Stage Cyclotron (ISSC+Booster Separated Sector Cyclotron), presently proposed and studied by Rubbia [A25, A26].

A5 TIME SCALE AND EFFORTS REQUIRED

A demonstration experiment could be performed at the existing accelerator at LANL with a molten salt system operating at $k_{\text{eff}}=0.96$ and at power level of $40 \text{ MW}_{\text{th}}$. Such an experiment would have to be accompanied by research and demonstration, at about the same technical effort as the experiment, on the required separations in the molten salt system. If properly financed, seven years hence, an integrated demonstration of the ADTT system could be in operation at $200 \text{ MW}_{\text{th}}$ level, with the deployment of the ADTT system beginning in about fifteen years.

A $3000 \text{ MW}_{\text{th}}$ ($1200 \text{ MW}_{\text{e}}$, $1060 \text{ MW}_{\text{e}}$ net to grid) ATW facility driven by a 1 GeV, 50 mA linac at $k_{\text{eff}}=0.96$ is estimated by the LANL group to cost about 2500 M\$ [A5, A27]. Such a unit is would to be able to handle spent fuel from four 1000 MW_{e} LWRs. The gross breakdown of the cost estimate is as follows [A5, A27]:

- Accelerator, turbines and other electrical equipment	900 M\$
- Blanket (subcritical reactor)	300 M\$
- Chemical plant (based on IFR cost estimates)	500 M\$
- Contingency and interest during construction	800 M\$

Rubbia is estimating [A29] that the prototype of his system based on fast neutrons and the Thorium cycle (only energy production) would cost about 1350 MUS\$ (with 250 MUS\$, i.e. 20 % of the costs, going into cyclotrons and ADS-specific equipment) for 1 GW_e, 10-15 mA unit. The time required to develop a demonstration facility (10-20% of full size) is optimistically estimated to 10 years. Considering the different basis for the accelerator cost estimates (the cyclotron proposed by the CERN group is estimated to be 250 M\$ cheaper than a corresponding linac [A28]) the total cost estimates from the LANL and CERN groups are surprisingly close. However, time and cost of licensing were not included in any of the estimates.

In the Japanese OMEGA program basic studies are to be performed to about year 2000. After this year pilot facilities will be constructed to demonstrate the P-T technology. The accelerator R&D program is divided into 2 phases: the first is construction of the Basic Technology Accelerator (BTA) with an energy of 10 MeV and a current 10 mA, the second is the construction of the Engineering Test Accelerator (ETA) with an energy of 1.5 GeV and an average current of 10 mA. Time scale - about 10 years.

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Biotite and chlorite weathering at 25°C. The dependence of pH and (bi) carbonate on weathering kinetics, dissolution stoichiometry, and solubility; and the relation to redox conditions in granitic aquifers

Maria Malmström¹, Steven Banwart¹, Lara Duro², Paul Wersin³, Jordi Bruno³

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January 1995

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Copper canister with cast inner component. Amendment to project on Alternative Systems Study (PASS), SKB TR 93-04

Lars Werme, Joachim Eriksson
Swedish Nuclear Fuel and Waste Management Co,
Stockholm, Sweden
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Marie Wiborgh (ed.)

Kemakta Konsult AB, Stockholm, Sweden
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Spent nuclear fuel corrosion: The application of ICP-MS to direct actinide analysis

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¹ Caledon-Consult AB, Nyköping, Sweden

² Studsvik Nuclear AB, Nyköping, Sweden
March 1995

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Palaeohydrological implications in the Baltic area and its relation to the groundwater at Äspö, south-eastern Sweden – A literature study

Bill Wallin

Geokema AB, Lidingö, Sweden
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Äspö Hard Rock Laboratory Annual Report 1994

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Feasibility study for siting of a deep repository within the Storuman municipality

Swedish Nuclear Fuel and Waste Management Co., Stockholm
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A thermodynamic data base for Tc to calculate equilibrium solubilities at temperatures up to 300°C

Ignasi Puigdomènech¹, Jordi Bruno²

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Cerdanyola, Spain

April 1995

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Investigations of subterranean microorganisms. Their importance for performance assessment of radioactive waste disposal

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June 1995

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Solute transport in fractured media – The important mechanisms for performance assessment

Luis Moreno, Björn Gylling, Ivars Neretnieks
Department of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden

June 1995

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Yvonne Ohlsson, Ivars Neretnieks
Department of Chemical Engineering and Technology, Royal Institute of Technology, Stockholm, Sweden

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Interactions of trace elements with fracture filling minerals from the Äspö Hard Rock Laboratory

Ove Landström¹, Eva-Lena Tullborg²

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² Terralogica AB

June 1995

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Roland Pusch

Clay Technology AB

February 1995

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Estimation of effective block conductivities based on discrete network analyses using data from the Äspö site

Paul R La Pointe¹, Peter Wallmann¹, Sven Follin²

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September 1995

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Temperature conditions in the SKB study sites

Kaj Ahlbom¹, Olle Olsson¹, Stefan Sehlstedt²

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Measurements of colloid concentrations in the fracture zone, Äspö Hard Rock Laboratory, Sweden

Anna Ledin, Anders Düker, Stefan Karlsson, Bert Allard

Department of Water and Environmental Studies, Linköping University, Linköping, Sweden

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Thermal evidence of caledonide foreland, molasse sedimentation in Fennoscandia

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Compaction of bentonite blocks. Development of technique for industrial production of blocks which are manageable by man

Lars-Erik Johannesson, Lennart Börgesson, Torbjörn Sandén

Clay Technology AB, Lund, Sweden

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Lennart Börgesson¹, Lars-Erik Johannesson¹, Torbjörn Sandén¹, Jan Hernelind²

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Björn Lagerblad, Jan Trägårdh

Swedish Cement and Concrete Research Institute

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Kristina Skagius¹, Anders Ström², Marie Wiborgh¹

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² Swedish Nuclear Fuel and Waste Management Co, Stockholm, Sweden

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Roy Forsyth

Caledon Consult AB

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Marcus Laaksoharju¹, Claude Degueudre², Christina Skärman¹

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² University of Geneva, Switzerland

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Survival of bacteria in nuclear waste buffer materials. The influence of nutrients, temperature and water activity

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December 1995

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December 1995

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