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Partitioning and transmutation Current developments – 2010

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

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January 2010

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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 authority, SSM (Swedish Radiation Safety Authority) including its predecessors SKI (Swedish Nuclear Power Inspectorate) and SSI (Swedish Radiation Protection Institute), as well as from Kärnavfallsrådet (Swedish National Council for Nuclear Waste), including its predecessor 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 production of more plutonium and other transuranium elements is desired. 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 do not, however, 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.

The field experienced renewed interest through the 1990ies, however strongly focused on accelerator-driven hybrid systems (ADS) for incineration of spent nuclear fuel. The driving political rationale for this interest was that ADS was seen as a politically acceptable research topic in countries with a negative political nuclear landscape. Hence, it could serve as a training ground for future nuclear technology also in a hostile environment. Because ADS was the essentially only activity in the field for a period of more than a decade, the concept P&T became synonymous with ADS in the public debate. Recently, the interest in fast critical reactors for incineration of spent nuclear fuel has

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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.

re-appeared on the scene. For instance, the joint European interest organization SNETP (Sustainable Nuclear Energy Technology Platform) has issued a strategic research agenda in which the construction and commissioning of a fast reactor for transmutation is the highest priority, and design of such a reactor is in progress. Thus, right now the interest in transmutation in critical reactors is larger than in ADS, and the research in ADS is to a large degree directed towards areas that are not technology-specific, i.e. where the results are useful regardless of whether critical or sub-critical accelerator-driven systems are employed.

In Europe the research has been 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 2004 /Ahlström et al. 2004/, and a third similar report was issued in 2007 /Ahlström et al. 2007/. The present report summarises the progress in the field through the years 2007–2009.

SKB has been the main sponsor of P&T research in Sweden since 1995 until the end of 2009. In October 2009, VR (Vetenskapsrådet, the Swedish Research Council) granted 36 MSEK to the GENIUS project (Generation-IV in Universities in Sweden), which is a project targeting next generation critical reactors. The first anticipated use of such reactors is, however, P&T of existing spent fuel from the present fleet of light-water reactors. Thereby, the activity level has increased dramatically just before this report was issued.

System studies of partitioning and transmutation

The previous status report /Ahlström et al. 2007/ 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.

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.

In addition, a number of transmutation-supporting projects are in progress. Each such project is far smaller than EUROTRANS, but in total they comprise a research volume comparable to EUROTRANS.

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[2] LBE = Liquid Bismuth Eutecticum.

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 ACSEPT (Actinide Recycling by Separation and Transmutation) that started in 2008 and shall be completed in 2012. The project has an overall budget of 23.8 M€, whereof the EC contributes 9.0 M€. The project is a so-called collaborative project within the seventh framework programme sponsored by the European commission. It constitutes a continuation of the project EUROPART within the previous framework programme. ACSEPT is divided into four domains:

1. Hydrochemistry
2. Pyrochemistry
3. Process development
4. Training

Sweden is represented by the nuclear chemistry group at Chalmers in Göteborg.

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 political situation regarding nuclear power has changed dramatically during the last three years, in Sweden, Europe and the World. The Swedish government has announced a change in the law, allowing new nuclear power reactors to be built. There is presently strong pro-nuclear majority in the European Parliament. The new German government has indicated the possibility that the operational time of the existing fleet can be prolonged. The new administration in the USA has halted some processes related to the repository in Yucca Mountain, because a policy change towards plutonium recycling is considered. All these changes are obviously of importance to P&T research and development.

Recently, the largest funding to research on new nuclear reactor technology in about 30 years has been granted by Vetenskapsrådet, responding to a direct request from the government. Thus, for the first time after the 1980 referendum, there is significant public funding to P&T research available in Sweden. SKB no longer is the sole financer; in fact, it is no longer the largest.

The gradual increase of the EC 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. Within the commission the expectations seem to be an unchanged level during the foreseeable future to ADS research, whereas research on fast reactors might increase.

It should be pointed out that the EC funding depends on the political situation in countries with strongly negative political opinion on nuclear power. As long as P&T research was synonymous with ADS, also nuclear-negative countries were willing to support it, because it was perceived as a cleaning-up activity, and not a way to continue nuclear power. With the recently increased interest in fast reactors for plutonium transmutation, and the associated prospect of increased reliance on nuclear energy, nuclear-negative countries might have problems to support increases in the P&T funding. Partly this effect can already be seen, with a large increase in the EC fusion research budget (an area still far from industrial deployment), and a more or less constant P&T funding.

If the political situation regarding the EC nuclear research funding is not changed, there is a significant possibility that countries with an interest to pursue fast reactor development (primarily France) will go ahead without EC support. Obviously, this would diminish the weight of the EC policy in this realm.

The research on P&T has 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 has been limited, but is now increasing, primarily because of the increased attention to fast critical reactors. 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.

P&T is no longer seen as a method to “clean up” as part of a nuclear phase-out; it is rather viewed as an integral part of a sustainable nuclear energy system, in which fast reactors play the major role in handling plutonium, and incineration of minor actinides by ADS is performed to reduce the radiotoxicity of the wastes from fast reactors. If ADS should be used at all, it seems today as there is close to global consensus that a double-strata concept is the most likely option.

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. It may also decrease the required volumes of high-level waste in the repositories (volumes of low- and intermediate-level waste will, on the other hand, tend to increase as a result of the partitioning processes).

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.

From a Swedish perspective it is important to participate in the international development and maintain a reasonable level of competence within the country. 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.

Recently, a generation change has taken place at the Swedish university research groups active in nuclear-power related research, and presently the activities grow rapidly, both due to increased interest in research and a larger need for education. The leading scientists in the new generation have all of them worked in projects supported by SKB and SKC, and most of them have been involved in P&T research. Thereby, the P&T research has already played a crucial role in the Swedish nuclear competence management.

Sammanfattning

Forskning och utveckling av metoder för separation och transmutation (S&T[3]) 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 avskiljer idag inte de övriga transuranerna[4] – neptunium[5], 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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[3] 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.

[4] 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.

[5] 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 i västvärlden under det tidiga 1980-talet försvann intresset för S&T mer eller mindre.

Intresset förnyades under 1990-talet, huvudsakligen inriktat mot acceleratordrivna system (ADS) för omvandling av långlivade ämnen i använt kärnkraftsbränsle. Den drivande politiska kraften bakom denna fokusering var att ADS sågs som en politiskt acceptabel forskningsverksamhet i länder med kärnkraftsfientlig politik. Därmed kunde ADS fungera som kompetenssäkring och -utveckling i kärnkraftsskeptiska länder. Eftersom ADS var i stort sett allenarådande vid denna tid blev begreppet P&T närmast synonymt med ADS i den allmänna debatten. De senaste åren har intresset för snabbreaktorer för transmutation återkommit. Ett exempel på detta är att den gemensamma europeiska intresseorganisationen SNETP (Sustainable Nuclear Energy Technology Platform) har antagit en strategisk forskningsplan i vilken utveckling och byggnation av en natriumkyld snabbreaktor för transmutation har givits högsta prioritet, och design av en sådan reaktor är i full gång. Därmed är intresset för snabba reaktorer nu större än för ADS, och forskningen inom ADS har i stor utsträckning inriktats mot frågor av icke teknologispecifik karaktär, dvs områden där resultaten är användbara inom såväl kritiska som acceleratordrivna system.

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/, och en tredje likartad rapport sammanställdes 2007 /Ahlström et al. 2007/. Den nu föreliggande rapporten sammanfattar den utveckling som skett under åren 2007–2009.

SKB har varit den huvudsakliga finansiären av forskning inom P&T i Sverige från 1995 fram till slutet av 2009. I oktober 2009 beviljade Vetenskapsrådet 36 MSEK till forskningsprojektet GENIUS (GenerationIV-forskning i UniversitetsSverige), vilket handlar om forskning kring nästa generation kritiska reaktorer. Den första tillämpningen av sådana reaktorer bedöms allmänt vara P&T av använt bränsle från dagens kärnkraft. I och med VRs satsning har alltså den svenska aktiviteten inom P&T ökat rejält just innan denna rapport färdigställdes.

Studier av system för separation och transmutation

I den förra lägesrapporten /Ahlström et al. 2007/ 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.

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. Förutom EUROTRANS pågår ett antal mindre forskningsprojekt där olika delfrågor studeras. Vart och ett är betydligt mindre än EUROTRANS, men sammantaget utgör de en jämförbar forskningsvolym.

Separation

Forskningen inom separation är inriktad mot två huvudlinjer – vattenkemiska processer och pyrokemiska processer. I Europa samlas insatserna inom EU-projektet ACSEPT (Actinide Recycling by Separation and Transmutation) som startade 2008 och avslutas 2012. Detta är ett s k kollaborativt projekt inom det sjunde ramprogrammet och utgör en fortsättning på projektet EUROPART inom det föregående ramprogrammet. Projektet har en total budget om 23.8 M€, varav EC tillskjuter 9.0 M€. ACSEPT är uppdelat på fyra domäner:

1. Hydrometallurgi (även benämt vattenkemi)
2. Pyrometallurgi (även benämnt pyrokemi)
3. Processutveckling
4. Utbildning och träning

Från Sverige medverkar kärnkemigruppen vid Chalmers.

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 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 politiska situationen för kärnkraft har ändrats dramatiskt under de tre senaste åren, i Sverige, Europa och Världen. Den svenska regeringen har annonserat en lagändring som möjliggör nybyggnation av kärnkraftverk. På det europeiska planet kan konstateras att den nya tyska regeringen har ställt i utsikt förlängd drifttid av befintliga kärnkraftverk. Såväl EU-parlamentet som kommissionen är idag i huvudsak kärnkraftspositiva. Den nya administrationen i USA har lagt vissa processer relaterade till slutförvaret i Yucca Mountain på is, detta på grund av att en policyändring till förmån för upparbetning övervägs. Alla dessa förändringar påverkar förutsättningarna för forskning och utveckling inom P&T av uppenbara skäl.

Nyligen har VR beviljat det i särklass största forskningsanslaget på 30 år till ny kärnteknik i Sverige, detta som en följd av en direkt instruktion från regeringen. Därmed finns för första gången efter folkomröstningen 1980 avsevärda statliga medel till forskning inom P&T i landet. SKB är därmed inte längre ensam finansiär inom området; SKB är faktiskt inte heller längre störst.

Den gradvisa ökning av EUs insatser 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 till ADS-forskning, medan medlen till forskning om snabba reaktorer skulle kunna öka.

Det bör poängteras att EUs finansiering beror starkt på den politiska situationen i länder med starkt negativ kärnkraftsopinion. Så länge S&T sågs som synonymt med ADS var även kärnkraftsskeptiska länder villiga att stödja detta i måttlig utsträckning, eftersom detta sågs som en ”uppstädning” snarare än en väg till fortsatt kärnkraftsanvändning. Med det idag kraftigt ökade intresset för snabbreaktorer för plutoniumtransmutation och de tillhörande möjligheterna till ökad användning av kärnenergi kan man tänka sig att kärnkraftsnegativa länder blir mindre intresserade att stödja forskning inom S&T. En sådan effekt kan redan ses, med en kraftigt ökad EU-finansiering av fusion (där vägen till eventuell industriell användning är mycket lång), och mer eller mindre konstant finansiering av S&T.

Om det politiska läget rörande EUs finansiering inte ändras finns en betydande risk att europeiska länder med intresse att utveckla snabbreaktorer (i första hand Frankrike) går vidare utan EU-stöd. Detta skulle naturligtvis försvaga EUs roll i sammanhanget.

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 har varit mer begränsat, men ökar nu, huvudsakligen på grund av den ökande uppmärksamheten kring snabbreaktorer. S&T ses mer som en intressant option inom framtida energisystem baserade på avancerade kärnreaktorer, avancerat kärnbränsle och avancerade kärnbränslecykler.

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. Det kan även minska behovet av förvarsvolym för högaktivt avfall (För låg- och medelaktivt avfall torde däremot volymerna öka på grund av separationsprocesserna).

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.

För svensk del är det viktigt att delta i 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.

Nyligen har de svenska universiteten genomgått en generationsväxling inom kärnteknik, och för närvarande ökar verksamheten snabbt, både på grund av ökat forskningsintresse och ökade utbildningsbehov. De ledande forskarna i den nya generationen har alla etablerat sig genom projekt stödda av SKB och SKC, och de flesta av dem har deltagit i S&T-forskning. Därmed har S&T-forskning redan spelat en avgörande roll för den svenska kärntekniska kompetensen.

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 do not, however, separate today the heavier transuranium elements – neptunium[6], 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 minor 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 in the Western world 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 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.

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[6] 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.

The programme in Sweden has until very recently mainly been 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 have been 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 6.5 MSEK/year from SKB, about 12 MSEK/year from VR, and 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 2-2.

Earlier reviews of the status of the efforts concerning P&T were published by SKB in 1998 /Enarsson et al. 1998/, 2004 /Ahlström et al. 2004/ and 2007 /Ahlström et al. 2007/. Short reviews of the previous development have also been given in SKB's RD&D-programmes 95, 98, 2001, 2004 and 2007 /SKB 1995, 1998, 2001, 2004, 2007/.

This current status report summarises the work reported in the years 2007–2009 and tries to assess the prospects for future development of P&T as seen from a Swedish perspective. 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 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 report is structured in four main chapters. Chapter 2 gives a short account of recent international and foreign work concerning P&T. Chapter 3 is devoted to presentation of some results from the Swedish work. Chapter 4 summarizes 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 2007 status report /Ahlström et al. 2007/.

2 Summary of some international studies since 2007

2.1 NEA

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, 2002, 2004, 2006, 2008/. The committee has also organised a number of expert groups that have reported on various aspects of partitioning and transmutation in OECD/NEA reports. The most important of these have been summarised in previous status reports /Ahlström et al. 2004, 2007/.

Under the auspices of the NEA Nuclear Science Committee (NSC), the Working Party on Scientific Issues of the Fuel Cycle (WPFC) has been established to co-ordinate scientific activities regarding various existing and advanced nuclear fuel cycles, including advanced reactor systems, associated chemistry and flow sheets, development and performance of fuels and materials, and accelerators and spallation targets. The WPFC has different subgroups to cover the wide range of scientific fields in the nuclear fuel cycle.

Created in 2002, the NEA Working Group on Lead-bismuth Eutectic (WG-LBE) technology is a WPFC subsidiary group which co-ordinates and guides LBE research in participating organizations while enhancing closer and broader-based collaboration. The aim is to develop a set of requirements and standards as well as consistent methodology for experimentation, data collection and data analyses. The results have been published in the form of a handbook /NEA 6195 2009/. Due to a rising interest in the Pb-cooled option in the Generation IV International Forum, the WG-LBE also decided to include data and technology aspects of both LBE and Pb. The current edition of the handbook is a state-of-the-art, critical review of existing data and discrepancies, open points and perspectives for both Pb and LBE technological development.

The Expert Group on Fuel Cycle Transition Scenarios Studies was created in 2003 to consider R&D needs and relevant technology for an efficient transition from current to future advanced reactor fuel cycles. The objectives of the expert group are: i) to assemble and to organise institutional, technical and economic information critical to the understanding of the issues involved in transitioning from current fuel cycles to long-term sustainable fuel cycles or a phase-out of the nuclear enterprise; ii) to provide a framework for assessing specific national needs related to that transition.

Two reports from this group have been published recently, representing different collaborations within the group, but also different approaches and views. The report *Nuclear Fuel Cycle Transition Scenario Studies* /NEA 6194 2009/ discusses issues related to future fuel cycles, and gives an overview of possible transition scenarios for Belgium, Canada, France, Germany, Japan, the Republic of Korea, Spain, the United Kingdom and the United States, at the time of writing for each. The key issues and technologies that are crucial to the deployment of advanced fuel cycles are also identified. The report is more concerned with discussing relevant aspects for scenario studies rather than presenting exact results. No overall results are presented, but only national results separately without unifying discussion. It can be noted that for the countries with a phase-out political situation, the scenarios studied represent essentially the views of the authors only.

In a second report, *Regional Fuel Cycle Synergies and Regional Scenarios for Europe* /NEA 6857 2009/, Salvatores et al. proposes a regional approach for the implementation of P&T in Europe. Especially it is pointed out that with ADS in a double-strata concept, i.e. in which ADS is used primarily for MA rather than TRU incineration, the needs for a single country might not motivate an ADS facility, whereas shared regional facilities might constitute a better use of resources.

The Belgian government asked the OECD/NEA to organize an international peer review of the MYRRHA project to provide an independent evaluation as part of the decision-making process. NSC composed a team of seven experts from seven countries that presented a report with views and recommendations in 2009 /NEA 6881 2009/. The team concluded unanimously that MYRRHA is an innovative and exciting project, that the facility would be World-unique, and could play a valuable role in the development of future advanced reactor systems. It was concluded that the design is so innovative that substantial uncertainties remain in the construction and financing. The probability that the costs will significantly exceed the expectations is considered substantial. Three possible options were identified:

- To give the project a full go-ahead decision, in which the Belgian government guarantees the funding of the project. Such a decision would involve a large uncertainty on the financial commitment.
- To give the project a go-ahead decision to perform detailed design work, where as many technical design aspects as possible should be freezed, thereby allowing budget estimations of reduced uncertainty before a full go-ahead decision be taken.
- To abandon the project due to costs and risks.

The middle option was recommended to the Belgian government, with the suggestion that a final decision be taken in two to three years.

The NEA Nuclear Development Committee (NDC) has hosted an expert group carrying out a study on *Strategic and Policy Issues Raised by the Transition from Thermal to Fast Nuclear Systems* /NEA 6352 2009/. A team of 17 experts from 11 countries and two international organizations (IAEA, EU) assessed various scenarios in deployment of fast reactors on a large scale as integral part of the nuclear energy system. Pål Efsing from Vattenfall Ringhals participated from Sweden. In short, the main findings were:

- For small countries, with a limited fleet of reactors, building the infrastructure required is unlikely to be cost effective. In those cases, regional solutions were deemed more probable.
- Countries with well-defined policies for once-through nuclear fuel cycles and advanced programs for geological direct disposal were considered less likely to adopt a fast reactor program, whereas for countries with little or no such plans fast neutron systems might offer an attractive strategy for waste management and disposal.
- Although fast reactors have been operated in a number of countries for some centuries total operation time, the technology still has not reached a maturity that allows industrial introduction without further R&D.
- Stable policies are a pre-requisite for successful introduction of a fast reactor fleet because of the long lead times required.
- Public acceptance was identified as mandatory. Non-proliferation issues need careful consideration.
- Human resource management via education and training is seen as a cornerstone.

Proton accelerator reliability has previously been identified as one of the possible limiting factors in the development of industrial ADS facilities. The energies required have been reached since long time, and the intensities are within reach. Up to now, however, no accelerators have ever been built with reliabilities even near what is needed for successful industrial ADS application. On the other hand, no proton accelerator application has hitherto needed such a high reliability. Thus, the question to what degree of reliability such accelerators can be developed is still and open issue.

Since 1998, the NSC has organized recurring workshops on the reliability of accelerators for nuclear energy applications. The most recent one was held in May 2009, and was organized in collaboration with SCK·CEN. The most important conclusion was that the experiences from the presently most powerful accelerator in operation, the Spallation Neutron Source (SNS), Oak Ridge, USA, are positive /NEA 6259 2008/. The number of beam trips, and the consequences thereof, have been reduced by fault-tolerating design. Reaching performance required by large-scale ADS applications is still far away, but significant progress has been made and no show-stopper has been identified.

2.2 IAEA

Within the framework of the project on *Technology Advances in Fast Reactors and Accelerator-driven Systems,* the IAEA has initiated a number of activities concerning the utilisation of plutonium and transmutation of long-lived radioactive waste, accelerator-driven systems, thorium fuel options, innovative nuclear reactors and fuel cycles, non-conventional nuclear energy systems, and fusion/fission hybrids.

The framework for all the IAEA activities concerning P&T is the Technical Working Group on Fast Reactors (TWG-FR). The TWG-FR acts as a catalyst for international information exchange and collaborative R&D. Given the common technical ground between plutonium utilisation R&D activities and the development of technologies for the transmutation and utilisation of long-lived fission products and actinides, both activities are performed within the framework of a single agency project: *Technology Advances in Fast Reactors and Accelerator-driven Systems*. Its present members are the following 14 IAEA member states: Belarus, Brazil, China, France, Germany, India, Italy, Japan, Kazakhstan, Republic of Korea, Russian Federation, Switzerland, United Kingdom, and United States of America, as well as the OECD/NEA, and the EU (EC). Belgium and Sweden are observers.

With regard to collaborative R&D, the IAEA has an ongoing (2002–2006) Co-ordinated Research Project (CRP) on *Studies of Advanced Reactor Technology Options for Effective Incineration of Radioactive Waste*, and has started a new CRP (2005–2009) on *Analytical and Experimental Benchmark Analyses of Accelerator-driven Systems (ADS)*. Recently, the TWG-FR has prepared a Technical Report on heavy liquid metal (HLM) thermal-hydraulics /IAEA 2007/.

2.3 EU-studies of transmutation

The research within EURATOM FP7 (2007-11) is composed of three major categories:

1. Fusion research
2. Nuclear Fission & Radiation Protection
3. Nuclear activities within the Joint Research Centres

The budget of these areas within FP6 and 7 is presented in Table 2-1.

In the realm of Nuclear Fission & Radiation Protection, three major thematic areas can be identified:

- **Radioactive Waste Management**. Geological disposal of long-lived radioactive waste and the reduction of toxicity of radioactive waste through partitioning & transmutation.
- **Reactor Systems**. Operational safety of existing reactor systems and the potential of future reactor systems for safer, more efficient power plants and competitive nuclear industry.
- **Radiation protection**. Especially risks from low doses, medical uses, emergency management etc.

In addition, key cross-cutting activities are financed:

- Support for research infrastructures.
- Retaining competences and know-how in all areas of nuclear science.

Table 2-1. EURATOM Framework (FP) Programs Budget (M€).

	FP6 (2002-06)	FP7 (2007-11)
Fusion Energy	824	1,947
Fission & Radiation Protection	209	287
JRC Nuclear Activity	319	517
Total	1,352	2,751

The EU Strategy in P&T for Sustainability of Nuclear Energy can be summarized as follows /Bhatnagar 2008/:

- Once-through cycle does not appear to be sustainable.
- Reprocessing of the spent fuel and transmutation of Minor Actinides in dedicated devices reduces radio-toxic inventory of the disposed waste in geological repositories.
- This has significant importance in non-proliferation strategy and radiological terrorism and reduces risks in case of an inadvertent human intrusion.
- A double-strata approach with sub-critical Accelerator Driven Systems (ADS) and/or critical Fast Reactors (FR) is being considered. A decision on the choice is planned in a couple of years.
- Geological disposal of the remaining waste (separation/transmutation losses) will be required.
- P&T is essential for the sustainability of nuclear energy.
- Geological disposal is indispensable for radioactive waste management.
- Both communities should work together for the future of nuclear energy.

The EU Waste Management Strategy for sustainability of nuclear energy is detailed below:

- Separation of main heavy metals reduces the volume and thermal output and extraction of heat-bearing (Sr and Cs) components permits a reduction in the needed size of the repository except possibly in salt-media that does not need this separation.
- Transmutation can reduce the half-life of most of the waste to be disposed of to a couple of hundred years overcoming the concerns of the public related to the long-life of the waste thus aiding the geological disposal community in securing a broadly agreed political consensus of waste disposal in geological repositories.
- Additional cost, additional secondary waste, activation products and intermediate-level waste and dose to workers in the process of reprocessing and transmutation itself will contribute to defining an optimal transmutation scheme.
- P&T has an added value of training many researchers in nuclear science and contributes to retaining of competence.
- Though the maximum eventual dose to human beings from a geological repository in normal scenarios is likely to be due to fission products, some evidence is appearing that minor actinides are also mobile.
- Generation IV safe advanced nuclear reactor concepts that burn waste and produce fuel for further use are gaining increased attention as a possible future course of action.
- It is clear that reprocessing of the spent fuel for a sustainable nuclear energy and fuel cycle would be required no matter what path of transmutation is followed.
- Efforts for the advanced partitioning processes should be reinforced towards pilot and test facilities for optimised separation processes in close cooperation with fuel fabrication teams and geological disposal (GD) community.
- The GD community should embrace the opportunity offered by P&T to ease the acceptance of geological repositories by society and make the best use of accepted sites by removal of long-lived and heat producing radionuclides from the waste.
- The GD community should especially take into account the requirements and accommodate the waste streams emanating from the advanced (minor-actinide) reprocessing systems with a view to transmutation whether in sub-critical or critical devices.

Table 2-2. 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	43.5
FP 7	2007–2011	~ 44

SNETP

The Sustainable Nuclear Energy Technology Platform (SNETP) was officially launched on the 21st September 2007, in the presence of EU Commissioners for Science and Research, J. Potočnik, and Energy, A. Piebalgs. SNETP is a framework to unite all stakeholders (public-private partnership) around a common vision of sustainable nuclear energy technology research, and an attempt to mobilise a critical mass of research and innovation effort. At present, SNETP has around 70 member organizations.

In layman's terms, Technology Platforms (TP) are a type of accredited EU lobbying organizations. If a large fraction of the stakeholders – private companies, governmental organizations, universities, non-governmental organizations – within a certain technology area can create a joint platform, this TP can get recognition from EC as a discussion partner. This means that the TP gets some administrative aid from the EC, and serves the EC with advice on e.g. research funding issues and policy development.

At the launch, the SNETP Vision Report was presented /SNETP vision report 2008/. It highlights the role nuclear energy plays in Europe's energy mix as the main provider of low carbon electricity (providing 31% of EU's electricity), and identifies future research, development and demonstration (RD&D) tracks that the nuclear fission sector must follow to address three objectives:

1. Maintain the safety and competitiveness of today's technologies,
2. Develop a new generation of more sustainable reactor technologies (Generation-IV Fast Reactors with closed fuel cycles)
3. Develop new applications of nuclear power such as the industrial scale production of hydrogen, desalination or other industrial process heat applications.

On 10th January 2007, the European Commission published a seminal communication, An Energy Policy for Europe /Energy Policy for Europe 2007/, which for the first time underlined the benefits of nuclear energy: low carbon emissions, competitiveness, and stable prices. In the context of an anticipated increase in use of nuclear energy in the world, the Commission also recognised that "there are therefore economic benefits in maintaining and developing the technological lead of the EU in this field". This communication was endorsed by the Council in March 2007, which also committed the EU to meet ambitious objectives by 2020 of 20% reduction in greenhouse gas emissions (compared to 1990), 20% renewable energies in the energy mix, and 20% reduction in energy consumption through better energy savings and management. In order to achieve these goals and realise the longer term vision of a low carbon society by 2050, the Commission identified R&D prospects of key low carbon energy technologies in a follow-up communication, the Strategic Energy Technology (SET) Plan, published on the 22nd November 2007 /EU SET 2007/.

Nuclear fission is cited together with other low-carbon technologies such as renewables and Carbon Capture and Storage (CCS) technology as one of the contributors to meet the 2020 challenges. By maintaining "competitiveness in fission technologies, together with long term waste management solutions", fission energy will continue to be leading low-carbon energy technology in Europe. This objective is to be achieved by acting now to "complete the preparations for the demonstration of a new generation (Gen-IV) of fission reactors for increased sustainability". From 2040 onwards, it is envisaged that this new generation of Fast Neutron Reactors will be operating in parallel to the advanced Gen-III Light Water Reactors now being built in Europe, thereby maintaining the current 1/3 share of nuclear electricity in Europe.

The SET plan has been followed up by SNETP in its more detailed Strategic Research Agenda (SRA), issued in 2009 /SNETP-SRA 2009/. In the SRA, sodium-cooled fast reactors (SFR) are identified as a proven technology, and should therefore be the first technology for a full-scale prototype. The design of a reactor in the 250–600 MW electric power range should be completed by the end of 2012, aiming at beginning operation in 2020. France has offered to host such a reactor, named ASTRID, and design is in progress. In parallel, research on two alternative critical fast reactor technologies, lead- and gas-cooled fast reactors, should be pursued. The aim is to select one of them in 2012 to be the second technology for deployment, and a prototype in the 50–100 MW thermal range should be in operation by 2020. Finally, the Belgian MYRRHA concept is identified as the first experimental demonstrator of ADS technology.

2.3.1 EUROTRANS

In the sixth framework programme of the European Commission, the largest integrated project on partitioning and transmutation was the EUROTRANS project, aiming at performing

- design and safety analysis of a 60 MWth MOX fuelled and lead-bismuth cooled sub-critical reactor driven by a 600 MeV proton beam, (XT-ADS),
- design and safety analysis of a 400 MWth sub-critical reactor with minor actinide based inert matrix oxide fuel (EFIT). Two coolants were considered, lead and gas,
- development and validation of methods for sub-criticality monitoring,
- fabrication and irradiation testing of minor actinide based inert matrix fuel,
- research on structural materials performance under irradiation and exposure to lead-alloy coolant,
- measurement and evaluation of selected nuclear data of relevance for modelling of accelerator driven systems.

XT-ADS

XT-ADS was conjectured as an experimental reactor for proving the concept of coupling a high power proton accelerator with a sub-critical core cooled by heavy liquid metal coolant. The reactor should also function as a materials irradiation testing facility, and was therefore to be designed to provide a sufficiently large fast neutron flux. The basis of the design was the MYRRHA concept previously developed by SCK-CEN in Belgium. Within EUROTRANS, more detailed attention was given to reduce peak linear ratings, core pressure drop, damage dose to the core barrel and the fuel reshuffling scheme.

Table 2-3 shows the more important parameters selected for the fuel pin design of XT-ADS. One may note in particular the height of the gas plenum. The height was increased from the MYRRHA design, to keep fission gas pressures low enough for high temperature creep of the T91 clad not to become a safety issue under transient conditions. A central hole was further added, to reduce fuel pin temperatures and improve performance under accidental conditions.

Each fuel assembly of XT-ADS contains 90 fuel pins with "standard" MOX fuel, having a plutonium fraction of 35 percent. Since the core should function as an irradiation device, several configurations were investigated. The so-called "clean core" loading map at equilibrium is shown in Figure 2-1. It contains 75 fuel assemblies and 8 empty positions, where experimental devices can be loaded. Three central assembly positions are dedicated to the insertion of the spallation target. 55 lead-bismuth filled assemblies surround the core and 45 reflector assemblies with steel pins protect the core barrel from fast neutrons.

At 57 MWth core power, the maximum neutron flux is 3×10^{15} /cm^2/s and dose rate in the experimental device positions is up to 20 dpa per year, satisfying the objective of using XT-ADS as a materials test reactor. The necessary proton beam power used for driving the reactor is 1.9 MW at current of 3.2 mA. Reactivity losses are about 110 pcm per day, whence a cycle length of 90 days is applied to limit reactivity losses to 1,000 pcm per cycle. A five batch reloading scheme was deviced, leading to a fuel residence time of 450 effective full power days and a fuel burnup of 33 GWd/ton.

Table 2-3. XT-ADS fuel pin geometry.

Fuel pin parameter	Value
Pellet inner diameter	1.60 mm
Pellet outer diameter	5.40 mm
Pellet-clad gap	0.10 mm
Clad thickness	0.475 mm
Clad outer diameter	6.55 mm
Pin pitch/diameter ratio	1.40
Fuel column height	600 mm
Gas plenum height	640 mm
Total fuel pin height	1,400 mm

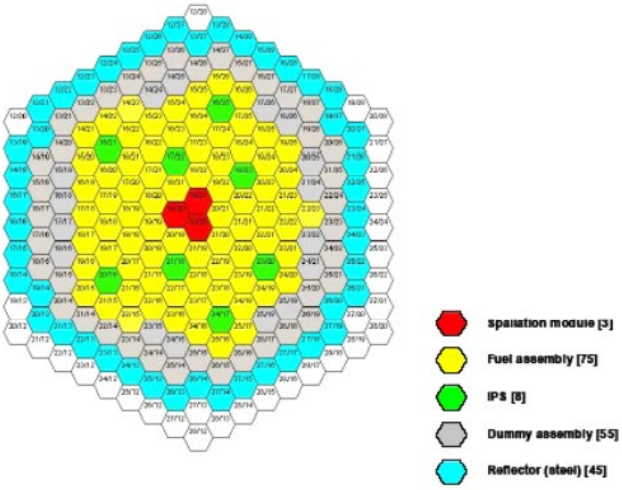

Figure 2-1. *XT-ADS core map. Note the asymmetrical configuration arising due to the choice of fitting the spallation target into the space of three (removed) central fuel assemblies.*

A preliminary cost estimate for construction of XT-ADS has been made by the industrial partners of EUROTRANS, leading to a figure of 684 M€, including decommissioning.

EFIT

EFIT (European Facility for Industrial Transmutation), in contrast to XT-ADS, is foreseen to be a prototype of an industrial facility for minor actinide burning. Two variants have been designed and analysed within EUROTRANS, the primary option relying on lead coolant and the back-up option on helium gas.

A major design criterion for EFIT was that it should feature a net plutonium production/destruction rate equal to zero. The rationale for this criterion is that the relatively costly fission energy produced in EFIT would be utilised for minor actinide burning exclusively. Further, the plutonium formed by transmutation of americium would be possible to recycle within the park of EFITs, thus not contaminating the first stratum of commercial reactors. Finally, the reactivity swing would to first order of approximation be equal to zero with such an approach, leading to benefits for both economy and safety /Glinatsis 2009/.

From the safety viewpoint it was considered that the lead-cooled EFIT should be able to utilize the high potential for natural convection of lead to permit not only decay heat removal, but also to survive an unprotected loss-of-flow accident, that is loss of pumping power without shut-down of the proton beam. Similar requirements were not demanded for the gas-cooled EFIT, where decay heat removal under loss of pressure is very difficult to manage. The helium cooled EFIT on the other hand features other advantages, such as the possibility of visual inspection of core integrity.

The sub-criticality level of EFIT was postulated to be keff $\leq$ 0.97 during normal operation. The requested margin of 3,000 pcm is a combination of margins for sub-criticality, accidental reactivity insertions and measurement errors.

The reference fuel for EFIT adopted by the design team is $(Pu,MA)O_{1.88}$ dispersed into a matrix of magnesium oxide (MgO). This composite “CerCer” fuel offers a relatively good neutron economy, sufficient thermal conductivity and low cost of fabrication. A significant drawback is the high vapour pressure of MgO, limiting the permitted temperature of the fuel to less than 2,100 K.

The termal hydralic and fuel zone design of the lead cooled core were made to ensure a maximum cladding temperature of less than 550°C during normal operation, for a core inlet temperature of 400°C and a linear rating limited to less than 21 kW/m. In combination with the zero net Pu production constraint, the relative fraction of plutonium in the reference core was determined at 45.7%.

Here, one should note that the actinide vectors assumed for fabrication of the first core load were those of spent LWR (UOX + MOX) fuel allowed to cool for up to 30 years. The adopted fuel hence features relatively low fractions of ^{241}Pu, and correspondingly higher inventories of ^{241}Am.

The design parameters for the EFIT reference lead cooled core are summarised in Table 2-4. Three fuel zones with varying inert matrix fraction and fuel pellet radii were introduced to keep the radial power peaking factor below 1.3. One may note in particular the gas plenum size, which is designed to accommodate helium gas being produced through the transmutation of ^{241}Am by neutron capture:

$$n + {}^{241}\mathrm{Am} \rightarrow {}^{242}\mathrm{Am} + \gamma \rightarrow {}^{242}\mathrm{Cm} + \beta \rightarrow {}^{238}\mathrm{Pu} + \alpha$$

Figure 2-2 shows the core map of EFIT. The space of 19 central fuel assemblies is used to accommodate the spallation target. The three fuel zones consist of 42, 66 and 72 fuel assemblies, respectively, each containing 168 fuel pins. Four rows of reflector assemblies filled with steel pins surround the core to reduce the damage dose to the core barrel.

Table 2-4. Design parameters for EFIT-lead.

Fuel pin parameter	Zone 1	Zone 2	Zone 3
MgO matrix volume fraction	0.570	0.500	0.500
Pellet radius	3.55 mm	3.55 mm	4.00 mm
Pellet-clad gap	0.16 mm	0.16 mm	0.16 mm
Clad thickness	0.60 mm	0.60 mm	0.60 mm
Clad outer radius	4.31 mm	4.31 mm	4.76 mm
Pin pitch/diameter ratio	1.581	1.581	1.422
Fuel column height	900 mm		
Gas plenum height	1,100 mm		
Total fuel pin height	2,100 mm		

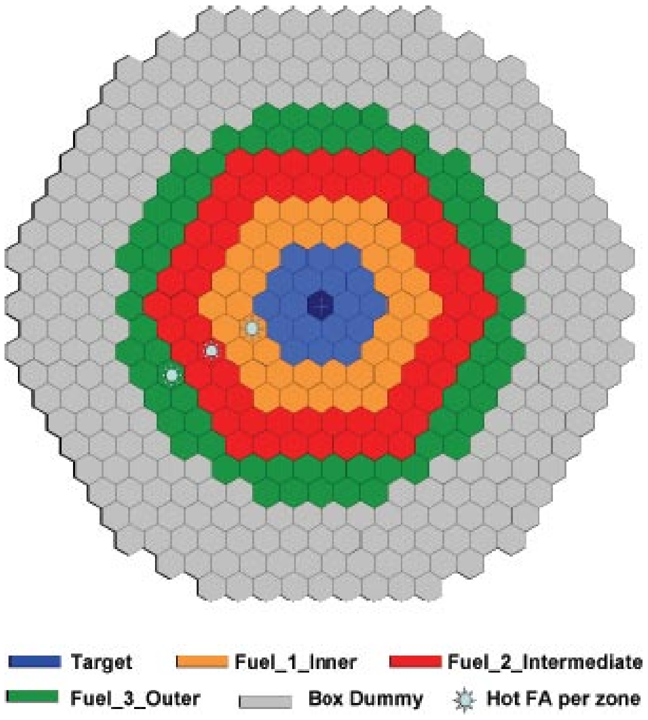

Figure 2-2. Core map of EFIT-lead. The spallation target is accomodated in the space of 19 central fuel assemblies.

The reactivity swing of this core was calculated to be about 200 pcm per one full power year of operation /Sarotto et al. 2008, Fokau et al. 2010/, which corresponds to an almost constant beam power of 11 MW. The estimated neutron source efficiency of 0.52 is much lower than that of XT-ADS, which partially is due to the large spallation target size. As neutrons created in spallation processes are moderated by in-elastic collisions in the target, their average energy drops below that of the average neutron energy in the core. Hence the probability for inducing fission is smaller than for the average neutron in the core.

Assuming that clad corrosion limits the residence time of the fuel to three full power years, a burnup of 78 GWd/ton of fuel is achieved in EFIT-Pb, corresponding to 14% reduction of minor actinides and an almost constant inventory of plutonium.

YALINA

In the YALINA-booster set of experiments, the aim was to find a reliable and accurate way of obtaining the subcriticality of an ADS. In these experiments, carried out at the zero-power coupled fast-thermal subcritical facility of JINPR in Sosny /Chigrinov 2004/, the Sjöstrand area ratio method /Sjöstrand 1956/ was applied in an extreme environment as a thorough validation for future offline use in a full-scale system. An important aspect of an ADS to be taken into consideration is its possible variation in source multiplication during operation due to for instance burnup. Thus, it is preferred to use a reactivity measurement method insensitive to source multiplication variations. The high flexibility of the subcritical facility allowed studying two configurations having the same subcriticality but different source multiplication factors. This was obtained by substituting highly enriched fuel in the centre of the core (close to the neutron source) by low enriched fuel at the core periphery. In this way the effective multiplication factor (k_{eff}) could be kept constant while altering the source multiplication factor (k_s). This was done with an effective multiplication constant of ~ 0.95. In addition, a deep subcritical configuration (~ 0.85) was investigated to identify possible problems when applying the methods during core loading.

Figure 2-3 show a cross sectional view of the YALINA-Booster set-up. The core consists of a central lead zone (booster), a polyethylene zone, a radial graphite reflector and a front and back biological shielding of borated polyethylene. The fast spectrum lead zone and the thermal spectrum polyethylene zone are separated by a so called thermal neutron filter, or valve zone, consisting of one layer of 108 pins with metallic natural uranium and one layer of 116 pins with boron carbide (B_4C), which are located in the outermost two rows of the fast zone. Hence, thermal neutrons diffusing from the thermal zone to the fast zone are likely to be absorbed either by the boron or the natural uranium. In this way, a coupling of mainly fast neutrons between the two zones is maintained.

The booster has two sub-zones, the inner and outer booster, with different fuel pin pitch. Both booster sub zones could be loaded with 36% enriched uranium oxide fuel. The thermal zone was loaded with uranium oxide with 10% enrichment.

The most important observation from the measurements made was the large deviation in obtained reactivity value when the Sjöstrand area ratio method was applied in different reactor regions. The absolute value of the reactivity when measured in the booster region can be up to a factor of two larger than the corresponding measurement in the thermal region or the reflector. A two-region point kinetic model of the system was constructed, which permitted to analyse and understand the physics behind this discrepancy. In order to obtain accurate correction factors, a novel, fully three-dimensional Monte Carlo approach was applied, which could be shown to be independent of the effective delayed neutron fraction. Furthermore, a direct estimate of the effective multiplication factor can be obtained /Berglöf 2010/. After applying the calculated correction factors, the spatial spread in measured reactivity was reduced from thousands of pcms to hundreds. Hence, one may conclude that sufficiently accurate reactivity measurements can be performed in sub-critical systems, as long as care is taken to locate detectors in positions far away from the accelerator-driven neutron source and other heterogeneous constituents.

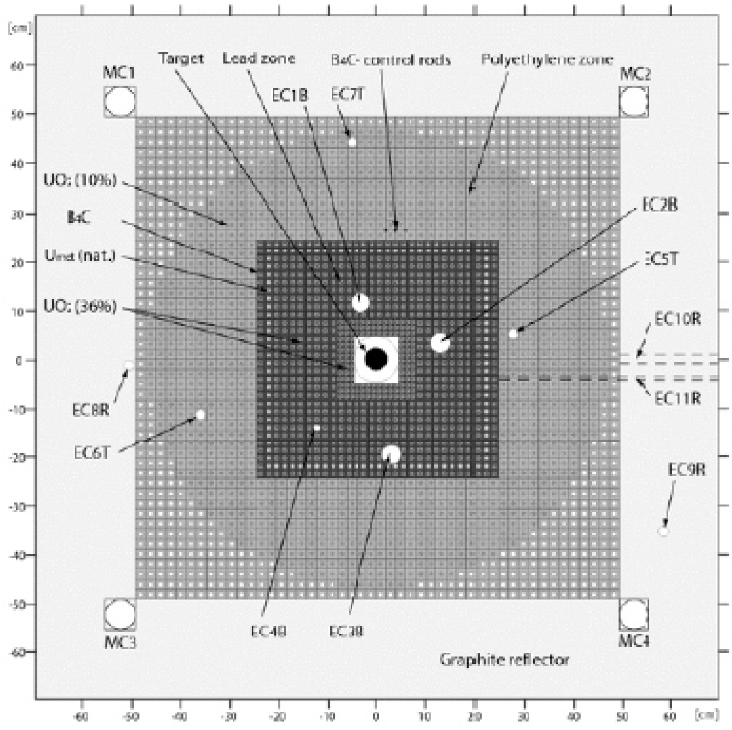

Figure 2-3. *Schematic cross-sectional view of the YALINA-Booster reactor core.*

GUINIVERE

In order to validate reactivity monitoring methodologies in a system more representative for XT-ADS, the GUINIVERE experiments are under preparation within EUROTRANS. Here, the VENUS critical facility in Mol has been rebuilt into a fast spectrum core with 30% enriched UOX fuel and simulated lead coolant. In addition a continuous beam D-T neutron generator and corresponding beam-line has been constructed. Design and safety studies were completed and the construction of the experiment is under way. In December 2009, the final steps for licensing were completed with the Belgian safety authorities and the fuel is expected to be loaded into the core in January 2010.

Fabrication of inert matrix composite fuels

The selection of the reference and back-up fuels for the industrial ADS prototype EFIT was made after an assessment of possible candidates with respect to the following properties /Wallenius 2006/:

- 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.

Carbide, metallic alloy, zirconia and thoria based fuels were discarded during the selection process, due to known difficulties with their dissolution in nitric acid. In the case of metallic fuels, the issue is related to the sodium bond present in the fuel design, while in the case of zirconia and thoria, the need for agressive catalysts like hydrofluoric acid, makes their dissolution incompatible with existing industrial processes in Europe.

Further, $MgAl_2O_4$ composites were discarded due to poor irradiation performance, and tungsten based cermets were found to hamper neutron economy too much. Within the fuel domain of EUROTRANS, the following ranking was made of possible fuels for EFIT

1. the composite cermet fuel $(Pu,MA)O_{2-x}$ – ^{92}Mo,
2. the composite cercer fuel $(Pu,MA)O_{2-x}$ – MgO,
3. the solid solution nitride fuel $(Pu,MA,Zr)^{15}N$,

where the safety case was strongest for the molybdenum based cermet fuel. The need for enrichment of molybdenum and nitrogen would however lead to additional cost penalties for the cermet and the nitride fuels. Issues related to high temperature stabilty of the MgO based cercer fuel and americium bearing nitride fuel will have to be assessed in more detail to ensure that these fuels eventually can be licensed as driver fuels of an industrial facility.

The perceived high cost for enrichment of molybdenum later formed the basis for the design domain of EUROTRANS to select the MgO based cercer fuel as the reference fuel for EFIT.

Having made the above selection, fabrication of $(Pu,MA)O_{2-x}$, $(Pu,MA)O_{2-x}$ – ^{92}Mo and $(Pu,MA)O_{2-x}$ – MgO was undertaken. Cermet fuels were fabricated in the MA-lab of ITU by infiltration of americium nitrates into porous plutonium dioxide beads, followed by thermal treatment to convert the nitrate to oxide, mixing with molybdenum powder, pressing and sintering. Figure 2-4 shows a cermet fuel pellet, featuring the typical dark oxide inclusions in the shiny metallic matrix /Haas et al. 2005/. The density of the pellets produced is 90–95% of the theoretical maximum.

Cercer fuels were fabricated in the ATALANTE lab of CEA, by co-precipitation of americium and plutonium oxalates, followed by calcination, mixing with magnesia powder, pressing and sintering /Jankowiak et al. 2008/. In fabrication of $(Pu_{0.5},Am_{0.5})O_{2-x}$ – MgO, the actinide compound was found to consist of a mixture of bcc and fcc phases. The measured oxygen to metal ratio varied between O/M = 1.61 to O/M=1.90, depending on the particular phase and fraction of inert matrix.

The investigation of americium bearing nitride fuels was carried out by JAEA, which formally became a partner of the EUROTRANS consortium. A solid solution $(Pu_{0.21}Am_{0.18},Zr_{0.61})N$ sample was prepared by manufacturing PuN and AmN powders by carbo-thermic nitridation of the respective oxides. These were then mixed with ZrN fabricated by hydridation/nitridation of metallic zirconium. Compaction and heat treatment under nitrogen/hydrogen for seven hours at 1,870 K then allowed to obtain a solid solution tablet /Takano et al. 2009/. Some losses of americium were observed during this process.

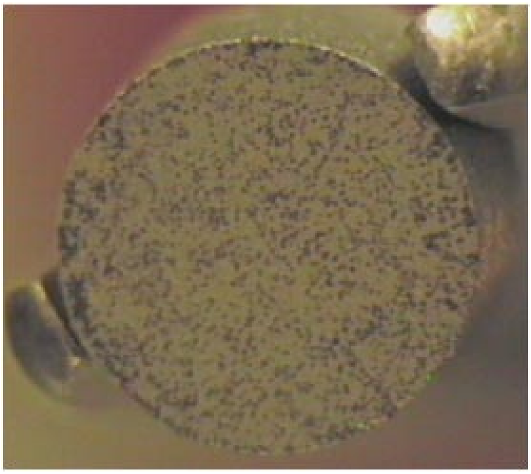

Figure 2-4. *A $Pu_{0.756}Am_{0.244}O_{2-x}$ – Mo pellet fabricated at ITU. The oxide phase represents 30% in volume.*

Thermo-physical properties of the above fuels have been measured during the course of EUROTRANS. In particular one may note the extremely high effective conductivty of the molybdenum based CERMET fuels, being of the order of 50 W/m/K for samples with 60 volume percent inert matrix /Fernandez et al. 2007/. The corresponding value for MgO based cercer fuels was measured to be 3–4 W/m/K. The expected value for the nitride fuel (calculated from conductivies measured for PuN, AmN and ZrN) is of the order of 20 W/m/K, depending on the sample density that may be achieved.

On the item of high temperature stability, Knudsen cell measurements performed under vacuum conditions show that Mg rapidly vapourises already at temperatures of 1,800–1,900 K, while the cermet fuel was shown to be stable up to at least 2,400 K /Delage 2009/. Under oxygen saturated conditions, the cercer will obviously exhibit considerably better stability.

FUTURIX

In the FUTURIX experiment, two MgO cercer and two Mo cermet fuel pins were irradiated for 240 full power days in the Phenix reactor, at linear ratings of 8–10 kW/m for the cercer fuel and 13–14 kW/m for the cermet fuels. This irradiation was the first true fast spectrum test for protypical ADS fuels. The results will become available as part of the post irradiation examination programme within the FAIRFUELs project.

Modelling of fuel performance

Within the domain for structural materials research, long term corrosion tests of selected steels to be used for cladding, core internals, heat exchangers and pressure vessel of XT-ADS and EFIT have been performed. Tests have been made both in lead-bismuth and pure lead, under oxygen concentrations representative for normal and accidental conditions, for different flow rates, mechanical loads and under irradiation with neutrons.

The steels selected for investigations within EUROTRANS were the ferritic-martensitic 9% Cr steel T91, and the austenitic stainless steel 316L. The former one was expected to provide better irradiation and corrosion resistance at high neutron fluences and temperatures up to 550°C, which is the maximum cladding surface temperature in XT-ADS and EFIT during normal operation. The latter features better mechanical properties under low neutron dose conditions and could be applied for e.g. the pressure vessel.

By now it is established that by maintaining an oxygen fraction in the lead coolant sufficiently large for protective oxide layers (spinel and magnetite) to form on the surface of steels, but low enough for the magnetite not to grow too thick, that integrity of steels can be maintained for temperatures up to 500°C. Suitable oxygen concentrations range from 1 to 2 ppm (weight). For lower concentrations (0.01 ppm), T91 and 316L are attacked by the liquid metal within 2,000 hours of exposure, even at a temperature of 450°C /Fazio 2008/. The thickness of the oxide layer grows with time and temperature, although the growth has been shown to be logaritmic (under flowing conditions) rather than parabolic. At 550°C, the thickness of the oxide may exceed 50 microns after 10,000 hours of exposure, while it may remain at 35 micros after the same exposure time at 500°C. The magnetite layer is sensitive to erosion and flow rates of 2.0 m/s or above will lead to its dissapearance.

For exposure times longer than 10,000 hours at 550°C, preferential dissolution of Cr in LBE occurs for T91, even with oxide scale formation. Therefore, better protection is required for the material to be able to operate in this temperature regime. At FZK (now KIT) in Karlsruhe, a technique for surface alloying the steel with FeCrAlY has been developed that appear to provide adequate protection, both at higher temperatures and higher coolant flowrates /Weisenburger et al. 2008/. The method is based on low pressure plasma spraying of FeCrAlY droplets on the surface of the steel, leading to formation of a surface layer of 30 micron thickness. This coating is porous and inhomogeneous, but can be treated with a pulsed electron beam (GESA), which melts the material to a depth of a few tenths of microns within ten microseconds, without affecting the structure of the bulk material. The resulting coating is homogeneous and features a very smooth surface. Exposed to oxygen containing liquid metals, the surface alloy forms a very thin and highly protective alumina scale. On the condition that the residual aluminum content is at least 4% by weight after the GESA treatment, T91 and

316 steels have been performing adequately for exposure times of ~ 10,000 hours at 550–600°C. In addition, no erosion of the alumina scale was observed in tests with LBE flowrates of up to 3.0 m/s /Müller et al. 2004, Weisenburger et al. 2008/.

The existence of liquid metal embrittlement has been confirmed in several independent investigations, and a significant reduction of creep rupture time under high pressure in LBE was observed /Auger et al. 2008/. This reduction however becomes less significant with reduced load /Jianu et al. 2009/.

Irradiation tests of T91 and 316L have been carried out in LBE environment. The TWIN-ASTIR experiment in BR-2 reached a dose of a 1.5 dpa at an irradiation temperature between 460 and 490°C /van den Bosch et al. 2009/. While a slight effect of liquid metal embrittelment was observed in post irradiation examination, no detrimental synergic effects from the combination of LBE exposure and neutron irradiation could be detected. In the SPEED-ASTIR follow up, irradiation of GESA-treated pressurised T91 tubes up to a dose of 3–8 dpa is planned. GESA treated material have also been irradiated in Phenix, assessing inter alia the impact of fast neutron doses up to 70 dpa on the integrity of the alumina scales.

Nuclear data

In EUROTRANS, UU participates with a joint Swedish-French experiment on neutron inelastic scattering, for which world-unique experimental equipment has been developed. The experimental work has been performed at UU, and analysis is in progress at LPC Caen, France.

2.3.2 RedImpact

Within the RedImpact project (coordinated by KTH), the impact of partitioning and transmutation on geological repository performance was investigated using various tools for simulation. Six different scenarios were investigated /Nabbi et al. 2007/:

A1) A once through open cycle with LWRs.

A2) Mono-recycling of plutonium in LWRs.

A3) Multi-recycling of plutonium in sodium fast reactors, using the European Fast Reactor (EFR) as reference.

B1) Multi-recycling of plutonium and minor actinides in sodium-cooled fast reactors.

B2) Mono-recycling of plutonium in LWRs and burning of minor actinides in Accelerator Driven Systems.

B3) Mono-recycling plutonium in LWRs, burning of plutonium in fast reactors and minor actinides in ADS.

These scenarios were considered in their equilibrium states (when isotopic vectors at fuel load remains constant from load to load). Transition scenarios were also investigated, using newly developed simulation tools.

Four different repository concepts were included in the study: Horisontal and vertical deposition in granite, horisontal deposition in salt and in clay.

The major findings may be summarised as:

- Full multirecycling of plutonum and minor actinides would result in significant reductions of heat load. In the case of granite or clay repositories, bentonite/clay temperatures must remain below 100°C. In these cases the gallery length for horisontally deposited canisters could be reduced by a factor of three, while the area of a repository with vertically deposited canisters could be reduced by a factor of six.
- Higher reduction factors (> 10) are possible to achieve if the fission products strontium and caesium are separated. A concept for their storage would however have to be developed in this case.

- It is important to note that the volume of intermediate level waste (ILW) would increase in the scenarios were reprocessing is applied. This resulting volume is of similar magnitude as the decrease in high-level waste (HLW).
- Concerning the reduction of radio-toxic inventory of the ultimate waste streams, it was confirmed that scenarios including multi-recycling of both Pu and MA, result in a reduction of the inventory by a factor of 100 once Cs and Sr has decayed, i.e. after 300 years. Multirecycling of Pu only reduces the inventory by less than a factor of 10. It should be noted that these values are not valid for transition scenarios where minor actinides already have been vitrified to some extent.
- As was well known already prior to RedImpact, partitioning and transmutation has no impact on the radiological dose resulting from a failure of barriers in the repository, since this impact is dominated by I-129. In all cases the estimated doses were orders of magnitude below natural background as well as regulatory limits.
- The impact of partitioning and transmutation on human intrusion scenarios was however found to be significant. The estimated dose to a geotechnical worker unintentionally extracting samples from a canister would be reduced by two orders of magnitude compared to the scenarios relying on LWR technology only.

2.3.3 GETMAT

The GETMAT project (Generation IV and Transmutation Materials) of the 7th framework programme is coordinated by Karlsruhe Institute of Technology (KIT). It addresses the need to qualify steels for use at high temperature in Generation IV reactors and accelerator driven systems. Five major work areas are pursued /Fazio 2008/:

Qualification of 9–12% Cr ferritic-martensitic steels

These are conventional steel grades produced at industrial scale, which have been studied earlier for application in advanced nuclear reactors. Example given, the TECLA, SPIRE and EUROTRANS projects have provided important data for their applicability in heavy liquid metal cooled reactor systems. In the MEGAPIE experiment, T91 was successfully used as construction material for the first liquid lead-bismuth spallation target at PSI, during four months of operation. Within GETMAT, the qualification of this class of steel will be extended, including post-irradiation examination on samples from MEGAPIE, the MATRIX fast neutron irradiation in Phenix, as well as the ASTIR and IBIS irradiations of samples irradiated in contact with LBE in BR2 and HFR.

Development and characterisation of Oxide Dispersion Strengthened (ODS) steels

The conventional 9–12% Cr ferritic martensitic steels are highly resistant to swelling and may operate to doses up to and above 200 dpa. However, these steels have inadequate mechanical properties at temperatures above 600°C. Austenitic (nickel bearing) cladding materials have better high temperature creep properties, but suffer from swelling at doses above 120 dpa, being a significant constraint for the burnup of the fuel in Generation IV reactors. Therefore it is of interest to investigate the applicability of ODS steels /Alamo et al. 2004/ for high temperature nuclear applications. Dispersing yttrium oxide nanoparticles to the bulk of ferritic and ferritic-martensitic steels, high temperature creep strength is considerably improved, and the range of application may possibly reach above 700°C. Irradiation tests also have shown that these steels retain ductility much better that their conventional counterparts /McClintock et al. 2009/. Since there presently exists no other major industrial application for this class of steels, major work needs to be accomplished on fabrication procedures, characterisation of microstructure, mechanical properties and irradiation performance. Within the GETMAT project, 9Cr and 14Cr ODS alloys will be manufactured using two different routes: powder metallurgy and innovative casting. The fabricated alloys will then be subject to mechanical testing and coolant compatibility tests. Further, they will be irradiated in different neutron and proton spectra to assess their resistance to radiation damage.

Qualification of welding procedures

A known issue with ODS steels is the difficulty in welding of the materials. Besides qualification of traditional fusion welding technologies (EB, TIG) alternatives are studied, such as magnetic pulse welding, diffusion bonding, friction stir welding, and explosive welding.

Development of a qualification procedure for FeCrAlY coatings for corrosion protection in heavy liquid metals at high temperature

As mentioned in the description of the EUROTRANS project, it has been found that the formation of iron oxide films on cladding steels is not adequate for protection of the steel for exposure times longer than 10,000 hours at 550°C. The technique for surface alloying the steel with FeCrAlY developed at FZK/KIT will be further refined and qualified in the GETMAT project, including irradiation testing of ferritic-martensitic, austenitic and ODS steels in fast neutron spectra (Phénix).

Multiscale modelling

Physical phenomena related to the synergistic effect of irradiation and environment are not linear. Incubation times or doses and thermally activated processes may determine the appearance of totally unexpected materials responses above a certain dose or temperature. Thus, a safe extrapolation of the behaviour of materials such as ferritic-martensitic steels to the envisaged in-service conditions must be based on some degree of physical understanding of the basic mechanisms acting from the atomic to the macroscopic level and determining their response to the applied environmental, thermal and mechanical loads, while being exposed to neutron irradiation. The modelling effort in GETMAT aims at understanding the physical mechanisms at the basis of the response to, mainly, irradiation of FeCr alloys, as model alloys for high Cr ferritic-martensitic steels.

2.3.4 FAIRFUELS

The FAIRFUELS project (Fabrication, irradiation and reprocessing of fuels and targets for transmutation) of the 7th framework programme is coordinated by Nuclear Research and consultancy Group (NRG). The project mainly concerns minor actinide bearing fuels, their fabrication and irradiation performance, including post-irradiation examinations and modelling of fuels tested in previous framework programme projects.

Technology for fabrication of americium bearing fuels and targets previously developed in programmes such as FUTURE and EUROTRANS will be further refined. The fuels to be fabricated for irradiation in FAIRFUELS will be based on a uranium dioxide matrix. In the MARIOS irradiation, the major objective is to investigate the accommodation of the helium being co-produced during transmutation of americium. In the SPHERE irradiation, spherical particles of $(U,Pu)O_2$ will be fabricated by the sol-gel method and then they will be infiltrated with Am solutions to reach contents applicable to homogeneous and heterogeneous recycling strategies (typically 5% and 20%). The MARIOS and SPHERE irradiations will last for 300 full power days in the High Flux Reactor in Petten.

Post-irradiation examinations will be performed on the CONFIRM, HELIOS and FUTURIX fuels irradiated in HFR and Phenix. Thermo-mechanical modelling of these experiments will be made with the MACROS code. In addition, modelling of fission gas and helium migration in MOX as well as in inert matrices (Mo & MgO) will be performed.

2.3.5 CDT

The Central Design Team (CDT project) in FP7 has the aim to design a fast spectrum transmutation experimental facility (FASTEF) able to demonstrate efficient transmutation and associated technology through a system working in sub-critical and/or critical mode. The project is coordinated by the Belgian research centre SCK·CEN.

The FASTEF design will be based on the work previously performed for the MYRRHA and XT-ADS projects. The major objectives of FASTEF are that it should

1. be operated as a flexible and high-flux fast spectrum irradiation facility,
2. be an experimental device to serve as a test-bed for demonstrating ADS technology and transmutation,
3. contribute to the demonstration of the Lead Fast Reactor technology.

By the end of the CDT project, it should be possible to take a decision on construction of the FASTEF/MYRRHA.

2.3.6 CONFIRM

The CONFIRM project (coordinated by KTH) was launched in the 5th framework programme to investigate safety, fabricability and irradiation performance of inert matrix nitride fuels for ADS. Due to the shutdown of Studsvik's R2 reactor, where irradiation of four (Pu,Zr)N fuel pins was supposed to have taken place, the completion of the project was much delayed. Eventually, an irradiation of shorter duration could be performed in the Dutch High Flux Reactor (HFR), and the project was concluded in August 2008. Other results from the project have been reported in /Ahlström et al. 2007/.

Two (Pu,Zr)N pins with 30% volume fraction of Pu were selected by NRG for irradiation in HFR with a target linear rating of ~ 450 W/cm at beginning of irradiation. The thermal design of the experiment was made to achieve cladding temperatures similar to those in liquid metal cooled reactors (700–800 K). The pins were irradiated for 6 cycles, corresponding to about 170 full power days. The CONFIRM irradiation sample holder was fabricated with a hafnium shield that removes the major part of the thermal neutron spectrum. In this way a (semi)fast spectrum is achieved which is more specific of a typical ADS spectrum. However, most fission power is still generated by the thermal part of the spectrum that remains despite the Hf-shielding. The CONFIRM irradiation was monitored using thermocouples. Fluence detectors were present to determine, after irradiation, the fluence and spectrum of the irradiation. The irradiation started on November 25th 2007 and was completed on the 22nd of June 2008. Neutron radiographies were made before and after irradiation. The radiograms are displayed in Figure 2-5, showing that the experiment is fully intact after the 6-cycle irradiation. Further, fuel stack elongations of 3.4–3.5% can be estimated.

Actual fluxes and cycle durations yielded a total fluence 13% higher than the pre-calculated value obtained with MCNP, and an estimated linear power of 47–49 kW/m for the CONFIRM fuel. Linear extrapolation of the pre-irradiation burn-up calculations then yields a calculated burn-up of 10.4% fission of the initial plutonium inventory /Wallenius 2009/.

Preliminary gamma-scan data of the CONFIRM pins show significant intensity peaks at the horisontal interface between the fuel and HfN end pellets. They further indicate that volatiles like ^{135}Cs have remained within the fuel stack and that release rates of ^{85}Kr to the gas plenum are significantly lower than predicted by the fuel performanc code used to model the experiment. If confirmed by gas analysis from puncturing tests, the CONFIRM fuel appears to be able to retain fission gases to a much larger extent than expected. Similar behaviour has been observed in post irradiation examinations of (Pu,Zr)N fuel irradiated in Russia and Japan /Arai et al. 2007, Golovanov 2005/. These experiments were however performed at lower heating rates than the CONFIRM irradiation.

2.3.7 PUMA

In the PuMA ("Plutonium and Minor Actinide Management in Thermal High Temperature Reactors") project, the feasibility of performing transmutation of plutonium and minor actinides in high temperature reactors has been investigated. The coated particle fuel used in these reactors offers unique safety features linked to stability and fission product retention properties.

The activities within the PUMA project were organized into four Work Packages, which concerned HTGR core physics analysis and qualification of analysis tools (WP1), HTGR Pu/MA fuel development, performance (and failure) modeling and testing (WP2), scenario studies (WP3), and project management and communication (WP4).

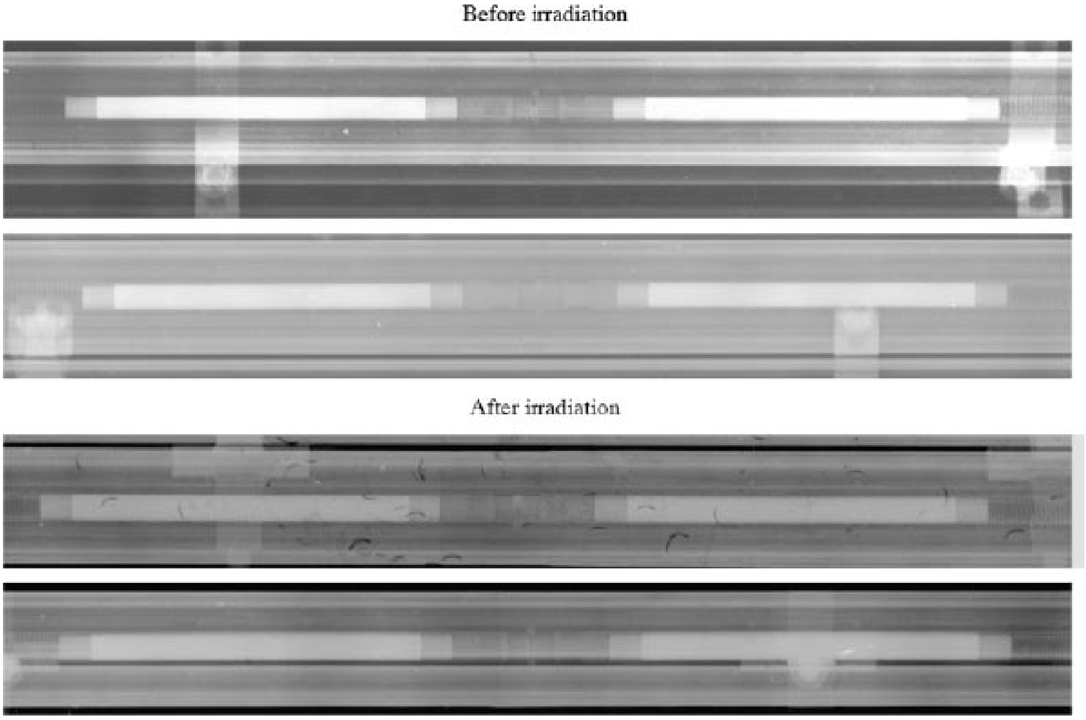

Figure 2-5. *Neutron radiograms of the CONFIRM fuel pins before and after irradiation.*

Investigation within the PUMA project concerned two types of HTGR reactors, PBMR-400 – pebble-bed (with fuel continuous reloading), and the GT-MHR (or derivative) having the prismatic block core design. Reference systems of both types were established and described. Within WP1, information about the computational codes used by the PUMA WP1 partners was collected and evaluated in order to identify deficiencies in the qualification of analysis tools.

Whole-core neutronic calculations were performed on the reference pebble-bed and prismatic HTGRs, loaded with reference Pu- and Pu/MA fuel. Pu/MA incineration performance as well as safety properties were investigated for several initial Pu and Pu/MA fuel compositions in combination with the reference reactor designs. It was demonstrated that there are no major show-stoppers for the incineration of Pu/MA in HTRs, although further development work has to be done. In addition, utilization of Thorium-Plutonium fuel in HTGR was investigated and consequently characteristics of Thorium-Pu fuels (dimensions, enrichment) were suggested.

Special attention has been dedicated to investigation of source convergence in the MCNP Monte Carlo calculations and results of these analyses indicate that this generic feature needs to be considered in future analysis of large, thermal systems.

WP1 additionally dealt with proliferation resistance of Pu/MA loaded HTGRs. According to the assessment, these types of reactors potentially offer superior performance in terms of efficiency, economy, inherent safety and proliferation resistance.

Within WP1, helium production in the coated particles was also modeled. Simulations showed that the He production is heavily dependent on location in the core. Two main processes producing alpha particles were identified and optimal loading scheme, leveling the He production in the core, suggested.

WP2 described the state of the art of production technology for Pu-bearing coated particle fuels and experiences from irradiation performance of these fuels in Europe. Results on release of He from irradiated UO_2 and MOX fuels as well as fresh Pu and Pu+Am fuels were reported. It was concluded that at HTGR operating temperatures, helium would almost completely be released from the kernel to the buffer layer. It would however not be released from the coated particle. Remarkably, aged coated particles exhibit an extremely high resistance to temperature. To model the stress in HTGR coated particle fuel, a stress analysis code has been developed. Also, thermo chemical properties of fuel kernels were modeled.

In the WP3, technical and socio-political characteristics of HTGR deployment, as well as future roles of HTGRs in a European nuclear park were described. Three fuel cycle codes were used and various deployment scenarios considered. As far as overnight costs are concerned, higher fuel burnup and higher thermal efficiency provide a significant economical benefit in comparison to light water reactors. Economics analysis of HTGR also favors modular construction due to lower risk to capital investment compared to a single large plant.

The PUMA project was coordinated by NRG and co-funded by the European Commision under the Euratom Research and Training Programme on Nuclear Energy within the Sixth Framework Programme (2002–2006), under contract no. FP6-036457.

2.3.8 ELSY

The ELSY (European Lead Cooled System) project in the 6th framework programme is coordinated by Ansaldo Nucleare. The major objective was to design a 600 MWe lead cooled Generation IV reactor with simplified technical engineering features, making it economically competitive (relative to the sodium fast reactor) and inherently safe. The large absolute expansion of lead makes it possible to design the primary system with a large fraction of natural circulation of the coolant. The high boiling temperature of lead further makes coolant boiling unlikely. Finally, lead is considerably less reactive with water than sodium, making it possible to avoid the use of an intermediate heat exchanger loop. Important technical challenges to be resolved include corrosion protection of structural materials and, in particular, finding an erosion resistant material for the pump impeller. The primary system design of ELSY is illustrated in Figure 2-6.

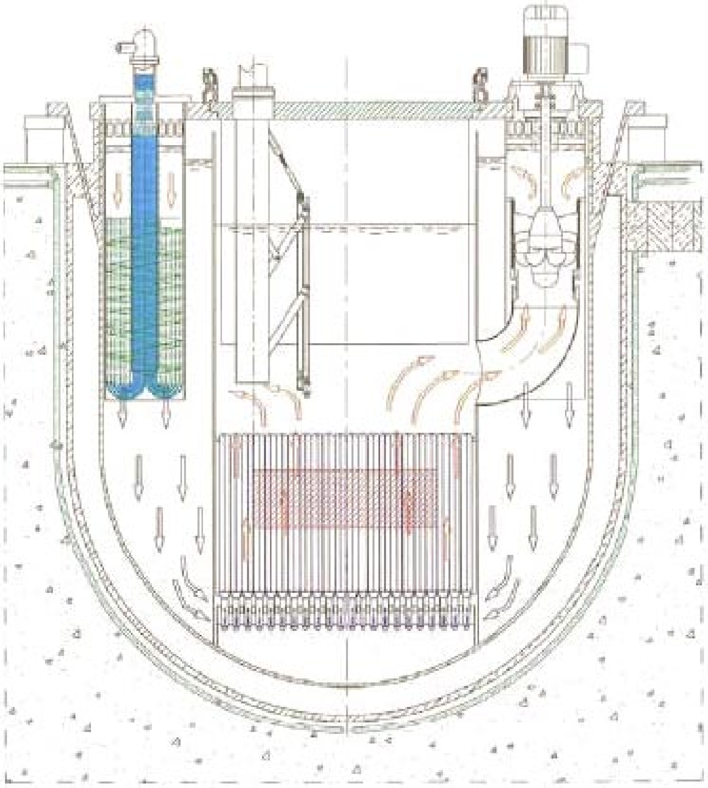

Figure 2-6. *Primary system design of ELSY.*

Two different core geometries have been investigated in detail: a wrapper free square assembly design brought forward by ENEA /Sarotto et al. 2007/ and a more traditional hexagonal assembly design made by SCK-CEN /Sobolev et al. 2009/. Using mixed oxide fuel, the reference designs rely on three fuel-zone cores. The square assembly design features plutonium content ranging from 13.4% in the inner zone to 18.5% in the outer. This plutonium content results in a conversion ratio (in terms of Pu-239 equivalent reactivity) equal to 1.0 and a positive reactivity swing of 700 pcm over three years. The fuel column height is 90 cm, the core contains 170 fuel assemblies, each containing 428 fuel pins. The average linear rating is 23 kW/m, and the core inlet/outlet temperatures are 400/480°C, respectively. The hexagonal assembly core contains 427 fuel assemblies with 216 fuel pins. The suggested plutonium content ranges from 14.9% to 17.4% and a negative reactivity swing of 700 pcm is calculated over three years of operation.

An alternative design based on the use of nitride fuel has been made, showing that the active core height in this case can be reduced to 70 cm, while maintaining a conversion ratio close to unity.

2.3.9 CANDIDE

The CANDIDE project (Coordination Action on Nuclear Data for Industrial Development in Europe) concerned nuclear data for such future reactors, with emphasis on Accelerator-Driven Systems (ADS) and Gen-IV type reactors. The project was carried out 2007-08 and was coordinated by Jan Blomgren, Uppsala University.

The primary importance of nuclear data is cost-cutting, i.e. with precise nuclear data, future reactors can be designed to reach high safety standards in a cost-effective manner. The project team was composed of key actors in the field, ranging from industry to academia and research centres. The industry partners defined the needs from the end-users' perspective, and their participation guaranteed that the work was application-oriented. The role of the non-industry partners was to assess the possibilities to provide data of sufficient quality to meet the needs of the technical application. The project involved reactor manufacturers (AREVA, France), electricity utilities (EDF, France and TVO, Finland), nuclear fuel producers (BNFL/Nexia, UK) as well as universities (Uppsala, Sweden and Budapest, Hungary) and research centres and technical support organizations (CEA, France; JRC-IRMM, EC; NRG, Netherlands; NRI, Czech republic; SCK·CEN, Belgium; CIEMAT, Spain and ITN, Portugal). Efficient dissemination has been guaranteed by close contacts with the IAEA and OECD/NEA Data Banks.

The major scientific endeavour of the project was to assess the present state-of-the-art of relevant nuclear data and to identify important areas where improvement can be made. The final outcome of the project is a report providing recommendations for future research in the field /Koning et al. 2009/.

A course on nuclear data is a major issue concerning improved training in the field. CANDIDE has therefore organized a course on nuclear data for young professionals. It is intended to provide a template for a regularly recurring event. Besides training, the course will contribute to European integration. The EXTEND course (European school on Experiment, Theory and Evaluation of Nuclear Data) took place in Budapest, Hungary during September 1–12, 2008. The course has been evaluated, resulting in very high scores from the participants.

In the assessment of nuclear data for future reactors, the first year was mainly devoted to identifying needs of future industry actors as well as data needs for foreseen research installations in the field. In parallel, work has been undertaken to review current best practices and methods for data production, as well as recent developments and near term prospects for improvements in nuclear data production.

In the second year, the work was focused on how to meet the demands identified during the first year. An important aspect of this is assessment of the status and evolution of resources. It is a well-known problem in nuclear data research and development that resources are declining. In the face of this trend it is crucial to establish which facilities will be available for measurements, which organisations will be engaged in evaluation and validation activities, and which personal resources will be available. The project team has reviewed the trends and commented on their impact on future nuclear data programs. This task will be of key importance to any prioritisation of new efforts and may result in important recommendations for training and maintenance of competences.

The most important generic conclusions and recommendations for nuclear data development are:

- *A long term commitment to modern nuclear data evaluation should be provided in Europe.* This concerns nuclear data evaluation that implements the latest advances in nuclear physics into high-quality nuclear data libraries for applied use, such as the JEFF-3 library. This should include a complete assessment of the uncertainties and uncertainty correlations in nuclear data (covariance matrices). If accomplished, this will allow better determination of safety and economical margins of both existing and future nuclear systems. This will also make it possible to relate advances in experimental and theoretical nuclear physics to the needs of industrial applications. *A special European targeted action for the production of high quality nuclear data libraries, including covariance data, for materials for advanced reactor design* is needed, to ensure that Europe's position remains at par with our competitors, who have recently taken such measures. This special effort should go well beyond the basic work performed as part of the OECD/NEA JEFF Project.

  Specific high-priority recommendations include:

  - Production of complete nuclear data libraries, including *a comprehensive set of reliable covariance matrices*, using both theoretical and experimental nuclear data information.
  - Development of systematic quality-assured data evaluation and validation methods, which guarantee consistent nuclear data libraries in which new experimental and theoretical information becomes directly and correctly available, for subsequent library updates.

  The production of covariance matrices is an extremely important task, which calls for a dedicated specially-funded action on the part of nuclear and reactor physics experts in close collaboration, rather than a broad collaborative FP project spanning the entire spectrum of nuclear data research. This is an area where targeted support from the EC could help bridge the current gap.

- *Provide and support the facilities that are capable to produce the required nuclear measurements and stimulate high-level measurements on key reactions of interest to advanced reactor development*, especially those measurements that demand higher accuracy than available from nuclear modelling, and critical data that serve as standards for large classes of other measurements. Specific high-priority recommendations include:
  - The stimulation of selected measurements that answer generally accepted high-precision nuclear data needs, such as those identified by the recent NEA SG-26 working group on nuclear data needs for advanced reactors and ADS, as categorized in the High-Priority Request List for nuclear data.
  - Insistence on a "culture change" in experimental nuclear physics, to deliver systematically to the international databases complete documentation of the experiments and all covariance information.
  - The stimulation of new integral measurements to test nuclear data in well-defined reactor-type spectra and to decrease the nuclear data uncertainties in cases where differential measurements do not suffice.

- *Provide the capability for advanced nuclear model development* to address the priority needs that cannot be met due to the lack of experimental facilities or because model calculations can provide certain important data in a more cost-effective way. Specific high-priority recommendations include:
  - Bring the predictive power of nuclear models for actinides to the same level as that for non-fissile nuclides. For this, consistent nuclear fission models and parameters for all important actinides need to be developed and made generally available. This will give the possibility to produce complete covariance data for actinides as well.
  - Development of consistent statistical methods to produce reliable covariance information from both theory and experiment.

- *Ensure flexible implementation of improved nuclear data libraries in nuclear technology and design.* Those companies or institutions that can assure and reduce the cycle time for innovations and quality improvements in nuclear data have a distinct advantage in either research or the industrial markets. This requires a modern approach to reactor software development, in particular regarding the handling of nuclear data. Obviously, the nuclear data community will benefit from rapid and flexible application of their results in actual reactor calculations, and the associated feedback will allow further improvements to be made. Specific high-priority recommendations include:

- Assist reactor code developers in developing easy upgradeable nuclear data library interfaces, for both static and dynamic system analyses. This should be pursued into the area of full-core coupled neutronic and thermo-hydraulic reactor calculations.
- Develop systematic approaches to integral validation and sensitivity studies, to ensure that improved nuclear data (e.g. better covariance matrices) can directly be tested on relevant integral measurements.

In addition to these cross-cutting issues, contemporary analyses of current reactors, GEN-IV reactors and ADS also give rise to specific issues. The most important recommendations for vertical nuclear data development are:

- *Fast neutron actinide cross-sections for both critical and sub-critical reactors.* There are strongly motivated requests for improvement of the nuclear data for ^{238}U (capture and inelastic) and the $^{238–242}$Pu isotopes (capture and fission). More precise measurements, fission model development, and careful data library evaluation including covariance data are called for.
- *Cross-sections for transmutation and target design in accelerator-driven systems.* Transmutation with sub-critical reactors, loaded with minor actinides, coupled with an accelerator are characterized by some specific nuclear data concerns:
  - Specific capture and fission measurements in the 1 eV to 1 MeV range for Am and Cm isotopes.
  - Well-chosen integral measurements for neutrons above 20 MeV.
  - Assessment of uncertainties of high-energy data (> 20 MeV).
- *High burn-up systems.* Increased burn-up scenarios will put a larger emphasis on the quality of fission product evaluations. In order to better assess the neutron absorption rate of the fission products, their cross sections, fission yields and radioactive decay properties need to be improved. Therefore, decay data and fission yield data need to be critically examined and future evaluations be accompanied by both uncertainty and covariance data.
- *Fast neutron cross sections for structural materials and coolants.* Modern nuclear data evaluations and precision measurements of inelastic scattering cross sections are required for important (system-dependent) structural materials, coolants and inert fuel elements (Na, Mg, Si, Fe, Mo, Zr, Pb, Bi). In particular, an accurate determination of the sodium void coefficient of an SFR requires improvements in the inelastic scattering cross sections for ^{23}Na and a complete covariance treatment.

In conclusion, *a substantial long-term investment in an integrated European nuclear data development program is called for, complemented by some dedicated actions targeting specific issues*. It can be expected that, as nuclear analysis and design methods improve, reactor designers will become more demanding in the targeted plant performance, which will result in more stringent requests on nuclear data evaluations, measurements, and validation. To be responsive, it is necessary to retain a critical mass of scientists in a variety of nuclear data related fields to maintain, develop and pass on their skills to the next generation of specialists. There are indications that, over the last few years, we have lost some of our expertise in the area of evaluation and data testing. There is also concern that the situation in this and other areas could rapidly deteriorate if no corrective action is taken. This deterioration will result from experienced people retiring or taking better career opportunities outside of their current research fields, and inadequate funding available to train replacements. As the situation appears to be fragile, it should be regularly reviewed. It is noted that significant enhancements in the nuclear data field can be generated through doctoral level student projects and postdoctoral research. Nuclear data students can additionally be a source for well-educated staff for the nuclear power industry and regulatory bodies, provided these positions are seen as good long-term career options.

2.3.10 EFNUDAT

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. EFNUDAT aims at providing a convenient platform to integrate all scientific efforts needed for high-quality nuclear data measurements in support of waste transmutation studies and design studies for Gen-IV systems that include an objective of producing less waste.

The EFNUDAT consortium groups 10 partners, equipped with nuclear data research infrastructures. The proposal unifies facility management, research community and stakeholders. The aim of EFNUDAT is to integrate all infrastructure-related aspects of nuclear data measurements and to provide access for external user to the participating facilties. Particular emphasis will be given to the following objectives:

- initiate networks leading to a stronger partnership in infrastructure management and exploitation,
- promote access and coherent use of all participating infrastructures to meet the scientific and industrial nuclear data requests,
- merge the complementary nuclear data measurement capabilities and expertise.

UU (initially Jan Blomgren, later Stephan Pomp) has held the role as network coordinator and board member. Thanks to funding via EFNUDAT, several international research groups have got beamtime costs for experiments at TSL covered. In fact, a significant fraction of the beamtime costs for the UU group have also been covered, because these experiments have been run in parallel with experiments of groups from abroad funded via EFNUDAT.

Evaluations of the EFNUDAT project have been very favourable. For instance, the CANDIDE assessment project has suggested a permanent activity of EFNUDAT type as one of the top priorities for successful future nuclear data research in Europe /Koning et al. 2009/.

2.4 EU-studies of partitioning

During the last decade and more several European framework programmes have been dedicated to the partitioning of nuclear waste for transmutation purposes. On the hydrochemical route they have been NEWPART (1996–1999) /Madic et al. 2000/, PARTNEW (2000–2003) /Madic et al. 2002, 2004/, EUROPART (2004–2007) /Hill et al. 2007/ and presently ACSEPT (2008–2011) /Bourg et al. 2009a/. During these years we have seen a change from the almost impossible task of separating trivalent actinides from lanthanides passing too high separation (extraction) and finally finding molecules with appropriate separation factor and distribution values /Ekberg et al. 2008/. Partially in parallel to these project investigations on pyrochemical partitioning processes have been performed PYROREP /PYROREP 2003/. From EUROPART and on the two different strategies have been treated within the same project.

2.4.1 EUROPART

The purpose of FP6 Integrated project EUROPART (FI6W-CT-2003-508 854) /Hill et al. 2007/ was to develop hydro- and pyro-metallurgical processes for the selective removal of actinides from spent nuclear fuels and their further conversion into precursors for new fuel fabrication, in order to reduce the radiotoxic inventory of the final wastes to be disposed of in deep geological repositories.

Among the hundreds of new ligands investigated (belonging to the families of functionalized calixarenes, diamides, tripodes, etc) a diglycolamide (TODGA) was successfully tested on a genuine PUREX raffinate for the co-extraction of actinides and lanthanides /Magnusson et al. 2009c/. Promising nitrogen ligands (BATBP) were designed for actinide/lanthanide separation, but they still require process optimisations.

In pyro-chemistry, lots of valuable thermodynamic data were acquired in molten chloride media, allowing comprehensive studies of actinides, lanthanides and other fission products. Two efficient processes were selected for the separation of actinides from lanthanides: *(i)* electro refining process on solid aluminium cathode in molten chloride, *(ii)* liquid-liquid reductive extraction in liquid aluminium-molten fluoride. Some reference flow sheets were assessed to help identify key issues and orientate R&D programs in the future.

Two methods were proposed to convert actinides from the partitioning solutions into fuels: co-precipitation and sol gel routes (internal and external gelation, and colloidal sol-gel processes).

From the hydrochemical point of view the process considered in and as a legacy of EUROPART is described in Figure 2-7.

Then general principle is that the uranium and plutonium (and possibly neptunium technetium and iodine) are separated from the waste in a modified PUREX process /McKay 1956/. The raffinate from this process is then transferred to the DIAMEX (DIAMmide EXtraction) (a process where the trivalent actinides and the lanthanides are separated from the rest of the fission and corrosion product by the use of e.g. a malondiamide DMDOHEMA) /Madic et al. 2002b/. The trivalent actinides are then separated from the lanthanides in the SANEX (Selective ActiNide EXtraction) process using e.g the BTP or BTBP class extractants /Madic et al. 2004/. The actinides are then supposed to be separated from each other in the SESAME (Selective Extraction of Americium by Electrochemical Method) process /Adnet et al. 1996, Donnet et al. 1998/. This was never realised within EUROPART other than as a biproduct of the development of actinide selective ligands like BODO /Andersson et al. 2003/. In all these processes the reference organic diluent is kerosene, octanol or similar aliphatic hydrocarbon. The actual hot test with an optimised SANEX process on actual PUREX raffinate was not realised until after EUROPART but then it was shown that the suggested process using CyMe4-BTBP in octanol worked /Magnusson et al. 2009b/. The major difficulty was the kinetics of extraction, which in this system is a bit on the sluggish side.

The pyro process at large was considered to follow the layout as described in Figure 2-8.

As outlined above two different areas were considered as the core of the project, i.e. electro refining and liquid-liquid extraction using molten salt and molten metals according to Figures 2-9 and 2-10.

The research was thus focussed on getting enough basic data to understand and design the processes designed above. It became clear that even after the finishing of EUROPART more data was needed before process testing could commence.

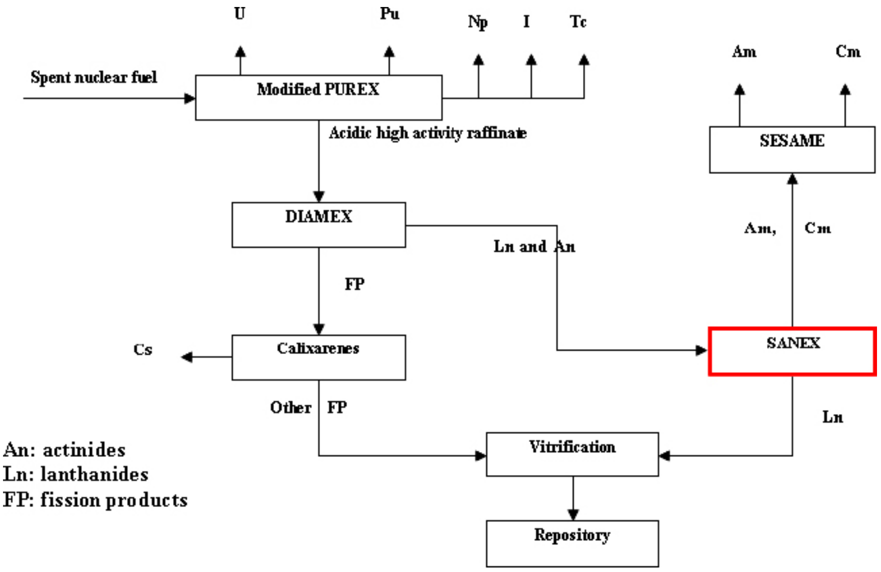

Figure 2-7. *Hydrochemical separation scheme in EUROPART.*

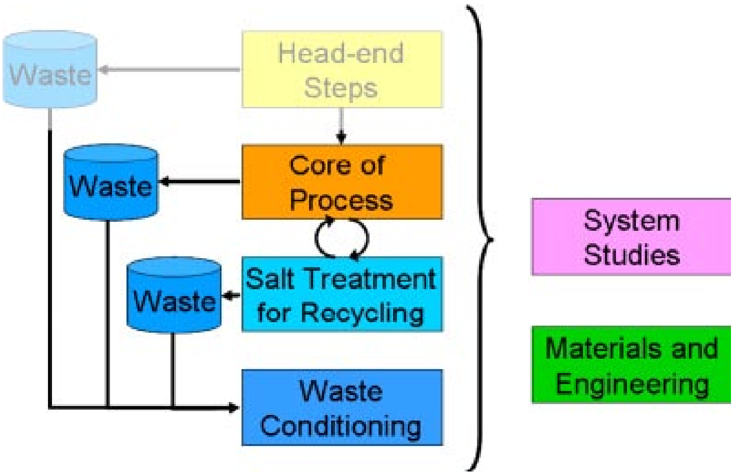

Figure 2-8. *Proposed pyrochemistry route.*

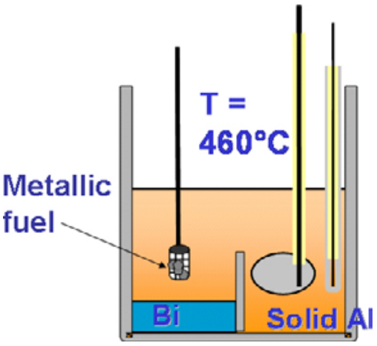

Figure 2-9. *Electrorefining on Solid Aluminium Cathode in molten chloride.*

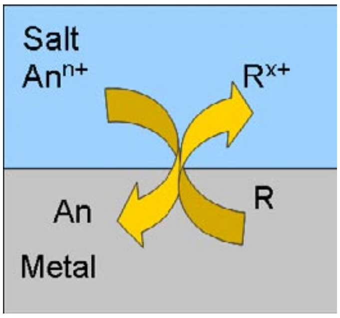

Figure 2-10. *Liquid-Liquid Reductive Extraction in molten fluoride.*

2.4.2 ACSEPT

The research on partitioning is following two main routes – hydrochemical processes and pyrochemical processes. In Europe the efforts are coordinated within the EU-project ACSEPT (Actinide Recycling by Separation and Transmutation) that started in 2008 and shall be completed in 2012. The project has an overall budget of 23.8 M€, whereof the EC contributes 9.0 M€. The project is a so-called collaborative project within the seventh framework programme sponsored by the European commission. It constitutes a continuation of the project EUROPART within the previous framework programme. ACSEPT is divided into four domains:

1. Hydrometallurgy (also denoted water chemistry)
2. Pyrometallurgy (also denoted pyrochemistry)
3. Process development
4. Training

Sweden is represented by the nuclear chemistry group at Chalmers in Göteborg. The ACSEPT project started in the spring of 2008 and has thus been active for just over one year. Very early in the project, as stated above, it was demonstrated that the classical SANEX process did work on genuine high active raffinate from the PUREX process /Magnusson et al. 2009b/. In principle the ACSEPT project is divided into four domains: Hydrometallurgy, Pyrometallurgy, Process and intergration and Teaching and education. The actual planning for the project is oulined below /Bourg et al. 2009b/.

Two strategies are proposed for the recycling of the actinides issuing from the various forms of future nuclear fuels (oxides, carbides and nitrides or metallic fuels): *(i)* their homogeneous recycling in mixed fuels (via a prior group separation of the actinides) and *(ii)* their heterogeneous recycling in targets or core blankets (via their selective separation from fission products).

Two major technologies have been explored so far to meet these challenges:

- Hydrometallurgical processes that benefit from more than 60 years of research and developments and a long-lasting proven experience at the industrial level.
- Pyrochemical processes first studied in the 50-60s for the treatment of spent fuel from Molten Salt Reactors and Breeder Reactors and more recently, with a renewed interest at the end of the 80s, for specific applications, but without reaching the industrial development level.

Hydrometallurgy

Many countries have developed over the past six decades hydrometallurgical processes to recover minor actinide elements (neptunium, americium, curium and higher actinides) in order to allow these metals to be used and to decrease the radiotoxic inventories of nuclear waste. However, none of these processes has ever been implemented at the industrial scale but the R&D has sometimes reached demonstration tests at the laboratory scale. Most of the partitioning strategies rely on a three-step approach:

- separation of U (and sometimes also Pu) from used fuel dissolution liquors,
- trivalent actinide and lanthanide co-extraction,
- trivalent actinide and lanthanide separation, this step being the most difficult of the three because of the similar chemical properties of these elements.

The processes developed around the world differ from the extracting systems involved in these different steps. To optimise these processes there is also the possibility to shrink two of these steps into a single one.

In the first step, tributyl phosphate (TBP) will be used. This substance is today used within the PUREX, UREX /Law et al. 2001/ and COEX /Baron 2009/ processes, developed in Europe and the US. As these processes are well advanced in these countries, ACSEPT does not intend to put any resources on this topic.

In the second step, malonamides, CMPO and TODGA are used in the DIAMEX, TRUEX /Alexandratos et al. 1993/ and ARTIST /Tachimori et al. 2002/ processes, respectively developed in Europe, US and Japan. TRPO (trialkyl-phosphine oxide) developed in China or UNEX (mixture of several extractants) jointly developed in Russia and the USA allows all actinides from U to Cm to be co-extracted from the dissolution liquors. The TALSPEAK /Weaver and Kappelmann 1968/ and reverse TALSPEAK processes are still the focus of intense research in the USA. The baseline of ACSEPT for this topic is clearly the achievements reached in the EUROPART project with the demonstration of the scientific viability of the An(III)+Ln(III) co-extraction using the TODGA extractant on a PUREX raffinate /Magnusson et al. 2009c/. All the progress in terms of optimization of the process will be measured against this baseline.

Finally, for the third step, a number of soft donor atoms containing extractants, such as polyaromatic nitrogen ligands (BT(B)P) or dithiophosphinic acids have been developed in Europe, China and Japan to selectively extract the trivalent actinides. Other soft donor atoms containing ligands, such as polyaminocarboxylates (HEDTA, DTPA) have been tested in France, the US (TALSPEAK /Weaver and Kappelmann 1968/) and JAPAN (SETFICS – Solvent Extraction for Trivalent F-elements Intragroup Separation in CMPO Complexant System) to selectively strip the trivalent actinides.

The current BTBP reference system developed during EUROPART, see above, for An(III) selective extraction does not fulfil all process development requirements but may be considered as the baseline where the project work could start from. Progress must be made in the following areas: stability against radiolysis, the rate of back extraction of americium An(III) (stripping) and process simplification to reduce the number of steps required. A reduction in the number of steps could result in a considerable reduction in the capital investment required to build an industrial plant, the size of the plant, the running costs and the final decommissioning costs.

In parallel, a new approach will be investigated: the selective stripping of trivalent minor actinides after their extraction by existing molecules (e.g. malonamides, DGA). The baseline from which this research will move forward from comes from the achievements of France, USA and Japan, and this concept has never been studied before in the previous EU framework projects. This idea is an alternative route if the An(III) selective extraction cannot be developed into a viable process.

The actinide group separation (i.e. Am-Cf) from used fuel dissolution liquors (probably after a bulk U partitioning cycle) has only been scarcely investigated in previous projects, and many challenging technological obstacles (e.g. solvent loading capacity, oxalate substitute) will have to be overcome before demonstration tests can successfully be carried out.

For each of these concepts for separation processes, the first milestone to measure the progress of research and development is clearly the selection of potentially successful extracting system based on the acquisition of extracting properties. Then, the second milestone is the decision to design flow sheets before going to experimental tests and this decision depends on the results obtained during the optimization of the extracting systems. The last milestones to measure the progress are directly linked to the success of the successive cold, then spiked and finally hot tests in terms of An recovery and decontamination factor against Ln.

Whereas chemical separation processes represent a common trunk of operations that are relatively independent of the nature and type of fuel treated, head-end operations, conversion of actinides from liquid to solid state are at the interface with the fuel, either for its fabrication or its treatment once used. Dissolution and conversion are fuel-dependent operations. In Domain 1 (DM1) dissolution studies will mainly focus on fuels dedicated to the actinide homogeneous recycling.

Although current generation of fuel forms are generally oxides, future fuels may be manufactured as oxide, carbide, nitride or metallic form. Features that all the future fuels envisaged for the homogeneous recycling have in common is that the Pu and minor actinide contents (respectively ~ 20 wt% and 5 wt%) will be higher than that normally encountered in current oxide fuels. The burn-up, and hence heat load and activity, of these fuels will also be much higher and will present new challenges in fuel handling and processing. In addition, the matrix and Pu content of the fuels may result in increased levels of Pu-rich insolubles, however, losses of TRUs to residues must be minimised to maximise the benefits gained from P&T fuel cycles.

The measure of the progress will be linked to successive milestones: the selection of technologies for advanced fuel dissolution, the feasibility of enhanced dissolution methods for recovery of Pu from residues and the feasibility of enhanced dissolution methods for treatment of advanced fuels.

The co-conversion of variously mixed actinide aqueous products into oxide, carbide or nitride powders, as starting materials for new fuels or targets will be the last step to close the loop. The objectives of the research to be carried out in this field are related to the development of methods for the co-conversion of mixtures of many actinides to solids containing many actinides. These mixed solids will be used for fuels for the Generation-IV (Gen-IV) reactors. Already mixed uranium and lanthanide oxides such as europium uranium oxide are known, it is possible that in the future mixed oxides containing both uranium and trivalent transplutonium metals will be made and used as fuels.

To reach these objectives, different co-conversion processes have been identified, such as co-precipitation and sol-gel routes. In addition, new alternative routes such as simultaneous denitration and conversion by impregnating solids, followed by thermal treatment, have shown promising initial results but have so far been less studied.

For all these processes, a common feature is the complexity of the initial mixed actinide containing solution, and there is a need to cover basic studies to understand and master the co-conversion processes. Furthermore, as co-precipitation processes are already well studied and advanced under national programmes, it has been decided to bring efforts on promising alternative routes that show interests for new fuels/targets and innovative fuel fabrication processes.

The measure of the progress will be linked to successive milestones: the selection of the conditions for stabilizing the oxidation states of polyactinide mixed solutions, the selection of potential co-conversion technologies and finally the fabrication of solid precursors, either powders or beads, from these selected technologies.

The main objectives of the hydrochemical domain will thus be to develop aqueous chemical processes in the various fields of used fuel dissolution treatment, separation process development and fuel precursors elaboration, and to demonstrate by the end of the project their technical feasibility at the laboratory scale on actinide solutions.

Pyrometallurgy

For more than 50 years, pyrometallurgy has been studied as an alternative strategy in the reprocessing of spent fuel. Indeed, it is considered as the reference route for molten salt reactor fuel reprocessing, and for the reprocessing of some types of fuels today envisaged for Gen-IV reactors that might not be compatible with current hydrometallurgical processes (metallic fuels for instance). Since pyrometallurgy also allows for reprocessing of high burn-up fuels/targets with short cooling times, it has also been considered as a potential technology for the recycling of high content minor actinides bearing targets, for instance in the ADS. As stated above, in FP5 PYROREP and in FP6 EUROPART projects, European scientists involved in the treatment of nuclear wastes, have carried out studies and research programmes together to increase the level of knowledge in pyrometallurgy with respect to process development, specific waste treatment and confinement.

A pyrochemical process currently includes several basic steps: separation of the fuel from the cladding material; dissolution of the material in a molten salt (except in the case of the electrorefining where no dissolution step is necessary); chemical or electrochemical reduction of oxide to metal; separation of the desired elements by electrowinning; transfer to an immiscible liquid metal phase or selective precipitation. These processes include not only a core process but also numerous ancillary operations: head-end steps, conversion of final products to ensure compatibility with subsequent refabrication techniques, decontamination of salt and metal fluxes prior to recycling and conditioning of specific waste materials in accordance with their chemical and radiological properties.

In PYROREP and EUROPART, the effort was put on basic data acquisition mainly in molten chloride and on core processes assessment. Two promising core processes were developed and assessed: electrorefining on solid aluminium in molten chloride and liquid-liquid reductive extraction in molten fluoride/liquid aluminium. In parallel, progress was made in the decontamination of spent chloride salt by fission products precipitation or filtration. Some original confinement matrices were also studied for waste conditioning. Finally, integration studies have been initiated in order to assess and to compare some selected process flow sheets and possibly to redirect R&D programs. These elements clearly represent the baseline against which progress and improvements will be measured.

Consequently, important technological blocks were identified as key scientific points to be studied in ACSEPT: head-end steps must be developed in fluoride and optimised in chloride. Some ancillary steps of the core process must be assessed (exhaustive electrolysis in chloride, actinide back extraction from aluminium). The need of an electrochemical process in molten fluoride as an alternative route has also been pointed out and important efforts will be devoted to this route. Progress is still expected in salt recycling (optimisation in chloride and development in fluoride) and specific waste conditioning (establishment of reference routes).

All these activities concentrated in Domain 2 (DM2) will allow making significant advances beyond the current state of the art in pyrochemical separation processes They will also bring to DM3 lab-scale tested technological blocks that will permit the elaboration of validated reprocessing schemes that could finally lead, in the future, to integral experiments and ultimate lab-scale demonstrations on genuine spent fuel. This work takes into account the diversity of the possible fuels of ADS or Gen-IV reactors, with a view to future demonstration at a pilot level at a longer term than the current hydrometallurgical processes. All the above expected scientific and technical advances will be measured by passing successfully different milestones.

Process and integration

Over successive European Framework Programmes novel partitioning technologies have broadly been demonstrated to be technically feasible at the laboratory scale with some current demonstrations performed with irradiated spent fuel. There are, however, significant challenges to tackle in turning the basic research and development into a deployable process at the pilot plant scale. Furthermore, there are challenges to face in process development and in integration of technology within the nuclear fuel cycle. The studies proposed in ACSEPT will deploy tools which will enable early feedback and guidance to lab scale studies. These feedback mechanisms can be used to positively select candidate separation processes for focused development, to refine the axis of research, and more generally to provide focus and rationalisation to the field of research. It thus directly benefits to the development and future implementation of the technologies, incorporating and using input from all contributing partners. This approach has been for the first time implemented and successively tested on pyro-chemical processes within EUROPART and will be extensively applied within DM3. Technical seminars for and by experts will be organized in the first year of the project to spread the knowledge among researchers and guarantee a good understanding and appropriation of the separation criteria to be fulfilled prior to developing processes.

A specific activity of ACSEPT will be devoted to emphasize integration between partitioning and transmutation.

ACSEPT will then endeavour to assess different concepts of fuel/target reprocessing operations in strong connections with fuel/target fabrication, and providing feedback on fuel and target so as to prepare future experimental programmes in a coherent P&T scheme.

These objectives are planned to be achieved by the following research activities:

- Design the fabrication technology beyond FP6 by the technologically very demanding fabrication and irradiation of Curium-containing pellets (CURIOS).
- Inert matrix fuels and targets, specifically designed for the purpose of minor actinide transmutation, are significantly different in composition compared to ordinary reactor oxide or mixed oxide fuels.

Several different types of targets and fuels are under investigation in FP6 programmes. The reprocessing aspects of the irradiated actinide targets have not been systematically studied before and ACSEPT thus proposes to address these challenges. ACSEPT contribution in this field will be limited to theoretical studies to evaluate and rank different types of targets with regard to their reprocessing capability and outline the potential key scientific issues that may be further studied in a future experimental programme. At t_0+12 months, an important milestone is expected in this respect.

Training and Mobility, Dissemination of Knowledge

ACSEPT project will seek to advance the integration of European education and training in the field of separation techniques, and actinide chemistry in particular, to combat the decline in student numbers and teaching establishments, young researchers thus providing the necessary competence and expertise for a sustainable development of nuclear energy.

DM4 will seek to improve not only motivations and skills of people but also make knowledge in the field of nuclear chemistry more transparent, improving the teaching methodology and providing an infrastructure that will support co-operative work among the members of the ACSEPT nuclear community. Of course strong links will be established with the on-going ACTINET NoE direct or derived actions or with any post-ACTINET initiative. Complementarities will be obviously sought so as to optimise EC funds.

The overall goal of education and training programme in ACSEPT is to enhance the knowledge in separation sciences within the participating community. A variety of skills and equipment exist within the consortium and thus cross-cutting education is one of the better forms of teaching. In addition to that also researchers outside the direct community should be given the possibility to learn from the experiences gained within ACSEPT. Therefore the main activities and budget allocation will rely on the mobility of personnel.

2.4.3 ACTINET

ACTINET-6 was funded as a “Center-of-Excellence” and has now been finished. ACTINET was a consortium gathering more than twenty-five European research institutions, and was dedicated to actinide sciences. The consortium was supported by the European Commission under a four-year contract established in March 2004 within the sixth Framework Program for Research and Technological Development.

Although the nuclear industry has now come to maturity and operates nuclear plants with a high level of safety and efficiency, some issues in the field of the back-end cycle and resource management remain open. Research and Development are needed to explore new concepts for nuclear energy generation that make better use of fissile material and generate less waste. One other major issue requiring intensive Research and Development programs remains a broadly agreed approach to waste management.

All these issues require expertise and improved knowledge of the processes involved in the behaviour of nuclear fuels in reactors, spent fuel treatment, as well as a comprehensive understanding of the processes governing the behaviour of high level waste in storage and disposal systems.

Advanced research in actinide sciences is therefore recognized as one essential endeavour for further development of a dynamic, competitive and sustainable knowledge-based nuclear industry.

The objective of ACTINET was to take steps in order to bring both research infrastructures and human expertise in Europe to an adequate performance level, thereby contributing to the development of the European Research Area in the fields of physics and chemistry of actinides.

The three lines of action were taken:

- Stimulating the emergence of a European infrastructure policy by pooling the major actinide laboratory facilities
- Promoting excellence by supporting ambitious shared research programs, taking advantage of the pooled facilities
- Increasing the attractiveness of actinide sciences among European students and young researchers, and allowing the next generation of actinide scientists and engineers to gain hands-on experience as part of their training.

Swedish scientists and research institutions participated in ACTINET-6: The group from CTH with a program dealing with actinide – lanthanide separations “The characterization of complexes formed during the kinetics experiments and irradiation experiments in organic systems containing 6,6’-bis(5,6-R-1,2,4-triazin-3-yl)-2,2’-bipyridine like molecules”. The project has so far resulted in three publications /Fermvik et al. 2009b, 2009c, Retegan et al. 2009/. In addition there are a number of articles in preparation.

The Wallenius group at KTH has been engaged in a number of projects ranging from thermodynamic data bases to studies of advanced fuel compositions.

Ingmar Grenthe has been a member of the scientific advisory group throughout the ACTINET-6 program.

The scientific achievements of the entire program has been judged very good and the program will be continued under the 7th framework activity in a slightly different form and the application under the title "ACTINET-3I" has been granted, but the research activities have not yet started. In the evaluation of ACTINET-6 it was pointed out that the link between the research groups and the end-users was too weak and this has been taken into account in the planning of the follow-up program.

2.5 Development in France

The activities in France are undertaken within the framework of the Act of June 28, 2006 (a sustainable management of nuclear materials and waste) that envisages a 15-year plan for the further development of the French nuclear programme. In short, the act is based on the following fundaments:

- A national plan for managing nuclear materials and radioactive waste should be presented by 2021.
- Long-term funding of radioactive waste management is guaranteed.
- A stepwise program for long-lived waste (high and medium activity) management along various approaches should be pursued:

  Partitioning & Transmutation:
  - 2012: Assessment of Fast Reactors / ADS, resulting in decision on technology choice.
  - 2020: Fast Reactor Prototype should be in operation.

  Retrievable Geological Repository:
  - 2015: Authorization decree.
  - 2025: Beginning of operation.

  Interim storage:
  - Creation of new facilities in 2015.

It can be noted that with such a time frame, the only realistic FR technology to launch is sodium-cooled fast reactors (SFR), because extensive experience has been collected during previous PHENIX-SUPERPHENIX operation, as well as at other similar reactors. Thus, this strategy forces a design of a new French SFR, ASTRID, to be completed by 2010, aiming at operation in 2020. Recently, SNETP has adopted a Strategic Research Agenda (SRA), which essentially replicates the French strategy concerning FR development.

At the Atomic Energy Committee meeting December 20, 2006, aspects of the FR/ADS research were detailed. Two types of fast reactors will be studied in parallel, sodium-cooled and gas-cooled reactors. Concerning the fuel cycle, three different options are considered:

- Recycling of U and Pu only. In that case MA are not dealt with further.
- Heterogenous recycling. In this case uranium and plutonium constitute one stream that is indefinitely recycled in FRs, whereas americium constitutes another stream that is incinerated in ADS. This scheme is very similar with the double-strata concept that constitutes the reference case in Japan (see section 2.7).
- Homogenous recycling, i.e. uranium, plutonium and minor actinides are re-cycled together in fast reactors of Gen-IV type.

Options should be kept open for R&D and demonstrations in the 2020s. In the report of 2021, an assessment of ways for a stepwise implementation should be presented.

In 2012, a report on the further development of the program should be presented. It is envisaged that scenario studies are undertaken to elucidate possible development lines. Some results of such ongoing scenario studies, performed in collaboration CEA-EdF, have recently been presented /Carré and Delbecq 2008/. The study is using the global scenario study by the World Energy Council made in 2003 as reference. Main results of the WEC report that are underlined by the CEA-EdF study are:

- A possible future dramatic increase in the LWR fleet could be hampered by scarcity of uranium. Furthermore, waste management could be a societal issue.
- With present-day recycling technologies, i.e. recycling plutonium once as MOX fuel, only about 10% savings in natural uranium consumption is gained.
- With multiple recycling of plutonium in fast reactors, the resources of natural uranium can be utilized 100 times better. There is, however, a risk of initial shortage of suitable plutonium from LWRs for fuel to FRs if FRs are introduced rapidly on a large scale. Moreover, the partitioning capacity might be insufficient for a large-scale FR deployment.
- LWRs will remain important throughout the 21st century. The most important reason to deploy FRs is to meet uranium scarcity. There is also room for more sustainable LWRs in a symbiotic fleet, if it can be proven to constitute an industrial option (after 2030).

The plutonium availability is very different in different countries. This motivates regional/national scenario studies. The ongoing French study will concentrate in the coming three years on a set of scenarios for the French reality. A complete set of new scenarios will be studied by 2012 to provide a report to the French Parliament.

Of special interest in the present report are the considerations of MA handling. Some limits of transmutation strategy are to be considered. It is considered that MA transmutation efficiency is potentially limited by:

- The amount of minor actinides vitrified before the implementation of the transmutation (~ 90 tonnes in 2040 in France).
- The amount of transuranics (mainly Pu) to be disposed at the end of nuclear industry (or at a chosen date to compare the different strategies).

It is pointed out that MA recycling is very challenging and therefore the impacts on all the facilities have to be assessed:

- Constraints on treatment facilities. The high activity might imply shielded fuel fabrication facilities, or even remotely operated facilities.
- Ageing of MA spent nuclear fuel in storage. For instance, the α activity of Cm can be cumbersome.
- Transportation cask design. Fuel cladding temperature and dose rate have to be considered.
- Fuel handling in reactor. Various aspects, like time before unloading and transportation (thermal load), impact on plant availability, impact on reactor safety (void coefficient in particular) have to be taken into account.
- The potential benefit of MA recycling on proliferation resistance will also be evaluated (strengthening of radiation barrier against diversion of nuclear materials, easier detection of nuclear materials, etc).
- Industrial feasibility of MA recycling and its cost (direct costs at every step of the whole fuel cycle, including disposal and reactors, and indirect costs such as the plants availability), compared with the cost of the reference option.
- Societal criteria will also be assessed in a more qualitative way (acceptability).

In parallel, EdF has adopted a strategy for renewal of the French nuclear fleet that can be summarized as follows. In the mid-term, two strategic complementary lines are pursued:

- Extending the existing reactors lifetime beyond 40 years.
- Preparing the fleet renewal beyond 2020 with the launching of EPR reactors (Flamanville 3 in 2012, Penly to follow).

In the long term, a two-step flexible approach is foreseen:

- To initiate this renewal (~ 2020) with earlier tested Gen-III reactors (i.e. additional EPRs).
- To introduce fast reactors (Gen-IV) by 2040, *if needed,* in a worldwide context resulting in an increased appeal to nuclear energy (sustainability).

Another scenario could be Gen-IV deployment by 2080, i.e. LWRs need to be the backbone of nuclear energy production for the entire century in that case.

2.6 Development in USA

In general, the policies on P&T have been rather volatile in the US during the last three-year period. Partly this reflects the change of administration early 2009, but also within the same administration notable changes in direction can be seen.

2.6.1 Transmutation

Global Nuclear Energy Partnership

The Global Nuclear Energy Partnership (GNEP) was launched during the Bush administration. The main motivation of GNEP is a strive to facilitate increased use of nuclear power globally, and simultaneously curbing illicit use of nuclear technology /McCarthy 2007/.

A main feature of GNEP has been internationalization of the nuclear fuel cycle. The aim has been to establish an internationally accepted mode of operation that would ensure easy access to fissile material for civil purposes and simultaneously discourage illicit use by making diversion and misuse of fissile materials more difficult, more costly, and acquisition of sensitive fuel cycle technologies more difficult to justify as part of a peaceful nuclear program. This should be accomplished by identification of three separate and distinct actors with separate functions in the system. *Fuel Suppliers* operate reactors and fuel cycle facilities, including fast reactors to transmute the actinides from spent fuel into less toxic materials. *Fuel users* operate reactors, lease and return fuel. *IAEA* provides safeguards and fuel assurances, backed up with a reserve of nuclear fuel for states that do not pursue enrichment and reprocessing.

To reach the nonproliferation goals, GNEP advocated the U.S. policy at that time of discouraging separated plutonium. As a consequence, there was a focus on aims to reduce this risk by pursuing used fuel recycling that does not produce separated plutonium. This should be accomplished by actions to discourage spread of enrichment and reprocessing capabilities, minimization of any further buildup of plutonium and eventually draw down existing stockpiles, improvement of proliferation resistance of recycling technologies, and development and incorporation of advanced safeguards. An essential part of GNEP is a pledge that U.S. will work with domestic and international industry to prove the recycling technologies needed to close the fuel cycle, minimize waste, and obtain more energy benefits.

Finally, it was pointed out that a global expansion of nuclear energy has begun, and moving to a closed fuel cycle was deemed a natural evolution that is necessary for better utilization of the uranium resources, reduced proliferation risk and reduced long-term environmental burden from nuclear waste. It was claimed that once the GNEP fuel cycle becomes the norm, it will reduce the credibility of a country's need for its own enrichment or reprocessing capability for peaceful purposes.

Advanced Fuel Cycle Initiative

Work in the USA on future nuclear cycles have been coordinated within the Advanced Fuel Cycle Initiative (AFCI), funded by the Department of Energy (DOE) and managed by the Idaho National Laboratory (INL). The Systems Analysis Campaign has been devoted to analyzing fuel cycle options, with the aim to provide guidance on system performance during transition to a closed fuel cycle. Some of the major findings are presented below /McCarthy 2008/:

- *Global demand for all energy will grow, and the global growth for nuclear energy will increase with or without CO_2 limits.* The global electricity consumption is estimated to increase 5-fold until year 2100. At the same time, nuclear power is expected to expand its global electricity market share by 25%. As a result, nuclear growth will challenge uranium and waste disposal resources.
- *Limiting CO_2 levels results in less fossil energy, and more nuclear power and renewables.* Carbon capture and sequestration technologies are key to fossil market shares. The more aggressive the CO_2 limits, the greater the importance of nuclear.
- *Nuclear energy is competitive with other sources with or without CO_2 taxes.* Recycle does not change this finding.
- *Domestically, nuclear energy is competitive with fossil energy.* The once-through cycle is judged to be potentially less expensive than coal, whereas closed fuel cycles are competitive with coal. Natural gas prices have greater uncertainty due to volatile fuel costs.
- *A U.S. carbon tax makes nuclear power more competitive.* Carbon taxes will hit coal the hardest. The uncertainty surrounding carbon taxes increases investment risk for all fossil baseload plants.
- *A closed fuel cycle will likely cost more than the once-through cycle.* Closed fuel cycles appear to cost about 10% more than once-through. Nuclear reactor and fuel cycle costs have, however, large uncertainties
- *Coordination is needed to avoid excess separated material inventories at the start of the transition.* Successful deployment of a fleet of fast reactors relies on the availability of suitable fuel. Thus, the partitioning and fuel production capability must be developed before the introduction of fast reactors in a coordinated fashion.

DOE development strategy

In 2009, DOE announced its development strategy, which was highly focused on gas-cooled reactors in general, and the Very High Temperature Reactor (VHTR) concept of the Generation-IV reactors in particular /DOE 2009/. The program presented the following expectations of the Gen-IV R&D:

- *High-temperature gas-cooled reactor technology.* Gas-cooled reactors are a revolutionary advance in reactor technology[7]. They are inherently safe, efficient, and can use less fuel than the current generation of light-water reactor designs. Gas reactors can be used to extend the benefits of nuclear energy beyond the electrical grid by providing industry with low carbon, high-temperature process heat for a variety of applications, including petroleum refining, biofuels production, and production of feedstock for use in the fertilizer and chemical industries.
- *Underlying Technologies.* Underlying technologies (fuels, materials, neutronic and thermofluid modeling) benefit the majority of reactor concepts and sizes. These technologies will receive limited but sustained Gen-IV support in cooperation with international R&D.
- *Hub for Modeling and Simulation.* The Modelling and Simulation Hub will focus on providing validated advanced modeling and simulation tools necessary to enable fundamental change in how the U.S. designs and manages nuclear facilities. The goal is to find ways to improve waste management, reduce proliferation risk, and lower the cost of nuclear facilities.

The following program accomplishments were foreseen for 2009:

- Continue the gas reactor fuel development, manufacturing, and qualification program. The first of eight irradiation experiments (AGR-1) has survived more than two years without any evidence of failure, and some of the samples will reach 18 percent burnup before the experiment is removed early next fiscal year.

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[7] This statement can be challenged by the fact that many of the early nuclear rectors were gas-cooled.
In particular, the UK nuclear power har until recently been dominated by the gas-cooled Magnox and AGR reactors.

- Complete preparations for the next fuel irradiation test, AGR-2, which contains fuel that was fabricated with production scale equipment, and complete the equipment design for the next series of tests, AGR-3 and -4, that will test fuel with hypothetical fuel manufacturing defects.
- Insert the first of six irradiation capsules to determine the properties of candidate nuclear-grade graphites, while under simulated conditions. Graphite is the primary reactor core structural material, and the Advanced Graphite Creep (AGC) experiment series is designed to qualify one or more commercial graphites for this mission.
- Complete environmental and mechanical property tests for potential Intermediate Heat Exchanger (IHX) metal alloys and continue to support industry-code committees in the development of test standards and design rules for the use of these alloys under Gen-IV conditions.
- Continue analytical computer modeling methods development in both physics and heat transport, and continue benchmarking of computer codes for gas reactor design concepts.
- Upgrade the laboratory-scale components and setup for demonstrating a promising advanced Brayton cycle energy-conversion system that uses supercritical carbon dioxide as the working fluid, and obtain data on the component behavior under supercritical conditions.
- Continue, in collaboration with international partners, the development of crosscutting benchmarking methodologies (economics, proliferation resistance and physical protection, and reactor safety).

For 2010, the plans included:

- Perform post-irradiation examinations of unique, highly-irradiated metallic material samples obtained from the Fast Flux Test Facility and the Phoenix Fast Reactor in France.
- Continue development of advanced materials such as ceramics, composite materials, and nano-structured ferritic materials for use in structural systems, fuel claddings, and other high-temperature applications.
- Complete the first Advanced Gas Reactor fuel irradiation experiment (AGR-1), perform post-irradiation examinations of the irradiated fuel specimens, and commence irradiation of the first fuel produced in large-scale production equipment (AGR-2).
- Continue irradiation of the first Advanced Graphite Creep (AGC-1) test experiment to provide data for nuclear graphite qualification.
- Continue the development of advanced gas reactor system-simulation software and initiate bilateral cooperation with Japan on the use of their experimental gas reactor as a test facility for code validation, operational experience, and as an instrumentation and controls test bed.
- Continue study of the use of liquid salt as a circulating fluid in primary and intermediate cooling loops, and demonstrate the technical and economic viability of an advanced Brayton-cycle energy-conversion system using supercritical carbon dioxide as the working fluid.
- Continue international R&D collaborations through the Generation IV International Forum, IAEA, and bi-lateral agreements.
- Continue advanced modeling techniques utilizing the Department's high-speed, parallel computers for the development of close-coupled neutronic and thermofluid codes.

Recent development

Soon after this program was announced, secretary Chu of the Obama administration – who just had taken office – announced a change of direction. The most important issue was a halt on the deployment of the Yucca Mountain repository. USA has had a once-through policy (like Sweden) since the Carter administration in the late 70's. The freeze of Yucca Mountain development was motivated by an intention to look deeper into the possibilities of a closed fuel cycle.

At the time of the writing, this new policy has not materialized in significant increases in the funding to fast-reactor research. For the fiscal year, a bill on 150 MUSD for Gen-IV research has recently been passed, which is comparable to the most recent previous years. It should be pointed out, however, that the recession in the US has strained the budget with severe cuts in many vital areas, and therefore it is difficult to judge the resolve to pursue Gen-IV research from this particular single year.

US Generation-IV priorities

Two of the six technology concepts identified in the Generation-IV Technology Roadmap have been identified to be the primary research and development candidates, the Sodium-Cooled Fast Reactors (SFR) and the Very-High Temperature Reactor (VHTR). Research and development (R&D) on the SFR is being conducted under the Fuel Cycle Research and Development Program (FCRD). VHTR R&D is being conducted under the Gen-IV Nuclear Energy Systems by the Office of Gas Reactor Deployment.

Recently, the Nuclear Energy division of the Department of Energy (DOE-NE) has announced its strategic goals, as formulated below on the DOE web site:

> While DOE is supporting research on several reactor concepts, priority is being given to the VHTR, a system compatible with advanced electricity, hydrogen production, and high temperature process heat applications. The VHTR concept is being pursued in the United States as the Next Generation Nuclear Plant (NGNP) in accordance with the Energy Policy Act of 2005. The emphasis on VHTR reflects its potential for being economically and safely integrated into industrial applications without emitting greenhouse gases. This fits within the goals of enhancing the security of our energy supply and doing so in an environmentally responsible manner. Fuel cycle options for the VHTR (a thermal-spectrum reactor) are more limited than for fast-spectrum reactors. Fast-spectrum reactors are a potential component in our long-term energy solution. Their mission strengths result from their superior ability to burn recycled nuclear fuel. Closing the fuel cycle by recycling will reduce quantity and radiotoxicity of nuclear waste and increase uranium fuel utilization. The lead laboratory for the Generation-IV program is Idaho National Laboratory (INL).

2.6.2 Partitioning

The work on advanced reprocessing in the U.S. is concentrated on development of the UNEX and UREX+ processes. Much work is devoted to a better understanding of the processes used for the UREX+ process streams, e.g. TRUEX, NPEX, FPEX and TALSPEAK.

/Gelis et al. 2009/ tested the extraction behaviour of actinides and lanthanides in three of these processes, namely TALSPEAK, TRUEX and NPEX. The test was performed with real LWR fuel that had been cooled for 25 years. The result for NPEX was > 99.99 Np and > 99.94% Pu recovery. TRUEX and TALSPEAK also behaved successfully.

/Nilsson et al. 2008/ investigated the influence of soft-donor complexing agents on the separation of Ln(III) from An(III) in the TALSPEAK process. The ligands tested were ethylenediamine tetraacetic acid (EDTA), N-(2-hydroxyethyl)ethylenediamine-N,N′,N′-triacetic acid (HEDTA), ethylenediamine tetramethylene phosphinic acid (EDTMP), diethylenetriamine pentaacetic acid (DTPA) and 2-(phosphonomethylthio)acetic acid (PMTAA). The results indicate that, although DTPA gives the best selectivity, all of the tested molecules showed a preference for Am.

/Grimes et al. 2008/ studied the behaviour of lactic acid in the TALSPEAK process. The results show that lactic acid partitions between the phases in a complex fashion.

/Nash 2008/ has published a thorough summary of the key features of the TALSPEAK process. The paper also discusses the TALSPEAK chemistry demonstrations to process scale, i.e. the CTH, DIDPA and SETFICS processes.

/Gong et al. 2008/ studied the reduction of TcO_4^- by acetohydroxamic acid (AHA) and how this reduction to Tc(II) affects the UREX process. Acetohydroxamic acid was also studied by /Tkac and Paulenova 2008/. They found that AHA has a considerably ability to form complexes with Pu even under strong acidic conditions. It was also proven that AHA forms extractable solvates with Pu(IV). /Paulenova et al. 2008/ modeled the distribution of Pu(IV) in the UREX extraction system with and without presence of AHA. Complexation of Ln and An by AHA was studied also by /Taylor et al. 2008/.

/Babain et al. 2008/ have investigated radionuclide partitioning in a modified UNEX process in cooperation with Russian scientists. The organic solvent was modified by using diamides of dipicolinic acid instead of CMPO. This measure increased the extraction capacity toward Ln and An metals. Selective stripping of An + Ln, Sr and Cs from the UNEX solvent have also been studied.

Some authors, e.g. /Laidler 2008/ and /Arm et al. 2008/ also discuss the GNEP (Global Nuclear Energy Partnership) initiative and its applications.

/Mincher et al. 2008a/ have investigated the radiation of the CCD-PEG (cobalt dicarbollide and polyethylene glycol) solvent intended for separation of Cs and Sr from dissolved nuclear fuel. ^{60}Co doses up to 432 kGy were used. The extraction of Cs was not affected while Sr extraction efficiency decreased with increasing dose. Stripping performance was not affected. /Mincher et al. 2008b/ also studied the radiation chemistry of 1-(2,2,3,3-tetrafluoropropoxy)-3-(4-*sec*-butylphenoxy)-2-propanol (Cs-7SB) that is used as a modifier in Cs and Sr extraction. Little decrease in extraction efficiency was found. It was concluded that the modifier acts as a radical scavenger that protects ligands from radiolytic attack.

Simultaneous separation of Cs and Sr from spent fuel by the FPEX process was investigated by /Law et al. 2008/. They used the extractants DtBuCH18C6 and BOBCalixC6. The process was successfully tested with a simulated waste solution.

/Ensor and Zimmermann 2008/ used the extractant bis(chlorophenyl)dithiophosphinic acid in combination with the synergist 4,7-diphenyl-1,10-phenantroline to extract Am(III) and Eu(III) from nitrate solutions. The results suggest that the system may be of interest for separation of Am(III) from Ln(III).

/Martin et al. 2008/ have studied the oxidation of Am to Am(VI) by Bi(V). The idea is to find a method for separation of Am from Cm by extraction of Am(VI) into 30% TBP in dodecane. It was found that 97% of the Am(III) was oxidized within 5 min but a much longer time was needed to obtain more than 50% Am(VI) in the solution.

/Meikrantz et al. 2008/ and /Garn et al. 2008/ have evaluated and tested 12.5 cm annular centrifugal contactors in TRUEX mass transfer studies. The obtained mass transfer efficiencies were in the vicinity of 100%.

2.7 Development of transmutation in Japan

The research on P&T in Japan is coordinated under the auspices of the Japanese Atomic Energy Agency (JAEA). The main strategy in Japan is the so-called double strata concept. In short, uranium and plutonium are re-cycled in LWRs, whereas minor actinides (and possibly long-lived fission products) are incinerated in ADS /Minato et al. 2009/.

The concept of the double-strata fuel cycle is shown in Figure 2-11. In this concept, the first stratum is based on commercial LWRs, in which recycling of uranium and plutonium is carried out, similar to existing MOX recycling. The second stratum is dedicated to incineration of MA by ADS (see Figure 2-12), and possibly long-lived fission products in general, and ^{129}I in particular. The mass flow of the actinides in the second stratum would be about a factor 100 smaller than the first stratum.

In the double-strata fuel cycle, MA-bearing fuel fabrication would not be easy because of the heat generation and the gamma, alpha and neutron emissions. To some extent this would be compensated for in the first stratum, because the partitioning of MA makes the recycled material used for fuel manufacturing in the first stratum less difficult to handle.

The Japanese program was reviewed in 2000 /Aoki 2002/, resulting in a recommendation to pursue studies of the double-strata fuel cycle concept in parallel with R&D on commercial FBRs. Recently (March 2009) another review has been undertaken. At present, this report is available only in Japanese /JAEA P&T review 2009/, but a summary is given below.

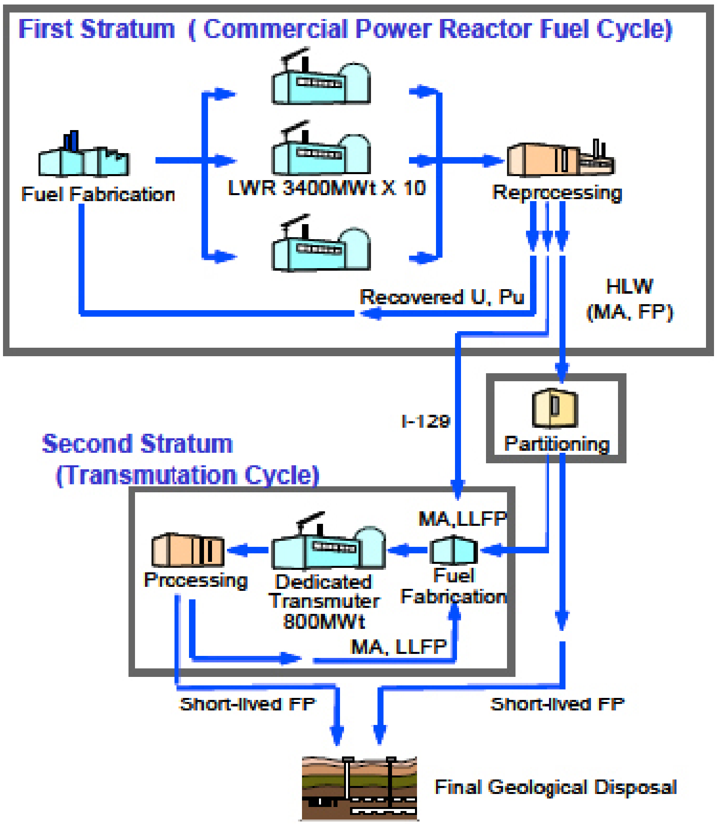

Figure 2-11. *The Japanese two-strata fuel cycle concept /Minato et al. 2009/.*

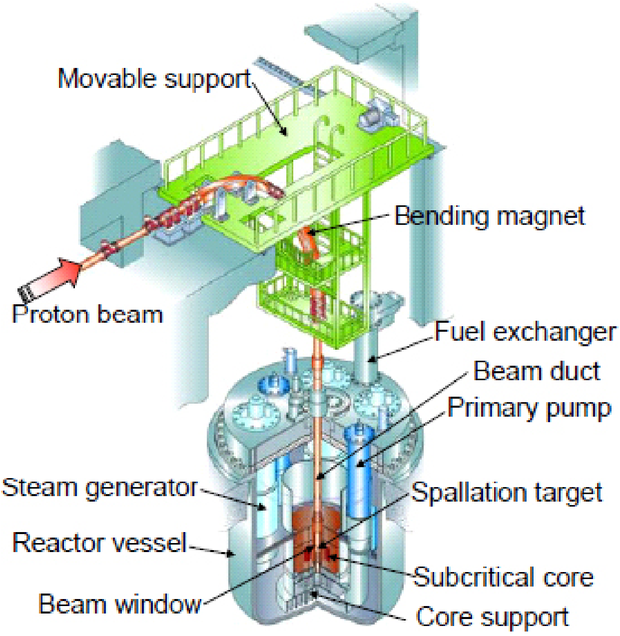

Figure 2-12. *The proposed ADS for MA incineration in the Japanese two-strata concept /Minato et al. 2009/.*

The report identifies three major areas of significance of the P&T technology: reduction of the potential hazard, mitigation of the requirement for geological repository site, and enhancement of the options in the design of the whole system of waste disposal. The report evaluated the current technology level of the P&T for both FBR and ADS. Although the technology level of some parts of the FBR cycle system are between basic research and engineering demonstration, the P&T technology in general is still in the basic research because of lack of experimental data for MA. It was, therefore, strongly recommended to accumulate the experimental data for MA as a common basis for both FBR and ADS.

The J-PARC accelerator facility under construction and commissioning is a cornerstone in the Japanese activities on P&T. A Transmutation Experimental Facility (TEF) in J-PARC is planned, although government approval has still not been given. TEF is planned to utilize a pulsed proton beam of initially 400 MeV, later to be upgraded to 600 MeV. TEF will consist of two satellite facilities: the Transmutation Physics Experimental Facility (TEF-P) and the ADS Target Test Facility (TEF-T). TEF-P is a critical facility which can receive a 10 W proton beam, where critical and subcritical experiments will be implemented for both ADS and FBR using MA-bearing fuels. TEF-T is a material irradiation facility using a 200 kW proton beam and an LBE spallation target. The accelerator complex J-PARC is in test operation at 300 kW target power. There are plans to increase to 600 kW. An recent overview is given by /Fujii and Arai 2009/.

There are, however, many other projects related to partitioning and transmutation in Japan, for instance on LBE technology and spallation target materials. Material development to mitigate the corrosion by LBE is underway, as well as post-irradiation examination (PIE) of spallation target materials.

/Tanaka et al. 2009/ report on an evaluation of the minor actinide recycling concept with Am-MOX fuel and development of fabrication of such fuels while /Kato et al. 2009/ report on an advanced process for fabrication of MOX fuel pellets. /Koyama et al. 2009/ have investigated plutonium production by transmutation of minor actinides. /Kinjo et al. 2009/ have performed a feasibility study on an upgraded Monju reactor core concept.

Many authors investigate advanced reprocessing as well as separation of trivalent actinides from trivalent lanthanides. An overview of wet reprocessing, in Japan and other countries, is given by /Tachimori and Morita 2009/. /Oigawa et al. 2007/ made an overview of P&T technology in Japan while /Nishihara et al. 2008/ studied the impact of P&T on LWR high-level waste disposal. /Morita et al. 2009/ reported on the development of an advanced reprocessing system based on precipitation. A computer code, PARC, for simulation of solvent extraction processes used for reprocessing was published by /Tsubata et al. 2009/.

/Matsumura et al. 2009/ investigated N,N,N´,N´-tetrakis(5-alkoxypyridin-2-yl)methylethylenedimaine (TRPEN) as a ligand for separation of trivalent minor actinides from lanthanides. The ligands were found to be efficient. /Ban et al. 2009/ have studied the radiolysis and extraction of certain dialkylamides. /Tachimori et al. 2008/ report on development of new CHON reagents for proliferation safe advanced reprocessing. Solvent degradation in reprocessing of MOX fuel and its effect on the plutonium purification cycle has been studied by /Kawaguchi et al. 2009/. /Ozawa et al. 2008/ report on a new back-end cycle strategy within the framework of the advanced ORIENT cycle, i.e. a fuel cycle that includes recovery of rare metal fission products like Ru, Rh and Pd. /Wei et al. 2009/ studied separation of Am and Cm by extraction chromatography using novel BTP extraction resins. /Shibata et al. 2009a, b/ report on a process involving uranium recrystallization.

Pyrochemical methods are also part of the Japanese P&T research program. Thus, /Moriyama et al. 2009/ have studied the influence of oxide ions on separation factors of actinides from lanthanides. /Hayashi et al. 2009a/ studied americium behaviour in a pyrochemical process for partitioning of spent nitride fuels. /Sato et al. 2009/ studied nitridation of U and Pu in a molten salt process. /Hayashi et al. 2009b/ have investigated electrolysis of AmN in an eutectic melt as well as renitridation of Am. The progress of a JAEA-CRIEPI study for metal pyroreprocessing has been reported by /Kitawaki et al. 2009/.

In nuclear transmutation some nuclides must be isotopically pure to avoid build-up of other, long-lived nuclides. To address this problem /Chaitikyan et al. 2009/ have reported on laser-chemical isotope separation of ^{135}Cs.

2.8 Development of transmutation in other countries

2.8.1 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[8] 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[9] 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 (U,Pu)N fuel intended for BREST was tested for the first time.

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/.

Russia seems to continue the successful irradiations of advanced nuclear fuels in the BOR-60 reactor. /Kosyakov et al. 2008/ suggest a method that uses the kinetic energy of fission fragments to obtain a separation between actinides and fission products in the fuel. /Gulevich et al. 2008/ performed a comparative study of ADS for minor actinides transmutation. The study included a lead-bismuth cooled ADS, a molten salt ADS and a heavy water ADS. The study still goes on and the first results indicate some advantages of the molten salt ADS.

Fission nuclear data is traditionally a strong Russian discipline, and a significant factor of all experiments on neutron-induced fission for FR as well as ADS applications are lead by Russian scientists, most often in ISTC projects.

2.8.2 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, HYbrid Power Extraction Reactor).

A scenario analysis for transmutation in fast reactors (Kalimer-600) compared to accelerator-driven systems has been performed by /Jeong and Ko 2008/. The result is that ADS as well as fast reactors can efficiently reduce the inventories of spent fuel and transuranium elements. /Jeong et al. 2008/ have also performed a dynamic analysis of a thorium fuel cycle in CANDU reactors. The results show that the amounts of spent fuel and TRU can be reduced with 15–38%. Based on the results of the comparison study, KAERI has switched focus from ADS to the critical fast reactor as the main transmutation system /Kim 2009/.

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

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

KALIMER-600 is a pool-type sodium-cooled reactor with a fast spectrum neutron reactor core. The core is loaded with U-TRU-Zr metal fuels. The reactor design life time is set to 60 years, which is evaluated to be long enough to provide the role imposed upon it without any degradation of the plant's efficiency. The net electricity output is 600 MWe and the net plant efficiency amounts to 39.4%.

KAERI is also performing a feasibility study on a 600-MWe sodium-cooled fast spectrum reactor core that can transmute TRUs produced by two LWRs of the same power and cycle length. The active core height is 90 cm at a cold state. A decrease in the conversion ratio to increase the transmutation rate is achieved by reducing the fuel rod outer diameter and the core height, and by using non-fuel assemblies such as void ducts. Unlike the KALIMER-600 break-even core, this transmutation reactor core does not have B_4C rods and dummy rods, while six ZrH_2 rods per assembly, which makes the core spectrum softer, are used to minimise the degradation of the fuel Doppler coefficient resulting from a reduction of the ^{238}U content.

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

3.1 Royal Institute of Technology (KTH) – reactor physics

KTH has performed research on transmutation of nuclear waste since the early 90's, with funding from SKB and EU. Initially, the work was focusing on design and safety analysis of accelerator driven systems /Wallenius et al. 2001, Eriksson and Cahalan 2002, Seltborg et al. 2003, Eriksson et al. 2005, Wallenius and Eriksson 2005, Fokau et al. 2010/. Subsequently, thermal-hydraulics of lead-bismuth coolant, advanced nitride fuel development, radiation damage physic in steels and Generation-IV reactor design and safety analysis have been added to the palette of competences /Olsson et al. 2003, Jolkkonen et al. 2004, Olsson et al. 2005, Ma et al. 2006, 2007, Wallenius 2009, Zhang et al. 2010/. In what follows, the major accomplishments during 2007–2009 are shortly outlined.

3.1.1 Design and safety analysis of a source efficient ADS with nitride fuel

As described above, the EUROTRANS project has resulted in the design of an industrial scale accelerator-driven system with excellent transmutation performance in terms of minor actinide burning. The application of oxide fuels, however has required a relatively large core size for the EFIT design. As the corresponding heat production in the spallation target demands a large target size for proper heat removal, the source efficiency of EFIT is relatively low.

The application of solid solution inert matrix nitride fuels, on the other hand, would allow significant reduction of the core size. As a consequence, the spallation target size may also be reduced, leading to improved source efficiency.

Fokau and co-workers have designed a nitride fuel ADS with lead-bismuth coolant and a core power of 200 MWth. The fuel pin design is based on that of EFIT-lead, but by adopting an austenitic steel cladding material the pin pitch could be reduced, making the core more compact, which results in an additional increase in source efficiency. Eventually a source efficiency 80% higher than the reference design of EFIT-lead could be achieved /Fokau et al. 2010/.

3.1.2 Source term estimations for XT-ADS and EFIT-Pb

A major advantage of lead coolant is that it will retain volatile fission products to a larger extent than sodium in case of a severe (core disruptive) accident. The source term (the inventory of radioactivity) available for release through a failing containment has been estimated by Jolkkonen for EFIT-lead and XT-ADS /Jolkkonen 2009/. Thanks to chemical bonding in the form of lead iodides and lead polonides, the release of these potentially problematic radioactive substances into the cover gas is significantly reduced. E.g. in EFIT-lead, the volatilised fraction of ^{131}I at 700°C is estimated to be less than 4 ppm, and that of ^{210}Po less than 0.03 ppm. The corresponding values in XT-ADS are estimated at 6 and 0.04 ppm for iodine and polonium. In the case of EFIT-Pb, the retention of iodine is good enough that a complete failure of the containment would not lead to significant dose to the public. In the case of XT-ADS, the much larger Po production from ^{209}Bi in the lead-bismuth coolant could lead to doses of the order of a few hundred mSv, whence the containment function would be essential for its safety performance.

3.1.3 Investigation of upper limits to americium concentration in sodium-cooled fast reactors

The ability of Generation-IV reactors to transmute legacy americium from the light water reactor park is limited by the adverse effects of americium on safety coefficients. This problem is particularly evident in the case of sodium-cooled reactors, where the positive temperature coefficient of the coolant is large even without americium in the fuel. In the literature, one may find that estimates for the permissible fraction of minor actinides in the fuel vary significantly (e.g. between 2 and 10%).

Only little transient analysis have however been made to support those claims. A detailed analysis of ULOF and UTOP transients in a model of a medium sized (1,500 MWth) SFR has been made by Zhang and coworkers, using the SAS4A code and reactivity coefficients calculated with SERPENT /Zhang et al. 2010/. It was shown that if the core power is kept at its original level, less than 3% of americium may be added to the fuel before a UTOP would lead to failure. Lowering the core power by reducing linear rating will however allow increase of the fraction of americium in the fuel, at the cost of electricity production.

3.1.4 Design of a nitride fuel version of the ELSY core

In the ELSY project (desribed elsewhere in this report), the reference core is based on standard MOX fuel. The application of nitride fuel would permit to reduce the core size and hence improve safety performance of the reactor. Wallenius has performed a neutronic assessment using the Monte Carlo code Serpent and arrived at a conclusion that the core height may be reduced by 22% using nitride fuel /Wallenius 2010/. This will allow increase of the fraction of americium in the fuel, as compared to the reference oxide core. Consequently, the nitride version of ELSY would have a higher potential for burning of legacy americium from LWR spent fuel.

3.1.5 The YALINA-booster experiments: results and interpretation

KTH has been a leading partner in the YALINA experiment describe above. Berglöf and co-workers have developed a novel approach to correct detector readings for spatial variation in neutron spectrum, which was shown to lead to a consistent method for determination of reactivity in sub-critical systems with strong heterogeneities /Berglöf 2010/. This method may be of particular use in e.g. XT-ADS, where the introduction of americium bearing fuel assemblies in a MOX core will lead to large variation in the local neutron spectrum.

3.1.6 Fabrication and characterisation of UN and (U,Zr)N fuel pellets

The success of the CONFIRM project has been an incentive for starting research on nitride fuel fabrication at KTH. High purity atmosphere glove boxes have been installed at a laboratory located in the material science department, and an existing furnace has been used to fabricate uranium and zirconium nitride powders from metallic source materials. The first UN and ZrN pellets were pressed in early fall 2009 (see Figure 3-1) and sintering tests will be performed in a dedicated boron nitride furnace under installation in January 2010. The aim of this activity is to optimise methods for fabrication and investigate properties of (U,Zr)N, including high temperature stability.

Figure 3-1. *UN (left) and ZrN (right) pellets manufactured in the fuel fabrication laboratory of KTH.*

3.1.7 Modelling of helium migration in CERMET fuels

One of the major candidate fuels for minor actinide burning in accelerator driven systems is the composite cermet fuel (Pu,Am,Cm)O_2–Mo, where the metallic molybdenum phase provides an effective thermal conductivity of the fuel 30 times largers than that of the oxide phase. This fuel type has recently been irradiated in Phenix (the FUTURIX experiment) and HFR (the HELIOS experiment). One possible concern is that the large amount of helium produced in the transmutation of americium would lead to excessive swelling. Therefore modelling of helium migration in the metallic molybdenum matrix is of high relevance. Runevall and Sandberg have calculated the probability for emission of He atoms from helium clusters that are expected to form in Mo vacancies. Using a combination of density functional theory (DFT) methods and rate theory, it was possible to produce a calculated spectrum of desorption of He as function of temperature. The calculation could be compared to experimental data, and was found to be in good agreement expect at the very highest temperature /Runevall and Sandberg 2009/.

3.1.8 Ab initio modelling of the Fe-Cr-C system

The useful properties of steels are due to a complicated microstructure containing iron and chromium carbides. Only some basic physical properties of these carbides are known with high precision, although the carbides may have a vital impact on the performance and longevity of the steel. To improve on this situation, /Henriksson et al. 2008/ performed extensive density-functional theory calculations of several carbides. The quantitative results were in perfect agreement with the relative empirical stability of the carbides. Also, in agreement with experimental data, it was found that $Cr_{23}C_6$ is not the most stable chromium-dominated carbide /Henriksson et al. 2008/.

In addition, to understand the behaviour of dissolved carbon in iron and chromium, first-principles density-functional theory calculations for carbon solution enthalpies, and diffusion activation enthalpies, in body-centered-cubic Fe and Cr were performed. The results for C in Fe compare well with experiments, provided that the effect of magnetic disordering is accounted for. Likewise, in Cr, the calculated solution and diffusion activation enthalpies agree well with available experiments. Further, first-principles calculations for the interaction energies between a solute (e.g. a Cr atom in bcc Fe) and an interstitial C atom were obtained. The results are in conflict with those inferred from internal friction (IF) experiments in disordered Fe-Cr-C alloys. A simple model of C relaxation in disordered Fe-Cr is used to compare theoretical and experimental IF curves directly, suggesting that a more extensive study of the energetic, thermodynamic, and kinetic aspects of C migration in Fe-Cr is needed /Sandberg et al. 2008/.

3.1.9 Natural circulation experiments in the TALL loop

The large absolute expansion of liquid lead and lead-bismuth with temperature, results in significant buoyancy forces, allowing for significant heat removal by natural circulation. Example given is EFIT-Pb designed to survive a loss of forced circulation for at least half an hour, relying on natural convection only. In certain flow regimes, natural circulation of heavy liquid metals can be unstable, however. Therefore, it is of importance to validate models used for design of the primary system. The 6.8 meter TALL loop at KTH mimics the primary circuit of a typical ADS design and is a unique facility in Europe for making such experiments. Ma and co-workers have studied how natural circulation is established after loss of forced circulation, as well as under start-up conditions. They were able to show that full natural convection is established within 8 minutes, while 50% of the asymptotic flow rate is achieved already within one minute /Ma et al. 2007/. No instabilities could be observed in experiments where forced circulation was turned off. A natural convection flow rate of 0.5 m/s was achieved for a temperature difference of 210 K between hot and cold legs, with the secondary cooling system working at full capacity.

3.2 Uppsala university – neutron research

The nuclear reactions group of the Department of Physics and Astronomy, 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. In addition to the work at TSL, the group is involved in experiments also at other facilities abroad.

A large effort during the recent three-year period has been the coordination of the CANDIDE project, described in section 2.3.9, and the coordination of networking activities in the EFNUDAT project, described in section 2.3.10.

3.2.1 Activities at TSL

A large data set on neutron-induced reactions at 96 MeV has been obtained at TSL in recent years. Using the SCANDAL setup /Klug et al. 2002/, elastic neutron scattering off a number of targets has been measured. Studied targets are H, D, C, Si, Y, Fe, Pb /Öhrn 2008/. The most recent development concerns a method to extract data on the (n,xn') reaction. A publication is in progress /Öhrn et al. 2009/. The production of light ions (p, d, t, ^{3}He and alpha particles) was measured with the Medley setup /Dangtip et al. 2000/. Such data are important for radioprotection (tritium), and provide information on the gas production in accelerator-driven systems. Studied target nuclei include Fe, Pb and U. With the latest publication /Tippawan et al. 2009/, the experimental research program at 96 MeV is finished.

In 2004 a new neutron beamline was put in operation at TSL /Pomp et al. 2005/. The design of this beamline allows measurements at 175 MeV, almost twice as high as previously, within a reasonable amount of beamtime. After a period with commissioning runs and installation of improved and additional shielding (see Figure 3-2) a series of experimental campaigns have taken place. So far Medley has collected data on light-ion production from C, O, Si, Fe, Bi and U. Analysis of the data sets is performed at Uppsala University, LPC Caen (France), Kyushu University (Japan), and Chiang Mai University (Thailand). Preliminary results for Fe and Bi show reasonable agreement between measurements and model calculations with the Talys code /Koning et al. 2008/ for protons. The production of deuterons and tritium is, however, overpredicted by Talys.

With financial support from the Swedish Research Council, the SCANDAL setup was equipped with larger CsI crystals to allow measurements at the higher energy. Test runs with the new CsI array have been performed successfully. Thus far elastic neutron scattering data on Si, Fe and Bi have been collected. The analysis of these data is in progress at UU.

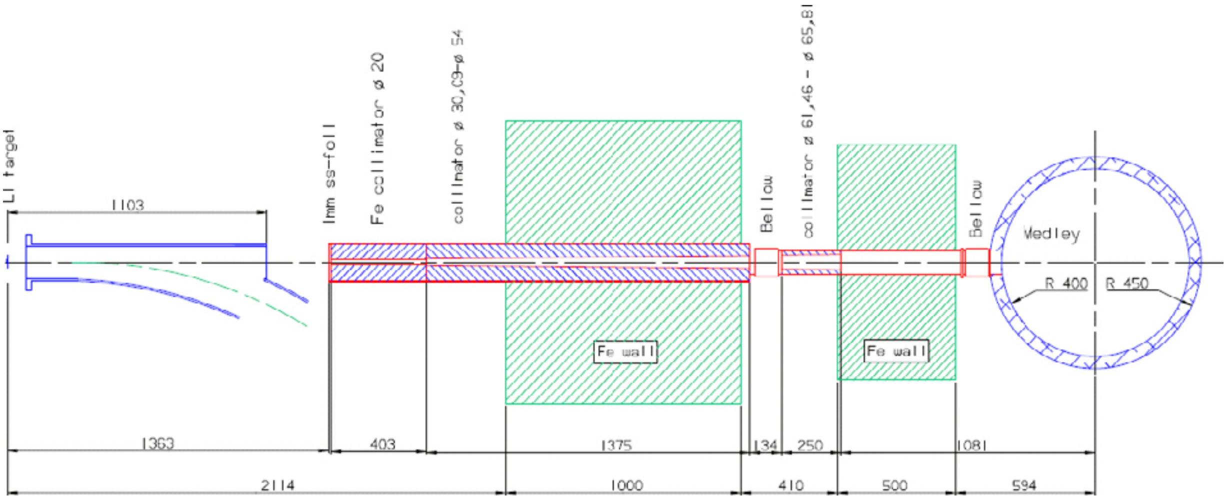


Figure 3-2. *Layout of the neutron beam at TSL (all dimensions in mm). The Li target is at the left and the Medley chamber on the right side. The SCANDAL setup is located further downstream after Medley and outside this figure. Next to Medley the recently installed additional iron wall can be seen. The beam pipe (red) is filled with an arrangement of collimator pieces (blue, hatched). The total distance between the centre of Medley and the Li target amounts to 4,618 mm.*

This work is supported from Swedish industry and authority through the KAT (1998–2002), NATT (2002–2006) and NEXT (2006–2010) projects. Most of the beam time at TSL for the Medley and SCANDAL runs were possible due to the financial support from the European Commission via the EFNUDAT project.

3.2.2 UU nuclear data activities elsewhere

Since 2007 the UU group is also actively involved in experimental activities conducted at other laboratories in Europe. Yields for neutron induced fission of ^{232}Th and ^{238}U at energies between 10 and 70 MeV have been measured at the quasi-monoenergetic beam at Louvain-la-Neuve, Belgium /Simutkin 2009/. Complementary measurements with the same experimental setup and for the target nuclei are currently underway at the white neutron source GNEIS in Gatchina, Russia. Data for neutron energies from threshold (about 1 MeV) up to 60 MeV will be analyzed. These activities are performed in close collaboration with the Khlopin Radium Institute, St. Petersburg, Russia.

Measurements of fission fragment properties of ^{234}U at neutron energies between 0.6 and 6 MeV are being performed at the Joint Research Centre of the European Commission in Geel, Belgium. The study of the ^{234}U(n,f) reaction is relevant even for present-day reactor applications since it involves the same compound nucleus as second chance fission of ^{235}U. This work has been taken up in the beginning of 2009 and first results have already been presented in workshops.

3.3 Chalmers University – nuclear chemistry

Chalmers University of Technology is involved in research regarding the separation of actinides and lanthanides as well as separation between the actinides themselves, as a partner in several European framework programs. Research on nuclear fuels and reprocessing began at Chalmers in 1946. The programs range from NEWPART in the 4th framework via PARTNEW and EUROPART to ACSEPT in the present 7th program. The aims of the projects have shifted during the last years from basic understanding to more applied research with focus once again on process development. This is clearly demonstrated by the fact that in the beginning it was not clear how these nitrogen donor heterocycles worked for separation of actinides from lanthanides. The first molecules used in this context were the commercially available terpyridine molecules that were extensively studied by e.g. /Spjuth 1999, Hagström et al. 1999/ and /Andersson et al. 2002/. Detailed studies relating to synergistic extraction using terpyridine and organic counter ions were performed by /Nilsson et al. 2006a/. As more and more results were gathered the molecule focus changed to the BTP family, which was studied by the Chalmers group and to a large extent also internationally. The last family studied and which currently is the most promising one is the BTBP family where much work has been performed in the Chalmers group. In many cases these basic studies are reported by /Ekberg et al. 2008/.

During the last three years the main research focus on Chalmers has been on the SANEX (Selective ActiNide EXtraction) process. This process aims at extracting trivalent actinides (An) into an organic phase with the use of a nitrogen donor ligand as extractant. The lanthanides (Ln) are left in the aqueous phase and a good An/Ln separation is achieved. Problems related to this concept are the major difficulty of separating the trivalent actinides from the chemically similar trivalent lanthanides, the kinetics of the system, the large dose given to the different organic phases etc. Chalmers' work within the development of the SANEX process has among other things been to investigate the properties of several different new extraction molecules, study the diluent effects and investigate the radiation behavior of various extracting systems. Due to new recruitments we have also started work on ligand design and development. This will decrease the response time between synthesis of new extraction ligands and their evaluation. Today the SANEX process is considered to be working on a basic scale after the successful implementation of a hot test /Magnusson et al. 2009b/ and the research has shifted towards a further developed process called i-SANEX. In an i-SANEX process actinides and lanthanides are co-extracted into an organic phase followed by selective back extraction of the actinides with a hydrophilic complexing agent. For this purpose, the development of viable hydrophilic complexing agents has also been initiated.

Another concept that has been introduced is the so-called GANEX (Group ActiNide EXtraction) process, and Chalmers has in the recent years started research within this field. The idea behind the GANEX process is to extract all the actinides as a group directly from dissolved spent fuel and then selectively strip the actinides for transmutation purposes. Our work regarding this process has been focused on the development and extensive tests of a new organic extraction system.

Aliphatic hydrocarbons are often used as diluents in the type of solvent extraction processes described above. Chalmers has chosen to mainly focus research on experiments with cyclohexanone as diluent. The solubility of BTBPs in cyclohexanone is higher than in aliphatic hydrocarbons, and the kinetics of the systems is generally faster.

Throughout this chapter the concepts of distribution ratio (D) and separation factor (SF) are frequently used in the context of extraction and they are defined as follows:

$D = [A]_{org} / [A]_{aq}$

D is thus the ratio of the total concentration of a substance, A, in the organic phase to its concentration in the aqueous phase.

$SF_{A/B} = D_A / D_B$

$SF_{A/B}$ is the ratio between the distribution ratios of two different metals (A and B) in the same system.

3.3.1 BTBP-chemistry

The use of nitrogen-containing heterocyclic molecules has been shown to efficiently separate actinides and lanthanides using solvent extraction /Ekberg et al. 2008/. The basic structure has changed over the years and currently the BTBP family is the most promising one. BTBP stands for *bis*-triazine-*bi*-pyridine, which refers to the nature of the central core molecule common to all of the molecules in the family. The different BTBPs mentioned in this report are illustrated in Table 3-1.

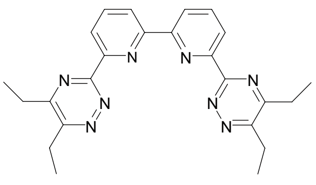
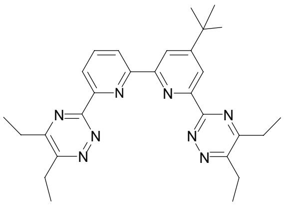
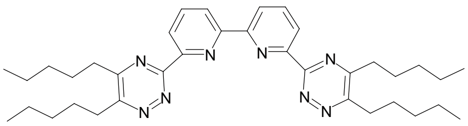
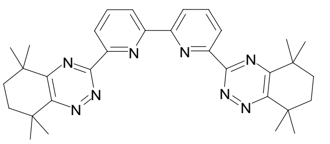
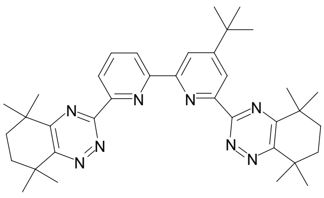
Recently, attempts have been made to synthesize a new type of BTBP with five-membered side rings instead of six-membered, which has previously been successful. However, problems were encountered when trying to form the C_3N_3 ring in the core molecule, probably due to ring strain in the side group, and the synthesis could not be completed.

Below follow descriptions and results of a number of studies done on different aspects of the BTBP chemistry.

Screening studies of a new molecule

When a new ligand has been developed and synthesized it is generally put through a number of different tests, a so-called screening of the molecule. The screening aims at establishing whether or not a new molecule is suitable as extractant and if it is worth to do more details studies on it. A new ligand, generically named MF2-BTBP, was synthesized at Reading University, UK. Different long chain alcohols were selected for the screening extraction experiments to serve as a comparison base with previous studies of $CyMe_4$-BTBP /Dubois et al. 2007, Retegan et al. 2007/, since MF2-BTBP has similar features with this ligand. A study of extraction kinetics in nitrobenzene was also performed. The extraction of Am and Eu using MF2-BTBP in long chained alcohols showed values similar to those obtained in the previous studies involving $CyMe_4$-BTBP /Retegan et al. 2007/ but this time a clear trend was found: the log D decreased with increasing the number of carbons in the chain. The molecule dissolved reasonably quick (5 minutes) in all the alcohols. This is not the case for the system containing the $CyMe_4$-BTBP molecule. Regarding the extraction kinetics; a system containing MF2-BTBP in nitrobenzene did not reach equilibrium even after 20 minutes; by comparison, the corresponding system containing $CyMe_4$-BTBP reached equilibrium after 15 minutes. To conclude; MF2-BTBP showed promising distribution ratios and separation factors but the extraction kinetics was too slow.

Table 3-1. Structures of the ligands used in the different studies /Foreman et al. 2006/.

Abbreviation	Chemical name	Structure
C2-BTBP	6,6'-bis(5,6-diethyl-[1,2,4]triazin-3-yl)-[2,2']bipyridine	
MF1-BTBP	4-tert-butyl-6,6'-bis(5,6-diethyl-[1,2,4]triazin-3-yl)-[2,2']bipyridine	
C5-BTBP	6,6'-bis(5,6-dipentyl-[1,2,4]triazin-3-yl)-[2,2']bipyridine	
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	
MF2-BTBP	4-tert-butyl-6,6'-bis-(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo[1,2,4]triazin-3-yl)-[2,2']bipyridine	

Thermodynamics of dissolution

An important aspect when using the BTBP molecules as extractants is the solubility of the molecule in the selected diluent. Different BTBPs have previously shown different solubility in different diluents and therefore dissolution experiments with C2-, CyMe$_4$-, C5-, MF1- and MF2-BTBP in cyclohexanone and octanol at different temperatures were performed to obtain entropy and enthalpy of dissolution (Table 3-2).

Table 3-2. Standard enthalpies and entropies of dissolution of several systems containing BTBP molecules dissolved in cyclohexanone or octanol). The uncertainties present one standard deviation.

System		ΔH^0 (kJ/mol)	ΔS^0 (J/mol)
CyMe$_4$-BTBP	cyclohexanone	37.2 ± 0.7	92 ± 2
	octanol	41.0 ± 0.8	99 ± 3
C5-BTBP	cyclohexanone	66.6 ± 0.5	207 ± 2
	octanol	51.4 ± 0.4	143 ± 1
C2-BTBP	cyclohexanone	22.3 ± 0.3	40 ± 1
MF1-BTBP	cyclohexanone	44.8 ± 0.5	123 ± 2
	octanol	37.7 ± 0.2	90 ± 1
MF2-BTBP	cyclohexanone	105.6 ± 2	346 ± 6

These can be estimated from the solubility by using Gibbs free energy change and the solubility constant. Entropy and enthalpy of dissolution were obtained for a number of systems and attempts to correlate the thermodynamic constants to extraction data were done /Spendlikova 2009/.

Mobility of BTBP-metal complexes

Since little is known of the nature of the metal complex present in the organic phase a series of electrical conductivity experiments were performed using solutions of metal complexes in nitrobenzene and cyclohexanone, both saturated with water. The aim of this study was to accept or reject the proposed hypothesis that "in nitrobenzene the metal species (and counter ions) formed by the extraction of europium nitrate by the BTBPs are separated ion pairs which are able to move freely, while in cyclohexanone these ions are present within contact 'pairs' which inhibits their free movement". As an initial test of the equipment and experimental protocol, the conductivity of tetraethylammonium perchlorate and a series of perchlorate salts of C2-BTBP complexes (copper(II), cadmium(II) and lead(II)) were determined. In addition the conductivity of the perchlorate salt of the $CyMe_4$-BTBP complex of lead was measured. Because mobility data for the ions present within tetraethylammonium perchlorate has been presented in the literature /Makrlik et al.1984/ and as this compound is readily available by the anion metathesis of tetraethyl ammonium chloride with perchloric acid, we have chosen to employ this salt as our standard within nitrobenzene. The three metal complexes were used to test the assumption that the mobility of a *homoleptic* BTBP complex is independent on the nature of the metal at the centre of the complex. As hoped the metal salts are strong electrolytes in nitrobenzene, and the ionic conductivities of the BTBP complexes are very similar. Using the Stokes equation and a literature value for the hydrodynamic radius of a perchlorate anion /Coetzee and Cunningham 1965/ it was possible to estimate the hydrodynamic radii of the metal complexes after making the assumption that these complexes exist as *homoleptic* BTBP complexes in nitrobenzene.

Diluent effects

It has been long known that changing the diluent highly affects the distribution ratios (D) and separation factors (SF) for metals with different extraction mechanisms /Mason et al. 1967, Peppard et al. 1971/. However, there have previously also been indications that even with the same extraction mechanism the SF may change when changing the diluent. A study /Retegan et al. 2008/ was made on $CyMe_4$-BTP and $CyMe_4$-BTBP, each of them dissolved in two different diluents: cyclohexanone and nitrobenzene. The four systems were compared in terms of D, SF and extraction kinetics (time to reach equilibrium). The kinetics proved to be faster in cyclohexanone than in nitrobenzene. Higher D for Am and Eu were observed in the nitrobenzene containing systems than in the cyclohexanone ones. This can probably be explained by the fact that an oxygen atom from the more polar nitrobenzene molecule can coordinate to the metal, thus increasing the lipophilicity of the complex and easing the extraction. As opposed to the D, SF is lower in nitrobenzene than in cyclohexanone for both ligands. Comparing the two ligands in cyclohexanone shows that the BTP extracts better than the BTBP, and it is primarily Am that shows higher D. BTP is thought to form 1:3 complex with actinides, while BTBP form 1:2 complex. Thus, in the case of BTP all nine coordination sites are occupied by BTP and nitrate and the complex is more lipophilic than the corresponding BTBP complex, which only occupies eight coordination sites.

ESI-MS measurements on Am and Eu complexes

In order to get a better understanding of the complexes formed between BTBPs and actinides and lanthanides, and to possibly extend the explanation of diluent effects, electrospray ionization mass spectrometry (ESI-MS) was used to characterize the complexes /Retegan et al. 2009/. Two extractants (C5- and $CyMe_4$-BTBP) in three different diluents (cyclohexanone, hexanol and nitrobenzene) were studied. The same type of complexes was observed in all systems. It was shown that $CyMe_4$ forms stronger complexes than C5, and also that the diluent affects the stability of the complex.

EXAFS studies of Am/Eu BTBP complexes in various diluents

Attempts were also made to study Am/Eu-BTBP complexes in octanol, cyclohexanone and nitrobenzene using EXAFS. The aim was to try to establish if the differences in separation factor (mentioned above) might be caused by differences in complex structure. No differences were observed. However, during measurements of the sample with the most different SF – (Am in nitrobenzene), problems occurred and no data could be obtained for this complex.

Recovery of BTBP from the organic phase

In a future industrial process it is of vital interest to recover and reuse the BTBP molecules. As an alternative to the evaporation of the diluent in the case of cyclohexanone a low temperature method operating at ambient pressure has been devised and tested. By treatment of a solution of a BTP or BTBP in cyclohexanone with an aqueous solution of sodium hydrogen sulphite ($NaHSO_3$) it is possible to convert the cyclohexanone into a crystalline solid that is less soluble in organic solvents without altering the extractant molecule /Young and Jencks 1978/. As the addition product of cyclohexanone and sodium hydrogen sulphite (the sodium salt of 1-hydroxy-cyclohexanesulfonic acid) is soluble in water, under suitable conditions it is possible to convert a cyclohexanone solution of a BTBP (or BTP) into an aqueous solution of the sodium salt of 1-hydroxy-cyclohexanesulfonic acid and solid BTBP. The BTBP can then be collected by filtration, washed with water and then dried before being reused.

3.3.2 Radiolysis

Since the separation process using the BTBP molecules takes place in a high radiation flux, the solvent, i.e. both the extractant and the diluent, must be resistant to radiation. A number of organic systems have been irradiated and then studied in terms of degradation products and the effect on extraction, and the studies are briefly presented below. Most solvents were irradiated using the ^{60}Co-source at Chalmers if nothing else is stated. If the ageing of the solvent was unknown, corresponding samples were placed in a flask outside the gamma source, at a remote distance, to act as a reference for investigation of the possible degradation over time of the molecule. Samples were taken from the solvents at the start of the experiments and then at different times during a longer time period.

Change in extraction behaviour during γ-irradiation of BTBP solvents

A number of systems have been studied at Chalmers in terms of how their metal extraction is affected by γ-radiation /Fermvik et al. 2007, 2008, 2009, 2009b, Fermvik 2008, Retegan et al. 2007b/. The most common behaviour is a decreasing extraction of both actinides and lanthanides as an organic system receives higher and higher dose /Nilsson et al. 2006b, Hudson et al. 2006, Magnusson et al. 2009, Hill et al. 2005/, but the opposite has also been observed /Retegan et al. 2007a, Fermvik et al. 2008/. Besides efficiently extracting trivalent actinides, many of the polydentate N-donor ligands also show high affinity for various transition metals, most of which are present in the HLLW solution resulting from the PUREX process. Therefore the effect on extraction behaviour towards palladium (Pd), silver (Ag), and cadmium (Cd) of an irradiated organic system containing 0.005 M C5-BTBP in cyclohexanone was also investigated /Fermvik et al. 2008/.

α- vs. γ-radiolysis

An extracting system will be exposed to different types of radiation, and experiments have been made to try to compare α- and γ-radiolysis of BTBP solvents. One study investigated the change in D_{Am} when solvents containing $CyMe_4$- or C5-BTBP in cyclohexanone were irradiated with low doses of alpha and gamma radiation, respectively. Both types of irradiation were given with a low dose rate and the total received dose was approximately 17 kGy. The γ-dose was given by the ^{60}Co source, and the α radiolysis was done by extracting ^{241}Am from an aqueous phase into the organic phase and then performing extraction tests at various times, i.e. after various doses. For $CyMe_4$-BTBP, there is no clear difference between the two types of irradiation, possibly the gamma radiation affects the system a bit more than alpha does. Both types of irradiation cause an increase in distribution ratio.

Unlike $CyMe_4$-BTBP, C5-BTBP is not stable over time /Fermvik et al. 2008/ and the distribution ratio of Am decreases as the solvent is ageing. This must be taken into account when evaluating data from radiolysis experiment, since the degradation of the solvent then is a combination of irradiation and ageing. The C5-BTBP solvent appears to be more affected by the alpha radiation than the gamma radiation. However, this is not the whole truth since C5-BTBP has been reported to be sensitive towards hydrolysis, and during the α irradiation the organic phase is in constant contact with the aqueous phase. In addition, degradation products from the radiolysis of water most likely affects the degradation of C5-BTBP and contributes to the change between α and γ irradiation in this experiment.

α-radiolysis experiments with ^{211}At

A method to perform α-radiolysis experiments with ^{211}At has been developed. ^{211}At is a short-lived isotope, hence enabling analysis of a "non active" solution after a certain period of time /Ekberg et al. 2010/. Experiments were done where astatine was dissolved straight into an organic phase containing 0.005 M C5-BTBP in cyclohexanone. A first set of extraction experiments were made to establish whether or not the astatine was bound to the BTBP ligand. The extraction of astatine showed to be independent of the absence or presence of BTBP ligands, hence the distribution ratio equals the distribution constant (K_D) and is a result only of the partitioning of astatine between the organic and aqueous phase. A second experiment was made with the aim to irradiate an organic solvent with α-radiation using ^{211}At and compare the resulting degradation products to those resulting from similar irradiation with γ radiation /Fermvik et al. 2009b/. In the analysis of the α irradiated solution the same degradation products were observed as when a similar solvent was exposed to approximately the same dose of γ radiation /Fermvik et al. 2009b/.

Radiolysis of a two-phase system

Radiolysis and degradation of an organic phase is very likely to be highly affected by the presence of an aqueous phase during irradiation. This will be the case in a future industrial process and it is thus important to not only study the irradiation of organic phases alone. A solvent containing 0.005 M C5-BTBP in cyclohexanone was irradiated in a ^{60}Co source at the Institute of Nuclear Chemistry and Technology in Warsaw, Poland, with a dose rate of 0.939 kGy/h. Irradiations were performed under three different conditions:

- Irradiation of organic phase alone.
- Irradiation of organic phase pre-equilibrated with aqueous phase.
- Irradiation of organic and aqueous phase in contact.

Extraction experiments showed that the extraction of Am decreases much more when any aqueous phase was present (condition 2 and 3). HPLC-analyses (performed by Institute of Inorganic Chemistry, Academy of Sciences of Czech Republic) showed that the concentration of C5-BTBP in irradiated samples was clearly decreasing with dose. And as could be expected from the extraction results, the decrease was much more pronounce when aqueous phase was present during irradiation. This is the opposite of what has been observed for other two-phase systems in ongoing work from other partners within the ACSEPT project. These differences in behaviour will be further investigated.

Effect of dose rate

It is not only the total dose given to an extracting system that is of interest, but also the dose rate, since this can vary within a wide range /Magnusson et al. 2009/. In order to investigate the dose rate dependence of radiolytic degradation, an organic system containing MF2-BTBP in cyclohexanone was irradiated with a total received dose of ~ 100 kGy but with a varying dose rate /Fermvik et al. 2009/. The low dose rate was ~ 15 Gy/h (at Chalmers) and the high dose rate was 1.2 kGy/h (Forschungszentrum Jülich, Germany). Since the total time of irradiation was very long for the low dose rate solution, a reference solution was used to take the possible ageing of the solvent into account. The system that was exposed to the low dose rate did not show a trend of either decreasing or increasing distribution ratios but instead the ratio between D for the irradiated solution and D for the reference solution was around 1 at all sampling points. This indicates that the solution is rather resistant towards radiolysis. The system that was exposed to a higher dose rate showed a slow decrease in distribution ratio; hence, the system seemed to be more affected by the higher dose rate.

The opposite behaviour was observed in another dose rate study /Fermvik et al. 2009b/, where the extractant was C5-BTBP and the diluent either hexanol or cyclohexanone. In both diluents, C5-BTBP was degraded to a larger extent with the low dose rate (12.5 Gy/h) than with the high (1,650 Gy/h). This may not be so surprising considering that a higher dose rate increases the possibility of recombination of radicals and thus decreases the ratio of further reactions with extractants. Another explanation for the results of this study is the fact that C5-BTBP is not stable with time. It degrades to a certain extent as it ages and since the lower dose rate means a much longer irradiation time, this also affects the results. The ageing was larger when cyclohexanone was used as diluent, but the radiolytic degradation was larger for the hexanol-based solvent.

Effect of storage temperature

In many cases the actual experimental procedure to make irradiations test varies between different labs. Some perform e.g. extraction experiments directly after irradiation, while other store their samples before conducting further experiments. To be able to make valid comparisons between different studies, and also within specific studies, it is important to be consistent regarding the time between irradiation and additional experiments, and also regarding the temperature during a possible storage time. To determine the effect of temperature during storage of samples between the irradiation and further experiments four organic solvents were irradiated and then stored for a long time before extraction experiments were carried out /Fermvik 2009/. Results were evaluated in terms of change in D_{Am}, D_{Eu} and $SF_{Am/Eu}$ after a certain time of storage. Samples were stored both in room temperature and in a freezer. Two of the solvents contained C5-BTBP dissolved in either hexanol or cyclohexanone and two solvents contained $CyMe_4$-BTBP in the same diluents. The behavior in the two different temperatures differs for different extractants as well as for different diluents. The results from these experiments highlight the importance of being consistent when conducting extraction experiments following irradiation and to be attentive when comparing different radiolysis studies.

Analysis of degradation products

It has often been proved that organic solvents somehow degrade as a result of irradiation and the effect on extraction has been studied a lot. But it has generally not been explained in detail how the solvents degrade, and what the degradation products look like. Analyses of irradiated solvents using HPLC-, APCI- and LC-MS have enabled the identification of a number of potential degradation products. These analyses were conducted at CEA, Marcoule in France and at the Institute of Inorganic Chemistry, Academy of Sciences of Czech Republic. Structures for degradation products resulting from radiolysis of solvents containing C5-BTBP in different solvents have been suggested /Fermvik et al. 2009b, 2010/, and a proposed degradation scheme for C5-BTBP is shown in Figure 3-3. The change in concentration of these degradation products as a function of the received dose has been studied /Fermvik et al. 2009b, 2010/, and the concentrations generally increase with increased dose. Products 1–4 in Figure 3-3 were the ones found in highest concentrations. Number 1 and 2 are formed first and then degrades further into products 3 and 4.

3.3.3 Water-soluble ligands for i-SANEX

As described in the introduction to this chapter, a development from the SANEX process is the so called innovative SANEX (i-SANEX). It aims at co-extracting An and Ln and then separate them by selective back-extraction of An with a hydrophilic complexing agent. A first attempt was done to develop two such hydrophilic ligands.

Design and synthesis of water-soluble ligands (PPA and PPPA) for i-SANEX

Two water-soluble ligands intended for the i-SANEX process were designed and synthesized; Pyridin-2-yl-phosphonic acid (PPA) and (6-Phosphono-pyridin-2-yl)-phosphonic acid (PPPA).

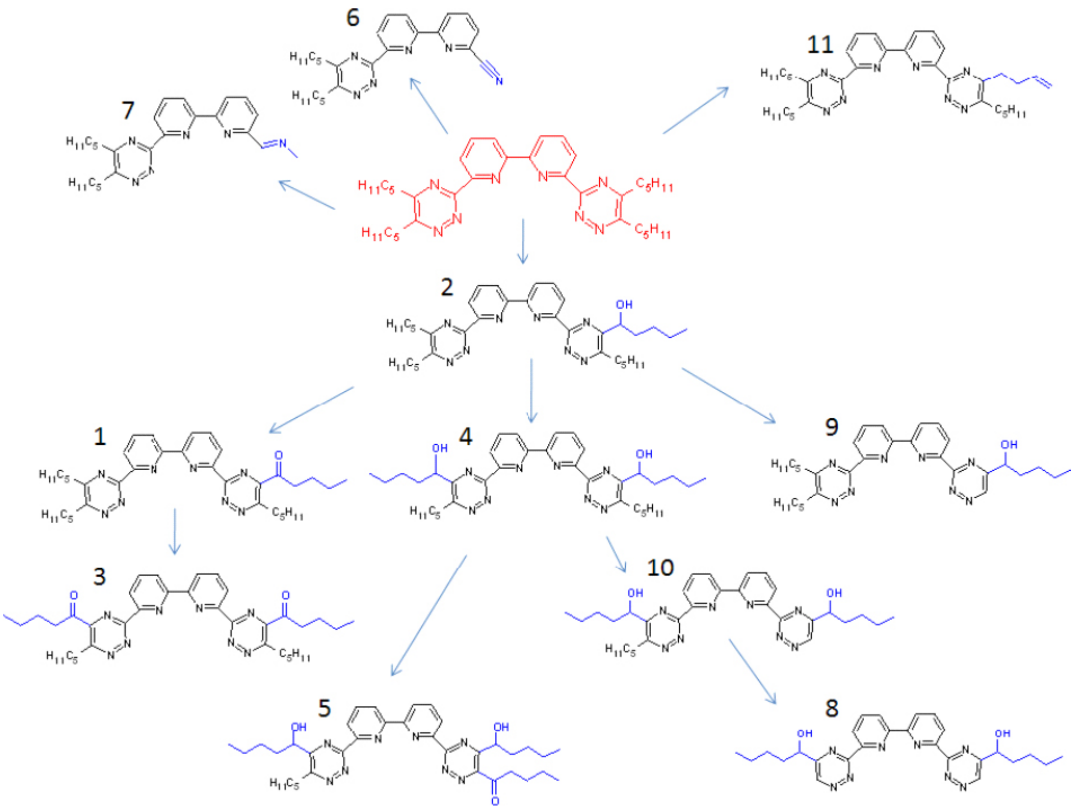


__Figure 3-3.__ Proposed degradation scheme for C5-BTBP (in red) during gamma irradiation /Fermvik et al. 2009b/.

Screening test

The water soluble ligands were tested according to the i-SANEX protocol established in the ACSEPT project. The D values for TODGA without any ligand added to the waterphase (D_{Am}=30 D_{Eu}=220) was only slightly affected by the PPA ligand (D_{Am}=20 D_{Eu}=110) which also gave rise to some 3rd phase formation. The addition of ligand PPPA however enhanced the separation factor from $SF_{Eu/Am}$=7.5 for just TODGA to $SF_{Eu/Am}$=16 but also heavily decreased the D-values for both Am and Eu (D_{Am}=0.002 D_{Eu}=0.03).

3.3.4 Development of a novel GANEX system

After the successful implementation of a hot test on both the DIAMEX and SANEX extraction system the separation procedure for spent nuclear fuel has been developed further. Now research is focused on a GANEX (Group ActiNide EXtraction) separation system. This system decreases the number of processes, which is an advantage for the final implementation. In this type of separation system the actinides should be extracted as a group from the lanthanides and the fission- and corrosion/activation products. The actinides can then be selectively stripped for transmutation purposes. The GANEX extraction is supposed to take place in a highly acidic environment after a separation of the uranium bulk from the dissolved spent fuel.

Formulation of the extraction system

A GANEX separation system for transmutation has been developed. In this separation system the actinides should be extracted as a group from the lanthanides and the fission- and corrosion/activation products. This has been achieved by combining BTBP with TBP in cyclohexanone.

The BTBP molecules have a well documented ability to separate trivalent and pentavalent actinides from trivalent lanthanides /Retegan et al. 2007b, Drew et al. 2005/. TBP on the other hand, has since the 50ies been known to extract uranium and plutonium /Burger 1958/. The role of the two molecules is to perform their respective extraction independently and to have as few interactions as possible between them. To see if the molecules were stable in a solution together, TLC (thin layer chromatography)-experiments were performed. There was also extraction tests performed to see if the molecules could extract independently of each other. Both these tests showed positive results.

Extraction tests

Extraction tests under process like conditions have been performed with the new GANEX solvent. The actinides are readily extracted ($\log(D_{Am})=2.19$, $\log(D_{Pu})=2.31$, $\log(D_U)=1.03$, $\log(D_{Np})=0.53$) and they are also separated from the lanthanides ($\log(D_{La})=-2.0$, $\log(D_{Ce})=-1.72$, $\log(D_{Nd})=-1.05$, $\log(D_{Sm})=-0.18$, $\log(D_{Eu})=-0.02$). Some fission products are also extracted by the new GANEX solvent. This is of course an unwanted scenario which is being dealt with in the next section. There is also some extraction of corrosion products that must be taken into consideration. A loading experiment showed that the D for the actinides extracted by the BTBP molecule is lowered but still sufficiently high.

Radiolytic stability

Samples of the GANEX solvent were irradiated in a ^{60}Co source with a dose rate of 0.94 kGy/h (performed at the Institute of Nuclear Chemistry and Technology in Warsaw, Poland), to evaluate the radiolytic stability of the new GANEX solvent. The solvent was irradiated with and without being in contact with the aqueous phase (4 M nitric acid) and after irradiation it was subsided to extraction tests and compared to a non irradiated sample which had been present in the same environment. The pure GANEX solvent seemed to be rather stable towards radiolysis but with a slight decrease in D for both Am and Eu. When the solvent was irradiated in the presence of the aqueous phase however, no decrease was observed; hence the aqueous phase seems to protect the organic phase to a certain extent. This behavior has also been observed for other organic systems /HYBAR N^{o}3 a, HYBAR N^{o}3 b/ but is the opposite to what was observed in the study of radiolysis in a two-phase system described above /Fermvik et al. 2010/.

Hydrolytic stability

The hydrolytic stability of the new GANEX solvent was tested by letting the organic phase stand in contact with an equal amount of aqueous phase (4 M nitric acid). After different lengths of time a portion of the organic phase was removed and subsided to extraction of Am and Eu from 4 M HNO_3. The solvent proved to be hydrolytically stable.

Flow sheet modeling

An excel flow sheet model for outgoing stream concentrations in a n stages counter current liquid-liquid extraction system has been made where D-values are the main input. The V_{org}/V_{aq} ratio as well as the amount of extraction stages (n) can be varied to optimize the metal concentrations in the outgoing streams.

3.3.5 Suppression/pre-extraction of fission products in the GANEX system

Among the fission products that were found to be extracted with the newly developed GANEX system, focus were put on three; Pd, Mo and Zr. Mo and Zr have a very high abundance in spent fuel /Serrano-Purroy et al. 2005, Malmbeck et al. 2000, Garcia Alonso et al. 1994/ and it is therefore interesting to inhibit their extraction. Pd is not equally abundant but besides being extracted by the system, ketones like cyclohexanone are able to reduce the palladium present in dissolved spent fuel to metallic palladium (palladium-black). Different methods to avoid the problem of fission product extraction in the GANEX system are presented below.

Extraction suppression and inhibition of formation of palladium black using methionine and bi-methionine

The mere presence of nitric acid is unable to inhibit the formation of palladium black in the presence of a ketone like cyclohexanone but when D,L-methionine (100 mM) is added to the aqueous phase, with cyclohexanone as the organic phase, no formation of metallic palladium is observed for several weeks. Without the methionine addition the formation occurs within a few hours. With the methionine addition to the aqueous phase the extraction of Pd with the GANEX solvent is also suppressed while no effect is seen on the extraction of e.g. Am and Eu. This suppression of extraction however only occurs when a low acidic water phase (1 M nitric acid) is used. When a stronger acid is used (4 M) the precipitation is stopped but the extraction of Pd is not suppressed. Due to this a new molecule, bi-methionine, was synthesized. This molecule (10 mM) effectively stopped the extraction of Pd even from a highly acidic water phase as well as it stopped the formation of Pd-black without affecting the extraction of e.g. Am and Eu. The effect of bi-methionine on the extraction of Zr and Mo- was also tested and with a large addition it seems as if the extraction of Mo is totally suppressed, this however must be further investigated. The D for Zr is somewhat lowered by adding bi-meth to the water phase thus reaching an acceptably low level.

Synthesis of bi-methionine

Using a procedure from the literature over 10 grams of 2-amino-4-[2-(3-amino-3-carboxypropylsulfanyl)-ethylsulfanyl]-butyric acid (bi-methionine) was prepared. In the original synthesis a mixture of hydrochloric acid, methionine and ethylene dibromide was heated before being reduced in volume and being neutralized by the addition of ammonia solution. The solid bi-methionine thus obtained required extensive washing with water before oven drying to remove all halide ions. (A 100 mM solution of the first batch of bi-methionine tested negative for halides with thallous nitrate.) In a new synthesis a partly successful attempt was made to isolate the hydrogen chloride salt of bi-methionine by recrystallization. The majority of the bi-methionine was isolated from the mother liquor after the addition of ammonia by concentration of the mixture at atmospheric pressure followed by cooling of the mixture. The polar nature of bi-methionine and allied substances complicates their analysis.

Pre-extraction with dioctyl sulfide

Another way to approach the problem with fission product extraction is to extract the troublesome elements before the actual GANEX process. This has been tested with dioctyl sulfide in kerosene. Pd (and also silver, another troublesome element) was readily extracted with the dioctyl sulfide as well as some of the Mo. The Zr was however not extracted, but the dioctyl sulfide still show good potential since it was found not to affect the actinides or lanthanides.

3.3.6 Synergic extraction of Eu

An earlier study /Sezgin 2003/ showed interesting results for the synergic extraction of europium by terpyridine (Terpy) and α-bromodecanoic acid (HA) in tributylbenzene (TBB). Therefore new sets of experiments were carried out to try to explain previous results and to extend the investigation /Aneheim et al. 2008/. The first step in the study aimed at determining whether or not the system showed the same behaviour with higher concentrations of Eu as with trace amounts, which was used in the old study. It was obvious that the maximum extraction of Eu did not occur at the same [HA]/[Terpy] ratio for the higher concentration of Eu as it did when using just trace amounts. If instead the extraction is plotted as a function of [HA], the maximum value of extraction coincides for the two different studies. Further experiments were carried out with other diluents, to see if the pattern was the same in diluents such as kerosene, toluene and cyclohexane. Kerosene proved to form a third phase when contacted with the aqueous phase so no extraction data could be obtained for that system. The system with toluene as diluent showed similar behaviour as the TBB system, while cyclohexane indicated a different behaviour and will require further studies.

3.3.7 Plutonium speciation under reducing conditions

After separation processes, before the production of new fuel, the actinides will be present in either mono- or polyactinide solutions. Depending on the solution conditions, e.g. pH, the oxidation states of the actinides may vary. Thus, for an optimal fuel production process, it is important to study the oxidation state of various actinides at various conditions (speciation). Plutonium is very interesting since it can have oxidation states +3, +4, +5 and +6 in a solution at almost the same time.

Pu speciation at reducing conditions has been performed /Ödegaard-Jensen 2009/. Pu(III) and Pu(IV) were separated using a combination of HTTA extraction and $KBrO_3$. One fraction was extracted by HTTA (selective for Pu(IV)) and to the other fraction $KBrO_3$ was added to oxidise Pu(III) to Pu(IV) which then was extracted. This was done using the fact that oxidation of Pu(IV) to Pu(VI) is slow[10]. Pu(VI) content was investigated using NdF_3 precipitation which selectively traps Pu(VI). Pu(V) can then be reduced to Pu(IV) and sorbed onto a filter.

3.3.8 Dissolution and recovery of molybdenum CERMETS

Advanced fuels for transmutation of minor actinides and plutonium are presently under investigation. One potential fuel type is a CERMET (CERamic-METallic composite) where the actinide bearing phase is distributed in a molybdenum matrix. Mo is chosen due to its neutronic and thermal properties e.g. high thermal conductivity and relatively low neutron capture cross section. In damaged fuel pellets and fabrication scraps both the Mo matrix and the actinide phase could be recovered and re-introduced in fresh fuels. In a first step the Mo should be dissolved and separated from the actinide phase and thereafter an appropriate method for Mo metal conversion should be applied. Experiments have been performed on leaching and dissolution of the above mentioned fuel type /Aneheim et al. 2009/.

3.4 Royal Institute of technology (KTH) – chemistry

3.4.1 Scientific objectives

A project has been initiated to analyse complexing agents, used for partioning, with NMR. The scientific objectives are:

- To investigate if and how detailed molecular information on structures, reaction dynamics and chemical bonding might be used to explain differences in the chemistry of Lanthanide(III) and Actinide(III) complexes and how these differences might be used in Ln(III)/An(III) separation technology.
- To investigate if and how NMR and quantum chemical methods (QM) might be used to achieve these objectives.

The combination of experimental and theoretical methods is a key element in these studies, where we have used experimental data for yttrium(III) and europium(III) to validate the corresponding information obtained by the use of quantum chemical methods. This validation provides the rationale for using QM to predict the properties of actinide(III) systems. We expect that this information will be a useful step when planning experimental studies in An(III) systems, both for the selection of extraction ligands and solvents.

The first phase of the project has been a detailed study of the properties of a beta-diketone extractant, tris[4,4,4-trifluoro-1-(2-thienyl)-1,3-butanedione] (TTA); we consider this as a good model for other acid ligands. This phase has now been finished and the results are presented in three manuscripts:

Structure and Dynamics of Binary and Ternary Lanthanide(III) and Actinide(III) – Tris[4,4,4-trifluoro-1-(2-thienyl)-1,3-butanedione] (TTA) – Tributylphosphate (TBP) Complexes. Part 1, The Structure and Thermodynamics of Y, Eu, Am and Cm Complexes as Studied by Quantum Chemical Methods and X-Ray crystallography, with V. Vallet, A. Fischer, Z. Szabó and I. Grenthe as authors.

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[10] The oxidation from Pu(IV) to Pu(V) is slow, whereas the oxidation from Pu(V) to Pu(VI) is faster.

The Structure and Dynamics of Binary and Ternary Complexes in the Y(III)/Eu(III) – TTA – Tributylphosphate (TBP) System in Chloroform as Studied by NMR Spectroscopy. Part 2, The Structure and Thermodynamics of 8- and 9-Coordinated Binary and Ternary Y – TTA Complexes Studied by Quantum Chemical Methods, with Z. Szabó, V. Vallet and I. Grenthe as authors.

Structure and Dynamics of Binary and Ternary Lanthanide(III) and Actinide(III) – Tris[4,4,4-trifluoro-1-(2-thienyl)-1,3-butanedione] (TTA) Complexes. Part 3, The Structure and Dynamics of Binary and Ternary Complexes in the Y(III)/Eu(III) – TTA – Tributylphosphate (TBP) System in Chloroform as Studied by NMR Spectroscopy and Quantum Chemical Methods, with Z. Szabó, V. Vallet and I. Grenthe as authors; these manuscripts will be submitted to Dalton.

3.4.2 Summary of results

Experimental X-ray structures of $Y(TTA)_3(OH_2)_2$ and $Eu(TTA)_3(OH_2)_2$ have been compared with QM structures in various model solvents mimicking H_2O, $CHCl_3$ and CCl_4. Experimental and theory based bond distances agree within the experimental errors, but the conformation of the TTA ligands vary slightly with the solvent. These results provides the validation of the QM method for the structure and bonding in U(III), Am(III) and Cm(III) TTA complexes. We found that there are small differences in chemical bonding between the various M(III) complexes, but for uranium(III), a significant electron delocalization from orbitals centered on U(III) to empty orbitals on the oxygen donors in TTA; this effect was not apparent between Y(III) and Eu(III) and very small for Am(III) and Cm(III). We conclude that it seems less likely that the differences in chemistry between Ln(III) and Am(III)/Cm(III) is due to differences in chemical bonding. Figure 3-4 shows a three-dimensional representation of one of the singly occupied orbitals located mainly on the TTA oxygens in $M(TTA)_3(H_2O)_2$; M = Eu(III) and U(III). The figure demonstrates a significant overlap for U(III) but not for Eu(III).

The NMR data demonstrate the presence of isomers of the $M(TTA)_3(OH_2)_2$ complexes and the geometry and relative energy of them have been obtained using QM methods. The mechanism for chemical exchange between them have been studied both experimentally and using QM, with good agreement in the experimental and QM calculated activation parameters.

We have made QM studies of the reaction energy for two-phase exchange reactions of the type:

$Ln(TTA)_3(OH_2)_2(org) + An(OH_2)_9^{3+}(aq) \rightarrow An(TTA)_3(OH_2)_2(org) + Ln(OH_2)_9^{3+}(aq)$

that can be compared with experiments. However, the QM methods at our disposal are not accurate enough to be useful and our QM partners are presently working to improve the computation methods.

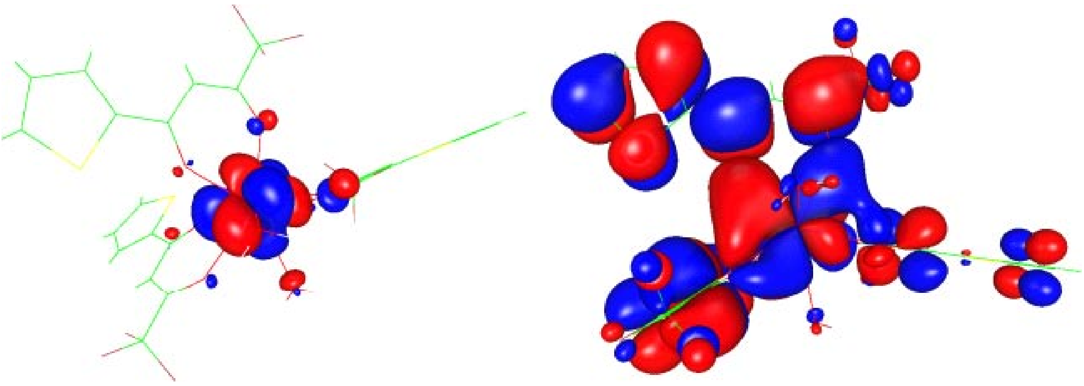

__Figure 3-4.__ Three-dimensional representation of one of the singly occupied orbitals located mainly on the TTA oxygens in $M(TTA)_3(H_2O)_2$; M = Eu(III) and U(III).

Addition of TBP resulted in a dramatic increase of the solubility of the binary Y(III) and Eu(III)-TTA complexes in the organic phase. The NMR data proved that two complexes formed in the ternary Y(III)-TTA-TBP system, $Y(TTA)_3(TBP)$ and $Y(TTA)_3(TBP)_2$, with the equilibrium constant, $4 \pm 2\ M^{-1}$, for reaction $Y(TTA)_3(TBP) + TBP \leftrightarrow Y(TTA)_3(TBP)_2$. The ^{31}P NMR data proved the presence of at least two structural isomers for $Y(TTA)_3(TBP)$.

Different ligand exchange reactions have been identified, very fast intramolecular exchange between the structure isomers, and a number of intermolecular exchange reactions between the free and coordinated ligands, the exchange between free and coordinated water and free and coordinated TBP is very fast, while the exchange between coordinated TTA and free HTTA is much slower. The rates of the exchange reactions and their activation parameters were determined by various dynamic NMR methods.

We suggest the same mechanism for the intermolecular HTTA/TTA exchange in both the binary and ternary systems with Y(III) and Eu(III). The first step is the fast coordination of HTTA to form the stable inner-sphere complex, which is followed by reactions that involve TTA chelate ring-opening/ proton transfer between entering and leaving ligand.

Based on our experiments in the Eu(III) system we can state that NMR spectroscopy is a very useful spectroscopic tool to study the formation and the ligand exchange dynamics of paramagnetic metal ion complexes.

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

As outlined in 2.3, the Sustainable Nuclear Energy Technology Platform (SNETP) is an organization where nuclear industry, research institutes, universities and other stakeholders are defining the needs for future R&D on nuclear power in Europe. One of the tree major pillars of SNETP is the development of Generation-IV fast-neutron reactors with closed fuel cycles – hence of direct relevance for the development of P&T. In the Strategic Research Agenda (SRA), issued by SNETP in May 2009, the priorites concerning R&D and deployment of Generation-IV reactors were presented /SNETP-SRA 2009/. It may be noted, that in the third call of FP7, the P&T project proposals to be evaluated for funding by the EC, are expected to respond to the needs defined in the SRA of SNETP.

SNETP has defined the strategy for fast-neutron reactor development with sodium-cooled fast reactors (SFR) as the first priority, considering the fact that the concept has been proven by operation of several such reactors in Europe. The lead cooled fast reactor (LFR) and the gas cooled fast reactor (GFR) are considered as alternative technologies. R&D topics for all three systems and their associated fuel cycle include

- Simplification of primary system design.
- Improved materials.
- Innovative heat exchangers and power conversion systems.
- Advanced instrumentation and in-service inspection systems.
- Enhanced safety.
- Partitioning and transmutation.
- Innovative fuels (minor actinide bearing).

Major demonstration projects are foreseen. These include the SFR prototype ASTRID, planned to be operating in France by 2020, and the construction of a demonstration facility for an alternative technology, either LFR or GFR. The choice of the latter should be made around 2012. In addition, ADS technology should be assessed in terms of its contribution to the closure of the fuel cycle, and may be demonstrated in the MYRRHA project in Belgium. These plans are reflected in FP7 projects recently having started or having been approved, such as the ESFR, LEADER, GOFASTER and CDT. The CDT project has been described above in Chapter 2 in this report.

The ESFR project (European Sodium Fast Reactor) is coordinated by CEA, and addresses viability and performance issues to support the development of a Generation-IV SFR, in particular the deployment of a demonstrator in 2020–2025. It includes activities on

- Fuel, fuel element, core and fuel cycle.
- Safety and security.
- Energy conversion system components and materials.
- Reactor system.

The project started in January 2009 and is co-funded with 5.8 M€ from the EC. It should here be noted that the ESFR project is well aligned with the French ASTRID project, carried out by a consortium consisting of CEA, AREVA and EdF, aiming at building an SFR in Marcoule (next to the site of Phénix), to start operation in 2020. The power level of ASTRID will be decided in 2010, while technological choices, such as steels for various components, should be selected by 2012, when also a formal decision to build ASTRID would be taken by the French government.

The LEADER project (Lead cooled advanced demonstration reactor) is coordinated by Ansaldo Nucleare, and has been allocated a funding of 3.0 M€ by the EC. The aim of the project is to perform the design of a low cost and fully representative scaled-down LFR prototype of suitable power. This design should be sufficiently detailed to allow in 2012 the choice of the LFR as the alternative fast-neutron reactor technology to be demonstrated in Europe by 2020. Much of the work would be based on the outcomes of the ELSY project described in Chapter 2. KTH participates in the LEADER project and coordinates the work package on education and training, in which the design of a low-power lead-cooled reactor for education and training (ELECTRA) will be performed by the participating PhD and Masters students.

The GOFASTER project is coordinated by AMEC (former NNC) and is funded with about 2 M€ by the EC. The aim of the project is to design a ~ 50 MWt helium-cooled fast reactor prototype (ALLEGRO), showing the feasibility to operate a gas cooled fast reactor.

4.2 Fuels

The development of fuels for ADS in Europe has presently reached the stage of post-irradiation examination of cermet and cercer oxide fuels irradiated in Phenix and HFR. In order to prove the feasibility of closing the fuel cycle with the selected fuels, methods for dissolution and reprocessing will have to be tested and validated. A proposal under the acronym ASGARD (Advanced fuelS for Generation IV reActors: Reprocessing and Dissolution) is under preparation for this purpose. The proposal is coordinated by Chalmers.

The on-going shift of focus towards transmutation in critical reactors, and in particular sodium-cooled reactors, means that French efforts on fuel development is being re-directed towards americium-bearing mixed oxide fuels and targets. Here, phenomena like internal clad corrosion and redistribution of fuel components are issues that require more detailed studies.

Nitride fuels are of interest in particular for application in lead cooled fast reactors. In the Swedish GENIUS project (Generation-IV research in Swedish Universities), fabrication of (U,Pu)N, (U,Pu,Zr)N and (Pu,Zr)N fuels will be performed in a new laboratory presently under construction at Chalmers. The methods used will be based on experience from fabricating and characterising (U,Zr)N fuels at KTH. The stability of (U,Zr)N fuels in contact with lead-bismuth at high temperature will be tested by KTH.

For the gas-cooled fast-reactor project, carbide fuel concepts with SiC-SiC cladding have been developed by CEA /Brunel et al. 2009/. This approach would require further validation, in particular when it comes to sealing of the clad and its irradiation resistance.

4.3 Materials

The development and validation of high-performance materials is crucial for the economic viability of Generation-IV reactors. For the fast-reactor concepts, the aim is to validate a material permitting both to increase the neutron dose to the cladding and to operate at high temperature. Austenitic stainless steels appear not to be able to meet the former objective. Conventional ferritic-martensitic steels however fail to meet the latter criterion. Hence, a major effort is presently devoted to developing methods for fabricating and welding oxide dispersion strengthened ferritic (12–14% Cr) and ferritic-martensitic (9% Cr) steels. This effort has been pioneered in Japan, and is now considered to be of major importance in French and European programmes.

For the lead-cooled fast reactor and ADS, the GESA method for making FeCrAlY surface alloying of steel components will need validation both in terms of irradiation performance as well as in terms of applicability on industrial scale.

Within the Swedish GENIUS project, a heavy liquid metal facility for corrosion studies will be constructed at KTH. Chalmers intends to study radiation damage structures in ODS steels with positron beam annihilation techniques. Further, modelling of swelling in austenitic model steels will be undertaken by KTH.

4.4 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.

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, but it seems unlikely that such an experiment can be performed before TSL closes.

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.

Recently, the CANDIDE project has resulted in recommendations for future work in the field (see section 2.3.9) /Koning et al. 2009/. Since the UU group is at cross-roads, these recommendations have been very useful in the identification of new research areas to go into, and the fact that UU has coordinated the project has resulted in very valuable contacts and visibility.

4.4.1 Future work for the UU group

The SCANDION particle therapy center under development in Uppsala will be operational around 2014. It is likely that TSL will then be closed down. There is a certain risk that the neutron beamline will be closed down even earlier. Thus, no new investment into the existing experimental setup at TSL is planned. Nevertheless, it is desirable to collect a data-set at 175 MeV that is as comprehensive as the 96 MeV data set. Therefore, especially measurements on elastic scattering from C and Y, and light-ion production from Ca should be made. With the large data sets at 96 and 175 MeV at hand, a systematic comparison of the experimental data with calculations from model codes are planned.

Up to now, the work at UU has been at very high energies where ADS is the only energy-related application. It is planned to continue with measurements of light ion production at lower energies, where data are of use also to Gen-IV systems, at another facility after the neutron beamline of TSL is closed. There are various options for this. One is to actively join the Neutrons For Science (NFS) collaboration at Spiral-II, GANIL, France. There, an intense white neutron beam with a maximum energy of about 30 MeV and quasi-monoenergetic neutrons up to 60 MeV will be available from 2012.

As described in section 3.2.2, experimental activities aiming at fission studies have been taken up by the UU group at other European facilities already several years ago. Another such activity will be measurement of independent fission yields at IGISOL, University of Jyväskylä, Finland. A measurement campaign on proton-induced fission of ^{232}Th at 25 MeV is planned for April 2010. In the second half of 2010 the IGISOL facility will be moved to a new beamline. This beamline is connected to the new K30 light ion cyclotron, which can deliver proton beams of up to 100 μA. This will allow for measurements of fission yields for neutron-induced fission. For this end a neutron production target with variable moderation will be developed. The aim is to measure fission yields for both thermal neutrons and neutron spectra similar to those in fast reactors. Such data have been identified as one of the top requests of the CANDIDE final report (see section 2.3.9) /Koning et al. 2009/.

These data shall then be used for the creation of a new nuclear data library of fission product yields from intermediate energy neutron induced fission (energy range 20–60 MeV) of actinides (^{232}Th, ^{231}Pa, ^{233}U, ^{234}U, ^{235}U, ^{238}U, and ^{237}Np). This work will be done in collaboration with Khlopin Radium Institute (KRI), St. Petersburg, Russia and the University of Jyväskylä. The project is led by Igor Ryzhov from KRI.

A new research activity which is currently started at UU concerns transportation of uncertainties in basic nuclear data through model codes and reactor core simulation codes to obtain macroscopic reactor parameters and their uncertainties (MACRO). This will be done with support from the Swedish Research Council, within the GENIUS project, and from Vattenfall AB. The realization of such a project has become possible due to the recent progress of nuclear model codes such as TALYS /Koning et al. 2008/ and improved computing power. Also this development is a response to the CANDIDE requests.

4.4.2 EU projects

A new nuclear data project called ANDES (Accurate Nuclear Data for nuclear Energy Sustainability) financed by the European Commission under the Euratom 7th framework programme (FP7) will start during spring 2010. This 3 M€ project got an excellent evaluation and will be realized as a collaborative project of 20 partners. This project contains four work packages that aim on "Measurements for advanced reactor systems (MARS)" (WP1), uncertainties and covariances of nuclear data (WP2), integral experiments for validation and constraints of nuclear data (WP3) and high-energy model validation between 150 and 600 MeV (WP4). UU will take part within WP4 with measurements at the TSL 175 MeV neutron beam.

Collaboration between ANDES partners and the GENIUS project is already underway. This holds especially for the GENIUS subproject MACRO and WP2 of ANDES. Our aim is to strengthen this link and make MACRO part of the follow-up project for ANDES.

The EFNUDAT project will be completed in 2010. Preparations for a follow-up project are in progress.

4.5 Partitioning

4.5.1 Overview of research directions

The separation of elements in nuclear waste is starting to become a mature science but there are still many obstacles to overcome. Traditionally there are two major strategies: hydro-processing and pyro-processing. Today the hydro-processing is the most promising one and also the one being developed for process scale. Ultimately, however, this may not be the final choice. One large uncertainty still to be addressed is how the future spent fuel will look. Depending on composition and burn-up of the fuel, hydro-processing may be unsuitable and thus leaving pyro-processing as the only option. In the light of this it is of vital importance that both these routes are continuously investigated to have preparedness for any future scenario that may arise. Another reason to argue for continuing to study both alternatives is the concept of a molten fuel reactor. In some of the conceptual sketches the fuel is continuously purified on line before it is recycled into the reactor again.

4.5.2 Hydro-processing

The hydro-processing has in the last years proven to be a viable method to reprocess the spent nuclear fuel of today to the level of separating out the trivalent actinides from the rest of the waste on a laboratory scale. In principle it is possible to start constructing a separation plant but there are many issues left like stability towards radiolysis and kinetics of extraction. These problems may be solved by designing the separation equipment in a different way, e.g. if the kinetics of extraction is too slow the contact time could be longer.

If the horizon is lifted a bit away from the European stage the picture is similar. For example, in the US a lot of effort has during many years been put on the TALSPEAK process. This process has been shown to work but many features are still not understood /Nilsson and Nash 2007, 2009/ and on this basis it may not be feasible to construct a full-scale separation plant.

Thus, in general, the hydrochemical-based reprocessing of nuclear waste is facing the development of industrialisation of the process. Basic studies are still needed but for the next decade we will see a clear move towards research focussed on the challenges associated with optimising a system and equipment for pilot plant development.

4.5.3 Pyro-processing

As mentioned earlier the pyro-processing is not as far come as the hydro processing and therefore the aim of the coming years is very different. There is still a great need to obtain data for possible different fuels and separation routes. Whether it will concern molten salt/molten metal separation or electrochemical separation does not matter. Currently there is no real reason to select one or the other, which is why efforts should be made in both directions to obtain enough flexible data and modelling ability to make pyro-processing able to face the uncertainties of future high burn-up fuels with possibly high minor actinide content.

There is also a materials problem that needs to be addressed in the future. The wear and tear of pyro-processing equipment is high and even if knowledge is available from the mining industry etc specific demands will be raised for materials used to process molten spent nuclear fuel. This is, however, a subject that is not typically addressed by nuclear chemists but highly skilled metallurgists that will have to join this part of process development.

4.5.4 NMR research at KTH

The KTH group has started investigation on extraction systems with uncharged ligands where nitrate is co-extracted from the aqueous phase into the organic phase. NMR experiments are planned that will allow studies of the exchange dynamics between the aqueous and the organic phase in a two-phase system. The QM activities will involve method development in order to be able to handle the actinide systems with sufficient accuracy to be useful for experimentalists.

4.6 Fusion-driven systems for transmutation

Fusion-driven systems (FDS) for transmutation constitute an alternative to accelerator-driven systems. In mainstream ADS concepts, spallation nuclear reactions are used for neutron productions. This typically means a proton beam of the order of 1 GeV energy impinging on a heavy-element target, resulting in an energy spectrum with a peak around 1 MeV and with a tail exhibiting a 1/E intensity distribution extending up to the incident beam energy. This energy spectrum is not optimal for incineration of nuclear waste, but the interest in spallation-based neutron production is because it presently represents the beat compromise between intensity and economy.

Ideally, neutrons should have an energy exceeding the thresholds for neutron-induced fission, but not much above, because higher energies causes radiation-shielding problems. Since all actinides have fission thresholds in the range up to about 2 MeV, the ideal neutron production reaction would be the deuterium-deuterium (D-D) fusion reaction, resulting in 2.5 MeV neutrons. This reaction, however, yields a relatively modest flux, and therefore the common choice is the deuterium-tritium (D-T) fusion reaction

$$D + T \rightarrow He^4\ (3.5\ MeV) + n\ (14.1\ MeV)$$

that produces neutrons with the energy 14.1 MeV. This reaction has a yield two orders of magnitudes larger than the DD reaction, and the energy is still not so high that the shielding problems are paramount.

The leading development lines of controlled nuclear fusion are based on the DT reaction. The ITER tokamak device, which is under construction at Cadarache in France, will produce more than 10^{20} neutrons per second in pulses lasting for 20 minutes or more. Such a high flux is higher than required for incineration of nuclear waste. Tokamaks can only be operated in pulsed mode, but other type fusion devices (mirror machines and stellarators) can operate in continuous mode. The latter are therefore more likely to be employed in a future production stage, whereas tokamaks are the presently prime option for plasma research and technology development. The ITER device will be a large toroidal plasma confinement device, with a 36 m long plasma column with a 2 m minor radius, and it has until recently been thought of as a pure fusion device intended to produce 700 MW thermal power. Neutron sources sufficient to drive a sub-critical reactor can be substantially smaller.

The rationale for FDS is the same as for ADS. From a reactor physics perspective, the only difference Is the energy spectrum of the neutrons. Thus, the main motivation for FDS is to use it for incineration of material that is difficult to handle in large quantities in critical reactors, i.e. elements like Americium whose fission is accompanied with a very small fraction of delayed neutrons, making criticality control difficult.

The origin of the hybrid breeder reactor idea is hard to trace, since the fusion research was classified in the 1950ies. Andrei Sakharov may have been the first researcher to discuss the possibility of a hybrid breeder reactor to amplify the output of the fusion neutrons /Sakharov 1990/. Hans Bethe proposed in 1978 a mirror-based hybrid reactor for production of fissile fuel /Bethe 1978, 1979/, requiring a fission mantle design that slows down the neutrons to avoid plutonium burning in the mantle surrounding the fusion machine. It should be pointed out that transmutation was not the intended use of these hybrid breeder reactors. Following the proposals of Bethe, the hybrid breeder reactor studies have also aimed at breeding new fissile material U-233 from Th-232, which can be used as fuel in a separate fission reactor. After the reactor accidents in Three Mile Island and Chernobyl, studies on hybrid breeder reactors were essentially abandoned.

It should be noted that the global civil fusion research is dedicated to pure fusion, i.e. the aim is to reach self-burning controlled fusion plasma conditions. It would, however, be much less demanding to develop a design in which the plasma is not self-sustained, but needs to be driven, whereas still being sufficiently intense to induce fission in a surrounding blanket consisting of, e.g. natural uranium. Such a device would reach net energy output for a much smaller and less costly plasma system.

The reason for not pursuing such a development is to a large degree political. In many countries where fission has been politically incorrect, fusion has been seen as a distinctly different technology. Involving fission as the major energy production in a hybrid fusion-fission device has therefore been negatively viewed by the proponents of the idea that fusion is markedly different from conventional nuclear power.

In principle, it is possible to operate an FDS with no multiplication in the system, but this means that each fission has to be induced by an externally produced neutron, resulting in a very large neutron source and correspondingly large costs and difficulties. Most designs therefore assume that a majority of the fissions are caused by fast neutrons produced in fission chain reactions of the elements to be incinerated, like in an ADS.

In subcritical systems driven by fusion neutrons, the fission power exceeds by a large factor the power output from the fusion neutrons. As high value of the effective neutron multiplication factor k_{eff} as possible is desirable to optimize power production, whereas reactor safety is improved by lower values. Sufficient reactor safety can be expected with $k_{eff} = 0.96$, and the produced fission power then exceeds the fusion power by a factor

$$\frac{P_{fis}}{P_{fus}} \approx 150$$

The primary fusion power is of minor importance in FDS, but the fusion neutrons control the level of fission reactions in the subcritical reactor core.

At present, the limiting factor for FDS development is the design of the plasma employed as neutron source. A number of technical solutions have been proposed, but none is close to industrial deployment. An introduction to the field and a survey of the proposed technologies are presented in a recent SKB report /Ågren et al. 2008/.

If a suitable fusion-based neutron source is developed, FDS might constitute an alternative to ADS. At that stage, the research challenges are essentially the same for the two concepts, i.e. materials and fuel issues can be expected to be major challenges, and the partitioning is not dependent of the core technology. The nuclear data development required is less demanding than for ADS, primarily not because the physics itself is easier, but because experiments are far less costly and the availability of experimental facilities is far better.

5 Short update of assessments in the 2007 report

In the previous status reports made for SKB in 2004 and 2007 /Ahlström et al. 2004, 2007/ 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 in the 2004 report. 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 items of the assessment are also updated or modified in the light of recent developments.

5.1 General

The most important change during the last few years is the rapidly increasing political acceptance of nuclear power. On a European level, the parliament and commission are now both positive to nuclear power in general, which has consequences also for the view on P&T. The EC Strategic Energy Technology (SET) Plan /EU SET 2007/ identifies nuclear fission as one of the most important future energy sources of Europe. The Strategic Research Agenda of the Sustainable Nuclear Energy Technology Platform /SNETP-SRA 2009/ outlines a strategy in which an industry-scale sodium-cooled fast reactor should be built around 2020, and research on parallel technologies (lead- and gas-cooled critical fast reactors, as well as ADS) should be pursued with the aim to demonstrate their potential.

These recent political changes have still not materialized in increased EC funding. 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.

It should be emphasized that until the last few years, P&T has been viewed by the EC as synonymous with ADS. With the recent comeback of critical Fast Reactors (FR), the situation might change. Within the commission the expectations seem to be an unchanged funding level during the foreseeable future, at least concerning ADS. The funding to FR research is more difficult to predict. P&T research with an ADS-focus has been politically acceptable for many nuclear-sceptical countries, whereas FRs are much harder to accept politically for anti-nuclear countries. It is therefore not unlikely that increased EC funding will be counteracted by some countries, resulting in that a larger fraction of the funding of FR development might be channelled outside the EC.

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. In the end of 2009, a large grant for research on Generation-IV reactors was awarded by Vetenskapsrådet to a consortium of Swedish universities, including all the groups active in P&T research with SKB support. This will lead to a significantly increased activity level in Swedish research on P&T.

The interest for R&D on ADS-based 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. The industry interest in FR development is rapidly growing.

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 participate in the international development and to maintain a reasonable level of competence in the country. 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 option for burning minor actinides in a second stratum, where LWR and FR constitute the first stratum.

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 to use lead as both primary coolant and 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.

Building and operation of a small experimental ADS-facility is necessary to confirm the concept. The Belgian MYRRHA concept has been identified by SNETP as the preferred choice for such a test facility /SNETP-SRA 2009/, but the Belgian government is not fully committed to the project. It would take another five to ten years to construct the facility, 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 levels required to meet this time target. It is thus unlikely that the time schedules suggested in the ADS-systems studies will be met.

The previous prediction that deployment of ADS-plants on an industrial scale is unlikely to occur before 2050 /Ahlström et al. 2004, 2007/ is still valid, but for another reason. Previously, this prediction has been based on the technical difficulties in combination with the relatively modest funding level. Our present prediction is rather based on the increased attention on FRs, that can be expected to have two side-consequences. First, FR development can be expected to solve many of the technical problems involved in ADS, in particular materials and fuel issues. Second, deployment of FR will most likely have higher priority than ADS development. Moreover, the final performance of fast reactors will be of seminal importance for the design of ADS. It seems generally accepted now that ADS should constitute a possible second stratum, with minor actinide incineration as main rationale. Thus, the amounts and composition of the waste streams from FR are of crucial importance for the ADS design. It seems therefore reasonable that ADS research is performed keeping a range of options open until the performance of FRs is clarified, and detailed design of an industrial ADS can thereby be expected to await an initial FR deployment and operation phase.

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, ASTRID, by 2020. This project has been adopted as the first step in the SNETP Strategic Research Agenda /SNETP-SRA 2009/.

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 data as those presented in the previous status reports /Ahlström et al. 2004, 2007/. 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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Note on Swedish references:

SKB Technical Reports (TR-yy-nn) and SKB Reports (R-yy-nn) are published in pdf-format on SKB's web-site *www.skb.se*.

Appendix 1

Reference group for P&T research and development

Members

• SKB	Fred Karlsson; *chairman*
• CTH – nuclear chemistry	Christian Ekberg
	Jan-Olov Liljenzin, *deputy*
• KTH – reactor physics	Jan Wallenius
• KTH – organic chemistry	Zoltán Szabó
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	Stephan Pomp, *secretary (from 2009-02-01)*

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• Kärnavfallsrådet	Eva Simic
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• SKC	Jan Blomgren *(from 2009-02-01)*

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