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Partitioning and transmutation Current developments – 2007

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

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Preface

This report is 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 teams that are 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 1. A late draft of the report has been made available for comments to the members and observers of the reference group. The editor and 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 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.

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[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.

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 (FP) 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, USA and Russia.

A review of the status of the efforts concerning P&T was published by SKB in 1998 /Enarsson et al. 1998/. A second status report was compiled by the Swedish reference group on P&T-research in the first half of 2004 /Ahlström et al. 2004/. The present report summarises the progress in the field through the years 2004–2006.

System studies of partitioning and transmutation

The previous status report /Ahlström et al. 2004/ summarised the results from a number of systems studies of partitioning and transmutation. The studies had been conducted on national and on international bases in Europe and USA as well as within OECD/NEA. Follow-up and supplement of the studies have been made by a couple of new studies within OECD/NEA and by a broad international overview within IAEA /NEA 2006a, IAEA 2004/. Furthermore in conjunction with the French decisions concerning the continued work on management and final disposal of high level waste an account has been given of the past fifteen years work of partitioning and transmutation /CEA 2005, CNE 2006, NEA 2006b/.

The NEA-study of advanced nuclear fuel cycles and management of radioactive wastes was performed by an international expert group during the years 2004–2005. The group makes i.a. the following conclusions:

- The picture presented in earlier NEA-studies is confirmed by the new study.
- Thus it is possible to establish a strategic development towards a maximum decrease of the waste source term and a maximum utilisation of the uranium resources.
- The fuel cycles that have been studied illustrate that the long-lived radioactivity may be reduced by burning primarily the plutonium and then the other transuranium elements. If the fraction of fast reactors is increased towards 100% it is also feasible to substantially reduce the uranium consumption per produced kWh electricity.
- Considering the width and flexibility of the advanced nuclear fuel cycles under development it should be possible to design and construct a safe and economical nuclear energy system that in an efficient way meets the demands on economizing with natural resources and a good waste management. The strategic choices will be based on specific national conditions as availability of uranium, availability of waste repositories, size of nuclear programme as well as social and economic considerations.

An issue particularly emphasised in the study is the importance of decay heat for the size of a repository. The high level waste from advanced nuclear fuel cycles requires considerably less volumes than the once-through cycle for light water reactors. Per produced kWh electricity the required volume may decrease by a factor of 5–20. A contributing reason is that advanced reactors work at higher temperatures and thus have better thermal efficiency. It is also presumed that caesium and strontium are separated and stored separately or used in some way.

The economical analyses included in the NEA-study give the same indications as in previous reports. The cost uncertainties are however considerable and exceed the differences between different fuel cycles included in the analyses.

The IAEA-study also confirms the conclusions drawn by earlier system studies. The report emphasises that the risks of proliferation of special nuclear material must be considered within all programmes for partitioning and transmutation (as well as partitioning and conditioning). International cooperation may play an important role in this context. Furthermore increased integration of the efforts on P&T and waste management is emphasised.

The extensive French reporting from 15 years of work following the special law of 1991 has formed the basis for the continued programme now decided by the French government. Important parts of this programme are continued efforts for development of partitioning and transmutation within a frame of a closed fuel cycle. The emphasis is on development of advanced reactors within the so-called Generation IV. A goal is construction of a modern fast reactor, probably sodium cooled, to be ready for operation in 2020.

The main French report /CEA 2005/ was reviewed by an expert group appointed by NEA. The review report /NEA 2006b/ includes some conclusions of general character i.a.:

- The research on fuel and targets is the weakest R&D-link with the P&T-area. This research is very important for fast reactors as well as accelerator-driven systems.
- A concern for the development of P&T is the shrinking infrastructure for R&D and in particular the lack of facilities for irradiation with fast neutrons. The development needs an adequate long-term financing.
- Accelerator-driven systems may play a role within a so-called double strata system where all plutonium is burned in commercial power reactors and the minor actinides in accelerator-driven systems.

Transmutation

The major European project specifically focusing on a transmutation system is the Eurotrans project. This four-year project started in April 2005 and has a budget of 43 M€ of which 23 M€ are contributed by the European commission. It has 47 participants from 14 countries of which 10 represent industries, 19 research centres and 17 universities in Europe. The universities are collectively represented in the project organisation by the ENEN (European Nuclear Engineering Network). Sweden is represented by KTH in Stockholm and by the neutron research group at Uppsala University. The project is focusing on accelerator-driven systems and is subdivided into five sub-domains addressing

1. Design.
2. Coupling of accelerators with sub-critical cores.
3. Minor actinide fuel development.
4. Structural materials behaviour under LBE[2]-cooling and fast neutron irradiation.
5. Basic nuclear data.

Eurotrans is an integrated continuation of the PDS-ADS[3] and other projects of the fifth frame work programme. The PDS-ADS was conducting preliminary design studies of accelerator-driven systems. Three different designs were considered:

1. An 80 MWth LBE cooled core with low linear rating (11 kW/m), originating from a proposal by Ansaldo[4]. A major design feature was argon gas bubbles to be introduced above the core in order to enhance natural circulation.
2. An 80 MWth He cooled core with medium linear rating (25 kW/m), originally proposed by Framatome[5].
3. A 50 MWth LBE cooled core with high linear rating (50 kW/m), originating from SCK-CENs[6] Myrrha design. This facility would also have the purpose of serving as an irradiation facility for materials testing, including thermal spectrum islands.

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[2] LBE = Liquid Bismuth Eutecticum.

[3] PDS-ADS = Preliminary Design Study of an Accelerator-Driven System.

[4] Ansaldo = Italian industrial company.

[5] Framatome = French nuclear industrial company (now partner with Siemens in Areva).

[6] SCK-CEN = Belgian national nuclear company operating the laboratories at Mol.

The PDS-XADS project was completed in 2005 and the major conclusions may be summarised as follows:

- The low rated design of Ansaldo featured the best safety characteristics, permitting heat removal by natural circulation during various accident scenarios. The low neutron flux would however result in poor performance in terms of minor actinide burning rates.
- The He cooled core might encounter problems with decay heat removal in case of depressurisation. The presence of a spallation target makes these problems more complicated to manage than in the case of a critical fast gas cooled reactor.
- The Myrrha design would meet difficulties in surviving certain types of accidents, due to the relatively low fraction of natural circulation in combination with a very high linear rating.

In total, the results were encouraging enough for the European research community to launch a larger effort in the sixth framework program, working towards the demonstration of transmutation in Europe by carrying out

1. An advanced design of a 50–100 MW experimental facility demonstrating the feasibility of transmutation in an accelerator driven system (XT-ADS), being possible to realise in a short-term (~10 years).
2. A conceptual design of a several hundred MWth European Facility for Industrial Transmutation of minor actinides (EFIT), to be realised in a longer term.

Beside the design studies the main part of the R&D-efforts for transmutation are devoted to studies of fuel and materials. The results from these efforts are applicable to fast reactors as well as accelerator-driven systems.

Some recent status of these studies are summarised in the report.

Partitioning

The research on partitioning is following two main routes – hydrochemical processes and pyrochemical processes. In Europe the efforts are coordinated within the EU-project Europart that started in 2004 and shall be completed in June 2007. The project is a so-called integrated project within the sixth frame work programme sponsored by the European commission. It constitutes a continuation of the projects Partnew, Calixpart and Pyrorep within the previous frame work programme. Partnew was focused on separation of actinides and lanthanides from water solutions by liquid extraction and use of nitrogen donating ligands as e.g. malonamides. Calixpart also worked with liquid extractions of water solutions but using calixarenes. Pyrorep studied pyrochemical methods for partitioning in salt or metal melts.

Europart is divided into six work packages – five in hydrochemistry and four in pyrochemistry. Participants are 24 organisations in 11 EU countries plus in addition Australia and Japan. Sweden is represented by the nuclear chemistry group at Chalmers in Göteborg. The Swedish research on partitioning is currently well integrated with the EU-projects.

The research on partitioning has made important progress in recent years. In some cases one has succeeded to separate americium and curium. Many challenges remain however. Within hydrochemistry one has achieved sufficiently good distribution and separation factors. The focus turns now towards development of an operating process. The search for ligands that give sufficiently good extraction and separation will continue but with less intensity. The emphasis will rather be on improving stability against hydrolysis and radiolysis. This may be achieved either by additives to the solvent or by selection of a proper solvent. The development of processes and equipment must be intensified.

Pyrochemical research is looking into methods for recovery of uranium and for separating fission products with large neutron cross sections. The objective is to avoid separation of plutonium from other transuranium elements and thus simplify the proliferation issue.

The future work is focused on improved selectivity and on technical development. Design of processes and equipment is difficult due to the aggressive properties of the melts and the relatively high temperatures required.

The fabrication of fuel for transmutation and the reprocessing of transmutation fuel require considerable development that cannot be conducted in full until the fuel has been better specified. Such development for an advanced fuel cycle will thus need additional time.

Some conclusions

The gradual increase of the efforts for research and development on P&T during the period 1990–2003 has not continued after 2003. The financial support from the European commission to P&T-projects has levelled off. It is unclear which support that really will be available in future programmes. Within the commission the expectations seem to be an unchanged level during the foreseeable future.

The research on P&T has still a prominent international position among R&D-programmes for future nuclear power and nuclear fuel systems. It attracts considerable interest among students in nuclear disciplines. The interest within the nuclear energy industry is limited. P&T is seen as an interesting option within a future energy system based on advanced nuclear reactors, advanced nuclear fuel and advanced nuclear fuel cycles.

The interest for P&T within the frame of a phase-out of the current nuclear power programme has more or less disappeared.

A successful development of P&T within the frame of advanced nuclear fuel cycles will not eliminate the need for deep geologic repositories for high level waste and for long-lived wastes. The complex processes will unavoidably create waste streams containing small amounts of long-lived radionuclides. The development may however decrease the demands on engineered barriers as well as the required volumes in geological repositories.

From a Swedish perspective it is important to follow the international development and maintain a reasonable level of competence within the country at least as long as an essential part of the electricity production is based on nuclear energy. The competence developed by research on P&T is valuable not only for evaluating the progress and potential within this field but also for development of safety and fuel supply at existing nuclear facilities.

The application of P&T for an effective and substantial decrease in the amount of long-lived elements that need final disposal implies the use of nuclear energy during a very long time – above 100 years.

Sammanfattning

Forskning och utveckling av metoder för separation och transmutation (S&T[7]) 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 slutförvar.

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 som på så sätt omvandlas till andra nuklider. 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 (avskiljs) 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[8] – neptunium[9], 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.

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.

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[7] 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.

[8] 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.

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

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, 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 et al. 1998/. En andra lägesrapport utarbetades inom referensgruppen för forskning och utveckling kring separation och transmutation i början av 2004 /Ahlström et al. 2004/. Den nu föreliggande rapporten sammanfattar den utveckling som skett under åren 2004–2006.

Studier av system för separation och transmutation

I den förra lägesrapporten /Ahlström et al. 2004/ redovisades vissa resultat från ett antal systemstudier över separation och transmutation. Studierna hade genomförts på nationell och internationell bas i Europa och USA samt inom OECD/NEA. Dessa studier har följts upp och kompletterats med ett par studier inom OECD/NEA samt en bred internationell genomgång inom IAEA /NEA 2006a, IAEA 2004/. Vidare har i samband med besluten i Frankrike om det fortsatta arbetet på behandling och slutförvaring av högaktivt avfall redovisats en sammanfattning av de senaste femton årens arbete på separation och transmutation /CEA 2005, CNE 2006, NEA 2006b/.

NEA-studien över avancerade kärnbränslecykler och hantering av radioaktivt avfall genomfördes av en internationell expertgrupp under åren 2004–2006. Gruppen drar bl a följande slutsatser:

- Den bild som framkommit från tidigare NEA studier bekräftas av den nya studien.
- Det är således möjligt att etablera en strategisk utveckling mot en maximal minskning avfallets (radioaktiva) källterm och ett maximal utnyttjande av uranresurserna.
- De bränslecykler som undersökts illustrerar att man kan reducera den långlivade radioaktiviteten genom att i första hand bränna plutonium och sedan övriga transuraner. Om andelen snabba reaktorer i systemet ökas mot 100% kan man också högst väsentligt reducera uranförbrukningen per producerad kWh el.
- Med hänsyn till bredden och flexibiliteten hos de avancerade kärnbränslecykler som är under utveckling bör det vara möjligt att utforma och bygga ett säkert och ekonomiskt kärnenergisystem som på ett effektivt sätt möter kraven på hushållning med naturresurser och en bra avfallshantering. De strategiska valen kommer att baseras på specifika nationella förhållanden såsom tillgång till uran, tillgängliga avfallsförvar, storlek på kärnenergiprogram samt sociala, och ekonomiska överväganden.

En fråga som betonas särskilt i studien är betydelsen av restvärme för erforderlig storlek på ett slutförvar. Det högaktiva avfallet från avancerade kärnbränslecykler kräver väsentligt mindre volym än direktdeponering av använt bränsle från lättvattenreaktorer. Per producerad kWh el kan volymbehovet minska med en faktor 5–20. En bidragande orsak är att avancerade reaktorer arbetar med högre temperatur och därför har bättre termisk verkningsgrad. Det förutsätter också att man förutom aktiniderna även separerar cesium och strontium och förvarar dessa ämnen i separata förvar eller nyttjar dem på annat sätt.

De ekonomiska analyser som ingår in NEA-studien ger samma indikationer som tidigare rapporter. Osäkerheterna i kostnaderna är emellertid avsevärda och överstiger skillnaderna mellan de olika analyserade bränslecyklerna.

IAEA-studien bekräftar också de slutsatser som dragits av tidigare systemstudier. I rapporten betonas att riskerna för avledning av klyvbart material (proliferation) måste beaktas särskilt inom alla program för separation och transmutation (och även separation och konditionering). Härvid kan internationell samverkan spela en viktig roll. Vidare efterlyses en ökad integration av insatserna på separation och transmutation med arbetet på kärnavfallshantering.

Den omfattande franska rapporteringen från nära 15 års arbete till följd av den särskilda lagen 1991 har legat till grund för det fortsatta program som nu beslutats. En viktig del av detta är fortsatta insatser på utveckling av metoder för separation och transmutation inom ramen för en sluten bränslecykel. Tonvikten ligger dock på utveckling av avancerade reaktorer inom den s k Generation IV. Man siktar på att en prototyp till en ”modern” snabb reaktor, troligen natriumkyld, skall stå klar år 2020.

De franska rapporterna granskades av en internationell expertgrupp inom NEA. Gruppens granskningsrapport /NEA 2006b/ innehåller några slutsatser som är av mer allmän karaktär bl a:

- Forskningen på bränsle och strålmål är den svagaste länken inom FoU-kedjan för separation och transmutation. Denna forskning är mycket viktig för såväl snabba reaktorer som för acceleratordrivna system.
- Ett bekymmer för utvecklingen av separation och transmutation är den krympande infrastrukturen för FoU och särskilt bristen på utrustning för bestrålning med snabba neutroner. Utvecklingen kräver adekvat långsiktig finansiering.
- Acceleratordrivna system kan spela en potentiellt användbar roll inom ett s k ”double strata system” där allt plutonium bränns i kommersiella reaktorer och ”minor actinides” i acceleratordrivna system.

Transmutation

Det största europeiska projektet som direkt inriktas på transmutation är det s k Eurotrans-projektet. Detta är på fyra år och startade i april 2005 med en budget på 43 M€ varav 23 M€ betalas av European Commission. Projektet har 47 deltagare från 14 länder, varav 10 representerar industrin, 19 forskningscentra och 17 europeiska universitet. Universiteten representeras kollektivt i projektorganisation av ENEN (= European nuclear engineering network). Sverige representeras av KTH i Stockholm och av gruppen för neutronforskning vid Uppsala universitet. Projektet är inriktat på acceleratordrivna system och uppdelas på fem s k domäner:

1. Utformning av systemet.
2. Koppling mellan accelerator och underkritisk reaktorhärd.
3. Utveckling av transuranbränsle.
4. Materialegenskaper vid kylning med flytande bly-vismut och bestrålning med snabba neutroner.
5. Grundläggande kärndata.

Eurotrans är ett s k integrerat projekt inom EU:s sjätte ramprogram och utgör en fortsättning på ett flertal projekt inom det femte och tidigare ramprogram.

Inom femte ramprogrammet studerades olika utformningar av acceleratordrivna system inom projektet PDS-XADS (= preliminary design study of accelerator-driven system). Tre olika utformningar studerades:

1. En 80 MW termisk LBE[10] kyld reaktorhärd med låg bränsleeffekt (11 kW/m) ursprungligen föreslagen av Ansaldo[11]. En speciell egenskap är att argonbubblor förs in ovanför härden för att förbättra den naturliga cirkulationen.

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[10] LBE = Liquid Bismuth Eutectic – flytande bly-vismut eutektikum.

[11] Ansaldo = Italienskt industriföretag.

2. En 80 MW termisk helium kyld reaktorhärd med intermediär bränsleeffekt (25 kW/m) ursprungligen föreslagen av Framatome[12].
3. En 50 MW termisk LBE kyld reaktorhärd med hög bränsleeffekt (50 kW/m) från SCK-CEN[13]:s Myrrha-projekt. Myrrha avses fungera som en bestrålningsanläggning för materialprovning och innefatta en zon med termiskt neutronspektrum.

Projektet PDS-XADS slutfördes under 2005 och de viktigaste slutsatserna var:

- Ansaldo-utförandet med låg bränsleeffekt hade de bästa säkerhetsegenskaperna och kunde kyla bort resteffekten med naturlig cirkulation i olika olycksscenarier. Det låga neutronflödet ger emellertid dåligt utbyte vad gäller förbränning av ”minor actinides”.
- De helium kylda utformningen kan ge problem med resteffektkylningen vid lågt gastryck. Närvaron av spallationskälla gör dessa problem svårare att bemästra än för en kritisk snabb gas kyld reaktor.
- Myrrha-utformningen har svårt att klara vissa olycksscenarier beroende på relativt låg naturlig cirkulation i kombination med hög effekttäthet.

Sammantaget ansågs resultaten dock tillräckligt lovande för att man skulle starta det större Eurotrans i det sjätte ramprogrammet. Syftet är arbete mot en demonstration av transmutation i Europa genom att genomföra:

1. En avancerad utformning av en 50–100 MW termisk experiment anläggning med acceleratordrivet system (XT-ADS) vilken är möjlig att realisera på kort sikt (ca 10 år).
2. En konceptuell utformning av en flera hundra MW termisk europeisk anläggning för industriell transmutation av ”minor actinides” (EFIT) som kan realiseras på längre sikt.

Förutom studier av utformning av systemen berör huvuddelen av FoU-insatserna på transmutation bränsle- och materialfrågor. Resultaten från arbeten inom dessa områden är tillämpliga på såväl på snabba reaktorer som på acceleratordrivna system.

Några intressanta resultat från de pågående projekten presenteras i rapporten.

Separation

Forskningen inom separation är inriktad mot två huvudlinjer – vattenkemiska processer och pyrokemiska processer. I Europa samlas insatserna inom EU-projektet Europart som startade 2004 och avslutas i juni 2007. Detta är ett s k integrerat projekt inom det sjätte ramprogrammet och utgör en fortsättning på projekten Partnew, Calixpart och Pyrorep inom det föregående ramprogrammet. Partnew inriktades på separation av aktinider och lantanider från vattenbaserade lösningar med vätskeextraktion och användning av kväveavgivande ligander som t ex malonamider. Calixpart handlade också om vätskeextraktion av vattenlösningar men baserat på calixarener. Pyrorep studerade pyrokemiska metoder för separation i salt- eller metallsmältor.

Europart uppdelas i nio arbetspaket varav fem inom hydrometallurgi och fyra inom pyrometallurgi. I projektet deltar 24 olika organisationer i 11 EU-länder samt dessutom Australien och Japan. Från Sverige medverkar kärnkemigruppen vid Chalmers. Den svenska forskningen på separation är f n väl integrerad med EU-projekten.

Forskning på separationsprocesser för transmutation har gjort viktiga framsteg på senare år. I vissa fall har man till och med lyckats separera americium och curium. Det återstår dock många utmaningar. Inom hydrometallurgi har man uppnått tillräckligt bra fördelningsfaktorer och separationsfaktorer. Fokus skiftar nu mot att utveckla en fungerande process. Sökandet efter ligander som ger tillräckligt bra extraktion och separation kommer att fortsätta men med mycket mindre intensitet. Tonvikten kommer i stället att ligga på att förbättra stabiliteten mot radiolys

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[12] Framatome = Franskt industriföretag; numera ingående i Areva (tillsammans med tyska Siemens).

[13] SCK-CEN = Nationella belgiska kärnforskningsföretaget.

och hydrolys. Detta kan ske antingen genom tillsatser till lösningsmedlet eller genom val av lämpligt lösningsmedel. Utvecklingen av processer och utrustning behöver intensifieras.

Inom pyrometallurgi pågår forskning på bl a återvinning av uran och på metoder att ta bort klyvningsprodukter med stora neutronabsorptionstvärsnitt. Syftet är att undvika separation av plutonium från andra transuraner och därigenom förenkla safeguardsproblemet. Det framtida arbetet fokuseras mot förbättrad selektivitet och mot teknisk utveckling. Utformning av processer och utrustning är besvärlig med hänsyn till smältornas aggressiva egenskaper och de relativt höga temperaturer som krävs.

Tillverkning av bränsle för transmutation och upparbetning av transmutationsbränsle kräver en betydande utveckling som inte kan genomföras fullt ut innan man har definierat vilket bränsle som skall användas för transmutationen. Utveckling av denna del av en avancerad bränslecykel kommer således att kräva ytterligare tid.

Några slutsatser

Den gradvisa ökning av insatserna på forskning och utveckling av S&T som skedde under perioden 1990–2003 har inte fortsatt efter 2003. Det finansiella stödet från Europakommissionen till S&T-projekt inom EU har planat ut. Det är oklart vilket stöd som verkligen blir tillgängligt i framtida program. Inom kommissionen synes man förvänta sig en oförändrad nivå under överskådlig tid.

Forskning inom S&T har fortfarande en framträdande position internationellt inom FoU för framtida kärnkraft- och kärnbränslesystem. Det tilldrar sig betydande intresse bland studenter inom kärntekniska ämnen. Intresset inom kärnenergiindustrin är mer begränsat. S&T ses mer som en intressant option inom framtida energisystem baserade på avancerade kärnreaktorer, avancerat kärnbränsle och avancerade kärnbränslecykler.

Intresset att genomföra S&T inom ramen för utfasning av existerande kärnkraftsprogram har mer eller mindre försvunnit.

En lyckad utveckling av S&T inom ramen för avancerade bränslecykler kommer ej att eliminera behovet av slutförvar för högaktivt och långlivat avfall. De komplexa processerna kommer oundvikligen att generera avfallsflöden med små mängder långlivade radionuklider. Däremot kan utvecklingen minska kraven på de tekniska barriärerna liksom den erforderliga volymen i geologiska slutförvar.

För svensk del är det viktigt att följa den internationella utvecklingen och upprätthålla en rimlig kompetensnivå inom landet åtminstone så länge som en väsentlig del av landets elproduktion baseras på kärnenergi. Kompetens som utvecklas vid forskning på S&T är värdefull inte enbart för att bedöma utveckling och potential inom detta område utan också för utveckling av säkerhet och bränsleförsörjning vid existerande reaktorer.

Tillämpning av separation och transmutation för att effektivt minska mängden långlivade ämnen som behöver slutförvaras innebär utnyttjande av kärnenergi under mycket lång tid – över 100 år.

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 since the early 1990ies. 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 1995/. 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, hence transforming them into stable or more short-lived isotopes. This process will release a substantial amount of energy and 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[14], 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.

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 (FP) have established

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[14] 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.

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, 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 chemical and biological engineering 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 FP's are shown in Table 1-1.

Earlier reviews of the status of the efforts concerning P&T were published by SKB in 1998 /Enarsson et al. 1998/ and 2004 /Ahlström et al. 2004/. Short reviews of the previous development have also been given in SKB's RD&D-programmes 95, 98, 2001 and 2004 /SKB 1995, 1998, 2001, 2004/.

Another Swedish review of the developments concerning P&T was made by KASAM and published in /KASAM 2004 pp 347–411/. Under the headline "Partitioning and transmutation – an alternative to final disposal An issue in focus" KASAM describes the basic principles of P&T, gives a summary of the state-of-the-art and of ongoing and planned research. Three different scenarios are discussed from a Swedish perspective:

- An exclusively Swedish transmutation system.
- A system where Sweden completely depends on the technology and resources developed in other countries.
- Partitioning and fuel fabrication abroad, transmutation in Sweden.

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

Programme	Time frame	Budget M€
FP 3	1991–1994	4.8
FP 4	1994–1998	5.8
FP 5	1999–2002	28.6
FP 6	2003–2006	38
FP 7	2007–2010	~ 40 ??

In the concluding remarks KASAM discusses the advantages and disadvantages of P&T and makes the following concluding statement:

> The application of P&T to Swedish nuclear waste will be a question for future generations. With present-day knowledge of this technology, it is not acceptable to interrupt or to postpone the Swedish nuclear power programme, citing P&T as an alternative. On the other hand, this possible future alternative reinforces the requirement that the repository should be de-signed so that waste retrieval is possible. According to the ethical principles that KASAM and others have established, each generation should take care of its own waste and not force future generations to develop new technologies to solve the problems. Therefore, it is reasonable for resources to be put aside for further research on P&T. This research could also pay off in ways which are of value for other areas, such as nuclear physics, chemical partitioning technology and materials technology. Swedish P&T research should be co-ordinated with the research and development being conducted in other countries. To, at this stage, allocate resources for further P&T research is also in line with the view that our generation should give future generations the best possible conditions to decide whether they want to choose P&T as a method for taking care of spent nuclear fuel, instead of direct disposal alone (in accordance with the KBS-3 method, for example).

This current status report summarises the work reported in the years 2004–2006 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 1.

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 concerning reporting about "current status" in the statutes for the aforementioned P&T reference group.

The report is structured in four main chapters. Chapter 2 gives a short account of recent international and foreign work concerning P&T. Chapter 3 gives a short account of some results from the Swedish work. Chapter 4 attempts to present some views on the need for future R&D important for P&T. The last chapter, Chapter 5 gives a short update of the assessments of the prospects for P&T presented in the 2004 status report /SKB 2004/.

2 Summary of some international studies since 2004

2.1 Advanced nuclear fuel cycles and radioactive waste management – NEA study

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 1990, 1992, 1994, 1996, 1998, 2000, 2002a, 2005, 2006c/. The committee has also organised a number of expert groups which have reported on various aspects of partitioning and transmutation in OECD/NEA reports. The first two of these studies – *Actinide and fission product P&T – status and assessment report 1999* /NEA 1999/ and *Accelerator-driven systems (ADS) and fast reactors (FR) in advanced nuclear fuel cycles. A comparative study* /NEA 2002b/ – were summarised in the previous status report /SKB 2004/.

A third NEA expert-group study was published in the spring of 2006 – *Advanced nuclear fuel cycles and radioactive waste management* /NEA 2006/. This study broadens the scope of the analyses carried out in previous NEA studies of potential P&T systems and advanced fuel cycles. It addresses waste management issues, providing performance assessment for some examples of high-level waste (HLW) repositories. The impact of different HLW isotopic compositions on repository performance is analysed. Furthermore the effect of advanced fuel cycle schemes on HLW repository capacity is assessed.

The expert group summarises the main findings as follows;

> The overall findings from the present study confirm the vision set up by previous NEA studies. In particular, they show that it is possible to establish a strategic progression towards a maximal reduction of the waste source term and a maximal use of uranium resources. The nine fuel cycle schemes and four variants investigated in the study illustrate the possibility to reduce the activity of the waste by burning plutonium, then minor actinides, and then to reduce specific uranium consumption (per unit of electricity generated) as the fraction of fast reactors in the mix is increased to 100%.
>
> Given the wide range and flexibility of advanced fuel cycles under development – represented in this study by a limited number of representative schemes – it should be feasible to design and implement safe and economic nuclear energy systems addressing efficiently natural resource and waste management issues. Strategic choices will be based on the priorities of policy makers which reflect country specific criteria such as characteristics of available waste repositories, access to uranium resources, size of the nuclear power programme, and social and economic considerations.

The fuel cycles analysed are:

Open fuel cycles:

1. Once through fuel cycle with enriched UO_2 in PWR (EPR[15]) burned to 60 MWd/kg.
2. Plutonium (Pu) recycled once in PWR as MOX[16] and burned to 60 MWd/kg.
3. Pu and neptunium (Np) recycled once in PWR as MOX and burned to 60 MWd/kg, (avoids separation of pure plutonium).

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[15] EPR = European pressurised water reactor marketed by Areva (e.g. Olkiluoto unit 3).

[16] MOX = Mixed OXide fuel.

4. Enriched UO_2 burned once in PWR to 35 MWd/kg, processed (OREOX) and burned once an additional 15 MWd/kg in Candu[17] (Korean Dupic scheme).

Partially closed fuel cycles:

5. Continuous recycle of Pu plus enriched uranium (MOX-UE) in dedicated PWR. UOX and MOX-UE are burned to 60 MWd/kg.
6. Same as the previous but including recycle of americium (Am) in part of the MOX-UE. Curium (Cm) is stored 100 years for decay to Pu and some U.
7. Once burning of enriched UO_2 in PWR to 60 MWd/kg and then continuous recycle of Pu as MOX in fast reactor (FR); MOX burned to 140 MWd/kg in each cycle. Am once recycled as special targets in FR and then disposed. Curium (Cm) is stored 100 years for decay to Pu and some U. FR optimised for MOX burning.
8. Same as previous but Am stored with Cm for 100 years and then recycled with Pu in FR.

Fully closed fuel cycles:

9. Once burning of enriched UO_2 in PWR to 60 MWd/kg and then continuous recycle of all transuranium elements (TRU) as metallic fuel (Zr matrix) in fast reactor (FR); fuel burned to 140 MWd/kg in each cycle. FR optimised for TRU burning.
10. So called double strata fuel cycle. Pu once recycled in PWR as MOX and then further recycled in FR as MOX. Minor actinides from PWR and FR recycled as nitride fuel in ADS. PWR and FR fuel reprocessed by Purex but ADS fuel by pyroprocessing. Burnup 50 MWd/kg in PWR, 185 MWd/kg in FR and 150 MWd/kg in ADS.
11. Variant of double strata cycle without FR. This means that Pu is burned in ADS after once recycle in PWR. Burnup in ADS 150 MWd/kg in this case.
12. FR breeder cycle based on gas-cooled fast reactor, pyroprocessing and continuous recycle of all actinides including U as carbide fuel burned to 100 MWd/kg in each cycle.
13. FR breeder cycle based on sodium-cooled fast reactor (EFR[18]), extended Purex-processing and continuous recycle of all actinides including U as MOX burned to 100 MWd/kg in each cycle.

In the fully closed fuel cycles only the processing losses will generate actinide-containing wastes. Thus a fairly broad range of current and potential future fuel cycles were explored. Fuel cycle were analysed with emphasis on primary and secondary waste generated at each step. The compositions, activities and heat loads of all waste flows were calculated and their impact on the waste repository concepts was assessed. The study also investigated economic aspects based on parametric calculations relying on available cost data and expert judgements. The fuel cycles were analysed under equilibrium conditions. The lead times necessary to reach equilibrium were not considered. These lead times will be rather long and have to be taken into account in a holistic assessment for specific fuel cycle planning.

The indicators used for comparative assessment in the study were:

- Uranium consumption.
- TRU loss/transfer to waste.
- Activity after 1,000 years.
- Decay heat after 50 years.
- Decay heat after 200 years.
- HLW, including spent fuel, volume.
- Maximum dose for repositories in granite.

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[17] Candu = CANadian Deuterium Uranium reactor.

[18] EFR = European Fast Reactor; sodium-cooled fast reactor design worked out by European industries.

- Maximum dose for repositories in clay.
- Maximum dose for repositories in tuff.
- Fuel cycle cost.
- Total electricity generating cost.

Some of the more interesting results were:

- The uranium consumption depends mainly by the fraction of fast reactors. All the cycles based on light water reactors have uranium consumptions differing by less than 15%. Fully closed fuel cycles based on fast reactors reduce uranium consumption by more than two orders of magnitude.
- TRU loss is the indicator which is the most sensitive to the details of the schemes. Multirecycling can decrease the TRU loss to waste by a factor of six. More advanced cycles may give a further reduction of up to two orders of magnitude of this loss.
- The activity of HLW after 1,000 years describes the radioactive source term after decay of the heat generating isotopes. After this time, the short-lived fission products have decayed strongly and the removal of actinides from the waste flux is very efficient in decreasing the source term. The fully-closed fuel cycle schemes reduce by almost two orders of magnitude the activity of HLW after 1,000 years.
- Decay heat is very important for the design of underground repositories. In granite, clay and tuff formations the maximum allowable disposal density is determined by thermal limitations. HLW arising from advanced fuel cycle schemes generates considerably less heat per unit of electricity produced than spent fuel arising from the reference PWR once-through scheme. The lower thermal output of HLW allows a significant reduction in the total length of disposal galleries needed. Separation of caesium and strontium would reduce the required repository size even further. For example, in the case of disposal in a clay formation the gallery length needed in the HLW disposal is reduced by a factor 3.5 through a fully-closed cycle scheme as compared with the reference PWR once-through scheme and by a factor 9 through a scheme including separation of caesium and strontium. In the case of granite the reductions in excavation volume would be similar.
- Extending the cooling time from 50 to 200 years will result in a drastic reduction of the thermal output of HLW from advanced fuel cycle schemes and, consequently, of the repository size needed.
- The volume of spent fuel and/or HLW to dispose of is an important parameter for the total capacity of a given repository site. This volume is reduced significantly by closed fuel cycle schemes as compared with the reference PWR once-through scheme. Further volume reductions, by an order of magnitude, are achievable by progressing towards deeper burning of minor actinides and lesser use of uranium.
- Differences in heat load and waste volume have a major impact on the detailed concept of the repositories. This in turn has technical and economic consequences. For example, a given repository might receive the waste from the production of 5 to 20 times more electricity if the electricity were produced by advanced reactors associated with advanced fuel cycle processes than if it were produced by light water reactors operated once through.
- The calculated doses for the different repository concepts are lower for the recycle schemes than for the once-through cycle. The reason is that iodine-129 is disposed with the fuel in the latter case but dispersed at reprocessing in the recycle cases. Should iodine-129 be captured and disposed of in the HLW repository the doses resulting from the scenarios would be about equal for all cases. In the very long-term, i.e. after a few million years, the total dose is lower in the case of the fully closed fuel cycles, because much smaller amounts of actinides, in particular uranium, have to be disposed.

- The economic analyses provide similar indications as previous studies. Fuel cycle cost is only a small fraction of total nuclear electricity generation cost. Nuclear power is a capital intensive technology and the investment cost – associated with building, refurbishing and eventually dismantling the reactor – exceeds by far the fuel cycle cost. Therefore, the impact of various fuel cycle schemes on the economics of nuclear electricity remains marginal under any set of assumptions. The total electricity generation costs vary by no more than 20% from one scheme to the other and the uncertainties on unit costs and other parameters which increase with the degree of innovation in the fuel cycle scheme are of the same order of magnitude. It is however noted that the uncertainties in cost parameters are large and the uncertainty ranges for the different schemes exceed the differences between the schemes.

An issue dealt with only in the main part of the report is the amount long-lived low and intermediate level wastes created in the different fuel cycles. It is pointed out that the volumes of such waste very much depend on the process for conditioning of the waste. Furthermore very few data exist on the secondary waste flows from real operation or from experiments. This creates large uncertainties. The numbers given in the report indicate however very clearly that the volumes of long-lived low and intermediate level wastes will be substantially larger in all recycle scenarios than in the once through cycle.

2.2 Physics and safety of transmutation systems – NEA study

Another expert group created within OECD/NEA has published a status report on Physics and Safety of Transmutation Systems /NEA 2006b/. The report describes the state of the art concerning the challenges facing implementation of P&T, scenario issues related to accelerator-driven systems (ADS), long-lived fission product transmutation and the impact of nuclear data uncertainty on transmutation system design.

Concerning the role of P&T and ADS the study gives the following main conclusions:

- P&T technologies offer the potential for a significant radioactive waste minimisation.
- P&T can be applied to widely different fuel cycle strategies as:
  - Sustainable development of nuclear energy.
  - Minimisation of waste arising from a legacy of spent nuclear fuel.
- P&T does not eliminate the need for deep geological storage whatever the strategy, but allows increasing its capacity, drastically reducing the burden and potentially improving public perception.
- Fast reactors offer the most flexible tool in order to implement P&T. The use of ADS can be seen as an option or a potential backup solution.
- Demonstration of P&T implies the demonstration of all "building blocks" of the strategy: adapted fuels, adapted reprocessing techniques, reactor behaviour when loaded with significant quantities of MA.

One chapter of the report is devoted to Scenario studies for P&T. Methodology as well as some case studies are described. In a time dependent (phase-out) scenario it seems that the relatively large number of facilities required point to an advantage of an (international) regional approach to ADS-based transmutation in order to achieve optimum use of the resources.

Concerning ling-lived fission products it is pointed out that the dominating long-term contributors to dose (risk) from a deep repository are often fission products as ^{129}I, ^{135}Cs, ^{99}Tc, ^{126}Sn and/or ^{79}Se. These are generally more mobile than the long-lived actinides and have relatively high radiological toxicity. The nuclides ^{129}I and ^{99}Tc can relatively easy be transmuted in nuclear reactors considering their neutron capture cross sections. Other fission products are judged to be difficult to transmute due to small cross sections and in case of ^{135}Cs also to the need for separation from other stable cesium isotopes. For ^{99}Tc it is generally judged feasible to use metallic Tc as a target material for transmutation. For ^{129}I there is however no consensus concerning a suitable target material. Candidate compounds for iodine targets that are studied are i.a. CaI_2, CuI, MgI_2, NaI and other. Experimental and theoretical studies of transmutation of ^{129}I and ^{99}Tc are reviewed.

2.3 Implications of P&T in radioactive waste management – IAEA study

IAEA has prepared a technical report discussing the implications of partitioning and transmutation on nuclear waste management /IAEA 2004/. The report concentrates on the radioactive waste aspects of P&T. The main purpose was to gather useful technical information on the expected long term consequences on waste management from present and potential future fuel cycle options. In the introduction it is pointed out that continued world-wide use of nuclear energy at the present or at an expanded level will produce increasing waste streams that will need to be disposed of. In the event of the use of the once through fuel cycle, repositories of the size of the one planned at Yucca Mountain will need to be constructed every seven to ten year world-wide.

The report contains chapters concerning:
- Potential impact of P&T on radioactive waste management.
- Non-proliferation aspects of P&T.
- Fuel cycles.
- Partitioning.
- Transmutation.
- Additional waste management considerations.
- Conclusions.
- Recommendations to decision makers.

Among the conclusions and recommendations the following points are noted:

- Full implementation of P&T can give a significant reduction of the inventory of trans-uranium to be disposed of in geologic repositories. Reduction factors of 100–200 of TRU inventory and radiotoxicity seems possible. The time scale for reaching the reference level of radiotoxicity (natural uranium in equilibrium with its daughter nuclides) might be reduced from about 100,000 years to 1,000–5,000 years.

- It is not likely that all nuclear states would have their own reprocessing and partitioning facilities to perform P&T. Similarly, disposal centres for specific waste types (e.g. hot targets, iodine) would probably not be available in all states. Some coordination between national responsibilities for such nuclear materials and wastes may be advantageous.

- P&T is at the R&D stage. Expected time scales for necessary development could be of the order:
  - Aqueous partitioning 5–10 years.
  - Pyrochemical partioning 10–15 years.
  - Special fuel and target fabrication 10–15 years.
  - Fast reactor reintroduction 20–25 years.
  - Development of accelerator-driven systems 25–40 years.

- Partitioning followed by conditioning of the minor actinides to fuel irradiation targets for future transmutation or to an inert matrix for long term storage or disposal is sometimes discussed as an interim strategy (P&C-strategy). Expected time for start of such an approach is of the order 5–10 years if using aqueous methods for partitioning and 10–15 years with pyrochemical techniques.

- The potential proliferation risks must be properly addressed at an early stage in any P&T or P&C programme. International organisations could play an important role in developing proliferation resistant principles and guidelines as well as in monitoring transfers and inventories of minor actinides and in planning and assessment of proper technical systems for surveillance.
- The study calls for more integration of the efforts of the P&T and waste management communities to reach a better common understanding of the long term issues involved.
- Recycling of plutonium in LWR will reduce the separated plutonium stocks and partially use its fissile content. Full recycling of plutonium is only possible when fast reactors become operational on an industrial scale.
- Transmutation of the minor actinides cannot yet be envisaged on an industrial scale. But it is prudent to continue research on this line in order to keep the possibility of future development of dedicated transmutation reactors open. This might open horizons for the role of transmutation in waste management.

2.4 EU-studies of transmutation

The European commission supports a number of studies concerning P&T under its so-called shared cost programmes. The major projects supported within the sixth framework programme are listed in Table 2-1. The costs and funding from EC (for the sixth FP time frame) are given in the the table. The projects are briefly presented in the ensuing text together with some results from projects under previous EU-programmes.

2.4.1 XADS and Eurotrans

In the fifth framework program, pre-conceptual studies of three different accelerator driven systems were performed within the PDS-XADS project (Preliminary Design Study of eXperimental Accelerator Driven Systems). This programme was coordinated by Framatome, having the main purpose of developing facilities where the principle of testing the coupling of an accelerator with a sub-critical core at power could be made. The fuel was considered to be standard fast reactor MOX, with the addition of a few experimental minor actinide assemblies. The three XADS:s studied may be characterised as

1. An 80 MWth LBE cooled core with low linear rating (11 kW/m), originating from a proposal by Ansaldo. A major design feature was argon gas bubbles to be introduced above the core in order to enhance natural circulation.
2. An 80 MWth He cooled core with medium linear rating (25 kW/m), originally proposed by Framatome.
3. A 50 MWth LBE cooled core with high linear rating (50 kW/m), originating from SCK-CENs Myrrha design. This facility would also have the purpose of serving as an irradiation facility for materials testing, including thermal spectrum islands.

Table 2-1. Major P&T projects in the sixth framework programme of EU /van Goethem et al. 2006/.

Project name	Purpose	Total cost M€	EC funding M€	Time schedule
Europart	Partitioning MA	10.3	6.0	2004–2007
Eurotrans	Design + R&D for ADS	43	23	2005–2007
Red-Impact	Impact of R&D on waste management	3.5	2	2004–2007
Actinet	Network of excellence for actinide research	10.5	6.3	2004–2007

The PDS-XADS project was completed in 2005 /Ansaldo Nucleare 2004, NNC 2004/, and the major conclusions may be summarised as follows:

- The low rated design of Ansaldo featured the best safety characteristics, permitting heat removal by natural circulation during various accident scenarios. The low neutron flux would however result in poor performance in terms of minor actinide burning rates.
- The He cooled core might encounter problems with decay heat removal in case of depressurisation. The presence of a spallation target makes these problems more complicated to manage than in the case of a critical fast gas cooled reactor.
- The Myrrha design would meet difficulties in surviving certain types of accidents, due to the relatively low fraction of natural circulation in combination with a very high linear rating.

In total, the results were encouraging enough for the European research community to launch a larger effort in the sixth framework program, working towards the demonstration of transmutation in Europe by carrying out:

1. An advanced design of a 50–100 MW experimental facility demonstrating the feasibility of transmutation in an accelerator driven system (XT-ADS), being possible to realise in a short-term (~10 years).
2. A conceptual design of a several hundred MWth European Facility for Industrial Transmutation of minor actinides (EFIT), to be realised in a longer term.

The Eurotrans project started in April 2005 with a total budget of 43 M€, out of which 23 M€ are funded by the EC. The project is subdivided into five sub-domains addressing /Knebel et al. 2006/:

1. Design.
2. Coupling of accelerators with sub-critical cores.
3. Minor actinide fuel development.
4. Structural materials behaviour under LBE-cooling and fast neutron irradiation.
5. Basic nuclear data.

Design

Experiences from the PDS-XADS project lead to the choice of a lead-bismuth cooled core with medium linear rating (15 kW/m) as a reference for the XT-ADS design. Further, it was decided that the system should be designed for localisation at SCK-CEN in Mol, Belgium. A review of several design options was made in the early stage of Eurotrans. The following design features were established in early 2007 /Areva 2006, SCK-CEN 2007/:

Table 2-2. Major design features of XT-ADS (March 2007).

Core power	57 MWth
Coolant	Lead-bismuth eutectic
Coolant inlet/outlet temperature	300°C / 450°C
Fuel	MOX (30% Pu)
Fuel column height	60 cm
Average (peak) linear rating	15 (20) kW/m
Average burnup	80 MWd/kg
Cladding	T91 ferritic martensitic steel
Accelerator	Linear
Proton beam energy	600 MeV
Proton beam power	2.0 MW
Spallation target	“windowless”
k-effective@BOL	0.95
In-pile experimental positions	8

The so called “windowless” design in practise means that a free surface of liquid LBE is separated from the accelerator tube end piece by an open tube containing vacuum. Thus, the accelerator tube window may be kept separated from high neutron fluxes at cold conditions, which is expected to significantly prolong its service time.

Detailed analysis of the safety performance, including design of the decay heat removal system is under way. A particular safety issue that has been pointed out is the consequences of a steam generator tube rupture. This kind of accident would cause the intrusion of steam bubbles into the primary circuit, with a potential for thermo-mechanical interactions that might be harmful for the integrity of the primary vessel and/or core structures. A comprehensive research programme to investigate the potential consequences of a steam generator tube rupture therefore is of high priority.

For the design of EFIT, it was decided that the fuel composition should satisfy the dual requirement of a high minor actinide burning rate, while keeping the effective plutonium burning rate at zero. Here one should remember that transmutation of americium partially leads to the production of plutonium, following the decay of ^{242}Am in its ground state (with a half life of 10 hours) into ^{238}Pu. Hence plutonium still contributes to the fission energy production. The plutonium in the spent ADS fuel will be of low quality, containing a high fraction of ^{238}Pu, which preferably is recycled in dedicated ADSs. In order for the first core load to resemble the equilibrium fuel as much as possible, it was suggested to use a transuranium composition deriving from spent LWR fuel (mixture of UOX and MOX) with a cooling time of 30 years. Such a fuel is essentially free of ^{241}Pu, and would thus suffer less from reactivity losses due to ^{241}Pu fission. Preliminary investigations made by ENEA for an oxide fuel dispersed in magnesia showed that with such a TRU composition, a Pu/MA ratio of 46/54 results in a zero net burning rate of plutonium combined with a destruction rate of minor actinides equal to 42 kg/TWh_{th} /Sarotto and Artioli 2006/. The potential for consumption of minor actinides from spent LWR fuel would hence be maximised. An additional advantage is that the reactivity swing is reduced to a minimum very close to zero.

The criteria for choosing the fuel form and composition for EFIT were elaborated within the 5th framework programme Future, as well as in the initial stages of the Aftra domain of Eurotrans /Wallenius 2006/. Taking into account a number of boundary conditions ranging from reprocessability to safety performance (see below), it was recommended to select two composite oxide fuels as the final candidates for the design of EFIT /Wallenius et al. 2006/.

1. $(Pu,MA)O_{2-x}$ – ^{92}Mo
2. $(Pu,MA)O_{2-x}$ – MgO

Where the molybdenum based CerMet (Ceramic-Metallic) composite fuel was judged to provide superior safety characteristics. The cost associated with enrichment of molybdenum however lead the participants in the Design domain to select the magnesium oxide based fuel as the primary candidate. In the spring 2007, ENEA suggested a three zone design of a 400 MWth core, where the combination of a varying inert matrix fraction and fuel pellet diameter resulted in a flat power profile. The major characteristics of this core are given in Table 2-3. It may be noted that the source efficiency (a measure of how effective neutrons created in the spallation target are to induce fission, as compared to the average fission neutron) is low for the suggested design. This is mainly a consequence of the large spallation target diameter, which leads to moderation and leakage of source neutrons. A smaller target size would improve the utilisation of the source neutrons. Its lower heat removal capability would therefore automatically be compensated for by a smaller demand for proton beam power, with a corresponding gain in cost for operation of the accelerator.

An attractive property of the design suggested by ENEA is the very small reactivity swing, permitting an almost unchanged proton beam power over the burnup cycle. The burnup rate is however comparatively low, which means that a residence time of more than seven years for the fuel would be required to reach an average burnup aim of 20% set by dose limitations for the clad. Present techniques for corrosion protection of the clad surface though are hardly validated for seven years of operation, hence the more modest burnup aim of 8% is suggested.

Table 2-3. Major design features of EFIT with MgO based fuel (March 2007).

Core power	400 MWth
Coolant	Lead
Coolant inlet/outlet temperature	400°C / 480°C
Fuel	$(Pu_{0.46},MA_{0.54})O_{1.9}$ – MgO
Matrix fractions	57/50/50
Fuel pellet diameter	7.1/7.1/8.0 mm
Fuel column height	90 cm
Average (peak) linear rating	15 (20) kW/m
Average burnup	78 MWd/kg
Cladding	T91 ferritic martensitic steel
Accelerator	Linear
Proton beam energy	800 MeV
Proton beam power	10 MW
Neutron source efficiency	0.6
Spallation target	“windowless”
k-effective@BOL	0.97
Reactivity swing	< 200 pcm per year

Ecats

In the Ecats domain of Eurotrans, experiments aiming at testing the coupling of an accelerator with a sub-critical core were planned, to proceed on the basis of the Muse experiments in the fifth Framework Programme. Initial plans of installing a tantalum target for a 140 MeV cyclotron proton beam in the Triga reactor of ENEA in Rome however were cancelled due to lack of national Italian funding for the project. Instead, experiments with d-t neutron generator sources will be conducted at the Yalina facility in Minsk. Further, the construction of a neutron generator driven zero sub-critical facility with simulated lead fuel is planned to be made in the Venus facility at Mol in Belgium, using enriched UOX fuel to be provided by CEA /ECATS 2007/.

Aftra

In the Aftra (Advanced Fuels for TRAnsmutation) domain of Eurotrans, methods for fabrication of minor actinide based fuels have been developed. When handling macroscopic quantities of highly active minor actinides like ^{241}Am, it is of importance to avoid the formation of dust, which means that traditional powder pressing methods are not suitable.

ITU in Karlsruhe has developed the so called “infiltration” technique, where porous beads are infiltrated with a solution of americium nitrate /Fernandez et al. 2003/. The beads must be insoluble in nitric media, thus limiting the selection to ceramic compounds like zirconium and/or plutonium oxide. The upper bound to the relative fraction of americium in the end product is given by the initial porosity, but in practice it has shown to be possible to achieve up to 30% volume fraction of americium by repetition of the infiltration process. The hence produced $(Pu,Am)O_{2-x}$ or $(Pu,Am,Zr)O_{2-x}$ beads may then be mixed with a metallic powder and pressed into composite “cermet” pellets which are sintered according to standard procedures. For the Futurix irradiation in Phenix[19], two cermet fuel pins were fabricated with the following composition /Donnet et al. 2005/:

- $(Pu_{0.80},Am_{0.20})O_{2-x}$ – Mo (86 vol% Mo)
- $(Pu_{0.23},Am_{0.25},Zr_{0.52})O_{2-x}$ – Mo (60 vol% Mo)

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[19] Phenix = French sodium-cooled fast reactor with 250 MWe power.

"x" denotes the degree of sub-stoichiometry resulting from release of oxygen during the sintering process. The final value of O/M depends on the ratio of Am to Pu in the sample. The final density of the material is 90–95% of the theoretical value, with a near 100% density achieved for the metallic matrix.

In the Atalante laboratory of CEA[20] in Marcoule, solid solutions of $(Pu,Am)O_{2-x}$ are fabricated by oxalic co-conversion of nitrates /Donnet et al. 2005, Pillon and Wallenius 2006/. With this method, arbitrary concentrations of americium are manageable. After mixing with magnesium oxide, pressing and sintering, composite "cercer" pellets with a density of 92–95% of the theoretical value are obtained. The phase structure of the actinide oxide was found to be a complex function of americium content. For equal fractions of Pu and Am, both a bcc phase with an O/M ratio of 1.93, and an fcc phase with O/M = 1.66 were found by X-ray diffraction analysis. The same analysis of $(Pu_{0.20},Am_{0.80})O_{2-x}$ – MgO pellets however unexpectedly revealed a monoclinic phase for the actinide oxide. For the Futurix experiment, two cercer fuel pins were fabricated with the following specifications:

- $(Pu_{0.50},Am_{0.50})O_{2-x}$ – MgO (80 vol% MgO)
- $(Pu_{0.20},Am_{0.80})O_{2-x}$ – MgO (75 vol% MgO)

Thermo-physical properties were measured for the composite oxides in 2006. The very high thermal conductivity of the molybdenum based cermet fuels (k > 50 W/(m·K)) could be confirmed, while the conductivity of the cercer fuel was found to be slightly lower than expected at low temperature /Staicu et al. 2006/. A major issue related to the high temperature stability of MgO was encountered during these measurements, when samples of $(Pu,Am)O_2$-MgO vaporised as temperatures exceeded 2,130°K. Even though this process would not occur in a closed system like a fuel pin, it may have consequences for the safety performance of EFIT, in case of clad failure. Vapour pressure measurements made on $(Pu_{0.8},Am_{0.2})O_{2-x}$ – Mo on the other hand, indicated that eutectic melting of the compound occurred at a temperature around 2,400°K.

In May 2007, irradiation of the four composite oxide fuel pins will start in Phénix, side by side with metallic alloy fuel pins fabricated by Idaho National Laboratory in the US. The irradiation will continue until the end of life of Phénix. The shut down is foreseen in 2009, mainly due to shortage of fast reactor MOX fuel supply.

In the fifth framework programme project Confirm, being coordinated by KTH, inert matrix nitride fuels were fabricated for irradiation at Studsvik. As the R2 reactor at Studsvik was shut down this experiment had to be moved to the HFR reactor at Petten, Holland. The (Pu,Zr)N pins manufactured by PSI[21] were shipped from Studsvik to Petten in March 2007, and irradiation of two segments will start during summer. Destructive post-irradiation examination is to be made by PSI as part of the Aftra programme.

Demetra

In the Demetra domain of Eurotrans, the combined effect of irradiation and heavy liquid metal corrosion on structural materials (fuel clad, primary vessel, accelerator beam window) is investigated. In the 5th FP Tecla project, it was demonstrated that by careful control of the oxygen content in the liquid metal, at a level of 1 ppm (weight), that a protective oxide layer may be maintained on the surface of ferritic-martensitic steels, preventing dissolution of the steel up to 7,200 hours at temperatures of 550°C. The thickness of the oxide shell depends significantly on the composition of the steel.

At longer exposure times or at higher temperatures (which may arise during accident scenarios), the dissolution kinetics may however be rapid, and additional protection measures needs to be applied. For this purpose, a surface alloying technique, based on spraying of aluminium vapour

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[20] CEA = Commisariat à l'energie atomique – French national nuclear research organisation.

[21] PSI = Paul Scherer institute in Switzerland.

on the surface of the clad, followed by pulsed electron beam treatment (GESA) is developed at FZK[22]. When immersed into LBE, a thin but protective layer of aluminium oxide forms on the surface, in perfect bonding with an Fe(Cr,Al) phase connecting the surface with the underlying steel. GESA treated austenitic steels tested in LBE do not exhibit any dissolution attack even up at 600°C (for an exposure time of 4,000 hours and a flow rate of 1.3 m/s) /Müller et al. 2004/.

At lower temperatures the successful operation of LBE loops for more than 27,000 hours in Europe have demonstrated that the oxygen control technique originally developed in Russia is suitable for long term applications. Most of the long term corrosion tests were however performed at flow rates of about 1 m/s, where effects of erosion are small. Corrosion tests of ferritic martensitic steels in LBE flowing with a velocity of 2 m/s at temperature of 550°C are now under way in the Corrida loop at FZK in Karlsruhe.

The oxygen control technique was developed for Soviet sub-marine reactors with an intermediate (partially moderated) neutron spectrum, and was only recently (successfully) tested in a fast spectrum lead loop experiment in BOR-60 /Efimov et al. 1998/.

An important activity in Demetra is to study the effect of irradiation on the GESA treated steels. A large number of specimens are under fast neutron irradiation in Phenix since April 2006. In Phenix, the insertion of an LBE loop was however not possible. Therefore, experiments where GESA treated steels are irradiated in LBE environment have been designed for the materials test reactors BR2 (Mol, Belgium) and HFR (Petten, the Netherlands). The ASTIR irradiation in BR2 is made at a typical operation temperature of XT-ADS (350°C) started in January 2006. In a second round, the temperature will be raised to 450°C.

Modelling of corrosion and irradiation phenomena are essential for gaining understanding of the physical processes leading to deterioration of cladding and other structural material. In CEA, a model for the corrosion kinetics in oxygen saturated LBE has been developed, which works well up to temperatures of 630°C but not yet above /Martinelli 2005/.

In a combined Swedish-Belgian effort, multi-scale modelling of irradiation induced defect production in Fe-Cr alloys has advanced significantly. Electronic structure calculations by /Olsson et al. 2003, 2006/ lead to the discovery of the negative mixing energy of the alloy for small Cr concentrations. Further calculations of formation energies for interstitial defects in the vicinity of Cr atoms revealed that Cr tends to bind to interstitials, which reduces their mobility. Diffusion coefficients for interstitial clusters calculated with a novel embedded atom method model of the alloy showed that the mobility of such clusters in the alloy is reduced by two orders of magnitude as compared with pure iron, correlating well with the experimentally measured swelling rates as function of chromium content. The result is a much improved understanding of basic mechanisms responsible for swelling (or rather the absence of swelling) in ferritic steels /Olsson et al. 2006, 2007, Terentyev 2006/.

2.4.2 Megapie

The successful completion of the Megapie experiment at PSI is a major milestone towards the implementation of a prototypical accelerator driven system /Latgé 2006/. The liquid lead-bismuth spallation target installed at PSI was developed in collaboration between nine research institutes in Europe, Japan, Korea and the USA. After six years of research, development and testing, it was put into operation on top of the proton beam of the PSI cyclotron in August 2006. The 800 kW proton beam deposited 580 kW heat in the target during its four months of operation, while producing spallation neutrons at a rate twice as high as achieved with the previous solid target used by PSI. Not only did the experiment show that a megawatt rated liquid metal spallation target is feasible to operate with a very high reliability (the target delivered neutrons 95% of the scheduled time), it also proved that the production of polonium is not a show-stopper for licensing of such a device.

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[22] FZK = Forschungs zentrum Karlsruhe in Gemany.

Figure 2-1. *The Megapie target prior to installation in the SINQ facility of PSI (from PSI press release).*

2.4.3 RedImpact

Over the last three years, a comprehensive study of the consequences of different back-end fuel cycle strategies have been undertaken within the frame of the European Commission's 6th framework programme, known as RedImpact /Gudowski et al. 2006b/. The main goal has been to examine the impact of partitioning and transmutation on waste management and geological disposal as well as to assess the economical, social and environmental aspects of different fuel cycle scenarios.

Six main scenarios, representing possible future European fuel cycles have been studied. In most cases both equilibrium and transition situations have been addressed. The first three scenarios should be considered as near at hand scenarios, utilising only existing, or near commercialisation technology. The remaining three are more complex scenarios, involving partitioning and transmutation:

- A1: Reference once through scenario.
- A2: Monorecycling of plutonium in PWRs.
- A3: Multirecycling of plutonium in fast reactors.
- B1: Fast neutron Gen IV scenario.
- B2: Simplified Double strata with PWRs and ADSs.
- B3: Full Double strata with PWRs, FRs and ADSs.

The detailed goals of the project are related to the following items:

- Overview and analysis of different **waste management strategies** including the type of geological disposal, irretrievability vs. retrievability, long-term vs. intermediate storage etc as well as identification of relevant operations.
- Elaboration of **industrial deployment scenarios** to identify the near to medium term technical options for partitioning and transmutation, their current technical applicability, the necessary research and development, and realistic schedules for deployment.
- **Performance indicators** for the different steps of the fuel cycles affected by partitioning and transmutation, e.g. radioactive releases from fuel cycles, dose to workers, long-term radiological impacts, and costs.

- **Quantification of waste streams** and waste reduction potentials based on simplified core calculations, elaboration of characteristics for proliferation, radioactivity, radiotoxicity, heat generation etc.
- **Impact on waste management** including geochemical behaviour, retention capabilities and radiotoxic potentials as well as requirements on waste inventories and the process of waste disposal under different strategies and implications on repository layout, repository operation and post-closure safety.
- **Fuel cycle modelling and (simplified) life cycle assessment** and consistent analysis of important operations in the fields of:
  - fuel-related aspects, such as ore demands and diverse fuel options,
  - reactor/transmuter operation of alternate/symbiotic systems,
  - waste management and partitioning,
  - waste features and masses for storage and disposal.
- **Synthesis and sustainability criteria** addressing sustainability issues and externalities in a quantified way by using Multi Criteria Analysis techniques (MCA).

The project has 23 partners from eleven countries. This has made available a unique collection of expertise on the wide range of technologies represented both within the existing set of European nuclear installations and among the proposed waste management strategies. Several on site visits have been performed to back-end fuel cycle facilities throughout Europe. Halfway through the project, a workshop on plutonium management was held at Cambridge University aimed at providing an interface between the project participants and society.

RedImpact has now come to an end. The final analysis is complete and a number of conclusions have been drawn. The main results of the project correspond to the goals described above, including radiotoxicity reduction calculations, heat generation and subsequent repository requirements for different waste forms, as well as the MCA of the identified indicators used to adequately describe the scenarios in a general context.

2.4.4 Other transmutation studies within EU

Generation IV fast neutron reactors may be used for transmutation of minor actinides in homogeneous (diluted in driver fuel) or heterogeneous (dedicated target assemblies at core periphery) modes. In the ELSY (European Lead cooled SYstem) project, a design of critical reactor with lead coolant is undertaken, that would permit transmutation of minor actinides from the LWR spent fuel inventory. The reference core is based on oxide fuel, while nitride fuels may allow for better performance, and are thus studied as an alternative solution.

In another EU-project called PUMA, the feasibility of burning minor actinides in high temperature reactors is studied. Major obstacles like reprocessability of Triso[23] fuel and fabrication of fuel containing significant fractions of californium would have to be overcome to make this concept workable.

2.4.5 Nuclear data

During the latest decade, the leading research on nuclear data for ADS has been undertaken in Europe including Russia. There is a striking imbalance around 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.

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[23] Triso = Tri-ISOtropic; multilayerfuel particle with coating of pyrolytic carbon and silicon carbide.

In the fifth EU framework program (FP5), a large fraction of the P&T research was devoted to nuclear data. Two large projects, Hindas and NTOF were financed by the EC /Hindas, NTOF/. Hindas, in which the Swedish participation was prominent, was a network of many facilities in Europe to remedy the lack of cross section data at energies above 20 MeV, i.e. above the energy range covered by traditional critical reactors. Hindas resulted in a wealth of new nuclear data and advanced the frontier significantly.

NTOF was primarily focused on the low-energy range. It was dominated by activities at CERN[24], Geneva, Switzerland, where a spallation neutron source was developed. The agenda comprised 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 was extended. At present, the facility is operational with parameters close to the specifications, and the first results have been presented.

In FP6, a notable shift in focus on European nuclear data research for ADS took place. With the successful completion of Hindas, it was concluded that the nuclear data requirements had to a large degree been fulfilled. Therefore the Eurotrans project was focused on other problem areas. Still, however, a work package on nuclear data was included, but at a lower ambition level than in FP5 /Knebel et al. 2006/.

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[25] in Uppsala.

2.5 EU-studies of partitioning

2.5.1 Partnew

The Partnew (New solvent extraction processes for minor actinides) project was carried out by 11 partners, coordinated by C. Madic (CEA) and co-funded by EC under the fifth Framework programme. Partnew was in principle a continuation and extension of an earlier European research programme with the acronym Newpart, which had produced some remarkable results. The project started 1st of November 2000 and lasted 36 months. During that time 189 different molecules were prepared and tested as minor actinide extractants and as reagents for An(III)/Ln(III) separations. More than 100 scientific papers were published in journals and proceedings. A rather comprehensive final technical report was also issued in June 2004 and is available from the EU /Madic et al. 2004/. Hence only a strongly abbreviated summary of some of the important results is given here.

Diamex[26] basic and process development studies

Several bis-malonamides were prepared and tested. Some of these showed interesting extraction properties. Their basic chemistry was further investigated. In addition several flow-sheets were developed and some of these were tested with a highly active concentrate. The successful operation of some of these was also shown in centrifugal contactor batteries but also using hollow-fibre modules.

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[24] CERN = Centre Europeen pour la recherché nucleaire.

[25] TSL = The Svedberg laboratory.

[26] Diamex = Diamide-based solvent extraction process developed and tested during Newpart.

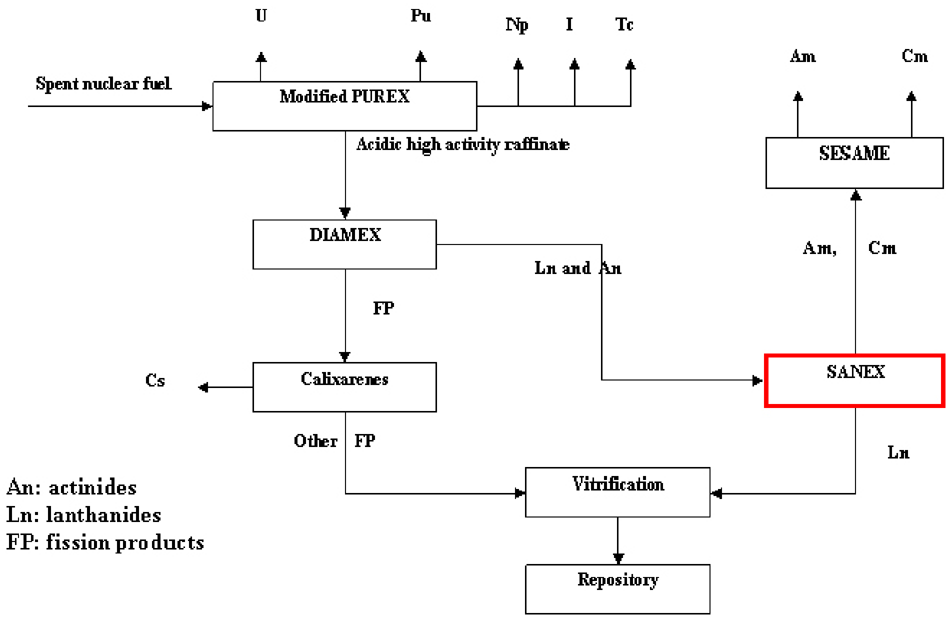

Figure 2-2. Main partitioning steps in spent nuclear fuel treatment.

Sanex[27] basic and process studies with polydentate N-bearing ligands

The new reagents prepared included both semi-BTPs[28] and new BATPs[29]. Numerous studies of their solvent extraction thermodynamics and mechanisms were made and supplemented by molecular modelling. BTP-based Sanex processes were tested in centrifugal contactor batteries using synthetic radioisotope-spiked highly active feed. However, the chemical stability of the tested molecules was not sufficient for their use in an industrial process.

Sanex studies with bis-(substituted phenyl)-di-thiophosphinic acid + neutral ligand synergistic mixtures

Unexpectedly high separation factors between An(III) and Ln(III) were observed in some of the tested systems. This was probably due to the entropy term in the thermodynamics of the extraction reaction. Also in this case, process flow-sheets were developed and successfully tested in centrifugal contactor batteries.

Sanex basic studies with new S-bearing ligands

Numerous molecules were prepared and tested. However, the only S-bearing ligands useful for An(III)/Ln(III) separations were found in a new group of bis-substituted-dithiophosphinic acids.

Am(III)/Cm(III) separation: basic and process development studies

Only two new systems were found to be promising, one based on a single malonamide, and one based on a synergic mixture containing a chlorinated di-phosphinic acid. Two separation flow-sheets were developed and tested using these two systems. Interesting results were obtained in both cases.

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[27] Sanex = Selective ActiNide Extraction; process in which actinides are separated from lanthanides.

[28] BTP = Molecule of the type *bis*-triazine-pyridine; semi-BTP refers to molecules corresponding to one half of a BTP-molecule.

[29] BATP = Type of hydrophobic tridentate nitrogen heterocyclic reagents. They are similar to the BTP’s but have additional annular rings (A= annular).

2.5.2 Europart

The sixth European framework programme Europart started in 2004 and was scheduled to finish at the end of 2006 /van Goethem et al. 2006, Madic et al. 2006/. However, due to the fact that all the work had not yet been performed it was extended to June 2007. This extension was, however, without extra financial support from the EC. The scope of the Europart project comprises the continuation of three different fifth framework projects, i.e. Partnew, Calixpart /Arnaud-Neu et al. 2004/ and Pyrorep /PYRO 2003/. As seen in the previous section Partnew dealt with separation of actinides and lanthanides from aqueous solution using solvent extraction and various nitrogen donor ligands or e.g. malonamides. Calixpart also utilised solvent extraction but in this case the ligands were of the calixarene type. In the case of Pyrorep the separation was made by molten salts or molten metal techniques, in some cases used as solvent extraction processes.

Europart is divided into 9 work packages (WP) which in turn are divided into a number of tasks, see below. There are 5 WPs in the domain of hydrometallurgy and 4 in the domain of pyrometallurgy. A total of 24 partners from 11 countries are involved in the research. Two non-EU countries Australia and Japan has joined the project during the elapsing years. The planned work was divided according to:

- WP1 aims are to define efficient systems for the extraction of trivalent actinides (Am to Cf) from HAW (high active waste) with new extractants of the families of polyamides, polydentate N-bearing ligands, S-bearing ligands and COSAN[30]s.
- WP2 has the same objectives as WP1, but the extractants to be studied belong to the domain of calixarenes, multi-podants, COSANs, etc.
- WP3 concerns the development of extraction processes with extractants of the same family as those of WP1 but devoted to the treatments of fuels or targets of future nuclear systems.
- WP4 has the same objectives as WP3, but the extractants to be developed are of the same families as those of WP2.
- WP5 is devoted to the conversion of the separated actinides into solid compounds (oxides, carbides, nitrides) to prepare targets or fuels.
- WP6 concerns the determination of basic chemical properties of actinides (An) and Fission Products (FP) in the form of chlorides and fluorides fused salts, and in metallic fused solvents.
- WP7 aims are to define An partitioning processes.
- WP8 is related to the definition of methods for the conditioning of spent salts.
- WP9 aims are to define system studies for the possible future industrial development of An-partitioning.

The main results and achievement of the Europart project so far are:

WP1 and WP3: Molecular modelling of complexation and extraction was done with several systems:

- $Eu(NO_3)_3L_2$ extracted complex with L= DMDOHEMA[31], the malonamide of the Diamex process.
- Complex of Lu(III) with polyaza-polyaromatic ligands.

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[30] COSAN = CObalt SANdwich.

[31] DMDOHEMA = N,N´-Di Methyl-N,N´-DiO ctyl-2-(2-HExyloxy-ethyl) MAlonamide.

Numerous extractants have been synthesised, e.g.

- TODGA[32].
- COSAN-CMPO[33]s.
- Bis-malonamides.
- Bis-diglycolamides.
- New BTBP.
- New BDP[34] nitrogen ligands.

Moreover, solid extractants were prepared, including TODGA-PAN[35].

For the co-extraction of An(III) + Ln(III) several extractant systems were studied:

- Bisdiglycolamides.
- TODGA + TBP[36] mixture.
- New COSAN-CMPOs.

For the An(III)/Ln(III) separation, studies were carried out with:

- Hemi-BTPs.
- $CyMe_4$-BTBP.
- Mixture of COSAN + CMPO + C5-BTBP.
- Mixture of $CyMe_4$-BTBP + DMDOHEMA.

For the co-extraction of An (at different oxidation states), solid TODGA-PAN and malonamide-PAN were studied. The structures of some complexes were studied, including:

- COSAN complexes.
- $M(nPr\text{-}BTP)_3$ complexes, with M = Am, Cm and Eu, ii) BTBP complexes with several Ln(III) ions.

The supramolecular organisation of solvents containing malonamide and extracted solutes was studied using SAXS and NMR techniques.

The study of the stability of extractants *vs.* hydrolysis and radiolysis was carried out with the following systems:

- DMDOHEMA.
- TODGA.
- Bis-diglycolamide.
- Representatives of the BTBP family.

Cold tests were performed:

- For the Diamex process using a hollow fibre module (HFM) pilot plant.
- With chromatographic separation on a column loaded with TODGA-PAN solid extractant.

Active tests have been carried out:

- For U/Pu splitting using the Purex process but with AHA complexant for the stripping of Pu from the loaded solvent.
- For the Purex treatment of a spent fuel, in order to generate HAW to be used later.

The compound $CyMe_4$-BTBP was found suitable for design of an An(III)/Ln(III) partitioning process. The TODGA + TBP mixture had been selected for the co-extraction of actinides An (III, IV, VI).

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[32] TODGA = TetraOctyl D iGlycolAmide.

[33] CMPO = C arbaMoylmethyl Phosphine Oxide.

[34] BDP = Butyl isoDecyl Phtalate.

[35] PAN = 1-(2-PyridylAzo)-2-Naphthol.

[36] TBP = Tri Butyl Phosphate.

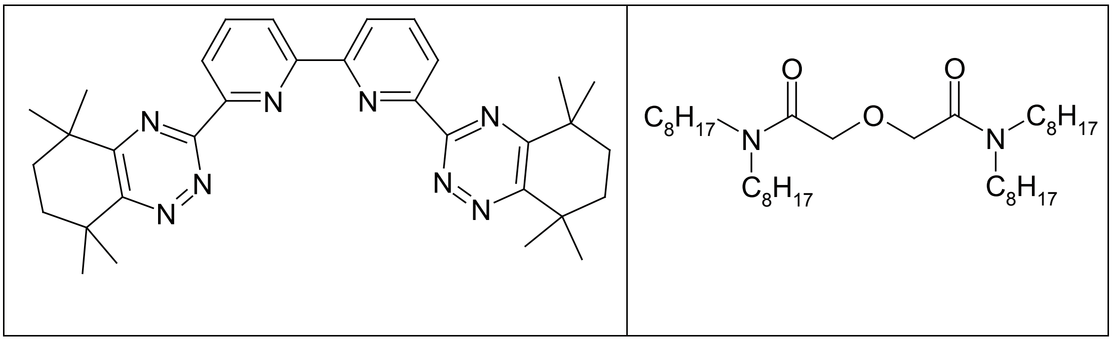

Figure 2-3. *Schematic picture of CyMe$_4$-BTBP (left) and TODGA (right).*

WP2 and WP4: Molecular dynamic behaviour of dicarbollide complexes of U(VI) and Eu(III) was studied in a mixture of water and chloroform. It was shown a great affinity of these complexes for the interface between the two liquids. Numerous new extractants were synthesised including :

- COSAN-CMPO.
- CMPO-calix[4]arenes.
- CMPO-calix[6-,8]arenes.
- COSAN-CMPO-calix[4]arenes.
- Tripodal ligands.
- Diglycolamide-calix[4]arenes.
- Picolinamide-N-oxide-calix[4]arenes.
- Cyclotriveratrylenes.

Complexation and extraction properties of the new ligands were studied including:

- Tripodal CMP and CMPO compounds.
- COSAN-CMPO-calixarene.
- CMPO tripodal ligand in synergistic mixture with C5-BTBP.
- CMPO-calix[4]arene.
- CMPO-calix[6]arene + COSAN synergistic mixture.

The radiolytic stability of the ligand picolinamide-calix[6]arene was studied by RPE[37].
The scaling up of the synthesis of ligands was done for:

- Bis-COSAN-bis-CMPO*t*Bucalix[4]arene.
- CMPO-calix[4]arene.
- Picolinamide-calix[6]arene.

Some new ligands have been selected for further developments:

- Upper rim CMPO-calix[4]arenes for the co-extraction of An(III)+Ln(III).
- Calix[6]arene picolinamide for the separation An(III)/Ln(III).
- COSAN-CMPO compound.

WP5: Several methods were used for the preparation of mixed An or surrogate oxides, including:

- Sol-gel.
- Precipitation.

Several systems were studied, mostly based on surrogates, including:

- Zr(IV)/Ce(III).
- Zr(IV)/Y(III)/Ce(III).

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[37] RPE = Resonance Paramagnetic Electronic.

- Zr(IV)/Y(III)/Nd(III).
- Th(IV)/Ce(IV).

The precipitates formed were characterised, and then calcined to obtain the oxides. The oxide beads or powders were then used to prepare pellets by pressing and sintering. Then, the formed pellets were characterised. Moreover, for the preparation of carbides instead of oxides, the sol-gel process was used and in the broth containing the salts to be transformed in hydroxides, black carbon was added and the beads formed incorporated the carbon. Thus, in the future, the treatment of these beads containing black carbon will generate metallic carbides.

WP6: The determination of activity coefficients of Pu, Gd and Ce in metallic Ga was performed successfully by electrochemical methods. Moreover, the basic properties of some metal ions in chloride and fluorides molten salts were determined by electrochemical methods. The work done included:

A) in LiCl-KCl melt, the electrochemical properties of :
- Np.
- Th.
- Ru.

B) in FLiNaK molten salt:
- UF4.
- NdF3.
- GdF3.

Moreover, the study of Zr behaviour in LiFAlF3/Al-Cu system (used for reductive extraction) was performed. Simulation tools for liquid cathode reduction were developed.

WP7: A HAW[38] has been prepared and was used for denitration/calcination and then chlorination and liquid-liquid reductive extraction test. Electrorefining test in LiCl+KCl melt was performed using an Al cathode. The bath contained: U(III), Np(III), Pu(III), Am(III), Y(III), Ce(III), Nd(III), Gd(III) and Zr(II or IV). A total of 3.72 g of An were deposited as metal on the Al cathode with a mass of 4.17 g. This is a most satisfactory result. Other tests of electro refining in the same melt were done with UZr alloy. Liquid-liquid reductive extraction was realized from fluoride melt. The salt melt contained the following elements: An = Pu, Am and Cm ; Ln = La, Ce, Sm and Eu. Two metallic solvents were tested:
- Al-Cu (78-22 mol%).
- Al.

The An were transferred to the metallic solvents very efficiently and almost no Ln were reduced. This test was thus very successful for An/Ln partitioning.

New experimental facilities were first tested or created, including:

- The PYREL II facility (ENEA), which was first cold tested. A total of 26.5 kg of Bi (liquid cathode) and 6.67 kg of LiCl-KCl eutectic was introduced in the plant for the test.
- A new glove-box was created at Nexia Solutions in order to test pyrochemical systems with An, including Pu.
- At NRI[39], an electrolyser was installed in a new glove-box.

At EDF[40] a computational tool was created in order to calculate the electro reduction with liquid cathode in fluoride media.

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[38] HAW = Highly Active Waste.

[39] NRI = Nuclear Research Institute, Czech Republic.

[40] EDF = Electricte de France.

WP8: Several decontamination methods of spent salts were studied:

- Precipitation.
- Electrolysis.
- Ion-exchange with zeolites.

For conditioning spent salts, sodalite was studied.

WP9 Several pyrochemical systems related to different concepts were studied:

- ADS spent fuel.
- MSTR[41] fuel cycle.
- IFR[42] spent fuel.

2.6 Non-EU partitioning work in Europe

Though the majority of the P&T research carried out in Europe is part of the Europart project there is also some independent work being done. One example is the work at Sellafield to use hydroxamic acid complexants to separate U from Np and Pu in advanced Purex flowsheets /Birkett et al. 2005/ and to improve Np routing in reprocessing using Purex /Birkett et al. 2006/. Overviews of this work have also been given by /Fox et al. 2006/ and /Taylor 2006/. A similar development at CEA aimed at separating three minor actinides (Np, Am, Cm) and three fission products (Tc, I, Cs) from the Purex waste stream /Dinh and Baron 2006/. The separation processes were tested by treatment of 13 kg of genuine UO_2 fuel at the Atalante facility. The grouped actinide separation concept is referred to as GANEX (Global Actinide Extraction). GANEX is described very briefly by /Varaine et al. 2004/.

NEA hosted an information exchange meeting on P&T in 2006 /NEA 2006c/. The contributions by /Salvatores and Boucher 2006/ and /Dujardin 2006/ give a good overview of P&T in an international perspective. Other efforts are reported by /Bilancia et al. 2005/ in Italy who used an eight stage centrifugal contactor battery for selective actinide extraction with a tri-synergic mixture. /Kannappan et al. 2006/ developed a new complex for separation of uranium from lanthanides. The complex contains a dinuclear unit composed of two UO_2^{2+} ions bridged by two phenoxide oxygens. /Reinoso-Garcia et al. 2005/ used CMP(O)- and N-acyl(thio)urea-tetrafunctionalized cavitands to separate Am(III) from Eu(III) using Br6-COSAN (bromodicarbollylcobaltate) as synergist. A high selectivity was obtained with tetrakis[(diethyl-N-methylcarbamoylmethylphosphate) propoxy] cavitand at pH 3.

Pyrochemical processes are an alternative to solvent extraction. A good overview of the role of pyro-processes in advanced fuel cycles has been published by /Nawada and Fukuda 2005/ of the IAEA. /Conocar et al. 2006/ tested a pyrochemical An/Ln separation process using fluorides and molten Al. Separation factors of more than 100 were obtained for Pu and Ln(III).

/Lambertin et al. 2005/ determined activity coefficients of Pu and Ce in liquid Ga as part of development of a separation process.

/Serp et al. 2005a,b/ studied the electroseparation of actinides from lanthanides in LiCl-KCl melts using solid Al or molten Bi electrodes. The Al electrode experiments yielded very good separation and high actinide recovery.

/Gaillard et al. 2005/ investigated the possibility to use two ionic liquids, BumimPF6 and BumimTf2N for electrodeposition and solvent extraction. The results are said to be promising.

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[41] MSTR = Molten Salt Test Reactor, USA.

[42] IFR = Integrated Fast Reactor, US concept see section 2.8.2.

2.7 Development in France

2.7.1 Background and general framework

In 1991, the French parliament adopted a law requesting that research on nuclear waste management should be made according to three options, out of which partitioning and transmutation was one. In 2005, CEA issued a report on results from the P&T research performed in France since 1991 /CEA 2005/. The report concluded that

- Aqueous processes for partitioning of minor actinides from Purex raffinate may be deployed on an industrial scale.
- Transmutation of fission products is impractical, and unnecessary when considering reductions in radio-toxic inventories.
- Transmutation (multi-recycling) of plutonium may be performed in existing commercial PWRs, employing slightly modified MOX fuel design.
- Fast neutron systems are necessary for successful transmutation of minor actinides. This task may be performed in homogeneous or heterogeneous modes using sodium cooled fast reactors based on the EFR (European Fast Reactor) design.
- Accelerator driven reactors may play a role in a Double Strata system where all plutonium is utilised in commercial LWRs and minor actinides are destroyed in ADSs. Such a system could be considered if there is a delay in the introduction of critical fast reactors.

The report was reviewed by an international expert group appointed by /NEA 2006/. Following the report, the French parliament adopted a new law in 2006 stating that France should conduct research on Generation IV reactors and accelerator driven systems. The technical feasibility of these systems should be assessed by 2012, leading to the construction of a prototype installation to be taken into operation before end of 2020 /Assamblée National 2006/. President Chirac had previously announced that CEA is given the task to deploy a prototype Generation IV reactor by this date.

In the fast reactor research programme of CEA, priority is given to the more mature sodium technology, while helium gas constitutes a secondary alternative /Pradel and Courtois 2006/. The research on ADS will be performed in an international context. For this purpose CEA has signed a bilateral agreement with SCK-CEN, Belgium to develop a "fast spectrum facility" for construction at Mol, Belgium, employing lead-bismuth or gas coolant.

2.7.2 Transmutation studies

A major focus in the French programme on P&T is fuel development. The sodium cooled prototype breeder reactor Phénix at Marcoule was restarted in 2003 with the explicit purpose of performing fast neutron irradiation experiments for transmutation. Among the experiments are /Fontaine et al. 2005/:

- CAPRIX (high plutonium content MOX fuel).
- MATINA (inert matrix irradiation performance).
- ECRIX (transmutation of americium in magnesium oxide inert matrix targets).
- CAMIX-COCHIX (transmutation of americium in yttria stabilised zirconia inert matrix. targets.
- Futurix-FTA (irradiation performance of ADS fuels with high content of americium).

The ECRIX irradiation was successfully completed in March 2006, reaching 34% fission of the original americium /Sudreau et al. 2006/. Destructive PIE[43] is under way at Marcoule. Results from the MATINA experiment confirmed the good irradiation performance of MgO, which

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[43] PIE = Post Irradiation Examination.

remains the reference inert matrix in the French programme. During measurements of thermal conductivity of the fuels fabricated for the Futurix-FTA experiment, it was however found that the MgO matrix evaporated at a temperature of 2,130°K.

For the fabrication of Futurix fuels, the co-precipitation method was applied as a dust-free mean to obtain a solid solution of plutonium- and americium oxides /Donnet et al. 2005/. The phase structure of the mixed actinide oxide was found to be a complex function of americium content (see section 2.4.1 subsection on Aftra). For the Futurix experiment, two cercer fuel pins were fabricated with the following specifications:

- $(Pu_{0.50},Am_{0.50})O_{2-x}$ – MgO (80 vol% MgO)
- $(Pu_{0.20},Am_{0.80})O_{2-x}$ – MgO (75 vol% MgO)

These fuel pins were loaded into Phénix in May 2007, together with Cermet fuels from ITU and metallic alloy fuels from INL[44] in the US.

Numerous other activities within the field of transmutation are being pursued, and only a few of them are mentioned here:

- Irradiation experiments funded by CEA have been conducted in BOR-60[45] in Russia.
- France is one of the major participants in the Megapie and Eurotrans projects.
- CEA is operating the lead-bismuth loops STELLA, COLIMESTA and CICLAD for development of lead-bismuth technology and corrosion protection testing at flow velocities up to 5 m/s.
- The Megapie target was fabricated by the French company Reel.
- Within Eurotrans, CNRS[46] is responsible for the development of high reliability components for linear proton accelerators.
- Areva[47] is contributing significantly to the design of XT-ADS in the Eurotrans project.

2.7.3 Partitioning studies

See sections 2.5, 2.6 and 2.10.

2.8 Development in USA

2.8.1 General framework

While the idea of partitioning and transmutation originated in the United States /Claiborne 1972, Foster 1974/, serious efforts were for many years hampered by the Carter-doctrine, in practise restricting government funded research related to separation of civilian plutonium. However, after the millennium shift the US government launched the Generation IV and the Advanced Fuel Cycle (AFCI) initiatives /DOE 2002, 2004/. A major goal of these programmes is to identify and develop technologies permitting to avoid construction of a second repository for civilian spent nuclear fuel after the long delayed Yucca Mountain in Nevada.

Following a few years of studies of transmutation in thermal, fast and accelerator-driven systems, the US senate suggested to give priority to fast spectrum reactors. In 2006, the US Department of Energy selected the sodium cooled fast reactor as focus for further development of a system capable of performing the task of transmutation /DOE 2006a/. This development is

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[44] INL = Idaho National Laboratory, USA.

[45] BOR-60 = Russian fast sodium cooled reactor with 60 MW thermal power /Efimov et al. 1998/.

[46] CNRS = Centre National de la Recherche Scientifique.

[47] Areva = French company for development and supply of nuclear installations.

currently implemented as an integral part of the Global Nuclear Energy Partnership (GNEP), where the use of advanced nuclear energy is promoted i.a. to stabilise and reduce carbon dioxide emission rates. Countries joining GNEP would be assisted in employing nuclear power for energy production under the condition that fuel cycle services are handled by a few nations with "secure" advanced nuclear capabilities. In 2006 expressions of interest for siting and building commercial prototype separation and fast reactor facilities were requested and collected. R&D on the underlying technologies for partitioning of spent fuel and transmutation fuel fabrication are meanwhile continued in the AFCI programme /DOE 2006b/.

2.8.2 Transmutation development

Within AFCI, priority has been given to development of metallic alloys fuels, a fuel concept originally considered for the Integral Fast Reactor (IFR) project /Wade and Hill 1997/. Metallic fuels have a large swelling rate, which may be accommodated by fabricating pins with a large fuel-clad gap, filled by liquid sodium to ensure heat removal. The low melting temperature of actinide metals is raised by alloying with zirconium. A significant problem when considering fuels for transmutation is the anomalously high vapour pressure of americium /Kennedy et al. 2004/. In order to avoid losses of Am, the traditional injection casting method had to be re-designed. By use of arc-melting, Idaho National Laboratory proved it possible to fabricate fuel slugs with up to twelve weight percent Am without measurable losses /Hilton et al. 2006a/. Thermo-physical properties of uranium free and low-fertile fuels were measured, and melting temperatures were found to be in good agreement with predictions of Thermo-Calc /Kennedy et al. 2006/.

Americium bearing nitride fuels were fabricated in Los Alamos using carbo-thermic nitridation of oxides /Voit et al. 2005/. Large losses of americium occurred when sintering at temperatures above 1,600°K under inert atmosphere. Sintering under nitrogen, as suggested by KTH /Jolkkonen et al. 2004/ was attempted, but did not allow to meet the target density of 85% TD[48]. Therefore, higher density pellets sintered in argon were used for irradiation experiments, in spite of americium concentrations being much below the initial target.

Test irradiations of Am bearing metallic alloy and nitride fuels were performed in the Advanced Test Reactor (ATR) in Idaho /Hilton et al. 2006b,c/. No pin failures were registered for up to 8% burnup. The metallic fuels exhibited rapid swelling followed by gas release, although with a later onset as compared to standard UPuZr fuels. The nitride fuels exhibited modest swelling and low gas release rates. Extensive cracking and relocation was observed, with potential concern for going to higher burnup. The set of irradiations in ATR is continuing with a final burnup target of up to 40%!

Sodium bonded pins with Pu-12Am-40Zr and U-29Pu-4Am-2Np-30Zr fuels were fabricated and shipped to France for fast neutron irradiation within the Futurix-FTA experiment in Phénix /Jaecki et al. 2006/. The irradiation started in May 2007. Uranium free and low fertile nitride fuels with sodium bond were also fabricated, but will be inserted in a separate irradiation rig at a later stage.

Concerning structural materials, tests of corrosion protection and corresponding modelling efforts have been made by Los Alamos using the Delta test loop /Li 2006/. Good correspondence for corrosion kinetics between model and experimental data were obtained. An aluminium alloyed Oxide Dispersion Strengthened (ODS)-steel loop for tests of corrosion in pure lead at temperatures up to 700°C has been constructed.

A study made by /Wigeland et al. 2006/ indicates that substantial increased drift loadings for high level waste can be achieved in a Yucca mountain type repository if the dominant heat generating elements are separated from the spent nuclear fuel and stored separately or transmuted. The drift loading could be increased by a factor of 4–5 if plutonium and americium is

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[48] TD = Theoretical density.

removed, by a factor about 40 if also cesium and strontium are removed and a factor of 90–225 if also the curium is taken away. The factors presumes a separation efficiency of 99 to 99.9%. The base for the calculations is standard PWR-fuel with 50 MWd/kg, 25 years separation/emplacement time and 100 years repository closure time. In a large nuclear programme P&T och P&C may thus offer a considerable increase in the terms of repository capacity needed per unit of electricity produced.

2.8.3 Partitioning development

An important part of the U.S. P/T research is focused on the UREX+ processes[49]. At present there are five such processes /Bresee 2006/ called UREX+1, UREX+1a, UREX+2, UREX+3 and UREX+4. The difference between them is the number of product fractions. All give U, Tc, Cs/Sr and fission product fractions. UREX +1 and UREX+1a also give a TRU (or TRU + Ln) fraction while e.g. UREX+4 gives Pu + Np, Am and Cm fractions. The UREX+1a process is designed to reduce the attractiveness of process streams to terrorist attack and to enhance the traceability of fissile material.

The UREX processes start with a >99.9% recovery of U. Then Cs and Sr are extracted by the CCD-PEG[50] or FPEX[51] processes /Riddle et al. 2005/. Tc and I are also removed from the waste. The waste then contains only the TRU elements including Pu and the rest of the fission products. Further partitioning can be done using the TRUEX[52] and TALSPEAK[53] processes. To improve the overall process complexation and redox chemistry of U(VI), Np(V) and Pu(VI) with acetohydroxamic acid has been studied by /Sinkov et al. 2006/.

/Law et al. 2006/ reported on separation of trivalent actinides from lanthanides in an acetate buffer solution using Cyanex 301 as extractant. Separation factors up to 960 were obtained but a drawback was that the distribution coefficients of lanthanides usually were too high to allow efficient separation.

Separation of trivalent actinides from lanthanides was also studied by /Peterman et al. 2006/ using a novel dithiophosphinic acid extractant. The results have not yet been published. Complexes of tri- and tetravalent lanthanide and actinide cations with octacoordinate soft donors were studied by /Jensen and Almond 2006/. /Chiarizia et al. 2005/ studied extraction of An(III) and Ln(III) with dialkyl-substituted diphosphonic acids like H_2DEH(MDP)[54] and H_2DEH(EDP)[55]. The fist one showed good extraction but little selectivity, while the second one exhibited less efficient extraction but much higher selectivity. Modified actinide extractants were also studied by /Villazana 2006/ who investigated derivatives of the CMPO extractant.

Cs and Sr separation from nuclear waste solutions have been studied by /Moyer et al. 2005/ and /Law et al. 2005/ using crown ethers and calixarenes, e.g. D(Me3C)CH18C6[56] and BOBCalixC6[57]. A flow sheet for spent nuclear fuel was developed based on these studies.

/Ozawa et al. 2006/ have applied for a patent on a solvent for actinide recovery from acidic solutions. The solvent contains a bidentate organophosphorous extractant in a polar diluent as well as another bidentate organophosphorous extractant, also in a polar diluent.

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[49] UREX = Uranium extraction.

[50] CCD-PEG = Dicarbollide/polyethylene glycol.

[51] FPEX = Fission Product Extraction.

[52] TRUEX = TRansUranium Extraction process.

[53] TALSPEAK = Trivalent Actinide Separation by Phosphorous reagent Extraction from Aqueous Komplexes.

[54] H_2DEH(MDP) = P,P'-di-(2-ethylhexyl)methylenediphospnic acid.

[55] H_2DEH(EDP) = P,P'-di-(2-ethylhexyl)ethylenediphospnic acid.

[56] D(Me3C)CH18C6 = 4,4',(5')-Di-(tertbutyldicyclo-hexano)-18-crown-6.

[57] BOBCalixC6 = Calix[4]arene-bis-(tertoctylbenzo-crown-6).

Overviews of the P&T field have been given by /Nash 2006a,b/ and /Choppin 2005/.

The U.S. also has significant amounts of old defence waste that is taken care of e.g. at Savannah River. Several papers have been published about this subject, e.g. /Campbell 2006, Chang 2006, Hobbs et al. 2005a,b, Long et al. 2006/. These papers mainly concern Sr and Cs separation from alkaline waste solutions. The results are less important for P&T as part of an advanced fuel cycle for power reactors.

A novel approach for reprocessing of nuclear fuel is reported by /Wai 2006/. In this work UO_2 is dissolved in supercritical CO_2 and the separations are then carried out in this medium. A demonstration project using CO_2, tributyl phosphate and nitric acid is described.

/Li and Simpson 2005/ report on an anodic process for electrorefining of irradiated EBR-II[58] driver fuel assemblies. The authors used a molten $LiCl\text{-}KCl\text{-}UCl_3/Cd$ system. Molten salts were also used by /Remerowski et al. 2005/ to recover $^{238}PuO_2$ from combustibles.

There seems to be no extensive work on ionic liquids in the U.S. in 2005–2006 but a review has been given by /Gutowski et al. 2005/.

/Tripp et al. 2006/ report on steam reforming for solidification of cesium and strontium products from an advanced aqueous reprocessing of nuclear fuel. The results show that these waste solutions can be converted into aluminosilicate materials.

A general observation when going through the U.S. work related to P&T is that there is no idea that corresponds to the European CHON principle. Most extractants, not to say all, are based on S and P.

2.9 Development on transmutation in Japan

2.9.1 General framework

Japan has been undertaking long term research on partitioning and transmutation on a national basis since the start of the Omega[59] programme in 1988 /Enarsson et al. 1998/. According to the "Framework for Nuclear Energy Policy" authored by the Japanese Atomic Energy Commission and approved by the Japanese government in October 2005, basic and fundamental R&D activities on P&T shall be carried out to reduce the burden of radioactive waste disposal.

In 2005, two of the major institutes performing research in these fields, JNC and JAERI, merged to form Japan Atomic Energy Agency (JAEA). JNC hitherto had been mainly responsible for developing the sodium cooled fast breeder reactor technology, where transmutation of minor actinides would be performed by homogeneous dispersion into the mixed oxide fuel. JNC operated the 100 MWth sodium cooled fast research reactor Joyo in Oarai, and was also responsible for construction and maintenance of the 250 MWe sodium cooled fast prototype reactor Monju.

JAERI on the other hand, performed development of advanced light water reactors for multi-recycling of plutonium /NEA 2003/, and suggested that minor actinide transmutation better be done in accelerator driven reactors, according to the double strata concept /Murata and Mukaiyama 1984, Tsujimoto et al. 2004/. For this purpose, the application of inert matrix nitride fuel and pyrometallurgical reprocessing is foreseen. The projected coolant for the ADS of JAERI would be eutectic lead-bismuth.

A third approach to P&T is taken by the Central research institute of the electrical power industry (Criepi), where metallic alloy fuels with homogeneously dispersed minor actinides would be employed in a sodium cooled fast reactor.

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[58] EBR-II = Experimental Breeder Reactor number 2; this Na-cooled reactor operated in Idaho from 1954 to 1994.

[59] Omega = Options Making Extra Gains of Actinides and fission products.

Following the creation of JAEA, the Japanese government has decided on a basic plan for science and technology including "intensive investment in fast reactor cycle technology" between 2006 and 2011. The responsible ministry therefore has suggested restarting the operation of Monju in 2008 in order to facilitate this development. Further, the technological development for extraction of minor actinides will be promoted.

2.9.2 Technical achievements

In 2003, irradiation of minor actinide bearing metallic alloy fuels (including Cm) commenced in Phenix in frame of the so called Metaphix experiment. The fuels were designed by Criepi and fabricated by ITU[60] in Germany. Non-destructive PIE results for low burnup fuel (2.5% fission in actinides) show no major problem. Irradiation up to 11% burnup is under way.

$(Pu_{0.2},Zr_{0.8})N$ fuels were successfully irradiated up to 15% fission in actinides in JMTR[61], at a linear rating of 40 kW/m. Post irradiation examination revealed lower swelling rates than expected. This is interpreted as a result of the low operating temperature, due to the high thermal conductivity of ZrN /Arai et al. 2006/.

Small samples of (Np,Pu,Am,Cm)N have been fabricated, showing that a solid solution nitride may be achieved including all these elements. In the case of inert matrix nitrides, fabrication of (Am,Zr)N samples reveal a two-phase structure, indicating limited solubility of AmN in ZrN. The thermal conductivity of AmN was measured for the first time at JAEA, and was found to be five times higher than that of the corresponding oxide /Nishi et al. 2006/.

MOX fuel pellets containing 5% americium were fabricated by JAEA for irradiation tests in Joyo. These tests began in 2006, and will be followed by power-to-melt experiments in 2008 /Minato et al. 2006/.

At Tokaimura, the construction of the J-PARC accelerator complex is proceeding. Two test facilities for ADS research are planned to be built in conjunction with the high power proton accelerator, namely a lead-bismuth spallation target test station with a power of 200 kW thermal, and a zero power sub-critical facility with minor actinide based fuel. A lead-bismuth loop for corrosion tests of steel clad materials is under operation since several years.

2.10 Development of transmutation in other countries

2.10.1 Russia

While Russia is operating both fast reactors and reprocessing facilities at full demonstration scale, the emphasis so far was mainly on proof of feasibility for fast reactor technology. E.g. does BN-600[62] use UOX fuel instead of MOX, and the separated plutonium from the Mayak reprocessing plant at Ozersk is not utilised as fuel for LWR.

In the nineties a major effort was devoted to the development of the BREST reactor concept, a lead cooled fast reactor, partially based on the lead-bismuth technology implemented in Soviet sub-marines. The BREST reactor was designed to be able to recycle its own production of minor actinides /Adamov et al. 1997/.

The BOR-60[63] reactor in Dimitrovgrad has been intensely used for innovative fuel irradiation experiments, including successful irradiation of PuO_2-MgO and (Pu,Zr)N fuels up to 20% burnup. A lead loop experiment was performed in BOR-60, where the lead bonded (U,Pu)N fuel intended for BREST was tested for the first time.

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[60] ITU = Institut für Transuranelemente, Karlsruhe operated by EU.

[61] JMTR = Japanese Materials Test Reactor.

[62] BN-600 = Russian sodium cooled fast reactor with 600 MWe power.

[63] BOR-60 = Russian sodium-cooled fast reactor with 60 MWth /Efimov et al. 1998/.

The Russian government has recently revitalised the programme for innovative reactors with the final aim of demonstrating the feasibility of closing the fuel cycle by means of partitioning and transmutation in fast neutron reactors. This means that the sodium cooled BN-800 reactor, which will be completed by 2012, should be loaded with MOX fuel. For this purpose, the construction of a new MOX fuel factory will be required. BN-800 would later be used for validation of the closed fuel cycle concept, possibly including a co-located reprocessing plant based on pyrochemical technology /Bychkov 2007/.

2.10.2 Korea

The Korean policy on spent nuclear fuel remains undecided, with intermediate storage at power plant sites assumed until 2016. A significant R&D programme on direct use of spent PWR fuel in Candu reactors (Dupic) has been running since 1991, and today Dupic pellets have been fabricated using PWR fuel with 65 MWd/kg burnup /Kim 2007/. Dupic fuel fabricated from lower burnup PWR pellets has been irradiated in the Hanaro research reactor. The spent Dupic fuel would then be pyrochemically processed into metallic alloy fuel for further use in sodium cooled fast reactors (Kalimer-600) or lead-bismuth cooled accelerator driven reactors (Hyper).

2.11 Other partitioning developments

2.11.1 Supercritical separation technique

In Korea work on supercritical CO_2 extraction of U(VI) from HNO_3 using a diamide extractant /Koh et al. 2006/ has been done. Carbon dioxide may be a promising substitute to normal solvent extraction due to its ease of recycling, and the fact that is leaves negligible amounts of secondary waste.

2.11.2 Pyrochemical separation techniques and electrorefining

In Japan a method for reprocessing of spent oxide fuels, characterized by carbon-chlorination treatment in combination with electrolysis, has been developed /Mizugushi et al. 2005/. /Minato et al. 2005/ have done a review of an experimental facility, TRU-HITEC (TransUranium High Temperature Chemistry), used for basic studies of the behaviour of TRU elements in pyro-chemical reprocessing of oxide fuels. Other Japanese patents describe a method for minor actinide recycling from oxide fuels /Fujita et al. 2006/ and an electrolytic device for recovery of useful components from spent nuclear fuels /Igakura et al. 2006/.

Ikegami has written a paper on a new fuel cycle concept named “Compound Process Fuel Cycle”, in which LWR spent fuels are multi-recycled in a fast reactor core without ordinary reprocessing but with only a simple dry pyrochemical processing with almost no increase of minor actinide content /Ikegami 2006/. Other recent work in pyroprocessing involves sulfurization, which basic concepts are described by /Sato and Tochiyama 2006/, and electrochemical reduction technology /Ohta et al. 2005/. Further articles describing electrorefining are also available /Kinoshita et al. 2005, Kato et al. 2006, Asakura et al. 2005, Hoshi et al. 2005/.

In Russia separation coefficients of uranium and rare earth metals in LiCl-KCl eutectic melt by electrochemical transient techniques have been investigated /Kuznetsov et al. 2005/. /Serebryakov et al. 2005/ have investigated the safety of reprocessing 15.5 kg spent BOR-60 fuel. Other techniques that also have been studied recently involve nitrate alkali melts /Gotovchikov et al. 2005/, molten carbonates with subsequent precipitation of rare earth fission products using phosphate /Griffiths et al. 2006/, electrochemical recovery of palladium from nitric acid solutions /Kirshin and Pokhitonov 2005/ and electrochemical transient techniques for determination of uranium and rare earth metal separation coefficients in molten salts /Kuznetsov et al. 2006/.

2.11.3 Ion-exchange chromatography

An extraction chromatographic method for the separation of actinides and lanthanides using a chlorometylated polymer support grafted with 4-ethoxy-N,N-dihexylbutanamide has been reported /Maheswari and Subramanian 2005/. More chromatographic studies have been done in Japan, involving e.g. adsorption characteristics on anion exchangers /Wei 2005/, minor actinides separation from high-level waste by CMPO/SiO_2-P extraction resin /Wei et al. 2006, Zhang et al. 2005/, separation of trivalent actinides from rare-earth elements using a pyridine resin embedded in high-porous silica beads /Ikeda et al. 2005/ and anion exchange in combination with electrolytic reduction and extraction chromatography /Wie et al. 2005/.

Other articles concerning chromatographic separation involve e.g. recovery of palladium from nitric acid solutions on anion-exchange resins /Korolev et al. 2005/, uranyl, thorium and lanthanum nitrate extraction using a polymer carrier and tri-isobutylphosphinosulfide (in Russian, /Pyartman et al. 2006/), sorption property determination of fibrous materials filled with anion exchangers and complexing sorbents used for concentrating and separating radionuclides /Myasoedova et al. 2006/, use of dipicolinate-based complexes for producing polystyrene resins for extraction of yttrium and heavy lanthanides /Chauvin et al. 2006/, thin-layer chromatography of actinides and lanthanides using silica gel and a mobile phase containing tributylphosphate, trioctylphosphine and di(n-butyl)dithiophosphoric acid /Curtui and Soran 2006/ and finally the Russian patent on reprocessing of liquid nuclear waste using an ion-selective sorbent after concentrating, ozonization and microfiltration /Chechel'nitskii et al. 2006/.

2.11.4 Magnetic separation

A nanoscale magnetite carrier with an adsorbed layer of dibenzo-18-crown-6, to remove radionuclides from nuclear waste solutions, has been studied /Ambashta et al. 2005/. In Japan a process for spent nuclear fuel reprocessing, which involves a magnetic separation of fuel and fission products, has been developed, patent written in Japanese /Sato et al. 2005a/. /Sato et al. 2005b/ have described a sulphide reprocessing scheme, which consists of oxidation, selective sulfurization and magnetic separation or selective dissolution steps. In India a magnetically assisted separation process using tiny ferromagnetic particles coated with Cyanex 923 was investigated /Shaibu et al. 2006/.

2.11.5 Solvent extraction

A literature review on spent fuel reprocessing technology in Japan has been written /Takahashi 2006/. Recent activities on aqueous partitioning in Japan have been summarized by /Kimura et al. 2006/. A presentation of IAEA activities in the area of P&T is also available /Stanculescu 2006/. In India /Dey and Bansal 2006/ have written a review on spent fuel reprocessing.

A theoretical study on structures and energies of trimethyl phosphate complexes with nitric acid and water has been done /Kim et al. 2005/. Molecular structures of β-diketones has also been made and confirmed by experiments /Umetani 2006/.

Experimental solvent extraction studies are available from a variety of countries around the world, e.g.

- chemical studies of rutherfordium using SISAK[64] /Polakova et al. 2006/,
- synergic extraction of lanthanides with a mixture of picrolonic acid and benzo-15-crown-5 in chloroform /Rehman et al. 2006/,
- recovery of Pd from nitric acid solutions using carbamoyl phospine oxides /Zaitsev et al. 2005/,
- extraction using adducts of complex inorganic acids with monosubstituted polyethylene glycols /Smirnov and Efremova 2006/,

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[64] SISAK = Short-lived Isotopes Studied with the AKufve-technique.

- separation of actinides and lanthanides using TPEN and its derivatives /Matsumura and Takeshita 2006/,
- extraction behaviour of trivalent lanthanide ions using mixtures of N-p-methoxybenzoyl-N-p henylhydroxylamine and 1,10-phenanthroline /Inoue et al. 2006/,
- extraction studies of TODGA[65] molecules /Ansari et al. 2005/,
- data presentation on dispersion-free solvent extraction of Pu using microporous hydrophobic polypropylene hollow fibre membrane contactors with TBP /Gupta et al. 2005/,
- synergic extraction of Zr and H using 4-acylbis(1-phenyl-3-methyl-5-pyrazolones) in the presence of neutral organophosphorus extractants /Reddy et al. 2006/,
- evaluation of calix-crown ionophores for selective separation of Cs from acidic nuclear waste /Mohapatra et al. 2006/.

Combining theory and experimental achievements will results in process development, which here is represented by the following work:

- recent development in China on rare earth separation /Yan et al. 2006/,
- a simplified solvent extraction process for U, Pu and Np recovery (in Japanese, /Nakahara et al. 2006/),
- the new reprocessing technology, FLUOREX, which combines fluoride volatility and solvent extraction methods /Kobayashi et al. 2005/,
- studies of Pu(IV) polymer formation /Kumar and Koganti 2005/,
- reprocessing of heavy water reactor fuels (involving a combination of Purex and THORAX[66]) /Kant 2005/.

There are some recent patents concerning P&T; extraction and recovery of Purex-refined products (in Russian, /Zil'berman et al. 2005/) and a method and device for reprocessing of plutonium rich MOX fuel (in Japanese, /Shimada et al. 2005/).

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[65] TODGA = N,N,N´,N´tetraoctyl diglycolamide.

[66] THORAX = THORium EXtraction process.

3 Recent results from Swedish R&D-programmes on P&T

3.1 Royal Institute of technology – KTH

The reactor physics group at KTH has been performing research on transmutation of nuclear waste during 15 years. Under the leadership of Waclaw Gudowski, the research group expanded from three persons in the early 90's to 15 in 2006. The scope of research has broadened from Monte Carlo simulations of advanced reactor core neutronics to transient safety analysis, multi-scale modelling of radiation damage in model alloys, thermo-chemistry of transmutation fuels, high temperature reactors and fuel-cycle economics. Six PhD theses on waste transmutation have been completed and 55 papers have been published in peer reviewed journals since 1999. The activities have mainly been funded by nuclear industry (in particular SKB) and by the European Commission. The group has been or is co-ordinating international projects in the 4th, 5th and 6th Framework Programmes (The IABAT, Confirm and RedImpact projects respectively). From November 2006, Janne Wallenius is head of the Reactor Physics division, employing seven senior scientists and seven PhD students.

Starting with the fifth framework programme, the division of nuclear power safety commenced experimental activities relevant for transmutation systems, building up the TALL facility, a lead-bismuth loop for thermal-hydraulic studies.

In what follows, the research activities at KTH since the previous status report in 2004 will be summarised. See also /Gudowski et al. 2004, 2005, 2006/.

3.1.1 System and safety studies of accelerator driven systems

Although the suitability of accelerator-driven systems for the purpose of higher actinide burning was suggested already in 1974 /Foster 1974/ and its proper placement in the Double Strata fuel cycle was well established prior to the millennium shift /Salvatores et al. 1998/, very little detailed design studies of ADSs with minor actinide based fuel existed in the open literature. In particular, no coherent comparison between different design options was available. At KTH, several PhD theses devoted to this subject were presented in recent years /Eriksson et al. 2005, Seltborg 2005, Westlén and Wallenius 2006b/.

Neutronic properties of lead-bismuth cooled accelerator driven systems (ADS) with different minor actinide based ceramic fuels (two composite oxides and one solid solution nitride) were studied using the Monte Carlo code MCNP /Wallenius and Eriksson 2005/. Adopting a TRU composition with 40% plutonium in the initial load, transmutation rates of higher actinides (americium and curium) in the order of 265–285 kg per GWth·year are obtained. The smallest reactivity swing is provided by the magnesium oxide based cercer fuel. The cercer cores however exhibit large coolant void worth, which is of concern in case of gas bubble introduction into the core. Nitride and cermet cores are more stable with respect to void formation. The poorer neutron economy of the molybdenum based cermet makes it however difficult to accommodate an inert matrix volume fraction exceeding 50 percent, a lower limit for fabricability. Higher plutonium fraction is thus required for the cermet, which would lead to lower actinide burning rates. The nitride core yields high actinide burning rates, low void worth and acceptable reactivity losses.

Safety characteristics of these designs were investigated using the fast reactor transient analysis code SAS-4A /Cahalan et al. 1994/, adapted by Argonne National Laboratory to permit simulations of source driven systems. Thermo-physical properties of the novel fuel types were parameterised. The accident analyses was carried out for response to unprotected loss-of-flow,

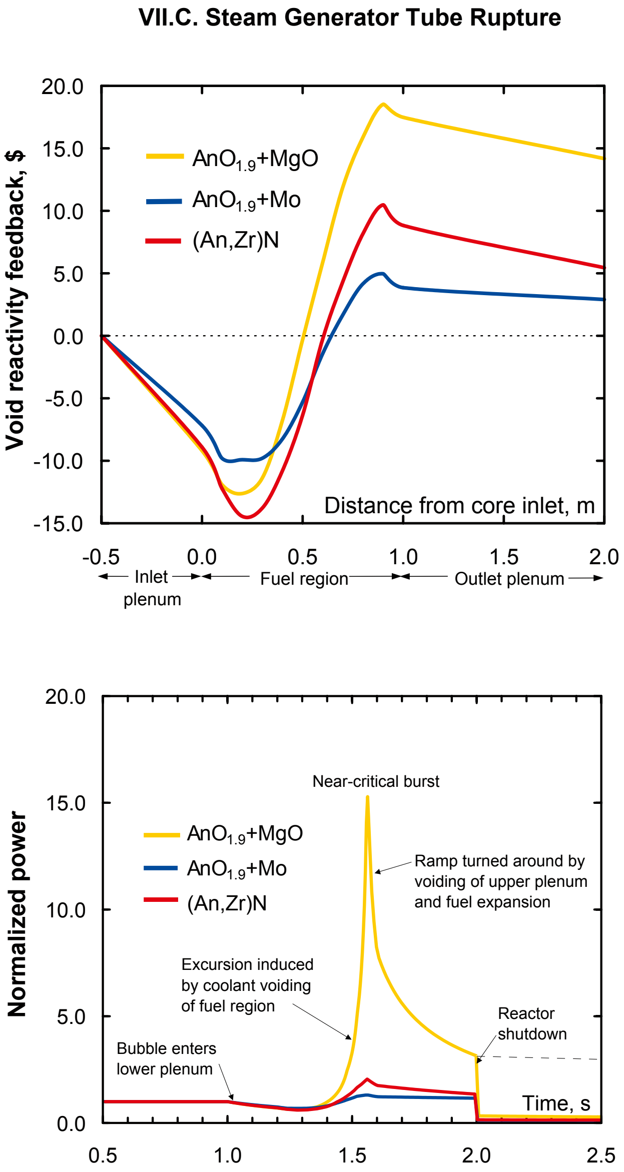


Figure 3-1. *Reactivity insertions and corresponding power excursions during transport of a gas bubble through sub-critical cores with different inert matrix fuels. From /Eriksson et al. 2005/.*

accelerator beam-overpower transients and coolant voiding scenarios. Maximum temperatures during transients were determined and compared with tentative design limits. Reactivity effects associated with coolant void, fuel- and structural-expansion, and cladding relocation were investigated. The studies indicated favourable inherent safety features of the CerMet fuel. Major consideration is given to the potential threat of coolant voiding in accelerator-driven design proposals. Results for a transient test case study of a postulated steam generator tube rupture event leading to extensive cooling voiding underlines the importance of a having a low coolant void reactivity value in a lead-bismuth system, despite the high boiling temperature of the coolant. It was found that the power rise following a voiding transient increases dramatically near the critical state, as illustrated in Figure 3-1. The studies suggest that a reactivity margin of a few dollars in the voided state is sufficient to permit significant reactivity insertions.

In earlier studies at KTH, the proton source efficiency was defined to remove ambiguities in the definition of the neutron source, when assessing the external neutron source efficiency in proton accelerator driven systems /Seltborg et al. 2003/. The proton source efficiency was then used to assess potential core designs. In a study on the impact of fuel composition on the proton source efficiency it was shown that nuclides with an even number of neutrons in general and ^{241}Am and ^{244}Cm in particular have favourable properties with respect to improving the proton source efficiency if they are placed in the innermost part of the core. Moreover, it was shown that loading the inner part of the core with only curium increases the proton source efficiency by 7%. Plutonium, on the other hand, in particular high-quality plutonium consisting mainly of ^{239}Pu and ^{241}Pu, was found to be a comparatively source inefficient element and is preferably located in the outer part of the core /Seltborg and Wallenius 2006/.

Comparing different options for the ADS coolant it was found that the proton source efficiency, is reduced by 10% when switching coolant from helium to lead-bismuth /Westlén and Seltborg 2006/.

The potential of gas cooled ADSs for minor actinide transmutation was studied for nitride fuels with traditional pin and pellet design /Westlén and Wallenius 2006a/ and for a titanium nitride coated particle fuel /Westlén and Wallenius 2006b/. Comparing to liquid metal cooled systems, a harder spectrum is achievable, which is beneficial for reducing the buildup of curium.

KTH is co-ordinating the safety work package in the Design domain of Eurotrans. One particular task is to assess the so called "source-term" of XT-ADS and EFIT. This includes estimations of the fractional release of radio-activity to the environment from a severely damaged core. A preliminary study has been made where the volatilities of caesium, strontium, iodine and polonium contaminants in liquid lead and lead-bismuth have been evaluated. This work includes predictions of the predominating chemical forms in solution and gas phase, with special emphasis on the degree of non-ideal properties in very dilute solution. These investigations make it possible to predict the equilibrium (i.e. maximal) vapour pressure of radiotoxic species over a coolant spill or breached reactor vessel at a given temperature. First assessments indicate that retention of volatile fission products in liquid lead is very good (due to formation of less volatile compounds with lead), and one might thus expect a small specific magnitude of the source term as compared to other reactor systems. In the case of XT-ADS, where significant amounts of polonium will be produced in the lead-bismuth coolant, an issue of concern may be the formation of volatile polonium hydride, if the irradiated coolant comes into contact with water.

3.1.2 Coupling between accelerator and sub-critical core

The reactor physics division is participating in a series of experiments performed at the Yalina facility in Minsk. The facility consists of a high intensity neutron generator coupled to a sub-critical zero-power core.

The main parts of the neutron generator are the deuteron ion accelerator and a Ti-T or Ti-D neutron production target, with a diameter of 45 mm, located in the centre of the core. The neutron energy is around 14 MeV for the DT- and 2.5 MeV for the DD-reaction. The neutron generator can be operated in both continuous and pulse mode and provides the possibility to generate pulses with frequencies from 1 Hz to 7 kHz with pulse duration of 2–130 microseconds. The maximum beam current in continuous mode is 2 mA with a beam diameter about 20 mm giving a maximum neutron yield of approximately $2 \cdot 10^{11}$ neutrons per second for the Ti-T target and $2 \cdot 10^{10}$ neutrons per second for the Ti-D target.

The first experiment with participation of KTH concerned validation of different methods for measuring sub-criticality in a thermal core with 10% enriched uranium dioxide fuel and polyethylene moderator /Persson et al. 2005/. The zero-power core is cooled by natural convection of the surrounding air. Here it was found that the pulsed neutron source slope fit method (PNS) provides the best estimate of reactivity (using MCNP simulations as the reference value). The source jerk method suffers from poor statistical accuracy but appears to overestimate reactivity, whereas the area method /Sjöstrand 1956/ results in underestimated values.

In a second experiment, a dual spectrum fast-thermal core denoted as Yalina-Booster was constructed. The core consists of a central lead zone, a polyethylene zone, a radial graphite reflector and a front and back biological shielding of borated polyethylene. The loading is 132 fuel pins containing metallic uranium of 90% enrichment, 563 fuel pins containing uranium dioxide of 36% enrichment, 108 fuel pins with metallic natural uranium and a maximum of 1,141 EK-10 fuel pins containing uranium dioxide of 10% enrichment. The loading of the latter fuel pins is made based on cylindrical symmetry as shown in Figure 3-2.

The fast-spectrum lead zone and the thermal-spectrum polyethylene zone are separated by a so called thermal neutron filter, which consists of one layer of 108 pins with metallic uranium and one layer of 116 pins with boron carbide (B_4C) which are placed in the outermost two rows of the fast zone. Thermal neutrons diffusing from the thermal zone to the fast zone will either be absorbed by the boron or by the natural uranium, or transformed into fast neutrons through fission in the natural uranium. In this way, a coupling of only fast neutrons between the two zones is maintained.

Detector channels are located in different parts of the core. The PNS slope fitting method showed excellent stability: less than 2% spread for all experimental channels. In the core region, the area method appeared to be rather stable, but in the reflector some deviation was found. Keeping in mind that this result may be reactivity dependent, this is in contrast to other studies, where the area method has shown to be more stable than the PNS slope fitting.

The ^{252}Cf-source based noise methods Rossi-α and Feynman-α did not reproduce the result from the PNS slope fitting, indicating the need of two-region models. The deviation increased with the subcriticality level.

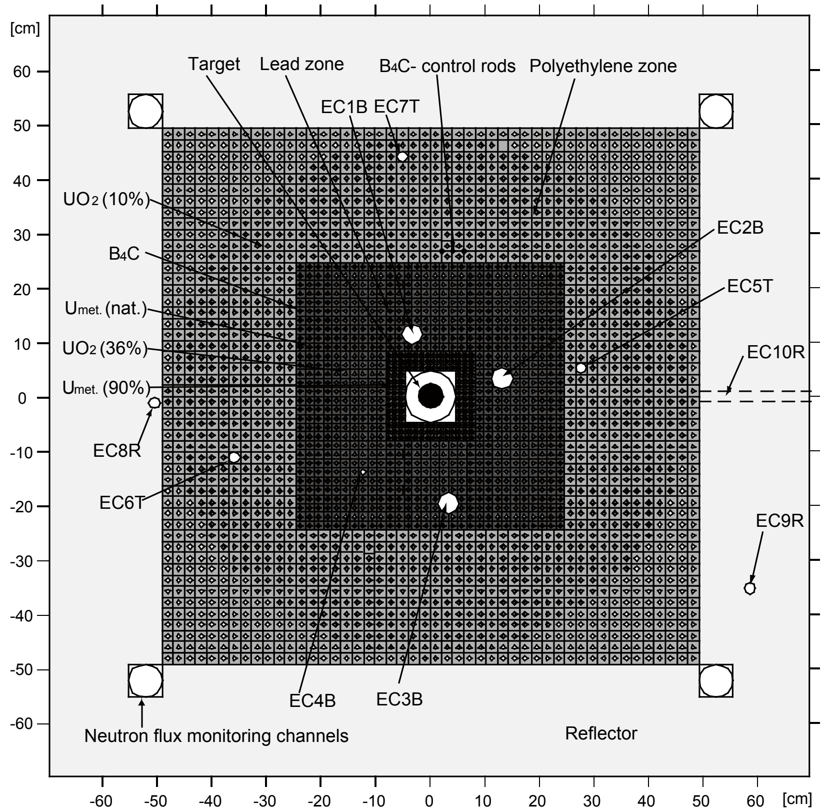


Figure 3-2. *Schematic cross-sectional view of Yalina-Booster.*

Prompt neutron decay constants obtained from the pulsed Rossi-α formula are in good agreement with those from the PNS slope fitting method. However, it is much simpler to directly find the prompt neutron decay constant from the PNS histogram than performing the Rossi-α analysis. Thus, the pulsed Rossi-α method is less attractive for reactivity monitoring of pulsed systems /Persson et al. 2007/.

3.1.3 Transmutation fuel studies

The criteria for choosing the fuel form and composition for EFIT were elaborated within the 5th framework programme Future, as well as in the initial stages of the Aftra domain of Eurotrans /Wallenius et al. 2006/. Taking into account

- fabrication,
- reprocessing,
- secondary waste stream,
- high temperature stability,
- transmutation performance,
- thermo-mechanical performance under nominal conditions,
- neutronic performance under nominal and accident conditions,
- margin to failure and
- ultimate behaviour under accident conditions

it was recommended to select two composite oxide fuels as the final candidates for the design of EFIT /Wallenius et al. 2006/:

1. $(Pu,MA)O_{2-x}$ – ^{92}Mo
2. $(Pu,MA)O_{2-x}$ – MgO

where the molybdenum based CerMet (Ceramic-Metallic) composite fuel was judged to provide superior safety characteristics. The cost associated with enrichment of molybdenum however lead the participants in the Design domain to select the magnesium oxide based fuel as the primary candidate.

In the fifth framework programme project Confirm (being coordinated by KTH) inert matrix nitride fuels were fabricated at PSI according to a procedure optimised at KTH using Thermo-Calc /Jolkkonen et al. 2004/. By switching gas phase during sintering from argon to nitrogen, it became possible to obtain a single phase material with low level of oxygen/carbon impurities and good mechanical properties. The characteristic golden colour of a nitride fuel is evident in Figure 3-3. These pellets were put into fuel pins and shipped to Sweden for irradiation in Studsvik. As the R2 reactor in Studsvik was shut down this experiment had to be moved to the HFR reactor in Petten, Holland. The (Pu,Zr)N pins manufactured by PSI were shipped from Studsvik to Petten in March 2007, and irradiation of two segments will start during summer. Destructive post-irradiation examination is to be made by PSI as part of the Aftra programme.

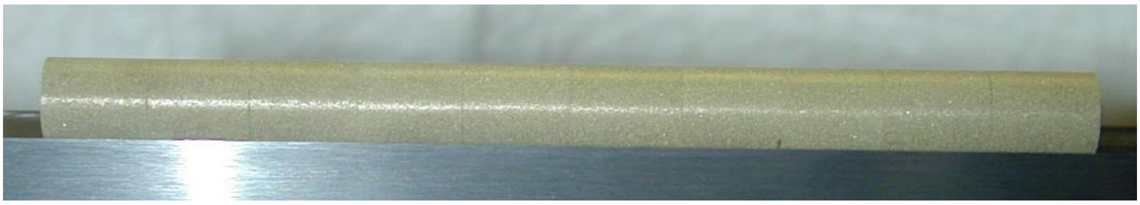


Figure 3-3. *(Pu,Zr)N fuel pellets fabricated at PSI following a procedure suggested by KTH.*

3.1.4 Thermal hydraulics experiments in lead-bismuth

TALL is an experimental facility constructed at KTH to study steady-state and transient thermal-hydraulics performance of LBE-cooled reactors, with the primary purpose of supporting the development of lead-bismuth-cooled accelerator driven systems. The flow can be by forced convection or by natural circulation and is used to study performance during representative accident scenarios. The specifications of TALL are set and modified to simulate prototypic thermal hydraulic conditions of an ADS, with well-conditioned flow and controllable power for thermal-hydraulic tests. The facility consists of a primary loop (LBE loop) and a secondary loop (oil loop). The LBE loop shown in Figure 3-4 is 6.8 m tall and the location of heaters and heat exchangers allows natural convection flows as should occur in the prototypic vessel. The LBE loop is of full height and has been scaled for prototypic (power/volume) ratio to represent all components, their LBE volume, pressure drops, flow velocity and heating rates corresponding to one tube of the selected heat exchanger design. In the 5th FP, experiments were performed to investigate the heat transfer performance of different heat exchangers, and the thermal-hydraulic characteristics of natural circulation and forced circulation flow under steady and transient conditions. One significant finding of these experiments is that the establishment of natural circulation may take longer time than expected /Ma et al. 2005/.

Figure 3-4. *The TALL facility at KTH.*

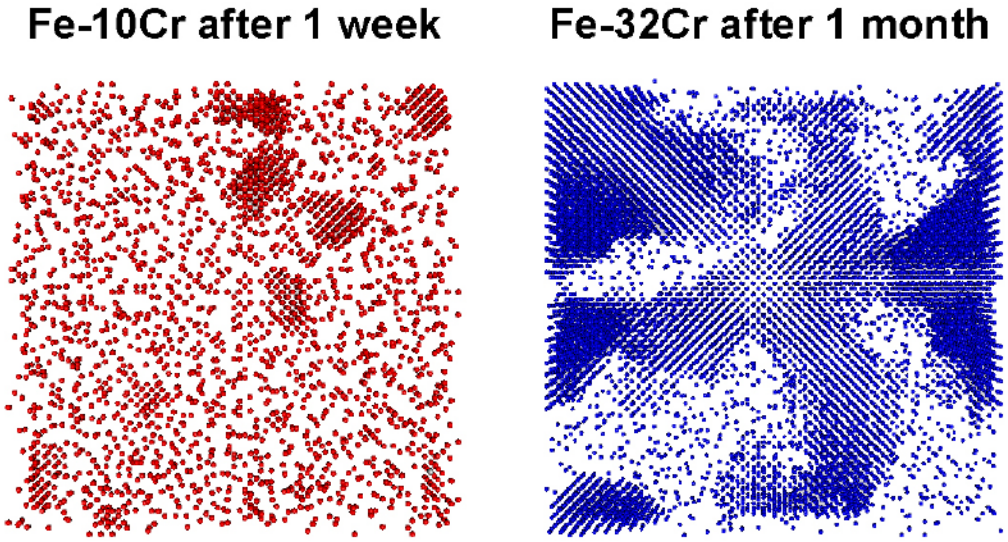
3.1.5 Radiation damage simulation

Significant progress was made in the multi-scale modelling of irradiation induced defect production in Fe-Cr alloys. This work has been done in close collaboration with Uppsala university, SCK-CEN, EdF and Helsinki University. Electronic structure calculations made with the EMTO-CPA code developed in Uppsala lead to the discovery of the negative mixing energy of the alloy for small Cr concentrations /Olsson et al. 2003, 2006/.

Further calculations of formation energies for interstitial defects in the vicinity of Cr atoms revealed that Cr tends to bind to interstitials, which will reduce their mobility /Olsson et al. 2007/. In order to enable simulation of defect cluster mobility in the alloy, a novel “two-band” embedded atom method model of the alloy was constructed where by assigning the change in sign of the mixing enthalpy to interactions between s-electrons of Fe and Cr, it became possible to reproduce the formation energy calculated with ab initio methods over the relevant range of Cr concentration. This model was applied in molecular dynamics simulation of defect formation energies, and it was shown that the “two-band” set of potentials were capable of reproducing the formation and binding energies of a wide range of interstitial configurations including one or two Cr atoms in bulk Fe. Using the potential in Kinetic Monte Carlo simulations of phase segregation, formation of the alpha-prime phase was observed with cluster characteristics typical for those observed in experiment. Figure 3-5 shows the positions of Cr atoms in originally random alloys after simulated ageing at 750°K /Olsson et al. 2005/.

For lower Cr concentrations than 9%, no segregation is observed. In Fe-10Cr, the spherical shape of the precipitates are those typical for the nucleation and growth mechanism, expected to occur for positive curvatures of Gibbs’ energy, while in Fe-32Cr, the precipitates have the diffuse shape typical for spinodal decomposition. The composition of the precipitates is similar to those found experimentally, with Cr concentrations ranging from 50–90%. While the present version of the two-band model enables for the first time to simulate the formation of the alpha-prime phase in Fe-Cr, it needs to be extended to take into account the effects of magnetism. This work is in progress.

Diffusion coefficients for interstitial clusters calculated with the two band model showed that the mobility of such clusters in the alloy is reduced by two orders of magnitude as compared with pure iron, correlating well with the experimentally measured swelling rates as function of chromium content. The result is a much improved understanding of basic mechanisms responsible for swelling (or rather the absence of swelling) in ferritic steels /Wallenius et al. 2007/.


Fe-10Cr after 1 week

Fe-32Cr after 1 month

Figure 3-5. *Positions of Cr atoms in initially random FeCr alloys after simulated thermal ageing.*

3.1.6 Fuel cycle economy – the cost for partitioning and transmutation

Within the RedImpact project, an update of previous fuel cycle cost assessments when including partitioning and transmutation has been made at KTH. A case study for the Swedish nuclear power system was made where a strategy with minor actinides and plutonium recycled in fast reactors was compared to a double strata scenario where plutonium is multi-recycled in LWRs using MOX with support of enriched uranium, and minor actinides are transmuted in accelerator driven systems /Runevall 2006/. Relative to the present Swedish once-through "cycle", the cost penalty for the fast reactor approach was estimated to 33%, and that of the ADS to 27%. The uncertainty in the cost estimate for partitioning and transmutation is large enough for the difference between the fast reactor and ADS approach to remain inconclusive. Still, there appears to be no reason to claim that introduction of ADSs for the specific purpose of minor actinide burning would be much more costly than to perform the same task in critical fast neutron reactors.

3.2 Chalmers University – nuclear chemistry

Chalmers is one of the 24 partners for Europart and thus much of the research made at Chalmers is in collaboration within this international frame. The use of nitrogen-containing heterocyclic molecules has been shown to efficiently separate actinides and lanthanides using solvent extraction. The basic structure has changed over the years and currently the BTBP family is the most promising one. BTBP stands for *bis*-triazine-bipyridine, which refers to the nature of the central core molecule common to all of the molecules in the family. A precursor to the BTBP family was the BTP family (*bis*-triazine-pyridine). Schematic figures of the chemical structures of the two types of molecules are presented in Figure 3-6. Most of the research at Chalmers involves these groups of molecules.

3.2.1 Basic extraction properties

A major part of the studies at Chalmers includes determination of basic extraction properties of the various extractants used for solvent extraction. The distribution ratios (D) for different metals and the separation factor (SF) between metals are calculated and compared for different extracting molecules. The most common metal pair studied is Am/Eu, which is used as analogue for the separation between trivalent actinides and lanthanides. Extraction of other actinides, such as Th, U and Np has also been investigated for a number of BTBP molecules. The conclusion of one of those experiments was clear and interesting: Np was the only metal extracted by all the investigated BTBP systems /Dubois et al. 2007/. An example of these results is shown in Figure 3-7, where the Th, U and Np extraction by different concentrations of CyMe$_4$-BTBP (6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo[1,2,4]triazin-3-yl)-[2,2']-bipyridine) in cyclohexanone is illustrated.

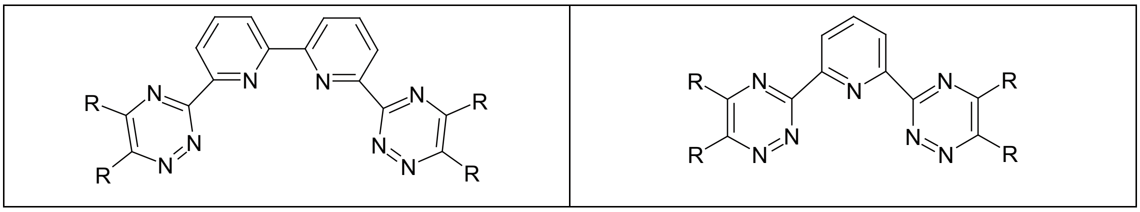


Figure 3-6. Chemical structures of BTBP (left) and BTP (right).

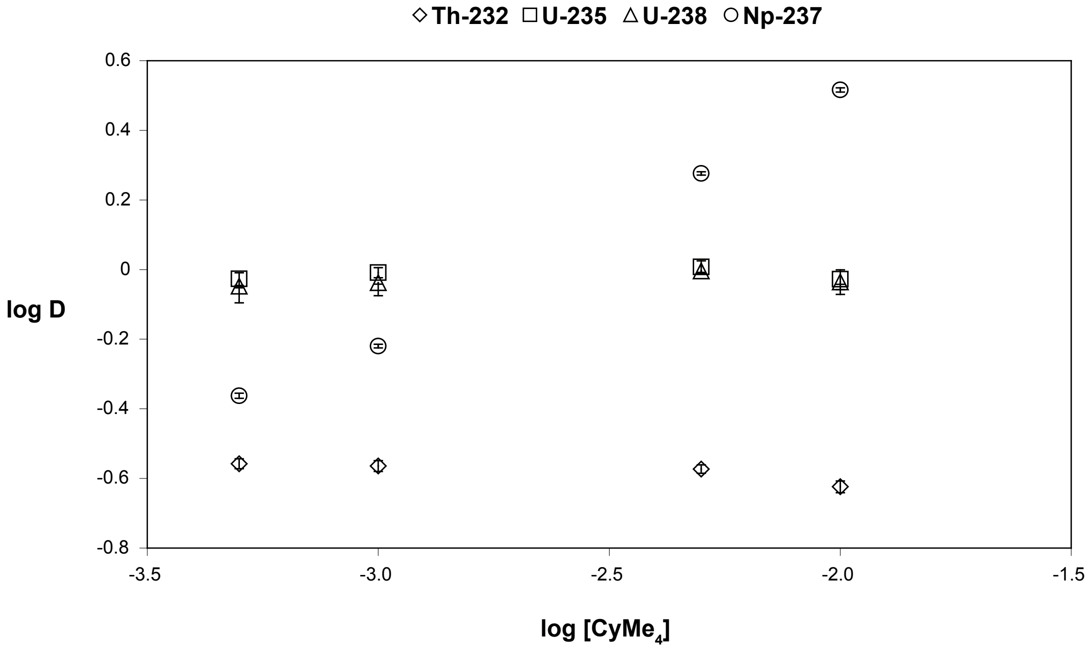


Figure 3-7. *Extraction of Th, U and Np with a varying CyMe$_4$-BTBP concentration in cyclohexanone. The aqueous phase consisted of 2 µM ^{234}Th, 2 µM natural U and 0.4µM ^{237}Np in 1 M HNO$_3$.*

The high level liquid waste solutions from a nuclear power plant contain a mixture of fission products, corrosion products and actinides. Extraction tests have been performed with synthetic solutions containing many fission products /Andersson et al. 2004, 2005/ as well as with nickel, which is a corrosion product that inevitably will be present in an industrial extraction system /Dubois et al. 2007/. Due to high distribution ratios for nickel with some nitrogen donor ligands it may be feared that the extraction of the actinides are hampered. In addition, nickel is a generally difficult element due to the combination of charge and valence, making it difficult to saturate and fear of formation of polynuclear complexes may exist.

By studying the extracting properties of several different BTP and BTBP molecules it has been possible to draw conclusions regarding the effect of varying the core molecule (BTP vs. BTBP) as well as varying the side groups (e.g. C5-BTBP vs. CyMe$_4$-BTBP) /Andersson et al. 2004, 2005, Dubois et al. 2007/.

The extracting properties of a certain system can be affected in many ways and a number of them have been studied. By changing the organic diluent the efficiency and selectivity of a molecule can be widely changed. One of the most promising molecules studied at Chalmers is CyMe$_4$-BTBP, and many different diluents have been tested together with this molecule. The diluent should obey the CHON-principle, i.e. consist of only carbon, hydrogen, oxygen and nitrogen, hence be totally incinerable and thereby more environmentally friendly than diluents including e.g. sulphur or phosphor. Other requirements for a suitable diluent include low flashpoint, low solubility in water etc. Tests have been made to find a new diluent to further improve the possible industrial separation process with CyMe$_4$-BTBP /Andersson et al. 2005/.

The effect on extraction by other system properties such as temperature, concentration of the extractant, nitrate concentration and ionic strength of the aqueous phase, has also been studied /Andersson et al. 2004, 2005, Dubois et al. 2007/.

To increase the extraction efficiency towards one or more metals a synergist can be added to the organic phase. The effect of two different synergists; α-bromodecanoic acid and 3,5-diisopropylsalicylic acid has been studied for a number of organic systems /Andersson et al. 2005/ and some results for a system containing CyMe$_4$-S-BTBP in decanol are shown in Figure 3-8.

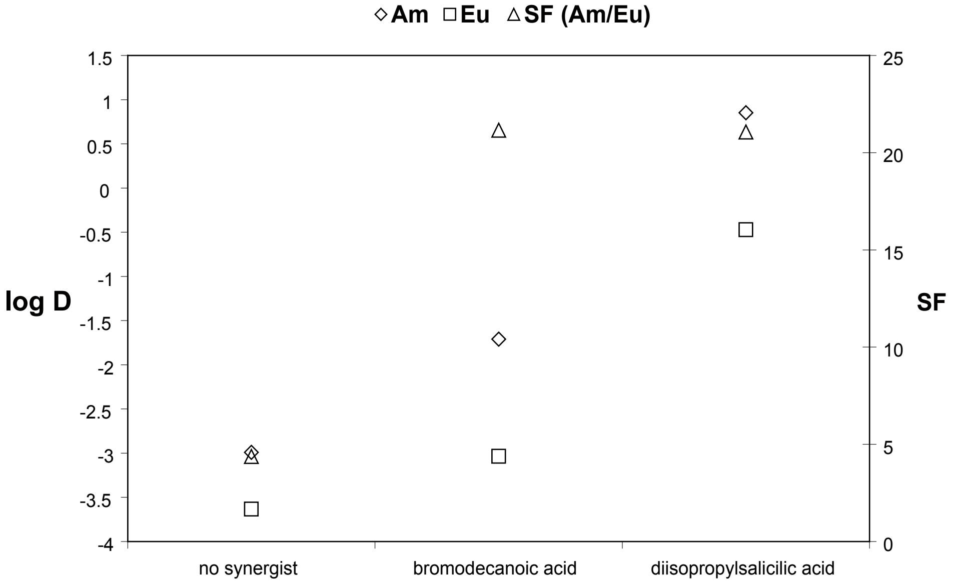


Figure 3-8. *The effect of addition of a synergist to a system containing 0.01 M* *$CyMe_4$-S-BTBP in decanol. The concentration of the synergist is 0.5 M and the aqueous phase consisted of 0.01 M* *HNO_3, 0.99 M* *$NaNO_3$* *and traces of* *^{152}Eu and* *^{241}Am.*

Kinetics

The kinetics of a system is important to study since it influences the contact time required to reach equilibrium between two phases in a potential extraction process. To get as much information as possible about a certain system it is desirable to make experiments at equilibrium. In the kinetic studies carried out it has for example been shown that the time to reach equilibrium varies a lot both with the extracting molecule as well as with the choice of solvent for a specific molecule /Andersson et al. 2004, 2005, Retegan et al. 2006/.

Stability towards irradiation

Since processes intended for the separation of spent nuclear fuel takes place in a high radiation flux, both the extractant and the diluent must be resistant to irradiation. A number of organic systems have been irradiated and the resulting effects on extraction have been investigated and compared to non-irradiated reference samples The latest BTBP molecules have been modified to be more resistant towards radiolysis and the most recent irradiation experiments (MF2-BTBP in cyclohexanone and octanol, respectively) show no change in extraction capacity of Am and Eu after a received dose of 32 kGy (in a dose rate of ~20 Gy/h). This can be compared to e.g. C2-BTP, which in most diluents looses almost all its Am extracting capacity after a dose of just a few kGy. Figure 3-9 shows how the distribution ratio of Am is affected with increased received dose for two different systems; C2-BTP and C5-BTBP, both dissolved in hexanol.

For some systems a change in SF with received dose has been observed. E.g. for a system containing C5-BTBP in cyclohexanone, the extraction of Am decreases more than the extraction of Eu as the dose increases, leading to a decrease in the SF between Am and Eu /Nilsson 2005/. This is an interesting result, since the degradation products seem to be selective towards Eu, and the system will be further studied. A difference in stability towards irradiation can also be seen when the solvent is varied. This behaviour has been studied, along with experiments regarding the possibility to inhibit degradation due to irradiation by adding various organic substances /Nilsson et al. 2006, Fermvik et al. 2006/.

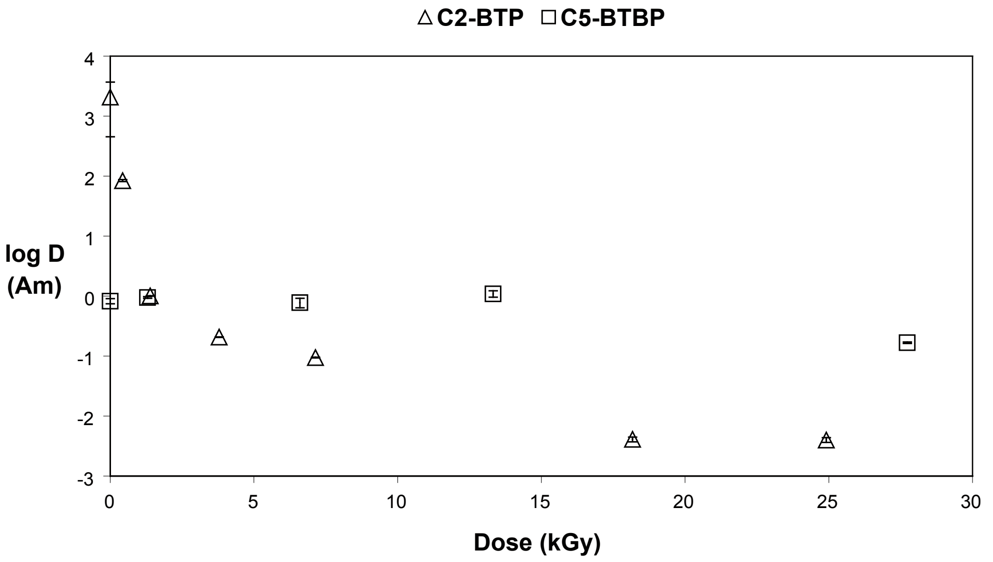


Figure 3-9. *Log D versus received dose for two different irradiated organic systems. The two systems contained 0.0018 M C2-BTP and 0.005 M C5-BTBP, respectively, both dissolved in hexanol and irradiated in a* ^{60}*Co source.*

3.2.2 Back-extraction

Back-extraction (or stripping) of a system is another aspect of the separation process that has been studied. In a working separation process the extracted metals should not form too strong complexes with the organic phase but be possible to back-extract to an aqueous phase, hence enabling reuse of the organic phase. The stripping of Am and Eu from different organic systems with glycolic acid as the stripping agent has been studied /Dubois et al. 2007/ as well as the stripping of Am with oxalic acid /Andersson et al. 2004/.

3.2.3 Lanthanide chemistry and nitrate complex formation

The basic extraction behaviour of lanthanides and the nitrate complex formation for some of them was investigated /Andersson et al. 2006a/. The organic system used consisted of 2,6-bis-(benzoxazolyl)-4-dodecyloxylpyridine (BODO) and 2-bromodecanoic acid (HA) in tert-butyl benzene (TBB) and the aqueous phase consisted of either 1 M perchlorate ion concentration (0.01 M H^+) or 5 M total ionic strength (0.01 M nitrate, 0.01M H^+ and the rest perchlorate). Trace activities of several lanthanides were used to spike the solutions. The stability constants of nitrate complex formation along the lanthanide series were estimated using a solvent extraction technique. The extraction of BODO/HA at low nitrate ion concentration showed a decreasing trend with increasing atomic number. A difference in the distribution ratio with respect to the ionic radius was seen in comparison of the light and the heavy lanthanides. A clear tetrad effect could also been seen along the series. Various extraction experiments with a series of lanthanides were also carried out for C5-BTBP /Nilsson 2005/.

3.2.4 Berkelium chemistry

The berkelium chemistry experiments were carried out at Lawrence Berkeley National Laboratory in Berkeley, USA. The berkelium was provided by Oak Ridge National Laboratory and it was trivalent under the chosen experimental conditions.

The experiments can be divided into two groups, namely general extraction properties of some nitrogen containing extractants (2,2':6',2-terpyridine (TERPY), BODO, C5- and CyMe$_4$-BTP) /Andersson et al. 2004/ and determination of stability constants for nitrate complex formation.

The stability constants for nitrate complex formation with trivalent berkelium were determined for the first time /Andersson et al. 2006b/. Comparison with other existing results for Am and Cm showed increasing stability constants with increasing atomic number. The trend was found to be similar with the one found for other complexants, such as thiocyanate and fluoride.

3.2.5 Process calculation program

A program for simulation of extraction in mixer-settlers has been developed and it has the ability to simulate several mixer settler batteries connected to a larger process. The basic idea of this program is that small volume elements are moved through the system in small time steps. New concentrations and flows are collected as soon as a volume is moved in a new part of the system. This process continues until a user decides that stop time is reached. The most powerful advantage with the programme is the possibility to acquire data of concentrations continuously and not necessarily at equilibrium and also the simulation can be made up to 20 different solutes in one single run. This makes it possible to study the kinetics of the system too. Changes in flow rates towards equilibrium can also be investigated /Andersson et al. 2004/.

3.2.6 Micro extraction device

A miniaturized system has been developed for performing fast and continuous liquid-liquid extraction. The apparatus consists of a micromixer with interdigital channels for intense mixing of the phases. The emulsion is subsequently fed into one to three filter units (teflon membranes with 0.5 µm pore size) for phase separation. The extraction device has an inner volume of 20 µL and a hold-up time of about 1 s at a flow rate of 60 mL/h. The micro scale equipment for liquid-liquid extraction is of basic science interest but also for industrial scale applications, when the extractant is available just in very small amounts /Andersson et al. 2004/.

3.2.7 Extraction with calixarenes

Calixarenes are a type of molecules primarily designed for separating caesium from high-level waste, but the possibility to use them for other types of separation has also been investigated. The name calixarene derives from calix, because the molecule resembles a vase, and from arene, referring to the aromatic building block. They have hydrophobic cavities that can hold small molecules or ions. The calixarenes studied during the past years are one wide rim and one narrow rim tetra-CMPO calix[4]arene. Figure 3-10 shows the chemical structure of the two molecules.

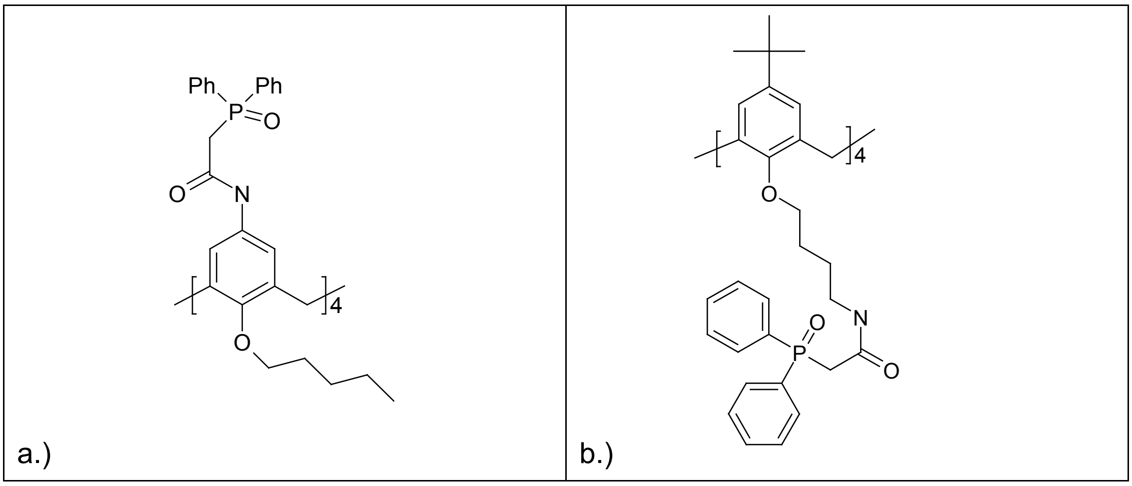


Figure 3-10. *Chemical structure of wide rim tetra-CMPO calix[4]arene (a) and narrow rim tetra-CMPO calix[4]arene (b).*

Basic extraction properties

The extracting properties of the calixarenes were investigated in a similar way as described above for the BTBP and BTP molecules. Experiments have included nitric acid concentration dependence, ligand concentration dependence and stability towards radiolysis etcetera /Dubois et al. 2007, Andersson et al. 2005/.

Systems with COSAN

COSANs (CObalt SANdwich) is another type of complexing agents used for caesium extraction. A study was made where the extracting properties towards Th, U, Np and Am were investigated for systems containing both a CMPO calix[4]arene and COSAN or chloroprotected COSAN /Dubois et al. 2007/.

3.3 Uppsala university – neutron research

The nuclear reactions group of the Department of Neutron Research, Uppsala University, is primarily active in measurements of cross sections of neutron-induced nuclear reactions. The experimental work is primarily undertaken at The Svedberg Laboratory (TSL) in Uppsala, where the group is operating two world-unique instruments, Medley and Scandal.

Two research projects supported by major Swedish nuclear energy actors in a wide sense, including SKB and SKI, have been carried out during the last few years. The project Natt, Neutron data for Accelerator-driven Transmutation Technology (2002-06), was focused on neutron elastic scattering data for ADS applications. The project Next, Neutron EXperiments for Transmutation (2006–2010), is dealing with fission and charged-particle production.

Both these Swedish projects have been closely linked to relevant EU projects. The FP5 project Hindas (High and Intermediate energy Nuclear Data for Accelerator-driven Systems) was carried out during 2000–2003 and comprised 16 universities or laboratories in Europe. Six experimental facilities were used to optimize the use of existing infrastructure /Hindas/. There was a strong Swedish participation in Hindas. The department of neutron research at Uppsala University lead one work package, and 5 out of the 16 Hindas partners performed experimental work at TSL.

In the present FP6 project Eurotrans (2004-08), UU participates with a joint Swedish-French experiment on neutron inelastic scattering, for which world-unique experimental equipment has been developed.

Analysis and documentation have 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 work has underestimated the probability for neutron scattering at 96 MeV 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.

Compelling evidence of the existence of three-body forces in nuclei has been obtained. The first publication on these matters from the group turned out to qualify on the top-ten downloading list of Physics Letters B, one of the very most prestigious journals in subatomic physics.

A new neutron beam facility with significantly improved performance has recently been built and commissioned at TSL. With this new facility, competitive equipment for research in 5–10 years from now is available.

During the last three years, five PhD theses with data from TSL have been successfully defended.

Several ISTC projects have been carried out at TSL in close collaboration with the INF group. This has matured to involve joint experimental work also at other facilities. For instance, a joint fission experiment is scheduled to take place in Louvain-la-Neuve, Belgium in June 2007.

Recently, the group has been active in the creation of two new EU projects. The EU project Candide (Coordination Action on Nuclear Data for Industrial Development in Europe) started January 1, 2007. The primary aim of the project is a nuclear data assessment for future reactors for waste incineration, i.e. accelerator-driven systems and Gen-IV type reactors. An important aspect of the project is that up to now, all nuclear data assessments have been targeting either critical or sub-critical system, whilst Candide attempts at a comparison on an equal footing. Jan Blomgren, INF, is project coordinator.

A network of European facilities for nuclear data measurements has been established via the EU integrated infrastructure initiative Efnudat (European Facilities for Nuclear Data Measurements). The neutron beam facility at TSL is one of nine facilities involved. See also the annual reports from the Uppsala group /Blomgren et al. 2004, 2005, 2006/.

4 Areas for further R&D on P&T

On an international or European scale the further development, permitting demonstration of P&T in pilot scale appears a reasonable objective for the coming 10–15 years. In practise, this corresponds to

1. Design and construction of fast neutron spectrum irradiation facilities, critical or sub-critical, using sodium, lead-bismuth or helium gas as coolant.
2. Performance assessment of one or two selected processes for minor actinide separation on the scale of ten kilograms per year.
3. Design and construction of a facility for fabrication of 10–20 tons of fast reactor MOX fuel, for loading into the fast spectrum irradiation facilities.

In addition, R&D on structural materials and minor actinide bearing fuels should be performed in parallel, to improve performance, increase safety margins and reduce costs for later implementation on industrial scale of P&T.

4.1 Transmutation in sub-critical reactors

In the 6th FP integrated project Eurotrans, the main design options for an experimental accelerator driven system (XT-ADS) with MOX fuel, lead-bismuth coolant and a thermal power of 60 MW have been determined, as described in section 2.4 above. During the second half of this project, the detailed design of the fuel elements, core, primary and secondary system will be fixed, including instrumentation, control and decay heat removal system. Reliability tests of accelerator components will be made and accident analysis for a number of postulated transients will be performed. In conjunction with assessments of the source term (release of radioactive elements following clad failure events) and containment function, this will lead to a preliminary safety analysis (PSAR) and licensing report for XT-ADS. Areas of particular significance for safety performance that were identified will be studied in more detail, such as:

1. *Possible consequences of steam generator tube rupture.* In order to take advantage of the absence of chemical reactions between lead-bismuth and water, the steam generator of XT-ADS has been located in the primary system, avoiding the use of an intermediate heat exchanger. Therefore, a steam generator tube rupture (SGTR) will lead to the introduction of water into the molten-lead bismuth. Rapid heating and steam bubble formation may then result in sloshing of the coolant followed by mechanical impact on primary circuit structures. Another consequence of relevance for safety is the potential steam bubble transport into the core, leading to reduced coolability, and in certain cases to positive reactivity insertions. It is therefore considered of high relevance to conduct a near-term research programme aimed on one hand at increased understanding of steam bubble formation mechanisms in lead, and on the other hand on asserting that the actual design of XT-ADS and other lead-cooled systems presently under development will not be vulnerable to SGTR events. Here, Sweden has the capacity to make important contributions with modelling oriented experiments in the TALL facility at KTH (see Chapter 3).
2. *Fuel failure detection methods.* In existing reactors, fuel cladding ruptures are detected by detection of delayed neutron emission from volatile iodine. In the preliminary analysis of the source term for XT-ADS performed by KTH, it was found that LBE is highly likely to retain any release of iodine from the fuel in the form of lead iodide. While this is highly beneficial for reduction of the source term, it will complicate the detection of failed fuel claddings. If fuel clad ruptures cannot be detected early, the core might have to be designed to cope with

the impact on coolability and reactivity caused by fission gas release, with potential negative impact on performance. A reliable system for fast detection of clad failures will therefore have to be designed on other grounds than release of iodine into the cover gas.

3. *Thermal deformation of core support grid.* The major design options for XT-ADS include a layout where fuel elements are loaded from the bottom, being locked to an upper core support grid by buoyancy forces. While this permits to simplify the fuel loading procedure in the presence of an off-centred spallation target loop, it results in a temperature gradient between inner and outer parts of the diagrid, causing bowing of the grid. The concomitant variations in geometry of the core during transients require a more sophisticated approach than usual for simulation of both thermal hydraulic and neutronic feedback.

4. *ADS-specific accidents.* A scenario specific to accelerator driven systems is the event when the accelerator is accidentally turned on after pumps have been tripped. The system thus should be designed to remove full power during the time requested for it to function without active response from operators (30 minutes), relying only on natural circulation. So far, XT-ADS was configured to respect the rapid creep limit of the reference T91 clad for fully developed natural circulation. Until natural convection has been fully established, there may however be a state of temporary undercooling. In sodium cooled reactors, a fly-wheel may be attached to pumps in order to increase their inertia, thereby preventing undershoot. In heavy liquid metal cooled systems, fly-wheels are however considerably less efficient. Therefore, other types of design measures would have to be conceived to ensure clad integrity during this type of accident. The TALL loop at KTH is today a unique device in Europe for determining the time to establish natural convection of lead-bismuth, and may therefore be instrumental for this analysis.

Within the Eurotrans project, the Ecats domain originally included experiments on coupling and dynamic feedback between a cyclotron driven neutron source and a Triga core at power in Rome. As the construction of the TRADE facility was cancelled due to lack of domestic support, a new zero-power coupling experiment has been launched at SCK-CEN in Mol under the acronym Guinivere (Generator of Uninterrupted Intense NEutrons at the lead VEnus REactor). It consists of a continuous beam version of the Genepi D-T neutron generator used for Muse in the 5th framework programme, coupled to a fast neutron core with 30% enriched UOX fuel and simulated lead coolant. In the Guinivere experiment, methods for determining power/beam current relation will be validated (with participation of KTH). The Belgian authority will also be given the opportunity to gain competence in licensing a fast neutron (zero-power) facility.

Although the research program intended to be implemented in Guinivere is of relevance for the development of XT-ADS, there is an evident lack of test facilities for coupling between a proton driven neutron source and a fast neutron core at sufficient power to test thermal feedback effects.

4.2 Fuels

In the 5th and 6th Framework Programmes, fabrication techniques for inert matrix fuels were developed which enabled to successfully manufacture uranium free solid solution nitride fuels (without americium) and composite oxide fuels (with americium), as described in Chapter 2. During spring 2007, these fuels will be loaded into the Phenix reactor in France and the High Flux Reactor in the Netherlands for irradiation testing. Results from post irradiation examination will become available in 2008 for the nitride fuels, and 2009–2010 for the composite oxide fuels. In addition, americiums bearing metallic alloy fuels manufactured in the United States are included in the Phenix irradiation experiment. The in-pile performance of the irradiated fuels will then be used to update thermo-mechanical simulation codes, which then may predict their behaviour at higher burnup and linear rating.

One of the major differences between a fuel dedicated to transmutation and a conventional fuel is the high level of helium production resulting from in-pile decay of ^{242}Cm. The impact of helium on fuel performance so far remains uncertain. Examination of fuels from the SUPER-FACT, T4, NIMPHE and BORA-BORA experiments indicates that helium release is higher than for fission gases (Xe, Kr), but still considerably less than 100%. It has been difficult to identify any consistent trend with respect to temperature and irradiation. If helium is retained in the fuel, contrary to present modelling assumptions, it would have an impact on predicted swelling rates and would also increase the degradation of gap conductance with burnup. In FP7, focus in modelling of fuel evolution will therefore be directed towards a better understanding of helium migration in actinide ceramic phases, as well as in the inert matrices of interest. This can be made by so called multi-scale modelling efforts, where a combination of electronic structure calculations on nanometre scale is combined with kinetic Monte Carlo simulations on the micro scale of grains.

So far, very little experience exists on the fabrication and irradiation performance of curium containing fuels. This lack is planned to be addressed in the seventh framework programme, by fabrication of oxide fuels containing a few grams of curium in the Atalante laboratory in France. A major challenge when considering application on industrial scale is the handling and management of curium, which simultaneously releases large amounts of heat (2.8 Watts per gram) and spontaneous fission neutrons ($1 \cdot 10^7$ neutrons per second and gram). Conventional methods for reprocessing and fuel fabrication therefore become impossible to apply /Pillon et al. 2003/. The high intrinsic neutron source makes remote handling mandatory in heavily shielded hot cells. In order to avoid excessive maintenance costs, robust and simplified manufacturing processes should therefore be devised for curium containing fuels. In particular, one should avoid powder metallurgy. Solutions could include infiltration of pellets or microspheres with a solution of curium nitrates, according to schemes developed at ITU /Fernandez et al. 2003/. An additional simplification, avoiding the step of pellet pressing, could be realised if fuels pins are filled directly with the microspheres using the vibropack method currently used for fabrication of MOX fuel for BOR-60 in Russia.

A difficulty encountered in the fabrication of nitride fuels is the high vapour pressure of americium nitride, leading to loss of material during sintering /Jolkkonen 2004/. Consequently, nitrogen gas should be used during the sintering stage, and the sintering temperature should be kept below 1,800°K /Jolkkonen et al. 2004/. These countermeasures however lead to a relatively high porosity in the end product. Methods improving the ability to sinter americium bearing nitrides should therefore be developed. One potential candidate is the use of electromagnetic vortex blending of the initial powders with rotating needles. This method is developed by the Bochvar institute in Moscow, and has enabled to reduce the residual porosity after sintering of (Pu,Zr)N pellets to levels below 10% /Skupov et al. 2006/.

The application of composite structures to improve the thermal conductivity of oxide fuels comes with a potential drawback in terms of eutectic melting at temperatures much below the melting points of the constituent materials. Measurements of eutectic melting on americium containing compounds are difficult to perform, and often only indirect evidence can be obtained. Recent measurements of americium vapour pressure over samples of $(Pu_{0.8},Am_{0.2})O_2$-Mo fabricated at ITU indicated that eutectic melting occur at 2,400–2,500°K /Staicu et al. 2006/. An improved understanding of basic processes involved in eutectic melting and other processes occurring at high temperature is of high value, not only for determining fabrication and irradiation parameters, but also for purposes of transient safety analysis. The facilities able to test nuclear fuels at very high temperature are however very few (mainly ITU in Germany). Therefore, KTH has submitted an application to the Swedish science council for construction of an advanced reactor materials laboratory (Karem). It is intended that the special competence of this laboratory would be the study of high temperature properties of nuclear fuels, including interactions with clad and coolant. A planning grant was awarded for 2007, and an application for funding of construction of the laboratory will tentatively be submitted in August this year.

4.3 Materials

Ferritic-martensitic (nickel free) steels were selected as the primary candidate for application in lead or LBE cooled transmutation systems, due to the combination of a low solubility in lead and a high resistance to radiation induced swelling. The 12% Cr steels developed for Soviet LBE cooled submarine reactors however suffer from severe embrittlement after irradiation by fast neutrons. As it is known that reduction of Cr content to 9% minimises the brittle-ductile transition temperature, the major effort in Europe has been focused on this class of steels.

In the fifth framework programme, fast spectrum irradiation of industrial 9Cr-1Mo steels (EM10, T91), experimental 9Cr-2W steels and the 9Cr-1W steel developed within the fusion programme were performed in BOR-60 up to a dose of 40 dpa. The best performance was found for 9Cr-2W steels, which however have no general industrial application today, and therefore are not available in large amounts.

The reduction of Cr and Si content (as compared to the Soviet submarine steels) might however lead to increased corrosion rates. Therefore the development of adequate corrosion protections methods has a high priority. A large number of LBE test loops have been constructed in Europe (including KTH). Extensive testing and optimisation of oxygen control have shown that is is possible to build and maintain a protective oxide layer on the surface of 9Cr steels up to a temperature of 820°K (550°C) for times extending to at least 6,000 hours.

The experience collected in irradiation experiments and corrosion test loops permitted the construction and operation of the Megapie spallation target at PSI in Switzerland during fall 2006. During four months this spallation target, constructed out of T91 steel, operated at a power of 800 kW with a reliability of 95%, which is considered to be a great success. The Megapie experiment thus has proven the principal applicability of liquid heavy metal spallation targets for high intensity neutron sources. A Megapie-II experiment is now under discussion, where the target design would be optimised to meet requirements for application in ADS or the European spallation source (ESS).

While oxygen control permits operation in LBE and lead up to 820°K under nominal conditions, other methods would have to be applied to increase the operating temperature, or to ensure integrity of the materials under transient conditions. At FZK, the so called GESA method for surface alloying is under development. The main principle is the spraying of Fe-Al vapour on steel surfaces, followed by electron beam polishing. This technique (in combination with oxygen control) is able to protect steel cladding tubes of both ferritic and austenitic type from corrosion attacks at temperatures up to 870°K (600°C) during tests lasting 6,000 hours /Müller et al. 2004/.

The stability of the surface alloy under irradiation conditions is tested in the fast neutron spectrum of Phenix during 2007/2008. The combined effect of irradiation and liquid LBE will be investigated in Belgian and Dutch materials test reactors. The test matrices of these experiments include oxide dispersion strengthened steels (ODS), holding a potential for improved mechanical properties at high temperature. Since high temperature creep is a major weakness of ferritic martensitic steels, it would also be of interest to validate the GESA technique for titanium stabilised austenitic steels like 15-15Ti. Such experiments might be performed in the seventh framework programme.

4.4 Partitioning

The art of separation for transmutation purposes has during the last years reached an important milestones in the research. Both the hydrometallurgy and the pyrometallurgy have taken rather big steps towards process development. In some cases it is even possible to get good separation between e.g. americium and curium. However there are many challenges still left.

Two pyrochemical methods have mainly been studied – the Fluoride Volatility Method (FVM) and electrodeposition.

FVM uses flame fluorination followed by condensation and rectification. This process can separate and recover uranium and other elements forming volatile fluorides. It is considered for reprocessing of spent oxide fuel. Here the functionality of the devices and the conditions of the process have been verified and studied. Future development along this path will follow in the close future and possibly also demonstration of feasibility.

In the field of electrodeposition from molten salts, the work has been focused on the development of a separation process for uranium considered for re-use and on methods for removal of undesirable fission products (with large neutron capture cross sections). Selected molten fluoride salts (e.g. LiF-NaF-KF, LiF-CaF_2) have been studied in order to find proper conditions for the separations – composition of melts, electrode materials, settings (applied separation potentials or voltage, current densities). Based on the results reached so far, uranium should be possible to remove by electrodeposition from the mixture of some lanthanides and maybe also from some other actinides. The further work will be focussed on the improvement of selectivity as well as technical development. Process and equipment design is not simple due to the aggressive nature of the melts and the rather high temperatures needed.

In the case of hydrometallurgy high enough distribution ratios and separation factors have been achieved. Now the focus is shifting towards realising a working process with its concealed difficulties. The search for ligands that give good extraction and separation will continue but with much less emphasis. The research will be divided into two parts – basic research and applied process research. The emphasis for basic research will be on improving the radiolytic and hydrolytic stability of the selected ligands. This could be done either by improving the ligand stability, by using additives to the diluent, or by selecting a suitable diluent. For this one needs to understand the basic chemistry that affects the radiation damage.

Until now, the aim in hydrometallurgy has been to obtain the desired separation performance using about five minutes of contact time. In the future a useful performance also with shorter contact times may be needed depending on which devices will be used for the separation process. The kinetic behaviour is not only important for a selective extraction but also for the stripping of the extracted elements. It was shown that many of the ligands used for very selective and effective extraction reacted very slowly when stripping extracted elements from the organic phase. This can either be improved by addition of complexing ligands to the aqueous phase or by making modifications to the organic phase used, i.e. modifying the ligand or the diluent.

Research on process and equipment will be intensified. The use of mixer-settlers is not considered for the aqueous partitioning at the moment but the alternative is not clear. Columns have their problems so some new equipment will be discussed and analysed within the coming years. However, if the work reverts back to the known concepts maybe mixer-settlers are not totally out at the end.

The processing of transmutation fuel itself is an area that has received some attention lately and must be brought more and more in the focus of the research. Presently there is no final and detailed knowledge of the physical and chemical requirements on fuel for a transmutation device. Hence the development will have to proceed along some hypothetical lines. However, regardless of the composition of the fuel, it will still have to be fabricated, irradiated and then dissolved after use.

The knowledge about how to fabricate currently used fuels is rather good, but how do we fabricate future fuels from the elements separated in the separation process?

There are two major fuel types to consider when discussing the separation process. Dissolution of the common nuclear fuel originating from the power producing reactor and the dissolution of the fuel from the transmutation reactor will require different studies. In the future several different nuclear fuels are foreseen e.g. carbides and nitrides. They require different dissolution strategies to make the dissolution effective and compatible with the following separation. Since the transmutation reactor is not well defined there are also several possible fuels and burnup. Dissolution techniques will have to be developed for these fuels along similar principles and demands as for the normal spent fuel. An issue requiring special attention is criticality in the dissolver.

Once the transmutation operation and its fuel are defined, the conversion rates and final fuel composition calculated it will be possible to optimize the corresponding separation process with regards to selectivity and yields. Until then, rather general separation processes have to be studied able to accept a considerable range of input material. This is unfortunate from the point of view of finalising a separation process as wished in e.g. France. On the other hand it is very much in the line with the preferences of other EU countries like the UK and Germany where the decisions and time schedule for the transmutation option are not as mature as in France.

4.5 Nuclear data

In the realm of ADS, two classes of nuclear data are clearly discernable, above and below 20 MeV. Above 20 MeV, no previous nuclear energy applications have been developed, and consequently the database is meagre. During the last decade, the situation on proton- and neutron-induced production of charged particles has improved considerably, and presently the situation is satisfactory for the demands as long as the aim is to build a demonstrator or prototype system. If a future full-scale ADS plant for large-scale incineration were to be built, the situation would, however, probably need to be revisited due to the higher demands of a production facility.

Concerning neutron scattering, there is still room for considerable improvement. The present upgrades at TSL are dictated to fill these demands. It can be foreseen that a 5–10 year experimental campaign is required to reach a situation resembling that of charged particles.

Up to now, all experimental activities have been focused on cross section measurements. The natural next step would be to carry out integral experiments, i.e. an experiment where the quality of the entire data library is assessed. This could for instance be measurements of neutron transmission through large blocks of various materials. Only one such experiment has been performed worldwide (in Japan), but not at the most interesting materials for ADS. TSL is well suited for such experiments, and it is conceivable that such integral experiments could be important in FP7.

At lower energies, the nuclear data situation is fundamentally different. The development of critical reactors has motivated large efforts in data production and therefore the present work is dedicated to filling important gaps in the literature. In general, the nuclear data status is satisfactory for uranium and plutonium, whilst there is room for improvement on the minor actinides (neptunium, americium and curium). At present and in the near future, the activities on americium dominate. This is due to two factors. First, americium is the nucleus that has the largest deficiencies in the nuclear data bases and second, it is probably the element where incineration in ADS is best motivated.

Nuclear data activities at lower energies could be expected to grow in a near future, because of the interest in Gen-IV reactor systems. The nuclear data required for development of Gen-IV are more or less identical with those needed for ADS.

If realization of a full-scale ADS or Gen-IV system would be carried out, another nuclear data activity might be motivated. The nuclear data on the most important elements, i.e. uranium and plutonium, were often produced thirty years ago or more. It is not unlikely that some of the key nuclear data in the adopted databases suffer from systematic errors. This might motivate some of these data to be revisited, taking advantage of the development of novel experimental techniques in the recent years.

5 Short update of assessments in 2004 report

In the previous status report made for SKB in 2004 /Ahlström et al. 2004/ an assessment of the prospects for P&T developments was made. The assessment covered the following aspects:

- Key R&D items.
- Key safety issues for.
  - ADS transmutation.
  - Partitioning.
  - Radiological protection.
  - Proliferation.
- ADS demonstration plant.
- Other demonstration facilities.
- Time and costs.
- Impact on waste management programme and nuclear power development.
- Acceptance.

These aspects were discussed at some length in a separate chapter. The progress made since 2004 does not change the main general content of this assessment and thus there is little need to cover all these aspects in the current report.

Based on the aforementioned assessment some conclusions concerning the P&T status were made in Chapter 6 of the 2004 report. It may be prudent to revisit and update these conclusions. In this context some of the 2004 items of the assessment are also updated or modified in the light of recent developments.

5.1 General

The gradual increase in R&D efforts on partitioning and transmutation (P&T) that occurred internationally during the period 1990–2003 has not continued after 2003. The financial support by the European Commission to P&T-projects within EU has levelled off and it is unclear how much support that will really be available in future programmes. Within the commission the expectations seem to be an unchanged level during the foreseeable future.

Research on P&T has still 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. P&T is more and more seen as an interesting option in future nuclear power systems based on advanced reactor designs, advanced nuclear fuel and advanced nuclear fuel cycles. The interest to pursue P&T for a nuclear "phase-out" programme is more or less nonexistent.

5.2 Impact on nuclear energy and waste programme

Successful development of P&T within an advanced nuclear fuel cycle 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. A recent NEA-study /NEA 2006a/ indicates for example that the volume requirements in a repository for high level waste (per unit of electricity produced) may be of 5 to 20 times more for electricity from light water reactors with once through fuel cycle than from advanced reactors with advanced fuel cycle processes. On the other hand the amounts of long-lived low and intermediate level wastes will be larger for fuel cycles with recycling of transuranium elements /NEA 2006a pp 67–70/.

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 produce a substantial part of the electricity by nuclear power.

Competence developed in research on advanced fuel cycles and waste management strategies including P&T 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 more than 100 years.

5.3 Acceptance

Advanced fuel cycles and waste management strategies including P&T on an industrial scale require large nuclear facilities that must be accepted and trusted by society. As such strategies involve very long lead times the trust and acceptance requirements become very strong. This means that the benefits must be perceived as very large and important.

5.4 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.

A feasibility and systems study of the partitioning processes covering technical but also proliferation, cost and safety aspects might be useful for guiding the further development in this area.

5.5 Transmutation

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

The main interest for transmutation seems to shift back on critical fast reactors. For some years it was largely focused on accelerator-driven systems (ADS). These are more and more seen as an interim option for burning minor actinides awaiting the full deployment of fast reactors as the main nuclear energy production device.

The basic technical components of ADS have not yet been fully defined. The currently preferred choices for the European demonstration plant EFIT studied within the EU-programme is lead as primary coolant and for the spallation target, a linear accelerator and composite oxide fuel, with MgO or metallic Mo as matrix. Other options are however still studied in particular concerning the fuel.

Some years ago both US and European studies concluded that a basic R&D programme of the order six years and costing several hundred million Euros was 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 however not reached the level presumed. 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 thus still 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. France has announced the intent to construct a modern sodium-cooled fast reactor before 2020.

Both ADS 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.

5.6 Safety and radiation protection

The implementation and construction of future advanced nuclear fuel cycle (including P&T) facilities must meet the same requirements from environmental and nuclear laws as other nuclear facilities.

The development of such facilities 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.

5.7 Time and costs

As is indicated 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. New assessments that have been published during recent years are all based on the same bases as those presented in the previous status report /Ahlström et al. 2004/. The new results do not change the picture presented there.

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 a 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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KASAM's reports are printed in the Swedish government's series SOUyy:nn. They are usually available in pdf-format on KASAM's web-site *www.sou.gov.se/kasam/*.

Appendix 1

Reference group for P&T research and development

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