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Partitioning and transmutation Current developments – 2004

A report from the Swedish reference group on P&T-research

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Preface

This report has been written on behalf of the Swedish reference group for research on partitioning and transmutation. The reference group has been assembled by SKB and its members represent the research teams active in this field at Swedish universities. The group also has observers from the authorities SKI and SSI as well as from KASAM – see Appendix IV. The draft of the report has made been available for comments to the members and observers of the reference group. The authors are, however, responsible for the contents.

Summary

The research and development on methods for partitioning and transmutation (P&T) of long-lived radionuclides in spent nuclear fuel has attracted considerable interest during the last decade. The main objective of P&T is to eliminate or at least substantially reduce the amount of such long-lived radionuclides that has to go to a deep geological repository for final disposal.

The radionuclides of main interest (concern) are those of the transuranium elements. These elements are formed in a nuclear reactor by one or more neutron captures in uranium atoms which then by subsequent radioactive decay are transformed to neptunium, plutonium, americium or curium. Even small amounts of elements heavier than curium are formed but these are of minor interest in this context. A few fission products (technetium-99, iodine-129) may also be of some interest for transmutation.

The long-lived radionuclides can be transmuted to more short-lived or stable nuclides by the use of nuclear physics processes. In theory and on laboratory scale several such processes are possible. In practice so far only transmutation by irradiation with neutrons can be achieved in macroscopic scale. Neutrons can cause fission in the transuranium elements and this process will release a substantial amount of energy. Thus transmutation on large scale of the transuranium elements from spent nuclear fuel must be done in a device similar to a nuclear reactor.

A prerequisite for transmutation by irradiation with neutrons is that the nuclides to be transmuted are separated (partitioned) from the other nuclides in the spent fuel. In particular the remaining uranium must be taken away unless you want to produce more plutonium and other transuranium elements. Separation of the various elements can at least in principle be achieved by mechanical and chemical processes. Currently there exist some large scale facilities for separation of uranium and plutonium from the spent fuel – reprocessing plants. These can, however, not separate the minor actinides – neptunium[1], americium and curium – from the high level waste that goes to a repository. Plutonium constitutes about 90% of the transuranium elements in fuel from light water reactors.

The objective of current research on partitioning is to find and develop processes suitable for separation of the heavier actinides (and possibly some long-lived fission products) on an industrial scale.

The objective of current research on transmutation is to define, investigate and develop facilities that may be suitable for transmutation of the aforementioned long-lived radionuclides.

The processes and facilities that could be implemented as results of such developments must meet very high standards of safety and radiation protection as well as have low environmental impact. They shall be economically viable and have good proliferation resistance. The large amount of energy released in the transmutation process should be

[1] Note: Neptunium can be separated with uranium if a minor adjustment of the operating conditions is made in the industrial PUREX process. This possibility is not used today as it would give increased costs for purification of recovered uranium.

used in a proper way. In other words the processes and facilities must be acceptable to society.

Research on P&T started already in the 1950ies when development of nuclear power gained momentum. In the subsequent years it was mainly tied to the development of the breeder reactor. As this development slowed down to a very low level in the early 1980ies the interest in P&T more or less disappeared.

The renewed interest through the 1990ies has caused some expansion of the programmes in this field in particular on an international level. In Europe this is focused on the R&D-programmes of the European Union (EU). The EU so-called framework programmes (FWP) have established a strong link between the various national programmes within the union and also in some other European countries. Other large programmes are going on in Japan (OMEGA), USA and Russia.

A review of the status of the efforts concerning P&T was published by SKB in 1998 /Enarsson 98/. This report summarises the work reported in the years 1998–2003 and tries to assess the prospects for future development of P&T as seen from a Swedish perspective.

Systems for partitioning and transmutation

During the last five years a number of system studies on P&T have been published. These studies give a good overview of the extensive work required for implementing P&T, of the relative large and complex system of facilities that are needed as well as of those problems that must be solved and the issues that must be addressed before such a system can be deployed.

The first of these system studies was done by an expert group within OECD/NEA and was published in 1999 /NEA 99/. Important conclusions from this study were i.a.

- The basic R&D for partitioning and transmutation requires long lead times and large investments in dedicated fast neutron devices, extensions of reprocessing plants and construction of remote controlled facilities for fuel fabrication.
- Partitioning of long-lived radiotoxic elements from spent nuclear fuel can be made in extensions of existing reprocessing plants but requires a large amount of work to be developed from laboratory to industrial scale,
- The transmutation with fast neutrons is more effective than in existing light water reactors. Transmutation of transuranium elements can best be achieved in fast reactors or in accelerator-driven systems with fast neutron spectrum.
- Partitioning and transmutation will not eliminate the need for a deep geological repository for certain long-lived radioactive wastes from spent fuel.

In the USA a certain interest arouse for transmutation using accelerator-driven systems in the early 1990ies. The centre of this interest was Los Alamos National Laboratory (LANL) that introduced the concept ATW – Accelerator-driven Transmutation of nuclear Waste. This gradually led to a broad study of ATW by the US Department of Energy at the request of US Congress. The study, published in the autumn of 1999 /DOE 99/, proposed a research programme for ATW that could be the start of a large scale concentration on such systems. Some parts of this work have started in particular those parts involving international co-operation. The programme as a whole has, however, not been accepted as a base for the US research on advanced fuel cycles or for future nuclear waste strategy. It was probably not intended as a complete

programme but more of an in depth evaluation of one of many possible scenarios for development of partitioning and transmutation.

In 2003 USA announced the so-called Advanced Fuel Cycle Initiative (ACFI) /DOE 03/, which aims at a broad study of fuel cycles for future nuclear power reactors – also named Generation IV reactors. This initiative is planned to be pursued in three phases – phase 1 basic evaluation; phase 2 – proof of principles; phase 3 – proof of performance. The programme is broadly laid out and includes review of all current reactor concepts and systems (LWR, HTR, ADS, FR, aqueous based reprocessing and pyrochemical reprocessing, etc).

Upon initiative from the research ministers in France, Italy and Spain a European technical working group – TWG – under chairmanship of Carlo Rubbia (Nobel laureate in physics and former Director General of CERN[2]) was formed in 1999. This working group proposed a plan for development of an accelerator-driven system (ADS) in Europe. The report from TWG was published in the spring of 2001 /TWG 01/. The ambition from the group was that the plan should form the base for continued EU-financed research work on ADS. In the plan the construction of a small experimental plant at 100 MW thermal power is proposed. The plant is envisaged to start operation in 2015. The cost for the first twelve years R&D-programme is estimated to M€ 980. In addition another M€ 180 are proposed for R&D on nuclear fuels especially suited to ADS. This first phase would then be followed by a second where a prototype for a full scale ADS is developed and constructed. After successful operation of this prototype industrial plants may be deployed starting around 2040. This study agrees fairly well concerning required times with the USDOE study. In details, however, the two studies have considerable differences. The US study has much more specified selections of systems for partitioning (pyrochemical process) as well as for transmutation (ATW cooled by liquid lead-bismuth eutectic) than the European one. The latter is delimited to transmutation with ADS, but keeps several parts of the ADS open for later choice of design – accelerator, coolant, etc.

The work within OECD/NEA continued within another expert group, which in the spring of 2002 published a comparative study of fast reactors and ADS for transmutation /NEA 02b/. The group reviewed a number of strategies for partitioning and transmutation of transuranium elements based on light water reactors, fast reactors and accelerator-driven systems. Among the general conclusions in the report are the following:

- While P&T will not replace the need for appropriate geological disposal of high-level waste, the study has confirmed that different transmutation strategies could significantly reduce, i.e. a hundred-fold, the long-term radiotoxicity of the waste and thus improve the environmental friendliness of the nuclear energy option. In that respect, P&T could contribute to a sustainable nuclear energy system.
- Very effective fuel cycle strategies, including both fast spectrum transmutation systems (FR and/or ADS) and multiple recycling with very low losses, would be required to achieve this objective.
- Multiple recycle technologies that manage Pu and MA either together or separately could achieve equivalent reduction factors in the radiotoxicity of wastes to be disposed.

[2] CERN = Centre Européen de la Recherche Nucleaire at French-Swiss boarder outside Geneva.

- The study shows that pyrochemical reprocessing techniques are essential for those cycles employing ADS and FRs where very high MA-content fuels are used.
- In strategies where Pu and MA are managed separately, ADS can provide additional flexibility by enabling Pu-consumption in conventional reactors and minimising the fraction of dedicated fast reactors in the nuclear system.
- In strategies where Pu and MAs are managed together, the waste radiotoxicity reduction potential by use of FRs and ADS is similar and the system selection would need to be made based on economic, safety and other considerations.
- Further R&D on fuels, recycle, reactor and accelerator technologies would be needed to deploy P&T. The incorporation of transmutation systems would probably occur incrementally and differently according to national situations and policies.
- Fully closed fuel cycles may be achieved with a relatively limited increase in electricity cost of about 10 to 20%, compared with the LWR once-through fuel cycle.
- The deployment of these transmutation schemes need long lead-times for the development of the necessary technology as well as making these technologies more cost-effective.

Some of the more important technical conclusions from this study are:

- To achieve the goal of reducing the amount of long-lived radionuclides in the waste by a factor of 100 or more the losses in each partitioning cycle must be less than 0.1%.[3]
- Transmutation of plutonium alone will decrease the long-term radiotoxicity by only a factor of five compared to spent fuel.[4]
- Transmutation can be achieved either by fast reactors or by different combinations of light water reactors, fast reactors and accelerator-driven systems.
- Physical limitations imply that very long time is needed to pursue a “complete” transmutation. This means that the indicated objectives can only be reached if the technology is used for at least 100 years.
- The use of pyrochemical methods implies new potential chemical and radiological risks that must be mastered.
- All transmutation strategies for light water reactors give considerable amounts of depleted and irradiated uranium that have to be managed. If this uranium not is considered as a resource for future fast reactors, the long-term radiological effects when radium, radon and its daughters grow in must be accounted for.[5]

[3] Comment: This implies that the “transmutation levels” presumed in the report are achieved. These depend on the fuel properties. Experiments in fast reactors have shown that the presumed “transmutation level” with e.g. oxide fuel is possible to achieve.

[4] Comment: Important factors in this context are the burnup and the waiting time between irradiation and the reprocessing. At high burnup and long waiting time plutonium-241 decays tp americium-241 and further to neptunium-237. In particular americium-241 is very important for the radiotoxicity in the time frame of a couple of thousand years.

[5] Comment: Depleted uranium from enrichment of natural uranium is of course obtained also without P&T and must be managed in a responsible manner.

- Transmutation of long-lived fission products involves many technical and practical difficulties. At present technetium-99 and possibly iodine-129 seem to be the major or only candidates.
- Experimental research on transuranium fuel is a highly prioritised subject. A bottle-neck for such research is the availability of facilities with high fast neutron flux.
- The basic research and development on fast reactors as well as accelerator-driven systems should be simpler if an international accord was obtained on the advantages and disadvantages of different coolants for such systems.

Partitioning

The most common method for partitioning of elements in spent nuclear fuel is liquid-liquid extraction. This implies using a mixture of aqueous and organic solvents where some elements are dissolved in the aqueous phase and some in the organic phase. The distribution properties are affected by the acidity of the aqueous phase, the presence of complexing agents, etc. This technology is applied in the large reprocessing plants in e.g. France, Japan and UK. At these plants uranium and plutonium are separated to the organic phase and the fission products to the acidic aqueous phase. Much of the research on partitioning is directed to further development of this kind of process to include also separation of the other so-called minor actinides in particular the trivalent actinides. For these three-valence actinides it is difficult to find suitable organic substances, which not concurrently separate lanthanides[6] from the fission products. The amount of lanthanides in the spent nuclear fuel is much larger than the amount of minor actinides. Furthermore many lanthanides have a large neutron capture cross section. It is therefore important that the lanthanides are effectively separated from in particular americium and curium. The report accounts for a number of substances that are or have been investigated.

The so-called PUREX-process, used in existing reprocessing plants, uses phosphor-based organic substances. A drawback with these is that they give considerable amounts of medium level wastes that are contaminated with long-lived radionuclides. The research is therefore currently directed on substances containing only atoms of carbon, hydrogen, oxygen and nitrogen – thus called CHON-reagents. Such reagents can be burned completely and will therefore give less solid waste volumes. The current research is mainly focused on this type of reagents.

The research on aqueous based partitioning processes did some progress during the recent few years. There are thus good prospects for development of a process recovering 99.9% of as well uranium as all transuranium elements from the spent fuel. The problem is above all the costs and the irradiation stability of the organic molecules.

When a process has been defined that is well functioning on laboratory scale the extrapolation to industrial scale is no big problem. The technical components needed for such a process are well tested in other applications.

The limited stability of organic molecules against the strong ionizing radiation from high level waste contributed early to the search for non-aqueous separation processes. The research was early directed towards pyrochemical processes. In the 1950ies such a process was constructed at the breeder reactor EBR-II[7] in USA. This reactor had metallic fuel and was cooled by liquid sodium. The reprocessing of the fuel was made

[6] Lanthanides are chemically very similar to actinides.
[7] EBR II = Experimental Breeder Reactor II at Idaho Falls in USA.

by controlled oxidative slagging to purify the melted fuel from most of the fission products. The purified fuel was then recast to new fuel rods that were re-circulated to the reactor. Another promising method was fluoride volatilization, which was tested in the MSRE[8] – project with fuel as melted salts. This method was further developed for light water reactor fuel, but abolished after great technical problems in the early 1970ies. The problems were i.a. large process losses, control of criticality, handling of small amounts of water vapour and oxygen as well as corrosion.

The technical difficulties caused the development of pyrochemical methods to strongly decrease through the 1970- and 1980ies. The interest in these methods was, however, renewed when the interest for P&T was increasing in the early 1990ies. The research increased again in particular in Japan but later also in USA, Russia and Europe. The pyrochemical methods are considered to be of particular interest for reprocessing/ partitioning of fuel giving very large irradiation doses, e.g. fuel with mainly americium, curium and plutonium with large fraction of heavy isotopes.

The development of pyrochemical partitioning processes requires also the development of equipment that is suitable for industrial application of such processes. Material and corrosion problems must be solved as well as technical and radiological safety issues. The road to industrial application is probably longer for the pyrochemical processes than for the aqueous based processes.

Transmutation

The radionuclides that are of primary interest for transmutation are the transuranium elements. These are most suitably transmuted by neutron fission. Several of the nuclides are, however, only fissionable by fast neutrons. Two different types of facilities are considered to obtain a sufficiently high fast neutron flux – fast reactors with a self-sustaining chainreaction and an accelerator-driven subcritical system (ADS). In the period 1950–1985 a number of fast reactors were constructed in several countries. The majority of them had liquid sodium as coolant. The largest was the French Superphenix with 1200 MW_e power. Only a few of these fast breeder reactors are still in operation.

During the 1990ies the interest for accelerator-driven systems increased substantially and currently there are considerable interests for studies of such systems. In Sweden almost all research on transmutation is focused on ADS.

An ADS consists of the following main components:

- A proton accelerator to give a strong current (from a few up to several tens of mA) of protons with about 1 GeV/proton energy.
- A spallation source that is hit by the proton current and where spallation[9] reactions with heavy nuclides (lead, bismuth, tungsten, tantalum etc) create a neutron flux of several tens of neutrons per proton.
- A subcritical nuclear reactor with fuel that contains the long-lived radionuclides to be transmuted. As the transmutation is caused by fission of the nuclides new neutrons are formed. For each incoming neutron from the source about 20 new neutrons are released and cause additional fission reactions. The design of the reactor shall be such that a self-sustaining chain reaction cannot occur.

[8] MSRE = Molten Salt Reactor Experiment at Oak Ridge National Laboratory, USA, during the 1960ies.
[9] Spallation is a nuclear reaction where a charged particle (e.g. proton) with high energy hits a nucleus and transforms this nucleus at the same time as numerous light particles are released (see further Section 4.1.2).

- Balance of plant systems to take care of the energy released by the fission processes and by radioactive decay and to supply electricity to the accelerator.
- Equipment to control and monitor the entire process with its safety systems.

Proton accelerators exist in two different principal designs – a linear accelerator (linac) or a cyclotron (with near circular particle tracks). Cyclotrons have some technical limitations concerning proton energy, beam current and flexibility. This has caused the interest to focus more and more on linacs. A linac for the required proton energy will be very long – several hundred metres – and puts high demands on several different technical parts as well as on radiation protection. Up to recently accelerators have mainly been built for pure research purposes where the requirements on availability have been modest. A plant for transmutation does, however, include large systems with high temperatures and built in inertia for changes in the system operating conditions. This in addition to requirements of efficiency and reasonable economy puts strongly increased demands on the availability of accelerators for ADS.

The spallation source is a central component in an ADS situated between the accelerator and the subcritical reactor. It is placed in a demanding environment with high level of radiation, high temperature, strongly varying pressure and requires very effective cooling. Different heavy materials may in principle be used for the source (lead, bismuth, tungsten, tantalum etc) but considering the cooling requirements and other factors the current main candidate is a eutectic mixture of lead and bismuth. An important issue is the design of the "window" between the source and the vacuum tube that brings the proton beam from the accelerator to the lead-bismuth target. The alternatives are a hot "window" close to the target or "windowless" design (meaning a cold window away from the hot target metal). The latter design puts high demands on maintaining sufficiently good vacuum in the access tube to avoid beam energy losses. A hot window on the other hand puts very high demands on strength and irradiation resistance of the metal in the window in order to limit replacements to once per year. Currently two spallation sources with liquid lead-bismuth are built or planned in Europe. One is MEGAPIE at PSI in Switzerland where a hot window design has been selected. The other one is for the MYRRHA-project in Belgium where a windowless design will be tested.

Different designs are being considered for the subcritical reactor. Fuel as well as coolant are still open choices. Water is not a possible coolant for a fast flux reactor. Possible options are gas (particularly helium) or liquid metal. Gas-cooling requires high pressure and is therefore often turned down due to the risk of loss of cooling. Earlier fast reactor programmes have accumulated considerable experiences on liquid sodium. The problems then encountered are obviously currently discouraging and the interest is more and more focusing on liquid lead-bismuth. This eutectic mixture has a relatively low melting point and a high boiling point and is also the main candidate for the spallation source. The latter simplifies of course the design of the systems. Experiences from reactors cooled with liquid lead-bismuth have only been obtained in Russia where seven submarine reactors have been built with this coolant. Their total operating time is about 70 reactoryears.

The research on fuel for ADS follows several parallel lines. Nitrides and alloys with inert metals, e.g. zirconium, attract considerable interest. The requirements on the fuel are i.a good heat conductivity to allow high power density, good irradiation resistance and ability to achieve high burnup – tens of percent compared to a few percent in current light water reactor fuel.

Fast reactors can, as already mentioned, be used for transmutation. During the 1990ies the research has i.a. been focused on burning plutonium in sodium cooled fast reactors. Specific types of fuel have been studied in France. In recent years the interest has also been directed towards fast gas-cooled reactors. The fast reactor may also be used for transmutation of other transuranium elements. This requires however special designs in order to prevent serious deterioration of the dynamic and safety properties of the reactor. If the fast reactor is designed as a breeder reactor it may be possible to achieve very high energy extraction from the mined uranium[10] as well as an effective transmutation of all transuranium elements.

In Russia there is also an interest for civil applications of fast reactors cooled with liquid lead-bismuth. Furthermore some work is done on ADS mainly financed through the so-called ISTC-programme[11].

As already mentioned a system with high fast neutron flux is of main interest for an almost complete transmutation of all transuranium elements. For transmutation of only plutonium also other systems may be used. French studies show that by using specially designed fuel assemblies the amount of plutonium generated by a light water reactor park can be limited and stabilised. Studies by General Atomics in USA and others have shown that it may be feasible to develop fuel for a high temperature gas cooled reactor that admits burnup up to 700 MWd/kg and burning of more than 90% of the actinide content including plutonium-239.

Some conclusions

The R&D efforts on partitioning and transmutation (P&T) have increased somewhat during the period 1998–2003. Research on P&T has taken a prominent role internationally in the R&D on future nuclear power and nuclear fuel cycle systems.

Despite the fact that partitioning and transmutation have been on the agenda for quite a few years there are still a number of issues that must be settled before the research and development can be given a clearly focused direction. The above mentioned studies in USA and Europe have tried to define the programme needed to get into such a position. The studies proposed research programmes for about six years at the cost of a couple of hundred million euros.

The construction of a small ADS experimental plant is a necessary step to develop and demonstrate the concept. This experimental plant should then be followed by a demonstration plant in almost full scale. Such a plant can at the earliest be ready in the mid-2030ies.

A number of circumstances have, however, contributed to slower speed, less intensity and lower funding than proposed in the studies.

There is no unanimous view on the objectives for partitioning and transmutation. Many see it as a way to achieve broad acceptance for nuclear power at large. Others promote it as a way to get out of the impasse for a deep repository in several countries. Others

[10] Studies within OECD/NEA state that a breeder reactor can extract up to 100 times more energy from each tonne of mined natural uranium than the current light water reactors with once through fuel cycle /NEA 02 p 230/.

[11] ISTC = International Science and Technology Centre, an organisation financed by EU, Japan, Russia and USA with the purpose to engage former Soviet nuclear weapons researchers in civil projects. Before Sweden became a member of EU, Sweden was one of the direct financing countries. Waclaw Gudowski, KTH, is member of the international expert group evaluating proposals concerning P&T for financing through ISTC.

again put a strong emphasise on the proliferation aspects of a growing stock of plutonium from light water reactors, from other reactor fuels and from dismantling of nuclear weapons.

There is no unanimous view on the need to develop ADS or for the role of ADS in a P&T-system. Some advocate that ADS should be used for burning of all transuranium nuclides from the present enriched uranium fuelled light water reactors. Others see the ADS as a supplement particularly suitable for burning minor actinides (americium, curium and neptunium), whereas the major part of the plutonium should be burned in light water reactors (or in fast reactors)

There is no consensus among experts on which technical route to follow for several parts of a P&T system.

The interest for P&T is mainly concentrated to the national research laboratories in USA, Europe, Japan and some other countries. The universities in many countries including Sweden also show a strong interest. P&T-research attracts considerable interest among students in nuclear science. Several important research programmes are going on at universities and research laboratories in several countries.

The nuclear industry, however, has shown only a limited interest in the development and is mainly visible in France and a few other countries with large nuclear programmes. The long-term interest in industry is much more focused on new types of reactors – often called Generation IV.

The national and industrial efforts on spent fuel and high level waste management in almost all countries are primarily focused on the resolution of the deep repository issues.

An integrated evaluation of these circumstances leads to the conclusion that it is unlikely that industrial ADS plants will be deployed before 2050.

A successful development of P&T will not make a deep geological disposal obsolete. The complex processes will inevitably generate some wastes containing small amounts of long-lived radionuclides. These wastes will require a deep geological repository. A successful development may, however, decrease the requirements on the engineered barriers in the deep repository and also decrease the volumes needed for disposal of wastes.

For Sweden it is important to follow the international development and to maintain a reasonable level of competence in the country at least as long as a substantial part of the electricity is produced by nuclear power. Competence developed in P&T research is valuable and useful also in the work on maintaining and developing safety and fuel supply for the existing light water reactors. It is also of importance for assessing the further development of the waste management programme.

The development of P&T for industrial application would as already mentioned take several decades. In order to transmute all nuclear fuel only from the already existing nuclear power plants would take at least another 100 years. P&T at an industrial scale requires large nuclear facilities that must be accepted by society.

The costs for P&T are not possible to determine with any good accuracy before the systems have been better defined and tested. The estimates that have been made indicate

an electricity production cost[12] that is between 10% and 50% above the costs from modern light water reactor designs with once-through fuel cycle. The investments needed in R&D and in new nuclear facilities are very large. They are, however, spread over long time and the major part should be looked upon as investments for energy production. It is not economically defensible to implement P&T without effectively using the energy released. This is particularly the case with transmutation of plutonium. Some experts have the opinion that it might be feasible to construct small ADS-plants specifically for burning americium and curium.

The introduction of a P&T-system for effective reduction of the amount of long-lived radioactive nuclides that must be disposed of in a deep geological repository thus requires a commitment to nuclear energy for a very long time. It may, however, on the other hand be looked upon as a long-term strategy for recycling and recovering energy from materials that otherwise are seen as very troublesome wastes.

[12] The estimated electricity production cost for a nuclear energy system with P&T is, however, still lower than the electricity production costs from systems with so-called renewable energy.

Sammanfattning

Forskning och utveckling av metoder för separation och transmutation (S&T[13]) av långlivade radionuklider i använt kärnbränsle har under senaste decenniet tilldragit sig ett växande intresse. Huvudsyftet med S&T är att ta bort eller åtminstone avsevärt minska mängden långlivade radionuklider som måste deponeras i ett djupförvar.

De viktigaste radionukliderna i detta hänseende är de s k transuranerna dvs ämnen tyngre än uran. Dessa bildas i kärnreaktorer genom att en eller flera neutroner infångas av uranatomer, som sedan via radioaktiva sönderfall omvandlas till neptunium, plutonium, americium eller curium. Även små mängder av ännu tyngre ämnen än curium kan bildas men dessa är av mindre betydelse i detta sammanhang. Några enstaka klyvningsprodukter (teknetium-99, jod-129) kan också vara av visst intresse för transmutation.

De långlivade radionukliderna kan omvandlas – transmuteras – till mer kortlivade eller stabila nuklider genom kärnfysikaliska processer. I teorin och i mycket liten laboratorieskala är flera sådana processer möjliga. I praktiken har hittills endast bestrålning med neutroner använts för transmutation i makroskopisk skala. Neutroner kan klyva kärnor i transuranatomer. Transmutation i stor skala av transuraner från använt kärnbränsle måste således ske i en anläggning som liknar en kärnreaktor. Eftersom kärnklyvningsprocessen frigör stora energimängder kommer anläggningen att likna en kraftreaktor.

En förutsättning för transmutation genom neutronbestrålning är att de nuklider som skall transmuteras separeras (avskiljes) från andra nuklider i det använda bränslet. I synnerhet måste man avlägsna kvarvarande uran om man skall undvika att det bildas mer plutonium och andra transuraner. Separation av de olika ämnena kan åtminstone i princip åstadkommas med mekaniska och kemiska processer. I dag finns det stora upparbetningsanläggningar som separerar uran och plutonium från varandra och från övriga ämnen i använt kärnbränsle. Dessa anläggningar kan emellertid inte avskilja de övriga transuranerna[14] – neptunium[15], americium och curium – från det högaktiva avfall som måste slutförvaras. Plutonium utgör ca 90 % av den totala mängden transuraner i använt kärnbränsle från dagens lättvattenreaktorer.

Målet för den pågående forskningen om separation (partitioning) är att finna och utveckla processer som är lämpliga för separation av tyngre transuraner (och eventuellt även vissa klyvningsprodukter) i industriell skala.

Målet för den pågående forskningen om transmutation är att definiera, undersöka och utveckla anläggningar som är lämpliga för transmutation av de nämnda långlivade radionukliderna i industriell skala.

[13] På engelska användes uttrycket partitioning = uppdelning eller avskiljning jämsides med eller i stället för separation. Den vanliga engelska (internationella) förkortningen är därför P&T för Partitioning and Transmutation.

[14] På engelska används uttrycket ”minor actinides” för att ange alla transuraner utom plutonium. Trots att neptunium är lättare än plutonium använder vi här ibland uttrycket ”tyngre transuraner” för dessa ämnen.

[15] Obs: Genom en mindre justering av driftbetingelserna i den existerande industriella upparbetnings-processen (PUREX) kan även neptunium avskiljas med denna process. Denna möjlighet utnyttjas inte idag eftersom det skulle ge ökade kostnader för rening av återvunnet uran.

De processer och anläggningar som kan bli resultat av denna utveckling måste möta mycket höga krav på säkerhet, strålskydd och miljöpåverkan. De måste vara ekonomiskt försvarbara och ge god säkerhet mot avledning av klyvbart material. De stora energimängder som frigörs i transmutationsprocessen bör utnyttjas på ett bra sätt. Processerna och anläggningarna måste accepteras av samhället.

Forskning om S&T startade redan på 1950-talet när kärnkraftsutvecklingen tog fart. Under de därpå följande decennierna var den främst knuten till utvecklingen av bridreaktorer. När denna utveckling minskade till en mycket låg nivå under det tidiga 1980-talet försvann intresset för S&T mer eller mindre.

Det förnyade intresset under 1990-talet har bidragit till en expansion av FoU-programmen särskilt på internationell nivå. I Europa fokuseras intresset till forskningsprogrammen inom EU. De s k ramprogrammen inom EU är starkt kopplade till de nationella programmen i medlemsländerna och några andra europeiska stater. Andra omfattande program pågår i bl a Japan (OMEGA), Ryssland och USA.

En genomgång av läget på S&T-forskningen gjordes på uppdrag av SKB under 1997–1998 och rapporterades i SKB R-98-06 /Enarsson 98/. Den nu föreliggande rapporten sammanfattar den utveckling som skett sedan dess.

System för separation och transmutation

Under de senaste fem åren har ett antal systemstudier kring S&T publicerats. Dessa studier ger en god överblick över det omfattande arbete som krävs för att realisera S&T, över det relativt stora och komplexa system av anläggningar som behövs samt över de problem som måste lösas och frågetecken som måste rätas ut innan ett system kan realiseras.

Den första av dessa systemstudier genomfördes av en expertgrupp inom OECD/NEA och publicerades 1999 /NEA 99/. Viktiga slutsatser från denna studie var bl a:

- Grundläggande FoU för separation och transmutation kräver långa ledtider och stora investeringar i anläggningar med snabba neutroner, kompletteringar av upparbetningsanläggningar och byggande av fjärrstyrda anläggningar för bränsletillverkning.
- Separation av långlivade radiotoxiska ämnen från använt bränsle kan göras i tillbyggnader till existerande upparbetningsanläggningar, men kräver stort arbete för att utvecklas från laboratorieskala till industriell skala.
- Transmutation med snabba neutroner är effektivare än i existerande lättvatten-reaktorer. Transmutation av transuraner kan bäst genomföras i snabba reaktorer eller i acceleratordrivna system med snabbt neutronspektrum.
- Separation och transmutation kommer inte att eliminera behovet av djupförvar för visst långlivat radioaktivt avfall från kärnbränsle.

I USA väcktes i början av 1990-talet ett visst intresse för transmutation med acceleratordrivna system. Centrum för detta intresse var Los Alamos National Laboratory, som lanserade konceptet ATW – Acceleratordriven Transmutation of nuclear Waste. Detta ledde småningom till en studie av ATW på begäran av kongressen och under ledning av USDOE[16]. Studien, som publicerades hösten 1999 /DOE 99/, utmynnade i ett förslag till forskningsprogram för ATW som kunde utgöra inledningen

[16] USDOE = United States department of energy.

på en storskalig satsning på ett sådant system. Vissa delar av detta arbete har sedan påbörjats särskilt de delar som rör internationellt samarbete. Programmet i stort har dock inte accepterats som bas för USA:s forskning på avancerade kärnbränslecykler eller framtida kärnavfallsstrategi. Det var säkert inte heller avsett som ett komplett program utan mer som en djupdykning i ett av en mängd möjliga scenarier för utveckling av separation och transmutation.

Under 2003 aviserades i USA det s k Advanced Fuel Cycle Initiative (AFCI) /DOE 03/ syftande till ett brett studium av bränslecykeln för framtida kärnkraftreaktorer – även kallade Generation IV reactors. Detta planeras genomfört i tre faser – fas 1 grundläggande utvärdering; fas 2 visa på principerna (proof of principle) under 5–6 år; fas 3 visa på funktion (proof of performance) under 15–20 år. Programmet är brett upplagt och innefattar genomgång av alla aktuella reaktortyper och system (LWR, HTR, ADS, FR; vattenbaserad upparbetning och pyrokemisk upparbetning etc).

Efter initiativ från forskningsministrarna i Frankrike, Italien och Spanien bildades under 1999 en europeisk arbetsgrupp TWG[17] under ledning av Carlo Rubbia från Italien (nobelpristagare i fysik och tidigare generaldirektör för CERN[18]). Denna arbetsgrupp föreslog en plan för utveckling av ett acceleratordrivet system (ADS) i Europa. Rapporten från TWG publicerades våren 2001 /TWG 01/. Ambitionen från gruppen var att planen skulle utgöra grund för det fortsatta EU-finansierade forskningsarbetet på ADS. I planen föreslås byggandet av en mindre experimentanläggning med ett ADS på ca 100 MW termisk effekt. Anläggningen avses bli driftklar till 2015. Kostnaden för de första tolv årens program för FoU samt konstruktion och byggande bedöms till 980 miljoner Euro. Därtill föreslås ytterligare 180 miljoner Euro för FoU på bränsle som är lämpat för ADS. Denna första fas skulle sedan följas av en andra där man utvecklar och bygger en prototyp till en ADS i fullskala. Efter framgångsrik drift av denna under några år skulle man slutligen kunna börja uppföra industriella anläggningar ca år 2040. Denna studie stämmer tidsmässigt ganska väl med den av USDOE gjorda studien. I sina detaljer innehåller de båda studierna dock avsevärda olikheter. Den amerikanska studien har betydligt mer specificerade val av system för såväl separation (pyrokemisk process) som transmutation (ATW kyld med flytande bly-vismut blandning) än den europeiska. Den senare är avgränsad till transmutation med ADS, men håller flera delar av detta system öppet för senare val av utförande – accelerator, kylmedel m m.

Arbetet inom OECD/NEA fortsattes inom en annan expertgrupp som våren 2002 kom ut med en jämförande studie av snabba reaktorer och ADS för transmutation /NEA 02b/. Gruppen gick igenom ett antal olika strategier för separation och transmutation av transuraner baserade på lättvattenreaktorer, snabba reaktorer och acceleratordrivna system. Bland de allmänna slutsatser som dras är bl a följande:

- Även om S&T inte kan ersätta behovet av ett geologiskt djupförvar för högaktivt avfall kan olika transmutationsstrategier signifikant (hundrafalt eller mer) reducera innehållet av långlivade radioaktiva ämnen som måste placeras i ett sådant förvar. Detta förbättrar miljöegenskaperna för kärnenergin och kan bidra till ett uthålligt kärnenergisystem.

[17] TWG = Technical working group.
[18] CERN = Centre Europeenne pour la Recherche Nucleaire – europeiska kärnforskningscentrum i Genève.

- Mycket effektiva metoder för transmutation i snabbt neutronspektrum och för multipel återcykling av kärnbränslet med mycket små förluster krävs för att uppnå dessa mål.
- Multipel återföring som hanterar plutonium och andra transuraner antingen separat eller tillsammans kan uppnå jämförbara reduktionsfaktorer för radiotoxicitet hos det avfall som måste slutförvaras.
- Pyrokemisk upparbetning (separation) är viktig för bränslecykler som innefattar bränslen med mycket högt innehåll av americium och/eller curium för förbränning i ADS eller snabba reaktorer.
- Separat hantering av plutonium och andra transuraner medger att plutonium till stor del kan förbrännas i vanliga reaktorer (lättvattenreaktorer) varigenom behovet av ADS och/eller snabba reaktorer minskar.
- Om plutonium och andra transuraner skall hanteras och bestrålas tillsammans får valet mellan ADS och snabba reaktorer baseras på ekonomiska, säkerhetsmässiga och andra faktorer.
- Vill man genomföra separation och transmutation krävs mer FoU framförallt på bränsle-, återförings-, accelerator- och reaktorteknik. Införandet av S&T bör troligen ske stegvis och i en takt avpassad till de nationella systemens förutsättningar.
- Man kan åstadkomma en helt sluten bränslecykel inom en begränsad kostnad-sökning på 10 % till 20 % av elproduktionskostnaden jämfört med den för lättvattenreaktorer med direktdeponering.
- Införandet av S&T-system kräver lång förberedelsetid för att utveckla tekniken och göra den kostnadseffektiv.

Några av de viktigare tekniska slutsatserna från denna studie är:

- För att uppnå målet att reducera mängden långlivade nuklider till avfallet med en faktor 100 eller mer måste förlusterna i varje separationscykel vara mindre än 0,1 %.[19]
- Transmutation av endast plutonium minskar den långsiktiga radiotoxiciteten med endast en faktor fem i jämförelse med använt bränsle.[20]
- Transmutation kan åstadkommas antingen med snabba reaktorer eller olika kombinationer av lättvattenreaktorer, snabba reaktorer och acceleratordrivna system.
- Fysikaliska begränsningar gör att det krävs lång tid att genomföra ”fullständig” transmutation. Detta innebär att man kan uppnå de beskrivna målen endast om man avser att använda tekniken i minst 100 år.
- Användningen av pyrokemiska metoder för separation innebär nya potentiella kemiska och radiologiska risker som måste behärskas.

[19] Kommentar: Detta förutsätter att man uppnår de ”transmutationsgrader” som den citerade rapporten förutsatt. Dessa beror i sin tur på bränslets egenskaper. Experiment i snabbreaktorer har visat att man kan uppnå den förutsatta ”transmutationsgraden” med t ex oxidbränsle.

[20] Kommentar: Viktiga faktorer i detta sammanhang är utbränningen och tiden mellan bestrålning och upparbetning. Vid hög utbränning och lång väntetid mellan bestrålning och upparbetning omvandlas plutonium-241 till americium-241, som sedan sönderfaller till neptunium-237. Särskilt mängden americium-241 är viktig för radiotoxiciteten i tidsperspektivet ett par tusen år.

- Alla transmutationsstrategier för lättvattenreaktorer ger avsevärda kvantiteter utarmat och bestrålat uran som måste tas om hand. Om detta uran ej betraktas som en resurs för framtida snabba reaktorer måste man även beakta dess långsiktiga radiologiska konsekvenser när radium, radon och dess döttrar växer in i uranet.[21]
- Transmutation av långlivade klyvningsprodukter innebär flera tekniska och praktiska svårigheter. För närvarande verkar teknetium-99 och möjligen jod-129 vara av de främsta eller enda kandidaterna.
- Experimentell forskning på transuranbränsle är ett högt prioriterat område. En flaskhals för sådan forskning är tillgången på anläggningar med högt flöde av snabba neutroner.
- Den grundläggande forskningen och utvecklingen på såväl snabba reaktorer som acceleratordrivna system skulle förenklas om man fick en bättre internationell samsyn om för- respektive nackdelar för olika slags kylmedel för dessa system.

Separation

Den vanligaste metoden för separation av ämnen i använt kärnbränsle är s k vätske-vätske extraktion. Denna innebär att man har en ”blandning” av vattenlösning och organiskt lösningsmedel där vissa ämnen löses i vattnet och andra i den organiska vätskan. Fördelningen av ämnen mellan vatten och organiskt lösningsmedel påverkas av vattnets surhet, närvaro av vissa reagens/komplexbildare m m. Tekniken används i de stora upparbetningsanläggningar som finns i bl a Frankrike, Japan och Storbritannien. Vid dessa separeras uran och plutonium till den organiska fasen och klyvnings-produkterna till den sura vattenfasen. Mycket av forskningen är inriktad på att vidareutveckla denna typ av process till att medge separation även av andra transuraner än plutonium, framförallt de trevärda. För några av transuranerna är det knepigt att hitta lämpliga organiska ämnen som inte samtidigt tar med sig lantanider[22] från klyvnings-produkterna. Mängden lantanider i använt lättvattenreaktorbränsle är mycket större än mängden americium och curium. Vidare har många lantanider stor förmåga att fånga in neutroner. Det är därför viktigt att man effektivt kan separera lantaniderna från americium och curium. I denna rapport redovisas ett antal olika typer av ämnen som undersöks.

Den s k PUREX-processen, som används i existerande industriell upparbetning, använder fosforbaserade organiska ämnen för att separera uran och plutonium från övriga ämnen i använt kärnbränsle. En nackdel med dessa är att de ger avsevärda mängder medelaktivt avfall som är förorenat med långlivade radionuklider. Man söker därför efter ämnen som innehåller enbart kol-, väte-, syre- och kväveatomer – kallas därför CHON-reagens. Dessa kan förbrännas fullständigt och ger därför mycket mindre mängd fast avfall. Den pågående forskningen är till stor del inriktad på denna typ av reagens.

Forskningen på vattenbaserade separationsprocesser har haft vissa framgångar under de senaste åren och man har gott hopp att utveckla en process som återvinner över 99,9 % av såväl uran som alla transuraner från använt bränsle. Problemen är framförallt kostnader och strålningsbeständighet hos de organiska molekylerna.

[21] Kommentar: Utarmat uran från anrikning erhålls givetvis även utan S&T och måste tas om hand på ett ansvarsfullt sätt.
[22] Lantaniderna är kemiskt lika aktiniderna.

När man har definierat en process som fungerar väl i liten skala är dock extrapolationen till industriell skala inte något stort problem. De tekniska komponenter som behövs i en sådan process är väl beprövade i andra tillämpningar.

De organiska molekylernas begränsade beständighet mot den starka joniserande strålningen från det högaktiva avfallet bidrog tidigt till att man försökte utveckla icke-vattenbaserade separationsmetoder. Forskningen inriktades tidigt på pyrokemiska processer. På 1950-talet byggdes en sådan process vid bridreaktorn EBR-II[23] i USA. Denna hade metalliskt bränsle och kyldes med flytande natrium. Upparbetningen av bränslet gjordes med en kontrollerad oxidativ slaggning efter det att bränslet smälts. Det renade smälta bränslet göts sedan direkt om till nytt bränsle som återfördes till reaktorn. En annan lovande metod var fluoridförångning som prövades vid MSRE[24] projektet med bränsle i form av smälta salter. Denna metod vidareutvecklades även för lättvatten-bränsle, men stötte på stora svårigheter i början på 1970-talet. Svårigheterna var bl a att hålla nere processförlusterna, kontrollen av kriticitet, hanteringen av små mängder vattenånga och syre samt korrosion.

De tekniska problemen gjorde att utvecklingen av pyrokemiska metoder starkt minskade under 1970- och 1980-talen. Intresset återupplivades emellertid parallellt med att intresset för S&T ökade i början av 1990-talet. Forskningen ökade framförallt i Japan men senare även i USA, Ryssland och Europa. De pyrokemiska metoderna bedöms framförallt vara av intresse för upparbetning/separation av bränsle som avger mycket höga stråldoser t ex bränsle med i huvudsak americium, curium och plutonium med stor andel tunga isotoper.

Utveckling av pyrokemiska separationsprocesser kräver även en teknisk utveckling av utrustning som lämpar sig för industriell tillämpning. Material och korrosionsfrågor måste lösas liksom olika tekniska och radiologiska säkerhetsfrågor. Vägen till industriell tillämpning är troligen längre för de pyrokemiska processerna än för de vattenbaserade.

Transmutation

De radionuklider som i första hand är av intresse för transmutation är transuranerna. Dessa transmuteras lämpligast ned kärnklyvning med neutroner. Flera av dessa nuklider är emellertid endast klyvbara med snabba neutroner. Två något olika typer av anläggningar övervägs för att åstadkomma ett tillräckligt starkt snabbt neutronflöde – en snabb reaktor med självunderhållande kärnreaktioner och ett acceleratordrivet underkritiskt system (ADS). Under perioden 1950–1985 byggdes ett flertal snabba reaktorer i flera länder. De flesta hade flytande natrium som kylmedel. Den största var Superphenix i Frankrike, en el-producerande anläggning med en effekt på 1200 MW_e. Endast ett fåtal av dessa snabba bridreaktorer är dock fortfarande i drift.

Under 1990-talet ökade intresset för acceleratordrivna system och för närvarande finns ett betydande intresse för studier av sådana system. I Sverige är nästan all forskning kring transmutation fokuserad mot ADS.

Ett ADS består av följande huvudkomponenter:

- En protonaccelerator som kan ge en stark ström (några få upp till 10-tals milliampère) av protoner med upp till ca 1 GeV energi per proton.

[23] EBR-II = Experimental Breeder Reactor II vid Idaho Falls i USA.
[24] MSRE = Molten Salt Reactor Experiment (smält salt reaktor experiment) vid Oak Ridge National Laboratory i USA under 1960-talet.

- En spallationskälla där protonströmmen träffar tunga atomkärnor (bly, vismut, wolfram, tantal eller liknande) och via spallation[25] frigör tiotals neutroner per infallande proton.
- En underkritisk kärnreaktor med bränsle som innehåller de långlivade radionuklider som skall transmuteras. Eftersom transmutationen sker genom kärnklyvning frigörs ytterligare neutroner. För varje neutron från spallationskällan bildas på så sätt ca 20 neutroner, som ger kärnklyvningar. Reaktorn utformas så att en själv-underhållande kedjereaktion inte kan uppkomma.
- Utrustning för att kyla bort det värme som frigörs och ta hand om energin på lämpligt sätt (t ex elproduktion). Utrustning för elmatning till acceleratorn.
- Utrustning för att styra, övervaka och kontrollera hela anläggningen.

Protonacceleratorer kan vara av två olika principutföranden – en linjär accelerator eller en cyklotron (cirkulär partikelbana). Cyklotroner har vissa tekniska begränsningar i fråga om protonenergi, strömstyrka och flexibilitet. Detta gör att intresset alltmer fokuseras mot en linjär accelerator. En sådan maskin för den önskade protonenergin blir mycket stor – flera hundra meter lång – och ställer stora krav på all möjlig teknik och även på strålskyddet.

Acceleratorer har hittills huvudsakligen byggts för rena forskningsuppgifter där kraven på tillgänglighet varit måttliga. En anläggning för transmutation innehåller emellertid stora system med hög temperatur och inbyggd tröghet för förändringar i systemens tillstånd. Detta tillsammans med krav på effektivitet och rimlig ekonomi ställer starkt ökade krav på tillgängligheten hos acceleratorer till ADS.

Spallationskällan är en central komponent i ADS placerad mellan acceleratorn och den underkritiska reaktorn. Den är utsatt för en krävande miljö med hög strålningsnivå, hög temperatur, starkt varierande tryck och krav på mycket effektiv kylning. Olika tunga material kan i princip tänkas för källan (bly, vismut, wolfram, tantal etc) men med hänsyn till kylningen och andra krav är för närvarande en blandning av flytande bly och vismut den främsta kandidaten. Alternativen är ett ”varmt” fönster bestående av en strålningsbeständig specialmetall eller ett ”kallt” fönster placerat en bit från spallationskällan. Det senare fallet ställer stora krav på upprätthållande av tillräckligt gott vakuum i ”tilloppskanalen” mellan fönstret och spallationsmaterialet så att protonerna ej förlorar energi på vägen. Ett ”varmt” fönster ställer å andra sidan utomordentliga krav på strålningsbeständighet och hållfasthet så att fönsterbyten kan begränsas till högst en gång om året. För närvarande byggs eller planeras två större spallationskällor i Europa med flytande bly-vismut dels MEGAPIE vid PSI i Schweiz, där man valt en konstruktion med ”varmt” fönster, och dels MYHRRA vid Mol i Belgien, där man ämnar prova ett utförande med ”kallt” fönster. Utformningen med ”kallt” fönster kallas ofta även ”fönsterlös” design.

Olika typer av utförande diskuteras för den underkritiska reaktorn. Såväl bränsle som kylmedel är ännu öppna för val. Vatten är otänkbart för kylning om man vill ha ett snabbt neutronspektrum. Tänkbara val är gas (särskilt helium) eller flytande metall. Gaskylning kräver högt tryck och förkastas därför av många med hänsyn till säkerheten mot kylningsbortfall. Flytande natrium har man betydande erfarenheter av från tidigare program för snabba reaktorer. De problem som därvid framkommit avskräcker tydligen,

[25] Spallation är en kärnfysikalisk process där partiklar med hög energi träffar atomkärnor och kraftigt omvandlar dessa samtidigt som ett flertal lätta partiklar frigörs.

åtminstone för närvarande, och intresset riktas alltmer mot flytande bly-vismut i eutektisk blandning. Denna blandning har en relativt låg smältpunkt och en hög kokpunkt och är dessutom första kandidat till material i spallationskällan. Det senare förenklar naturligtvis utformningen av systemen. Erfarenheter från reaktorer kylda med flytande bly-vismut finns endast i Ryssland där man byggt och drivit reaktorer med detta kylmedel i sju stycken atomubåtar. Sammanlagda drifttiden är ca 70 reaktorår.

Forskningen på bränsle för ADS följer flera parallella linjer. Nitrider och legeringar med inerta metaller t ex zirkonium tilldrar sig betydande intresse. Kraven på bränslet innebär bl a god värmeledningsförmåga som medger hög effekttäthet, god bestrålningsbeständighet och möjlighet till mycket hög utbränning – 10-tals procent jämfört med några få procent i dagens lättvattenreaktorbränsle.

Snabba reaktorer kan, som redan nämnts, användas för transmutation. Under 1990-talet har forskningen bl a varit inriktad på förbränning av plutonium i natriumkylda snabbreaktorer. Speciella typer av bränsle studerades i Frankrike. Under senare år har intresset i bl a Frankrike också riktats mot gaskylda snabbreaktorer. Snabbreaktorn kan också användas för transmutation av andra transuraner. Detta kräver dock speciell utformning så att man inte allvarligt försämrar reaktorns dynamiska och säkerhetsmässiga egenskaper. Utformar man snabbreaktorn som en bridreaktor kan man dels uppnå en mycket hög energiutvinning från det uran som man tar upp ur jordskorpan[26] och dels en effektiv transmutation av alla transuraner.

I Ryssland intresserar man sig också för civila tillämpningar av snabba reaktorer kylda med flytande bly-vismut. Dessutom görs vissa insatser på ADS i huvudsak finansierat via det s k ISTC-programmet[27].

Som redan har nämnts är system med högt flöde av snabba neutroner av störst intresse om man vill åstadkomma en nära nog fullständig förbränning av alla långlivade transuraner. Nöjer man sig med att transmutera plutonium kan man emellertid också komma långt med andra system. Franska studier visar att man med speciell utformning av bränsleelementen kan begränsa och stabilisera mängden bildat plutonium i en park med lättvattenreaktorer. Studier av General Atomics i USA m fl har visat att man kan utveckla bränsle till en gaskyld högtemperaturreaktor som medger utbränning upp till 700 MWd/kg och förbränning av mer än 90 % av aktinidinnehållet inklusive plutonium-239.

Några slutsatser

FoU-insatserna på separation och transmutation har ökat något under perioden 1998–2003. Forskning om S&T har intagit en framträdande roll internationellt inom FoU över framtida kärnkraft- och kärnbränslecykelsystem.

Trots det faktum att separation och transmutation har stått på agendan under ganska många år finns det fortfarande ett antal frågor som måste klaras ut innan forskning och utveckling på området kan ges en klart fokuserad inriktning. Ovan nämnda studier i USA och i Europa har försökt att definiera det program som krävs för att komma i ett

[26] Studier inom OECD/NEA anger att man med en bridreaktor kan ta ut upp till 100 ggr mer energi per ton natururan jämfört med en lättvattenreaktor med direktdeponering (engångscykel) /NEA 02b sid 230/.
[27] ISTC = International Science and Technology Centre; en institution finansierad av EU, Japan, Ryssland och USA med syftet att engagera tidigare sovjetiska kärnvapenforskare i civila projekt. Innan Sverige blev medlem i EU deltog Sverige direkt som en finansiär av ISTC. Waclaw Gudowski, KTH är med i den internationella grupp som utvärderar projektförslag rörande S&T som föreslås för finansiering av ISTC.

sådant läge. I studierna föreslogs forskningsprogram för ca sex år till kostnader på ett par hundra miljoner Euro.

Byggandet av en mindre experimentanläggning med ADS är ett nödvändigt steg för att utveckla och demonstrera konceptet. Denna experimentanläggning bör sedan följas av en demonstrationsanläggning i nära full skala. En sådan kan stå klar tidigast i mitten på 2030-talet.

Ett antal faktorer har emellertid bidragit till att farten, intensiteten och anslagen för insatserna är mycket mindre än vad som föreslogs.

Det finns ingen enhetlig syn på målen för separation och transmutation. Många ser det som ett sätt att nå en bred acceptans för kärnenergi i stort. Andra, som ett sätt att komma ur den blockering som drabbat djupförvarsfrågan i flera länder. Återigen andra fäster stor vikt vid spridningsriskerna med en starkt växande lager av plutonium från lättvattenreaktorbränsle, från andra reaktorer och från avveckling av kärnvapen.

Det finns ingen enhetlig syn på behovet av att utveckla acceleratordrivna system eller för dessa systems roll vid S&T. Några förordar att ADS skall användas för att transmutera alla transuraner från uranbränsle i nuvarande lättvattenreaktorer. Andra ser ADS som ett komplement som är särskilt lämpat för att transmutera neptunium, americium och curium medan det mesta plutonium bör förbrännas i lättvattenreaktorer (eller i snabba reaktorer).

Det finns ingen konsensus mellan experter om vilken teknisk inriktning man bör följa för viktiga delar av ett S&T-system.

Intresset för S&T är huvudsakligen koncentrerat till forskningslaboratorier i USA, Europa, Japan och några andra länder. Universiteten i många länder inklusive Sverige visar också stort intresse för S&T. Forskningen tilldrar sig betydande intresse bland studenter i kärntekniska vetenskaper. Flera viktiga program pågår vid universitet och forskningslaboratorier i många länder.

Kärnenergiindustrins intresse för denna utveckling är mycket begränsat och huvudsakligen märkbart i Frankrike och några få andra länder med stora kärnkraftprogram. Industrins långsiktiga intresse är mer fokuserat på nya typer av reaktorer – ofta kallade Generation IV.

De nationella och industriella insatserna på använt kärnbränsle och högaktivt avfall i nästan alla länder är fokuserade på att lösa frågor kring geologisk djupförvaring.

En sammanvägning av dessa olika omständigheter leder till slutsatsen att det är osannolikt att ADS-anläggningar i industriell skala kan tas i bruk före 2050.

En framgångsrik utveckling av S&T kommer inte att göra djup geologisk förvaring föråldrad. De komplexa processerna kommer oundvikligen att ge visst avfall med små mängder långlivade radionuklider. För detta avfall behövs djupförvar. En framgångsrik utveckling kan emellertid minska dels kraven på de tekniska barriärerna i djupförvaret och dels de volymer som behövs för deponering av avfall.

För svensk del är det viktigt att följa den internationella utvecklingen och att upprätthålla en rimlig kompetens inom landet så länge som Sverige har en betydande andel av kärnkraft i elproduktionssystemet. Kompetens utvecklad genom S&T-FoU är värdefull och användbar även i arbetet att underhålla och utveckla säkerhet och bränsleförsörjning för de existerande lättvattenreaktorerna. Den är också viktig för att bedöma den fortsatta utvecklingen av kärnavfallshanteringen.

Såsom redan nämnts kan utvecklingen av S&T för tillämpning i stor skala förväntas ta flera decennier. För att sedan transmutera allt kärnbränsle bara från de nu existerande kärnreaktorerna kommer det att krävas ytterligare åtminstone 100 år. S&T i industriell skala kräver stora kärntekniska anläggningar som måste accepteras av samhället.

Kostnaderna för S&T är inte möjliga att förutse med någon god säkerhet innan systemen definierats bättre och provats. De uppskattningar som gjorts pekar på en elproduktionskostnad[28] som ligger mellan 10 % och 50 % över den från dagens lättvattenreaktorer. De investeringar som krävs i FoU och i nya anläggningar är mycket omfattande. De är dock utspridda över lång tid och större delen får ses som investeringar i energiproduktion. Det är icke ekonomiskt rimligt att genomföra S&T utan att nyttiggöra energin. Detta gäller framförallt transmutation av plutonium. Vissa experter anser att man kan bygga mindre ADS-anläggningar för transmutation av americium, curium och neptunium.

Införande av ett S&T-system för att effektivt reducera den mängd långlivade radionuklider som måste slutförvaras i ett geologiskt djupförvar kräver således en bindning till kärnenergi för mycket lång tid. Det kan å andra sidan ses som en långsiktig strategi för återföring och återvinning av energi från ämnen som annars betraktas som mycket besvärligt avfall.

[28] Den uppskattade elproduktionskostnaden för ett kärnenergisystem med S&T är dock fortfarande lägre än elproduktionskostnaden för system med s k förnybar energi.

Contents

1 Introduction

The research and development on methods for partitioning and transmutation (P&T) of long-lived radionuclides in spent nuclear fuel has attracted considerable interest during the last decade. The main objective of P&T is to eliminate or at least substantially reduce the amount of such long-lived radionuclides that has to go to a deep geological repository for final disposal.

The radionuclides of main interest (concern) are those of the transuranium elements – neptunium, plutonium, americium and curium – see e.g. /SKB 95/. These elements are formed in a nuclear reactor by one or more neutron captures in uranium atoms and subsequent radioactive decay. The reduction of long-lived radionuclides can be achieved by transmutation of the nuclides by the use of nuclear physics processes. In theory, several such processes are possible. In practice so far only transmutation by irradiation with neutrons can be made in macroscopic scale. Neutrons can cause fission in the transuranium elements and this process will release a substantial amount of energy. Thus transmutation on large scale of the transuranium elements from spent nuclear fuel must be done in a device similar to a nuclear reactor.

A prerequisite for transmutation by irradiation with neutrons is that the nuclides to be transmuted are separated (partitioned) from the other nuclides in the spent fuel. In particular the remaining uranium must be taken away unless you want to produce more plutonium and other transuranium elements. Separation of the various elements can at least in principle be achieved by (mechanical and) chemical processes. Currently there exist some large scale facilities for separation of uranium and plutonium from the spent fuel – reprocessing plants. These can, however, not separate the heavier transuranium elements – neptunium[29], americium and curium – from the high level waste that goes to a repository. Plutonium constitutes about 90% of the transuranium elements in fuel from light water reactors.

The objective of current research on partitioning is to find and develop processes suitable for separation of the heavier actinides (and possibly some long-lived fission products) on an industrial scale.

The objective of current research on transmutation is to define, investigate and develop facilities that may be suitable for transmutation of the aforementioned long-lived radionuclides.

The processes and facilities that could be implemented as results of such developments must meet very high standards of safety and radiation protection as well as have low environmental impact. They shall be economically viable and have good proliferation resistance. The large amount of energy released in the transmutation process should be used in a proper way. In other words the processes and facilities must be acceptable to society.

[29] Neptunium can be separated with uranium if a minor adjustment of the operating conditions is made. This possibility is not used today. In fact almost 30 % of the neptunium is, however, already recovered in the uranium purification part of the plants and then returned to the high level waste stream together with trace amounts of plutonium and uranium.

Research on P&T started already in the 1950ies when development of nuclear power gained momentum. In the subsequent years it was mainly tied to the development of the breeder reactor. As this development slowed down to a very low level in the early 1980ies the interest in P&T more or less disappeared.

The renewed interest through the 1990ies has caused some expansion of the programmes in this field in particular on an international level. In Europe this is focused on the R&D-programmes of the European Union (EU). The EU so-called framework programmes (FWP) have established a strong link between the various national programmes within the union and also in some other European countries. Other large programmes are going on in Japan (OMEGA), USA and Russia.

The programme in Sweden is mainly financed by SKB and was initiated in the early 1990ies. At present research on partitioning is made by the nuclear chemistry group of the department of materials and surface chemistry at Chalmers University of Technology in Göteborg. The research on transmutation is made at the departments of nuclear and reactor physics and of nuclear safety at the Royal Institute of Technology (KTH) in Stockholm. Research on basic physics data for transmutation is made at the department of neutron research at Uppsala University. All these Swedish research teams are participating in projects that are partly financed by the European Commission of EU. They are also engaged in other international cooperation. Of particular interest are some projects in former USSR states financed through the International Centre for Science and Technology (ISTC) by EU, USA, Japan, South Korea and Norway. The projects related to transmutation are closely followed by the KTH physics team. Some of the cross section measurements financed by ISTC are performed at the facilities in Uppsala in close contact with the Uppsala researchers. The current level of funding for the P&T-research in Sweden is about 5 MSEK/year from SKB and another about 3.5 MSEK/year from EU and other sources.

The efforts in Sweden are small compared to the international activities. The main programmes in Europe are those financed by EU and the national programme in France. The EU budgets for the recent FWP's are shown in Table 1-1.

A review of the status of the efforts concerning P&T was published by SKB in 1998 /Enarsson 98/. Short reviews of the previous development have also been given in SKB's RD&D-programmes 95, 98 and 2001 /SKB 95; SKB 98; SKB 01/.

In its decision concerning the RD&D-programme 2001 the Swedish government pointed out that SKB should "…continue to follow the technical development concerning different alternatives for management of nuclear waste within the framework of the RD&D-programme." /Gov 02/

Table 1-1. P&T-budget in EU-programmes.

Programme	Time frame	Budget M€
FWP 3	1991–1994	4.8
FWP 4	1994–1998	5.8
FWP 5	1999–2002	28.6
FWP 6 – tentative	2003–2006	30–35

Following its review of the RD&D-programme 2001, KASAM[30] proposed "…that SKB should present, in RD&D-Programme 2004, a more detailed basis for the evaluations made of the size of SKB's financial support to research and acquiring of information on transmutation" /KASAM 02/.

This report summarises the work reported in the years 1998–2003 and tries to assess the prospects for future development of P&T as seen from a Swedish perspective.

The report has been co-authored by members (and their associates) of a Swedish reference group on P&T-research established by SKB. This group includes members from the research teams that are active in Sweden on this subject and a chairman appointed by SKB. The Swedish authorities have appointed observers to the reference group. The current composition of the group is given in Appendix IV.

The objectives of this report are to:

- Present the current situation on P&T.
- Give a base concerning P&T for SKB's next RD&D-programme.
- (Fulfil the requirement in the statutes for the aforementioned P&T reference group.)

The report is structured in five main chapters. Chapter 2 summarises recent work concerning a "complete" P&T-system. Chapter 3 gives an account of the current research concerning partitioning and Chapter 4 a corresponding account concerning transmutation. Chapter 5 attempts to present views on the prospects for future development of P&T from various aspects such as R&D issues, safety issues, needs for demonstration plants, time and cost, impact on waste management systems and acceptance by society. The last chapter, Chapter 6, presents some conclusions or recommendations that the authors believe are justified at the current stage.

Appendix I describes the current EU-projects on P&T in some detail. Appendix II summarises the P&T-related projects in Russia that are financed by ISTC. Appendix III gives a brief account of the P&T research at the Swedish universities.

A list of acronyms used or occurring in the text is given in Appendix V. Most of the acronyms are also explained in footnotes on the page where they first occur.

[30] KASAM = Swedish National Council for Nuclear Waste.

2 Recent P&T-systems studies

2.1 NEA – actinide and fission product P&T – status and assessment report 1999

The nuclear development committee within the OECD/NEA has followed the development of P&T since its restart in the early 1990ies. A series of information exchange meetings have been organised /NEA 90; NEA 92; NEA 94; NEA 96; NEA 98; NEA 00; NEA 02a/. In 1996 the committee set up an expert group representing twelve countries and three international organisations. The aim was to make a systems analysis of the P&T-strategy. The group reported its findings in the end of 1998 at the information meeting in Mol /NEA 98/ and in an OECD/NEA report /NEA 99/. The following brief summary is based on that report.

The group states that much of the current work in this area is at an early stage of development. It is difficult to predict the outcome or associated timescales of current research and development or of the costs associated with development and operation of technologies under study. The structure of a total system study and the related requirements for information are outlined in the report but the primary focus for the present is on review of the potential technologies for P&T together with a critique of the development status and prospects for eventual industrial application.

The report is subdivided in two main parts – a general part for the non-specialist and a technical part discussing details on several technical issues in partitioning, transmutation and long-term waste management. Three principal fuel management schemes are discussed in some detail:

1. Once through fuel cycle – meaning final disposal of the spent fuel after only one cycle of irradiation in a reactor core (and usually with some cooling time after irradiation).
2. Plutonium recycling in current light water reactors called “conventional fuel cycle”.
3. Advanced recycling of (some) transuranium elements in advanced reactors.

The main findings of the expert group are summarised in the last two chapters of the general part under the headlines “Critical evaluation” and “General conclusions”.

Concerning the critical evaluation the report says:

- P&T may take its place in a future nuclear fuel cycle industry either as an additional activity or as a partial replacement of present fuel cycle activities.
- The P&T option can be chosen only if a conventional or advanced fuel cycle is operated. But during the R&D phase all countries interested in a development of the fuel cycle or waste management can participate in an international effort.
- The P&T strategy should be oriented towards the gradual elimination of long-lived radionuclides from waste streams for which decisions in the field of waste disposal should be taken. But this approach will lead to a reduction of the radiotoxic inventory in the waste at the expense of a limited increase of radiotoxic inventory in the reactor cores and in the fuel cycle facilities unless a comprehensive actinide incineration route such as accelerator-driven transmutation is set up.

- In the course of a long-term nuclear programme there might be a gradual shift from LWR-UO_2 fuel to LWR-MOX fuel to be followed by the introduction of fast reactors in the nuclear power plants and by the use of accelerator-driven systems.
- Partitioning of minor actinides is an additional step to U and Pu recycling in MOX fuel. In order to carry out partitioning operations a number of additional industrial facilities will have to be designed and constructed. But the present state of the art in partitioning is at the laboratory and hot-cell scale of development.
- Partitioning is a long-term venture which needs newly designed facilities for treatment of high level liquid waste or an adapted PUREX extraction cycle with direct separation of all long-lived radionuclides.
- Partitioning of minor actinides from high level liquid waste reduces the long-term radiotoxic inventory of vitrified high level waste and contributes to improving the hazard perception of the high level waste to be disposed of. However, it does not modify the technical aspects of waste. The fission products (^{137}Cs and ^{90}Sr) determine the heat output of high level waste, while leach rates of the transuranium elements, which determine the radiological impact in the long term, are controlled by their low solubilities.
- Partitioning of minor actinides (Np, Am, Cm) and selected fission products (Cs, Sr, Tc, etc) creates more opportunities to improve the nuclide-specific conditioning of long-lived radionuclides. The increased thermodynamic stability of new waste forms offers, in principle, better perspectives for very long-term storage of transuranium elements in comparison with vitrification which has been designed especially for complex mixtures of minor actinides and fission products, Further development of partitioning methods and technologies is strongly recommended in order to broaden the technical basis for engineering assessment of partitioning.
- Transmutation is a general term, which covers incineration, i.e. transformation into fission products, and transformation by neutron capture into another radionuclide or a stable isotope. The development of this technology may have important spin-offs for other nuclear programmes.
- Some transmutation reactions produce a majority of non-radioactive nuclides, some generate a variety of radionuclides with a range of half-lives. The produced nuclides should have a shorter half-life than the target, or a lower radiotoxicity, or lead in some cases to the production of an other mother nuclide with less radiological impact.
- Transmutation of minor actinides is in the short term not always conducive to the reduction of the short term radiotoxic inventory.
- Transmutation in thermal reactors proceeds in general by neutron capture. Recycling of industrial quantities of minor actinides requires an increased enrichment level with an accompanying increase in the nuclear power production cost. Yet, heterogeneous recycling of minor actinides in LWR-MOX fuelled reactors is possibly a viable route as long as surplus Pu is available to compensate for the negative reactivity induced by the minor actinides. However, on this transmutation route, the overall radiotoxicity of the fissile and fertile materials of the fuel cycle does not decrease significantly.

- Transmutation of minor actinides in fast neutron reactors reduces the radiotoxicity since all minor actinides are fissionable to a certain degree. The higher the mean neutron energy the higher the "incineration" yield. But for reasons of reactor safety the acceptable minor actinide loading and the resulting fission and transmutation yields are limited. The theoretical energy potential of the minor actinides is about 10% of that of plutonium generated in UO_2 fuel. The fast neutron spectrum devices (fast reactors, accelerator-driven systems) are more promising than LWR s for the transmutation of minor actinides.
- Some long-lived fission products have been considered as candidates for partitioning and transmutation. ^{129}I, ^{135}Cs, ^{99}Tc and ^{93}Zr are the most frequently mentioned.
- Partitioning of some fission products is already achieved industrially; iodine is removed from spent fuel solutions by sparging, scrubbing and separate treatment before disposal into the oceans (in some countries the discharge of iodine is not allowed). If the discharge of ^{129}I to the ocean should become difficult to continue over a long period of time, conditioning and perhaps transmutation ought to be re-examined. The transmutation of ^{135}Cs and ^{93}Zr is impossible without isotopic separation.
- The theoretical basis for Tc separation has been described but no practical implementation has been started. Transmutation of ^{99}Tc is possible in thermal reactors and in thermalized sections of fast reactors. However, the transmutation half-life in a thermal spectrum device is fairly long (15 to 77 years). Unless neutrons will become available at marginal cost in future nuclear facilities (ADS) this option should not get first priority in R&D.

The report presents the following general conclusions:

- Fundamental R&D for the implementation of P&T needs long lead times and requires large investments in dedicated fast neutron devices (fast reactors (FR), accelerator-driven systems which may complement or substitute for operating LWRs), extension of reprocessing plants and construction of remotely manipulated fuel and target fabrication plants.
- Partitioning facilities for minor actinides and some long-lived fission products could be designed and constructed as extensions to existing reprocessing plants.
- Partitioning methods for long-lived radiotoxic elements have been developed on a laboratory scale but much work is still to be done on improving and scaling up the partitioning technology in order to make it comparable with industrial reprocessing practices.
- The short-term impact of partitioning would be to reduce the long-term radiotoxic inventory of the resulting high level waste at the expense of an increase of the operational requirements for the nuclear facilities concerned.
- Fast neutron-spectrum devices (dedicated fast reactors or accelerator-driven system facilities) are more efficient than current LWRs for recycling and transmuting long-lived radionuclides. The accelerator-driven system may play an important role in "incinerating" the actinides and some fission products from spent LWR-MOX or FR-MOX fuel in order to reduce the long-term radiotoxic inventory of the wastes.

- Recycling of plutonium and minor actinides could stabilise the transuranium inventory of a nuclear power plant park. Multiple recycling of the transuranium elements is a long-term venture in which it may take decades to reach equilibrium transuranium inventories.
- Conditioning of separated long-lived nuclides in appropriate matrices which are much less soluble than glass in geological media, or which could serve as irradiation matrix in a delayed transmutation option, is a possible outcome for the next decades.
- P&T will not replace the need for appropriate geological disposal of high level waste, irradiated transuranium concentrates and residual spent fuel loads from a composite reactor park.

2.2 US – a roadmap for developing ATW (ADS) – October 1999

2.2.1 The study and its main results

In 1999 the US Congress directed the United States Department of Energy (DOE) to study the accelerator transmutation of waste (ATW) and to prepare a "roadmap" for developing this technology. The DOE Office of Civilian Radioactive Waste Management managed this task.

The report /DOE 99/ was prepared by the experts from US national labs and DOE. It addressed the following objectives:

- Identify the technical issues that must be resolved.
- Propose a schedule and program to resolve these issues.
- Estimate the cost of such a program.
- Propose collaborative efforts with other countries and other programs developing ADS technology.
- Assess the institutional challenges of an ATW program.
- Assess the impact ATW technology could have on the civilian spent nuclear fuel program.
- Assess areas of development that could have benefits to other ongoing programs.
- Estimate capital and operational life-cycle costs to treat spent fuel.

The results of US roadmapping were summarized in eight action items:

1) Six-year, science-based R&D programme described to address the key technical issues.

The programme would be capable of reducing the technical risk and assessing the technical viability of the ATW technology. The key technical issues identified are:

- lifetimes of proposed materials and components,
- reliability of components,
- degree of partitioning and separations, and
- quantification of long-lived radioactivity generated in operations, including spallation products.

2) R&D plan to resolve issues developed and schedule identified.

Three years: complete systems and trade studies to justify major technical choices, complete an institutional analysis; five years: complete a pre-conceptual system design, complete a detailed research, development, and demonstration (RD&D) plan to begin after year six; at the end of six years, complete initial R&D and assess technical viability of ATW.

3) Cost of six years of R&D estimated.

Total cost about M$ 281 consisting of:

- systems studies, M$ 18,
- accelerator R&D, M$ 58,
- separations R&D, M$ 55,
- target/blanket R&D, M$ 123,
- programme management, M$ 26.

4) Opportunities for collaboration identified.

Potential for active technical and financial collaboration with ongoing programmes in Europe, Russia, and Asia.

5) Institutional challenges identified.

Significant challenges for US policies (e.g. nonproliferation of nuclear weapons and reprocessing), long-term programme management commitments, long-term financing commitments, regulatory and environmental implementation, and public acceptance.

6) Impacts on the civilian spent fuel programme assessed.

Some impact of ATW, if successful, on the first repository programme, could reduce potential long-term radiation doses from repository wastes by a factor of about 10. However, a repository is still required due to the presence of defence wastes, which are not readily treatable by accelerator transmutation of waste, and the long-lived radioactivity generated by ATW operations. A reduced inventory of plutonium and other transuranics in the repository could decrease the likelihood of a future unauthorized attempt to recover fissile material.

7) Benefits to ongoing programmes identified.

Supportive to programmes in nuclear nonproliferation, nuclear safety, waste management, high-power accelerators, nuclear chemical processes, nuclear fuels, next-generation nuclear fuel cycles, materials science, and nuclear technology in general.

8) Capital and operational life-cycle costs to treat spent fuel estimated.

Total life-cycle cost to treat 87 000 tonnes of commercial spent fuel: approximately G$ 281 (G$ 2 R&D, G$ 9 demonstration, and G$ 270 post-demonstration design, construction, operation, and decommissioning). Such a large upfront expenditure commitment will be a major challenge. Over the life-time of ATW plant operations, much of the capital, operational, and development and demonstration costs may be offset by the sale of electricity. However, when the time value of money is considered, this offset may be small. Total time: 117 years, of which R&D (initial 8 years) and demonstration comprise the first 27 years, and post-demonstration period activities comprise the following 90 years.

The system evaluated in the cost assessment consisted of eight (8) ATW stations each having the following facilities:

- Two accelerator units – each linear proton accelerator with 1 GeV and 45 mA (i.e. 45 MW).
- One spent fuel processing facility consisting of a modified PUREX system for separation of uranium only and an electrometallurgical system for separation of the transuranium elements from the fission products. Capacity 175 MTU/yr of fuel from LWR
- Eight transmuter units forming four power blocks with two units each. Steel clad metallic fuel with nominal composition 23% TRU and 77% Zr, cooled with liquid Pb-Bi. Spallation source is liquid Pb-Bi – the same as the coolant that also cools the window of the source. The thermal power of each transmuter unit is 840 MW_{th} giving 6720 MW_{th} per station. The efficiency is 37% giving a gross electric output of 2486 MW_e of which 285 MW_e will be used for the accelerators and other plant equipment.
- Four electric generation units and one station switchyard.
- One ATW fuel unit for preparation/fabrication/recycle/refabrication of ATW fuel and for target processing/fabrication/recycle/refabrication, including waste treatment and packaging.
- One waste transport and disposal function for ATW radioactive wastes – ATW station to disposal sites and emplacement.

The life cycle of each of the eight ATW stations would be 76 years from the start of design to final decommissioning. Some details of the costs are given in Section 5.5.

2.2.2 Conclusions from the ATW roadmap study

The conclusions from the ATW roadmap studies were summarized as follows:

- A repository is an essential element of the nuclear fuel cycle with or without ATW deployment. It is required in the US for disposal of defence high-level waste, DOE-owned spent fuel, and civilian spent fuel.
- Applying an ATW system to commercial civilian spent fuel as described in this report, would reduce its contribution to the dose predicted in the total system performance analysis for the Yucca Mountain repository project. The defence high-level waste and DOE-owned spent fuel contents of the repository would then dominate long-term dose, and predicted peak doses would be reduced by a factor of ten. The inventory of fissionable materials from commercial spent fuel in the repository could be reduced by a factor of 1000. The volume of waste packages associated with commercial spent fuel would be slightly reduced. The 82 000 t of chemically separated uranium could be suitable for near-surface disposal at a low-level waste site or non-shielded storage for potential future use in advanced reactors.
- For a deployment-driven R&D and development schedule, a program resulting in a near full-scale prototype to demonstrate the ability to deploy an ATW system would require approximately 20 years and cost about M$ 11 000. Public acceptance as well as several other complex institutional issues would have to be addressed in the process leading to a decision for deployment.

- The roadmap developed for deployment of ATW systems for partitioning/ transmuting of commercial spent fuel depends on implementation scenarios that were assumed for decades into the future, and it therefore has large uncertainties. For the aggressive implementation scenario assumed in the roadmap study, several decades and several tens of billions of dollars will be required; the scale of energy recovered and its value in the marketplace is comparable to that of large power plants currently operating. Much of the capital, operational, and D&D[31] costs may be offset by the sale of electricity.
- Auxiliary benefits would be derived from development of ATW technology. Among them is the opportunity to participate in and influence international efforts in areas of nuclear nonproliferation, safety, and waste management.
- Active participation by the US in international ATW R&D efforts could have positive impacts on cost and schedule for US ATW programmes and may provide access to facilities/capabilities not available in the US.
- Although no "show stoppers" are identified, a several-year science- and technology-based R&D programme would be prudent to increase the knowledge base needed for ATW. Such a program would likely provide additional benefits in related technologies (e.g. materials science, accelerators, etc).

2.2.3 Recommendations from the ATW roadmap study

The US Administration made no recommendation with regard to the proposed research programme. DOE stated: Should there be a national policy decision to pursue this technology, the expert group has identified a science-based research programme to address the technical issues cited in the report. The pace and funding for a research programme would then have to be carefully evaluated and planned in light of the currently unproven technologies involved, the potential benefits that may be gained, and current and evolving Government budget realities.

An initial science and technology development pathway involving international collaboration was recommended by the ATW Steering Committee.

Recommendation 1: An initial period of up to six years should be undertaken for trade and system studies, for two distinct purposes. The first would be to evaluate ATW within the framework of nonproliferation, waste management, and economic considerations. The second would be to evaluate the efficacy of the numerous technical options for ATW system configuration, trading off such factors as accelerator type and power, size (heat rating) of each transmuter module, the neutron spectrum characteristics and effectiveness for transmutation, type of nuclear fuel and coolant, technologies for materials separation under proliferationresistant conditions, etc. These trade and system studies would refine the goals and requirements for an ATW system.

Recommendation 2: During the six-year period, science-based R&D should address the key technology issues identified during the roadmap preparation for each of the system elements (partitioning, recycle, accelerator, spallation target, and transmuter), identified in Section 2 of this Report to Congress[32]. Special attention should be directed to the high-priority issues identified earlier. The science-based R&D and trade studies

[31] D&D = US Department of Defence.
[32] Refers to /DOE 99/.

should support each other mutually, e.g. establishing feasibility and identifying other options.

Recommendation 3: Recognizing existing programmes in Europe and Japan, the above recommendations should be undertaken in the form of strong international collaboration at the level of system studies, system configuration, and on science-based R&D. After establishing technology preferences, opportunities for collaboration on international ATW technology demonstration efforts should be explored.

Recommendation 4: A total funding level of approximately M$ 281 is needed for support of the initial six-year science- and technology-based R&D programme. This level will allow deliverables projected for the six-year program to be met.

Recommendation 5: At the end of the fifth year or the beginning of the sixth year of an initial R&D programme, the programme would be able to prepare for Congress the following deliverables:

- A reference ATW system definition at a preconceptual level.
- A description of the status of ongoing science and technology efforts highlighting major interim results and needs.
- A development and demonstration plan for ATW including further use of existing US and/or international facilities and identification of demonstration facilities to be constructed.
- A preliminary design and cost for such demonstration.
- Institutional analysis defining the strategy for addressing and dealing constructively with issues such as regulation, public acceptance, etc.

Some of the conclusions and recommendations of the roadmap have already been implemented, e.g. much closer collaboration between Europe and USA has been established exemplified in US participation in a few European projects like MEGAPIE, MUSE, TECLA, nuclear fuel projects etc. Moreover US scientists are actively participating in the preparation of projects for the 6th FWP.

The US roadmap as a whole has, however, not been accepted as a base for US research efforts on advanced nuclear fuel cycles and/or waste management strategies.

2.3 A European roadmap for developing ADS – April 2001

In 1998 the Research Ministers of France, Italy and Spain, set up a Ministers' Advisors Group on the use of accelerator driven systems (ADS) for nuclear waste transmutation. This led to the establishing of a technical working group (TWG) /Rubbia 99/. The mission of TWG was to identify the critical technical issues and later also to prepare a "roadmap" for a demonstration programme to be performed within 12 years.

An extended TWG – consisting of experts from Austria, Belgium, Finland, France, Germany, Italy, Portugal, Spain, Sweden and the JRC[33] – started its work in the spring of 1999 and issued a report in April 2001 /TWG 01/. The report identified necessary steps to start the construction of an experimental accelerator driven system towards the end of the decade. This was considered as an essential prerequisite to assess the safe and efficient behaviour of such systems for a large-scale deployment for transmutation purposes in the first half of this century.

[33] JRC = Euratom Joint Research Centre; part of the EU cooperation.

The first goal of the TWG was to propose a technological route to reduce the long-term potential hazards associated with nuclear waste, based on the transmutation of nuclear waste in ADS; and to assess the impact of this approach in the reduction of the radiotoxicity of nuclear waste. The second and main goal of the TWG was the preparation of a technical programme, with cost estimates, which would lead to the realisation of an experimental ADS within 12–15 years, covering the 6th and 7th Framework Programmes within EU. The Report /TWG 01/ sketched a programme plan (roadmap) leading to the development of innovative fuels and reprocessing technology, a co-ordination of human resources and experimental facilities, a training ground for young researchers, spin-offs in the fields of accelerators, spallation sources, liquid metal technology, radioisotope production and actinide physics and chemistry. The third goal of the TWG was to identify possible synergies within the scientific community, and to indicate potential spin-offs, showing how competence can be maintained and developed in the field of nuclear energy research.

In the transmutation scenario two options were considered; the waste being either recycled and transmuted in available conventional critical reactors (homogenous fuel recycle option, with no separation of plutonium and minor actinides), or in dedicated burner reactors. In the double strata fuel cycle option, plutonium is kept separated from the minor actinides and 5 to 20% of dedicated burner reactors in the reactor "park" would be required. If plutonium and the minor actinides are kept together, the fraction of dedicated burner reactors would be approximately 20%.

The report identified several reasons why ADS should be seriously considered for future transmuters. An ADS provides more control and flexibility in the design and operation of a subcritical reactor driven by an external neutron source than is achievable for a critical reactor. The external neutron source for the subcritical reactor implies that almost any fuel composition can be used in the system. This is particularly valuable when the reactor shall be used to transmute large amounts of minor actinides (MAs). The report indicates that ADS has good potential for waste transmutation and for reducing the amounts of waste and thereby reducing the requirements on deep geological repositories. Assuming a separation efficiency of 99.9% of the long-lived transuranium isotopes from the waste, followed by their complete transmutation in a dedicated burner reactor, the radiotoxicity may reach the levels of rich natural uranium ores in about 500–700 years.

ADS cannot only burn large quantities of minor actinides per unit (in contrast to critical reactors), but also generate heat and electricity in doing so. In addition, schemes have been proposed, in which also some of the long-lived fission products can be destroyed.

The report emphasized that there are many safety issues, which are common to both critical and subcritical reactors, e.g. appropriate cooling during normal operation or decay heat removal.

The report presented a scenario for development and deployment of ADS in Europe. It is pointed out that the introduction of accelerator-driven systems requires three steps of development before deployment on industrial scale:

- A comprehensive mid- and long term R&D program, to develop the single elements and components of the system. This includes development of new fuels and fuel cycle systems as well as suitable materials for the various parts of the system.
- Planning, design, construction and operation of an experimental accelerator-driven system for the demonstration of the concept.

- Planning, design, construction and operation of a large size prototype accelerator-driven system.

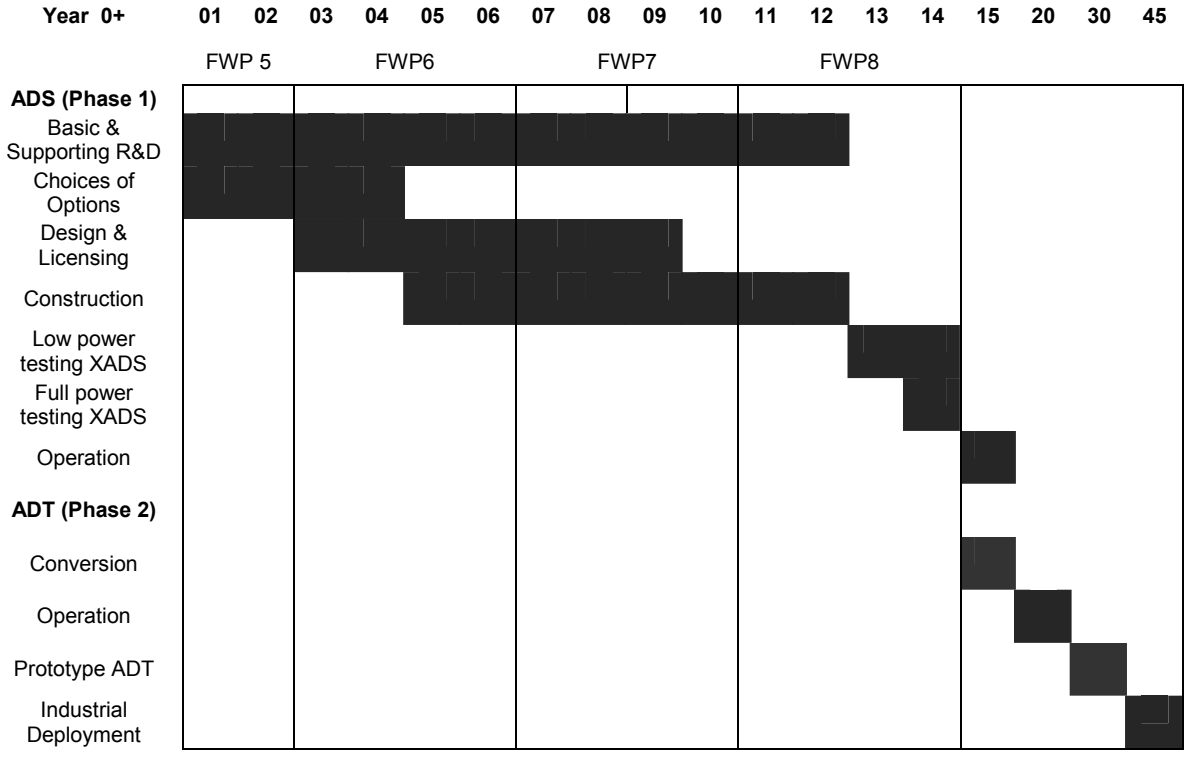
Following a first phase of R&D focused on the understanding of the basic principles of ADS, the European programmes should be streamlined and focused on a practical demonstration of the key issues. These demonstrations should cover high intensity proton accelerators (beam currents in the range 1–20 mA), spallation targets of high power (in excess of 1 MW), and their effective coupling with a subcritical core. In the field of fuels and materials, the realisation of representative minor actinide based fuels and targets, the assessment of their physical-chemical properties and behaviour under irradiation, together with the assessment of the related processing methods, become a priority for a credible waste transmutation programme. A proposed time schedule for the development of ADS technology in Europe is reproduced in Table 2-1.

It was suggested that during the first few years (2–3 years), a broad systems analysis should be performed on two concepts under consideration: the liquid Pb-Bi cooled system and the He cooled system. After a decision on the most suitable concept, to be made prior to 2004, a detailed design of the ADS could be started. For the first five to seven years, the R&D shall concentrate on:

a) The development of high intensity accelerators and megawatt spallation sources, and their integration with a subcritical reactor into a unified facility.
b) The development of advanced fuel fabrication and reprocessing technology.

Start of construction of an experimental ADS could be 2008 and start of operation could be 2013 according to the proposed plan.

Table 2-1. Time schedule for the development of an experimental ADS and accelerator-driven transmutation (ADT) technology in Europe.



The fuel to be used in the first phase of operation will be conventional mixed oxide fuel. Use of existing fuel from SNR-300[34] or Superphenix[35] could be envisaged. Innovative and dedicated minor actinide fuel will be tested in the accelerator-driven system and will replace the mixed oxide fuel in a second phase of operation (XADT). This mode of operation is envisaged for 2025. Around 2030 construction of a prototype could be started. This prototype has to have all features of the ADS to be deployed at a later stage (power, coolant, fuel etc). After successful operation of the prototype, the system could be deployed on an industrial scale starting around 2040.

The TWG also prepared estimates of costs for the suggested development and deployment scenario. The first stage (5th FWP) was estimated to M€ 50 over 3 years, with 50% being funded by EC not including the costs of the development of high-power accelerators, which are not funded by the EU.

The estimated costs for the development of an experimental accelerator driven system are summarized in Table 2-2. The total costs covering R&D, engineering design, construction, and fuel is estimated to M€ 980 over a twelve year period until 2012.

Table 2-2. Estimated costs (M€) for the development of a 100 MW_{th} accelerator-driven system.

Year 2000+	1	2	3	4	5	6	7	8	9	10	11	12	Total
	5th FWP		6th FWP				7th FWP						
Basic &Support R&D	30		90				70				10		200
Engineering Design	5		75				60				10		150
Construction	0		80				300				70		450
Fuel	0		10				120				50		180
Total	**35**		**255**				**550**				**140**		**980**
R&D for dedicated fuel	5		70				70				35		180*

* Estimated to 2012 for development of dedicated fuel and fuel processing.

2.4 EU – system studies 1998–2003

The fifth Framework Programme of the European Atomic Energy Community (EURATOM) has had two specific programmes on nuclear energy, one for indirect research and training actions managed by the Research Directorate General (DG RTD) and the other for direct actions under the responsibility of the Joint Research Centre (JRC) of the European Commission (EC). The strategic goal of the first one, "Research and training programme in the field of nuclear energy" has been to help exploit the full potential of nuclear energy in a sustainable manner, by making current technologies even safer and more economical and by exploring promising new concepts. This programme included a key action on nuclear fission (aside of activities in thermonuclear fusion), research and technological development (RTD) activities of a generic nature on radiological sciences, support for research infrastructure, training and accompanying measures. The key action on nuclear fission and the RTD activities of a generic nature

[34] SNR-300 = German liquid sodium cooled fast reactor at 300 MW_e built in the 1980ies but never taken in operation.
[35] Superphenix = French liquid sodium cooled fast reactor at 1200 MW_e operating from the late 1980ies for a about 10 years but closed after repeated technical problems.

are being implemented through indirect actions, i.e. research co-sponsored and co-ordinated by DG RTD, but carried out by external public and private organisations as multi-partner projects. The total budget available for these indirect actions during FWP5 has been M€ 193 /Bhatnagar 02/.

Partitioning and Transmutation (P&T) of long-lived radionuclides in nuclear waste has been one of the most important research areas of the EURATOM fifth (1998–2002) as well as it is for the sixth (2002–2006) Framework Programmes (FWP). The main objective of this research work is to provide a basis for evaluating the practicability, on an industrial scale, of P&T for reducing the amount of long lived radionuclides to be disposed of.

The emphasis in the recent EU-studies of transmutation has been on accelerator-driven systems (ADS). In FWP5, there have been 13 projects in the area of P&T with a total budget of about M€ 69 of which the EU contribution was about M€ 29. A network ADOPT has co-ordinated the activities on an experimental ADS design project with those of four clusters, one cluster on chemical separation

- Partitioning.

and three clusters on transmutation:

- Basic studies (BASTRA).
- Technological studies (TESTRA).
- Fuel studies (FEUTRA).

Each of these clusters has been formed by 3–4 projects – see Figure 2-1.

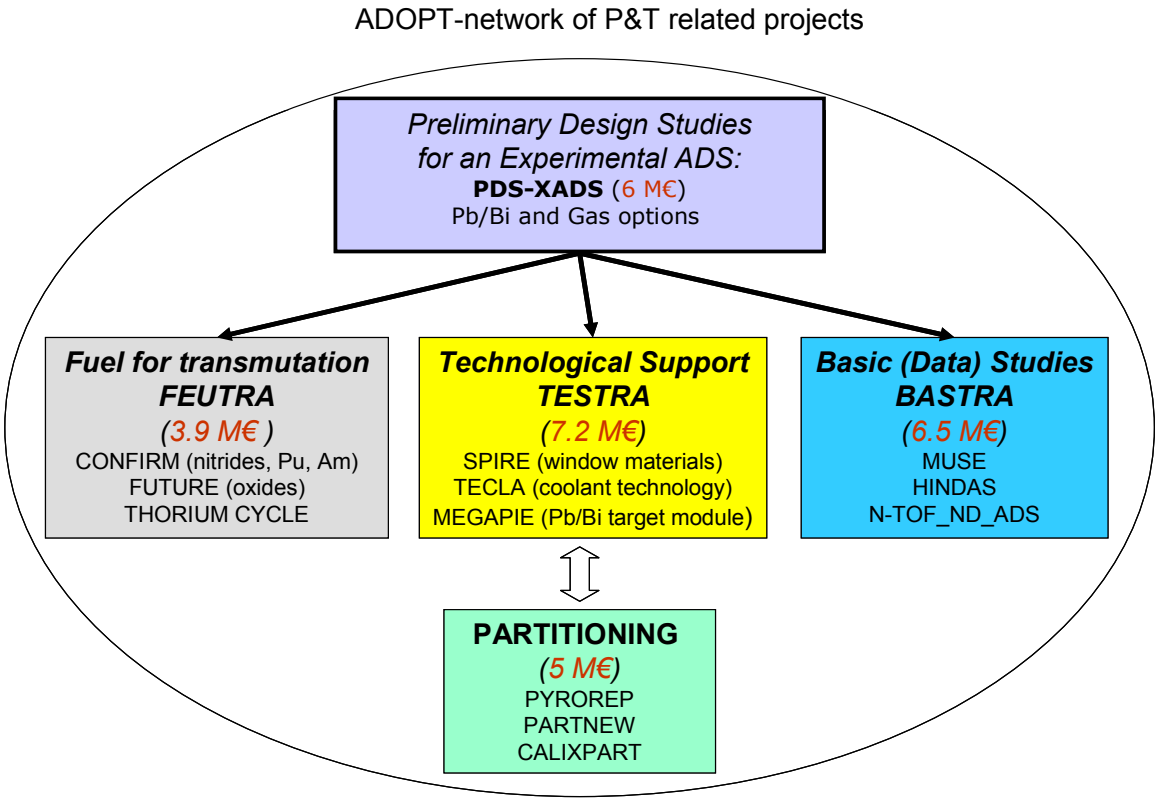


Figure 2-1. *ADOPT-network unifying a P&T cluster project structure in the 5th FWP of EU. Numbers in red indicate funding from the EC.*

The structure of EU's projects in the FWP5 can be presented in the following way:

2.4.1 ADOPT network

The ADOPT (Advanced Options for P&T) network consists of 16 partners co-ordinated by SCK·CEN from Belgium and funded with k€ 400 from the European commission. The objectives of the network (see Figure 2-1) have been to:

(i) Formulate actions with a view to promote consistency between FWP5 funded projects and national programmes.
(ii) Review overall results of the FWP5 projects.
(iii) Identify gaps in the overall programme of P&T research in Europe.
(iv) Provide input to future research proposals and guidelines for R&D orientation.
(v) Maintain relations with international organisations and countries outside the EU involved in P&T and ADS development.

ADOPT has rather unwillingly converted itself into a proposal for an integrated, ADS-transmutation directed project within FWP 6 – EUROTRANS.

2.4.2 Design studies of an experimental ADS

A critical first step for demonstrating the practicability of a transmuter is to demonstrate operation of an ADS with an accelerator coupled to the neutron spallation target located in the centre of the subcritical multiplying core. For this purpose an experimental plant – XADS – has been foreseen. The aim of the PDS-XADS[36] project, with 25 partners and co-ordinated by the French company Framatome, has been well documented studies with supporting evidence to choose and adopt the most promising technical concepts for XADS. The PDS-XADS has also:

- Addressed the critical points of the entire system.
- Identified the research and development (R&D) required in support.
- Defined the safety and licensing issues.
- Assessed the preliminary cost of the installation.
- Consolidated planning of the XADS development.

The assessment and comparison studies of the different conceptual designs of the main systems (accelerator, spallation target, subcritical core, primary system) will allow identification of the best solution, which could be studied in detail during the next phase of the design activities. The studies were focused on three principal ADS – systems:

- Pb/Bi cooled ADS "demonstrator" (~ 80 MW_{th}) – called Ansaldo-design.
- Gas cooled ADS – called Framatome design.
- Small scale Pb/Bi cooled multipurpose ADS ~ 20–30 MW_{th} – called MYRRHA-design.

The PDS-XADS project is still running and one of its final products is a list of resolved and unresolved key technical issues important for an engineering design of XADS. Of the three investigated concepts the liquid Pb-Bi cooled ADS was given the largest attention and a lot of work has been done to assess that concept. The conclusions are that there are no visible show-stoppers for development of any of the investigated

[36] PDS-XADS = preliminary design study of XADS.

concepts. However, it is concluded that a Pb-Bi cooled ADS will have less nuclear safety problems to be resolved than a gas cooled ADS. The final conclusions, however, will not be available until the final report of PDS-XADS project is published.

An Integrated Project proposal for FWP 6 tries to unify transmutation activities within EU in one single project – EUROTRANS – planned to start in 2005.

2.5 Summary of US developments following the roadmap

After the roadmap report /DOE 99/ the US DOE established a national Advanced Accelerator Application (AAA or 3A) programme which merged two different research programmes in USA: Accelerator-driven Transmutation of nuclear Waste (ATW) and Accelerator Production of Tritium (APT). ATW was focused on the accelerator applications for the commercial nuclear waste transmutation, while APT was developing accelerator technology for tritium production, a material needed for the nuclear weapons inventory.

The AAA program was rather short-lived and within a couple of years it was replaced by the Advanced Nuclear Fuel Cycle Initiative /DOE 03; Herczeg 03/ (see Figure 2-2). The Advanced Fuel Cycle Initiative (AFCI) was initiated by a common action of the directors of the US national laboratories: Argonne National Laboratory (ANL), Los Alamos National Laboratory (LANL), Brookhaven National Laboratory (BNL), Oak Ridge National Laboratory (ORNL), Sandia National Laboratory (SNL), Idaho National Engineering and Environmental Laboratory (INEEL), Idaho Accelerator Centre (IAC), and the Savannah River Site (SRS). It was announced in 2003 out of the following concerns:

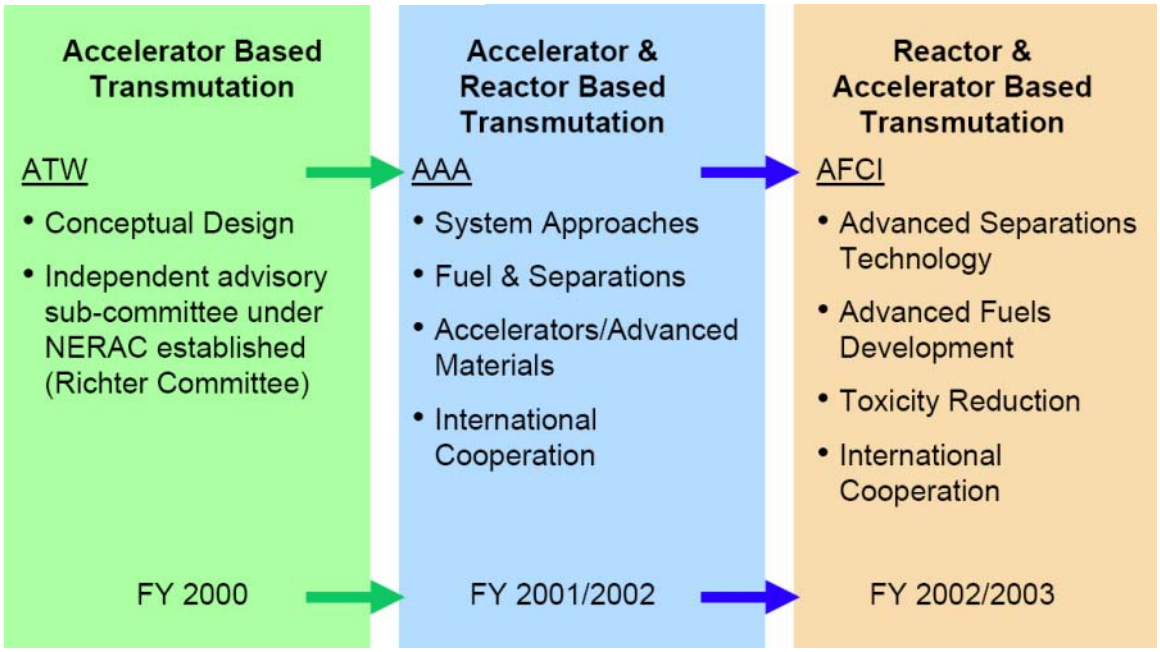


Figure 2-2. *Evolution of the US transmutation programme from ATW to AFCI /Herczeg 03/.*

1) Rapidly eroding US capabilities to sustain and develop nuclear fuel cycle technologies.
2) An advanced fuel cycle effort being an important component in reinvigorating nuclear energy in USA aside of Yucca Mountain waste repository and new commercial reactor initiatives.
3) Long term collaboration in nuclear technology with Russia addressing challenging topics of mutual concern such as:
    - consider technical alternatives to the use of MOX in materials disposition including consideration of a multipart plutonium management approach that develops options for cooperation on long-term proliferation-resistant technology to prevent formation of and/or eliminate plutonium,
    - engage Russian scientists and engineers in peaceful nuclear activities,
    - provide insight into Russian nuclear policies and programs and
    - make relevant Russian expertise and facilities available to the US.
4) Demonstration of US seriousness regarding nuclear energy and fuel cycle.

AFCI is developing the technology base for waste transmutation and should demonstrate its practicality and value for long-term waste management. AFCI's transmutation technology will also explore the potential to extract energy from nuclear waste and make it available to the national power grid.

An important part of the AFCI's mission is to revitalize declining US nuclear infrastructure. The AFC Initiative includes plans to provide capabilities for demonstration of waste transmutation and advanced nuclear technologies such as those for Generation IV reactors. The programme will establish and support a national university program to reenergize development and training in nuclear engineering and related fields, and develop research partnerships to rebuild a declining national nuclear science technology base.

The Advanced Fuel Cycle Initiative Programme includes a team of national laboratories, industrial partners and universities. Participating laboratories are mentioned above. Industry partners include Burns & Roe Enterprises, Inc (BREI) and General Atomics (GA). University Partners include University of Nevada Las Vegas, University of Michigan, University of California Berkeley, and University of Texas.

While pursuing the major programmatic areas of transmutation, the AFCI Programme is to revitalize the US nuclear science and engineering infrastructure in a very broad frame. The new AFCI nuclear facilities will be capable to perform many tasks in nuclear technology, including medical isotope production, special purpose isotope production, Generation IV reactor experiments, experimental loops to test different types of reactor coolants, fuels and materials, and access for special purpose experiments by university students, industry, and international collaborators.

The AFCI is divided in three phases with the following objectives:

- Phase I − Basic Technology Evaluation, comprising the results obtained in a frame of ATW and AAA programmes.
- Phase II − Proof of Principle: two parallel paths − 5–6 Years R&D − to provide information for decision makers.
- Phase III − Proof of Performance in 15–20 years.

Results of the Phase I can be shortly described as:

- The system studies have significantly narrowed the technical options.
- Development of the proliferation resistant technologies:
  - Separations − UREX demonstration of uranium separation at 99.999% purity.
  - Nitride and metal fuels fabricated for irradiation testing.
- International cooperation has provided US with approx. M$ 100 in research and experimental data:
  - In multinational collaboration.
    - EU – MEGAPIE project.
    - ISTC Pb-Bi target and technology development.
  - Bilateral collaboration with CEA.
- Revitalization of nuclear research.
  - 20 fellowships awarded to MS students working in areas related to transmutation (AAA initiative).
  - Many Ph D fellowships added to AFCI programme.
  - University of Nevada – Las Vegas (UNLV) has established major research programme to support transmutation R&D and generate new scientists/engineers.

Phase II is planned for about 5–6 years of R&D and the main objective is to provide a basis for decisions on "proof of principle phase". Two parallel paths are envisaged:

- Path 1: Intermediate term (up to 2015), using current reactor technology management.
  - Reducing high-level waste volumes.
  - Optimizing economics and performance of the planned geological repository.
  - Reducing the technical need for a second repository.
  - Reducing long-term inventories of plutonium in spent fuel.
  - Enabling the proliferation-resistant recovery of the energy contained in spent fuel.
- Path 2: Long term (about 2030) – using fast reactor technology including accelerator driven systems.
  - Reducing the toxicity of spent nuclear fuel.
  - Reducing the long-term heat generation of spent nuclear fuel.
  - Providing a sustainable fuel source for nuclear energy.
  - Supporting the future operation of Generation IV nuclear energy systems.

Phase III – Proof of Performance, is planned for about 15–20 years and will deliver results for both paths.

For the path 1:

- Final design, licensing and operation of a commercial spent fuel treatment facility.
- Evaluation of the LWR Lead-Test-Assembly experiment.
- Final design, licensing, and operation of a commercial (LWR/ALWR) fuel fabrication facility.

For the path 2:

- By the year 2007 decision is to be taken on final transmutation technology and determination of the testing programme – fast reactor, ADS, or both.
- Engineering scale demonstration of advanced pyroprocessing.
- Design and operation of the demonstration fuel fabrication Facility.

It is assumed that an international collaboration will have a significant off-set on the costs.

The final outcome of the successful AFCI programs is formulated as:

- Elimination of the technical needs for a second geological repository.
- Confirmation of advanced fuel cycle designs required for successful deployment of Generation IV nuclear energy systems.

Potential cost savings resulting from AFCI are estimated to be about G$ 35–50, coming mainly from optimization of the geological repository needs and costs.

Recently, the AFCI program together with the Generation IV initiative became parts of a broader program, the Nuclear Hydrogen Initiative (NHI), to develop hydrogen production systems and coupling them to nuclear heat sources. The primary focus of the hydrogen production is in the Next Generation Nuclear Plant (NGNP), a very high temperature gas-cooled thermal reactor.

2.6 NEA – ADS and FR in advanced nuclear fuel cycles – 2002

2.6.1 Introduction

In 1999 the nuclear development committee of OECD/NEA solicited another expert group for further study of P&T and advanced fuel cycles. The scope was mainly the clarification of the roles and merits of the fast reactor and the fast-spectrum accelerator-driven system (fast ADS) with regard to their application as actinide and fission product burners. The study included as well the assessment of the development status of the ADS with emphasis on reactor technology and safety, fuel cycle technology, trends in electricity cost, and general feasibility. The study was published in 2002 /NEA 02b; Wydler 01; Byung-Chan 02; Bertel 02/. By concentrating on transmutation strategies with fully closed fuel cycles, i.e. the particularly effective transmutation strategies, the study complements the P&T status and assessment study published in 1999. The essential differences between the varieties of proposals for implementing such strategies are evaluated with the help of a set of seven representative “fuel cycle schemes”, which are analysed in a consistent manner using reactor and fuel cycle parameters agreed by the expert group. The schemes are shown in Figure 2-3.

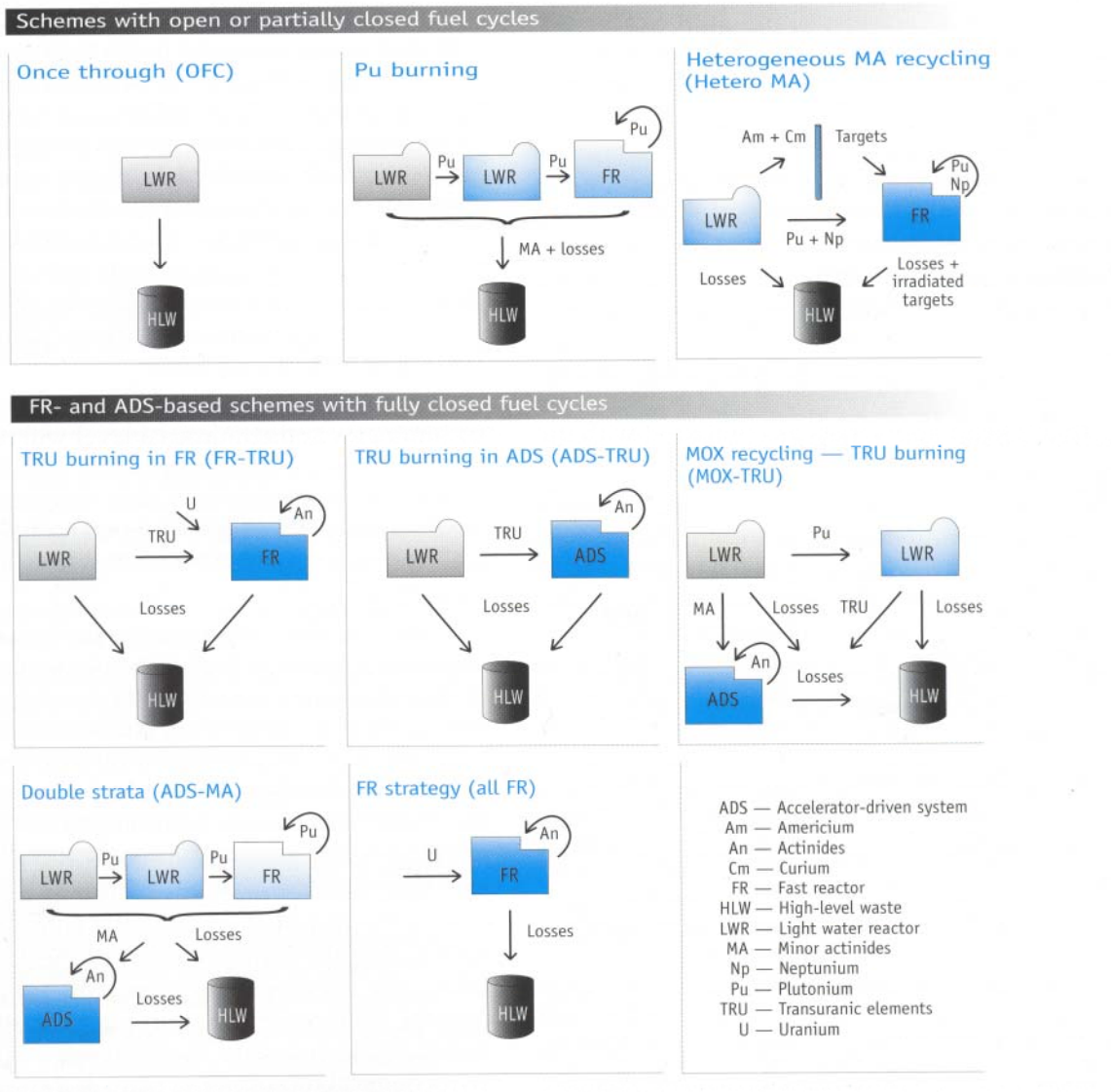


Figure 2-3. *Overview of the fuel cycles analysed in the report. (Reproduced from NEA News 2002 – No 20.2 p 21.)*

2.6.2 General comments and conclusions

The concluding chapter of the report from this study starts with some general comments concerning the development of advanced fuel cycles and P&T[37]:

The principle of sustainable development requires the fuel cycle of future nuclear energy systems to be closed for plutonium as well as minor actinides to ensure the production of fission energy with limited amounts of natural resources (i.e. uranium) and long-lived radioactive waste. It also requires a safe and cost-effective nuclear energy production. The resource efficiency and waste reduction goals together can ultimately only be reached by the introduction of advanced reactor systems with a significant fraction of fast reactors. For well-known reasons, however, a massive

[37] The following quotation uses some abbreviations frequently: ADS = accelerator-driven system; FR = fast reactor; MA = minor actinides; LWR = light water reactor; TRU = transuranium elements.

substitution of existing LWR-based, by such advanced, reactor and fuel cycle technology is not a realistic near-term scenario.

Partitioning and transmutation (P&T), which could address the high-level radioactive waste issue now and prepare the ground for a more resource-efficient nuclear energy system in the future, may become an attractive and appropriate intermediate strategy on the way to the ultimate goal of the sustainable nuclear energy system. In this context, the accelerator-driven system (ADS) can play an interesting role as a minor actinide or transuranics (TRU) burner. The interest in such burners is, of course, coupled with the P&T system and will diminish with an increase in the fraction of fast reactors in the park because, in a FR-dominated reactor park, dedicated burners will no longer play an essential role.

The general conclusions from the study are summarised in the following principal messages:

- While P&T will not replace the need for appropriate geological disposal of high-level waste, the study has confirmed that different transmutation strategies could significantly reduce, i.e. a hundred-fold, the long-term radiotoxicity of the waste and thus improve the environmental friendliness of the nuclear energy option. In that respect, P&T could contribute to a sustainable nuclear energy system.
- Very effective fuel cycle strategies, including both fast spectrum transmutation systems (FR and/or ADS) and multiple recycling with very low losses, would be required to achieve this objective.
- Multiple recycle technologies that manage Pu and MA either together or separately could achieve equivalent reduction factors in the radiotoxicity of wastes to be disposed. The study shows that pyrochemical reprocessing techniques are essential for those cycles employing ADS and FRs where very high MA-content fuels are used.
- In strategies where Pu and MA are managed separately, ADS can provide additional flexibility by enabling Pu-consumption in conventional reactors and minimising the fraction of dedicated fast reactors in the nuclear system.
- In strategies where Pu and MAs are managed together, the waste radiotoxicity reduction potential by use of FRs and ADS is similar and the system selection would need to be made based on economic, safety and other considerations.
- Further R&D on fuels, recycle, reactor and accelerator technologies would be needed to deploy P&T. The incorporation of transmutation systems would probably occur incrementally and differently according to national situations and policies.
- Fully closed fuel cycles may be achieved with a relatively limited increase in electricity cost of about 10 to 20%, compared with the LWR once-through fuel cycle.
- The deployment of these transmutation schemes need long lead-times for the development of the necessary technology as well as making these technologies more cost-effective.

2.6.3 Technical conclusions

The report from the NEA group lists the detailed conclusions for some important technical areas as follows:

Role of ADS in actinide transmutation strategies

- All transmutation strategies with closed fuel cycles could, in principle, achieve high reductions in the actinide inventory and the long-term radiotoxicity of the waste, and these are comparable with those of a pure fast reactor strategy. With respect to these reductions, the potentials of the FR and the ADS are very similar.
- Under the assumptions used in the study, these strategies can achieve a more than hundredfold reduction in the long-term waste radiotoxicity and even higher reductions in the heavy metal and TRU losses to repository, compared with the once-through fuel cycle.
- The reduction factors are primarily determined by the fuel burn-up and the reprocessing and fuel fabrication losses. An ambitious goal for the recovery of all actinides (99.9%, as already achieved for uranium and plutonium) must be set, if the quoted reduction factors are to be realised.
- Multiple recycling of plutonium without minor actinide transmutation is useful for the management of plutonium, but cannot qualify as a transmutation strategy because it reduces the long-term waste radiotoxicity by only a factor of about five[38].
- With regard to actinide waste production and technological aspects, the TRU burning in FR and the double strata strategies are similarly attractive. The former can gradually evolve to a pure fast reactor strategy, but requires high initial investments in fast reactor and advanced fuel cycle technology. The latter confines the minor actinides to a small part of the fuel cycle, but calls for particularly innovative technology for this part of the fuel cycle.
- The subcritical operation of an actinide burner with a fast neutron spectrum offers interesting additional parameters of freedom in the core design. In particular, the possibility of operating such a burner with a uranium-free (or thorium-free) fuel supply allows the fraction of specialised transmuters in the reactor park to be minimised.
- A further advantage of the subcritical operation mode is the tolerance of the system against degradations in the safety characteristics of the core. Both of these advantages are of particular relevance for systems which burn pure minor actinides, e.g. minor actinide burners in a double strata strategy.
- Transmutation systems with partially closed fuel cycles, e.g. systems in which minor actinides are separated from the fuel and recycled in special “target” pins, are technologically less demanding and do not require an ADS, but cannot achieve the high transmutation effectiveness of systems with fully closed fuel cycles.
- Physical limitations associated with the production and destruction of in-pile and out-of-pile fuel inventories imply very long time constants for the start-up and final shut-down phase of new fission-based nuclear technologies. This implies that

[38] Comment: Important factors in this context are the burnup and the interim storage time of the fuel before separation of the plutonium. By high burnup and long interim storage time the ^{241}Pu will decay to ^{241}Am and further to ^{237}Np. In particular the content of ^{241}Am is important for the waste radiotoxicity in the time frame of a few thousand years.

transmutation technology, with or without ADS, can fulfil its promises only if it is introduced with the intention of using it for at least a century.

- For a nuclear energy scenario with a finite time horizon, the full benefit from transmutation can be realised only if, in the shut-down phase, the TRU inventory is burnt and not put to waste. Due to the low power-specific heavy metal inventory of the respective burner, the TRU burning in ADS strategy features a lower steady-state TRU inventory and, in the shutdown phase, can burn this inventory more quickly than the other investigated strategies.

Fuel cycle technology

- Actinide transmutation implies the handling of unusual fuels with very high decay heats and neutron source strengths. A significant effort is required to investigate the manufacturability, burn-up behaviour and reprocessability of these fuels. This applies particularly to fuels with high minor actinide content, which can probably be reprocessed only with the help of pyrochemical methods[39]. These methods have to be further developed to tolerate from ten to more than twenty times higher decay heat levels than those encountered in the pyrochemical reprocessing of fast reactor fuels.
- The introduction of pyrochemical processing techniques at the industrial level will require the development of new process flow-sheets and the use of potentially very corrosive reagents in hostile environments. These processes will generate chemical and radiological hazards which will have to be mitigated.
- The PUREX aqueous reprocessing can be considered as valid for the FR-MOX fuel in the plutonium-burning and double strata schemes. Reprocessing of this fuel within short cooling times and with the required high recovery yields, however, will require the plutonium dissolution yield to be improved and the PUREX flow-sheet to be modified.
- Due to the high radioactivity of FR-MOX fuel, its handling will require measures to be taken to reduce the radiation doses in the fabrication plant and during the transportation of the fuel assemblies. The increased requirements for shielding, and preference for short transportation paths, of multiple recycled fuels also favour the pyrochemical reprocessing method.
- All transmutation strategies which include LWRs in the reactor mix produce large streams of depleted and irradiated uranium. If this uranium is not considered as a resource for future fast reactors, its long-term radiological impact has also to be taken into account[40].

ADS technology and safety

- For all ADS-based transmutation strategies, important technological challenges exist with regard to the accelerator, the target, the subcritical reactor and new types of safety issues.
- On the whole, the development of accelerators is well-advanced, and beam powers of up to 10 MW for cyclotrons and 100 MW for linacs now appear to be feasible.

[39] Comment: This implies that dilution with other fuel, e.g from LWR, is not a feasible option.
[40] Comment: By the same argument − depleted uranium is obtained from enrichment of uranium (independent of P&T) and must also be handled in a responsible way.

However, further development is required with respect to the beam losses and especially the beam trips to avoid fast temperature and mechanical stress transients in the reactor.

- Various problems related to the accelerator-reactor coupling have still to be investigated. Thereby, special attention has to be given to the target and especially the beam window, as these components are subjected to complex stress, corrosion and irradiation conditions which are not encountered in normal reactors.
- While the reactor physics of subcritical systems is well-understood, the issues regarding the dynamic response to reactivity and source transients require further investigation because they are the area of greatest difference between critical and subcritical systems.
- The presence of an external neutron source which can vary very rapidly, in combination with very weak reactivity feedbacks, especially from the Doppler effect, implies fast and (depending on the subcriticality level) violent responses to control actions which puts additional demands on the control actuators, the fuel behaviour, and the heat removal processes. In particular, the fuel should be capable of adiabatic heat storage to buffer any sharp changes.
- If hypothetical core disruptive accidents have to be taken into account in the safety analysis of an ADS, a prompt negative feedback mechanism for quenching such accidents has to be developed.

Cost of actinide transmutation

- Fully closed fuel cycles may be achieved with a relatively limited increase in electricity cost of about 10 to 20%, compared with the LWR once-through fuel cycle. In case of partially closed fuel cycles, e.g. only closed for plutonium, the cost increase is about 7%.
- Among the fully closed fuel cycle strategies investigated in the present study, TRU burning in FR and the double strata strategy feature the lowest increases in system-wide electricity cost relative to the LWR once-through fuel cycle.
- Fuel cycle strategies which involve the use of ADS-technology show an overall economic benefit by burning as much plutonium as possible in less expensive, more conventional systems, i.e. MOX-LWRs and MOX-FRs.
- Especially the TRU burning in ADS strategy would benefit from lower accelerator beam costs. Reducing the accelerator costs by a factor of three would halve the electricity cost increase for this strategy.
- The economic incentive to increase the burn-up fraction in the minor actinide and TRU burners beyond a value of 15% becomes marginal. Further reductions in fuel losses to repository at an acceptable system-wide energy costs, therefore, are to be obtained preferentially by increasing the fuel recovery rate.
- For the closed fuel cycle strategies, the advanced technology contribution to the system-wide electricity cost is in the range of 10 to 50%. If all non-LWR technology is considered as advanced, the advanced technology cost contribution for the closed fuel cycle strategies lies in the range of 30 to 50%.
- The cost analysis confirms the long-term potential of P&T for reducing the transuranics inventory and the radiotoxicity of the waste with a rather limited

increase in the electricity generation cost despite that this cost increase may not be acceptable in today's market environment. It also means that transmutation may become affordable with only a limited cost increase if a reduction in the actinide waste radiotoxicity becomes important for the society.

Fission product transmutation

- Excess neutrons produced by critical and subcritical burners can, in principle, be utilised to transmute fission products. With the neutron fluxes available in these systems, it is theoretically possible to transmute the long-lived fission products; the transmutation of the more abundant short-lived fission products, however, is impracticable due to insufficient transmutation rates. This means that transmutation, in principle, allows the mitigation of the long-term risk from fission products in a geologic repository, but cannot significantly reduce the heat generation and mass of the disposed fission products.
- Maximising the supplier-to-burner reactor support ratio can result in an insufficient neutronic potential for transmuting the long-lived fission products of the entire reactor park. If the transmutation would be limited to ^{129}I and ^{99}Tc, all TRU burning strategies could, theoretically, accomplish the task.
- In practice, the necessity of isotopic separations and difficulties in the preparation of targets present difficult obstacles for the fission product transmutation, which currently reduce the number of candidate nuclides to only one or two, i.e. ^{99}Tc and, possibly, ^{129}I. So far, the feasibility has been established only for ^{99}Tc. This means that, for the remaining long-lived fission products, partitioning followed by immobilisation in an especially stable matrix may remain the only realistic method for reducing their radiological impact.

R&D needs

- Experimentation on fuels is a priority. No concept can be considered seriously if the appropriate fuels are not defined and proven, i.e. characterised, fabricated, irradiated and reprocessed.
  - Since fuels play a central role in all scenarios of waste minimisation and nuclear power development, an international share of efforts around nitrides, oxides and metals should be organised in order to ensure an optimum use of resources in the few existing laboratories which can handle very active fuels.
  - In this context, the availability of irradiation facilities, in particular fast neutron facilities which can produce high damage rates in the specimens, is a key issue and major concern. Again, an international initiative could be envisaged to harmonise programmes and to allow the best use of existing resources to be made. Identification of the experimental irradiation needs in such a shared international fast-spectrum facility would be a worthwhile undertaking.
- Demonstration at appropriate scale of the performance of pyrochemical processes (level of losses, secondary waste, etc) is needed in order to assess in more detail the technical-economic viability of certain fuel cycle options.
- In the field of basic R&D supporting FRs as well as ADS, the discussion around the coolants for fast-spectrum systems would benefit from a better international agreement on pro and cons of the different options.

- Improved modelling tools to simulate the materials behaviour under (mixed) irradiation conditions (and possibly high temperatures) may prove to be a very valuable approach and a sharing of expertise and benchmarking within an international context may be advocated.
- Safety analysis of ADS should identify the possible paths to exclude hypothetical core disruptive accidents (HCDA) in ADS. If such a HCDA has to be taken into account in the safety analysis of an ADS, a prompt negative feedback mechanism for quenching such an accident has to be developed.
- In addition to this R&D, countries embarking on an ADS-based closed fuel cycle strategy should envisage a demonstration experiment, which allows the ADS concept to be validated from operation and safety viewpoints.
- And last but not least, Performance assessment studies for a geological disposal site using a P&T source term are necessary in order to seek clarification of the cost/benefit analysis of such advanced fuel cycles, including geological disposal.

2.7 System studies in other countries 1998–2003

2.7.1 Japan

In Japan /Takano 02; Takano03a/ the objective of P&T is oriented to removal and transmutation of mainly minor actinides and long-lived fission products. Plutonium is to be used as a nuclear fuel material under the Japanese nuclear energy policy.

Most of the research efforts related to partitioning and transmutation in Japan are concentrated at Japan Atomic Energy Research Institute (JAERI). JAERI, together with JNC[41] and CRIEPI[42], have since many years been carrying out partitioning and transmutation studies under the OMEGA program (Options Making Extra Gains of Actinides and Fission Products). The progress of the OMEGA program was reviewed in 1999 by the Atomic Energy Commission's Advisory Committee on Nuclear Fuel Cycle Back-End Policy and the report was issued in March 2000. The review emphasised the necessity of such R&D works as future system design and the development of the implementation scenario of P&T, basic experiments to demonstrate the feasibility of the processes, and engineering scale experiments in order to obtain safety data of these systems.

OMEGA program

The R&D areas (relevant for P&T) covered by the OMEGA Program are as follows[43]:

- Physical and chemical properties of MA and fission products.
- Partitioning of radioactive elements from HLW of reprocessing process.
- Transmutation: nuclear and fuel property data of MA, system design studies, reactor fuel.
- Accelerator target development, development of high power accelerator for transmutation.

[41] JNC = Japan Nuclear Cycle Development Institute
[42] CRIEPI = Central Research Institute of the Electric Power Industries, Japan.
[43] Comment: One of the original OMEGA goals was the recovery/recycle of valuable elements from the irradiated fuel.

JAERI, being the main institute in Japan to carry out the development of partitioning processes, nitride fuel technology, and some basic studies to support the ADS development has the following R&D program:

- In the subcritical core design and technology of an 800 MW_{th} ADS, the cross-flow between spallation target and ductless fuel assemblies is being studied by thermal-hydraulic and structural analysis.
- In neutronic design studies, efforts are continued to reduce radial power peaking and burnup reactivity swing, and to evaluate an adequate subcritical level.
- In the lead-bismuth eutectic technology, loop experiments are being started to obtain material corrosion and erosion data.
- In the superconducting linear accelerator technology a prototype cryomodule is being built to examine its performance.

The cost and benefits of P&T technology development are studied on the following possibilities:

(1) The long-term radiotoxicity of high-level waste (HLW) is reduced by a factor of two-hundred and the inventories of minor actinides are reduced by a factor of one hundred which gives significant reduction of the volume of required repository.
(2) Sr and Cs separated from HLW are intermediately stored and processed, reducing very significantly the heat load at the waste storage.
(3) The transmutation of Pu reduces significantly long term nuclear proliferation concerns.
(4) P&T technology is estimated to add a relatively modest 5–10% to the electricity cost, though this value depends on the cost estimates of geological disposal and fuel cycle (costs of double strata fuel cycle are used here as a reference level).

Basic studies to support the ADS R&D for P&T technology in JAERI have been carried out on the basis of the double-strata fuel cycle concept as shown in Figure 2-4. As transmuter of nuclear waste JAERI has proposed an 800 MW_{th}, liquid lead-bismuth cooled ADS.

JAERI continues to carry out the development of partitioning processes, nitride fuel technology, and some basic studies to support the ADS development. Recently, more and more university and industry groups have got involved in R&D activities on ADS, not only from the nuclear energy community but also from nuclear physics and accelerator communities.

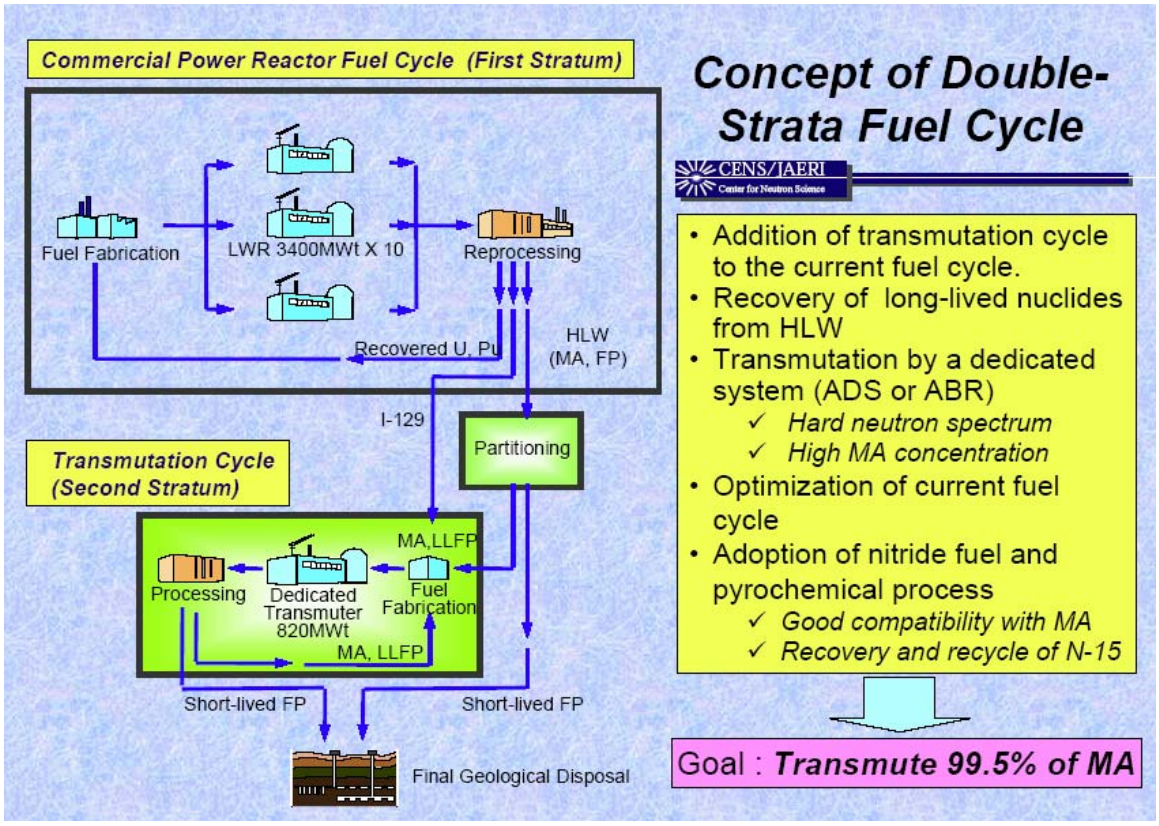


Figure 2-4. *Concept of the double-strata fuel cycle based on ADS.*

Core design of 800 MW_{th} ADS

A core design study of an 800 MW_{th} ADS – see Figure 2-5 /Oigawa 04/ – was performed to find a preferable peaking factor and burnup reactivity swing. In the JAERI reference design core, ductless type fuel assemblies are adopted to avoid a blockage accident by lead-bismuth coolant and to enhance the cooling capability of the decay heat.

Lead-bismuth eutectic technology and material development

JAERI selects lead-bismuth eutectic as the first candidate for the spallation source and core coolant materials in the ADS system, because it has some advantageous characteristics such as chemical stability, low melting and high boiling temperature, high neutron yield by spallation reactions, small neutron capture cross sections and less positive void reactivity than sodium. However, lead-bismuth is corrosive against structural materials as austenite stainless steel for higher temperature than 500°C and is erosive against materials in high flow speed, beyond 2 m/s.

Corrosion of structural materials

For target, materials and operation tests of a Pb-Bi system, a material test loop and stagnant test devices have been installed for Pb-Bi technology development, delivering important results in static and dynamic conditions. However, most of the tests were conducted without the oxygen control, and the next step of those experiments addresses various conditions with oxygen control and various Pb-Bi flow rates.

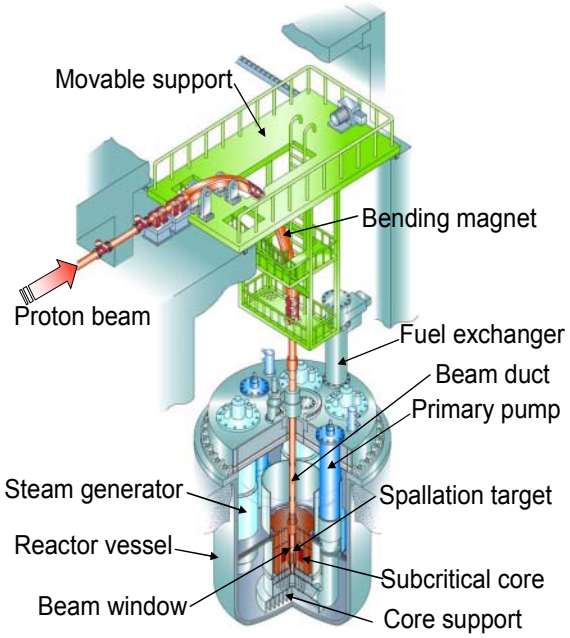


Figure 2-5. *Conceptual view of 800 MW_{th} LBE-cooled ADS designed by JAERI.*

Thermalhydraulics at beam window

One of the critical problems in the design of the ADS is the viability of the beam window. JAERI and MES (Mitsui Engineering and Shipbuilding Co Ltd) are building collaboratively a thermal-hydraulic loop to investigate the prediction accuracy of heat transfer coefficients of the hemispherical shape of the beam window.

Coordination of research

The annual research plans and schedules for the next five years for the research and development of the P&T technology in JAERI are coordinated among the relevant groups such as the Research Group for Advanced Fuel, the Research Group for Actinides Science, the Research Group for Separation Chemistry and the Nuclear Transmutation Group to draw the road map towards the demonstration of the P&T technology.

Transmutation experimental facility

In order to study the basic characteristics of the ADS and to demonstrate its feasibility from viewpoints of the reactor physics and the spallation target engineering, JAERI plans to build the Transmutation Experimental Facility (TEF) in the Tokai Research Establishment under a framework of the J-PARC project (Japan Proton Accelerator Research Complex) – see also Section 4.1.1. The construction of the TEF is scheduled to start around 2007 /Oigawa 04/.

TEF consists of two experimental facilities: the Transmutation Physics Experimental Facility (TEF-P) and the ADS Target Test Facility (TEF-T). The TEF-P is a zero-power critical/subcritical facility where a low power proton beam will be available to research on reactor physics and controllability of the ADS. The TEF-T is a material irradiation facility which can accept a maximum 200kW–600MeV proton beam into the spallation target of Pb-Bi eutectic.

The principal mission of TEF-T is to demonstrate the feasibility of the high-power spallation target system and to research the material compatibility in the LBE with irradiated environment. The irradiated structural material of the target vessel, as well as the irradiated material test pieces, will be examined from viewpoints of tensile strength, ductility, fatigue, fracture toughness etc. In addition to these tests, the effects of the corrosion and the erosion by LBE and the spallation products will be studied by changing the parameters such as the temperature, the irradiation period, the flow speed and the oxygen concentration in LBE. The experiences to be gathered at TEF-T will be valuable for learning how to operate and handle the high-power spallation target.

JAERI participates actively in the International MEGAPIE Project in Switzerland.

2.7.2 France

The French program on partitioning and transmutation has its background in the 1991 law where research on P&T of minor actinides is required to evaluate its merits as a complementary waste management strategy. The program employs a substantial number of scientists and engineers at the research institutes CEA and CNRS. French industry also takes on active interest with COGEMA and Framatome in the forefront.

The research program has many branches, but the main focus (concerning transmutation) is on

a) Development of novel fuel assemblies for multi-recycling of plutonium in existing PWRs.

b) Design of gas-cooled fast neutron reactors.

c) Fuels and targets for transmutation in fast neutron reactors and ADS.

Other activities are also pursued, though with less intensity, like R&D on the other types of generation IV reactors, materials research for ADS (MEGAPIE, TECLA and SPIRE projects) and the MUSE experiments on subcriticality control. In this summary, the three major research areas listed above will be shortly reviewed.

The French R&D-programme on waste management is closely followed by a national commission that regularly issues evaluation reports. In June 2003 the 9th such report was published. On P&T the commission states as follows /CNE 03/:

Research on advanced separation of minor actinides and caesium have resolutely entered the phase of technical feasibility demonstration. This stage started on solid scientific and organisational bases. However, keeping to the programme schedule before 2005 for the Atalante facility (Marcoule) will require close watch on the CEA's[44] part.

On the other hand, studies on transmutation are in a preliminary stage. They concern the simulation of transmutation systems and the assessment of the evolution under irradiation of the inventories listing the transmutable elements within the context of scenario studies. At the same time, three experimental activities are being carried out. The first one concerns the manufacturing and behaviour of targets made with samples of minor actinide compounds and long-lived fission products placed in an irradiation reactor such as Phenix[45]. The second one deals with the subsystems of an accelerator-

[44] CEA = Commissariat a l'Energie Atomique, France.
[45] Phenix = French fast sodium cooled reactor with 250 MW_{el} power, in operation since the mid-1970ies.

assisted subcritical system (linear accelerator, spallation target, subcritical core) which is considered as a tool enabling the massive transmutation of minor actinides, even certain long-lived fission products. Finally the third one deals with basic studies (materials, nuclear data)

To date the demonstration of transmutation technical feasibility is not completed, particularly when tackling the difficult issue of manufacturing and reprocessing of irradiation targets loaded with americium and curium. Some ceramic compounds of americium that are under study seem promising. Only preliminary results concerning irradiations in Phenix will be available before 2006 – and the Commission appreciates the fact that it is starting in 2003 for a first campaign at maximum power equivalent to 120 full power days. It is a first essential step, but the crucial step of the study on the behaviour of a pin and a complete assembly in a fast neutron reactor can only be truly envisaged with the development of a dedicated reactor (as Super-Phenix could have been). As for the subcritical systems, the effort led at a European level continues with the 6th Framework programme, and the new element of this year is the perspective of a European demonstrator from the MYRRHA project. There is a broad consensus for it on the part of the European scientific community, and the CEN·SCK, an organism in charge of nuclear research in Belgium, has agreed in principle.

Finally, studies on transmutation aim at first treating already produced waste that extend today to some future reactors – so called 4th generation. One of their objectives is to be able to self-cycle their own long-lived radionuclides, even to produce less, as it is the case in the thorium cycle. The national scientific community, within the shared research group Gédépeon, is presently re-orienting itself in that direction.

Fuel assemblies for multi-recycling of plutonium in PWRs

Standard MOX fuel assemblies cannot be used for multi-recycling of Pu in LWRs, since the presence of Pu-240 and Pu-242 suppresses the thermal neutron spectrum in these assemblies. Consequently, the void coefficient increases and boron/control rod worth decreases with increasing fraction of these nuclides. However, in the last five years, CEA has suggested novel PWR fuel designs that would enable multi-recycling of plutonium /NEA 03/. In the MIX concept, MOX fuel rods are fabricated with enriched uranium. Hence the magnitude of the thermal flux is preserved, and safety coefficients remain within acceptable values. The disadvantage of this approach is that the number of fuel pins fabricated subject to the same safety standards as MOX fuel would be several times higher, increasing the cost penalty. In the CORAIL concept /Youinou 99; Youinou 01/ 84 MOX pins are positioned in the periphery of an assembly with 180 standard UOX fuel pins in the centre – see Figure 2-6. Since the MOX rods are located within one to two thermal diffusion lengths of the UOX rods, the CORAIL assembly is more tolerant to the poor plutonium quality arising from multi-recycling. The CORAIL concept is now considered as the reference for Pu management in LWRs. Since it does not involve any innovative design of the fuel itself, it is a low cost option for Pu recycling that could be tested on industrial scale in the near term (next 5–10 years).

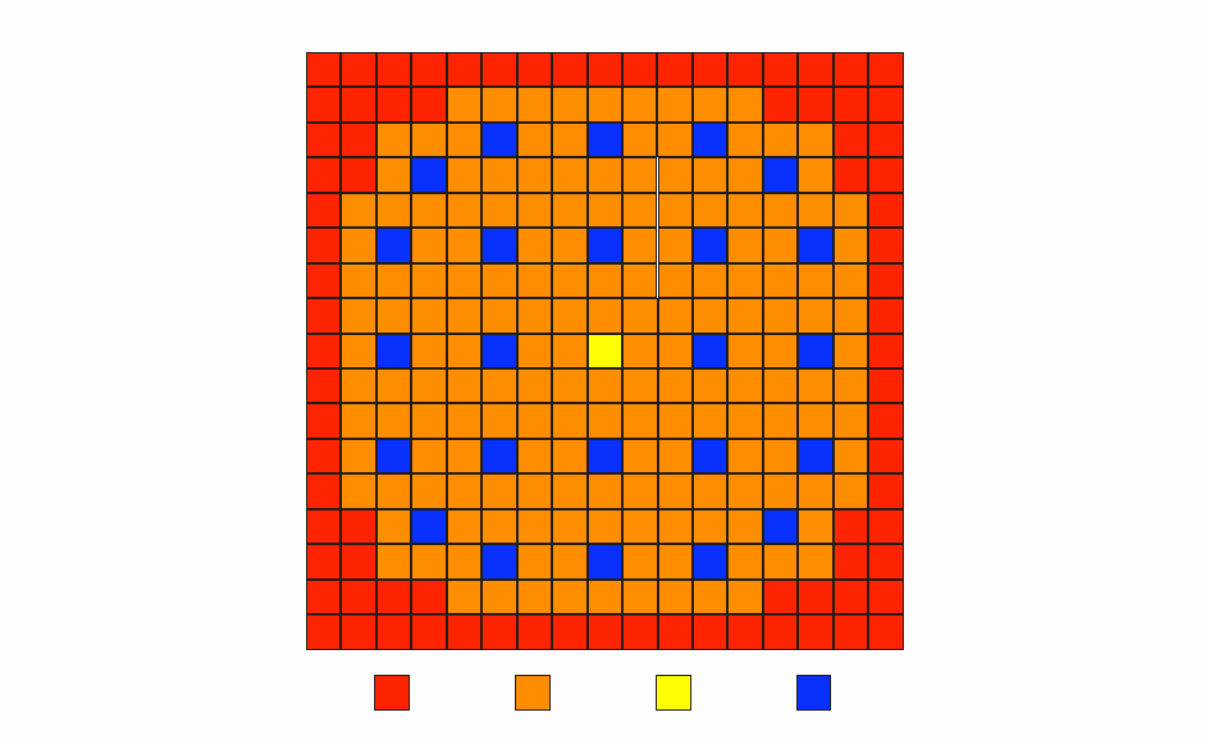


Figure 2-6. *CORAIL concept – 17*17 PWR fuel assembly with 84 peripheral Pu-rods (red) and 180 U-rods (orange). (Blue is control rod thimbles and yellow is instrument tube thimble.)*

Other, more innovative concepts are under investigation, like the inert matrix CERMET fuel for PWRs. In this case, the matrix would be zirconium metal. The plutonium oxide inclusions would have to be coated by a protective layer, chromium being under investigation as a potential coating material /Buffe 03/. An issue that would have to be solved in this case is the poor solubility of plutonium oxide in nitric acid.

Design of gas-cooled fast neutron reactors

The CEA management has redirected most of the research on advanced nuclear fission reactors towards gas-cooled fast neutron reactors. Among the advantages of this reactor type is a good chemical compatibility between the coolant (helium) and air, and a much smaller sensitivity of the coolant void coefficient to the presence of americium in the fuel. A larger loading of minor actinides in the fuel is thus possible, as compared to sodium-cooled reactors.

The main disadvantage of this system is the poor heat removal capability of the coolant, which requires operation under high pressure (40–80 atmospheres). A loss of coolant accident requires active shutdown systems to operate within a short grace time. The corresponding low mass rating of the fuel leads to very long residence times in order to achieve an appreciable burn-up. For example, recent design proposals provide not more than five percent burn-up during ten years of operation /Garnier 03/.

The high neutron leakage of the gas-cooled core also requires a fuel providing a better neutron economy, in order to achieve a breeding ratio in the proximity of unity. CEA is primarily investigating nitrides and carbides for this purpose, and a fabrication line for uranium-plutonium nitride is planned to be constructed at Cadarache in the near future /Chauvin 03/.

A possibility to improve the performance of the gas-cooled reactor would be the development of cladding materials with better high-temperature stability. Refractory materials like zirconium carbides are under consideration in this context.

Fuels and targets for transmutation in fast neutron reactors and ADS

As mentioned above, the emphasis in the French R&D program on fast reactors is on uranium-plutonium fuels. The transmutation of americium is foreseen to be made in so called target assemblies, where americium oxide is dispersed in an inert matrix. By introducing moderator pins into the target assembly, a high burn-up fraction is assumed to be possible. In the reference design, the inert matrix is magnesium oxide, and the moderator pins contain zirconium hydride /Pillon 02/. AmO_2-MgO target pins have been fabricated according to various methods (for details see Section 4.1.4), and the so called ECRIX irradiation has already started in Phenix. The advantages of this concept are that existing reactor types may be used for americium management, and that a high mass reduction of actinides can be achieved in a single irradiation (75–80%). The main disadvantage is that the irradiation in a thermalized spectrum leads to production of highly neutron active curium and californium isotopes. Hence recycling of such a target is not possible, and the toxicity reduction becomes rather modest. Typically, a factor of ten is achievable, compared to a factor of hundred in multi-recycling scenarios. In addition, the perturbation that the presence of moderators introduces into the driver core leads to penalties in terms of high power peaking factors.

CEA also participates in the development of fuels for ADS. $(Pu,Am)O_2$ powder and pellets have been fabricated within the FUTURE program, and irradiation of two fuel pins with $(Pu,Am)O_2$ dispersed in MgO is planned to take place in Phenix starting late 2006.

2.7.3 Belgium

The Belgian parliament endorsed in 2002 a law on the nuclear phase-out. Besides urgent needs of electricity supply, existing power plants are planned to be shut down after 40 years of operation and no licenses for new plants will be granted. The law, however, also says that this decision can be reconsidered if the electricity availability and independency are put into danger and if the decision is jeopardizing the obligation of Belgium under the Kyoto protocol to reduce its CO_2 production to 7.5% below the level during the reference year 1990. In a contemporary decision the Belgian government stated clearly that knowledge and competency in nuclear science should be kept alive in Belgium among others through the support to the Belgian Nuclear Research Centre – SCK·CEN. SCK·CEN has a key role in nuclear research in Belgium including a very active research in transmutation. Since 1998, SCK·CEN in partnership with IBA[46] and many European research laboratories, is designing a multipurpose ADS for R&D applications – MYRRHA facility – and is conducting an associated R&D support programme /Abderrahim 03; Abderrahim 04/. MYRRHA – mentioned a few times in this report also in European research context – is an Accelerator Driven System (ADS) under development at Mol in Belgium and aims to serve as a basis for the European experimental ADS to provide protons and neutrons for various R&D applications. MYRRHA consists of a proton accelerator, a liquid Pb-Bi spallation target and a Pb-Bi cooled, subcritical fast reactor. At the start of the project, the cyclotron technology was the first choice for the accelerator due to the proximity of IBA and its

[46] IBA = Ion Beam Applications SA.

expertise in the field. Since 2002, taking into account the developments achieved within the FWP5 PDS-XADS, the MYRRHA team has made of the linac option their first option but keeping the cyclotron in stand by. The capital investment for the cyclotron is a factor three lower compared to the linac. In a first stage, the project focuses mainly on demonstration of the ADS concept, safety research on subcritical systems and nuclear waste transmutation studies. At a later stage, the device will also be dedicated to research on structural materials, nuclear fuel, liquid metal technology and associated aspects and on subcritical reactor physics. The MYRRHA project shall demonstrate the ADS concept at a reasonable power level, of 50 MW_{th}, and should demonstrate the technological feasibility of MA and LLFP transmutation under realistic conditions. Subsequently, MYRRHA will be used for nuclear research on applications such as radioisotope production.

The MYRRHA project couples a proton accelerator with a liquid Pb-Bi windowless spallation target, surrounded by a Pb-Bi cooled subcritical reactor in a pool type configuration within a standing vessel (Figure 2-7) The spallation source is fully immersed in the reactor pool and interlinks with the core but its liquid metal contents is separated from the core coolant. This is a consequence of the windowless design presently favoured. The core pool contains a fast-spectrum subcritical core cooled with liquid Pb-Bi eutectic (LBE) and thermal islands regions located in in-pile sections (IPS) in the fast core. The core is fuelled with typical fast reactor fuel pins composed of MOX type fast reactor fuel (similar to Superphenix or BN600[47] fuel rods) with total Pu-contents limited at maximum to 30%. The device as shown in Figure 2-7 with the double-wall pool containment vessel is surrounded by a biological shield to limit the activation of the surrounding soil as the MYRRHA subcritical reactor will be installed in an underground pit.

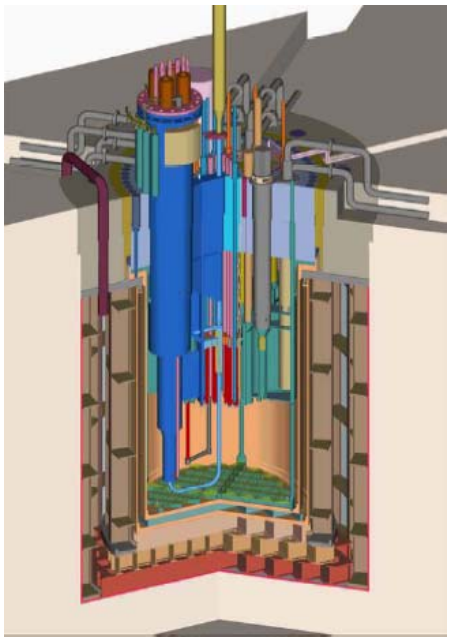


Figure 2-7. *MYRRHA primary vessel.*

[47] BN600 = Russian sodium cooled fast reactor at 600 MW_e.

An accelerator designed for MYRRHA is a high power linac with a proton beam of 350 MeV and 5 mA current intensity based on the work in the project PDS-XADS. This proton beam allows to reach a fast neutron flux of $1 \cdot 10^{15}$ n/cm².s (E > 0.75 MeV), which is necessary for the MA irradiation position. These performances were regarded as being within the reach of the extrapolated cyclotron technology of IBA. Nevertheless, taking into account the conclusions of the PDS-XADS project related to the accelerator reliability to be achieved for the ADS application, the linac option is now the favoured solution for the MYRRHA accelerator.

The MYRRHA project is intended to fit into the European strategy towards an ADS Demo facility for nuclear waste transmutation. As such it should serve the following purposes:

- ADS concept demonstration: coupling of the three components at rather reasonable power level (few ten's of MW_{th}) to allow operation with representative thermal feed-back and reactivity effects mitigation.
- Safety studies for ADS: to allow beam trips mitigation, subcriticality monitoring and control, optimisation of restart procedures after short or long stops, feedback to reactivity injection.
- MA transmutation studies: Requires high fast flux level ($\Phi_{> 0.75\ MeV} = 10^{15}$ n/cm²·s).
- LLFP transmutation studies: Requires high thermal flux level ($\Phi_{th} = 1–2 \cdot 10^{15}$ n/cm²·s).
- Medical radioisotopes: Requires also high thermal flux level ($\Phi_{th} = \sim 2 \cdot 10^{15}$ n/cm²·s).
- Material research: Requires large irradiation volumes with high constant fast flux level ($\Phi_{> 1\ MeV} = 1–5 \cdot 10^{14}$ n/cm²·s).
- Fuel research: Requires irradiation rigs with adaptable flux spectrum and flux level ($\Phi_{tot} = 10^{14}–10^{15}$ n/cm²·s).
- Initiation of medical and new technological applications such as proton therapy and proton material irradiation studies.

In mid-2002, the MYRRHA pre-design documents were submitted to an International Technical Guidance Committee for review of the pre-design phase. The conclusions and recommendations were as follow:

- No "show stoppers" are identified in the project.
- Give more attention to safety case studies and iterate to the pre-design before entering the detailed engineering phase.
- Address some R&D topics that can lead to timing bottlenecks very soon such as fuel pin and assembly development and qualification.
- Make a decision on the accelerator option (cyclotron vs linac) and eventually revisit beam parameters.

Responding to this recommendation some key design issues have already been addressed:

- The LBE corrosion by leaving the major part of the system at "cold" conditions and limiting the LBE velocity below 2.5 m/s.
- Criticality control during core loading by leaving the spallation target in position and loading from underneath.
- Avoiding spallation source window break by choosing the windowless design.
- Application of robotics and ultrasonic visualisation for service and maintenance activities.

MYRRHA is a challenging facility triggering a renewal of R&D activities within the fission community. Its development attracts young talented researchers and engineers looking for challenges.

2.7.4 Spain

With nine nuclear power plants and a total of 7.8 GW_{th} installed power, Spain is the fifth European country in the ranking of energy production and consequently of nuclear waste generation. Following the request of Spanish Senate, Spain has to make a revision of the radioactive waste management policy in 2010. Although with an initial preference for the direct disposal, the P&T option is still under consideration as a complementary technology for the management of the spent fuel /CIEMAT; Gonzalez 04/.

The Spanish programme in P&T is focused on creation of infrastructures and the acquisition of basic know-how, through participation in international projects of EU and on the NEA and IAEA international organizations working groups.

Spanish Science and Technology Ministry, MCYT, has recently established Partitioning and Transmutation as a strategic research line for the coming years. Previously ENRESA, the Spanish body responsible of radioactive waste management, incorporated P&T in its R&D programme. Both the MCYT plan and ENRESA selected CIEMAT (Research Centre for Energy, Environment and Technology) as its reference research centre for P&T. CIEMAT develops the R&D activities on P&T in collaboration with ENRESA, the Madrid Polytechnic University and several other university groups.

A separate project was created at CIEMAT – Fission driven by ACcElerator and Isotopes Transmutation (FACET), at present part of the Nuclear Innovation Programme – focused on the following topics:

- Developing computer simulation tools for ADS and transmutation.
- Participating in experiments related to ADS and transmutation.
- Evaluation of ADS configurations for radioactive waste transmutation.
- Evaluation of advanced fuel cycles including transmutation.
- To accumulate know-how and to be ready to provide neutronic advice to ADS projects and on fuel cycles with transmutation.

Collaboration with the Karlsruhe Joint Research Centre Institute of Transuranium elements (ITU) was established to study chemical processes with minor quantities of actinides and fission products. The lines of research are oriented towards advanced separation activities, using both hydrometallurgical and pyrometallurgical processes.

As part of the transmutation programme, CIEMAT has signed a collaboration agreement with the French Commissariat de l'Energie Atomique (CEA), making it possible to participate in important experiments and have access to the major nuclear infrastructure. The objective is to develop nuclear transport simulation models and perform joint experiments to obtain basic nuclear data (cross-sections), as well as to carry out integral verification experiments and the simulation of transmuting systems. At present, preference is given to subcritical transmuting systems driven by accelerators, and to the separation processes associated with them.

2.7.5 Korea

Korea /Yoo 02/ has already taken a small step towards a more effective fuel cycle through the DUPIC project (Direct Use of PWR spent fuel In CANDU), which recycles spent PWR fuel in CANDU reactors taking advantage of the very good neutron economy in those reactors. Currently transmutation, equally to direct disposal, is investigated in order to find an effective solution for the long-term management of spent fuels. Korea, like Japan, has difficult problems with siting of a geological repository due to a large population in a relatively small country and a strong opposition against any proposed site. Korean Atomic Energy Research Institute (KAERI) conducts a long term R&D transmutation project with a HYPER (HYbrid Power Extraction Reactor) Accelerator Driven System in focus. The HYPER system is, like many ADS systems in other countries, based on a liquid Pb-Bi cooled subcritical core of 1000 MW_{th}, fast neutron spectrum and metallic fuel.

The partitioning study is focused on the development of pyroprocessing technology based on the electrolysis of molten salts. The basic study currently being conducted at KAERI includes some experimental tests of lab-scale electrorefining and electrowinning by employing so far only non-radioactive materials.

Korean strategy of partitioning and transmutation

Pyrochemical separation methods, though still in the R&D stage, are attracting large interest in Korea as a prospective partitioning method for long-lived radionuclides. It is considered that pyrochemistry has noticeable advantages over wet processes, especially in terms of proliferation resistance as well as economy. According to KAERI's long-term R&D programme, KAERI will develop transmutation technology based on a hybrid transmutation system as well as pyrochemical technology as the basis of the P&T cycle.

The basic concept of the Korean transmutation system implies the use of the transuranium elements to be transmuted also as a nuclear fuel material generating power for electricity production. This puts some technological constraints on the transmutation system. The first requirement is separation of uranium from the spent fuel to avoid buildup of new transuranium elements. Moreover, burnup and other nuclear limitations require recycling of the transuranium elements in those systems.

A conceptual study of transmutation system

Korea is studying an accelerator driven system – HYPER, see Figure 2-8 – aiming at transmutation of transuranic elements and the fission products ^{99}Tc and ^{129}I.

Some parts of the conceptual study was performed during 1997–2000. The work during 2001–2003 has been focused on the evaluation of key unit systems of the HYPER. A conceptual design of the HYPER system will be completed during 2004–2006.

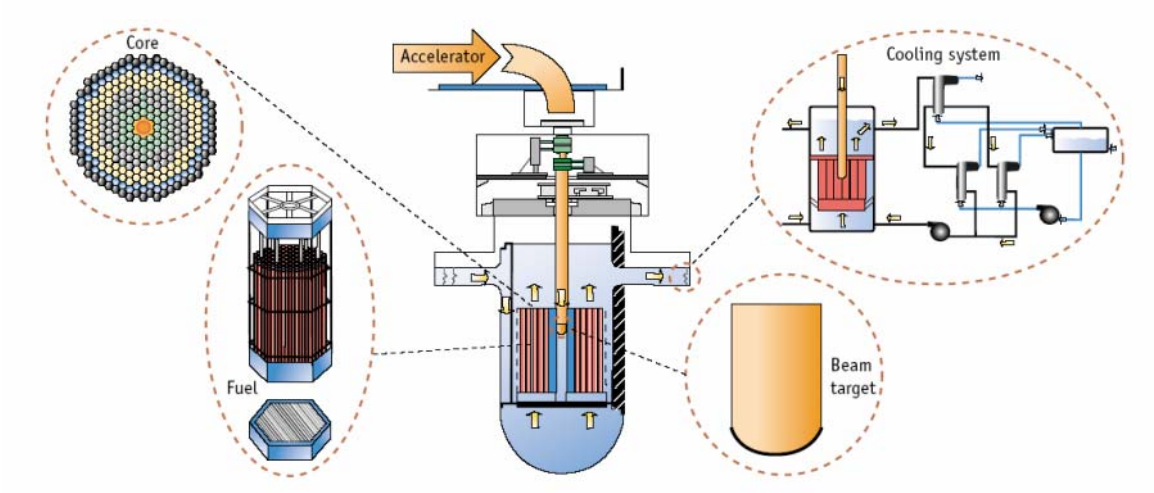


Figure 2-8. *Schematic diagram of the "HYPER" (Hybrid Power Extraction Reactor) accelerator-driven system (Source: KAERI, Rep. of Korea).*

HYPER core

HYPER is designed to transmute TRU as well as long-lived fission products at the same time by fast neutron irradiation.

In order to keep the relative assembly power within the design value the core should be divided into three different TRU enrichment zones. The fuel of low TRU fraction (26% TRU – 74% Zr) is designed to load in the innermost zone whereas the fuel of high TRU fraction(46% TRU – 54% Zr) will be loaded in the outermost region. The use of burnable absorbers in the core was found to reduce the reactivity swing by 38%.

Various optimisation studies on the concept of the core are now underway in as follows:

- Optimisation of height-to-diameter ratio.
- Optimisation of subcriticality level.
- Optimisation of spallation source.
- Development of 3-D kinetics code, HITE (3-dimensional hexagonal finite element code for transient and steady state).

In the analysis of the HYPER system, the partitioning process is assumed to achieve the rates 99.9% of uranium and 90% of lanthanide eliminations from LWR spent fuel (discharged burn up ≅ 33 000 MWD/MTU), respectively. As a result, the ratio of TRU to uranium will be 9**:**1 in HYPER.

HYPER fuel

One of the basic requirements for the selection of HYPER fuel is a good compatibility with the partitioning process. As the loss of radioactive nuclides cannot be avoided at each recycling process, a high burn-up capability is required as a selection criterion in order to minimise the loss. A metallic fuel has been selected because it has relatively good compatibilities with the dry partitioning process, high burn-up requirement, and also fast neutron spectra. Either a TRU-Zr alloy or a (TRU-Zr)-Zr dispersion type are considered as candidate fuel. In the case of the dispersion fuel, particles of 90 weight percent TRU – 10 weight percent Zr metal alloy are to be dispersed in a Zr matrix.

Pb-Bi Coolant and target

A lead-bismuth (Pb-Bi) eutectic alloy was selected as coolant material for the HYPER system because Pb-Bi is rather benign in terms of chemical reactivity and can be used as a spallation target also. A preliminary design study has been performed for its thermal hydraulics in the core.

2.7.6 Russia

Russia has an optimistic and creative approach to the future of nuclear power. The Russian long time scenario is based on fast reactor development /Lopatkin 02/. Almost all research focused on ADS is carried out with ISTC funding – see Appendix II.

Russian activities, aimed at setting up in the future a fuel cycle of nuclear power with reasonably minimised quantities of radioactive waste subjected for final disposal, are being carried out by Ministry of Atomic Energy (Minatom) as part of the general Strategy for development of national nuclear power. These studies do not comprise Accelerator Driven Systems, which are formally left for ISTC funded activities.

Several key missions of an official Russian plan deserve attention:

- In the next 20–40 years, advanced thermal reactors running on enriched uranium until the economically acceptable reserves of natural uranium are exhausted.
- Reprocessing of all spent fuel from thermal reactors to separate plutonium and long-lived nuclides.
- Development of a new generation of fast reactors which will meet the requirements placed on innovative reactors for large-scale electricity production (economic efficiency, safety, minimised radwaste, proliferation resistance).
- After 2030, deployment of a system of innovative fast reactors, using plutonium separated from spent fuel of thermal reactors, and solution with their help of the totality of problems associated with transmutation of long-lived nuclides.

Transmutation tasks in the Russian strategy comprise the following activities:

- Studies of various scenarios of nuclear power development with a certain focus on radiotoxicity reduction at the back-end of the fuel cycle.
- Development of radiochemical technologies appropriate for homogeneous or heterogeneous transmutation scenarios.
- Update and improvement of the nuclear data libraries for MA and LLFP.
- Alternative approaches to MA and LLFP transmutation.

Nuclear power development scenarios and assumptions

Scenario studies have been performed in Russia focused on nuclear power transition to the desired fuel cycle, depicted in Figure 2-9. Thermal reactors in that study were exemplified by VVERs and RBMKs. Fast reactors were represented by blanket-free BREST-1200 (lead-cooled) or BN-800 (sodium-cooled) with U-Pu-MA nitride fuel and core breeding ratio of 1–1.05. The closed fuel cycle of fast reactors can ensure transmutation of actinides both generated in the fast reactors themselves and contained in the spent fuel of thermal reactors. The latter actinides are incorporated in the fuel of

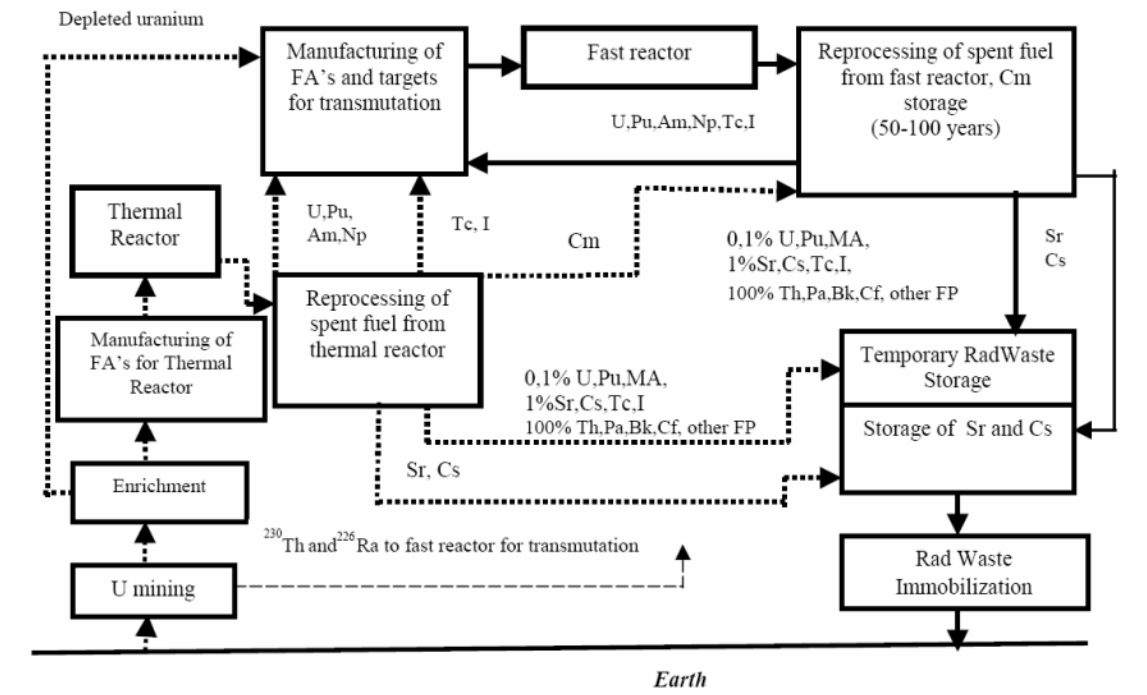


Figure 2-9. *Russian scenario for the future fuel cycle.*

the first charge. Transmutation of minor actinides in fast reactors accompanies energy generation, leaving the efficiency of the nuclear power system practically unaffected. Transmutation of LLFP (Tc and I) may also be achieved in FR but is considered to be less important at the current stage of the conceptual studies.

The studies concluded that transmutation of minor actinides is fully feasible in the system of thermal and fast reactors under consideration. It was assumed that construction of new thermal generating capacities would not proceed beyond the year 2020 (meaning VVER-1000s with the operating life of 50 years). Beginning in 2030, BREST-1200 power reactors would be put in operation at a rate of 1 unit per year. The spent nuclear fuel of thermal reactors will have accumulated ~ 570 tonnes of plutonium (including 320 tonnes of ^{239}Pu and ^{241}Pu) and 120 tonnes of MA. By the year 2100, using this plutonium together with their own small breeding, fast reactors in Russia can raise their capacity to 82.8 GW_e. Minor actinides from spent nuclear fuel of thermal reactors will be included in the first charges of fast reactors until the year 2080 and will be completely "incinerated" in the closed cycle of fast reactors towards the end of the 21st century. It has been shown that, given some 3% of MA in the starting fuel charge as loaded, about 40% of MA will burn up over the first fuel irradiation time.

In 2002, scenario studies have continued on future nuclear power development. Some cases are under investigation, which, in addition to the above, envisage introduction of thermal reactors with the Th-U fuel cycle after the year 2050.

P&T technologies

The initial round of R&D to support development of U-Pu-MA fuel reprocessing methods for the new generation of fast reactors, including transmutation of minor actinides, was carried out in 1999–2001 in Russia. The development was based on the following assumptions:

- Uranium and plutonium should not be separately extracted at any stage of the process (technological support of the non-proliferation regime).
- Neptunium and americium are not removed from the fuel (uranium + plutonium) during reprocessing, or – if removed – are taken back for recycling; these elements may also be added to the regenerated fuel.
- In reprocessing, it is advisable to extract separate fractions of Cm, Cs and Sr for subsequent cooling, and of I and Tc for subsequent transmutation.
- The disposed waste must have no more than 0.1% of U, Pu, Np, Am and Cm; 100% of the remaining actinides (Th, Pa, Bk, Cf); 1% of Cs, Sr, Tc and I.

At the initial stage, the feasibility of meeting the above requirements with the use of the following procedures was explored:

- Aqueous technologies:
  - Modified PUREX process.
- Dry processes:
  - Electrolysis of molten chlorides with reduction of actinides to metals or to nitrides – LINEX process.
  - Metallurgical process, with nitrides kept intact at all stages of reprocessing.
  - Pyrochemistry (ion exchange reactions) in molten fluorides and chlorides.
  - Dry fluoride volatility process.
  - Re-crystallisation in molten molybdates and phosphates.

No technology examined has shown fundamental difficulties for commercialisation with a production rate of 25–50 t of irradiated fuel per year; and the main requirements (see above) can be met under nominal conditions of the process. It is also possible to address the radwaste partitioning requirements by combining various methods. The difference in the costs of fuel processing by the examined methods does not exceed ± 10%. According to economic estimations, the cost of a plant for fuel reprocessing and refabrication will make no more than 15% of the cost of the nuclear power plant with two BREST-1200 units (2x1200 MW_e) to be catered for by this plant. Studies are in progress on non-aqueous methods, including ion-exchange reactions and electrolysis processes. Work on dry volatility process and advanced PUREX processes have been suspended due to their relatively easy transformation for the plutonium extraction.

In the recent years, RIAR has been engaged in studies (DOVITA Programme) on fuel manufacturing technology, in manufacture of mock-up fuel rods with mixed $(U,Np)O_2$, $(U,Pu,Np)O_2$ and $(U,Pu,Am)O_2$ fuel and their irradiation in BOR-60 to a burn up of 13–20% as well as in post irradiation examination of these fuel rods. These efforts are in fact the first practical attempts at MA transmutation; they provide experience of handling fuel with considerable MA amount (up to 5%) and demonstrate the effect of minor actinides on the irradiated fuel qualities.

Optimisation of MA and LLFP transmutation procedures and modes

Computational studies on modes of MA and LLFP transmutation in fast reactors have been performed at RDIPE and IPPE for the last 3 years, with focus on nuclear safety issues. Currently, for various reasons, transmutation of minor actinides is considered as small additions (no more than 3% by mass) to the core fuel (homogeneous approach). In this case, there is no need for setting up special production of targets for the separated MA fractions. Heterogeneous transmutation of minor actinides (as separate fuel assemblies) is also under investigation. Transmutation of LLFP will take place in the fast reactor blanket, while the neutron spectrum will have to be made considerably "softer".

Alternative approaches to MA and LLFP transmutation

It is believed in Russia that fast reactors alone are capable to transmute the minor actinides accumulated in spent nuclear fuel of thermal reactors and self generated in fast reactors. On the other hand, computational studies show that under certain circumstances (increase in the pre-reprocessing average spent fuel cooling times to 50 years and more, higher burn up of thermal reactor fuels, etc) there may appear problems with transmutation of minor actinides inside starting charges of fast reactors. In such a case, various approaches are possible including provision of dedicated reactors for transmutation of long-lived nuclides, e.g.

- Subcritical blankets with a target driven by an accelerator – ADS.
- Dedicated blankets of thermonuclear reactors – fusion driven transmutation.
- Critical reactors with liquid fuel – molten-salt and liquid metal reactors.
- Critical reactors of traditional design with solid fuel.

2.7.7 China

It is estimated that around the middle of this century China requires an installed capacity up to 1 200 GW_e electricity /Xialing 01; Zhao 02/. The nuclear power is officially regarded as the best choice in China to complement available potential of fossil and hydropower based power generation system.

Right now only small amounts of spent fuel from NPPs have been accumulated in China. But the situation will change drastically in the future according to a prediction of nuclear energy development in China. The annual generation of waste is estimated to 2275 m^3, 7500 m^3 and 10 000 m^3 respectively for the year 2004, 2010 and 2020.

Because of the limited uranium resources in China, China pursues the closed fuel cycle policy for nuclear power. Strategic decisions have been taken to construct the first Chinese commercial spent fuel reprocessing plant around 2020 and to develop the fast breeder reactor technology along with accelerator-driven systems.

A Chinese National Project to develop fast reactors and ADS is divided into three phases. The first phase will end at about 2005 when a first Chinese sodium cooled fast reactor – CEFR (Chinese Experimental Fast Reactor) will reach criticality. For the second phase a prototype (Prototype Fast Breeder Reactor – PFBR) of a 300 MW_e Modular Fast Reactor is under consideration. In the third phase it is planned to develop and deploy a Large Fast Breeder Reactor (LFBR) and a Modular Fast Breeder Reactor (MFBR), being suitable for MA burning.

In parallel to the fast reactor development a five year ADS-development programme was launched in 1998.

The main objectives of this programme were:

- Accelerator technology development:
  - Development of an ion source with a 60 mA proton beam.
  - Development of accelerator modules for a superconducting 1 GeV linac.
- System studies of ADS:
  - A medium size sodium cooled fast breeder subcritical core driven by an accelerator with of 1 GeV energy and intensity of 16 mA.
  - A pressurised heavy water moderated core (CANDU type) fuelled with Th-U and U-Pu fuels.
- Spallation Target studies concentrated on Pb (Pb-Bi) target.
- Experimental work on VENUS facility. This facility consists of a pulsed-neutron generator and a subcritical assembly. The pulsed-neutron generator is a 300 keV Cockcroft-Walton type accelerator for generating 14 MeV neutrons by $^{3}H(d,n)^{4}He$-reactions. The facility is used to study the neutron flux distribution, neutron energy distribution, the neutron enhancement, the reactor fission rate and k_{eff}.

The next ADS development phase is now under discussion and will most probably continue in the directions as described above.

3 Current research concerning partitioning

Traditionally, partitioning techniques used in the nuclear industry are dominated by liquid-liquid extraction based on a combination of organic and aqueous solutions. Organic molecules are degraded or destroyed by high-energy radiation, which complicates the design of processes and increases operating costs. Hence, numerous attempts have been made over the years to develop viable non-aqueous partitioning alternatives, which would have much smaller sensitivity to radiation – but this work has only had limited success.

3.1 Aqueous partitioning techniques

The main aqueous technique considered for partitioning of actinides, lanthanides and long-lived fission products is liquid-liquid extraction (see 3.1.1–3.1.3) – which has been used in industrial scale reprocessing of spent nuclear fuel for more than 50 years – but also chromatographic separations, ion exchange separations, ionic liquids, supercritical fluid extraction and supported membrane extraction (see 3.1.4) are referred to as aqueous techniques. The aqueous processes have in common that the separation can be strongly affected and optimized by changing solution properties like acid concentration, redox potential, and the presence of complexing agents. They often show a high selectivity for the desired elements and a high yield, i.e. small losses. Many of the techniques and reagents are widely used for other process applications, e.g. metal production, where a long time trend has been to convert from pyrochemical processes to liquid-liquid extraction. Thus a lot of experience and proven equipment are available. Other advantages are that a properly designed liquid-liquid extraction process has a very low environmental impact, is continuous, and that it can be designed to operate for many years by only remote control. A drawback is radiolytic destruction, or alteration, of the reagents when processing highly radioactive materials, which requires effective clean-up systems on-line. Such clean-up operations produce low-level waste in case the reagents used are not completely incinerable – which is often the case today. This is the reason for the current drive to develop new separation reagents containing only carbon, hydrogen, oxygen, and nitrogen atoms – so called CHON-reagents.

The work on aqueous partitioning techniques since the previous P&T report /Enarsson 98/ has thus to a large extent been concentrated on reagents confirming to the CHON-principle, i.e. reagents that are completely incinerable. In addition, considerable work on phosphorous containing reagents has also been performed. We will treat these two general types, non-CHON and CHON reagents in that order followed by a review of separation processes. Finally some aqueous based alternatives to solvent extraction are reviewed.

3.1.1 Phosphor-based ligands

Overview

Extracting agents or synergists containing phosphor were among the first ones used for extraction of actinides and later also for lanthanides. The most well known is TBP (tri butyl phosphate) which is used together with nitric acid in the PUREX[48] process for reprocessing of spent nuclear fuel from the late 1940ies. As emphasis shifted to separation of the different 5f elements in the 1950ies and 1960ies the development of acidic organophosphorus extractants increased significantly. Phosphoric, phosphonic and phosphinic acid esters containing at least one conjugating proton were extensively investigated for the separation of various metals, but particularly for trivalent lanthanides and actinides. The premier of these is HDEHP (di-(2-ethylhexyl)phosphoric acid), see Figure 3-1, which is still extensively used.

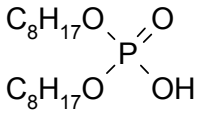


Figure 3-1. *Schematic picture of HDEHP.*

However, HDEHP cannot facilitate a complete separation of lanthanides and actinides since their cation radii and their distribution coefficients overlap. To solve this problem the TALSPEAK (Trivalent Actinide Lanthanide Separation by Phosphorus Extractant and Aqueous Complexes) process was developed. The aqueous medium is lactic acid and DTPA (diethylenetriaminepentaacetic acid) and the extractant is HDEHP. Later, the extraction of trivalent actinides within a PUREX style process was desired and a new type of extractant was used, CMPO (octyl(phenyl)-N,N-diisobutyl-carbamoylmethylphosphine oxide). The process based on these agents, the TRUEX (TRans Uranium EXtraction) process, was developed. Most kinds of commonly used phosphor containing agents are shown in Figure 3-2.

[48] PUREX = Plutonium Uranium Redox Extraction.

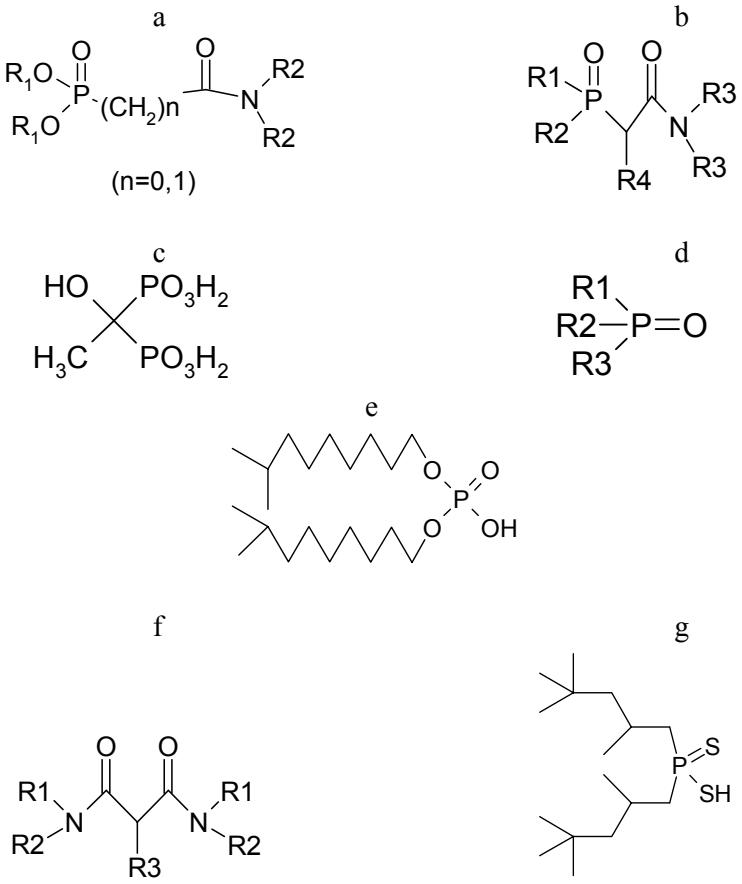


Figure 3-2. *Structures of extractants and complexants: a) carbamoylphosphonate (n=0) and carbamoylmethylphosphonate (n=1), b) carbamoylmethylphosphine oxides, c) 1-hydroxyethane-1,1-diphosphonic acid, d) trialkylphosphine oxide, e) diisodecylphosphoric acid (DIDPA), f) bridge-substituted tetraalkylmalonamide, g) bis(2,4,4-trimethylpentyl)dithiophosphinic acid (Cyanex 301). (Figure 3-2 from /Math 01/.)*

Phosphine oxides

The most extensively developed classes of reagents for recycling and isolation of minor actinides are the carbamoyl phosphonates and phosphine oxides /Hor 99/. It is known that the extracting power of neutral organophosphorus compounds increases in the order phosphates < phosphonates < phosphinates < phosphine oxides /Bur 58/. Unfortunately the synthesis difficulties increase in the same order. Almost 100 derivatives of this kind of extractants have been developed but the most studied is CMPO, which positive features are its relatively small affinity for trivalent actinides, relative acid independence and the adaptability to a variety of acid media. On the other hand it is relatively expensive and has some limitations in its phase compatibility /Nas 02a/. The CMPO class has been extensively studied for several years but is still the focus for many different studies, extracting capability for actinides and lanthanides /Rao 98; Kara 03; Mura 01/, other metals and the degradation products of CMPO and their extraction ability /Math 98a/ and using slightly unusual techniques such as water-in-oil micro emulsions /Nag 00/. It has been shown that exchanging the nitric acid media to perchloric acid media enhances the extraction using CMPO systems /Kul 02/.

A very good separation within the lanthanide group has been achieved using the CMPO-TBP mixtures with a DTPA (diethylenetriamine pentaacetic acid)-nitrate solution /Kom 99/, and suggested process flow sheets have been developed for e.g. recovery of Am and Cm /Kom 98a/. In order to find a more cost effective system other reagents have been studied, e.g. dialkyl metylphosphonates /Chm 02/. There also exists a kind of merge between the pyridine type of extractants discussed below and phosphino groups. Although the results are not outstanding, they call for further studies and then mainly in the US /Nas 02b/. By mixing several trialkyl phosphine oxides there is hope that a great number of radionuclides may be extracted in one step. However, being a rather undefined mixture the practical application still has some time to come /Han 03/.

Phosphoric acids

Acidic extracting agents based on phosphorous are nearly as common as the phosphine oxides and one of the most well known is HDEHP (di-(2-ethylhexyl)phosphoric acid). By changing some groups of the molecule new derivatives can be made and there exist many of these generally written as $H_2DEH[R_i]$ where R_i describes the added groups which may be MDP for methane diphosphonic acid or BDP for butyl diphosphonic acid. One such compound, (diphenylphosphenylmethyl)phenylphosphinic acid, was thoroughly studied with respect to its extraction ability for e.g. most of the lanthanides, thorium and uranium with very promising results /Tur 02/. More general studies covering several metals such as iron, lanthanides and some actinides were performed at Argonne National Laboratory. In addition they were also studying the complexation mechanisms and the stoichiometry of the reactions /Chia 97; Chia 98; Chia 01/. The extracting agent DIDPA (diisodecylphosphoric acid) is sometimes used together with TBP for extraction of both lanthanides and actinides. The elements are then selectively stripped using hydrazine carbonate and good separation factors (at least > 3) have been observed both between actinides and lanthanides as well as within the actinide group /Wat 02a/. A thorough study on the complexation behaviour of di(chlorophenyl)dithiophosphinic acid and a variety of O-bearing co-extractants was made. This work contains the stoichiometry of the extracted and stripped complexes /Ion 01/. Other mixtures of extracting agents that show promising separation (Am/Eu > 20), contain bischlorodithiophosphinic acid with TBP, TOPO (trioctyl phosphineoxid) or TBPO (tributyl phosphineoxide) /Mod 98a/.

One common group of extractants is the Cyanex reagents. Although there exist Cyanex compounds belonging to the phosphine oxide group, e.g. Cyanex 923 (trialkylphosphine oxide) most of the ones used for partitioning of nuclear waste are phosphoric acids. One of their main advantages is that they are more stable than similar thiophosphororic acids /Math 01/. The most often used Cyanex compounds are Cyanex 272 (bis(2,4,4.trimethylpentyl) phosphinic acid), Cyanex 301 (bis(2,4,4-trimethylpentyl)dithiophosphinic acid) and Cyanex 302 (bis(2,4,4-trimethylpentyl)monothiophosphinic acid). A study has been made on the stoichiometry of the extracted lanthanide and actinide complex using these ligands and it was not surprisingly found that three Cyanex molecules bind to a metal ion in all cases except when using rather high nitric acid concentration where nitrate ions also participated in the extraction /Jen 02/. These ligands may also be used in synergy as demonstrated by Reddy et al, who used Cyanex 301 together with Cyanex 923 and obtained good results also investigating the stoichiometry of the extracted complex /Red 99/. The extraction of Am and Eu using Cyanex 301 alone was also studied /Zhu 96/.

Calixarenes

In the 1940ies a group of substances called [1_n]metacyclophanes with the common name calix[n]arenes was discovered. However their popularity did not increase greatly until the 80's and they are now the subject to intense studies in many countries. Their chemical structure can vary considerably but in general they are fairly complex molecules. A good description of calixarenes in general and their extraction behaviour is given in /Lud 00a/. Several authors have studied the separation of actinides and lanthanides using different calixarenes and the results are promising, but the synthesis of the different molecules is sometimes rather difficult and may thus not be suitable for industrial processes /Rom 99; Lud 00b; Ata 02; Doz 02; Ard 00/. The calixarenes are still interesting from a basic research perspective since extremely high selectivity can be obtained. Other elements such as technetium and cesium have also been removed from acidic liquid waste /Gru 99/. Some attempts have been made to merge to effects of the CMPOs and the calixarenes /Del 98; Del 99/.

3.1.2 Non phosphor-based extractants

Extractants based on pyridine

N-donor extractants promise a higher chemical stability than sulphur compounds. 2-4-6-tri(2-pyridyl)-1,3,5-triazine (TPTZ) was the first reported effective N-donor extractant, being a solvating compound and combined with 2-bromodecanoic acid as an anion source /Mus 84/. The ligands have then been improved with substitutes to be more chemically and radiolytically resistant, as well as to increase the distribution ratios and separation factors. Several molecules have been investigated regarding their extractive properties in different systems /Kol 99a; And 02; And 03a; And 03b; Nil 02; Hag 99; Hud 03; Bou 02; Char 02; Cor 98/. The influence of the length of the substituent alkyl chain and effect of diluents have also been studied /Kol 99b; Wei 03/. Different modifiers can also be added to the system to increase the solubility of the ligand (and more specific the protonated species).

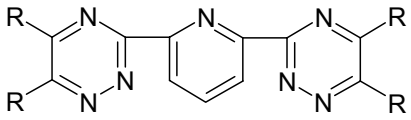


Figure 3-3. *2,6 bi(5,6-dialkyl-1,2,4-triazin-3-yl)pyridine.*

6-methyl-2-(2-pyridyl)benzimidazole, also a pyridine based extractant, is one example of studied 2-substituted benzimidazoles /Kol 97/. The separation factor between Am and Eu has been investigated as a function of anion and decreases in the order thiocyanate > nitrate ~ perchlorate ~ iodide > thenoate > 2-(2-pyridylazo)-1-naphtolate.

Monoamides

Amides are resistant toward radiolysis and their complete burnability leads to smaller amounts of waste. Their main degradation products show no interference in the separation of U(VI) and Th(IV) from fission products /Mow 01/ and can easily be washed out of the extractant. In /Mow 01/ the disubstituted monoamides N,N-dihexyloctaneamide and N,N-dihexyl(2-ethyl)hexylhexanamide have been studied regarding extractive properties in different systems.

Malonamides

The main advantages of the malonamides compared to organophosphorous extractants are their innocuous radiolytic products and their ease of incineration and synthesis. The structure of the malonamide has a great influence on the metal extraction. Solubilities, electronic effects and steric hindrances together influence the degree of extraction. In one study it was indicated that one of the substituents on the nitrogen atoms should be a methyl group to minimise the steric hindrance and the other nitrogen substituent should have a long carbon chain to make the molecule more lipophilic /Spj 97/.

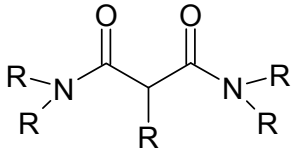


Figure 3-4. *Basic structure of a malondiamide.*

IR measurements indicate that the metal is coordinated to the carbonyl oxygen atoms rather than to the nitrogen /Nig 94/. The extraction is increased if the central methylen carbon has alkyl or oxyalkyl groups attached /Mow 02/. Malonamides with lower basicity show the best metal extraction, which is the opposite of corresponding organophosphorous extractants and monoamides, at least at low acidity /Spj 00/. The extraction mechanism is changed from a coordinative mechanism at low acidity to an ion-pair mechanism at higher acidity (1–14 M HNO_3) /Chan 97/. Kinetic studies indicate that diffusion controls the reactions. The extraction rates for lighter lanthanides are 50% higher than for heavier lanthanides /Wei 01/

Bicyclic diamides

The novel bicyclic diamide 3,9-diaza-3,9-dioctylbicyclo[4.4.0]decane-2,10-dione has been studied /Lum 03/. Both calculations and experiments show that the two amine oxygen atoms are ideally situated for binding to trivalent actinide or lanthanide ions and forming strong complexes. The extraction of Eu is 10^7 higher than conventional malonamide extractants, but also Am is strongly extracted.

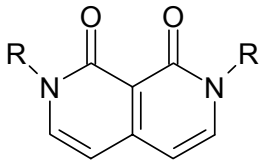


Figure 3-5. *Basic structure of a bicyclic diamide.*

Diglycolamides

The extraction behaviour of several diglycolamides has been studied. The extractability of diglycolamides for actinides is higher than that of malonamides /Sas 00; Sas 01/. Introducing ether oxygen (which plays an important role in the chelation to actinides /Nar 99/) into the main frame increases the extractability, as does also the substitution of oxygen atoms to ether sulphur atoms. The distribution ratios gradually increase with increasing atomic number in the lanthanide group, which is the opposite trend to that obtained for other diamides /Sas 02/.

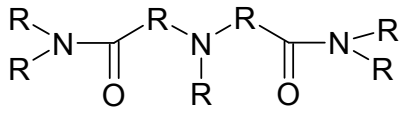


Figure 3-6*. Basic structure of a diglycolamide.*

N,N,N',N'-tetraoctyl-3-oxapentanediamide extracts both actinides and lanthanides completely from aqueous nitric acid solution /Sas 01/.

The influence of the length of the alkyl chain was investigated, showing an optimum for this diglycolamide. A longer alkyl chain suppresses the extraction but increases the solubility. The influence of diluent was also studied. Solvents with higher dielectric constants gave higher distribution ratios due to the fact that aromatic or halogenated solvent might react with the donor oxygen of the diglycolamides. The distribution ratios increase with increasing nitrate concentration. The separation between Am and Eu is sufficient, but separation of some other elements may be difficult to perform.

Diamines

Tetrapodal hexadentate ligands have higher selectivity and chemical stability than other conventional N-donor ligands. Water molecules that coordinate strongly around the metal ions are removed by encapsulation of the ligand.

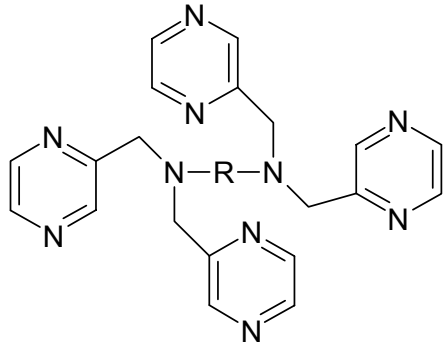


Figure 3-7. *Basic structure of tetrapodal hexadentate ligands.*

The correlation between the ligand architecture and the metal ion binding selectivity is a rather new area of research and have been studied for this type of ligands in e.g. /Wat 02b; Karm 02/ and /Jen 00/. The N,N,N',N'-tetrakis(2-pyridylmethyl)ethylenediamine (TPZEN) shows a separation factor at ~ 100 between Am and Eu at pH ~ 4. Nitrate ions are involved in the extracted species.

PARTNEW

The European PARTNEW program concerns the partitioning of minor actinides from high level liquid radioactive wastes. The main scientific goals are to study possible solvent extraction processes for the partitioning of minor actinides, to investigate the preferential extraction of minor actinides over the other elements and to design a 2-cycle process including 1) a SANEX process for separation of An/Ln and 2) an Am/Cm separation process. The design of a 3-cycle process including 1) a DIAMEX process for actinide + lanthanide separation from acidic HLW, 2) a SANEX process for An/Ln separation and 3) an Am/Cm separation process was also studied considering two principal feeds for the process: 1) the acidic High Active Raffinate (HAR) issuing the reprocessing of UOx or MOx spent fuels and 2) the High Active Concentrate (HAC) produced after concentration/denitration of HAR.

The research has involved the preparation of new solvent extracting agents containing polyamides, heterocyclic nitrogen- and sulphur-bearing ligands. Methods have been established on traditional and combinatorial chemistry (synthesis and screening tests). The reactivity of the new ligands versus the target metal ions has been studied and computer models of the complexes formed developed. A library of molecules and corresponding data has been established /Mad 03; PAR 03/.

3.1.3 Separation processes using solvent extraction

The DIAMEX (DIAMide EXtraction) process, which is intended for separation of actinides and lanthanides from acidic HLW (High Level Waste), has been investigated by several authors. Courson et al /Cou 00/ performed a centrifugal extractor experiment with simulated HLLW (High Level Liquid Waste) corresponding to a raffinate from the PUREX process. Four extractors were used for extraction and four for stripping. A hot batch test with genuine HLLW was also performed. Malmbeck et al /Malm 00/ performed a hot test with a 16 stage centrifugal extractor battery using genuine PUREX raffinate. DMDBTDMA (DiMethyl DiButyl TetraDecyl MalonAmide) was used as extractant in these tests. Serrano-Purroy et al /Ser 03/ used a similar extractor battery for a test with simulated HAC obtained by concentration and denitration of HAR obtained from PUREX reprocessing of MOX fuel. In this case DMDOHEMA (2-(2-hexyloxy-ethyl)-N,N'-dimethyl-N,N'-dioctyl-malonamide) was used as extractant. In conclusion, the DIAMEX process is reported to represent an excellent compromise between good extraction and back-extraction properties. Decontamination factors of > 300 for minor actinides and 100–230 for lanthanides were achieved, as well as an Am and Cm recovery of about 99.9%. Oxalic acid scrubbing was efficient in preventing co-extraction of molybdenum and zirconium. A process optimization is, however, needed to prevent co-extraction of technetium, ruthenium, palladium and neptunium. In the HAC test /Ser 03/ oxalic acid and HEDTA showed to prevent co-extraction of molybdenum, zirconium and palladium. Geist et al /Gei 03a/ also studied the DIAMEX process with DMDOHEMA as extractant but instead of centrifugal extractors they used hollow fiber modules (HFM) as liquid-liquid phase contactors. The process was tested with a synthetic PUREX HAR raffinate spiked with americium. Both extraction and back-extraction of americium worked very well and a decontamination factor of 20 000 was obtained together with a 99.995% extraction yield.

The ALINA (or SANEX IV) process uses a solvent containing chlorine-substituted aromatic dithiophosphonic acids + tri octyl phisphine oxide (TOPO) in an aromatic diluent for An(III)/Ln(III) partitioning. It was developed and thoroughly investigated by Modolo and Odoj /Mod 98a; Mod 98b; Mod 99; Mod 02/. It was found that aromatic dithiophosphinic acids are more stable to hydrolysis and radiolysis than aliphatic dithiophosphinic acids like Cyanex 301. Good separation factors were obtained. Geist et al /Gei 03b/ used counter-current centrifugal extractors and HFM to test the SANEX-IV (or ALINA) process with di(chlorophenyl) dithiophosphinic acid as selective extractant for trivalent actinides. Am decontamination factors of > 1000 and 25 000 were found using centrifugal extractors and HFM, respectively. HFM has also been used by Geist et al /Gei 03c/ to investigate the SANEX-III process that uses n-Pr-BTP (2,6-di(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine) as solvating extractant. This system works well but is impeded by a slow chemical reaction. BTPs (bis-triazinyl-pyridines) were also investigated by Hill et al /Hil 02/ for use in the SANEX process.

The diamide-like ligand TODGA (TetraOctylDiGlycolAmide) has been proposed by JAERI. It has a higher affinity for An(III) and Ln(III) than the diamide of the DIAMEX process. TODGA has been studied by Modolo et al /Mod 03/ in batch tests and in tests with centrifugal extractors. It is promising, but it requires high oxalic acid concentrations to prevent Zr extraction.

In Japan Arai et al /Ara 97/ ran the TRUEX (TRansUranium EXtraction) process with n-dodecane as solvent instead of carbon tetrachloride. Three flow sheets were tested: the original Argonne National Laboratory flow sheet with extraction, scrub and strip, flow sheet 2 with an extra scrub and flow sheet 3 with strong nitric acid as scrubbing solution. The processes were tested with a synthetic waste solution mainly containing lanthanides (Nd was used as model element for the actinides). Small mixer-settlers were used. flow sheet 3 was found to be the best since no oxalate precipitations were observed. Ozawa et al /Oza 98/ tested new salt-free reagents like hydrazine oxalate, hydrazine carbonate and $(CH_3)_4NOH$ to separate actinides and fission products in the TRUEX and SETFICS flow sheets. Hot tests were performed in a mixer-settler battery. A similar approach was chosen by Chitnis et al /Chit 99a; Chit 99b/ who tested the strip of actinides, mainly Pu and Am, from the TRUEX solvent using a mixture of formic acid, hydrazine hydrate and citric acid. The experiments were performed batch wise as well as in a small mixer-settler battery. Koma et al /Kom 98b/ successfully demonstrated the usefulness of the SETFICS process by using a real TRUEX product solution and a mixer-settler battery.

In the US Spencer et al /Spe 99/ dissolved sludge from Oak Ridge National Lab and used it to evaluate the TRUEX process. The partitioning of Eu, Pu, Cm, Th and U was found to work almost as predicted by the TRUEX Generic Model. Hg was, however, found to be strongly extracted. The Hg problem was studied by Fiskum et al /Fis 01/ by using actinide containing waste produced at the INEEL reprocessing facility. The TRUEX process was used with an addition of L-cysteine to the feed solution. The L-cysteine was found to block Hg extraction without affecting the extraction of other elements significantly.

Also in the US, Del Cul et al /Del Cul 97/ ran a modified TALSPEAK process using mixer-settlers and solutions containing e.g. Zr, Mo and Nd. The modification was that citric acid was used instead of glycolic or lactic acid. It was found that the use of citric acid eliminated or greatly reduced the deleterious effects caused by impurities like Zr.

Romanovski et al, Law et al and Herbst et al /Rom 01; Law 01; Her 02/ have tested the UNEX flow sheet in batch and centrifugal contactor experiments. UNEX is designed to separate cesium, strontium and actinides from acidic radioactive waste. It uses chlorinated cobalt dicarbollide for Cs extraction, polyethylene glycol for Sr extraction and diphenyl-*N,N*, dibutylcarbamoyl phosphine oxide for actinide extraction. Actinides are stripped with DTPA. Lanthanides seem to be extracted together with the trivalent actinides.

Jianchen and Chongli /Jia 01a/ performed centrifugal contactor tests of the TRPO (trialkyl phosphine oxide) process that uses a mixture of trialkyl phosphine oxides as extractant. The process recovers trivalent actinides and lanthanides. The same authors also performed centrifugal extractor tests /Jia 01b/ on strontium partitioning from HLLW by DHC18C6 (dicyclohexano-18crown-6).

Guiragossian et al /Gui 03/ suggest a new hydrometallurgical method for partitioning of nuclear waste. The method is based on electrolysis. There are, however, several questions concerning the flow sheet (actinides that do not precipitate in alkaline solution, actinides that float on top of mercury etc) and it will not be further discussed here.

3.1.4 Alternative separation techniques to solvent extraction

Although solvent extraction is the dominating separation technique when dealing with nuclear applications there are some other methods and complements to solvent extraction available and under development. Many of these systems still require similar or the same extracting agents as regular solvent extraction.

Alternative diluents

Some investigations of alternatives to organic diluents have been made.

Polyethylene glycol (PEG) based aqueous biphasic systems have been tested /Paw 99/. The interest for PEG instead of conventional oil-water systems is that PEG is cheap, easily available and non-toxic in nature. Also it does not involve organic diluents, which are usually volatile, inflammable and toxic. By using phosphotungstic acid as extracting agent and a salt-rich aqueous phase, separation factors of around 10 can be obtained at optimal conditions.

Attempts to use ionic liquids (IL) have been made /Vis 03; Nak 03/. ILs are composed of organic cations and either organic or inorganic anions that remain liquid over a wide temperature range, including room temperature. IL characteristics can be dramatically adjusted by changing the anion type, or subtly altered by changing the length or number of alkyl groups appended to the cation. One benefit of the IL is, as with PEG, that they are not volatile or flammable. Typical ILs are non-coordinating and cannot extract metal ions from the aqueous phase by themselves. Therefore an extracting reagent is needed. Experiments using CMPO and TBP have been made and distribution ratios of Am^{3+}, Pu^{4+}, Th^{4+} and UO_2^{2+} are all at least one order of magnitude higher than compared to similar conditions using dodecane /Vis 03/. A number of lanthanides, La, Ce, Nd, Sm, Eu, Gd, Ho and Er, has been extracted using CMPO dissolved in an IL and results show that all tested lanthanides are extracted better, particularly the light lanthanides, than with dodecane as organic phase /Nak 03/. One problem with IL is that the solubility of CMPO is much lower than in dodecane. Also, the toxicological and safety aspects of these new solvents are not yet known. Thus care should be exercised in the use and study of these chemicals /Vis 03/.

Supercritical fluids as an alternative to organic diluent were investigated /Murz 01; Murz 02/. Supercritical fluid extraction (SCE) is based on the fact that the density of a gas in supercritical state is close to that of a liquid. Strong dissolving power of these fluids, having viscosity close to that of gases, provides high penetration ability of fluids and rapid extraction /Murz 01/. Carbon dioxide (SC-CO_2) is the most frequently used agent in supercritical extraction. The solubility of an uranyl trifluoroacetylacetonate complex into SC-CO_2 was investigated /Murz 01/ and the solubility of UO_2^{2+}, Pu(IV), Np (IV), Am (III) and Eu(III) in complexes with different extracting agent such as β-diketones and TBP into SC-CO_2 was investigated /Murz 02/. The studies showed that the solubility was high but also that SC-CO_2 cannot be considered as an inert solvent due to partial degradation of complexes in the supercritical fluid.

Mathur and Choppin have investigated the use of paraffin wax as organic diluent in solvent extraction /Math 98b; Math 98c/. The extraction is performed at elevated temperature so that the wax is melted and with quick cooling the wax solidifies and can be easily separated from the aqueous phase. When performing extraction at 65°C and using TBP as extracting reagent UO_2^{2+} and Pu^{4+} was extracted to 96–99% and ~ 98% respectively /Math 98b/. Eu^{3+} at the same conditions was not extracted as trivalent actinides and lanthanides are not extracted by TBP. When using TOPO as extracting agent at similar conditions it was possible to recover ~ 90% of Eu^{3+} and ~ 99% of Pu^{4+} and UO_2^{2+} /Math 98c/. In laboratory scale this technique proved easy but in macro scale reprocessing schemes this system may be handicapped by the necessity of heating and cooling the paraffin wax.

Alternative extraction systems

Supported liquid membrane (SLM) extraction

By associating an extracting agent on a SLM extraction and concentration of the extracted elements can be performed in one step /Cri 99; Hos 99/. SLMs are constituted by two aqueous phases separated by an organic phase impregnated on a microporous support. The feed solution contains radioelements and salt buffer. The second aqueous phase is the stripping solution, which receives the concentrates of the elements being extracted. Using neutral carriers, the driving force of the transport is the difference of salt concentration between the feed and the stripping solution. If the extracting agent is selective towards the cations of the salt buffer or protons, it is possible to transport radionuclides, even against their own concentration gradient. By using phosphorous compounds as extracting agents and a feed solution of 4 M $NaNO_3$ and 1 M HNO_3, extraction of Pu and Am up to 100% and Np to 73% was achieved /Cri 99/. A SLM impregnated with highly purified Cyanex 301, a phosphinic acid, was able to effectively separate Am from Eu. Since the feed volume was much larger than the product volume, americium was transported against its concentration gradients and was concentrated into the product solution /Hos 99/.

Composite sorbents and chromatographic separation

The use of solid-phase extraction instead of an organic phase for a two phase system has been investigated /Gra 99; Man 02; Bar 00; Nai 02; Hor 97; Matt 01; Buc 97; Aal 02; Malo 01/. One solid extractant that has found wide use is TRU-resin™, which is synthesized by Eichrom Industries, Inc, USA. This is a polymer (polyacrylate) bead that has been impregnated with CMPO and TBP /Gra 99/, which yields an actinide specific sorbent material. Some research is being made for improving this type of solid extractant by changing the inert support material into e.g. polyacrylonitrile /Man 02/, poly(4-vinylpyridine) /Bar 00/ or silica gel /Nai 02/. Improvements can also be made by changing the extracting agent /Hor 97; Nai 02; Bar 00/ for targeting specific elements. These solid extractants can easily be packed in columns for chromatographic separation. One technique for separating actinides by chromatography is sequential injection (SI) /Gra 99/. After the sample has been loaded in the column, different wash solutions can be introduced to the column one after another (hence sequential) to rinse out specific elements. This system can serve as basis for an automated radiochemical separation workstation by using standard laboratory equipment.

If a small amount of ferromagnetic material is encapsulated in a solid extracting bead the separation of the solid material from an aqueous phase could be made with magnets. This has been performed /Matt 01; Buc 97/ and a magnetically assisted chemical separation (MACS) process has been investigated. The advantage with magnetic particles in a column is the ability to utilise magnets to suspend the solids so that it does not clog the column. It would also be possible to make magnetic fluidised beds. Matthews et al /Matt 01/ tried to increase the extraction and selectivity of actinides over lanthanides by combining CMPO with a calix[4]arene on a magnetic particle. This resulted in an increase of extraction for Eu(III), Am(III) and Ce(III) compared to using CMPO alone but no specific separation between actinides and lanthanides was observed.

Ion exchange resins

Tertiary pyridine-type anion exchange resins have been investigated for separation between actinides and lanthanides /Nur 99; Suz 03/. Nur et al showed that the trivalent elements Ce, Gd, Tb and Y did not adsorb on the pyridine resin while the actinides U, Am and Cm did /Nur 99/. These results show that the actinides can be separated from lanthanides and also some separation between americium and curium and also between curium and uranium was observed. Similar results were found by Suzuki et al where a separation factor of around 1.7 was found for americium and curium at optimal conditions /Suz 03/.

3.2 Non-aqueous partitioning techniques

By non-aqueous partition techniques it is generally understood chemical processing at elevated temperatures. Thus, such processes are also often called pyrochemical. The major attraction of these processes lies in the fact that metals, oxides, or salts are used, which in general are much more resistant to damage by ionizing radiation than molecular liquids like water or organics. Another favourable property is that much higher concentrations can be handled, decreasing the size of the plant. A third good property has also been advocated, i.e. the assumption that pyrochemical processing of spent nuclear fuel can be designed in such a way that separation and production of pure plutonium can be avoided, made very difficult, or perhaps nearly impossible to achieve[49].

3.2.1 Systematic of non-aqueous separation processes

Non-aqueous separation processes can be classified as follows:

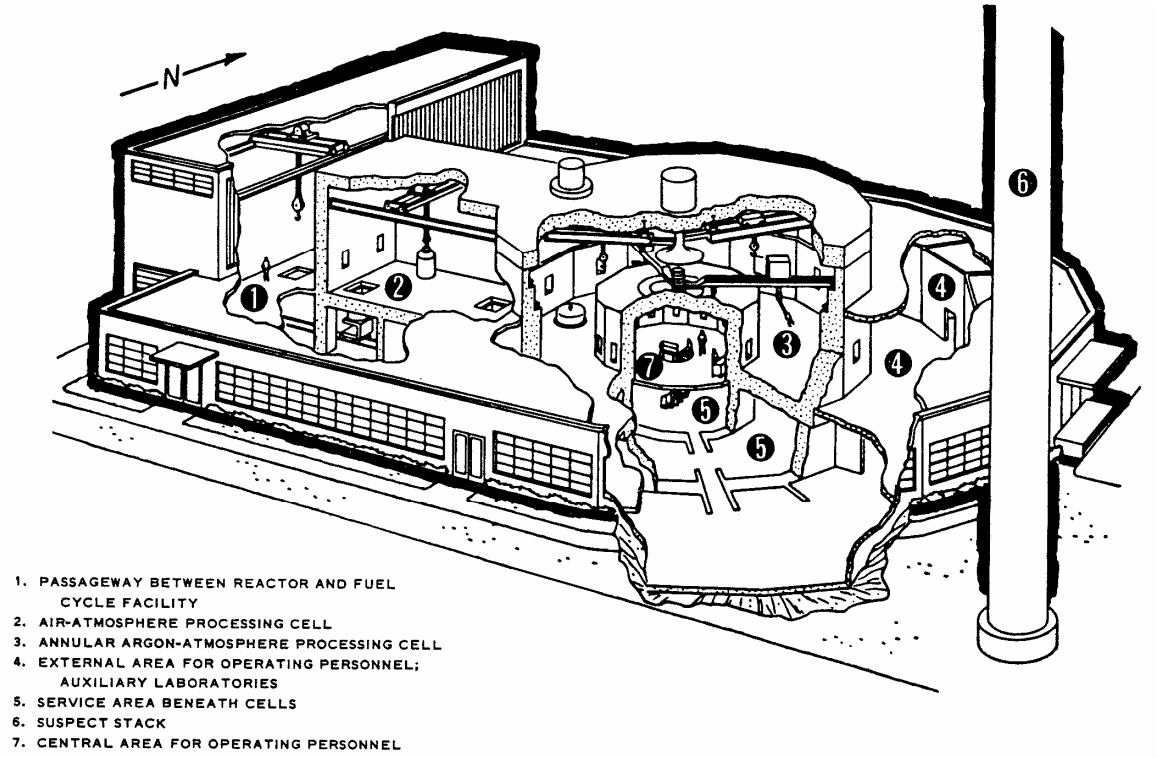
- **Distillation**
  Of special interest is the possibility to separate the carrier metal, e.g. zinc, from a mixed liquid metal melt by distilling away the carrier metal, thus recovering the dissolved metal(s), e.g. U and/or Pu, from the melt in a rather pure form.
- **Reactive vaporisation**
  The reaction used has been fluorination of a fluoride melt by a HF/F_2-gas mixture thereby generating volatile UF_6 and PuF_6, which distil from the salt-melt and can be recovered by cooling of the escaping gas to near room temperature.

[49] Comment: Some experts seriously question the validity of this argument.

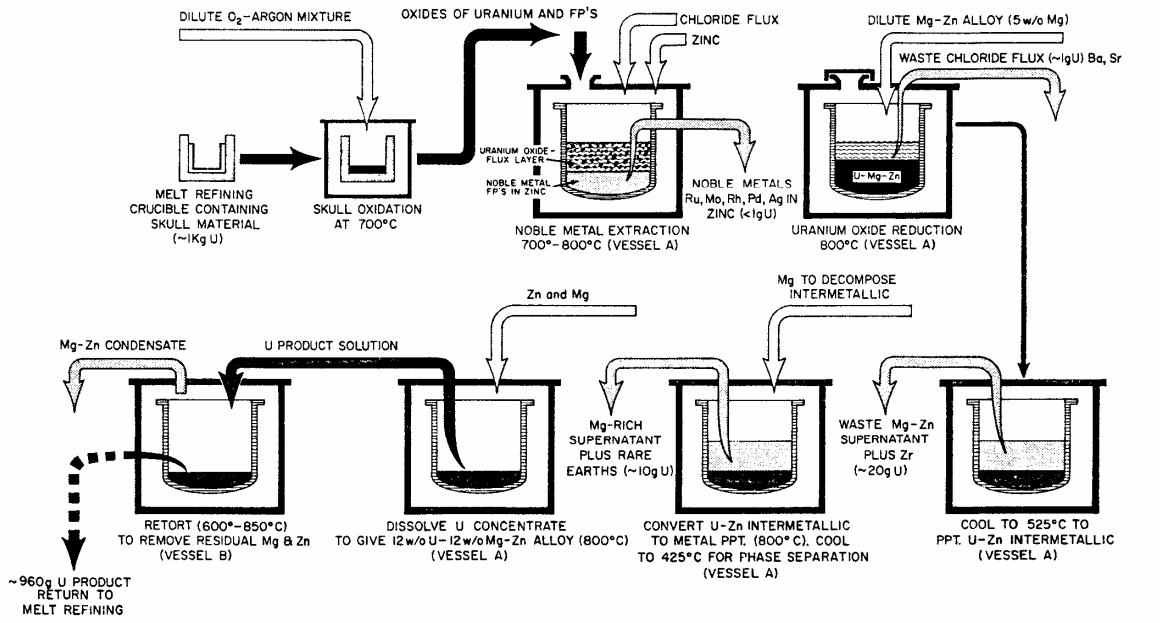
- **Slagging**
  Slagging utilizes the transfer of elements from a metal melt into a separate layer consisting of slag-forming substances added to the molten metal. Often the transfer is controlled by adding reducing material to the metal melt or by blowing oxygen containing gas through the melt. Slagging is in large scale industrial use for non-nuclear metal purification, e.g. in copper and steel production.
- **Two-phase extraction**
  It was early realized that liquid-liquid extraction can be carried out using a molten salt phase and a molten metal phase. It has several similarities with the better known aqueous/organic based liquid-liquid extraction currently in large scale use in reprocessing. In order to control the distribution of elements between the molten salt and the metallic phase two possibilities exist. One can choose one of several possible carrier metals and one can change the redox potential in a given system by adding or removing a strongly reducing component, usually Li-metal. The choice of salt-melt seems to be of secondary importance for the differences in distribution of elements between the two phases at a given temperature.
- **Ion exchange**
  No recent development work on the use of solid ion-exchanging materials for separations of elements from molten salts has been found.
- **Electrolysis (electrorefining, electrowinning)**
  By passing a current through a molten salt it is possible to precipitate many metals on the cathode surface. Because different elements have different deposition potentials it is possible to recover one element, or a group of elements, as an oxidic or a metallic deposit on the cathode. The combination of molten salt and cathode material as well as the physical state of the cathode, solid or molten, set the limiting cathode potential (relative to the melt) for deposition of each element in the melt. Electrolysis of molten salts is in large scale industrial use for production of several metals, e.g. aluminium and magnesium.

3.2.2 Historic development

The knowledge of reprocessing of spent nuclear fuel by non-aqueous processes was probably at its height in the western world in the early 1960ies. One of the major achievements in the late 1950ies was the design and operation of the pyrochemical reprocessing plant as part of the EBR-II experimental breeder reactor project in the US. That plant used controlled oxidative slagging to purify a melt of spent metallic fuel from most of the fission products, a process in many ways similar to one used routinely in the steel industry. The purified molten metal was directly recast into new fuel elements. Typical plutonium losses to wastes were above 5% /Lin 61/. A schematic drawing of the pyrochemical processing plant is shown in Figure 3-8 /Bur 64/.
The lower part of that figure also shows the treatment of skulls from the main pyroprocessing operation made in order to better recover the uranium in the fuel.
A more promising process was fluoride volatilization, which was tested in the MSRE project on molten-salt fuel /Wei 70/ and further developed in the US for use in reprocessing of LWR UO_2 fuel on an industrial scale in the Midwest Fuel Recovery Plant.



Cutaway View of EBR-II Fuel Cycle Processing Facility



EBR-II Liquid Metal Process for Reclamation of Melt Refining Skulls

Figure 3-8. *The pyrochemical fuel processing plant at EBR-II /Bur 64/.*

Difficulties were found to keep process losses very low, to design processes with inherent criticality control, to manage the adverse effects of small amounts of water vapour and oxygen, and to control corrosion. Events during start-up of the Midwest Fuel Recovery Plant, USA, /GE 66; GE 73; GE 74; Rog 77/ and during operation of a

pilot plant in Fontenay aux Roses, France, made it natural in the 1970ies to shut down further research and development of pyrochemical processes for partitioning in the western countries in favour of aqueous separation, which was believed to be safer. However, R&D and applications in the nuclear field still remained in the Soviet Union. The surviving industrial nuclear applications in the West were production of uranium hexafluoride for use in uranium enrichment, production of metallic uranium and plutonium, and routine purification of recycled weapons grade plutonium metal /Kni 81/.

With the passing of time, renewed interest in non-aqueous separations arose, mainly within the Japanese OMEGA project, but also in the US and in EU. However, because the US did later on decide to abstain from civilian recovery and recycling of plutonium, the work in the US on non-aqueous reprocessing has by now almost ceased /Ino 02/.

3.2.3 Review of recent work

Much of the current R&D work funded by the European Union aims at recovery and understanding of the old knowledge and technology. At the same time it is generating a new generation of chemists with knowledge of how to apply high temperature chemistry to reprocessing and separation of elements from nuclear fuel, nuclear waste, and irradiated targets. Within the OMEGA project, CRIEPI, JAERI, and others have worked on the development of such elementary processes and equipment that could perhaps be used to design a reprocessing plant for metallic fast breeder fuel or for partitioning of high level waste from aqueous reprocessing.

Molten salts used

The salt melts studied since our last P&T review are LiF, LiF-NaF, LiF-BeF_2, LiF-CaF_2, LiF-AlF_3, LiF-NaF-KF, and LiCl-KCl. The tendency seems to have been to move away from salt systems containing beryllium, because of the poisonous nature of that element, and also to some degree from fluoride towards chloride melts. Eutectics are often used in order to reduce the melting point of the salt mixture.

Electrode development

Electrodes studied for use as cathodes have been cadmium, bismuth, zinc, and silver as solids or in molten form /Bou 03; Ham 03; Ino 02; Sak 98; Zve 03/. The physical state of the cathode seems to be important. Malmbeck and co-workers at ITU have reported that separation of U and Pu is difficult on molten zinc cathodes from fluoride melts because the deposition potentials are very close, whereas a good separation of U from Pu is possible using solid zinc cathodes in fluoride melts where the deposition potentials are nicely separated /Malm 03/. Similar results for solid and liquid cathodes are given in a summary paper by Inoue /Ino 02/, see Figure 3-9. Hence, a co-processing of fuel using liquid cathodes can probably rather easily be changed to a separate recovery of U and Pu by a change of electrode material or by a change from a liquid to a solid state of the same electrode material. Thus, it is difficult to prove that any molten salt electrowinning process is totally safe against minor modifications which would permit a clandestine recovery of pure plutonium. Many of the systems studied seem to be quite sensitive to the adverse effects of small amounts of oxygen or traces of water vapour, see e.g. /Ham 03/.

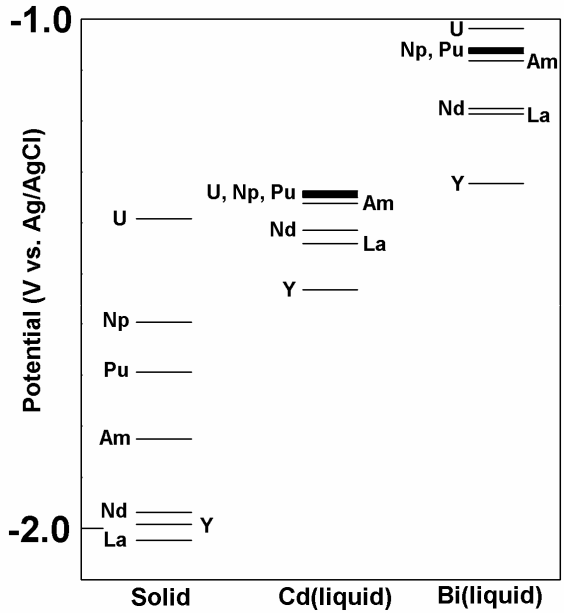


Figure 3-9. *The effect of cathode material on deposition potentials /Ino-02/.*

Liquid-liquid extraction

Distribution constants, K_d-values, for many elements between molten LiCl-KCl and molten Zn(Li) or Bi(Li) have been measured /Mor 98/. The K_d-values are more favourable for a separation between elements when using Bi(Li) than when using Zn(Li) as molten metal phase. However, both lanthanides and actinides have lower K_d-values in the salt-Bi(Li) system than in the salt-Zn(Li) system. These effects are interpreted as due to differences in inter-metal bonding between the carrier metal and the dissolved metals. As a result of these observations, Kinoshita et al tested separation of lanthanides from actinides with multi-stage extraction using the LiCl-KCl/Bi(Li) system /Kin 99/. Their results indicate that a counter current process in principle should permit a separation and recovery of at least 99% of the actinides with a purity acceptable for use as FBR fuel. However, their mass balances indicate a total loss of TRU elements of no less than 10%, perhaps as deposits on equipment walls. Such losses would be quite unacceptable in a real P&T process, where the total recovery of many actinides need to be at least 99.99%.

Process development

Few new variants of reprocessing/separation schemes have been proposed since our last P&T status report. One complex, but also rather complete, flow sheet has been suggested by CRIEPI for recovery of minor actinides from HLLW, see Figure 3-10 /Ino 02/. Most of the steps in this flow-sheet seem to be under development within the OMEGA project, but it is unclear how far this work has proceeded.

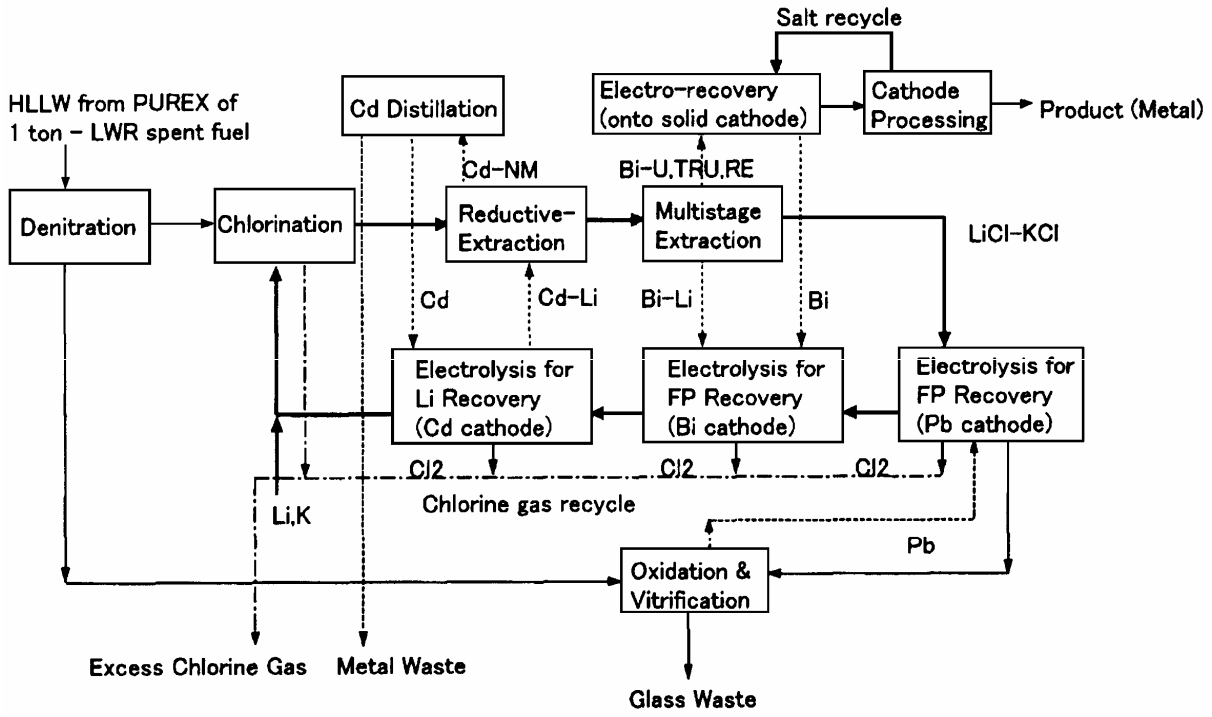


Figure 3-10. *Suggested flow-sheet for partitioning of HLLW /Ino 02/.*

Equipment

Small scale equipment for emf-measurements of redox potentials in melts has been designed and used /Bou 03; Ham 03; Sak 98; Zve 03/. Single-stage liquid-liquid extraction equipment in laboratory scale has been developed and used /Bou 03; Ino 02; Kin 99; Mor 97; Mor 98; Sak 98/. A few types of new electrolysers for electrowinning of actinides have been developed and tested /Bou 03/. However, no equipment suitable for large scale use in a reprocessing/separation plant seems to have been developed and tested /Ino 02/. Hence, there is quite a large technical development and testing of equipment needed before non-aqueous processes are ready for consideration for industrial applications in the nuclear field.

4 Current research concerning transmutation

As mentioned in the introduction, the objective of transmutation in the present context is to transmute long-lived radionuclides with neutrons into nuclides that are stable or have shorter half-lives. The main nuclides of interest for transmutation are the transuranium elements. The most effective nuclear reaction for this is nuclear fission. Capture reactions will only lead to transmutation from one long-lived nuclide to another via decay of some interim short-lived nuclides. Many of the transuranium nuclides are only fissionable in fast neutron spectra for example the plutonium isotopes with even atomic mass number. This means that for an effective transmutation a strong fast neutron flux is needed.

Two types of facilities are considered for creating such a strong neutron flux – a fast reactor with a self-sustaining chainreaction and accelerator-driven subcritical reactor system (ADS). Several sodium-cooled (self-sustaining) nuclear reactors were built in the time frame 1950–1985. The largest was Superphenix in France, an electricity producing plant with 1200 MW_e power. Only a few of these systems are, however, still in operation. In the early 1990ies the interest for ADS started to increase and currently there is a relatively strong interest to explore such systems. The transmutation research in Sweden is almost exclusively focused on ADS-systems. This overview therefore starts with an account of current research on ADS and then continues with some comments on fast reactors and other conceivable options.

4.1 Accelerator driven systems

Accelerator-driven systems consist of the following major components:

- A proton accelerator to give a strong current (from a few up to several tens of mA) of protons with about 1 GeV/proton energy.
- A spallation source that is hit by the proton current and where spallation reactions with heavy nuclides create a neutron flux of several tens of neutrons per proton.
- A subcritical reactor with fuel that contains the long-lived radionuclides to be transmuted by the neutrons. Due to the fission of these nuclides new neutrons are formed and for each incoming neutron from the source about 20 new neutrons are obtained and cause additional fission reactions. The design of the reactor shall be such that a self-sustaining chainreaction cannot occur.
- Balance of plant systems to take care of the energy released by the fission processes and by radioactive decay and to supply electricity to the accelerator.
- Equipment to control and monitor the entire process and its safety systems.

The current research is mainly focused on the three first mentioned parts of an ADS-system

4.1.1 Accelerators

Choice of the basic accelerator concept

The present state-of-the-art in accelerator technology shows that only two basic accelerator concepts of are able to deliver beam intensities in the mA range – sector-focused cyclotrons and linear accelerators (linacs).

For cyclotrons, proton energies of 600 MeV are well established through e.g. the experience with the machine at PSI[50] in Switzerland. From this, it is also considered in the cyclotron community that a beam intensity of about 5 mA should be safely reachable. However, extrapolating up to 10 mA is more questionable, and might require a complex of at least two cyclotrons with the two beams being funnelled together. A given cyclotron cannot be expanded in energy. Boosting the energy from 600 to 800 MeV would thus require full replacement of the final and main stage or installation of a post-accelerator, a not cost effective operation. Energies in the GeV range with currents at or above 10 mA (industrial transmuter) are beyond the practical working limit of a cyclotron. Furthermore, a cyclotron is basically a CW[51] machine and the requirement to provide pulses for neutronics measurements is a major difficulty for a cyclotron of such power. None of all these limitations are present in a linac where intensities can reach above 100 mA without an intrinsic energy limit.

The strategy to achieve good reliability relies on over-design, redundancy and fault-tolerance. This approach requires a highly modular system where the individual components are operated substantially below their performance limit. A superconducting linac, with its many repetitive accelerating sections grouped in "cryomodules", conceptually meets this reliability strategy. It further allows keeping the activation of the structures rather low, which is important for radiation protection and maintenance, whereas the extraction channel of high power cyclotrons in this respect is of a considerable concern.

For these reasons, it has been concluded that the cyclotron solution for an XADS presents a number of difficulties if not impossibilities: funnelling, pulsing, beam trips, double-machine scheme, intrinsic current limitation, energy upgrading that precludes this solution despite its advantages such as lower price, proven technology at the MW level as demonstrated by PSI, and compactness. Therefore, the reference design is a superconducting linac.

However, it should be pointed out that cyclotrons of the PSI type should be considered as the natural and cost-effective choice for preliminary low-power experiments, where availability and reliability requirements are less stringent. CW linear accelerators must be chosen for demonstrators and full-scale plants, because of their potential in terms of availability, reliability and power upgrading.

Accelerator specifications for an experimental ADS (XADS)

Studies performed in the frame of the Technical Working Group on ADS (see Section 2.2) and then continued in the European Project PDS-XADS (see Section 2.4.2) have determined the main technical specifications for the XADS accelerator – see Table 4-1.

[50] PSI = Paul Scherrer Institute, Swiss national research laboratory.

[51] CW – Continuous Wave; this term was originally describing the Radio Frequency (RF) providing the energy (acceleration) to the beam in a linear accelerator. The term is commonly used as synonym for a 100% duty cycle particle accelerator.

Table 4-1. XADS proton beam specifications.

Max beam intensity	6 mA CW on target (10 mA rated)
Proton energy	600 MeV (includes 800 MeV upgrade study)
Beam entry	Vertically from above preferred
Beam trip frequency	Less than 5 per year (exceeding 1 second)
Beam stability	Energy: ± 1%, Intensity: ± 2%, Size: ± 10%
Beam footprint on target	Gas-cooled XADS: circular ∅ 160 mm LBE-cooled XADS: rectangular 10 × 80 mm^2 MYRRHA: circular, "donut" ∅ 72 m
Intensity modulation	0.2 ms "holes" in CW beam for neutronics measurements, repetition frequency 0.01–1 Hz

The XADS accelerator belongs to the category of the so-called HPPA (high-power proton accelerators). HPPA are presently very actively studied (or even under construction) for a broad use in fundamental or applied science. The overall performance of the subcritical system will be critically determined by a strict adherence of the XADS accelerator to its specifications. Compared to other HPPA, many requirements are similar, but it is to be noted that the reliability specification, i.e. the number of unwanted "beam-trips", is rather specific to the use as driver for an ADS. Therefore, the ADS accelerator studies for the reference design had to integrate this stringent requirement from the very beginning, taking into account that this issue could be a potential "show-stopper" for ADS technology in general.

The XADS accelerator reference layout

A reliable linac

The proposed reference design for the XADS accelerator, optimized for reliability, is shown in Figure 4-1[52]. It is composed of a "classical" proton injector source (ECR[53]) + a normal conducting RFQ[54] structure). Additional warm IH-DTL[55] or/and superconducting CH-DTL[56] structures are used up to a transition energy. At this point a fully modular superconducting linac accelerates the beam up to the final energy.

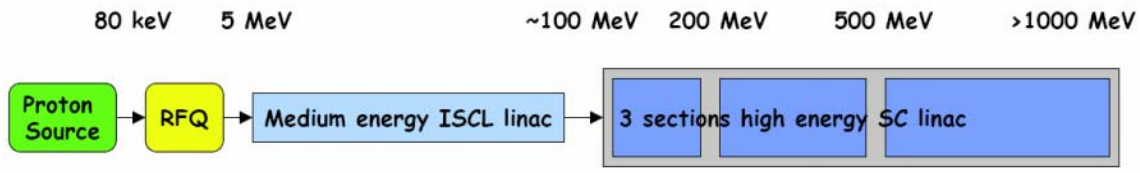


Figure 4-1. *A very schematic layout of a linear accelerator for ADS.*

[52] ISCL = Independently phased Supercoducting Cavity Linac.
[53] ECR = Electron Cyclotron Resonance, one of the most efficient ionization methods currently used in high intensity ion sources.
[54] RFQ = Radio Frequency Quadrupole, a low-energy linear structure which simultaneously assures bunching, focusing and accelerating and thus overcomes space charge effects.
[55] DTL = Drift Tube LINAC, a structure in which the particles are accelerated in the gap between two consecutive drift tubes. Focusing can be made inside the drift tubes. Reliable and well proven design. Since the tubes have to increase length with the particle energy so that the phase in the accelerating gap is kept constant, a DTL becomes inefficient at high energy. IH DTL = Interdigital H-mode Drift Tube Linac, where Interdigital H stands for a geometrical arrangement of accelerating modules.
[56] CH-DTL = Crossbar H-mode Drift Tube Linac, where, Crossbar H-mode stands for a geometrical arrangement of accelerating modules.

Up to the transition energy, fault-tolerance is guaranteed by means of a "hot stand-by" spare. Above this energy, "spoke" and, from 100 MeV on, "elliptical" cavities are used. Beam dynamics calculations for this part have shown that an individual cavity failure can be handled at all stages without loss of the beam. Besides this fault-tolerance, another remarkable feature of the concept is its validity for a very different output energy range: 350 MeV for the smaller-scale XADS require 9 cryomodules of β=0.65 elliptical cavities; in order to obtain 600 MeV, simply 10 more cryomodules have to be added (7 with β=0.65 and 3 with β=0.85) and 12 additional (β=0.85) boost the energy to 1 GeV. Therefore, already the small-scale XADS accelerator provides full demonstration not only of the 600 MeV XADS (and could be converted to it), but even for an industrial scale machine.

A superconducting linear accelerator can fulfil a priori the specifications for the XADS /Mueller 03/. This linac can be used for all different versions of XADS and it is also representative for an industrial machine. The proposed machine is reliable through the rigorous implementation of a highly modular system with derated components operated in a fault-tolerant way.

Japanese accelerator studies

In the beginning of the 2000ies, the accelerator work within the OMEGA project was merged into the Neutron Science Project, which in its turn became a part of a large project called J-PARC[57]. The purpose of the J-PARC is to pursue advanced science and nuclear technology including nuclear transmutation. Figure 4-2 shows the schematic layout of the J-PARC. The facility is being constructed at the JAERI/Tokai site. The accelerator complex for the Joint Project consists of a 600-MeV linac, a 3-GeV RCS[58] and a 50-GeV synchrotron. The linac plays two roles; one is to inject the beam to the RCS, and the other is to provide the beam to an accelerator-driven nuclear waste transmutation system – ADS. The high-energy part of the 600-MeV linac uses superconducting (SC) cavities, which can be a prototype of the future CW accelerator for the ADS applications. The 3-GeV, 1-MW beam is provided to the pulsed spallation neutron experiment. It is also used for muon science. The 50-GeV beam is used for particle and nuclear physics. It is also extracted for neutrino experiments, which are conducted at the SUPERKAMIOKANDE detector located 300 km from the Tokai site. In this way, the high-intensity proton accelerators will intensively and efficiently promote a wide variety of science and engineering fields. The facility includes upgradeability to a 5-MW neutron source, which is allocated to the second phase of the project.

As shown on Figure 4-2 the 600 MeV proton beam from the linear accelerator will be introduced into the Transmutation Experimental Facility (TEF).

Superconducting linear accelerator technology

The proton accelerator developed for the ADS should be of high intensity, more than 20 MW, with good economical efficiency and reliability. Taking this into account, JAERI regards the superconducting linear accelerator (SCL) as the most promising choice.

The SCL consists of a series of cryomodules, which contain two units of super-conducting cavities made of high-purity niobium. JAERI and KEK (High Energy

[57] J-PARC = Japanese Proton Accelerator Complex.
[58] RCS = Rapid Cycling Synchrotron.

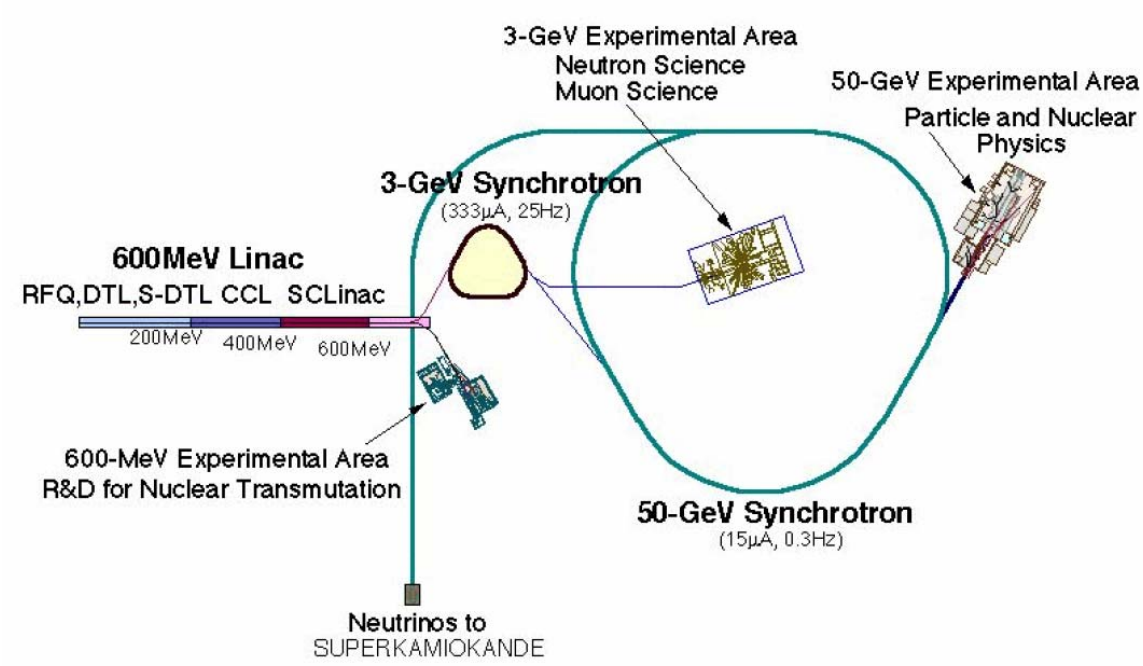


Figure 4-2. *Schematic view of the Joint Project accelerator complex.*

Accelerator Research Organization) co-operate on building a prototype cryomodule and examining its performance in electric field and helium cooling.

4.1.2 Spallation source

A spallation source is a key component of an Accelerator-Driven System − ADS. Spallation is a generic expression for certain types of nuclear reactions that occur when energetic particles (e.g. protons, deuterons, neutrons, pions, muons, etc) interact with an atomic nucleus − the target nucleus. In this context, "energetic" means kinetic energies larger than about 300 MeV per nucleon. The initial collision between the incident projectile and the target nucleus leads to a series of direct reactions (intranuclear cascade) whereby individual nucleons or small groups of nucleons are ejected from the nucleus. At energies above a few GeV per nucleon, fragmentation of the nucleus can also occur. After the intranuclear cascade phase of the reaction, the nucleus is left in an excited state. It subsequently reaches a ground state by "evaporating" nucleons, mostly neutrons. In short, spallation can be described as a nuclear reaction in which the energy of the injected particle is so high that more than two or three particles are ejected from the target nucleus and both its mass number and atomic number is changed.

The spallation process occurs in two stages, intranuclear cascade and evaporation. For thick targets, high-energy (above 20 MeV) secondary particles (plus their progeny) can undergo further spallation reactions. For some target materials, low-energy (below 20 MeV) spallation neutrons (i.e. the cascade-evaporation neutrons) can enhance neutron production through low-energy (n,xn) reactions. For heavier nuclei, high-energy fission can compete with evaporation in a highly excited nucleus. Tantalum, tungsten, and lead are examples of materials that can undergo an effective spallation/ high-energy fission.

A spallation neutron source is a very useful and attractive neutron source for many scientific purposes like neutron scattering research and condensed matter investigations.

Spallation neutrons for neutron scattering purposes have been very successfully and routinely used at some accelerator centres, like the Paul Scherer Institute in Switzerland, the Rutherford Laboratory in United Kingdom, the Los Alamos National Laboratory etc. A very new generation of the neutron spallation sources are now being constructed in USA – Spallation Neutron Source (SNS) in the Oak Ridge National Laboratory and in Japan – JPARC-project.

The spallation source or "target unit" is one of the most innovative and demanding components of the ADS. It constitutes the physical and functional interface between the accelerator and the subcritical reactor. It is simultaneously subject to severe thermal-mechanical loads and damage due to high-energy heavy particles. The spallation module design should be based on a balanced optimisation between neutronic efficiency, material properties (physical, chemical) and thermal-hydraulic performances under the conditions imposed by safety, reliability and lifetime.

Different concepts are presently under investigation:

a) Liquid metal target.
   i. Target with window.
   ii Windowless target.
b) Solid target of metal with high density and very good heat conduction properties (tungsten, tantalum).

For the liquid metal only two choices have been seriously considered: mercury and liquid lead-bismuth eutectic. While the neutron scattering community decided of a various reasons for the mercury source, the ADS community considers liquid lead-bismuth (LBE) as the primary choice. The advantages of LBE are chemical inertia, high boiling temperature, relatively low melting temperature (123.5°C), good heat conductivity and no immediate volume expansion upon solidification. A disadvantage of the LBE is generation of ^{210}Po, a short-lived hazardous alpha emitter formed by neutron irradiation of bismuth.

For the coupling between the accelerator and the spallation source there are two basic options. The acceleration of the proton beam must take place in very high vacuum (1 μPa), while a hot liquid spallation target (source) obviously will ruin any vacuum due to evaporation. Therefore, the accelerator high vacuum and the target must be separated by a window. If this window is in direct contact or close proximity with the spallation liquid, it is often denoted a "window target solution" or "hot window". There are wide-spread concerns about the durability of such a window due to the very harsh environment it has to sustain. If such a window breaks, it might cause severe damage to a large part of the entire system. Therefore, so called "windowless" or "cold window" solutions have been proposed, in which the accelerator high vacuum is terminated by a cold window at a distance before the spallation target, and the spallation liquid has a free surface. The pressure above the liquid Pb-Bi surface in this case is of the order 1–10 mPa. In both cases, the main problem is not that the global balance for heat removal cannot be fulfilled for reasonable flow parameters, but that the creation of hot spots near flow stagnation points could either jeopardize the window or produce localized boiling which would deteriorate the (low) vacuum conditions at the free surface.

Present development projects for window spallation targets at power levels of around ~ 1 MW power (MEGAPIE, Russian target – ISTC 559) /Fazio 03; Dedoul/ try therefore to cool the window in a cross flow, thereby avoiding the risk of stagnation points in an axis-symmetric arrangement. This option may represent a difficult

challenge for extrapolation to larger target volumes in an even more crowded environment. The design of an adequate window to separate the forced flow of several bars from the vacuum of the proton beam line is one of the major challenges of the ADS design.

A windowless design development, also in the ~ 1 MW power range (such as planned for the MYRRHA development project) /Abderrahim 03/ can avoid the above difficulties. In this case, however, new difficulties are introduced. The free surface, being the beam vacuum interface, has to fulfil the requirements that metal and spallation product evaporation and out-gassing do not unsuitably increase the pressure in the few metres of the beam line adjacent to the subcritical core. Indeed no effective vacuum pump can be installed in this region of the core. A too high particle density would eventually lead to thermal loading of the beam line from secondary plasma formation with run-away character and even beam blocking. The difference to a solid interface beam line is the access to particle reservoirs potentially orders of magnitude higher than for solid interfaces.

A first prototype of an ADS 1 MW liquid Pb-Bi spallation target has been manufactured by the Institute of Physics and Power Engineering (Obninsk, Russia) in collaboration with KTH and Los Alamos National Laboratory – see Figure 4-3 /Dedoul/. The original goal of this project, funded by the International Science and Technology Centre, was to test this spallation target at the LAMPF accelerator in Los Alamos. Unfortunately, due to funding problems and changing priorities in USA, this target (which is now at the University of Nevada, Las Vegas) still waits for its irradiation experiment. In spite of still lacking on-beam experiences, this project has already had an impact on the development of Pb-Bi technology in Europe through the disclosing of proprietary and/or military-classified know-how about the corrosion control technology for liquid lead-bismuth.

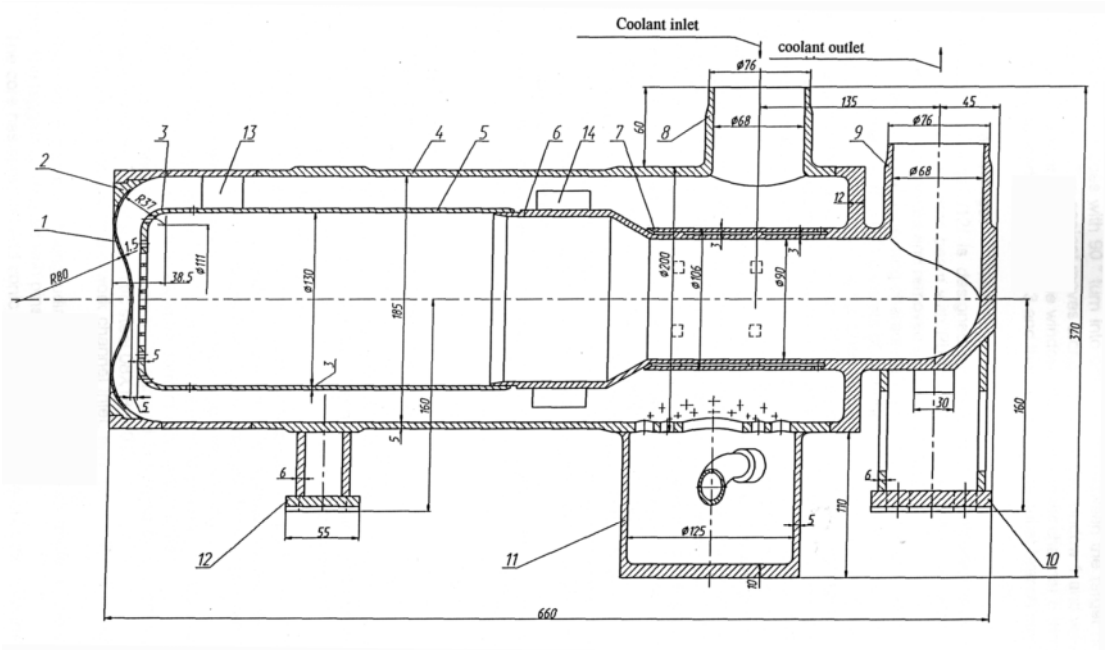


Figure 4-3. *A 1 MW Pb-Bi spallation target manufactured in Russia. Main components of the target: 1 – window, 2 – window support, 3 – diffuser plate, 4 – target hull, 5 – inner channel.*

So far two European projects have been established in order to customize the Pb-Bi technology for ADS development. These are at present the two forerunners in Europe for development of intense spallation sources:

- In the MEGAPIE project, carried out by a partnership of laboratories, a Pb-Bi target will be irradiated to create a pure spallation source by the SINQ proton beam of PSI at 590 MeV and 1.8 mA. The design uses a window for which the highest irradiation dose will be of the order of 10 DPA[59] in the 6 months of the actual experiment planned for coming years. The MEGAPIE spallation source should start operation in March 2005 and deliver neutrons for a neutron research community until the end of 2005.
- In the MYRRHA project, a small ADS facility for irradiation purposes. It is under design at the SCK·CEN. A windowless Pb-Bi spallation source design has been chosen to cope with the geometrical constraints to achieve high performance of the subcritical core. A 350 MeV, 5 mA proton beam will generate the neutrons in the target subjected to a current density around 150 $\mu A/cm^2$. Experiments to verify the physical and thermal-hydraulics behaviour of the windowless configuration are already being performed in water and planned in LBE. The MYRRHA facility is planned to be operational in 2008–2010.

4.1.3 Subcritical reactor

The use of accelerator-driven subcritical reactors was originally suggested for transmutation of higher actinides (americium and curium), as the introduction of these elements in high concentration into the fuel of critical systems would yield unacceptably small safety margins /Foster 74/. For the same reason, the Double Strata fuel cycle suggested by JAERI and refined by CEA, employs accelerator driven systems (ADS) for management of minor actinides /Murata 84; Takizuka 89; Salvatores 98/.

Attempts have also been made to design ADSs for the purpose of transmuting the entire transuranium stream from spent LWR fuel, typically a mixture of 80–90% of Pu with 10–20% minor actinides /Rubbia 97; Wallenius 01; Yang 01; Yoo 02/. The motivation for such a "two-component" strategy is mainly the lack in particular countries of developed and/or accepted strategies for plutonium recycling in existing reactors. The fraction of power produced in nuclear facilities dedicated to waste management (~ 20%) would also be smaller than in a LWR plus fast reactor strategy /NEA 02b/. The associated cost penalty, however, turns out to be comparatively large.
See Section 5.5.

Accordingly, the focus of ADS development in the European Community is for deployment as burner of higher actinides and low-quality plutonium. Plutonium from spent LWR fuel would then be multi-recycled in critical reactors. A possible European Double Strata power park is displayed in Figure 4-4.

[59] DPA = displacements per atom.

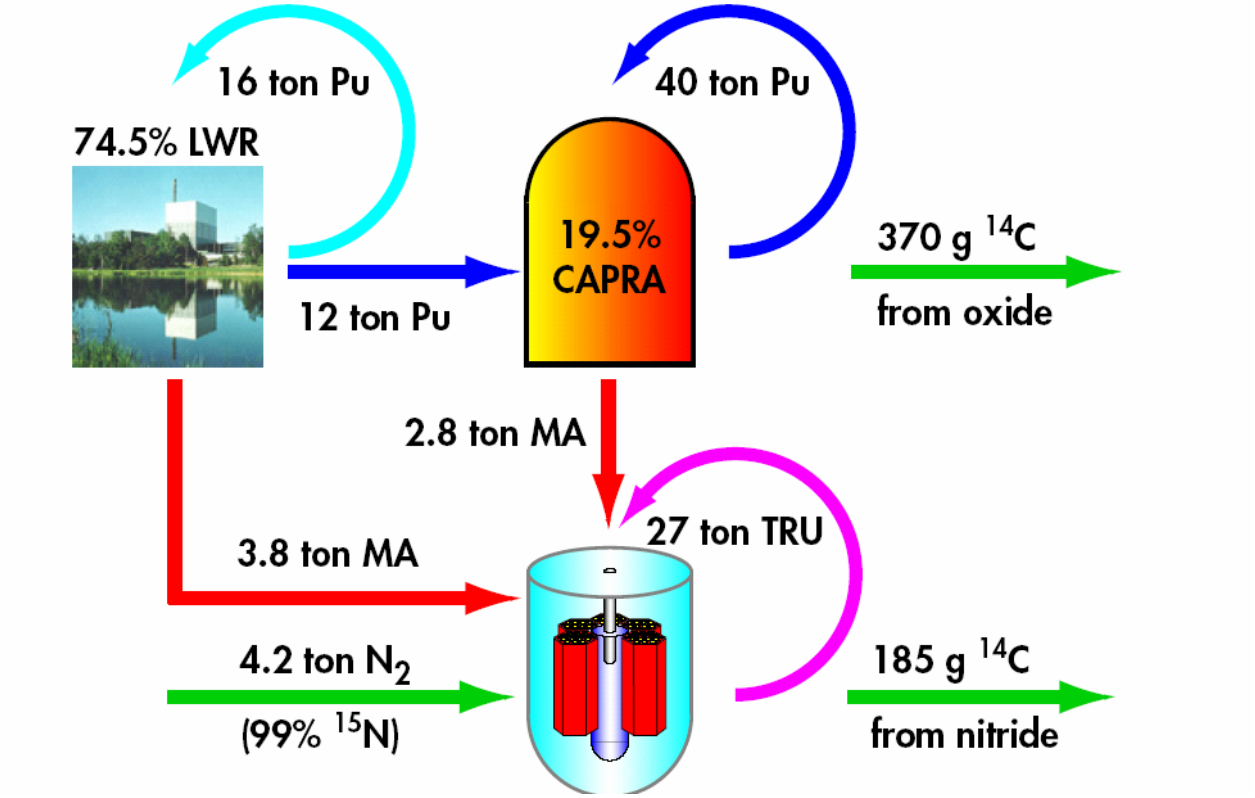


Figure 4-4. *Double strata fuel cycle including LWR, FR and ADS. (CAPRA is a special designed fast reactor, FR, for Pu-burning.)*

While the EU funded project PDS-XADS mainly concerned the design of an experimental ADS with standard MOX fuel, the fuel development projects CONFIRM and FUTURE have included design and safety studies of an 800 MW_{th} ADS with higher actinide based fuel. The core power was chosen as a function of the minimum permissible subcriticality level (max $k_{eff} = 0.97$) and spallation target power (30 MW_{th}). As scoping studies showed that sodium coolant could not provide acceptable safety margins for a core of this size /Wallenius 03a/, lead-bismuth eutectic (LBE) was selected as reference coolant. Concerning the fuel, a feed stream of 40% plutonium, 50% americium and 10% curium was adopted. In the first irradiation cycles the Pu would come from spent LWR-MOX fuel, while later, it is substituted with the Pu present in spent ADS fuel containing a large fraction of the heavier non-fissile isotopes. Keeping the Pu fraction of 40% constant, the reactivity losses during irradiation can be minimised, and do not exceed 1700 pcm[60] even for a burnup of 20% /Smith 03/. Hence, no fuel shuffling will be needed, simplifying fuel management considerably.

In the scoping studies a series of potential inert matrices for the fuel was investigated /Wallenius 03a/. Thorium and zirconium oxide could be discarded for reasons of poor reprocessability combined with poor safety performance during loss of coolant accidents. Metallic matrices like tungsten and natural molybdenum have high melting temperature but too high absorption cross sections. Chromium featured outstanding neutronic characteristics, but turned out to form a low melting eutectic with lanthanide oxides, /Fernandez 03/. Three candidates were identified that fulfilled minimum

[60] pcm = pro cent mille, i.e 10^{-5} (change in k_{eff}).

requirements concerning fabricability, reprocessability, thermal conductivity and neutronic safety, being magnesium oxide, molybdenum-92 metal and zirconium nitride. The two former would be a matrix of actinide oxide dispersions, while the latter would be in solid solution with actinide nitride.

Linear ratings of these fuels were set as a function of their thermal conductivity and melting temperature. For the detailed design study, 25 kW/m was assumed for the CERCER (MgO), 30 kW/m for the CERMET (^{92}Mo) and 35 kW/m for the nitride.

The pin diameter was determined as a compromise between the wish for a high inert matrix fraction and a low residence time. The latter condition is due to the corrosion rate of the steel cladding in LBE, which is known to be 20–40 microns per year at T = 470°C /Barbier 01/. In order to reach at least 20% burnup during a residence time of less than three years, the pellet diameter should thus be as small as possible. On the other hand, for reasons of fabricability, the matrix fraction must be at least 50 volume percent. Consequently, the pin diameter was chosen as the smallest diameter allowing for a 50% volume fraction of inert matrix in the fuel. For the oxide fuels this corresponds to an inner/outer clad diameter of 6.0/6.8 mm, while for nitride fuel smaller values of 5.0/5.7 mm are possible.

The pin pitch was finally determined from transient calculations of unprotected loss of flow accidents. Figure 4-5 displays the clad temperature for P/D = 1.5[61]. As can be seen, the clad temperature exceeds 1 330 K after a few tens of seconds, if the accelerator cannot be shutdown. Accordingly, P/D = 1.60 was selected for the oxide fuels, and P/D = 1.75 for the nitride /Eriksson 04/.

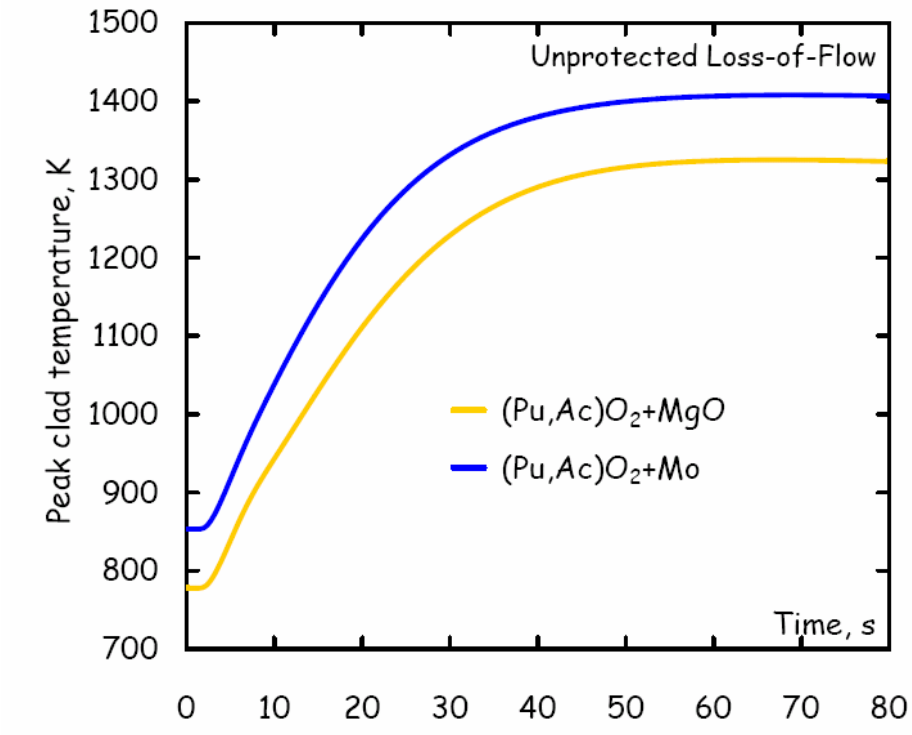


Figure 4-5. *Clad temperature following unprotected loss of flow accident.*

[61] P/D = pitch to diameter ratio (pitch is centre to centre distance between individual fuel rods).

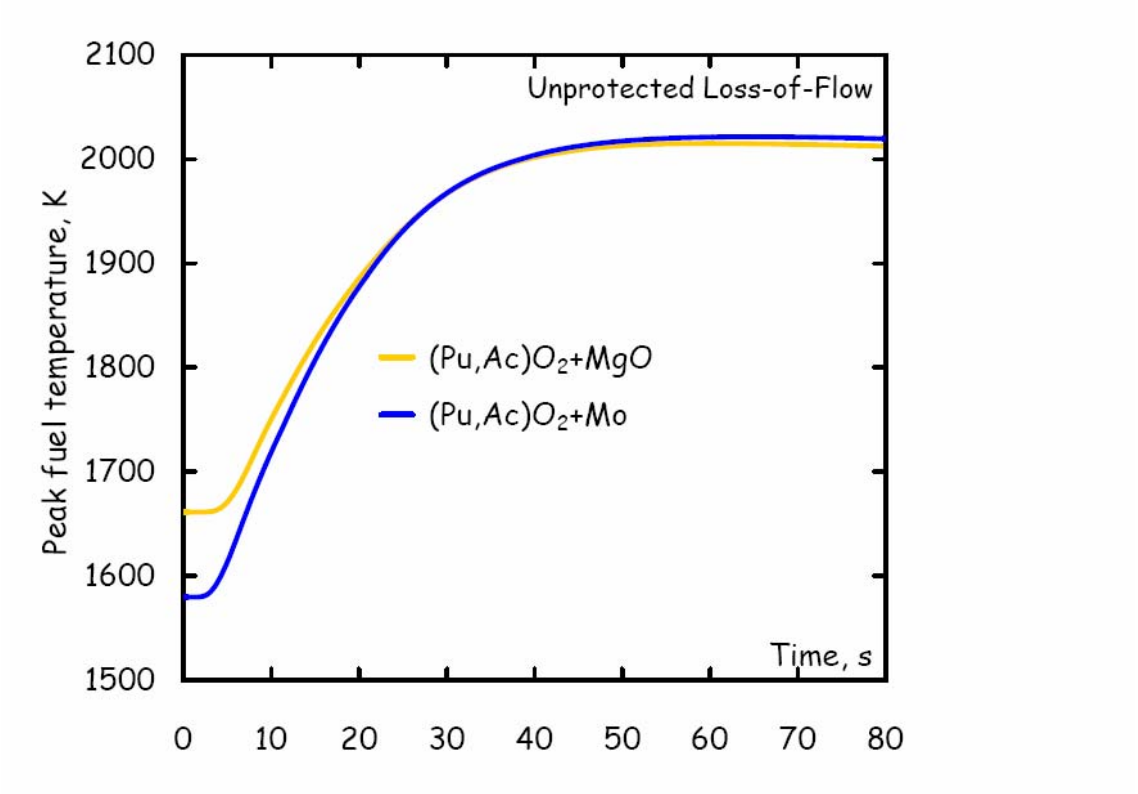


Figure 4-6. *Fuel temperature following unprotected loss of flow accident.*

Concerning the fuel, accidental insertion of the full accelerator current at BOL[62] turns out to be the limiting event. Figure 4-6 indicates that these fuels would survive a 50% overpower, with a margin to melt of at least 100 Kelvin for the CERCER fuel, and 500 Kelvin for the CERMET /Eriksson 04/.

Reference designs for an 800 MW_{th} subcritical core with three different fuels are thus now available within the European program. Further tests of fabricability, high temperature stability and irradiation performance of these fuels will lead to the selection of one of them as primary candidate for the ~ 300 MW_{th} ADS prototype with minor actinide based fuel to be designed within the sixth FWP.

For further information and discussion of the subcritical reactor see Section 5.3 on ADS demonstration plant.

4.1.4 Fuel

The European research program on fuels for transmutation has within the last few years been focusing on uranium-free fuels for burning of plutonium and minor actinides. Two major paths have been followed:

I. Inert matrix composite oxide fuels and solid solution nitride fuels for multi-recycling of minor actinides and dirty plutonium[63] in accelerator driven systems, in accordance with the Double Strata scenario.

[62] BOL = Beginning of life; EOL = End of life; "life" is fuel life in reactor; often also BOC and EOC respectively indicating start and end of cycle between two refuelling operations.
[63] "Dirty plutonium" = plutonium with a mix of isotopes containing a large fraction of even-number isotopes (238,240,242). Multiple recycling will result in such plutonium composition.

II. Inert matrix composite oxide targets for once-through irradiation of americium in thermalized assemblies residing in fast neutron reactors.

The uranium free approach is justified not only in order to minimise production of plutonium. As can be seen in Tables 4-2 and 4-3 the presence of americium in the fuel is strongly detrimental for both the Doppler coefficient as well as the effective delayed neutron fraction[64]. Two of the major advantages pertaining to the use of U-238 as matrix for fissile material are thus absent for americium bearing fuels.

Table 4-2. Doppler coefficient in a sodium cooled pin lattice as function of Am concentration /Wallenius 04/. Note that the temperature feedback of the fuel vanishes when Am is introduced into the fuel, while U-238 is not required to obtain a sizeable feedback when americium is absent.

U	Th	Zr	Pu	Am	Δk/ΔT [pcm/K]
0.80	-	-	0.20	-	- 0.52
0.70	-	-	0.20	0.10	- 0.18
0.60	-	-	0.20	0.20	- 0.05
-	0.80	-	0.20	-	- 0.42
-	0.70	-	0.20	0.10	- 0.07
-	-	0.80	0.20	-	- 0.39
-	-	0.70	0.20	0.10	- 0.01

Table 4-3. β_{eff} as function of U-238 and Am concentration /Wallenius 04/. The introduction of americium lowers the ratio β_{eff}/β significantly, even when U-238 is present.

U	Zr	Pu	Am	β [pcm]	β_{eff} [pcm]
0.80	-	0.20	-	470	390
0.60	-	0.20	0.20	400	270
-	0.80	0.20	-	350	300
-	-	0.40	0.60	320	170

[64] Fraction of delayed neutrons to total neutrons from fission = β.
Relative probability that a fission will be caused by a delayed neutron = β_{eff}.

ADS fuel development

In the Double Strata Scenario /Murata 84; Salvatores 98/, the americium and curium produced in critical power reactors is supposed to be multi-recycled in dedicated minor actinide burners. An advantage of this approach is that handling of these highly active elements is constrained to a very small part of the nuclear power park. Calculations show that the fraction of power produced in reactors managing minor actinides could be as low as 4–6%. Performing multi-recycling of Pu either in fast neutron burner reactors /Languille 95/ or in innovative LWR MOX assemblies like CORAIL /Youinou 01/, the cost penalty for performing P&T can be minimised. The poor reactivity coefficients of MA based fuel, in conjunction with a very small effective fraction of delayed neutrons, however, makes subcritical operation of such cores imperative.

In the European Union 5th Framework Program, the CONFIRM and FUTURE projects have focused on developing inert matrix fuels for accelerator driven systems. CONFIRM is coordinated by the department of Nuclear and Reactor Physics at KTH, and includes modelling, safety analysis, fabrication, property measurements and irradiation of nitride fuels. A major advantage of solid solution nitride fuel is its high thermal conductivity, being a factor of 5–10 larger than for corresponding oxide fuels. Further, the dissolution of PuN and the reference matrix ZrN in nitric acid is much easier than for plutonium and zirconium oxides. Nitrides are however more complex to fabricate, and may have problems with high temperature stability.

AEA-T in the UK has performed pressurised high temperature tests of UN and (U,Zr)N. While the uranium nitride samples dissociated into uranium metal and nitrogen gas at $T \approx 2680$ K, the (U,Zr)N samples remained intact up to their melting point. Solidus and liquidus temperatures were measured to be 2880 K and 3030 K, respectively /Thetford 03/. From these numbers, it can be inferred that (Pu,Zr)N should remain stable up to 2800 K.

PSI in Switzerland started fabrication of (Pu,Zr)N pellets in 2002, using carbo-thermic nitridation of oxide powders. Here, carbon is added to the oxide, in order to form CO during heating in nitrogen gas. Using the standard procedure for removal of excess carbon, it however turned out to be difficult to obtain pure materials, i.e. residual oxygen and carbon levels below one weight percent. Modelling made at KTH indicated that switching gas phase during the decarburisation stage from Ar-H_2 to N_2-H_2 would suppress formation of ZrC and hence improve the result /Jolkkonen 03/. In 2003, PSI changed the fabrication route accordingly, and was able to obtain nitride powder with significantly lower impurity levels. After pressing and sintering at 2000 K, a density of 80% of the theoretical maximum was achieved. Figure 4-7 shows ($Pu_{0.2}$,$Zr_{0.8}$) pellets that have been ground to a diameter of 5.55 mm. Four (Pu,Zr)N pins, two with 20 molar percent Pu and two pins with 30 molar percent Pu, have been fabricated by PSI and were delivered to Studsvik in December 2003. These four pins will be irradiated in a hafnium-shielded rig in Studsvik's R2 reactor starting 2004. The irradiation has been modelled by Serco Assurance with the fuel performance code NITRAF, indicating that fuel temperatures at the planned linear power (45 kW/m initially, rising to 55 kW/m half through the irradiation) will not exceed 2300 K. By end of 2005, a burnup of 12% fission in actinides will be achieved.

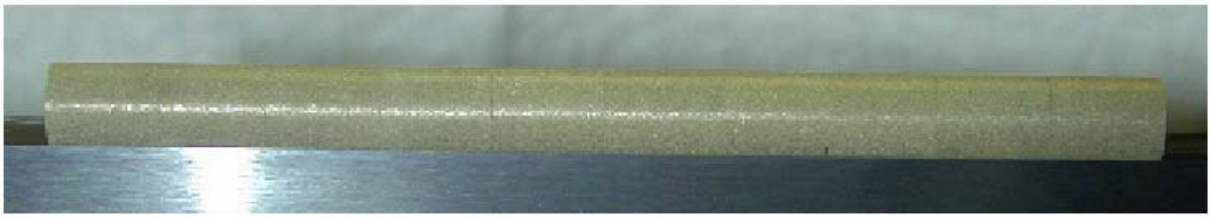


Figure 4-7. *($Pu_{0.2}Zr_{0.8}$) pellets fabricated by PSI. Pellet diameter 5.55 mm. The pellets will be irradiated in Studsvik starting in 2004, with a target burnup of 12% fission in actinides.*

The thermal diffusivity and heat capacity for (Pu,Zr)N samples with high impurity levels have been measured by CEA in Cadarache. Figure 4-8 displays the thermal conductivity inferred from those measurements. It is encouraging to note that the obtained conductivity was even slightly higher than previous theoretical assessments. The values may be compared to the conductivity of $(Pu,Zr)O_2$, being about 2 W/m*K. Note further that the pellets remained stable (under one bar of nitrogen) up to 2400 K.

When the Minor Actinide lab opens for operation in early 2004, ITU will fabricate (Am,Zr)N using carbothermic nitridation of oxide microspheres obtained with the Sol-Gel method. The high temperature stability of AmN remains a question mark. In Los Alamos as well as in JAERI, samples of AmN were evaporating quickly when performing sintering under inert gas, at temperatures exceeding 1600 K.

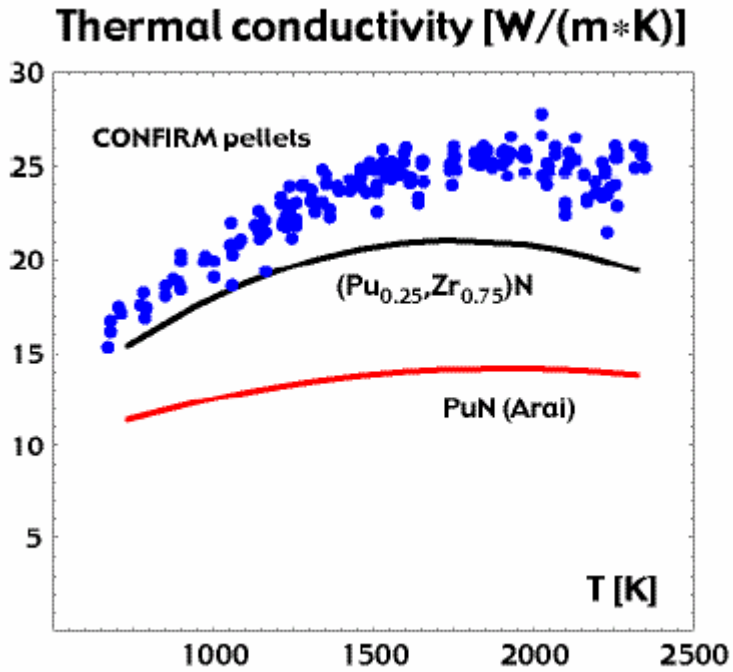


Figure 4-8. *Thermal conductivity of ($Pu_{0.25}Zr_{0.75}$) pellets with high impurity levels of oxygen and carbon (~ 1 weight percent each), measured by CEA.*

Modelling made by KTH has shown, however, that AmN should remain stable under 1 bar of nitrogen up to T = 1800 K /Jolkkonen 03/. Recent data from JAERI confirm that losses of Am during sintering under nitrogen at this temperatures are lower by at least an order of magnitude /Takano 03b/. Fabrication of (Pu,Am,Zr)N thus appears to be feasible. Los Alamos has already fabricated such material, and irradiation tests will take place in the Idaho Advanced Test Reactor in 2004 /Crawford 03/. Stability of the compound during transients must be proven though before one can guarantee the applicability of nitrides for the purpose of americium transmutation. Hence, the FUTURE project has dealt with fabrication of $(Pu,Am)O_{2-z}$, and different approaches of improving the thermal conductivity of oxide fuels. Various types of matrices have been investigated, including magnesium oxide and different metals. While fabrication requirements set a lower limit of 50 volume percent for the matrix, the high cross section for neutron absorption in these materials set upper limits to their concentration in the fuel. Tungsten and natural molybdenum, e.g. are metals that have both high melting temperature and high thermal conductivity, but are not compatible with the actinide vector determined by safety and reactivity conditions /Wallenius 03a; Smith 03; Wallenius 04/. Chromium would be the outstanding choice from the neutronic view-point, but may have problems related to fabricability as well as irradiation resistance. It has therefore been decided to focus fabrication and irradiation tests of $(Pu,Am)O_{2-x}$ dispersed in magnesia and ^{92}Mo. Enrichment of the lightest molybdenum isotope would decrease the absorption cross section of the fuel significantly. It would also suppress the undesired production of ^{99}Tc by orders of magnitude. In addition, industry (URENCO) already produces ^{92}Mo in amounts sufficient for fabrication purposes.

Initial fabrication tests of $(Pu,Am)O_2$ at CEA in Marcoule have shown that a solid solution can be obtained in a large range of compositions. Test of composite fuel fabrication will start soon at CEA as well as at ITU, with the objective of having four oxide pins (two of each kind) ready for irradiation in Phenix by end of 2006. The FUTURIX irradiation will further include metallic fuels fabricated by Argonne-West, and nitride fuels from Los Alamos. It will thus be a very comprehensive, side by side test of four different ADS fuel candidates in a fast neutron spectrum.

Americium targets for irradiation in fast reactors

An alternative strategy for transmutation of americium has been investigated within the EFTTRA (European Fuels and Targets for Transmutation) collaboration since the beginning of the 1990s. Homogeneous mixing of americium into the fuel of sodium cooled fast reactors was early on shown to deteriorate the safety properties /Tommasi 95/. Adding moderated americium targets to the periphery of CAPRA[65] or EFR[66] cores, however, allows maintaining acceptable safety coefficients /Beaumont 99/. A fairly extensive research program has hence been devoted to theoretical simulation of high burnup irradiations and experimental irradiation testing of such targets. In the so called T4 experiment, pellets of magnesium aluminate spinel ($MgAl_2O_4$) containing 11 weight percent ^{241}Am were fabricated by the newly developed infiltration method at ITU. Two pins were irradiated in the High Flux Reactor in Petten up to 28% and 57% burnup (fission in actinides), respectively /Klaasen 02/. A large swelling (18 and 29 volume

[65] CAPRA = special (French) design of fuel assemblies for burning Pu in a fast reactor.
[66] EFR = European Fast Reactor; design of a fast power reactor made by a industry consortium.

percent) of the pellets was detected in spite of the low temperature in the targets. This swelling has been attributed to amorphisation of the spinel phase. Hence, for further irradiation testing of americium targets, yttrium-stabilised zirconium and magnesium oxide will be used as matrix. The irradiation of Am microdispersed in MgO (ECRIX) commenced in Phenix 2003, while targets where solid solutions of americium and yttrium stabilised zirconium are micro and macro dispersed in magnesia (CAMIX & COCHIX) are intended to start in late 2004.

Since the transmutation of americium in this concept takes place in a moderated spectrum, a significant fraction of the material is converted to curium and californium. The neutron activity from spontaneous fission of californium becomes extremely large, and the reprocessing of americium targets irradiated to high burnup is therefore not considered to be feasible. As a fast reactor environment is required to provide the high neutron flux, the maximum achievable burnup is limited by radiation damage to the cladding in the target pins. Recent calculations /Newton 03/ have shown that the limit to burnup is likely to be 75%, rather than the 90% originally projected. Consequently, the reduction of radiotoxicity in this approach becomes rather modest (a factor of 10), and the time needed to store the irradiated target pins before the radiotoxicity becomes comparable to that of rich uranium ores is of the order of 10 000 years. This should be compared to the period of less than 1000 years required to store the secondary waste arising from multirecycling of minor actinides in ADS.

4.1.5 Materials

Radiation damage

Structural materials like fuel clad, fuel element wrapper tubes, spallation target window and containment in an ADS will suffer radiation damage, leading to degradation of mechanical properties at high temperatures (radiation induced and thermal creep, swelling) as well as low temperatures (embrittlement). The behaviour is temperature, dose, dose rate and strongly composition dependent.

In the case when the interface between the accelerator tube and the spallation target (the “window”) is located in contact with the liquid lead-bismuth, the cycle length of ADS operation will be determined by the life time of this window. Early estimations suggested that damage caused by a one megawatt proton beam, in combination with neutron irradiation, could lead to brittle failure of the window within one year /Konys 03/. Therefore, an important task in the development of ADS is to find or develop a steel that is resistant to radiation damage arising from a mixed proton and neutron spectrum. In the 5th FWP SPIRE project, several candidate materials have been investigated before and after irradiation. Ferritic (nickel free) steels were selected on the basis of their higher threshold dose for swelling and better compatibility with lead-bismuth.

The composition of the steels studied is displayed in Table 4-4. Theoretical investigations of Fe-Cr model alloys have also been performed, in order to acquire a better understanding of basic phenomena leading to damage.

Table 4-4. Chemical composition of the main ferritic-martensitic steels investigated in SPIRE. The so called "9-chromium" steels EM10 and T91 are known to become less brittle under irradiation, while 12-chromium steels like HT9 are more corrosion resistant.

Name	Cr	Mo	C	Mn	Si	Ni	V	Nb	N
EM10	8.8	1.05	0.11	0.50	0.37	0.18	–	–	0.02
T91	8.3	0.95	0.11	0.38	0.43	0.13	0.20	0.08	0.02
HT9	12.0	0.96	0.19	0.60	0.42	0.56	0.30	–	0.06

Impact of spallation elements on mechanical behaviour

The production of spallation elements in the window, like titanium, phosphorous and sulphur, is expected to embrittle the proton beam window. Reference steels with 9% Cr concentration were doped with these elements during fabrication. It was found that titanium may be soluble up to 0.1 weight percent, whereas sulphur very quickly segregated into precipitates. Phosphorous tended to segregate at grain boundaries. In all cases, a degradation of toughness and impact properties was observed.

One of the steels (T91) proved to be less susceptible to hydrogen embrittlement than other steels tested (EM10, HT9). This was interpreted as a higher probability for hydrogen to be trapped at grain boundaries and carbide/matrix interfaces.

Mechanical properties of samples that were implanted with spallation elements through ion beam irradiation, exhibited a spectacular dependence on implantation temperature. At a temperature of 250°C, a complete loss of ductility was obtained for helium concentrations exceeding 2500 appm[67]. At higher temperatures, the loss of ductility was much less pronounced. Hence, it is recommended that the operational temperature of the beam window should not be less than 350°C.

Proton and neutron irradiations

A large number of steel samples (~ 500) have been irradiated by 600 MeV protons at PSI in Switzerland at temperatures ranging from 90°C up to 350°C /Dai 03/. The maximum dose obtained was 12 DPA[68]. The loss of ductility was found to be larger than in corresponding neutron irradiations. 12-chromium steels like HT9 were shown to be severely embrittled already at this modest dose. From post-irradiation analysis, the lifetime of T91 subjected to 1 MW of beam power could be projected to be of the order of 5–6 months. This result will have a major impact on the forthcoming MEGAPIE experiment, where a Pb-Bi cooled target with a T91 window will be installed and used as a neutron source at PSI.

[67] appm = atom parts per million.
[68] DPA = Displacements Per Atom.

Samples of T91, EM10 and HT9 doped with sulphur and phosphorous were irradiated in the Russian fast neutron reactor BOR-60. Doses up to 40 DPA were obtained, and post-irradiation examination showed that the doped specimens had lower ductility than the reference steels.

Basic studies

Theoretical modelling of defect production and evolution in pure metals and binary alloys may be done using a multi scale approach, where experimental data are combined with calculations on micro-and macro-scale. Within the SPIRE project, KTH has developed a set of Embedded Atom Method potentials for Fe-Cr alloys, describing effective interactions between the constituents. The potentials for the pure elements were fitted to recent experimental data for elastic constants, vacancy formation energies and thermal expansion coefficients. For the alloy, ab initio data for mixing enthalpies calculated at Uppsala University were used /Olsson 03/. Using these potentials in molecular dynamics simulation of thermal aging, it was found that the model correctly could reproduce experimentally measured temperature, time and concentration dependence of Cr precipitation in binary Fe-Cr alloys /Wallenius 03b/. This opens the possibility for direct calculation of precipitate induced hardening. Figure 4-9 shows how the distribution of Cr atoms in a Fe-20Cr alloy changes with time at a temperature of 800 K.

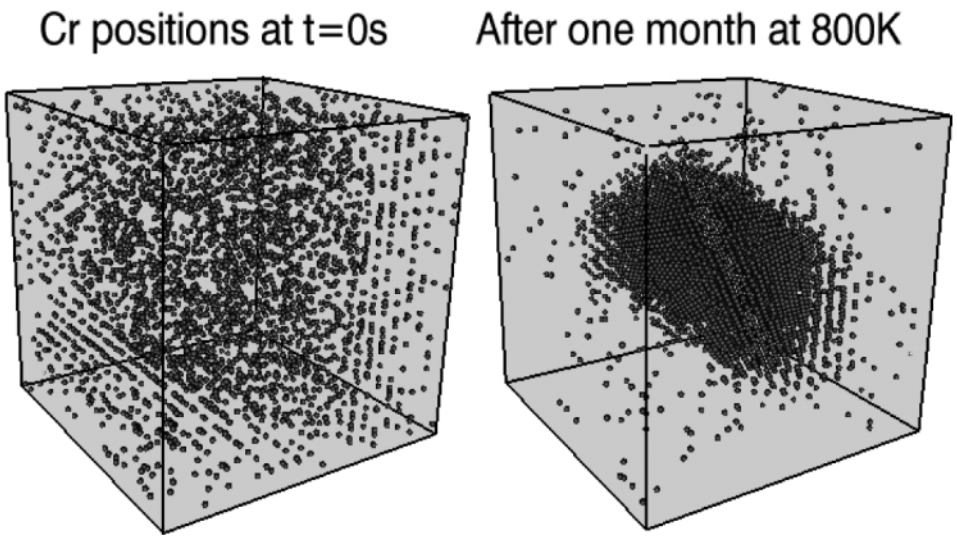


Figure 4-9. *Precipitation of Cr atoms in an initially random alloy, simulated with Kinetic Monte Carlo using a potential fitted to Fe-20Cr. No precipitation is observed using the potential fitted to properties of Fe-5Cr, in agreement with the phase diagram.*

4.1.6 Safety

Background

World-wide, there have been various studies performed on the safety of an ADS for transmutation of minor actinides. These studies have been of very preliminary nature, since a detailed design of the ADS was not available. The currently on-going European studies of an experimental ADS (XADS) conducted safety assessments of detailed designs developed for XADS cooled either by lead-bismuth eutectic (LBE) alloy or by He gas. These designs have subcritical reactors fuelled with conventional PuO_2-UO_2 fast reactor fuel (from the SUPERPHENIX), and a thermal power range of 50 to 80 MW_{th}. Their purpose is to prove in principle that transmutation of minor actinides can be achieved by irradiating them in a fast neutron spectrum ADS. The fact remains, however, that a subcritical reactor fuelled completely with minor actinides has still not been shown to be fully compatible with the safety and regulatory requirements in Europe even at the power level of 50 to 80 MW_{th}. The reactivity coefficients for actinide fuel may need to be thoroughly examined before a reliable safety analysis could be performed for an actinide fuelled ADS.

Since the objective of this report is to describe the 'State-of-the-Art', the following description is based on work currently on-going with in the European Project PDS-XADS. This work includes assessment of the safety of XADS designs currently under development in Europe. Two of these designs are cooled by LBE, the Ansaldo-design of 80 MW_{th} capacity /Ansaldo 01/ and the MYRRHA-design of 50 MW_{th} capacity developed by SCK·CEN /Benoit 03/. The third design has been developed by NNC in UK and Framatome and is cooled with He gas /Giraud 03/. The basic structure of the LBE cooled designs is a pool type system with a subcritical core at the bottom of the vessel, containing a spallation source (also LBE) in its middle, which is also at the end of a proton beam pipe connected to the accelerator. The LBE is cooled in heat exchangers built into an annulus region near the top of the vessel, employing a diathermic fluid and water, respectively. This secondary coolant is cooled by air coolers in the Ansaldo-design. There is no electric power generation in the LBE cooled XADS. There are differences between the Ansaldo-design and the MYRRHA-designs, e.g. the former employs much lower power density or linear power than the latter design.

Integrated safety assessment

The safety assessment work in the PDS-XADS-project is performed by several partners with KTH (department of nuclear safety) being the leader of the safety analysis work. The main objectives are /Carluec 04/:

- Develop an integrated safety approach common to both the LBE- and the gas-cooled concepts.
- Develop an evaluation methodology for both alternatives, i.e. the LBE and the gas-cooled designs.
- Produce safety analysis reports for each of the designs with the identification of the design features required to meet the XADS safety objectives.

The rationale for the integrated safety approach is quite similar to that practiced for the current LWR plants i.e. defence in depth, single failure criterion and specified safety goals. The PDS-XADS is a subcritical fast reactor; thus, it has the inherent advantage that reactivity-initiated accidents (RIA) (which are important for fast reactors) may be prevented by an appropriate choice of the subcriticality level. The safety evaluation approach required the specification of the design-base conditions (DBC) and the design-extension conditions (DEC). These were specified for both the LBE-cooled and the gas-cooled designs. Again, guidance in their specification was derived from the safety regulations for the LWRs and for the fast reactors.

From the specifications of the DBC and the DEC, a specification of the main safety issues became possible. It should be noted here that consideration of the severe accidents and their attendant concerns in design are also a part of the work. However, the approach, so far, is to prevent them and manage their consequences. Additional considerations are currently believed not be so fruitful, particularly for the LBE-cooled design, since basic phenomenology and data for the interaction of core melt and coolant are lacking. Evaluation of the severe accidents for the gas-cooled design can take guidance from such analysis performed previously for the gas-cooled fast reactor (GCFR) designs.

The specification of the safety evaluation methodology followed the practice previously employed for the fast reactor designs, i.e. best estimate methodology with appropriate safety margins, as determined by an uncertainty analysis. Towards this focus, an evaluation was made of the computer codes available for performing an adequate, best-estimate safety analysis and for producing preliminary safety analysis reports (PSARs). It was found that, fortunately, several codes were available which could be adapted for the analysis of the DBC and DEC transients. These include /Carluec 04/:

(i) RELAP-5, which has been modified for the LBE coolant by ANSALDO.
(ii) RELAP-5 – PARCS, coupled code also modified for the LBE-coolant.
(iii) TRAC-M code of USNRC, recently modified for the LBE-coolant by the LANL.
(iv) EAC (European Accident Code) developed at the JRC-Petten.
(v) SAS-4, SASS-SYS codes modified to include LBE.
(vi) SIMMER code, which can model the hypothetical core disruption accident (HCDA) for a fast reactor.
(vii) SIM ADS code developed at FZK.

Production of the PSARs requires analysis of not only the design-base accidents, but also of a host of transients. Towards this purpose, a list of DBC transients was developed. The design-extension condition and transients have currently been specified as the DBC transients except that the power reduction, affected by the shut-down of the accelerator, does not occur.

Since the RIA looses its prime significance for the subcritical reactor, the more important safety issues are:

(i) Heat removal through natural circulation.
(ii) Loss of heat sink.
(iii) Reliability of, and time to shut down of the accelerator.
(iv) Ingression of water in the gas-cooled design and of oil in the LBE-cooled design.
(v) Fuel assembly blockage, possibly due to corrosion products.
(vi) Recriticality potential; this effect is important and different from critical reactors since there is no external control of reactivity (control/shut-down rods) in the current XADS designs.

The reactor shutdown is affected only through the shutoff of the accelerator beam.

Other consideration which are important are that:

(a) There should be no cliff-edge issues regarding DEC conditions.
(b) Severe accidents should be prevented and no safety equipment should be employed which cannot function during a severe accident.
(c) There are presently no containment strength requirements on the design of XADS.
(d) There should be no action required outside the reactor boundary to limit the radioactive releases.
(e) The residual risk should be defined; the accidents pertaining to the residual risk do not need to be analyzed, however, a limit should be defined for the residual risk.
(f) The required performance of the mitigative systems should be determined.

DBC and DEC transients

A list of 3 operational, 11 protected and 12 unprotected transients has been defined for the LBE cooled XADS and a list of 3 operational, 14 protected, 11 unprotected and 3 shutdown transients has been defined for the gas cooled design. These transients were also required to consider both the beginning of cycle (BOC) and the end of cycle (EOC) fuel loading. These transients are listed in Table 4-5 and Table 4-6 /Sehgal 03a/. The analysis of these transients should provide sufficient results to write a credible PSAR.

The partners shared the responsibilities of performing analysis with their codes (some mentioned in the lists) for various transients. The coverage was such that inter-comparisons could be made so as to constitute a “limited” benchmarking of each other’s calculated results, since the codes employed have relatively different capabilities. Good inter-comparisons were obtained and it was evident that the results obtained are reasonably credible and that no errors or large uncertainties exist.

Table 4-5. List of transients to be analysed for LBE-cooled XADS (Ansaldo-design).

Transient number	Transient	Description	Burnup state								Transient already analysed by ANSALDO
			BOC	EOC	ENEA	PSI	JRC	FZK	FZK	FZK	RELAP5
					RELAP5+ PARCS	TRAC-M	STAR-CD, CFD, EAC2	SIMMER	SAS4-ADS	SIM-ADS	
Operational transients											
O - 1	Shutdown	Plant taken to ambient (30°C)	×	×						×	From HFP to HZP
O - 2	Shutdown with target flooded	Target is flooded and then plant taken down to ambient (30°C)	×	×			×			×	
O - 3	Startup	Plant is taken from CZP to HFP	×	×						×	From HZP to HFP
Protected transients											
P - 1	PLOF	Complete loss of all forced/ enhanced circulations in primary and secondary (oil) systems	×	×	×	×				×	×
P - 2	PTOP	300 pcm jump in reactivity at HFP	×	×				×		×	
P - 3	PTOP	300 pcm jump at CZP	×	×						×	
P - 4	PLOH	Complete loss of both secondary trains	×	×	×	×				×	
P - 5	PLOF+PLOH	Loss of gas and secondary loops lost	×	×	×					×	×
P - 6	LOCA	Primary vessel leaks, level in primary drops by 2 m, (partial) loss of nat. circ. in primary	×	×		×	×				×
P - 7	Over-cooling of primary side	Core inlet temp. drops 150°C in 450 sec.	×	×						×	×
P - 8	Inlet blockage of SA w/o radial heat transfer	Flow area of peak SA reduced to 2.5%, no radial heat transfer assumed	×	×	×		×			×	
P - 9	Blockage of SA with radial heat transfer	Flow area of peak SA reduced to 2.5%, radial heat transfer assumed	×	×			×				×
P - 10	Spurious beam trips	Beam trips for 1,2,3…10 sec. intervals	×	×	×			×	×		×
P - 11	HX tube rupture	Secondary oil leaks into primary side	×	×		×					
Unprotected transients											
U - 1	ULOF	Complete loss of all forced/ enhanced circulations in primary and secondary (oil) systems	×	×	×	×	×	×		×	Various partial ULOFs
U - 2	UTOP	300 pcm jump in reactivity at HFP	×	×			×	×		×	
U - 3	UTOP	300 pcm jump at CZP	×	×			×			×	
U - 4	ULOH	Complete loss of both secondary trains	×	×	×	×	×			×	
U - 5	ULOF+ULOH	Loss of gas and secondary loops lost	×	×	×					×	
U - 6	Unprotected LOCA	Primary vessel leaks, level in primary drops by 2 m, loss of nat. circ. possible	×	×		×	×				
U- 7	Unprotected over-cooling of primary side	Core inlet temp. drops 150°C in 450 sec.	×	×						×	No feedback
U - 8	Unprotected blockage of SA w/o radial heat transfer	Flow area of peak SA reduced to 2.5%, no radial heat transfer assumed	×	×	×		×	×		×	With radial heat transfer
U - 9	Unprotected inlet blockage of SA with radial heat transfer	Flow area of peak SA reduced to 2.5%, radial heat transfer assumed	×	×			×				
U – 10	Unprotected HX tube rupture	Secondary oil leaks into primary side	×	×		×					
U - 11	Beam Overpower to 200% at HFP		×		×			×	×	×	×
U - 12	Beam Power Jump to 100% at HZP		×	×	×				×		×

Table 4-6. List of transients to be analysed for He-cooled XADS (NNC/CEA/Framatome).

Transient number	Transient	Description	Burnup state							Transient already analysed by NNC/CEA/ Framatome
			BOC	EOC	ENEA	PSI	JRC	Framatome	FZK	???
					RELAP5+ PARCS	TRAC-M	STAR-CD, EAC2, HEXNOYN	STAR-CD CFD	SIM-ADS	
Operational transients										
O - 1	Shutdown	Plant taken to ambient (30°C)	×	×					×	
O - 2	Shutdown with target flooded	Target is flooded and then plant taken down to ambient (30°C)	×	×					×	
O - 3	Startup	Plant is taken from CZP to HFP	×	×					×	
Protected transients										
P - 1	PLOF	Complete loss of all forced systems (primary, DHRS and secondary)	×	×		×		×	×	
P - 2	PLOF	Loss of forced primary and secondary systems but DHRS active	×	×		×			×	
P - 3	PTOP	400–500 pcm jump at HFP (target flooding, water ingress, air ingress)	×	×	×				×	
P - 4	PLOH	Complete loss of all heat transfer into secondary and DHRS, only primary blower active	×	×		×			×	
P - 5	PLOH	Complete loss of all heat transfer into secondary, DHRS and primary blower active	×	×		×			×	
P - 6	PLOF+PLOH	All blowers lost and secondary loops lost (station black-out)	×	×		×			×	
P - 7	PLOCA, primary vessel leak	Primary blower active	×	×						
		P-7_30 cm^2							×	
		P-7_100 cm^2				×			×	
		P-7_500 cm^2							×	
P - 8	PLOCA, primary vessel leak	All blowers off (both prim.+DHS blowers), station black-out	×	×						
		P-8_3 cm^2							×	
		P-8_30 cm^2				×			×	
		P-8_100 cm^2							×	
		P-8_500 cm^2							×	
P - 9	PLOCA, primary vessel leak	Prim. blower off, 1 of the 2 DHS blowers active, 2MW heat removal. w_DHRS to be defined by Bernard	×	×				×		
		P-8_3 cm^2							×	
		P-8_30 cm^2							×	
		P-8_10 0cm^2							×	
		P-8_500 cm^2							×	
P - 10	Blockage of SA w/o radial heat transfer	Flow area of peak SA completely blocked, –1000 pcm insertion after clad melting	×	×	×					
P - 11	Spurious beam trips	Beam trips for 1,2,3…10 sec. Intervals, Fuel damage possible.	×	×						
P - 12	Core compaction at HFP	–2000 pcm added at HFP	×	×						
P - 13	Inst. Loss of pressure without DHR		×					×		
P - 14	Unprotected LOCA		×	×				×		
Shutdown state transients (protected)										
P - 15	PTOP	5000 pcm jump at CZP in shutdown state (target flooding, water ingress, air ingress)	×	×						
P - 16	Fuel handling error	–1000 pcm at CZP in shutdown state	×	×						
P - 17	Loss of forced convection	Coolability of SA held above core at CZP						×		

Unprotected transients											
U - 1	ULOF	Complete loss of all forced/ enhanced circulations in primary and secondary systems	×	×		×	×		×	
U - 2	UTOP	400–500 pcm jump in reactivity at HFP (target flooding, air ingress)	×	×	×		×		×	
U - 3	UTOH	Complete loss of all heat transfer into secondary, blower on	×	×		×	×		×	
U - 4	ULOF+ULOH	All blowers lost (prim. & DHRS) and secondary loops lost	×	×		×	×		×	
U - 5	ULOCA, primary vessel leak	Prim. blower active	×	×						
		U-5_3 cm^2							×	
		U-5_30 cm^2							×	
		U-5_100 cm^2				×			×	
		U-5_500 cm^2							×	
U - 6	ULOCA, primary vessel leak	Blowers off (both prim. & DHS)	×	×						
		U-6_3 cm^2							×	
		U-6_30 cm^2				×			×	
		U-6_100 cm^2							×	
		U-6_500 cm^2							×	
U- 7	Unprotected over-cooling of primary side	Core inlet temp. drops 50°C in 50 sec.	×	×						
U - 8	Unprotected blockage of SA w/o radial heat transfer	Flow area of peak SA completely blocked, Insert 1000 pcm insertion after clad melting, no radial heat transfer assumed	×	×	×					
U - 9	Beam Operpower to 200% at HFP		×				×			
U – 10	Beam Power Jump to 100% at HZP (DHRS blowers active)		×	×			×			
U - 11	Core compaction at HFP	200 pcm added at HFP	×	×						

Some preliminary conclusions from the analyses

It was found that the Ansaldo-design behaves remarkably well for both DBC and DEC transients, except for the DEC transient: Unprotected loss of heat sink coupled with loss of flow. In that transient there can be core damage after 30 to 60 minutes. Since this transient has very low probability, it could be assigned as a residual risk. There may be some relief in the consequences if there is a strong natural circulation in the secondary loop. It is important to have a good design of the secondary loop.

An analysis of the postulated transients for the gas-cooled design indicated that the peak clad temperature may be ~ 800º C for normal operation at the 80 MW_{th} thermal rating. In a postulated loss of flow transient, temperatures could reach 950ºC. These values are significantly beyond the 700ºC limit for the operation of the clad. The gas-cooled design was re-examined, recently, because of these results. The remedies appear to be (1) to roughen the surface of the clad to increase the heat transfer coefficient, thereby decrease the temperature difference between the clad temperature and the gas coolant. Additionally (2) a gagging (orificing) scheme is being designed to reduce the radial peaking factor, thereby reducing the temperature of the hottest pin in the core. Thus, these innovations can provide large reduction in clad temperature during normal operation and transients and it appears that the gas-cooled XADS design may be able to tolerate the design base accidents and transients successfully. Further analyses of the gas-cooled design are proceeding. Previous safety analysis work performed on the gas-cooled fast reactor (GCFR) can serve as a model. It should be noted here that the

gas-cooled XADS design cannot tolerate unprotected transients, i.e. the accelerator trip has to be 100% effective and immediate. Delays larger than a few seconds can also lead to high temperatures in the cladding.

The MYRRHA-design by SCK·CEN was evaluated on a very preliminary basis. This design is LBE-cooled, however, it is a high performance design, employing much larger linear power rating than the Ansaldo design. Thus, it will not be as forgiving as the Ansaldo design. The MYRRHA design also has significantly less difference in height between the thermal centres than the Ansaldo design, which provides much less capability for natural circulation cooling.

4.2 Fast critical reactors

From a pure technological viewpoint, recycling of plutonium in fast critical reactors appears very attractive. Not only does the fast spectrum allow for multi-recycling of plutonium without any support of ^{235}U, but it also leads to a smaller production of americium than if Pu would be multi-recycled in thermal systems. The CAPRA design study made by a European collaboration in the 1990ies showed that existing fast breeder reactors without major difficulties may be converted into plutonium burners /Languille 95/. By introducing moderator pins into the core and reducing the pin diameter it was shown that the fraction of ^{238}U in the oxide fuel could be reduced to 65% while still maintaining acceptable safety parameters /Beaumont 97/. The CAPRA core could thus consume a substantial amount of plutonium coming from spent light water reactor fuel. Assuming that Pu burning in CAPRA cores would be part of a Double Strata fuel cycle, the fraction of power produced in fast critical reactors would be 19% /Salvatores 98; NEA 02b/.

The recycling of americium in fast critical reactors is, however, a much more problematic issue. Adding americium to the fuel inevitably leads to a decrease in Doppler feedback, increase of the sodium void coefficient, and a reduction of the effective delayed neutron fraction. Table 4-7 shows explicitly how these parameters are deteriorated when minor actinides with a MA/Pu ratio of 1/7 are included in the CAPRA fuel. The resulting values are on the limit of being acceptable. It can be noted that this minor actinide fraction corresponds to the waste stream from spent UOX fuel. Increasing further the relative fraction of Am by, e.g. MOX recycling in LWRs, would mean that CAPRA cores could no longer consume all americium coming out of the LWRs.

Table 4-7. BOL safety parameters of a CAPRA core with 4% minor actinides in the fuel, compared to that of a minor actinide free core.

Core	CAPRA with MA	CAPRA without MA
Beta-effective	310 pcm	380 pcm
Doppler constant	–890 pcm	–1260 pcm
Coolant void coefficient	+0.22 pcm/K	+0.14 pcm/K
Coolant void worth	+460 pcm	–70 pcm

Similar conclusions have been made for the metallic fuelled Integral Fast Reactor design of Argonne National Laboratory /Wade 97/, as well as the lead cooled Russian BREST reactor /Adamov 97/.

Therefore, closing the fuel cycle using fast critical burner reactors only, appears to require abandoning of MOX recycling in LWRs entirely. Hence, the fraction of fast neutron reactors in the power park would have to be as large as 37%, which would yield a significant increase in the cost penalty for P&T, compared to the Double Strata Fuel cycle /NEA 02b/.

It should be noted that the Generation IV initiative concerns deployment of an entirely new reactor park, without any LWR component. The Generation IV reactors would thus only consume the minor actinides produced in their own cores – which may be feasible. The cost of electricity generation in a nuclear park with 100% Generation IV reactors would, however, be much higher than in any P&T scenario with a base load of LWRs for commercial power production.

4.3 Other transmuters

Transmutation, in the sense of reducing the amount of transuranium nuclides going to a deep repository and/or "incineration" of weapons usable isotopes (^{239}Pu) can be performed in different types of critical reactors. In theory also the irradiation with other types of elementary particles than neutrons can cause transmutation. In practice such irradiation is not applicable in macroscopic scale for technical, economical and other reasons.

A number of studies, in particular in France, have been made of increased burning of plutonium in light water reactors. Some of these have been summarised by a working group in NEA /NEA 03/. The studies show that with a modified design of the MOX-fuel assemblies in a PWR it is possible to strongly limit or even reduce the accumulating amount of plutonium from a reactor park like the French. A once-through fuel cycle in a PWR gives a net production of about 30 kg Pu per TWh_e whereas with some of the advanced Pu fuel assembly designs studied a net consumption of 60–130 kg Pu per TWh_e may be reached. The implementation of some of these designs might be an interim strategy to keep the stock of plutonium limited. See also Section 2.7.2.

Other reactor types considered to be candidates for a reactor-transmuter concept are the Deep Burn Modular Helium Reactor (DB-MHR) and Molten Salt Reactor.

General Atomics in USA with some international partners (including KTH) have proposed and are developing the DB-MHR concept aimed at transmutation of some of the transuranium nuclides from spent LWR fuel. This concept is based on the High Temperature Gas Cooled reactors which were developed and demonstrated in 1960ies. A typical plant of a DB-MHR nuclear facility would use four modules of 600 MW_{th} each to burn Pu from Light Water Reactors and, at the same time, to produce electricity and/or generate high temperature process heat. An essential feature of the DB-MHR is the use of ceramic-coated fuel particles (TRISO) that provide very strong containment and are highly resistant to irradiation, thereby allowing very extensive burnup levels in one or two passes /Talamo 04/.

Each module consists of a gas-cooled graphite-moderated DB-MHR. The attractive features of the DB-MHR are:

- High burnup, up to 700 MWd/kg, with better than 90% destruction of actinides (including ^{239}Pu).
- High conversion energy efficiency (about 50%).
- Great flexibility in the choice of the fuel type: fertile and non-fertile cores (e.g. Th, U or Pu).
- Spent fuel with TRISO particles is considered to be a good waste form.

Plutonium and other transuranium destruction would be performed in DB-MHR, without multiple reprocessing, thus limiting the generation of secondary waste. Over 90% incineration of ^{239}Pu is considered by some nuclear engineers to be a sufficient incentive for construction of such a reactor. Detailed analyses of DB-MHR performance indicate, however, that at least one reprocessing pass of spent TRISO particle will be required to achieve all-over optimization of actinide transmutation. Deep-Burn technology could be available for deployment in a relatively short time.

The Molten Salt Reactor concept designed for an efficient TRU-burning system has also been "revisited" in last years, particularly in France and in Russia. The foundations of the molten salt technology, based on fluoride salts of uranium and thorium dissolved in Be-Li fluorides, were developed at Oak Ridge National Laboratory. "Nuclear feasibility" of this concept was demonstrated in the Molten Salt Reactor Experiment with four years of successful operation in 1965–1968.

The idea of a fluid-fuel reactor is appealing with its potential for an effective consumption/transmutation of nuclear fuel (most of the constraints which limit burn-up in solid fuel are relaxed), inherent safety features regarding supercriticality accidents etc. The molten salt fuel at operating reactor fissile concentrations can provide inherent protection against criticality accidents during handling. For thermal neutron spectrum designs, a graphite moderator is required for criticality so that criticality can occur only in the core.

Molten salt reactors can be designed with or without on-line processing, or with only partial processing. For liquid fuel reactors with reprocessing, some of the common nuclear reactor terminology is even not applicable. Except for some start-up periods the molten salt reactors operate at an equilibrium steady state. The fuel concentration and content do not vary with time. Fissioned/consumed fuel is replenished by feeding or by breeding. The term "fuel burn-up", commonly used for solid-fuel reactors, thus has not the same meaning, as there is no specific amount of energy generation related to a particular identifiable original amount of fissile material. For the same reason, there is no excess reactivity to compensate for burn-up or for the continuous poisoning caused by fission product accumulation as in solid fuel reactors. Also decay heat problems for the MSR are not as severe as for normal solid fuel because of small concentration of UF_4 in the fuel salt (l–2% mole).

Molten salts reactors can operate at high temperatures and low pressures, and they possess favourable heat transfer properties. These properties result in high thermal efficiency for the reactor and absence of safety hazards associated with high pressures. The salts are chemically stable and non-flammable, averting fire hazards, and there are no energetic chemical interactions between the salts and water.

One advantage of the fluoride-based MSR is the potential for an integrated fuel recovery capability. The processing is based on the high volatility of UF_6. By sparging the salt with fluorine, uranium can be removed as UF_6, which can then be converted back to UF_4 and recycled into a fresh batch of fuel salt. The residual salt, free of uranium, could be subjected to any of a number of processes to remove fission products and concentrate them. The carrier salt components (lithium, beryllium, fluorine) could also be isolated and recycled if that would be economically desirable. All of these steps could be made independent of the reactor operation.

There are of course also some drawbacks with fluid-fuel reactors. Some examples are that radioactive gaseous fission products must be collected and contained, that fairly large systems outside the reactor will contain fission products and must have increased shielding, that these same systems will be contaminated by some fission products, that radiation protection of plant operating and maintenance staff will be more difficult/expensive etc.

A few Molten Salt Reactor concepts have been considered for transmutation purposes:

- THORIMS-NES[69]-concept /Furukawa/, composed of: a Molten-Salt Reactor, fissile-fuel producers by spallation/fission reactions of 1 GeV-proton – Accelerator Molten-Salt Breeder (AMSB) and a pyroprocessing plants.
- "TASSE"[70] /Slessarev 99/ – a thorium fuelled accelerator driven subcritical concept developed at CEA optimized for both energy production and TRU-incineration. This concept has not been developed beyond a pre-conceptual stage but brought a lot of refreshing attitudes to a molten salt reactor.
- AMSTER[71] – is a new design of a molten salt critical reactor aimed to burn TRUs with a uranium and/or Th support. AMSTER is based on the same salt composition as MSBR, i.e. 61% LiF + 21% BeF_2 + 18% AcF_4 (Ac stands for actinides). Several fuelling options are envisaged for AMSTER:
  - Core at equilibrium: TRU from spent PWR fuel with continuous feed of $^{235/238}U$ and/or Th.
  - TRU incinerator: core for which the TRU inventory is higher than that of the LWR spent fuel and which is kept constant by adding continually ^{235}U and TRUs.

  AMSTER shows a good performance compared to other transmutation systems having a low TRU inventory with relatively small losses.
- MOSART[72] concept under development in Russia, funded currently by ISTC. The system is fuelled only by TRUs (without fertile Th or U isotopes); has high transmutation efficiency and can be fuelled with a wide range of TRU compositions from LWR spent fuel. MOSART has a pool type design with graphite as moderator/reflector material and a salt composition[73] of 15% LiF + 58% NaF + 27% BeF_2.

[69] Thorium Molten-Salt Nuclear Energy Synergetics.
[70] Thorium based Accelerator driven System with Simplified fuel cycle for long term Energy production.
[71] Actinides Molten Salt TransmutER.
[72] Molten Salt Reactor Transmuter.
[73] The choice of this composition results compared to others in a significant reduction of undesirable tritium production in the core.

By now a lot of interesting research is being done in this project on salt solubility, chemistry of TRU-fluoride salts and material compatibility in support to a detail conceptual design of the reactor.

One MOSART reactor would be able to transmute in equilibrium TRU-waste from four LWR reactors. MOSART reactors might be considered as a "clean-up" reactor for a once-through nuclear fuel cycle.

4.4 Nuclear data for transmutation

The nuclear data needs for transmutation can roughly be divided into three areas, characterized by the energy of the incident particle. These will be referred to as low energy (up to 20 MeV), intermediate energy (20–200 MeV) and high energy (above 200 MeV).

At low energies, the most important nuclear data are cross sections for neutron-induced fission and capture reactions. At high energies, proton-induced neutron production cross sections are the most important data, while at intermediate energies, a wide range of various nuclear reaction cross section data are of importance, and there is no specific reaction of dominating importance.

The development of the conventional nuclear power as well as nuclear weapons and fusion prompted a large nuclear data campaign from the end of World War II until today. This has resulted in large global data files, comprising a few million data points, which are compiled and evaluated by well-established procedures. These data libraries have traditionally had an upper energy limit of 20 MeV, which is dictated both by the needs of the applications and the availability of measurements. Electrostatic accelerators, which are easy to operate and inexpensive, can provide neutrons up to about 25 MeV. Above this energy, much more costly and technically challenging techniques have to be used for neutron production.

The needs of traditional applications are visible in the low-energy data base. For, e.g. light water reactors, there is an obvious need for high-quality data on uranium and plutonium isotopes, while data on, e.g. curium are less important due to the low curium content in the reactor. If considering dedicated waste burners, where curium might be a much more abundant element than in power reactors, the quality of the curium nuclear data accordingly becomes more important.

With a slight over-simplification, it can be said that the low-energy nuclear data for light water power reactors are today of a sufficiently good quality, but for other applications, like waste transmuters, there are still needs on some specific cross sections. The data that are important to get the full system performance under acceptable control depend on the purpose. For ADS, two main options have been discussed where clearly identified data needs are at hand. These are incineration of minor actinides, especially americium and curium, and transmutation of fission products. In both cases, main candidates for new nuclear data measurements are the elements to be incinerated. Which element and isotope is most important is more difficult to assess. This issue needs sensitivity analysis, i.e. a simulation of the proposed technical system where the uncertainties of various performance parameters are studied versus the nuclear data input, varied within the data uncertainty.

Up to now, only a few such sensitivity studies have been performed. One general effect on which there is consensus is that for lead- or lead-bismuth cooled systems, the presently limited knowledge of the inelastic cross sections on lead isotopes results in large uncertainties in the system performance ($\Delta k_{eff} \approx 0.02$) /Car 97; Dahl 02/. Experimental measurements of these cross sections to remedy this uncertainty are being planned within the proposed upcoming EUROTRANS project.

Below 20 MeV, one single cross section can be decisive for the performance of the total technical system, to a large degree because of strong resonances. This is, however, a typical low-energy effect. Above 20 MeV, it is physically impossible to have sharp resonances in nuclear cross sections. Instead, cross sections always change in a rather smooth fashion. As a consequence, one single cross section cannot make a dramatic difference. Instead, knowledge of the general behaviour is what is important.

This means that the general approach to the entire problem is different above 20 MeV. It is impossible, in practice and even in theory, to measure all the reactions taking place in sufficient detail to use data as direct input to system modelling. Instead, nuclear theory has to be used, and the role of experiments is to provide key data for theory development. This means that to provide maximum benefit from a given investment, a coordinated effort with a balance of theory development and data measurements should be undertaken.

There is a notable division in nuclear data at about 200 MeV, which is due to theory as well as the experimental situation. At energies below 200 MeV, the interaction of an incoming neutron with a nucleus is determined to a large degree by collective effects in nuclei, i.e. all the protons and neutrons in the nucleus – and their internal interactions – contribute to the process, resulting in an exceedingly complex situation. Moreover, in this energy range, the motion of protons and neutrons inside the nucleus is chaotic, making deterministic theory breaking down. Hence, precise measurements are indispensable, since theory cannot even in principle determine some important parameters, like the energy, width and cross section of important resonances. At energies above about 200 MeV, the interaction is much less complex. An incoming neutron interacts with just a single proton or neutron in the nucleus, or a cluster of a few such particles. Accordingly, the reaction has a much simpler and smoother character, and theory based on the fundamental interaction of protons and neutrons can be used to provide reasonably good predictions of the neutron-nucleus interaction. This transition is obviously not sharp, but 200 MeV is the traditional thumb-rule for the low-energy limit for the validity of this type of theories.

In addition, there is a notable experimental limit at about 200 MeV. There are – and has been – a relatively large number of accelerators with maximum energies close to 200 MeV available for this type of research. Furthermore, there are a number of accelerators operating in the 600–2000 MeV domain, but only a few machines provide beams in the 200–600 MeV range. At present, no relevant accelerator in Europe operates in the 200–600 MeV range.

The nuclear data needs above 200 MeV are primarily motivated by the neutron production by proton-induced spallation. Thus, proton-induced neutron production reactions are of paramount interest. In addition, high-energy protons impinging on a heavy metal target will induce a very large range of nuclear reactions, which essentially all of them result in radioactive products. Thus, the total activation of such a target can be significant, and needs to be studied.

The situation is more diverse in the intermediate energy range. When protons impinge on a target, they do not necessarily produce neutrons in one single step. Instead, protons can produce other particles, which in turn produce neutrons. To get a reasonable precision in the final neutron production of the system, these intermediate reactions need to be known. This means that a wider span of reactions needs to be investigated. Furthermore, the theory situation is not as good as for higher energies, but not as complex as for low energies.

Since 2000, two large EU-supported projects have been active in the field of nuclear data for ADS, NTOF and HINDAS /NTOF; HINDAS/. NTOF is primarily focused on the low-energy range and dominated by activities at CERN, Geneva, Switzerland, where a spallation neutron source has been developed /Bor 02/. The agenda comprises mostly capture and fission cross section measurements in the eV to keV neutron energy range. The development of the facility was significantly delayed compared with the original time table, and therefore the project has been extended. At present, the facility is operational with parameters close to the specifications, and the first results have been presented.

HINDAS (High and Intermediate energy Nuclear Data for Accelerator-driven Systems) comprises 16 universities or laboratories in Europe /Kon 02/. Six experimental facilities are used to optimize the use of existing infrastructure. The HINDAS project, which has recently been completed, has resulted in a dramatically improved nuclear data situation in the 20–2000 MeV range.

There has been a strong Swedish participation in HINDAS. The department of neutron research at Uppsala University has led one work package, and 5 out of the 16 HINDAS partners have performed experimental work at The Svedberg Laboratory (TSL), Uppsala /Klu 02/.

In parallel with the EU activities, ISTC has financed important activities on nuclear data for transmutation. It should be especially pointed out that our present knowledge of fission cross sections above 20 MeV is heavily dominated by ISTC-supported data. From a Swedish perspective, it is noteworthy that a significant fraction of these results has been produced by Russian groups working at TSL.

There is a striking imbalance over the World on nuclear data for ADS. Europe, including Russia, dominates heavily. The other large nuclear energy countries, i.e. USA and Japan, have only limited research in this field, in spite of previously having hosted important activities.

5 Prospects for P&T developments

The successful development and introduction of partitioning and transmutation will depend on a broad research and development programme as is evident from the various studies described in the previous chapters. It is also evident that the system to be developed includes a number of important components and facilities that not yet are very clearly defined. This chapter tries to assess the prospects for the development of P&T from some important aspects. The first section addresses some of the key technical issues that have to be resolved. The sections following thereafter discusses some other issues that also will be of decisive importance – safety, needs for major demonstration plants, time and costs, impact on waste management programme as well as nuclear power programme at large and last but not least acceptance. Such a discussion can never be the final word but only a temporary assessment of the current situation.

5.1 Key research and development items

Despite the fact that P&T has been on the agenda for a rather long time there are still a number of issues that must be resolved before any really focused R&D-effort can start on P&T. The so-called roadmaps that were developed in USA and in Europe some years ago – see Sections 2.2 and 2.3 above – tried to define the programme needed to reach such a decision point for the ADS. Some efforts have since been done along the route proposed by these studies as described in the previous chapters. A number of things have however contributed to that the speed, intensity and funding of the efforts are much lower than suggested in the proposals.

- There is no unanimous view on the objectives for P&T. Some see it as a way to gain broad acceptance for nuclear power at large. Others promote it as a way to get out of the impasse for a deep repository in several countries. Others again put a strong emphasise on the proliferation aspects of a growing stock of plutonium from LWR and other reactor fuel as well as from dismantling of nuclear weapons.
- There is no unanimous view on the need to develop ADS for P&T or for the role of ADS in a P&T-system. Some advocate that ADS should be used for burning all transuranium nuclides from the present UOX-fuelled LWRs. Others see the ADS as a supplement particularly suitable for burning minor actinides (americium, curium and neptunium) and some plutonium with low fissile content, whereas the major part of the plutonium should be burned in LWR or in fast reactors. In the long run if nuclear power shall be a sustainable energy source for a substantial part of the world fast reactors may be the only reactors needed and then ADS will only be an interim solution.
- The interest for P&T is mainly concentrated to the national laboratories in USA, Europe, Japan and some other countries. The universities in many countries including Sweden also show a strong interest. The nuclear industry, however, has shown only a limited interest in the development. Their long-term interest is more focused on what is called the Generation IV type reactors.

- The national and industrial efforts on spent fuel and HLW management in almost all countries are primarily focused on the resolution of the deep repository issues. One important argument for this is that geological disposal of some wastes will be required also if P&T is developed and implemented.
- There is no consensus among experts on which technical route to follow for several parts of a P&T system.

Partitioning

Looking first at partitioning there are two basic systems that are advocated – liquid-liquid extraction systems and (non-aqueous) pyrochemical and/or electrochemical systems. These are described in Chapter 3.

The successful partitioning of minor actinides from real high level liquid reprocessing waste was first demonstrated some 20 years ago using industrially available phosphor-based reagents at a small pilot plant /Lil 84; Per 84a; Per 84b/. However, the process tested was felt to be too complicated and too expensive for any large scale use – approximately increasing the cost of reprocessing by 50%. At the same time research was begun on reagents that do not contain phosphorous. The reason is that with a completely incinerable reagent mixture it will be possible to transform most of the medium active waste from solvent degradation and clean-up into such a form (oxides) that it can be recycled back to the dissolver. Musikas coined the concept of CHON reagents, i.e. reagents containing only carbon, hydrogen, oxygen and nitrogen atoms /Mus 85/. Much of the research on a TBP[74] replacement reagent has been centred on diamide molecules because these have extraction properties for U and Pu in strong nitric acid solutions which are comparable to TBP and because they satisfy the CHON requirement. It was then obvious that similar CHON-reagents ought to be developed for the minor actinide recovery operation.

This development took an unexpected turn when a new class of reagents, the BTP:s, were discovered by Kolarik and co-workers /Mad 00/. The BTP:s can selectively remove americium and curium from strongly acidic high-level liquid waste without extracting any significant amount of the lanthanides. However, small scale pilot plant testing with synthetic and real high level waste showed some unfavourable properties; the chemical and radiological stability is not very good and a few elements, like Pd, are also extracted and very difficult to remove again. Furthermore, the synthesis of the tested reagent is complicated and rather expensive. Hence, much of the more recent research has been aimed at finding out if and how the basic BTP structure can be simplified and/or modified in order keep the good properties but to avoid the undesirable extraction of some elements, to increase stability, and to reduce production cost /Mad 02/.

It is very likely that more research has to be performed on selected diamides which can replace TBP in PUREX-like reprocessing or be used to co-extract minor actinides and lanthanides. More research is also still needed on BTP-like molecules, some of which might even be able to separate americium from curium once both elements are separated from the rest of the fission and corrosion products present in the high-level waste. Each time a good candidate reagent is found in the more basic research it is necessary to develop the appropriate process flow sheet and to test this using simulated or real HLW. Finally, the size of the partitioning process might be reduced if the HLW can be

[74] TBP = tributylphosphate.

pre-concentrated and partially denitrated before treatment. However, this poses new problems such as formation of precipitates and colloids which have to be removed and purified separately.

With regard to pyrochemical processing, much remains to be done before its suitability for use in P&T can be assessed. In principle, all the chemical knowledge needed is probably available even if much is buried in old technical reports and forgotten. Retrospective literature studies would probably be rather useful. It is also desirable that all the available knowledge from existing large scale non-nuclear industrial uses of pyrochemical processes and equipment should be collected and evaluated with regard to its possible use in the nuclear field. The basic research on pyrochemistry also needs to concentrate on the development of suitable equipment which should have a long life expectancy, an ultra-high reliability, be continuous, be remotely operated, and be maintained without any manual intervention. Once such equipment is developed and tested for unit operations in pyrochemistry it would put certain requirements on the process chemistry which then may have to be developed further.

Furthermore, new sensitive measurement methods must be developed which permit accurate on-line determination of the tiny concentrations of minor actinides left in the processed HLW and in other waste streams from the partitioning process.

In summary, the adequacy of separation of uranium, TRU, and long-lived fission products from spent LWR-UOX, MOX fuel, FR and/or ADS transmuter fuel still need to be proven on an industrial scale. The goal for separation efficiency of 99.9% seems to be within reach at least for the aqueous process, but it has not been demonstrated yet on a large scale.

Transmutation

As has been pointed out earlier in this report, effective transmutation of all transuranium radionuclides can only be achieved in a fast neutron spectrum. This means that a fast reactor or an ADS with a fast neutron spectrum is needed.

Fast reactors have already been developed and operated since many years. Following the slow-down or decline in construction of nuclear power plants, most of these fast reactors have been closed. The majority of these reactors were based on cooling with liquid sodium. A common reason for closing many of the reactors was technical problems associated with the sodium cooling. These problems have made many experts sceptical towards sodium cooling.

This situation has resulted in that other earlier rejected coolants like He-gas and in Russia liquid Pb-Bi are revisited in studies of future fast reactors. The major interest in many countries is, however, focused on the exploration of the ADS. In principle the same coolants as for a critical fast reactor could be used for the subcritical reactor part of an ADS. The ongoing European study of an experimental ADS (PDS-XADS[75]) is evaluating both a liquid Pb-Bi cooled design[76] and a gas-cooled design. It seems that the preference will be the liquid Pb-Bi cooled system. The strong advocates for gas-cooling mainly come from the industry whereas the research community has a strong preference for liquid Pb-Bi cooling.

[75] PDS-XADS = preliminary design study of an experimental ADS; EC funded project to be finished in 2004 – see Sections 2.4.2 and 5.3.

[76] The mixture of lead and bismuth is an eutectic mixture with much lower melting point than lead. This mixture is often called LBE for liquid Lead-Bismuth Eutectic. In this report sometimes LBE and sometimes liquid Pb-Bi is used but with the same meaning.

The development and demonstration of ADS technology face several technological challenges:

- Understanding the behaviour of fuel and structural materials in complex and aggressive environments must be a prime R&D activity. The challenges posed by materials development are particularly harsh both for FR or ADS transmuters, where very high neutron fluxes, liquid metals, and high temperatures shall co-exist.
- The primary and secondary proton damage to fuel and structural materials close to the spallation source in ADS is an additional and essentially new domain that has only limited resemblance to the proton damage experienced in existing spallation target sources for neutron physics research.
- ADS systems may be expected to operate with high availability for 60 years. New equipment and components must be designed and tested to assure lifetime reliability and availability.
- The technological feasibility of high-power proton accelerators needs further investigation, especially in relation to the desired reliability.
- Accelerator reliability and trip-free performance are obviously required for a nuclear power system. Beam trips would cause thermal stresses in fuel and structural materials, and if frequent, cause low cycle fatigue failure of the structures to become a safety and operating issue.
- The proposed use of relatively new types of coolants (lead, LBE or gas), especially in combination with new fuel types and cladding materials, demands specific attention to the thermal-hydraulics and core mechanics in normal and abnormal conditions.
- New techniques and tools must be developed to control the coolant chemistry, the subcriticality level monitoring (by beam power or moving control rods), the coupling between power level and accelerator beam power, etc, and especially to increase the reliability of the accelerator.
- In-service inspection, maintenance and repair, in particular reliably and economically.

Development of fuel and waste management strategies for a P&T system is also an area with many key items for R&D:

- Quantification of total long-lived radioactivity generated in the ADS transmuter system, including spallation products from the ADS, and the implications for waste streams and waste forms from partitioning, recycling, operation and decommissioning will be needed. (A European project funded by EC – RED-IMPACT – taking a first general look on these aspects has been just started – see Appendix I.)
- The impact of these waste forms on the design, performance, operation and costs of the deep repository should be evaluated in order to assess the potential benefits of transmutation in these respects.

- The fabrication of dedicated fuels needs intensive further research in particular development of fuel containing high fraction of TRU and/or minor actinides. While oxide fuel forms may be used in the short term, others such as nitride and metal may be more suitable in the longer term. A particular challenge is development of fertile-free fuel containing Am and fuel with high content of Cm. This may imply completely new designs of plants.
- In case of nitride fuel, specific processes are also needed in order to recover the costly enriched ^{15}N[77].
- Besides development of the fuel form itself, attention should be paid to the fabrication process, i.e. the need for dedicated fabrication plants, increased requirements for shielding and criticality control, optimisation of processes for small batch sizes, possible co-location of fabrication and reprocessing plants at an ADS site.
- Last but not least irradiation facilities with high fast neutron fluxes are needed for time consuming but very important irradiation validation of new fuels.

Integral experiments on ADS are needed in several scales to ultimately validate the feasibility of coupling of a proton accelerator with a subcritical core. Some very low-power physics experiments are already underway; the MUSE-experiment in France with fast neutrons supported by EC and at the Yalina facility in Minsk with thermal neutrons supported by ISTC. Both these use intense neutron generators. Under planning and preparation are experiments where existing proton accelerators are being coupled to low or medium power subcritical cores − Subcritical Assembly experiment in Dubna, Russia proposed for funding to ISTC, and the TRADE-experiment in Casaccia, Italy, proposed for funding to EC.

5.2 Key safety issues

5.2.1 ADS transmutation

The current evaluations of the safety of the LBE cooled XADS are generally favourable in the sense that the DBC and DEC transients have not been found to produce adverse affects. Moving from low thermal power to higher power machines with accompanying larger power density may produce adverse conditions for some DEC transients, in particular those involving failure to shut down the accelerator for a considerable time. The ability of natural circulation to remove heat in all situations, in particular the start and stability of natural circulation of LBE, which is a very high density liquid, may be a difficult issue. Loss of heat sink, in which the secondary coolant is lost or its pumping stops, coupled with natural circulation in the primary circuit has been found to be a transient, which could lead to core damage. Experimental data for this transient will be obtained in the TALL[78] facility for validation of the codes and for assessment of the consequences if there is a delay in the shut off of the accelerator.

The use of water as the secondary coolant in the heat exchangers of the ADS creates the hazard of a leak of water at high pressure into the pool of lead-bismuth. This event can create a pressure wave in the primary system and possibly transfer steam to the core

[77] The use of natural nitrogen with > 99% ^{14}N should be avoided as it reacts with neutrons to form ^{14}C.
[78] TALL = experimental facility for liquid Pb-Bi at KTH in Stockholm.

region, where it may generate a power spike. Effects of such events need to be evaluated, experimentally and computationally.

Perhaps the largest unknown factor in the safety area is the progression and consequences of severe accidents, which may involve melting of fuel and its interaction with the structures and the LBE in the core and vessel. In case the fuel (oxide) melts and floats on the LBE, it could accumulate in the heat exchanger, where if water is present in the primary side, it could go critical. Such severe accident scenarios and possibilities have still to be defined, studied and prevented or mitigated against. Currently, there is no information available for severe accidents in LBE-cooled ADS.

5.2.2 Partitioning

The total safety of reprocessing plants based on aqueous/organic solvent extraction is apparently acceptable today because several such plants are in operation around the world. However, recycling of MOX-fuel and eventual transmutation targets always poses special nuclear safety issues, the most important being criticality control in various parts of the plant. In general, criticality limitation has in the past been achieved by dilution of, e.g. MOX feed with a suitable amount of normal spent UOX fuel and/or addition of neutron poisons, like gadolinium, to the dissolver. In eventual new plants it is also possible to improve criticality control further by building, or lining, the new equipment in hafnium metal thus permitting a higher concentration of fissile nuclides in the process solutions at a given geometry. The major non-nuclear hazards in such plants are fires or rapid uncontrolled reactions between organics and nitric acid. Both accident types have occurred in the past, e.g. in Great Britain and in the former Soviet Union /Mus 92/.

The safety of pyrochemical separation plants not yet designed is difficult to assess. Most of the processes tested on a laboratory scale have shown a poor material balance, which could point towards unexpected deposits on equipment surfaces. It is not easy to predict if such losses are acceptable or dangerous. However, some general features can be pointed out that may be of safety concern. These are listed below for some types of unit operations typical of the pyrochemical separations now considered for use in the nuclear field.

Fluoride volatilization

Fluoride volatilization requires the handling of elementary fluorine, a substance reacting very violently with most other elements and most organic materials. Hence it poses a potential fire and explosion hazard. To avoid undesired reactions and fires the equipment handling fluorine has to be built from special nickel alloys, aluminium oxide and teflon. A drawback of teflon in a separation plant is the very poor radiation resistance of this material, which turns into a liquid already after a moderate radiation dose.

When using fluoride volatility it is very important to prohibit the leaking of oxygen and water vapour into the system. Should that occur, solid oxyfluorides of any gaseous actinide fluorides present are immediately formed as deposits on equipment and pipe walls. This can rapidly cause local criticality problems. Apparently this was one of the phenomena that caused the permanent shut-down of the Midwest Fuel Recovery plant /GE 66; GE 73; GE 74; Rog 77/. To prevent leakage into the system, the best would be

to operate above surrounding atmospheric pressure instead of below, however this would lead to a rapid release of radioactive material in case of any large leak.

Molten metal-molten salt redox extraction

The partitioning of various elements between a metal melt and a salt melt offers an elegant way to separate many elements present in spent fuel or irradiated transmutation targets using equipment somewhat similar to that used in some parts of aqueous reprocessing plants. In order to control the redox potential in such systems metallic lithium is normally used. Metallic lithium is very reactive towards oxygen and water, somewhat similar to metallic sodium. Hence molten lithium or melts containing metallic lithium can in principle always cause fires if they would leak out of the equipment used or larger amount of air leak into the equipment. The potential hazards are not totally different from those posed by liquid sodium cooled fast reactors.

Electrochemical plate-out

Most of the suggested pyrochemical separation processes uses electrolysis from molten salts as a means to recover the actinides in metallic form. Early designs used cathodes of molten cadmium, but newer designs use liquid or solid zinc instead. Cadmium is a relatively good neutron poison in contrast to zinc. One of the safety issues in electrolytic deposition from molten salts is the dendrites formed on the electrode surface. If these fall off and accumulate on the bottom of the electrolyser, criticality problems may arise with time.

5.2.3 Radiological issues

It has often been stated that P&T would necessarily increase the radiation dose to man considerably in the short-time perspective while perhaps reducing the potential dose somewhat in a very long time perspective.

Transport of spent fuel and radioactive waste

Due to weight and capacity limitations on radiation shielding during transport, and the necessary handling, it is very likely that the doses from such transports will depend on the amount and on the distance that spent fuel, high level waste, and other highly radioactive materials have to be transported. Thus, from a purely radiation protection perspective the best alternative would be to locate reactors, reprocessing plant, separation facilities, fuel and target fabrication plants, separation and transmutation facilities, and the final waste conditioning and disposal facilities in close proximity. From an economic point of view, considering scaling effects, and other practical reasons, a complete co-location is not very likely. Hence one would expect that the total radiation dose to man from P&T would increase in comparison with direct disposal of spent fuel if current techniques and methods are used due to an increased amount of highly radioactive material that will have to be transported over a longer distance. However, the relative increase will depend on eventual improvements in radiation shielding during transport, in transport methods, and in modes of handling.

Reprocessing of spent fuel

Reprocessing plants are very large and existing industrial units. It is unlikely that the radiation dose to man from operation of these existing facilities will decrease dramatically for a given amount of fuel processed. However, the releases to the environment of radioactive nuclides from such plants can be reduced considerably if more elaborate – and more expensive – methods for cleanup of low level waste streams are introduced. The techniques for this certainly exist, but are regarded as too costly today. As an example, off-gases from the dissolver can – after removal of water and nitrous oxides – be condensed to liquid form, distilled, and the elements containing radioisotopes, mainly Kr and Xe, recovered and stored as compressed gas in cylinders in a special medium time waste storage facility until their radioactivity has decayed to an acceptable level. The removal of Kr and Xe from dissolver off-gas by several methods has been tested on a pilot scale and also used on an industrial scale at the Idaho Chemical Processing Plant /Ben 81; Mer 72/. Currently, the removal of Kr from reprocessing off-gas is mandatory in the US /Ben 81/. Should new large industrial reprocessing plants be built to satisfy the requirements of a large scale P&T strategy, their radiological impact can be reduced in comparison with now existing plants – if desired – as far as economy permits.

Partitioning

The primary recovery of minor actinides and eventual other elements considered for transmutation should be made in plants co-located with the existing reprocessing plants or being an integral part of the reprocessing plant if new such plant have to be built. Reagents fulfilling the CHON-principle have the potential to reduce the amount of medium level and low level waste in comparison with the PUREX process. The extra radiation dose from the partitioning operation depends mainly on economic factors, but can from a purely technical point of view be made as low as desired.

In case of pyrochemical processing of spent fuel and/or irradiated targets principles similar to those valid for aqueous processes apply. Equipment repair or replacement is expected to be the operations that yield the highest doses to man in both aqueous and pyrochemical processes. It is not unlikely that the practical life of pyrochemical equipment will be shorter than for equipment used in aqueous processing – most of the latter being designed without any moving mechanical parts and expected to last some 30 years or more. However, it is still unclear to what extent, and how frequent, equipment repair and/or replacement will be necessary in any new pyrochemical plant and if this can be accomplished purely by remote control or not. The operating experience from pyrochemical separation plants in the nuclear field is rather limited and thus it is difficult to predict how easy it will be to keep doses to personnel very low.

Partitioning of recycled transmutation fuel can be made in separate plants or by recycling these fuels to the normal reprocessing operations. In both cases the extra radiation doses to man and environment from partitioning depends mostly on what is economically justifiable in terms of radiological protection.

Fuel fabrication

It is clear that the fabrication of transmutation fuel must be made in remotely operated plants. The extra radiation dose to man and environment depends mainly on the amount of radiation shielding used and on the possibility to use remote maintenance of all equipment. In both these cases, the economy will be the most important factor which will limit the possible dose reduction.

Transmutation

Transmutation of minor actinides can be made either in fast reactors or in special ADS systems. Both types of plants have to be designed and newly built for the transmutation of suitable targets.

The radiation doses to man and environment from fast reactors of conventional design are relatively well known. Should it turn out to be desired, these levels can probably be further reduced, but at a considerable economic cost.

Because no large scale ADS-systems have ever been built and operated, it is difficult to assess the radiation doses to man and environment from such plant. It is also not well known how far, and by what means, the doses from operation, maintenance, and releases to the environment can be reduced. It is however clear that the large accelerator facility requires substantial radiation protection measures in addition to those needed for a common nuclear reactor. It is evident that a trade off between protection and economical considerations will also in this case define the measures taken.

Waste handling

The transmutation of long-lived nuclides into shorter lived ones must in principle increase the radiation level from the waste in comparison with the original un-transmuted targets. Hence, handling and disposal of transmutation waste could perhaps require more elaborate remote handling techniques than needed for spent fuel. On the other hand, the radiation level of such waste is expected to decrease relatively fast and an intermediate storage for some decades could reduce radiation doses from the final disposal operation.

Conclusions

If the radiation doses to personnel, and the releases to the environment, are kept as high as permitted in all the operations needed in a complete P&T system it is obvious that P&T will result in a considerable increase in the short-time radiation dose to man and in radioactive releases to the environment. This is partly compensated by a reduction of potential future doses.

On the other hand, the new operations and plants needed in a P&T system might be designed with the objective to obtain much smaller radiation doses to man and smaller releases to the environment than the maximum permitted ones. Hence it is not a necessity that P&T will implicate any significant increase in short-time radiation doses or in releases to the environment.

5.2.4 Proliferation

The management of any fissile material as uranium, plutonium, spent nuclear fuel etc raises some concern for illicit use of the material for nuclear weapons proliferation. The use of transmutation technology will need reprocessing of the spent reactor fuel. An important factor for the abandoning of reprocessing in USA in the late 1970ies was the concern for such proliferation. This concern will be an important factor when assessing the feasibility of this transmutation.

There are several aspects of proliferation to consider in the assessment. The first is the separation of plutonium from the remaining uranium and from the fission products. This makes the plutonium more accessible as the gamma-radiation from the spent fuel

(almost) is eliminated. The plutonium is relatively easy to concentrate and thus easier to transport or divert. The safeguards measures for separated plutonium must therefore be much stricter and comprehensive. The other aspect is that the plutonium will be taken out of the high level waste that shall be placed in a deep repository. The risk for future recovery of plutonium from the spent fuel and to use it for weapons purposes does then not exist anymore.

Several ways to make the fuel cycle of transmutation systems more proliferation resistant have been proposed. They are based on avoiding element-specific separation. One of the approaches aims at co-processing plutonium and minor actinides. After initial separation of the uranium from the LWR spent fuel, the actinides are recycled in transmuters with a closed fuel cycle using pyrochemical reprocessing without further actinide separation.

Another strategy to reduce proliferation concerns can be to design a transmuter (or reactor) with a very long cycle without refuelling. This strategy, however, is difficult to implement for a fertile-free fuel concept.

In practice it probably will cause some technical problems and also increased costs to avoid separation of plutonium from the other actinides. This is of course the case for the fuel cycle strategies, which plan for burning plutonium in LWR or in FR and use ADS only for burning of the minor actinides. One may also argue that having developed and deployed partitioning processes which separate transuranium elements from uranium and fission products there is a risk that someone wants to go one step further and separate the transuranium elements from each other. This can be done in much smaller scale than the other separation.

The issue of proliferation and safeguards is an intrinsic part of the nuclear fuel cycle and it is no guarantee that any brand of P&T may make this issue any easier to handle than in the current situation. Neither are there any strong arguments that it should be worse.

5.3 ADS demonstration plant

As described in Chapter 2, so-called roadmaps for development of the ADS were published in USA 1999 and in Europe 2001. Both these studies emphasise that the first major step towards the implementation of an ADS should be the construction of a demonstration facility of some reasonable minimum size. The US study proposed a system (called ATF) with a linear accelerator, a spallation source and a liquid Pb-Bi-cooled subcritical reactor with a power of 30 MW_{th}. The estimated time for design and construction of this facility would be nine years after the initial six years of basic R&D. The European study does not define the basic design of the major plant components but leaves some option open for accelerator and for coolant. The thermal power envisaged for a European experimental ADS (XADS) is about 80 MW and the target date for start of operation given in the study is incidentally 2015 or 15 years after programme start.

The US programme has not yet opted for construction of an ADS demonstration facility and it remains to be seen what place this route will be given in the US programme.

Part of the ADS R&D strategy outlined in the European roadmap has been already adopted by the European Commission in their 5th FWP as well as in the call for proposals in their 6th FWP.

The 5th FWP project PDS-XADS studies the preconceptual design of the experimental ADS-plant with the objectives to be able to identify the most promising technical options, to define the corresponding R&D needs and to reassess the cost of the facility. Three different ADS systems are investigated:

- Two 80 MW_{th} ADS – one Pb-Bi cooled ADS and one gas-cooled ADS.
- One smaller (50MW_{th}), multipurpose design based on the Belgian MYRRHA project.

The PDS-XADS-project addresses important issues necessary to be resolved in order to be able to select one specific conceptual design of an ADS. A result and conclusion of the PDS-XADS project is a list summarizing the most important conclusions and/or concerns to be taken into account for the conceptual design of a European XADS. All the current results of the PDS-XADS project together with the results of other ADS-related European projects indicate that a conceptual design of the XADS intended to be prepared during the 6th FWP will focus on a liquid Pb-Bi cooled ADS at about 100 MW_{th}.

Neither the US nor the European research programmes follow the aforementioned roadmaps which were prepared a few years ago. This means that the date for decision on the construction of any ADS demonstration plant has been delayed in comparison to the proposals. It is, however, clear that any serious effort towards the use of ADS for transmutation must include such a demonstration of the system at an early stage in the development.

In Europe it is obvious that national and other interests play an important role in the selection of projects that are funded under the European Union FWPs. It has to be remembered that 50% or more must come from other sources than the EU budget. This implies that the prospects for any strong and coherent efforts to speed up the route to an ADS demonstration plant are rather poor in the current situation. In the US the competition from the so-called Generation IV programme will probably be a major roadblock for the rapid development of ADS.

5.3.1 Design of ADS demonstration plant

The current set of European projects developing the ADS design and technology is centred in the FWP5. The results achieved so far have recently been evaluated in order to develop the outlines of a large Integrated Project (IP) for FWP6 named EUROTRANS. This proposed project is based on the significant conclusion that the focus of the future ADS design will be on a LBE-cooled ADS to be developed in two steps. First an experimental system, with power rating of 80 to 100 MW_{th}, would be designed. This could be based on the Belgian MYRRHA design concept. The second step would be design of a larger scale system with the thermal rating of 250 to 300 MW_{th}, which would fully employ minor actinide fuel. The overall objective is to demonstrate that transmutation of the minor actinides can be achieved. No economic targets have been specified for any of these proposed systems. It should be noted that the gas-cooled and lead-cooled concepts would be regarded as backup solutions.

It is needless to say that the LBE cooled ADS will be a complex and hi-tech machine. There is no experience base presently in the West, with manufacture, construction and operation of a LBE-cooled reactor system. The Russian experience has been only with non-commercial applications of a critical reactor system employing LBE-cooling with

unknown safety and regulatory standards. Thus, the task is challenging. The studies pursued so far in the FWP5 projects have not identified any 'show-stoppers', but a number of key design, technical, material, operation and safety issues, have been identified and resolution (solution) for many have been proposed or achieved. The following sections briefly describe the various components of this topic and indicate the key issues for each of these.

Primary coolant system

For the LBE-cooled ADS, a pool-type configuration is used in order to contain the primary coolant in a simple boundary (the reactor vessel). This eliminates all the problems related to out-of-vessel primary coolant. In the Ansaldo design, enhanced natural circulation is employed to circulate the LBE. However, the feasibility and efficiency of a gas-lift pump are getting doubtful because of the difficulty of operation. More attention is now switched to the application of mechanical pumps.

The primary system of the LBE-cooled ADS subcritical reactor will have the following main components:

- In-vessel fuel handling system.
- Exchangers.
- Main and safety vessel.
- Reactor internals.
- Proton beam interface.
- Reactor vessel air cooling system.

Secondary coolant system

Compared to the primary system, the secondary system is not so sophisticated. Its purpose is to cool down the primary coolant in intermediate heat exchangers or steam generators. A key issue is how to choose the working fluid for the secondary system.

In order to avoid the violent interaction of the LBE with high-pressure water, the Ansaldo design does not use water as the secondary coolant. Instead, a high temperature organic fluid (called Diphyl-THT with the boiling point of 355°C) is employed at atmospheric pressure. The fluid has a low vapour pressure. It has been claimed that no violent interaction exists between the LBE and Diphyl-THT. Moreover, in order to be isolated from the power grid and to facilitate the experiments, the Ansaldo design does not have steam generators to produce steam and electricity. Air coolers are used to dissipate the heat to atmosphere.

The XADS will not produce electricity; however, an industrial scale machine should produce electricity to be economic. Thus, the designs in FWP6 will consider electricity as a product for the future ADS. Therefore steam generators should be designed. Perhaps it is not a good idea to directly position the steam generators into the LBE pool and hence an intermediate loop should be designed. An alternative concept is to employ double-wall tubes in steam generators as was done in the EBR-II plant. The final solution should be the result of extensive investigations, and the experience of FBRs and LWRs.

Core design

The core design of an LBE-cooled ADS employs the classic core design disciplines, i.e. neutronic, thermal-hydraulic and thermo-mechanical design.

For the neutron source characterization, a theoretical basis was developed for several neutronics parameters, which may be used to characterise subcritical systems that are driven by an external neutron source. The interface between the external neutron source and the reactor core was defined in such a way that in both deterministic and Monte Carlo analyses a consistent external neutron source be used.

The LBE-cooled core is shaped as a hollow cylinder of a few rows of fuel assemblies arranged in a honeycomb-like pattern. The fuel assemblies surround the inner cavity, which houses the spallation source unit. The outer fuel assemblies are surrounded by a sort of buffer region, which consists of a few rows of dummy elements. The purpose of this buffer region is to soften the neutron spectrum affecting the internal vessel structures. The ADS core must remain subcritical under any circumstance, which may range from normal operation to accident conditions, i.e. the Design Base Conditions (DBC) and the Design Extension Conditions (DEC). The operational subcriticality range is established in order to stay away from criticality ($k_{eff} = 1$) with adequate margin under normal operational conditions so that postulated accidents (DBC and DEC), which may lead to large temperature changes and positive reactivity insertions, may also be accommodated without attaining criticality. The core multiplication factor for normal operation at full power and BOL is set at k = 0.984–0.014 = 0.97 (i.e. a subcriticality margin of 3%). A 0.97 core multiplication factor is considered sufficiently low to ensure the safe operation of the LBE XADS without need of shutdown rods.

For the XADS study, Superphenix (SPX) region 2 fuel pellets (~ 20% Pu enrichment) are used for the fuel assemblies loaded in the inner region, while SPX type fuel pellets with higher Pu enrichment are used in the remaining fuel assemblies loaded in the outer region of the core.

The core design challenges for the future (FWP6) LBE-cooled ADS will be enhanced if a fully minor actinide fuel core is to be installed. It is known that a minor actinide fuelled core may have very small values of the effective delayed neutron fraction, of the Doppler and of the coolant density coefficients[79]. It is also clear that these reactivity coefficients are difficult to calculate with sufficient accuracy, due to the inaccuracies of the basic nuclear data at high energy. The progress made in nuclear data measurements may be sufficient for reasonably accurate determination of the reactivity coefficients. Even if the reactivity coefficients are known with adequate accuracy, the control of the ADS core fuelled with minor actinides would be challenging.

Accelerator

Present-generation accelerators are low-powered and not designed to be coupled to a subcritical reactor. Their availability and beam stability are poor and they need to be substantially improved for ADS application[80]. A superconducting linear accelerator, composed of independently powered superconducting cavities, and a doubly achromatic beam line penetrating the reactor vertically from above has been assessed to be the most suitable concept. This generic solution is valid for both the gas-cooled and LBE-cooled ADS concepts.

[79] See also Sections 4.1.3 and 4.1.4 in this report.
[80] See also Section 4.1.1 in this report.

For the XADS, the required beam characteristics are: 600 MeV final energy, 6 mA maximum beam current for operation (10 mA for the demonstration of concept), very high reliability (less than a few beam trips per year) – see further in Section 4.1.1. A 350 MeV proton accelerator has been specified for the MYRRHA design concept.

The future (FWP6) design most probably, will be a 600 MeV superconducting linac /Mueller 03/. The key issues for the accelerator still remain to be (i) the high current needed and (ii) its stability and reliability.

Spallation source unit

Three different spallation source concepts have been proposed and investigated: a window source configuration with a liquid source (LBE) for LBE- and gas-cooled core concepts and a windowless source configuration with a liquid source (LBE) for the LBE-cooled core concept. Some evaluation of a solid source for the gas-cooled core concept has been made[81].

The window source was the preferred concept at the beginning of XADS R&D. A lot of activities have contributed to the demonstration of a window source. For example, the MEGAPIE project is focusing on theoretical and experimental studies on the window source design. It is found that one technical difficulty in window source design is that the membrane at the window cannot withstand the high temperature due to the irradiation of the proton beam.

As a result, recently a windowless source is now preferred for the LBE cooled XADS. The design is still at a premature stage and the details are not clear. Extensive investigations on the windowless source are needed.

In the beam line very high vacuum conditions are desired in order to avoid the secondary plasma formation from the interaction of beam with gases. However, at the free surface of the "windowless target", LBE evaporation due to the hot spot and emanation of spallation products will sustain some gas/vapour pressure. Thus, key issues in the windowless spallation source design are the maintaining of sufficient vacuum conditions in the beam region by a vacuum pumping system and the safe collection of radioactive spallation products through the vacuum system.

LBE chemistry and control

Lead-bismuth eutectic (LBE) is used as both coolant and spallation material of the XADS because LBE has exceptional chemical, thermo-physical and neutronic properties and is well suited for nuclear coolant and spallation material applications. However, LBE is not fully chemically compatible with common steels used in nuclear installations, which therefore requires specific protective measures against corrosion.

Among the corrosion protection measures, "in-situ" oxidation and artificial coatings, were studied and it has been shown that the "in-situ" oxidation of EU nuclear grade steels (both austenitic and martensitic) show a "critical" temperature above which the protection is not perfectly effective. Conversely, some coatings (mainly Fe-Al alloys) show an excellent resistance in the liquid metal even at high temperatures in oxidising environment. With the progress of technologies, a new generation of oxygen sensors and systems capable to control the oxygen content in the LBE are being developed and qualified. Since the oxygen control can effectively establish and maintain a protective

[81] See also Section 4.1.2 in this report.

film on the steel surface, it is expected that an oxygen control system will be provided in the LBE-cooled XADS. Oxygen control at large scale and for large durations, however, has not been demonstrated so far.

The corrosion also produces some solid impurities, which will contaminate the LBE. So, for the purpose of the control and management of the solid impurities, dedicated devices and systems need to be developed to take away the solid impurities. Some reliable instruments and a better knowledge of aerosol and solid slugs generated by LBE corrosion are becoming available for small systems

Clearly, chemistry and corrosion control of LBE system is a large concern during the operations. The restrictive temperature regime of operations (allowed maximum high and minimum low) does not allow abnormal regimes of operations. The operational flexibility is not very large.

Materials issues

As mentioned above, LBE is not fully chemically compatible with common steels used in nuclear installations. Furthermore, the irradiation characteristics, especially at the spallation source unit, are different from any previous reactors. Thus tests and evaluations are necessary before the material is used with LBE[82].

So far, martensitic steels (T91, EM10 and 9Cr2W) and austenitic steel AISI 316/316L have been extensively tested for the LBE application /Benamati 03/. The “in situ” protection oxidation technique has proved to be effective to protect these materials against the LBE corrosion up to a temperature of 535°C. Aluminized coating can protect the Stainless Steel 316 L and the Ferritic Steel T91 up to a temperature of 600°C. For the T91, however, aluminization seems to be difficult to apply. For protective film, the temperature limit is fixed at ~ 500°C. “In situ” protection technique for the materials is suggested as the recommendation when the LBE flow velocity is expected to not exceed 2 m/s. It is not clear whether the aluminization will affect the mechanical properties.

The degradation of the mechanical properties of irradiated T91 steel has been addressed. The effects of spallation elements (Ti, S, P, He, Ca) that can be produced in the beam window made of T91 steel were investigated. The production of He, Ca, Ti, S in the martensitic steel is simulated by ion implantation, and the associated effects on microstructure and mechanical properties are studied. It is shown that the consequences of He on the tensile properties of the steel are strongly dependent on the implantation temperature.

Data on microstructure and mechanical properties after irradiation under a prototypical neutron and proton mixed spectrum have been collected as well. In that case, martensitic steels (T91, EM10 and 9Cr2W) are irradiated in the temperature range from 90 to 350°C for doses up to 12 DPA. The associated tensile and impact tests performed on the irradiated specimens prove that the magnitude of hardening and the associated loss of ductility increase with the dose even if a ductile fracture mode is obtained in general.

[82] See also Section 4.1.5 in this report.

Based on the previous study, as reference structural materials the austenitic steel AISI 316L will be considered for the vessel components and the martensitic T91 steel for the fuel claddings and the target window. However, further studies on the structural materials for target, vessel, core components and fuel claddings in terms of corrosion kinetics, corrosion protection and mechanical properties degradation (e.g. fatigue, creep, etc) with and without irradiation are needed.

A great amount of theoretical and experimental investigation is slowly contributing to the resolution of the material issues. More than half the tasks of the project TECLA (Technology, Materials and Thermal-Hydraulics for Lead Alloys), for example, focus on the material issues.

Summarizing, it appears that the 316 and T91 stainless steels would be suitable if an adequate oxygen control is maintained and temperature limits are observed. The LBE velocity of 2 m/s is another limit to watch. Overall, the restrictions imply strict controls on steady state and transient operation. The decrease in material ductility, due to irradiation, coupled with chemical reactions may limit the life of some components, in particular the target.

Thermal-hydraulics

Thermal-hydraulics is an important issue for the ADS design. Due to its high atomic number and high density, the characteristics of LBE flow and heat transfer will be different from the performance of the well-known coolants water and sodium. Better understanding, in this respect, is needed for the thermal-hydraulic design of an LBE-cooled XADS. There are not sufficient published data available for the LBE heat transfer, fluid flow, pressure drop, etc, for the parameters and systems of interest.

During the FWP5 some thermal-hydraulic/multi-purpose test facilities (such as CIRCE, KALLA, TALL)[83] have been designed and commissioned. It can be expected that more data will become available from their experiment campaigns.

CFD[84] simulation is also very active in the LBE thermal-hydraulic study. The project ASCHLIM (ASsessment of the Computational fluid dynamics codes for Heavy LIquid Metal) uniquely focus on the CFD part. It is clear that the CFD plays an important role in the LBE thermal-hydraulic study. The main tools are the commercial CFD codes. However, detailed data needed for validation of the CFD codes for heavy liquid metals, currently is not available.

The thermal-hydraulic experimental and computational studies have focussed on the spallation source unit and the primary system of the ADS. The key issues to be addressed are:

- Free surface flow at the windowless spallation source unit.
- Recirculation at the windowless spallation source unit.
- Flow resistance and heat transfer in fuel rod bundles.
- Natural circulation: its stability, time for its initiation, its potential for decay heat removal, etc.
- Temperature and velocity profiles both in core and in spallation source unit.
- Interaction of the LBE with the secondary loop coolant.
- Heat exchanger thermal hydraulics.

[83] CIRCE at Ansaldo in Italy, KALLA at FZK in Germany, TALL at KTH in Sweden.
[84] CFD = Computational Fluid Dynamics.

- Integral investigation (natural circulation, transients for decay heat removal, IHX/SG tube rupture, …).
- Thermal hydraulics coupled with oxygen control and solid impurities formation and removal.
- Measurement techniques development for LBE temperature, velocity, flow rate, level, etc.

The available facilities in Europe will contribute data for increasing the knowledge-base for the above topics and resolving some of the doubts about efficiency of heat removal. The experiments, however, are difficult to perform at prototypic conditions of temperatures of ≥ 500ºC.

In-service inspection and repair

In-Service Inspection and Repair (ISI&R) is another important issue for the ADS since the associated provisions allow to ascertain that the components involved in the safety demonstration meet their design specifications during their life time (including their availability) and that troubles can be detected before they become significant. Their repairability also has to be analysed. ISI&R approach is also used to meet reliability assessment objectives (optimization of the maintenance and inspection program, protection of plant investment and plant reliability). ISI&R systems for the LBE ADS have not been properly addressed so far. Perhaps robots would be needed and if so will have to be designed for operation in quite severe duty.

5.4 Other demonstration facilities

Besides a demonstration facility for the actual ADS transmuter, there are also needs for demonstration of other major facilities of a future P&T-system.

One important area is the production of fuel. Today, there is no laboratory for production of large quantities of fuel with americium admixture. At the Atalante laboratory at the CEA installation in Valrhô-Marcoule, France, americium targets (AmOx dispersed in MgO) are produced for irradiation in the Phénix reactor /Clefs 46/. This facility can be used for handling americium in moderate quantities. For curium, the situation is even more difficult because of the strong neutron emission. At present, only gram quantities can be handled. Therefore, CEA has stressed the need for development of techniques for handling significantly larger quantities, and envision a demonstration facility before going to full-scale production of fuels rich in americium and/or curium /Bouchard 03/.

Both the roadmaps presented in Chapter 2 advocates the advantages of pyrochemical reprocessing of the irradiated fuel from an ADS. Similar arguments can be raised for irradiated fuel from a critical fast reactor.

The US roadmap proposes as a necessary step the construction of demonstration spent fuel/ADS fuel reprocessing, fuel/target fabrication and waste management facility (FTF). This should be developed and constructed in parallel to the ADS demonstration facility (ATF).

The European roadmap proposes the design and construction of a small-scale pyrochemical reprocessing plant to be started around 2020.

5.5 Time and costs

As has been pointed out by several of the systems studies described in Chapter 2, the time for development, construction, deployment and operation of any P&T system is long compared even to the currently assessed lifetime of a nuclear power plant. The studies give numbers of about 30–35 years for research, development and demonstration and another 90–100 years for construction and operation before the stock of spent fuel from presently existing nuclear power plants have been processed and transmuted. These times are probably on the optimistic side assuming a fairly rapid commitment for intensive development of P&T. The time frame is in general not very much dependent on the technical route that is selected. Given the current state of knowledge one would, however, believe that fast sodium-cooled reactors could be deployed earlier than an ADS-plant or liquid Pb-Bi cooled fast reactors. Gas-cooled fast reactors might need something in between the other alternatives. It is unlikely that industrial deployment of ADS will occur before 2050.

A number of assessments of the costs for P&T in particular of ADS transmutation has been published in the last few years.

The US roadmap study includes an estimate of the costs for transmuting 87 000 tonnes of spent nuclear fuel from the current US LWR park. The costs are summarised in Table 5-1.

The report also points out that the 279 000 million US-dollars are about the same amount as the assessed value of the electric energy that the system would produce.

The assessment of R&D costs for ADS made by TWG for the European roadmap study have been summarised in Table 2-2. They are M€ 980 for the first 12 years. This agrees incidentally fairly well with the M$ 170 plus M$ 1000 for the accelerator plus transmuter from the US study in Table 5-1. From Table 2-2 one should also note that the costs for the 6th FWP are estimated to M€ 255 and for the 7th FWP to M€ 550. This is far above what might be available from EC funding. The funding under FWP 5 was M€ 28.6 and the expectation from FWP 6 is about M€ 30–35. This means that most of the financing for such an ambitious programme must come from national funding by the EU members.

Table 5-1. Total system life-cycle costs by system phase and system element. All numbers in the unit 1000 million (10^9) undiscounted 1999 US-dollars.(R&D-phase 8 years; Demonstration phase 19 years; Implementation phase 90 years. Some details of the assumed system are give in Section 2.3) After /DOE 99/ page 7.3.

Specification	R&D	Demonstration	Implementation			Total
			Capital	Operation	D&D	
Accelerators	0.17	3	11	44	0.6	59
Transmuters	1.0	2	30	49	3	86
Separation	0.5	2	9	41	1	53
Fuel fabrication	0.0	0.6	2	41	0.2	44
Site support	0.0	1	1	31	0.1	33
Retrieval+Trpt+Disposal	0.0	0.1	0	4	0	4
Integration	0.07	1	0	0	0	1
Subtotal	2.0	9	54	210	5	279

Another type of cost assessment was made by the OECD/NEA expert group comparing ADS and fast reactors in advanced fuel cycles /NEA 02b pp 205–230/. This study has been summarised in Section 2.6. They compare the costs for the different fuel cycles shown in Figure 2-3. The result is summarised in Table 5-2. The costs are given as "cost of electricity" = COE in US mills/kWh. The costs were calculated for fuel cycles with equilibrium flow of materials in each of the cycle scenarios analysed. The table also gives relative cost values R_{cost} compared to the once through cycle with $R_{cost} = 1$. Included in the table is also a comparison of what is called the loss of transuranium elements to the repository R_{loss} where the once-through fuel cycle has $R_{loss} = 1$, meaning that all TRUs are sent to the repository. R_{loss} is thus a measure on how well the transmutation may succeed in the different advanced fuel cycles.

One observation from this table is that the use of ADS seems to substantially increase the cost without really improving the effectiveness of transmutation. Another observation is that double strata fuel cycle with burning plutonium in LWR and FR and only the minor actinides in ADS may be a more viable option considering the decrease in some safety margins when burning minor actinides fast reactors.

A similar cost analysis for ADS transmutation fuel cycles has been made by Westlén at KTH /Westlén 01/. He compares the once-through cycle with two transmutation cycles using an ADS core design proposed by KTH. One of the transmutation cycles presumes that all TRU-burning is made in ADS (as is the case also in the US roadmap study). The other case assumes that plutonium is recycled in the LWR. Some results are shown in Table 5-3.

Table 5-2. Comparison of electricity costs and transmutation effectiveness for various fuel cycles. From /NEA 02b p 219/.

Fuel cycle	COE US-mills/kWh	R_{cost}	R_{loss}
Once through	38.0	1.00	1.000
Pu-burning LWR+FR	40.7	1.07	0.288
TRU-burning in FR	42.4	1.12	0.0035
TRU-burning in ADS	53.5	1.41	0.0057
MOX-recycle + TRU-burning in ADS	49.4	1.30	0.0055
Double strata	44.2	1.16	0.0047
All FR strategy	56.9	1.50	0.008

Table 5-3. Comparison of electricity for various fuel cycles. From /Westlén 01 p 50/.

Fuel cycle	COE US-mills/kWh	R_{cost}
Once through	24.5	1.00
TRU-burning in ADS	37.2	1.52
MOX-recycle + TRU-burning in ADS	33.4	1.38

All these cost studies are of course at this stage very uncertain. They give, however, one or two common messages. A necessary economic prerequisite for P&T is that the energy released by the transmutation reactions is being used for electricity production or for some other useful purpose. Further, the recycling of plutonium in LWR (or possibly sometimes in the future in FR) is substantially less costly than to burn all the transuranium elements in ADS.

5.6 Impact on waste management programme and nuclear power development

As pointed out in the introduction to this report, the objective of partitioning and transmutation is to eliminate or at least substantially reduce the amount of long-lived radionuclides that has to go to a repository for final disposal. This implies that a successful outcome of the ongoing research and development on P&T may have a considerable impact on in particular future waste management programmes but also on the general development of nuclear power and nuclear energy.

The future introduction of a P&T-system would have *inter alias* the following consequences for a waste management system in comparison to the current once-through fuel cycle for LWR:

- The waste form spent nuclear fuel would be replaced with other waste forms. These would contain only a small fraction (less than one percent) of the (long-lived) transuranium nuclides.
- The "new" waste forms would still include the main part of the fission products, in particular cesium and strontium. These would be included in some stable matrix as vitrified high level waste. The volumes of packaged high level waste could probably be considerably smaller than the volumes of packaged spent nuclear fuel.
- There would also be other waste forms from the partitioning processes similar to intermediate and low level waste from reprocessing. Furthermore, there would be waste streams from the handling and fabrication of nuclear fuels or targets with the transuranium elements and any other nuclides to be transmuted.
- In addition there will be radioactive waste from activation products in used and decommissioned equipment of the transmutation plant. The waste forms would be similar to those from operation and decommissioning of nuclear reactors.
- Some or all of the aforementioned waste forms would have to be placed in a deep geological repository. As the total amount of transuranium elements in waste is reduced by a factor of 100 or more compared to LWR spent nuclear fuel the demand on the barrier system in the repository is correspondingly less. This implies that the barrier system can be simplified. If higher temperatures can be permitted due to the different waste forms it may also be possible to further reduce the volume of the repository,

The used uranium from the spent nuclear fuel would have to be treated as waste or be recycled in some type of nuclear reactor. If the uranium is treated as a waste it would have to be placed in some kind of repository with due consideration to the eventual in-growth of radium and its daughters. If the uranium is recycled, it will mean a more efficient use of the uranium that was originally mined, but also the creation of more transuranium elements that may have to be transmuted. In general, burning or

transmutation of plutonium and other transuranium elements implies increased efficiency in utilisation of the mined uranium. The ultimate step in this respect is the use of breeder reactors offering the potential to get on the order of 100 times the energy per unit of mined uranium compared to what is obtained in LWR by the once-through cycle.

The implementation of P&T would of course also have an impact on the costs of nuclear power. This issue is discussed in Section 5.5.

Research and development on P&T has an impact on the general development of nuclear science. It involves interesting problems in many disciplines, e.g. nuclear physics, reactor physics, nuclear chemistry, materials science, nuclear engineering and nuclear fuel design. The challenging problems and goals have already increased the attraction for the nuclear science disciplines at the universities. This contributes to a broadening of competence that may be of benefit to all nuclear energy development, in particular for safety and performance of nuclear fuel and waste management. In the short term this may turn out to be an important impact for the current nuclear power programme.

A project REDIMPACT, partly financed by the European Commission within the sixth FWP, has recently started. The project will assess the waste streams and impacts in terms of waste forms, waste volumes, costs, environmental effects, etc for various nuclear fuel cycles. This project is scheduled for completion by the end of 2006. It is coordinated by the nuclear and reactor physics department of KTH, Stockholm and includes 23 different organisations (among them SKB) from 10 countries.

An implementation of P&T in Sweden could hypothetically follow any of several different scenarios. As long as construction of new nuclear power facilities are not allowed in Sweden a feasible scenario is the shipment of spent fuel from the existing reactors to some other country for reprocessing followed by burning of the transuranium elements in a foreign transmuter and if necessary reshipment of the conditioned wastes for final disposal in a Swedish repository. Another possibility is the recycling of plutonium in (some of) the existing reactors applying some of the multi-recycle concepts recently suggested – see Sections 2.7.2 and 4.3. The minor actinides and the plutonium not suitable for LWR recycling could then be transmuted in a foreign transmuter. All wastes would go to a Swedish repository. A third scenario could be to change the law and permit building a few dedicated transmuters in Sweden. Assuming that plutonium is recycled in existing reactors applying some of the multi-recycle schemes it would be sufficient with an ADS park with a power of 1 GW_e for burning americium, curium and poor-quality plutonium. This corresponds to about three subcritical reactors. One could then also consider a dedicated separation facility for the treatment of spent ADS fuel on a relatively small scale (a few tonnes per year). This separation plant could be co-located with the ADS plants. There are of course also other more long-term scenarios that might be possible.

A common feature for all scenarios with full implementation of P&T is that they require a transition to more advanced fuel handling as well as continued use of nuclear power for at least one hundred years. A considerable effort in education of operators for advanced nuclear facilities would be necessary and create a set of very interesting jobs. Advancing the competence in the nuclear field certainly could be beneficial for sustainability of the nuclear power industry in general.

5.7 Acceptance of P&T

P&T-systems include several large nuclear facilities. The construction of such systems requires permits under several laws – in Sweden the more important laws are the Environmental code /SFS 1998:808/, the Act on Nuclear Activities /SFS 1984:3/ and the Radiation Protection Act /SFS 1988:220/. The current policy in several countries including Sweden is to not permit the construction of new nuclear power plants and/or nuclear fuel cycle facilities. A prime prerequisite for acceptance of P&T-facilities is that the necessary permits are obtained. Thus, in several countries a change of the current policy and law is necessary for the acceptance.

The acceptance of an advanced industry is strongly influenced by both the real and the perceived benefits and drawbacks of the industry.

The potential benefits from P&T are in particular:

- The elimination of > 99% of the long-lived transuranium elements from the waste to be deposited. These elements dominate the radiotoxicity of spent nuclear fuel for in the order of one hundred thousand years. Although the construction of deep repositories for the remaining waste will still be necessary, the construction might be easier and/or cheaper.
- The elimination of fissile plutonium that potentially might be diverted for use in nuclear weapons.
- The production of large amounts of nuclear energy that might be used for electricity production or other energy needs in society. This energy would amount to at least about 30% of the energy taken out of the fuel before, but potentially it could be more than 100 times the energy taken out of the uranium originally mined /NEA 02b/.
- P&T as a part of the use of breeder reactors for increased efficient use of mined uranium would give man a very long-term (thousands of years) sustainable energy source.

These potential benefits are balanced by some unavoidable drawbacks that must be mastered:

- The development of P&T to maturity for industrial deployment will take several decades – at least 30 years according to recent assessments. The necessary R&D will require large funds – of the order of M€ 10 000 /DOE 99; TWG 01/.
- The deployment of P&T will require substantially increased handling and management of large amounts of radioactive materials. This implies significantly increased potential radiation protection and nuclear safety hazards to be controlled.
- The siting of large-scale industrial facilities has been increasingly controversial during the latest three decades. This is in particular true for nuclear power, fuel cycle and waste management facilities. Today it would be very difficult in almost any country to get acceptance for building any P&T-facility on a new site. It might be possible to get acceptance on some sites that are already hosting a nuclear facility. Under current environmental legislation in many countries, acceptance by a strong majority of the local population is a prerequisite for the siting of any nuclear facility.

- Even if the funds for development would be available, there is still an uncertainty whether P&T will be technically mastered on an industrial scale and be commercially feasible.

In the long run, acceptance of P&T will depend on the perceived benefits versus the perceived drawbacks. These perceptions will depend on the evolution of society at large and on the evolution of the energy supply industry.

It is difficult to believe that the benefits of P&T could be perceived to be so great that it would gain general acceptance after a serious evaluation in countries where a policy of short-term phase out of nuclear power production prevails. The technology is very complex; it takes about a century to transmute the existing transuranium elements; the costs are substantially larger than for the direct disposal of the fuel; a deep repository will anyhow be needed; the technology will not be available until most of the phase out has occurred. On the other hand if the current plans for phase out are finally abolished, the lifetime of existing plants are extended and their replacement by new nuclear power plants will be allowed and seriously considered, the situation might be different.

However, in countries that already have a commitment to the long-term use of nuclear power, R&D on P&T may contribute to the development of more sustainable nuclear fuel cycles. P&T may then be a not absolutely necessary but prudent part of such a very long-term sustainable fuel cycle and waste management strategy.

It seems to us that the acceptance of deployment of P&T-facilities implies the acceptance of a continued commitment to the use of nuclear power. This means several generations of nuclear power plants beyond the currently operating plants. The acceptance of use of P&T-facilities may, from a Swedish perspective, also be possible if the services will be available under reasonable conditions in other countries or within international co-operation. Such use does, however, also imply acceptance of a commitment to this technology for the order of 100 years!

6 Some conclusions

6.1 General

The R&D efforts on partitioning and transmutation (P&T) have increased somewhat seen internationally during the period 1998–2003. In particular the financial support by the European Commission to P&T-projects within EU has increased considerably.

Research on P&T has taken a prominent role internationally in the R&D on future nuclear power and nuclear fuel cycle systems. It attracts considerable interest among students in nuclear science. Several important research programmes are going on at universities and research laboratories in several countries.

The interest for R&D on P&T in the nuclear industry is limited. It is mainly apparent in France and a few other countries with major nuclear power programmes.

6.2 Impact on nuclear energy and waste programme

Successful development of P&T will not make a deep geological repository obsolete. The complex processes will inevitably generate some waste streams with small amounts of long-lived radionuclides.

Successful development of P&T may, however, decrease the requirements on in particular the engineered barriers as well as on the volumes required in a deep geological repository.

For Sweden it is important to follow the international development and to maintain a reasonable level of competence in the country at least as long as we have a substantial part of the electricity produced by nuclear power.

Competence developed in P&T research is valuable and useful also in the work on maintaining and developing safety and fuel supply for the existing light water reactors. It is also of importance for assessing the further development of the waste management programme.

The implementation of partitioning and transmutation to effectively reduce the amount of long-lived radionuclides that must be placed in a geological repository necessitates a commitment to nuclear power for a very long time, at least over 100 years.

6.3 Acceptance

P&T on an industrial scale requires large nuclear facilities that must be accepted by society.

6.4 Partitioning

The major development efforts on partitioning are still concentrated upon aqueous processes. Pyrochemical separation processes attract considerable interest, but they may be reserved for special types of fuel with very highly radioactive contents.

The progress made in partitioning is promising. The goal to get a separation efficiency of 99.9% of the actinides from other elements in the spent fuel seems to be possible to reach.

A problem with the aqueous partitioning processes so far is the cost and stability of extraction agents.

Extrapolation of an aqueous partitioning process to industrial scale is rather straightforward once the flow sheet has been defined and tested on laboratory scale.

Pyrochemical processes needs further development. Before such processes can be extrapolated to industrial scale new types of equipment will require full scale tests with inactive and radioactive material.

6.5 Transmutation

There is broad agreement that effective transmutation/burning of all the transuranium elements can only be achieved in fast neutron spectra.

In several countries the main interest for transmutation is focused on accelerator-driven systems (ADS). In other countries the focus remains on critical fast reactors.

The basic technical components of an ADS have not yet been fully defined. Several options are still open and studied – for example type of primary coolant (liquid lead-bismuth, liquid sodium, helium gas), type of fuel (oxide, nitride, metal), type of accelerator (cyclotron, linear) and spallation source materials and design. The preferred options in the ongoing studies of an experimental ADS within EU seem to be a linear accelerator and liquid lead-bismuth eutectic coolant for the subcritical reactor as well as for spallation material.

US and European studies conclude that a basic R&D programme of the order six years and costing several hundred million Euros is required to define the base for construction of an experimental ADS facility.

Building and operation of a small experimental ADS-facility is necessary to confirm the concept. It would take another five to ten years to construct. It should then be followed by development and construction of an almost full scale demonstration plant that might be completed in the middle of the 2030ies.

The funding level of the R&D-programmes on ADS have not (yet) reached the level presumed in the studies presenting the above tentative time schedule. It is thus unlikely that the time schedules suggested in the ADS-systems studies will be met.

Deployment of ADS-plants on an industrial scale is very unlikely to occur before 2050.

Fast reactors can be used either as burners of plutonium and other transuranium elements or as breeders for most efficient use of the mined natural uranium.

Both ADSs and fast reactors need improved fuel, improved material and other improved equipment before deployment on an industrial scale. New fuel, new material and new equipment will need full scale testing before being used in an industrial plant.

6.6 Safety and radiation protection

The implementation and construction of future P&T facilities must meet the same requirements from environmental and nuclear laws as other nuclear facilities.

The development of P&T systems involves the resolution of many issues concerning safety and radiation protection. As the systems are under basic development it is unlikely that all such issues have been identified.

The ADS, properly designed, may have the advantage over the fast critical reactor that reactivity initiated accidents can be disregarded. Some anticipated transients in the ADS studied so far may however cause fuel damage. These transients need further analyses. Perhaps the largest unknown in the safety area is the progression and consequences of severe accidents, which may involve fuel melting. In the case oxide fuel melts and floats on the liquid lead-bismuth coolant the further development includes potential severe consequences. Such severe accident scenarios and possibilities have still to be defined, studied and prevented or mitigated against.

6.7 Time and costs

As is indicated in some of the conclusions above, the development of any P&T-system will require several decades. The deployment and operation of the necessary industrial facilities will take another 100 years or more before the stocks of long-lived radionuclides from the currently existing nuclear power reactors have been transmuted.

The costs for P&T are not possible to assess with any confidence at the current stage. A systems study in USA indicates that the total long-term cost for a P&T system over about 130 years would be some 280 000 M$ including R&D, investments and operation of an ADS-based system for the current US reactor park. This cost is on the same order as the estimated value of the electricity that could be generated by the ADS-transmutation plants included in the system.

The efficient use of the energy released by the transmutation process is essential for any possibility to achieve economic viability for P&T. This is particularly the case for transmutation of plutonium. Some experts have the view that minor actinides might be transmuted separately in dedicated ADS-plants where the generated heat could be dumped.

It seems that for future large scale nuclear energy production a system with light water reactors may dominate for considerable time. They may be supplemented by fast reactors for burning of plutonium and perhaps by ADS-facilities for transmutation of other transuranium elements as well as of some plutonium with high content of heavy isotopes.

In the long term, the uranium prices may increase due to depletion of the low-cost resources or due to demands for very strict environmental measures in the mining. Then fast breeder reactors may take over the role as leading nuclear power producers. Such reactors are also feasible for transmutation of the heavier transuranium elements.

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Appendix I

R&D-projects within EU-programme

The general structure of the EU fifth framework programme (FWP 5) is presented in section 2.4 of the main part of this report. That section as well as other parts of the report also includes descriptions of the project PDS-XADS and of some other EU-projects. This Appendix I gives more details of some of the projects within FWP 5 and also of some projects that have started in FWP 6.

1 Partitioning projects

The PARTITION cluster included three projects listed in Table I-1.

Table I-1. *PARTITION cluster projects*

Acronym	Subject of research	Coordinator (country)	No. of partners	Start date & duration	EC funding (M€)
PYROREP	Pyrometallurgical processing research	CEA (F)	7	Sept 2000 36 months	1.5
PARTNEW	Solvent extraction processes for minor actinides (MA)	CEA (F)	10	Sept 2000 36 months	2.2
CALIXPART	Selective extraction of MA by organic matrices	CEA (F)	9	Oct 2000 40 months	1.4

The first one, PYROREP, aimed at assessing flow sheets for pyrometallurgical processing of spent fuels and targets. Two methods, salt/metal extraction and electrorefining, investigated the possibility of separating actinides from lanthanides. Materials compatible with corrosive media at high temperature have been selected and tested.

The two other projects dealt with the development of solvent extraction processes to separate minor actinides (americium and curium) from high-level liquid waste (HLLW). In the project PARTNEW**,** the minor actinides were extracted in two steps. They were first co-extracted with the lanthanides from HLLW (by the DIAMEX processes), then separated from the lanthanides (by the SANEX processes). Basic studies have been performed for both steps, in particular synthesis of new ligands and experimental investigation and modelling of their extraction properties. The radiolytic and hydrolytic degradation of the solvents were also studied and the processes are tested with genuine HLLW.

The CALIXPART project dealt with the synthesis of more innovative extractants. Functionalised organic compounds, such as calixarenes, were synthesised with the aim of achieving the direct extraction of minor actinides from HLLW. The extraction capabilities of the new compounds were studied together with their stability under irradiation. The structures of the extracted species were investigated by nuclear magnetic resonance (NMR) spectroscopy and X-ray diffraction to provide an input to the molecular modelling studies carried out to explain the complexation data.

1.1 PARTNEW – New solvent extraction processes for minor actinides

The goal of the PARTNEW project was to define the sustainable, environmentally friendly process with high potential for industrial application, which would be selective for actinide separation from the effluents issuing from the reprocessing of spent nuclear fuels, either uranium oxide or mixed oxide (MOX) fuels /PARTNEW/.

The objectives of the project were to:

- Study possible solvent extraction processes.
- Investigate the preferential extraction of actinides over the other elements that are present in the wastes.
- Design a two cycle process including:
    - a Selective ActiNide EXtraction (SANEX) process, to separate selectively the trivalent actinides (An = americium and curium) from the highly acidic waste feeds and
    - an Am(III) / Cm(III) separation process.
- Design a three cycle process involving:
    - the DIAMide Extraction (DIAMEX) process for the selective extraction of An(III) and Ln(III) (lanthanides(III)) mixtures from the feeds derived from acidic effluents produced after the reprocessing of the spent nuclear fuels done by the PUREX process,
    - a SANEX process for An(III) / Ln(III) separation from acidic feeds and
    - an Am(III) / Cm(III) separation process.
- Consider two principal types of feeds for the processes:
    - the highly active raffinates (HAR) generated after the reprocessing of uranium oxide or MOX fuels and
    - the highly active concentrates (HAC) produced after concentration and denitration of the HARs.

The work included fundamental chemical research work, process development and qualification using real genuine high active wastes.

The scientific tasks for the chemistry of solvent extraction were to:

- Prepare new solvent extraction reagents containing polyamides, heterocyclic nitrogen- and sulphur-bearing ligands.
- Establish methods based on traditional and combinatorial chemistry for the design, synthesis, development and testing of the molecules.
- Understand the extraction, reactivity, kinetics and other thermodynamical properties of the developed new ligands.
- Computer model the complexes or ion-pairs formed leading to ligand design.
- Compare calculated and experimental data for the establishment of quantitative structure-property relationships.
- Establish and develop a library of molecules and data for the principal aim of the project.

The scientific tasks for the process development were to:

- Study the radiolytic and hydrolytic stabilities of the new molecules to be used within the processes.

- Define solvent extraction processes including the composition of the different solutions (organic and aqueous), the hydraulics of the mixtures and the organic to aqueous flow ratios and the extraction, scrubbing and stripping together with solvent clean-up sections. The extraction devises tested were mixer-settles, pulse columns, centrifugal contactors and hollow fibre modules.
- Define computer models of the processes in the steady and transient states.

The partners involved four universities and seven national European laboratories.

1.2 EUROPART – EUROpean research program for the PARTitioning of minor actinides from high active wastes from the reprocessing of spent nuclear fuels

EUROPART is the merged continuation in FWP 6 of PARTNEW, CALIXPART and PYROREP. It was started in January 2004 and will last for three years /EUROPART/. The separation techniques that will be studied can be divided into hydrometallurgy and pyrometallurgy. The main axes for the research will be:

- The partitioning of MA (minor actinides, Am-Cf) from high burn-up UOX and multi-recycled MOX fuels.
- The partitioning of all actinides together for recycling, for advanced dedicated fuel cycles, *i.e.* the double strata concept, and the ADS (Accelerator Driven System).

The research is divided into nine different work packages.

The objectives of the EUROPART research related to the hydrometallurgy are:

- Definition of processes for the joint partitioning of trivalent actinides (An), from Am to Cf that are contained within aqueous highly active raffinates (HAR) and/or highly active concentrates (HAC) issuing from the reprocessing of nuclear fuels, such as high burn-up UOX and MOX.
- Definition of processes for the individual separation of the trivalent Am/Cm/Bk/Cf ions from the product generated by the first processes.
- Definition of reprocessing processes for the treatment of spent fuels from advanced dedicated nuclear systems, such as those of the “double strata concept”, i.e. ADS.
- Definitions of methods for the co-conversion of An for fuel or target preparation.

The objectives of the EUROPART research related to the pyrometallurgy are:

- Determination of basic properties of An in molten halides that are the media to be selected for the development of pyrometallurgical partitioning processes.
- Definition of partitioning processes of An from high level wastes issued from the reprocessing of UOX and/or MOX spent fuels by the PUREX process.
- Definition of processes for advanced dedicated fuel cycles.
- Definition of conditioning methods for the wastes to be generated by the partitioning processes.
- Definition of the overall organisation of the methods to be implemented within pyrometallurgical partitioning processes.

There are 24 participating laboratories including a European nuclear research laboratory, national nuclear research laboratories, universities, nuclear industrial companies and industrial companies. The participants are:

CEA, France – coordinatior
BNFL, United Kingdom
Chalmers Technical University, Sweden
CIEMAT, Spain
Czech Technical University, Czech Republic
Centre National de la Recherche Scientifique, France
Consejo Superior de Investigaciones Cientificas, Spain
Electricite de France, France
ENEA, Italy
Forschungszentrum Julich,Germany
Forschungszentrum Karlsruhe, Germany
Instutute for Transuranium Elements (EC), Germany
Institute of inorganic chemistry, Academy of Sciences of the Czech Republic, Czech Republic
Institute of Nuclear Chemistry and Technology, Poland
Johannes Gutenberg University, Mainz, Germany
Katchem spol, S. r. o. , Czech Republic
Nuclear Research Institute Rez plc., Czech Republic
Politecnico di Milano, Italy
Universidad Autonoma de Madrid, Spain
Universita degli studi di Parma, Italy
Universite de Liege, Belgium
Universite Louis Pasteur, France
University of Reading, United Kingdom
University og Twente, The Netherlands

1.3 ACTINET (Network for Actinide Sciences)

ACTINET is a network of excellence established by the 6th framework program. It is designed to enhance the collaboration within the union with respect to laboratories being able to handle actinides. Few laboratories possess knowledge and tools in actinide science. None of them cover the full spectrum at the scale required by the technical challenges and the interaction between the laboratories is scarce.

In this context, the general objective of ACTINET is to gather the concerned (European) scientific community through a network, aiming to reach sustainability in a few years. Knowledge dissemination, education and training activities through the network will ensure a high quality and thus high expertise of actinide chemistry in Europe. More specifically the goals of ACTINET are to:

- Coordinate the use of major actinide facilities to the European scientific community.
- Improve human mobility between member institutions, in particular between academic institutions and national laboratories.
- Promote excellence through a selection process of R&D and training activities.

The number of laboratories included in this network is 27 research institutes and universities from Belgium, Cyprus, Czech Republic, Denmark, Finland, France, Germany, Netherlands, Spain, Sweden, Switzerland and UK.

2 Transmutation projects

2.1 BASTRA cluster

Three projects have been grouped into the cluster of Basic Studies on Transmutation (BASTRA) (See Table I-2). The MUSE project aimed to provide validated analytical tools for subcritical neutronics, data and a reference calculation tool for ADS study. The experiments have been carried out by coupling a pulsed D-T/D-D neutron generator source (GENEPI) to the MASURCA facility loaded with MOX fuel operated as a subcritical system with different coolants (such as sodium and lead). Cross-comparison of codes (benchmarking) and data have been extensively done. Experimental reactivity control techniques, related to subcritical operation have been developed. See further section 2.4 in this Appendix I.

The other two projects dealt with nuclear data. These are presented in sections 2.2 and 2.3 below.

Table I-2. Basic *studies for transmutation (BASTRA) cluster projects*

Acronym	**Subject of research**	**Coordinator** (country)	**No. of partners**	**Start date & duration**	**EC funding** (M€)
MUSE	Experiments for sub-critical Neutronics validation	CEA (F)	13	Oct 2000 36 months	2.0
HINDAS	High and intermediate energy nuclear data for ADS	UCL (B)	16	Sept 2000 36 months	2.1
n-TOF - ND-ADS	ADS nuclear data using time of-flight facility	CERN(CH)	18	Nov 2000 36 months	2.4

2.2 HINDAS – High- and Intermediate-energy Nuclear Data for Accelerator-driven Systems

HINDAS is a joint European effort, which gathers essentially all European competence on nuclear data for transmutation in the 20-2000 MeV range /HINDAS/. The program has been designed to obtain a maximal improvement in high-energy nuclear data knowledge for transmutation. This goal can only be achieved with a well-balanced combination of basic cross section measurements, nuclear model simulations and data evaluations. The three elements iron, lead and uranium have been selected to give a representative coverage of typical materials for construction, target and core, respectively, especially relevant to ADS, as well as a wide coverage of the periodic table of elements.

The HINDAS project ran over the period 2000-09-01 – 2003-11-30, and has been coordinated by Jean-Pierre Meulders, Université Catholique de Louvain, Louvain-la-Neuve, Belgium. The total EU funding was 2.1 MEUR. To this should be added matching funding, equipment and infrastructure from the participating countries. The coordinator was supported by a management committee and by a scientific committee. A final report of the project is under preparation.

In total, 16 universities or laboratories participated. Of these, 6 have experimental facilities. This means that HINDAS involved essentially all relevant European laboratories in its energy range. This distribution and coordination of experiments at many laboratories made the work very efficient. What is noteworthy is that HINDAS involved many partners and even laboratories that have previously not been involved at all in activities on nuclear data for applications. Thus, HINDAS has widened the field of applied nuclear physics.

All results have been published in regular refereed physics journals. In addition, all relevant data have been implemented into the regular nuclear data bases. Continuous contacts have been kept with nuclear data evaluators; in fact some of the collaboration members are also established data evaluators themselves.

The HINDAS project has been carried out in eight work packages (WP). WP 1-3 concerned experiments in the 20-200 MeV range, WP 4-6 dealt with 200-2000 MeV experiments, and WP 7 and 8 were devoted to theory for the 20-200 and 200-2000 MeV regions, resp.

The division into two energy ranges is natural, since there appears to be a transition region around 200 MeV for the theoretical models. Below this energy the theoretical calculations have to include direct interactions, as well as preequilibrium, fission and statistical models, whereas at higher energies the intra-nuclear cascade model, together with fission and evaporation models, have to be considered. As a coincidence, the experimental facilities and the measurement techniques are also different below and above about 200 MeV.

The experimental WPs were structured according to type of particles produced. This means that for each energy range, there were WPs on production of light ions, neutrons and residues respectively. Below, the WPs are described in some more detail:

1) **Light charged-particle production induced by neutrons or protons between 20 and 200 MeV (Lead contractor: Université Nantes, France).**

The double-differential cross sections for proton- and neutron-induced production of hydrogen and helium ions on iron, lead and uranium isotopes have been measured at UCL-Louvain, TSL-Uppsala and KVI-Groningen. These measurements aimed at providing essentially complete data in both emission angle and ejectile energy. Such double differential cross sections constitute a very stringent test for theoretical models in this energy domain. In addition, charged-particle multiplicities in proton-induced reactions have been measured at KVI.

2) **Neutron production induced by neutrons or protons between 20 and 200 MeV (Lead contractor: Uppsala University, Sweden).**

Neutron elastic scattering measurements, (p,xn) and (n,xn) measurements on iron, lead and uranium have been performed at UCL-Louvain and TSL-Uppsala. Elastic scattering measurements are useful not only for optical model development, but can also be used directly for neutron transport calculations. The (n,n) and (n,xn) measurements made use of novel detection techniques, while (p,xn) reactions have been studied using NE213 detectors with time-of-flight techniques. The efforts within WP2 have to a large degree been linked to the Swedish KAT/NATT projects (see Appendix III).

3) Residual nuclide production induced by neutrons and protons between 20 and 200 MeV and production of long-lived radionuclides (Lead contractor: Hanover University, Germany)

Measurements of proton-induced production of residual nuclei have been carried out at PSI, while neutron-induced production has been studied at UCL-Louvain and TSL-Uppsala, where also neutron-induced fission has been measured. For the short-lived residual radionuclides, cross sections have been determined using activation techniques. The production of long-lived radionuclides has been studied by Accelerator-Mass Spectroscopy (AMS) after chemical separation at ETH[85].

4) Light charged-particle production above 200 MeV (Lead contractor: FZ Jülich, Germany)

The proton- and deuteron-induced production cross sections of protons and alpha particles have been measured with a 4π silicon ball detector at the COSY accelerator in Jülich. Experiments on thin targets aimed for tests of the intra-nuclear cascade model, while thick-target studies have been focused on benchmarking transport codes. These measurements will also help to evaluate gas production in the window and structure materials of an ADS, which will give implications for the lifetime of such components. With the PISA setup, it has also been possible to measure total and double-differential cross sections for production of spallation products.

5) Neutron production induced by protons above 200 MeV in thin and thick targets (Lead contractor: CEA-Saclay, France)

Double-differential neutron production cross sections, for both thin and thick targets, have recently been measured at CEA-Saclay using time-of-flight or magnetic spectrometer techniques. At FZ Jülich, multiplicities of neutrons up to 150 MeV have been studied event-wise with a 4π liquid scintillator, using both thin and thick targets. The two experiments are complementary, both for technical and physics reasons. E.g., comparisons can be made between the directly measured multiplicities with those inferred from integration of the double-differential data.

6) Residual nuclide production above 200 MeV in inverse kinematics (Lead contractor: GSI, Germany)

Proton- and deuteron-induced nuclide production has been measured in inverse kinematics, i.e., a lead or uranium beam hits a liquid hydrogen or deuterium target, and the spallation products are identified in flight using a high-resolution magnetic spectrometer. In this way all spallation products, irrespective of half-life, can be measured. These new data will be useful when calculating the radioactive inventory, the radiotoxicity and the breeded impurities in a realistic spallation target of an ADS. The technique of inverse kinematics is novel in nuclear data measurements for applications. (It is noteworthy from a Swedish perspective that via the participation in HINDAS, the technology has now been transferred to TSL for other experiments.)

[85] ETH = Eidgenössische Technische Hochschule in Zürich, Switzerland.

7) Nuclear data libraries and related theory (Lead contractor: NRG[86], Netherlands)

This work package concerned nuclear model calculations for analysis of the experimental data provided by WP 1-3, i.e., between 20 and 200 MeV. Special emphasis has been put on providing as complete information as possible on cross sections for all possible outgoing channels of iron, lead and uranium, and to construct improved nuclear data libraries, extending to 200 MeV.

8) High energy models and codes (Lead contractor: Université de Liège, Belgium)

This work package has been devoted to theory for WP 4-6, i.e., above 200 MeV, and regarded mainly intra-nuclear cascade models and evaporation and fission models. The main objective has been the development of powerful and accurate tools to calculate nucleon-nucleus spallation reactions.

2.3 NTOF

The n_TOF Nuclear Data for ADS project /NTOF/, which started November 1, 2000, is primarily focused on experimental studies of nuclear data at a new neutron spallation source at CERN, the European Organization for Nuclear Research. The main activity of the project is to provide experimental neutron data in the resolved resonance region. In addition, data evaluation is an integrated part of the project. The total EU funding is 2.4 MEUR.

The first year of activity of the project was characterized by the commissioning of the facility, CERN n_TOF. This facility, which started operation in April 2001, is capable of delivering an extremely intense pulsed neutron beam ($\approx 10^5$ neutrons/cm^2/pulse at the experimental station), generated by the high-intensity 20 GeV proton beam (7×10^{12} protons/pulse) of the CERN PS accelerator complex. An experimental area has been constructed at 187.5 m distance from the lead spallation target, which is equipped with a neutron beam monitoring system, several sets of detectors for neutron induced reaction cross section measurements, a data acquisition system and front-end electronics, a neutron escape-line, and a control room. By using time-of-flight techniques, n_TOF is a white neutron source covering an energy range from 1 eV up to the incident beam energy. The intensity essentially follows a 1/E dependence, which makes measurements at high energies very time-consuming. Thus, the competitive edge of the facility is in the 1 eV- 1 keV range. Given the very low repetition-rate of the proton pulses (≈1 pulse of 6 ns width every 2.4 s), the facility is particularly suitable for measurements of capture cross sections of radioactive isotopes.

The project was severely delayed at the initial phase. For instance, there were some significant technical problems with the n_TOF facility, especially concerning the background situation. Now a very low ambient background level, suitable for operation of the presently available and planned detection systems, has been obtained after a re-design of the shielding of the experimental area in September 2001. As a consequence of the first-year problems, the project has been prolonged by about one year.

Besides the measurements at CERN, additional measurements are undertaken at the GELINA pulsed TOF neutron facility of JRC-IRMM[87] (Geel, Belgium), at the neutron

[86] NRG = Nuclear Research and consultancy Group
[87] IRMM = Institute for Reference Materials and Measurements

sources of FZK[88] (Karlsruhe, Germany), NCSR-INP[89] (Athens, Greece) and ITN[90] (Lisbon, Portugal).

Below, the key issues of the project are outlined:

- Measurements with a precision of a few % of the appropriate capture, fission cross sections for elements with relatively well known cross sections (though the knowledge at high energies is still limited), i.e., ^{197}Au, $^{24\text{-}26}$Mg, ^{207}Pb, ^{56}Fe, ^{235}U, and ^{238}U.
- Determination with a precision of a few % of the capture cross sections for isotopes relevant to the Th-fuel cycle, i.e., ^{232}Th, ^{231}Pa, ^{233}U, ^{234}U, and ^{236}U.
- Determination with a precision of a few % of the capture cross sections for some transuranic isotopes, ^{237}Np, ^{240}Pu, ^{242}Pu, ^{241}Am, ^{243}Am, and ^{245}Cm.
- Determination with a precision of a few % of the capture cross sections of specific long-lived fission products, such as ^{151}Sm, ^{99}Tc, ^{121}I, ^{79}Se, and further on measurements in the lead region, i.e., ^{204,206,207,208}Pb and ^{209}Bi.
- Determination with a precision of a few % of the fission cross sections of ^{232}Th, ^{231}Pa, ^{233}U, ^{234}U, ^{236}U, ^{237}Np, ^{241}Am, ^{243}Am, ^{245}Cm.
- Measurements of precise (n,xn) cross sections using activation techniques on ^{233}U, ^{232}Th, ^{231}Pa, ^{232}U, ^{239}Pu, ^{241}Pu, ^{241}Am, ^{243}Am, ^{237}Np, and ^{205}Pb.
- Measurements of the total cross sections of ^{237}Np, ^{129}I, ^{239}Pu, and ^{240}Pu.
- Measurements of capture and fission cross sections at given neutron energies with mono-energetic beams of the isotopes ^{232}Th, ^{233}U, ^{237}Np, ^{241,243}Am, and ^{99}Tc, ^{129}I, ^{79}Se, ^{151}Sm, and ^{135}Cs.
- Evaluation of the measured cross sections with standard evaluation procedures in co-operation with the official nuclear data centres.
- Development of a data dissemination system fully compatible with the standard simulation programs, incorporating multi-platform data visualization software, user-friendly access interfaces and cybernetic tools, i.e., Internet, CD, etc.
- Dissemination of the results, i.e., to make measured and evaluated cross sections publicly available in rapid, complete and easily accessible nuclear databases, ensuring conformity and standardization in co-operation with the official nuclear data centres.

2.4 MUSE

MUSE-experiments were designed around an existing experimental reactor - MASURCA at CEA/Cadarache, reconstructed for neutronic studies of accelerator driven systems. The MASURCA core is rather small, the height being 60 cm and the radius ranging from 45 to 51 cm, depending on the core configurations. The reactor power is low (maximum 5 kW) and the core cooling is provided by air.

The core of MASURCA has a very flexible design and several different core configurations are possible. The core can be loaded with different fuels (e.g. thorium, uranium and plutonium), different coolants can be used (sodium, lead, gas) and different levels of criticality are possible, both critical and subcritical.

[88] FZK = Forschungs Zentrum Karlsruhe, Germany
[89] National Centre for Scientific Research – Institute of Nuclear Physics, Athens Greece
[90] Instituto Tecnológico E Nuclear, Portugal

The MUSE program started at CEA/Cadarache (with the sponsorship of EdF and Framatome) in 1995 with the short MUSE-1 experiment. In MUSE-2 (1996) diffusing materials (sodium and stainless steel) were placed around the external source in order to modify the neutron importance of the source and to study the effects from it. In both MUSE-1 and MUSE-2 an intense ^{252}Cf neutron source was used as external source. In MUSE-3, two years later, the californium source was replaced by the neutron generator SODERN/GENIE26, producing 14 MeV neutrons by (d,t)-fusion reactions. Several levels of reactivity were investigated and MOX (UO_2-PuO_2) fuel enriched with 25 % plutonium was used. Experiments with different "buffer zones" (sodium, stainless steel and lead) were performed.

In the on-going MUSE-4 experiments (started in 2000 in international collaboration via the 5th Framework Programme) the neutron generator GENEPI, especially developed for the MUSE-4 experiments, was introduced Figure I-1. With its improved performances (in terms of the quality of the neutron pulse and the source intensity) and the use of both (d,d)- and (d,t)-reactions, it expands the possibilities and improves the quality of the experiments. Accurate dynamic measurements based on the pulsed mode operation of the GENEPI will also allow new experimental reactivity determination of the subcritical multiplying media.

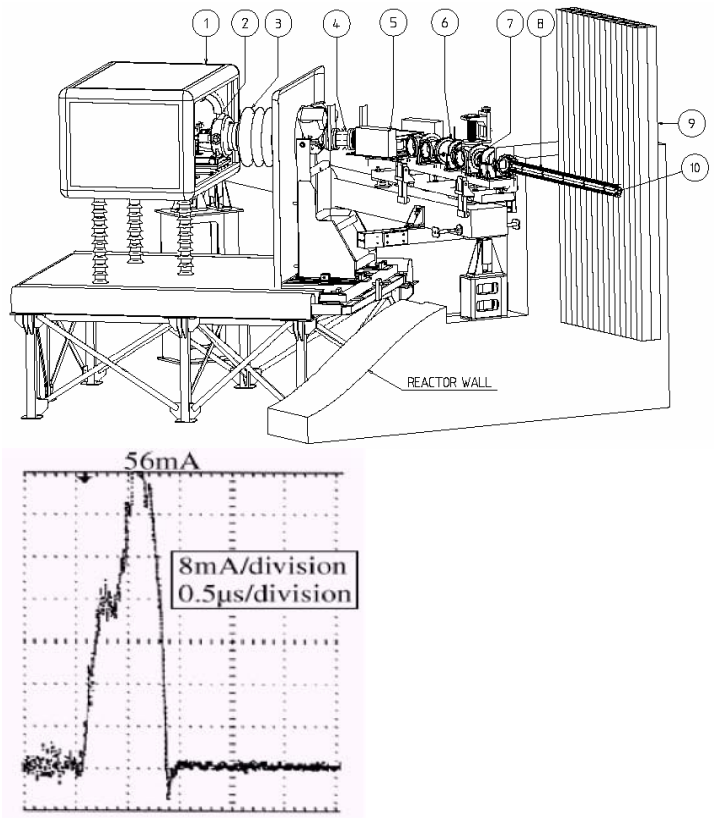

Figure I-1. ***A)*** *View of GENEPI. The HV terminal (1), the duoplasmatron (2), the accelerating tube (3), three electrostatic quadropoles (4,6,7), the dipole (5), the beam line (8) inside the reactor (9) towards the tritium target (19).* ***B)*** *The deuteron beam pulse.*

The geometry of the second subcritical configuration of MUSE-4 (Sc2) is shown in 0. The axial (z-direction) dimension of the fuel is 60.96 cm, except in a 21.2 cm wide channel above and below the lead buffer and the accelerator tube (in the y direction), where it was extended by 10.16 cm. The sodium-steel reflector ends at z = ±61.76 cm. There is also a 10.16 thick axial shield (not shown in the figure) above and below the Na/SS reflector. The neutron source is surrounded by a lead "buffer" medium, in order to simulate the diffuse properties of a spallation source. The GENEPI deuteron accelerator tube is introduced horizontally at the core mid-plane and the deuterium or tritium target is located at the core centre (filled rectangle). The fuel in MUSE-4 is MOX fuel with 72% ^{238}U, 21% ^{239}Pu and 5% ^{240}Pu plus small amounts of some other actinides.

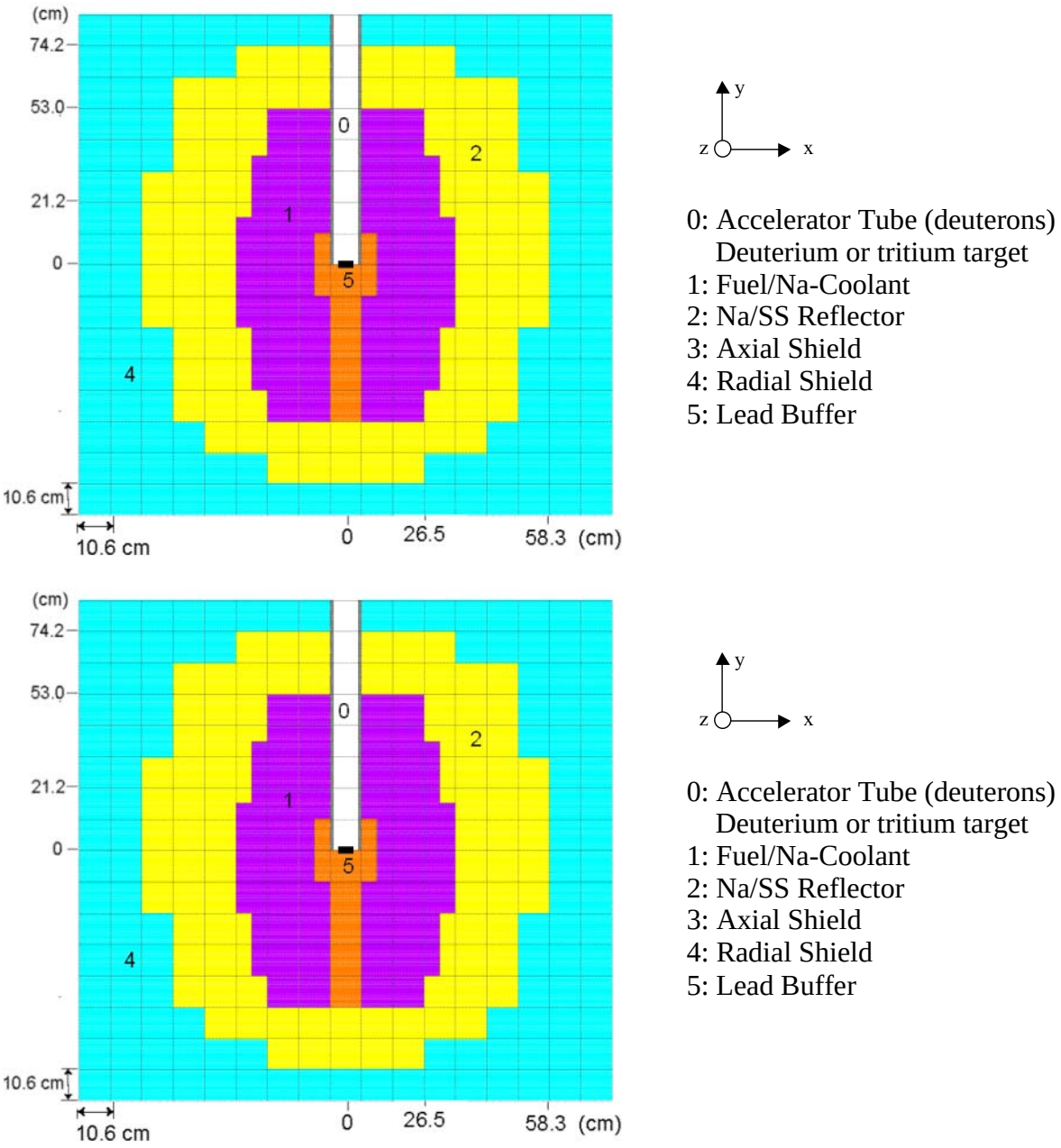

Figure I-2. *x-y cross-sectional view of the second subcritical configuration (Sc2) of the MUSE-4 experiments. The core is composed of a number of quadratic subassemblies with the dimension 10.6*10.6 cm.*

The following six different experimental configurations have been be studied in the MUSE-4 experiments:

- One critical reference configuration (GENEPI shut off). The reactivity will be experimentally determined by classical pilot rod shutdown measurement.
- Three subcritical configurations (Sc0 with k_{eff} ~ 0.994, Sc2 with k_{eff} ~ 0.97 and Sc3 with k_{eff} ~ 0.95)
- Two complementary asymmetrical configurations with k_{eff} around 0.95 and 0.93, obtained from the reference configuration and Sc1, by complete insertion of one of the safety rod.

The main results obtained up to date :

The main results obtained up to date :

- Experiments confirmed qualitatively a validity of theoretical models used for description of the MUSE-experiment
- All methods used in determination of MUSE subcriticality agree well for k_{eff} between 0.99 and 1.00. For k_{eff} well below 0.98 discrepancies between different methods increase. It remains to assess by further analyses which theoretical methods are most suitable for determination of deeper subcriticality
- All methods show good sensitivity for detection of small changes of reactivity

As concerns the various analysis methods, the measurement results show first, that the use of noise methods driven by the intrinsic source was difficult at subcriticality levels representative of an ADS (very low noise/signal ratios => measurement duration >5 h).

Then, with the pulsed neutron source, the results indicate that:

- The Rossi-α method can be used if pulsed source frequency is not higher than around 2 kHz. On the contrary, an analysis with the stochastic and deterministic Feynman-α methods should need frequencies higher than the GENEPI maximum frequency.
- The analysis in current mode using the Cross Power Spectral Density - CPSD is more delicate compared to the analysis based on pulse mode. It is also limited by both the acquisition chain used at MASURCA and the performances of GENEPI (higher intensity necessary),
- Rossi-α and Pulsed Neutron Source - PNS methods seem promising because of their easy set-up. Good statistics have been also obtained. Advanced kinetic models have to be defined to strengthen the use of these methods at deep subcriticality levels.

MUSE-project will be finished at the end of 2004.

2.5 TESTRA cluster

Four projects are grouped in the cluster of Technological Studies on Transmutation (TESTRA) (see Table I-3). This cluster focused on the investigation of radiation damage induced by products of spallation reactions in materials, of the corrosion of structural materials by lead alloys and of fuels and targets for actinide incineration.

Table I-3. *Technological studies for transmutation (TESTRA) cluster*

Acronym	Subject of research	Coordinator (country)	No. of partners	Start date & duration	EC funding (M€)
SPIRE	Effects of neutron and proton irradiation in steels	CEA (F)	10	Aug 2000 48 months	2.3
TECLA	Materials and thermalhydraulics for lead alloys	ENEA (I)	16	Sept 2000 36 months	2.5
MEGAPIE TEST	A megawatt heavy liquid metal spallation target experiment with proton beam	FZK (D)	17	Nov 2001 36 months	2.4
ASCHLIM	Computational fluid dynamics codes for heavy liquid metals	SCK·CEN (B)	14	Dec 2001 12 months	0.12

The SPIRE project addressed the irradiation effects on an ADS spallation target. The effects of spallation products on the mechanical properties and microstructure of selected structural steels (e.g. martensitic steels) have been investigated by ion beam irradiation and neutron irradiation in reactors (HFR in Petten, BR2 in Mol and BOR60 in Dimitrovgrad). Data representative of mixed proton/neutron irradiation are being obtained from the analysis of the SINQ spallation target at the Paul Scherrer Institute in Villigen (CH).

The objective of TECLA project was to assess the use of lead alloys both as a spallation target and as a coolant for an ADS. Three main topics have been addressed: corrosion of structural materials by lead alloys, protection of structural materials and physics-chemistry and technology of liquid lead alloys. A preliminary assessment of the combined effects of proton/neutron irradiation and liquid metal corrosion is being carried out. Thermal-hydraulic experiments are being performed together with numerical computational tool development.

The major objective of the MEGAPIE-TEST project has been to develop and validate the design and operation of a heavy liquid metal (Pb-Bi) spallation target at a level of a megawatt. The project aimed to provide a comprehensive database from single-effect experiments, a full-scale thermal-hydraulic simulation experiment, and the first beam-on experiments. In parallel, numerical computational tools have been validated for Pb-Bi target design. The studies included neutronic calculations, materials, corrosion, thermal-hydraulics, structure mechanics, liquid metal technology, safety and licensing issues. Prospects on the extrapolation and applicability of the obtained results to an ADS spallation target have also been given.

The ASsessment of Computational fluid dynamics codes for Heavy LIquid Metals (ASCHLIM) project aimed at bringing together various actors (industry, research institutions and university) in the field of heavy liquid metals both in the experimental and numerical fields and creating an international collaboration to:

- Make an assessment of the main technological problems in the fields of turbulence, free surface and bubbly flow and
- Co-ordinate future research activities in this area. The assessment has been made on the basis of existing experiments whose basic physical phenomena have been analysed through the execution of calculation benchmarks using commercial and research codes.

2.6 FUETRA cluster

Three projects were put together in this cluster (see Table I-4).

Table I-4. *Fuel studies for transmutation (FUETRA) cluster*

Acronym	Subject of research	Coordinator (country)	No. of partners	Start date & duration	EC funding (M€)
CONFIRM	Uranium-free nitride fuel irradiation and modelling	KTH (S)	7	Sept 2000 48 months	1.0
FUTURE	Development of transuranics oxide fuels for transmutation	CEA (F)	7	Dec 2001 36 months	1.7
THORIUM CYCLE	Development of thorium cycle for PWR and ADS	NRG (NL)	7	Oct 2000 48 months	1.2

The objectives of the CONFIRM project have been to develop methods for fabrication (such as carbo-thermic reduction process) of uranium-free nitride fuels (Pu,Zr)N and to model and test their performance under irradiation up to 20 % burn-up in the Studsvik R2 reactor. Carbo-thermic processes are also being used for the production of (Am, Zr)N pellets at ITU, Karlsruhe. Successful high temperature (≈2 500°C) stability tests of (U,Zr)N have been made and a study of C-14 production has been completed.

The main objective of the FUTURE project has been to study the feasibility of oxide compounds $(Pu,Am)O_2$, $(Th, Pu, Am)O_2$ and $(Pu, Am, Zr)O_2$ irradiated as homogeneous fuel for an ADS. The R&D programme has been largely devoted to the synthesis of the compounds, their characterisation (thermal and chemical properties at relevant temperatures) and the development of fabrication processes. Modelling codes have been developed to calculate the fuel performance. The input data for the codes have been based on experimental results. The fuel behaviour under accident conditions is being analysed using the experimental data obtained at high temperatures.

The objective of the project THORIUM CYCLE has been to investigate the irradiation behaviour of thorium/plutonium (Th/Pu) fuel at high burn-up and to perform full core calculations for thorium based fuel with a view to supplying key data related to plutonium and minor actinide burning. Two irradiation experiments have been carried out:

1. Four targets of oxide fuel (Th/Pu, uranium/plutonium, uranium and thorium) fabricated, irradiated in HFR in Petten and characterised after irradiation,
2. One Th/Pu oxide target is also irradiated in KWO reactor at Obrigheim (D).

Though this project was accepted for funding in the area of "safety and efficiency of future systems", it has been grouped with the FUETRA cluster.

2.7 RED-IMPACT

The project is part of the 6th FWP. The objectives of the 3-year RED-IMPACT project (total budget M€ 4 including an EC contribution of M€ 2.0) are to:

- Assess the impact of P&T on geological disposal and waste management.
- Assess economic, environmental and societal costs/benefits of P&T.
- Disseminate results of the study to stakeholders (scientific, general public and decision makers) and get feedback during the course of the study.
- Iterate and refine the work based on stake-holders' feedback to achieve full impact of this study on the implementation of the waste management policy of the European Community.

The work of the project is subdivided into six work packages:

- WP1: Waste management and transmutation strategies will be reviewed and a number of representative scenarios will be selected for in-depth impact studies.
- WP2**:** Feasibility of the industrial deployment of selected scenarios will be made and their impact on waste management will be studied.
- WP**3:** Assessment of waste streams, waste features, leach resistance, heat generation, reprocessing capability etc. will be studied for selected fuel cycles.
- WP4: Assessment will be made of the benefits and costs of P&T/C in advanced fuel cycles for waste management and geological disposal.
- WP5**:** Economic, environmental and societal assessment of fuel cycle strategies will be performed.
- WP6: Synthesis and dissemination of results of the above studies will be made to stakeholders.

The Consortium of RED-IMPACT has a very broad multi-disciplinary scientific and industrial background with 23 participants (as shown on Figure I-3) representing:

- Waste Management Agencies/Companies: ENRESA, NIREX, RAWRA, SKB
- Nuclear Industries and Utilities: BN, BNFL, COGEMA, EA, FRAMATOME, KKP-EnBW
- Research Institutes and Universities: CEA, CIEMAT, CITON, FZJ, GRS, KTH, ITU, NRG, NRI, USTUTT, SCK·CEN, VUJE

This 3-year study is geared to produce relevant results which should be complimentary to NEA/OECD, IAEA and some other national studies.
RED-IMPACT is coordinated by Kungliga Tekniska Högskolan.

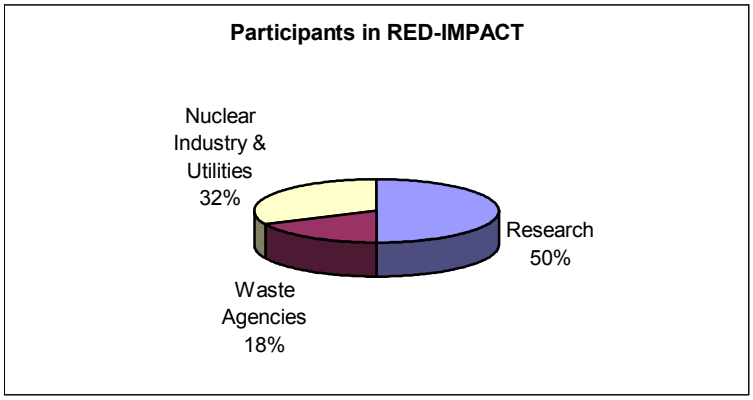

Figure I-3. *RED-IMPACT participants distribution on sectors.*

3 References Appendix I

EUROPART 6th Euratom Framework Programme, contract F16W-CT-2003-508854.

HINDAS HINDAS, 5th Euratom Framework Programme, contract no. FIKW-CT-000-0031

NTOF n_TOF-ND-ADS, 5th Euratom Framework programme, contract no. FIKW-CT-2000-00107, coordinator A. Mengoni, CERN, Geneva, Switzerland and University of Bologna, Italy

PARTNEW 5th Euratom Framework programme, contract FIKW-CT2000-00087.

Appendix II

P&T projects within the ISTC programme

Russian Institutes working through ISTC projects or under direct contracts with foreign collaborators are performing extensive experimental and theoretical studies on alternative approaches to transmutation, its fuel cycles and associated technological issues. In recent years about 30 ISTC Projects dealing with the transmutation concepts, nuclear data and technologies have been completed /Condé 03/.

ISTC projects of particular importance for transmutation are:

1 1 MW Spallation Target Project (# 559)

This project has been completed in Obninsk. A 1 MW liquid Pb-Bi spallation target has been manufactured and off-beam commissioned. The target was shipped to the University of Nevada, Las Vegas in mid 2002 and it is still undecided by the US-side if and when this target will be irradiated /Dedoul 01/.

2 Project SAD (# 2267)

Subcritical Assembly in Dubna (SAD), a construction of a low power ADS (20-30 kW), fuelled with MOX and driven by an existing 660 MeV accelerator. The facility will consist of a lead or lead-bismuth target, a blanket with U-Pu fuel, a lead reflector, shielding, and experimental channels. The target is to be exposed to a proton flux with the energy of 660 MeV and power of 0.5 kW, the thermal power of the blanket is set at ~20 kW, and $k_{eff} \approx 0.95$. SAD is planned for research in physics and technology to support the nuclear method for transmutation of Pu, MA and LLFP. /Gudowski 01; Gustov 02/.

3 Combined Radiochemical and Activation Analysis of Long-Lived Nuclear Waste Transmuted in Fast Reactors and High Energy Accelerators (# 1372)

The project includes radiochemical analysis and activation measurements of the isotopic composition changes of minor actinide samples irradiated in fast reactors and by the radiation field from a massive lead spallation neutron source. Comparative analysis of the radioactive isotope transmutation efficiency in fast neutron reactors and accelerator driven systems has been carried out. /Krivopustov 03; Kotchetkov 02/.

4 Experimental and Theoretical Studies of the Yields of Residual Product Nuclei Produced in Thin Pb and Bi Targets Irradiated by 40-2600 MeV Protons (# 2002)

The project is aimed at experimental and theoretical studies of the independent and cumulative yields of residual product nuclei in high energy proton irradiation of thin targets of highly enriched isotopes and natural Pb and natural Bi. /Titarenko 03/.

5 Improvement of Corrosion Resistance of Constructional Steels in Liquid Pb and Pb-Bi Alloys by Means of their Surface Modification with the Help of Pulsed Electron Beams and Protective Coatings (# 2048)

The purpose of the project is to develop and ground an effective way to protect constructive steels from corrosion in liquid Pb and Pb-Bi melts at temperatures higher than 500ºC via modification of their surface properties with the help of pulsed intense electron beams. /2048/.

6 Molten Salt Project (# 1606)

Stage 1: Experimental Mock-up of Molten Salt Loop of Accelerator-Based Facility for Transmutation of Radioactive Waste and Conversion of Military Plutonium. *Stage 2*: Experimental Study of Molten Salt Technology for Safe, Low-Waste and Proliferation Resistant Treatment of Radioactive Waste and Plutonium in Accelerator-Driven and Critical Systems. This project is carried out in close co-operation with EU MOST project. The objectives of this project are:

- Reactor physics & molten salt based fuel cycle investigations.
- Experimental studies of key physical and chemical properties of molten fuel salt.
- Corrosion studies in natural convection loop.

The first experimental data, have already been obtained for the selected Na-Li-Be/F salt system and include phase behaviour, solubility of plutonium trifluorides /oxides and transport properties. The construction of corrosion loop made from Ni-based alloy for studies with PuF_3 and redox control in Na,Li,Be/F system has been completed and experiments are under way. /Ignatiev 03a; Ignatiev 03b; Zherebtsov 04/.

7 References Appendix II

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Appendix III

R&D-projects within Swedish programme

1 Neutron data for Accelerator-driven Transmutation Technology – research at Uppsala University

The project NATT, Neutron data for Accelerator-driven Transmutation Technology, is performed within the nuclear reactions group of the Department for neutron research, Uppsala University. The activities of the group are directed towards experimental studies of nuclear reaction probabilities of importance for various applications, like transmutation of nuclear waste, biomedical effects and electronics reliability. The experimental work is primarily undertaken at The Svedberg Laboratory (TSL) in Uppsala, where the group has previously developed two world-unique instruments, MEDLEY and SCANDAL.

NATT is supported as a research task agreement by Statens Kärnkraftinspektion (SKI), Svensk Kärnbränslehantering AB (SKB), Ringhalsverket AB and Totalförsvarets forskningsinstitut (FOI). The project started 2002-07-01 and runs for four years, with a total budget of 6.0 MSEK. The primary objective from the supporting organizations is to promote research and research education of relevance for development of the national competence within nuclear energy.

The aim of the project is in short to:

- Promote development of the competence within nuclear physics and nuclear technology by supporting licentiate and PhD students.
- Advance the international research front regarding fundamental nuclear data within the presently highlighted research area accelerator-driven transmutation.
- Strengthen the Swedish influence within the mentioned research area by expanding the international contact network.
- Provide a platform for Swedish participation in relevant EU projects.
- Monitor the international development for the supporting organizations.
- Constitute a basis for Swedish participation in the nuclear data activities at IAEA and OECD/NEA.

A reference group consisting of Per-Eric Ahlström (SKB), Benny Sundström (SKI), Thomas Lefvert (Vattenfall AB, observer), Fredrik Winge (BKAB) and Katarina Wilhelmsen (FOI) is monitoring the project.

Project NATT was preceded by the project KAT (Kärndata för Acceleratorbaserad Transmutation, i.e., nuclear data for accelerator-driven transmutation). The contract on financial support to the KAT project was for four years, the period 1998-07-01 – 2002-06-30.

The activities within NATT have been closely linked to the EU project HINDAS (see separate description in Appendix I).

Recent highlights:

- Analysis and documentation has been finalized of previously performed measurements of elastic neutron scattering from carbon and lead at 96 MeV. The precision in the results surpasses all previous data by at least an order of magnitude. These measurements represent the highest energy in neutron scattering where the ground state has been resolved. The results show that all previous theory works have underestimated the probability for neutron scattering at the present energy by 0-30 %.
- A new method for measurements of absolute probabilities for neutron-induced nuclear reactions with experimental techniques only has been developed. Previously, only two such methods have been known.
- One student has reached his PhD exam, and one is scheduled for PhD dissertation June 2004. Two PhD students are expected to reach licentiate degrees in 2004.
- TSL has built a new neutron beam facility with significantly improved performance for these – and similar – activities.
- A new instrument for measurements of inelastic neutron scattering has been built, tested and found to meet the specifications. This work has been performed in collaboration with two French research groups from Caen and Nantes. The instrument will be used for a series of experiments during the coming years.
- Evidence of three-body force effects has been found in neutron-deuteron scattering. In a longer term, this can be of large importance in the development of theories of complex nuclei based on fundamental forces.
- Previous work by the group on nuclear data for assessment of electronics reliability has lead to a new industry standard in the USA.

2 Current research on transmutation at KTH-Nuclear and reactor physics

The research on safety of Accelerator-Driven Transmutation Systems (ADS) at the Department of Nuclear and Reactor Physics at the Royal Institute of Technology in Stockholm (KTH) has been largely determined by the programme of the European projects of the Framework Programmes in which KTH is actively participating. In particular:

a) ADS core design and development of advanced nuclear fuel optimized for high transmutation rates and good safety features. This activity includes even computer modelling of nuclear fuel production. Three different ADS-core concepts are being investigated:
   i. Conceptual design of Pb-Bi cooled core with nitride fuel – so called Sing-Sing Core developed at KTH.
   ii. Pb-Bi cooled core with oxide fuel – so called Ansaldo design for the European Project PDS-XADS.
   iii. Gas cooled core with oxide fuel – a design investigated for the European Project PDS-XADS.

b) Analysis of ADS-dynamics and assessment of major reactivity feedbacks.

c) Emergency heat removal from ADS.

d) Participation in ADS experiments including 1 MW spallation target manufacturing, subcritical experiments MUSE (CEA-Cadarache), YALINA subcritical experiment in Minsk and designing of the subcritical experiment SAD in Dubna.

e) Material studies for ADS, in particular theoretical and simulation studies of radiation damage in high neutron (and/or proton) fluxes.

f) Computer code and nuclear data development relevant for simulation and optimization of ADS, special efforts were put in the frame of the European Project PDS-XADS to perform sensitivity studies of the different nuclear data libraries.

g) Studies of transmutation potential of critical reactors in particular High Temperature Gas Cooled Reactor..

The conceptual design of a subcritical transmutation system adjusted to once-through fuel cycle has been developed as a so called Sing-Sing Core (SSC). This subcritical actinide burner is characterized by:

- Pb/Bi-eutectic coolant with compatible structural materials.
- Actinide mononitrides, (U,TRU)N in ZrN matrix, combining good thermal stability together with compatibility with water-based reprocessing techniques.
- Application of burnable absorbers (B_4C) and enlarging core batch-wise, thus reducing the reactivity swing.
- Optimised distribution of fuel and burnable absorber in individual core zones, minimising power peaking and increasing fuel averaged linear power.
- Choice of a core design with large pitches, mitigating consequences of unprotected transients; at the same time, coolant void worth becomes negative resulting with very good safety characteristics.

Extensive safety studies have been performed for this ADS concept including:

- Accident analysis including several scenarios like: the unprotected transient overpower (TOP), the unprotected loss-of-flow (LOF), the unprotected loss-of-heat-sink (LOHS).
- Temperature and void reactivity coefficients.

To complete the safety studies a emergency heat removal concept has been developed employing various inherent safety features. These studies were completed successfully with a PhD thesis.

Transmutation concepts require advanced nuclear fuels. Special efforts have been put into research on nitride fuel. A European project has been created under the KTH coordination with the objectives to manufacture, test, irradiate and post-irradiation analyse Pu-nitride and Am-nitride fuels as promising candidates for transmutation fuels. This EU-project – CONFIRM - is currently running and Pu-N fuel is being irradiated in Studsvik.

Moreover a successful methodology has been developed at KTH to model synthesis and manufacturing of nitride fuels.

Simulation tool development and nuclear data evaluation and processing methodology is a particularly important milestone giving the research group at KTH a uniquely complete and independent tool for advanced transmutation simulations. The central part of this simulation system is a Monte Carlo Burnup code – MCB, an original KTH contribution to the development of Monte-Carlo based burnup codes. A complete

burnup calculation and/or material density evolution in time can be done taking full advantage of reliable 3D neutron transport Monte Carlo methodology coupled with Bateman burnup. A continuous or batch fuel feed/extraction can be simulated including a possibility of reloading and shuffling of the fuel elements. The code uses extensive data libraries that cover nuclide decay schemes, continuous energy transport and reaction cross-sections, isomer state formation ratios, incident energy and target nucleus dependent fission product yields, and radioactive hazard indexes. This code has been extensively benchmarked and a validation process is underway. The MCB code needs temperature dependent, continuous energy neutron/gamma cross sections. To address those needs KTH customized and developed nuclear data evaluation and processing codes giving the possibility to create new nuclear data libraries based on the newest versions of existing evaluated nuclear data files like ENDFB6.8, JEF3.0 and JENDL3.3. Moreover a methodology of generating neutron group constants has been customized for reactor kinetics codes used in transient analysis.

The economical costs and benefits associated with a nuclear waste transmutation strategy have been assessed at KTH identifying the cost drivers of a partitioning and transmutation strategy, and estimating the cost of electricity generated in a nuclear park with operating accelerator driven systems – see Table 5-3 in section 5.5 of the main part of this report.

Addressing an important issue of radiation damages in high, mixed neutron-proton fields a research subproject on Molecular Dynamics study of radiation damage in Fe-Cr alloys resulted in a clear understanding of a microscopic role of Cr ions in radiation induced swelling processes.

Reactor Physics Group at KTH is an active participants and/or coordinator of many European projects in the 5th FWP. In particular:

- CONFIRM and FUTURE projects developing nitride and oxide fuels, respectively. KTH's focus is on some properties of those fuels in ADS systems, like: reactivity evolution, void worth, expansion coefficients, linear rating, limits to burnup, equilibrium composition, etc.
- MUSE - project, an experiment with a subcritical reactor (at CEA/Cadarache) driven by an intense neutron generator. KTH's focus has been on neutron source importance and neutron flux time evolutions.
- PDS-XADS - project developing a nuclear design for experimental accelerator driven system. KTH's involvement has been focused on several key issues: uncertainty evaluation of the nuclear design, assessment of the possibility to adopt neutron absorbing/reflecting elements at core periphery for dpa-reduction alternatively for Po-generation reduction, neutronic calculations for a gas-cooled ADS.
- SPIRE-project investigating materials for a spallation target window. KTH's contribution has been in theoretical studies of Fe-Cr alloys using Molecular Dynamics.

KTH has been also active in a few European network-projects: Molten Salt studies in a MOST-project, coordination of the European transmutation research through the ADOPT-network, ITEM -network - development a multiscale modelling for simulation of radiation effects for Virtual Test Reactors (VTR).

In the EU 6th KTH is coordinating a sizeable European project "Impact of Partitioning, Transmutation and Waste Reduction Technologies on the Final Nuclear Waste Disposal" – RED-IMPACT, with 23 partners representing 10 European countries.

KTH has actively participated in important international reports and roadmaps. In particular in preparation of:

- A European Roadmap for Developing Accelerator Driven Systems (ADS) for Nuclear Waste Incineration, The European Technical Working Group on ADS, ENEA 2001. /TWG 01/.
- Accelerator-driven Systems (ADS) and Fast Reactors (FR) in Advanced Nuclear Fuel Cycles. A comparative Study, NEA/OECD 2002. /NEA 02b/.

Between 2001-2003 KTH's group has published over 30 scientific articles in regular journals, including Physical Review, participated in several large international conferences presented at least 10 invited papers.

3 Current research on transmutation at KTH – Nuclear Power Safety Division

Nuclear Power Safety Division of KTH (NPS) is actively participating in the activities concerned with ADS RD&D. The division is a partner of both PDS-XADS and TECLA projects.

The tasks at NPS focus on thermal-hydraulics and safety issues concerned with the XADS. It should be noted that NPS is the coordinator of the Safety work-packages of both the PDS-XADS and the proposed EUROTRANS project.

TALL test facility and experimental results so far

According to the work tasks, a medium-scale test facility, named Thermal-hydraulic ADS Lead-bismuth Loop (TALL), was designed and constructed at NPS. The test facility is composed of a primary loop (LBE loop) and a secondary loop (oil loop). It is 6.8 m tall and the placement of heaters and heat exchangers is prototypic, and thereby allows significant natural convection flows. The scaling of the primary loop was based on the conceptual XADS design. It has been scaled to represent all the components, their LBE volume, and pressure drops, the flow velocity, the heating rates and the height corresponding to one tube of the heat exchanger design chosen. Thus, the data obtained should be valuable for validation of computational codes and models for the thermal-hydraulic performance of prototypical LBE-cooled accelerator-driven system for transmutation in steady-state and transient operation. Flows in some accidental regimes will also be modelled.

The facility came into operation in September 2003. Since then, investigations on LBE flow and heat transfer have been carried out for a straight tube heat exchanger and a U-tube heat exchanger, respectively. The characteristics of flow resistances and overall heat transfer coefficients were obtained. The details of the TALL and experiments are reported in /Sehgal 03c/. The following conclusions can be noted:

- The TALL test facility is well controlled and thermal-hydraulic conditions can be adjusted properly.
- The main operational parameters, such as LBE flow rate and temperature level, meet the design requirement.
- The oxygen level is measured, which allows the management of the oxygen concentration to an acceptable level.

- The LBE inlet temperatures of the heat exchangers are up to 450 ^{o}C, with the outlet temperature of the core tank up to 500 ^{o}C.
- Pressure drop through the straight tube heat exchanger appears to be higher than that calculated by Techo et al. correlation for a smooth tube, but in good agreement with that calculated by Moody correlation which takes the effect of surface roughness into account /Kakac 87/.
- Pressure drop through the U-tube heat exchanger is higher than that obtained in the straight tube heat exchanger. The pressure drop through the U-tube heat exchanger can be predicted by the correlation developed in the experiment /Sehgal 03c/.
- For both heat exchangers, the heat transfer coefficient of secondary (glycerol) flow is much smaller than that of primary (LBE) flow, and hence dominates the determination of the overall heat transfer coefficient.
- At high LBE temperature level, the heat transfer coefficients of glycerol in the secondary side of the heat exchangers are much higher than those calculated by available correlations. This might mainly result from subcooled boiling of the glycerol. It is strongly suggested that the subcooled boiling should be taken into account when designing the heat exchanger for ADS, if a lower boiling point coolant is employed for the secondary coolant.
- In general, the U-tube heat exchanger has a better heat transfer performance than the straight tube heat exchanger, especially for the secondary flow. Actually, the heat transfer resistance is very low for the primary flow, so the heat transfer enhancement should be considered for the secondary side.

Test plan of transients

Since KTH is the coordinator of safety issues in PDS-XADS project, the transient experiment and analysis are of interest. Using the TALL test facility, specific experiments will be carried out to study transients with reference to safety issues of ADS. While simulating decay heat in the core tank, the temperature and flow rate transient characteristics are measured during the following conditions:

a) Loss of heat sink (switch off the pump of the secondary loop).
b) Loss of external driving head (switch off the pump of the primary loop) − completely passive mode of LBE loop operation.
c) Switch on and off the heater to simulate accelerator trips.
d) Blockage in pipes.
e) Sudden increase in power.

The above experiments will provide first data for the safety evaluations of ADS design which can be employed for validation of safety analysis codes particular to an LBE-cooled reactor.

Analysis with REPLAP5

RELAP5/MOD3.2.2 code which was modified by ANSALDO for the LBE coolant is employed to perform pre-test analysis for transient experiments on TALL facility. Preliminary results concerned with start-up and shut-down of pump were obtained. The calculation was documented in /Sehgal 03b/. More calculations and validation will be done after the transient experiments.

Coordination of Safety Issues of the XADS

Another major part of KTH contributions is coordinating the activities concerned with the safety issues of the XADS. NPS has experience in LWR safety, fast reactor safety, licensing, public safety, etc., which lets it uniquely qualified to provide the contribution.

Safety is always the first consideration of a nuclear reactor. A safety approach needs to be proposed for the XADS based on the European licensing requirement for comparable nuclear facilities. The considerations include the plant siting, operational safety, design-base safety, design-extension safety and severe accident safety & management.

So far, KTH has been coordinating the activities to define the design-base condition (DBC) and design-extension condition (DEC) transients and accidents for the XADS design and, then to perform analysis of such transients and accidents in order to develop the safety analysis reports (PSAR). The main conclusions for the PDS-XADS are reported in subsection 4.1.6 of the main part of this report.

4 Current research on partitioning at CTH

The current work in the CTH group may be divided into two main areas being basic chemistry and process development. The main focus is on understanding and examining the basic chemical properties of extraction systems both by experiments and theoretical derivations.

Basic chemistry

Basic chemical properties of various chemical compounds

We have studied the chemical and extractive properties of several nitrogen containing synergists. Results have been published for 2,2':6',2''-terpyridine and 2,6-bis-benzoxazolyl-4-dodecyloxylpyridine. Many other compounds have been tested too, synthesized at the University of Reading, but the results have not yet been published.

Nitrate complex formation

Many effects we have seen when varying the nitric acid concentration may be attributed to the varying strength of the metal-nitrate complexes present in solution. We have studied the nitrate complex formation with Pm, Eu, Am and Cm. The results will be published in Radiochimica Acta soon. We have also made additional experiments with the rest of the lanthanides to determine their nitrate complex formation constants. The results are promising.

2-bromodecanoic acid

Many of the systems we have studies comprise a ligand and an extraction agent, This agent is in many cases 2-bromodecanoic acid. Thus, the basic chemical properties of 2-bromodecanoic acid (acid constant, dimerisation constant and distribution coefficient) have been studied. It is important to be familiar with how this extracting agent works when using it in combination with different nitrogen-containing synergists. The results give important knowledge about how the extraction mechanism works.

Theory of solubility parameters

Prediction of the distribution of a metal between organic and aqueous phases may be modelled using solubility parameter theory. It was shown that distribution ratios using mixtures of diluents could be modelled with good accuracy.

Process development

A mixer-settler battery has been set into operation and modified for process studies of the ligands developed within the PARTNEW project. First process parameters were determined using an analogue system and then these values were used to design a process for separation of the desired elements. However, due to the cost of the custom made ligands this set-up was not tested. Hopefully, using a newly developed micro separation unit these tests may be performed within EUROPART.

Continuation

The work will proceed much along the lines described above but eventually the focus will shift to more process and computer simulation efforts as we are closing in to a working system. More focus will also be given to real-system parameters such as radiation stability and loading problems with the different systems deemed promising in the scooping studies.

5 References to Appendix III

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TWG 01 A European Roadmap for Developing Accelerator Driven Systems (ADS) for Nuclear Waste Incineration. April 2001. Report prepared by European technical working group of experts on ADS under chairmanship of Carlo Rubbia

Appendix IV

Members and observers of the Swedish reference group for P&T-research

Members:

• SKB	Fred Karlsson; *chairman*
• CTH – nuclear chemistry	Jan-Olov Liljenzin (Christian Ekberg, *deputy*)
• KTH – nuclear and reactor physics	Waclaw Gudowski (Jan Wallenius, *deputy*)
• KTH – nuclear power safety	Bal Ray Sehgal
• Uppsala universitet – neutron research	Jan Blomgren; *secreterary*
SKB – programme manager	Lars Werme, SKB

Observers:

• KASAM	Sören Norrby (Mats Lindman, *deputy*)
• Ministry of environment	Ansi Gerhardsson
• SKB	Per-Eric Ahlström
• SKI	Stig Wingefors
• SSI	Helmuth Zika

Appendix V

List of acronyms

AAA (3A)	Advanced Accelerator Application
ABR	Actinide Burner fast Reactor
ADOPT	Advanced Options for P&T (EU project)
ADS	Accelerator Driven System
ADT	Accelerator Driven Transmutation
AEA-T	AEA Technology, British company (former U.K. Atomic Energy Authority)
AFCI	Advanced Fuel Cycle Initiative (US programme)
ALINA	Actinide - Lanthanide INtergroup separation in Acidic medium
ALWR	Advanced Light Water Reactor
AMS	Accelerator Mass Spectroscopy
AMSB	Accelerator Molten Salt Breeder
AMSTER	Actinides Molten Salt TransmutER
ANL	Argonne National Laboratory
Ansaldo	Italian company, with experience in design, construction and servicing of nuclear power plants
APT	Accelerator Production of Tritium
ASCHLIM	ASsessment of the Computational fluid dynamics codes for Heavy LIquid Metal
ATF	Accelerator Test Facility
ATW	Accelerator driven Transmutation of nuclear Waste
BASTRA	BAsic Studies on TRAnsmutation (EU cluster of projects)
BN	Russian fast neutron reactor (Bystryj Neutronij); BN-600 fast sodium cooled power reactor with 600 MW_e
BNL	Brookhaven National Laboratory
BOC	Beginning Of Cycle
BOL	Beginning Of Life
BOR	Russion fast reactor (also called BOR-60) with 60 MW_{th}
BREI	Burns & Roe Enterprises, Inc.
BREST	Bystryj Reactor so Svicevym Teplonositelem; Russian lead-cooled fast reactor concept
CALIXPART	Selective Extraction of Long Life Nuclides from High Activity Liquid Waste by Organized Matrices: Removal of Minor Actinides and a Process for Cesium Extraction
CAMIX	Composites of AMericium In PheniX
CANDU	Canadian Deuterium Uranium reactor
CAPRA	French design of fast reactor fuel for Pu-burning
CDF	Computational Fluid Dynamics
CEA	Commissariat a l'Energie Atomique, France
CEFR	Chinese Experimental Fast Reactor
CERCER	CERamic CERamic composite
CERMET	CERamic METal composite
CERN	Centre Europeennee pour la Recherche Nucleaire, Switzerland
CIEMAT	Research Centre for Energy, Environment and Technology, Spain

CIRCE	CIRCuito Eutettico; large scale facility for heavy liquid technology development operated at ENEA, Brasimone, Italy
CITON	Centre of Technology and Engineering for Nuclear projects; Romanian nuclear research institute
CMPO	CarbamoylMethyl-Phosphine-Oxide
CNRS	Centre national de la recherché scientifique
COCHIX	Concept Optimised target in Phénix
COE	Cost Of Electricity
COGEMA	Compagnie général des matères nucléaires
CONFIRM	Collaboration on Oxide and Nitride Fuel Irradiation and Modelling
CORAIL	French design of PWR fuel assembly for Pu burning
COSY	Accelerator i Jülich
CPSD	Cross Power Spectral Density
CRIEPI	Central Research Institute of the Electric Power Industries, Japan
CTH	Chalmers Tekniska Högskola, Chalmers technical university, Gothenburg
CW	Continuous Wave
DBC	Design Base Conditions
DB-MHR	Deep Burn Modular Helium Reactor
DEC	Design Extension Conditions
DG RTD	Research Directortate General (EU)
DIAMEX	DIAmide EXtraction
DOE	Department of Energy (USA)
DOVITA	Dry reprocessing, Oxide fuel, Vibropac, Integral, Transmutation of Actinides; Russian concept for an optimised fuel cycle for actinide burner reactor
DPA	Displacements Per Atom
DUPIC	Direct Use of PWR spent fuel in CANDU
EA	Environmental Assessment
EC	European Commission
ECR	Electron Cyclotron Resonance
ECRIX	French heterogeneous recycling experiment in Phenix
EdF	Electricité de France; French national power producer
EFR	European Fast Reactor
EFTTRA	European Fuels and Targets for TRAnsmutation
ENRESA	Empresa Nacional de Residuos Radiactivos, Spain
EOC	End Of Cycle
ETH	Eidgenössische Technische Hochschule, Zürich, Schweiz
EU	European Union
EURATOM	European Atomic Energy Community
EUROTRANS	Integrated Transmutation project, EU 6th FWP
FACET	Fission driven by ACeElerator and Isotopes Transmutation
FEUTRA	Fuel studies for transmutation, cluster including CONFIRM, THORIUM CYCLE and FUTURE
FOI	Totalförsvarets Forskningsinstitut, Swedish Defence Research Institute

FoU	Forskning och Utveckling (Swedish for R&D)
FR	Fast Reactor
Framatome	French company, with experience in design, construction and servicing of nuclear power facilities
FTF	Fuel/Target Fabriction
FUTURE	EU project for fuel studies in particular fuel for ADS
FUTURIX	Phenix irradiation collaboration between DOE, CEA, ITU and JAERI
FWP	FrameWork Programme (European Union)
FZJ	Forschungszentrum Jülich
FZK	Forschungszentrum Karlsruhe
GA	General Atomics
GCFR	Gas Cooled Fast Reactor
GELINA	Geel Electron Linear Accelerator
GENEPI	GEnérateur de Neutrons Pulsé Intense
GRS	Gesellschaft für Anlagen- und Reaktorsicherheit
GSI	Gesellschaft für Schwerionenforschung mbH, Germany
HAC	High Active Concentrate
HAR	High Active Raffinate
HCDA	Hypothetical Core Disruptive Accidents
HINDAS	High and Intermediate energy Nuclear Data for Accelerator driven Systems
HITE	3-dimensional hexagonal finite element code for transient and steady state
HLLW	High Level Liquid Waste
HLW	High Level Waste
HPPA	High Power Proton Accelerator
HTR	High Temperature Reactor
HYPER	Hybrid Power Extraction Reactor
IAC	Idaho Accelerator Centre
IAEA	International Atomic Energy Agency
IBA	Ion Beam Applications, Belgian private company
IL	Ionic Liquid
INEEL	Idaho National Engineering and Environmental Laboratory
IP	Integrated Project
IPPE	Institute of Physics and Power Engineering
IPS	In Pile Sections
IRMM	Institute for Reference Materials and Measurements
ISI&R	In-Service Inspection and Repair
ISTC	International Science and Technology Centre, Moscow
ITEM	Improvement of TEchniques for Multiscale modelling of irradiated materials; EU network in FWP 5
ITN	Instituto Tecnológico E Nuclear, Portugal
ITU	Institute of Transuranium elements, Germany
JAERI	Japan Atomic Energy Research Institute
JINR	Joint Institute for Nuclear Research, Russia
JNC	Japan Nuclear Cycle Development Institute
JPARC	Japanese Proton Accelerator Complex
JRC	Joint Research Centre

KAERI	Korean Atomic Energy Research Institute
KALLA	KArlsruhe Lead LAboratory
KASAM	Statens råd för kärnavfallsfrågor, Swedish National Council for Nuclear Waste
KAT	Kärndata för Acceleratorbaserad Transmutation (Nuclear Data for Accelerator driven Transmutation)
KEK	High Energy Accelerator Research Organisation, Japan
KKP-EnBW	Kernkraftwerk Phillipsburg, Germany
KTH	Kungliga Tekniska Högskolan, Royal Institute of Technology, Stockholm
KWO	Kernkraftwerk Obrigheim, Germany
LAMPF	Los Alamos Meson Physics Facility
LANL	Los Alamos National Laboratory
LBE	Lead Bismuth Eutectic
LFBR	Large Fast Breeder Reactor
LINEX	Lithium Nitrate Extraction of actinides
LLFP	Long Lived Fission Products
LOF	Loss Of Flow
LOHS	Loss Of Heat Sink
LWR	Light Water Reactor
MA	Minor Actinides
MACS	Magnetically Assisted Chemical Separation
MASURCA	Reseach reactor at Cadarache, France
MCB	Monte Carlo Burnup code
MCYT	Spanish Science and Technology Ministry
MEDLEY	Facility for neutron beam irradiation in MEDical and other applications at TSL in Uppsala, Sweden
MEGAPIE	Spallation target experiment at PSI, Switzerland
MES	Mitsu Engineering and Shipbuilding Co. Ltd.
MFBR	Modular Fast Breeder Reactor
Minatom	Ministry of Atomic Energy, Russia
MOSART	Molten Salt Reactor Transmuter
MOST	Review of Molten Salt Technology; EU network in FWP 5
MOX	Mixed Oxide, UO_2 mixed with some fraction PuO_2
MSBR	Molten Salt Breeder Reactor
MUSE	EU project on subcritical measurements in fast neutron spectrum at MASURCA
MYHRRA	ADS test and irradiation facility proposed to be built at SCK·CEN, Belgium
NATT	Neutron data for Accelerator driven Transmutation Technology
NCSR-INP	National Centre for Scientific Research – Institute of Nuclear Physics, Athens Greece
NEA	Nuclear Energy Agency within OECD
NERAC	Nuclear Energy Research Advisory Committee at US DOE
NMR	Nuclear Magnetic Resonance
NNC	National Nuclear Corporation, British nuclear industrial Co

NITRAF	Nitride irradiation modelling
NIREX	British nuclear industry owned organisation which sets policy for the disposal of nuclear waste, UK
NPP	Nuclear Power Plant
NRG	Nuclear Research and consultancy Group**,** Netherlands
NRI	Nuclear Research Institute, Czech Republic
NTOF	Neutron Time of Flight, measurement device at CERN
OECD	Organisation for Economic Cooperation and Development
OMEGA	Options Making Extra Gains of Actinide and Fission Products
ORNL	Oak Ridge National Laboratory
P&T	Partitioning and Transmutation
P&T/C	Partitioning and Transmutation / Conditioning
PARTNEW	Partitioning: New Solvent Extraction Processes for Minor Actinides
pcm	pro cent mille (10^{-5})
PDS-XADS	Preliminary Design Studies for an Experimental ADS
PEG	PolyEthylene Glycol
PFBR	Prototype Fast Breeder Reactor
PISA	Proton Induced Spallation
PNS	Pulsed Neutron Source
PSAR	Preliminary Safety Analysis Report
PUREX	Plutonium Uranium Redox EXtraction
PWR	Pressurized Water Reactor
PYROREP	Pyrometallurgical Processing Research Programme
R&D	Research and Development
RAWRA	Sprava ulozist radioaktivnich odpadu; waste management company in Czech Republic
RBMK	Russian acronym for large boiling water graphite moderated channel reactor (Chernobyl type reactor)
RCS	Rapid Cycling Synchrotron
RD&D	Research, Development and Demonstration
RDIPE	Research and Development Institute of Power Engineering, Moscow, Russia
RED-IMPACT	EU project to study impact of P&T using ADS
RFQ	Radio Frequency Quadrupole
RIA	Reactivity Initiated Accidents
RIAR	Research Institute of Atomic Reactors; Dimitrovgrad, Russia
RTD	Research and Technological Development
S&T	Separation och Transmutation
SAD	Subcritical Assembly in Dubna
SANEX	Selective ActiNide EXtraction
SCANDAL	SCAttered Neutron Detection Assembly; at TSL in Uppsala, Sweden
SCE	Supercritical Fluid Extraction
SCK·CEN	Belgian Nuclear Research Centre
SCL	SuperConducting Linear accelerator

SETFICS	Solvent Extraction for Trivalent f-element Intra-group Separation in CMPO-complexant System (carbamoylmethyl-phosphineoxide)
SINQ	Swiss Spallation Neutron Source
SKB	Svensk kärnbränslehantering AB, Swedish Nuclear Fuel and Waste Management Co
SKI	Statens kärnkraftsinspektion, Swedish Nuclear Power Inspectorate
SLM	Supported Liquid Membrane
SNF	Spent Nuclear Fuel
SNL	Sandia National Laboratory
SNR	German sodium cooled power fast reactor for 300 MW_e never completed
SNS	Spallation Neutron Source
SPIRE	EU project for study of materials changes due to particle irradition
SPX	Superphenix
SRS	Savannah River Site
SSC	Sing Sing Core
SSI	Statens strålskyddsinstitut, Swedish Radiation Protection Authority
TALL	Thermal-hydraulic ADS Lead-bismuth Loop
TALSPEAK	Trivalent Actinide Lanthanide Separation by Phosphorus Extractant and Aqueous Complexes
TASSE	Thorium based Acceleratoro driven System with Simplified fuel cycle for long term Energy production
TECLA	Technology Materials and Thermal-Hydraulics for Lead Alloys
TESTRA	TEchnological Studies on TRAnsmutation
THORIMS-NES	Thorium Molten Salt Nuclear Energy Synergetics
TOF	Time Of Flight
TOP	Transient Over Power
TRADE	TRiga Accelerator Driven Experiment, Italian project.
TRISO	Tri-ISOtropic; multilayer fuel particle coating of pyrolytic carbon and silicon carbide
TRU	TRansUranium elements
TRUEX	TRransUranium EXtraction
TSL	The Svedberg Laboratory, Uppsala Sweden
TWG	Technical Working Group
UNEX	UNiversal solvent EXtraction
UNLV	University of Nevada – Las Vegas
UOX	Uranium OXide
URENCO	Uranium enrichment company with facilities in the U.K., the Netherlands and Germany
UREX	URanium EXtraction
USDOE	United States Department Of Energy
USTUTT	University of Stuttgart, Germany
UU	Uppsala University
VENUS	Chinese subcritical assemblydrivenm by a neutron generator

VTR	Virtual Test Reactor
VUJE	Engineering, design and research organisation in Trava, Slovakia
VVER	Russian acronym for PWR of Russian construction
WP	Work Package (EU)
XADT	Experimental Accelerator driven Transmutation
YALINA	Subcritical assembly in Minsk, Belarus