

## ACSEPT—Partitioning technologies and actinide science: Towards pilot facilities in Europe

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### ABSTRACT

Actinide recycling by separation and transmutation is considered worldwide and particularly in several European countries as one of the most promising strategies to reduce the inventory of radioactive waste and to optimise the use of natural resources. With its multidisciplinary consortium of 34 partners from 12 European countries plus Australia and Japan, the European Research Project ACSEPT (Actinide reCYcling by SEparation and Transmutation) aims at contributing to the development of this strategy by studying both hydrometallurgical and pyrochemical partitioning routes.

ACSEPT is organised into three technical domains:

- (i) Considering technically mature aqueous separation processes, ACSEPT works to optimise and select the most promising ones dedicated either to actinide partitioning (for the heterogeneous recycling of actinides in ADS target or specific actinide bearing blanket fuels in fast reactor) or to grouped actinide separation (for the homogeneous recycling of the actinides in fast reactor fuels). In addition, dissolution and conversion studies are underway taking into account the specific requirements of these specific fuels.
- (ii) Concerning pyrochemical separation processes, ACSEPT focuses on the enhancement of the two reference cores processes selected within FP6-EUROPART. R&D efforts are also devoted to key scientific and technical issues compulsory to set up a complete separation process (head-end steps, salt treatment for recycling and waste management).
- (iii) By integrating all the experimental results in engineering and system studies, both in hydro and pyro domains, ACSEPT will deliver relevant flowsheets and recommendations to prepare for future demonstrations at a pilot level.

After more than two years of work, significant progress was achieved in process development with the demonstration of the SANEX and innovative SANEX flowsheets. Chemical systems were selected for GANEX and are under study. In addition, efforts were made to increase collaborations, mutualise and homogenise procedures and share good practices. Based on these assessments, it is now time to look at the future challenges to overcome.

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A training and education program is implemented to share the knowledge among the partitioning community, present and future generations of researchers. Specific attention is paid to the funding of post-doctorate fellowships, two having been appointed respectively at the end of 2008 and at the end of 2009. Through this training and education programme, the first ACSEPT International Workshop was organised last March in Lisbon, Portugal. It gave an emphasis to young researchers' contributions (two thirds of the contributions) and allowed young scientists to meet and exchange with international recognised experts.

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## 1. Introduction

When considering sustainable energy development worldwide, one of the challenges of the nuclear energy is the minimisation of long lived radioactive waste but also the optimisation of the use of natural resources with an increased resistance to proliferation. Partitioning and Transmutation (P&T), associated to a multi-recycling of all transuranics (TRUs), should play a key role in the development of a sustainable nuclear energy through GENIV fast reactor systems associated with a closed cycle (Fig. 1). It would also permit the transition from the currently practiced mono-recycling of plutonium in Light Water Reactors (LWR) to actinides (U, Pu, Am, Cm) recycling in GenIV reactors, thereby allowing minimisation of radiotoxicity in the ultimate waste.

Considering this evolution towards closed fuel cycles, it has been considered as a priority to strengthen the links and synergies between partitioning and transmutation as well as with geological disposal or interim storage activities. This evolution towards more integration should thus materialise and be consistent with the building of a European vision on P&T and more globally on future sustainable nuclear systems, such as those considered within the European Sustainable Nuclear Energy Technology Platform (SNE-TP – [www.snetp.eu](http://www.snetp.eu)).

Despite the different national strategies envisaged for managing nuclear wastes, the options for actinide recycling implementation show a significant common trunk which allows drawing a consensual European roadmap for research and development activities as well as for future pilot-scale fuel cycle facilities. Actually, P&T has been initially pointed out in numerous studies as the strategy that can relax the constraints on the geological disposal, and reduce the monitoring period to technologically and manageable time scales (Fig. 1). Moreover, even when considering the phase out of nuclear energy, the combination of P&T and dedicated burner such as Accelerator Driven System (ADS) technologies, e.g. at regional scale, would allow the minimisation of the long lived waste to be ultimately disposed of (Fig. 1).

To implement this strategy at the horizon 2040–2050, it is expected around 2012 to review national positions, as well as the impact of the recycling of actinide on geological repository in terms of requirements and capacity. Scenarios for the recycling of actinides implementation and the evaluation of options such as homogeneous/heterogeneous recycling should also take into account the national capacities in fuel reprocessing and fuel fabrication, notably during the transient period. A review of a grouped recovery of trans-uranium elements (TRUs) vs. minor actinides (MA) selective separation should also be undertaken. Priorities should also be given following the review of ADS vs. critical fast reactor systems potentialities and their coolant options. All these reviews should lead to take decisions on demonstration facilities to be built at a time horizon 2015–2020.

In line with these timescales, the FP7 ACSEPT Collaborative Project (2008–2012) provides a R&D framework structured to develop chemical separation processes compatible with fuel fabrication techniques, in view of their future demonstration at the pilot level, so as to offer technical solutions to the various fuel cycle options envisaged today.

The work-programme of ACSEPT was then built to develop and design both aqueous and pyro processes to be implemented in future fuel cycle pilot demonstrations, on the basis of the progresses achieved within EUROPART in the development of treatment processes answering the needs of future fuel cycles (heterogeneous recycling, homogeneous recycling, treatment of refractory materials). In addition to the hydrometallurgical and pyrometallurgical domains, a third domain devoted to process integration and systems studies was implemented in ACSEPT to answer the specific challenge of future pilot designs. A global vision of ACSEPT in terms of process development is given in Fig. 2.

The objectives of ACSEPT are addressed by a multi-disciplinary 34 members consortium composed of European universities, nuclear research bodies and major industrial players from 11 EU Countries plus Switzerland, Australia and Japan (Table 1). All the participants are renowned experts in their field, with a long-standing experience in partitioning or fuel fabrication techniques. They are often accompanied by young researchers, who are well educated and trained, thus preparing for future generations of scientists.

Moreover, ACSEPT gathers world-class nuclear laboratories (JRC-ITU, ATALANTE facility at CEA, NNL Central Laboratory... ) with modern equipments that proved to be operational and reliable. Other laboratories e.g. also in universities equipped with glove-boxes with sometimes more flexibility and fewer constraints are included in the infrastructure park.

## 2. Background of ACSEPT

For several decades, many European countries have chosen to investigate the strategy of a closed fuel cycle, currently involving (i) the reprocessing of spent nuclear fuels to recover uranium and plutonium and (ii) the vitrification of long lived radionuclides including MAs (the final waste being expected to be disposed of in deep geological repositories). Nevertheless, the sustainability of an advanced fuel cycle relies on the possibility to maximise the energy usage of nuclear spent fuel and to provide improved waste forms for long-term storage: the removal of minor actinides (*i.e.* Np, Am, Cm, Cf) with U and Pu from the waste and their recycling in fast neutron reactors, will significantly reduce the radio-toxic inventory as well as the heat content. Therefore, the separation of all significant trans-uranium (TRU) elements and their incineration in fast neutron reactors or ADS is a key feature of advanced fuel cycles. However, new reprocessing technologies (spent fuel dissolution, actinide separation and fuel refabrication) are required to address the challenges, which cannot be met by current generation of chemical plants. Two strategies are proposed today for the recycling of the actinides issuing from the various forms of future nuclear fuels (oxides, carbides, nitrides, metallic or inert matrix fuels): (i) their homogeneous recycling in mixed fuels (via a prior grouped separation of the actinides: GANEX concept) 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:

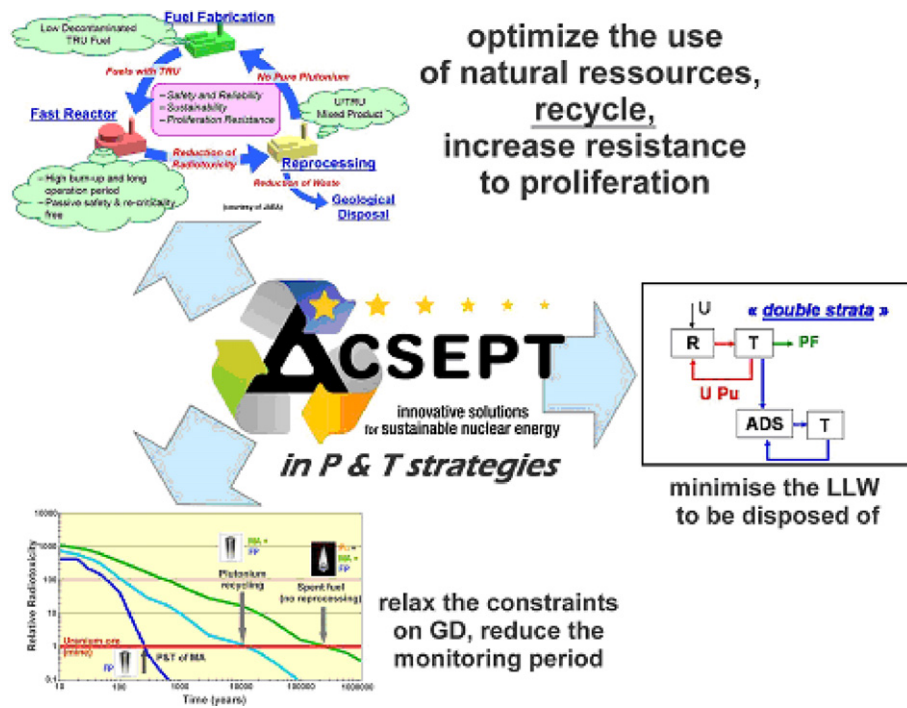


Fig. 1. ACSEPT promises to have a positive and beneficial impact for Europe, especially to ease the management of nuclear waste and to increase the public acceptance of nuclear electricity production.

- 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 (EBR-II fuel treatment at Idaho National

Laboratory), but without reaching the industrial development level.

2.1. Hydrometallurgy

As regard to partitioning processes, many countries have developed for the past 4 decades hydrometallurgical processes to recover

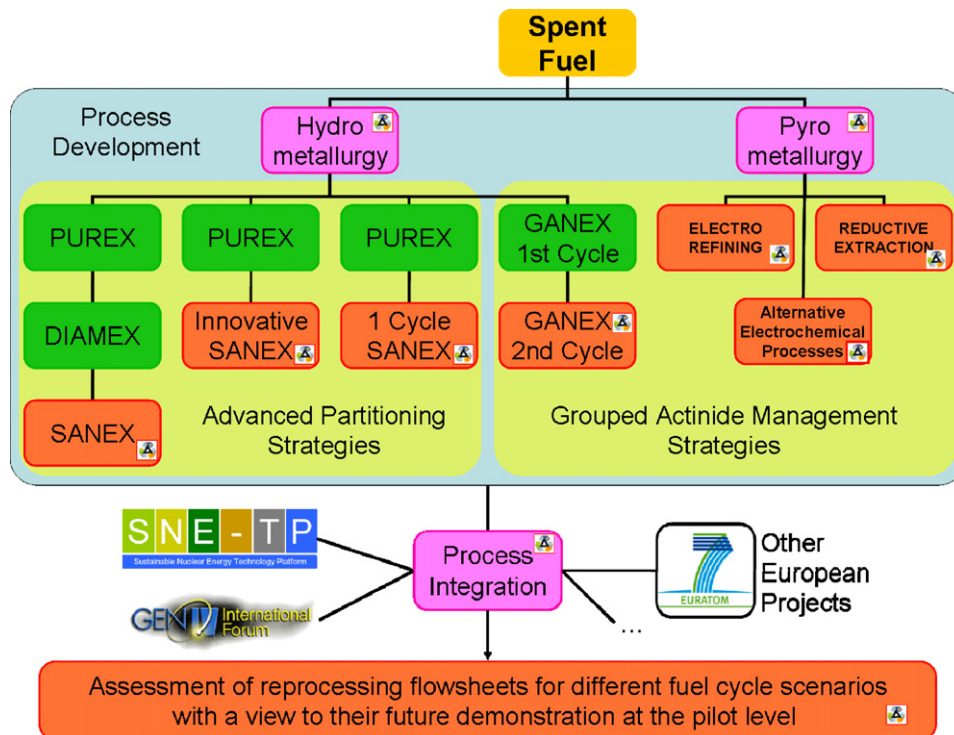


Fig. 2. ACSEPT provides a structured R&D framework to develop chemical separation processes, with a view to their future demonstration at the pilot level.

**Table 1**  
ACSEPT partners.

National Nuclear Research Laboratories Commissariat à l'Energie Atomique et aux Energies Alternatives	CEA	France
Centro de Investigaciones Energeticas, Medioambientales y Tecnologicas	CIEMAT	Spain
Ente per le Nuove Tecnologie, l'Energia e l'Ambiente	ENEA	Italy
Forschungszentrum Juelich GmbH	FZJ	Germany
Karlsruhe Institute of Technology	KIT-INE	Germany
Instituto Tecnológico e Nuclear	ITN	Portugal
National Nuclear Laboratory	NNL	UK
Nuclear Research and Consultancy Group	NRG	Netherlands
Paul Scherrer Institut	PSI	Switzerland
Australian Nuclear Science and Technology Organisation	ANSTO	Australia
Universities and/or National Fundamental Research Laboratories		
Chalmers Tekniska Hoegskola Aktiebolag	Chalmers	Sweden
Centre National de la Recherche Scientifique	CNRS	France
Czech Technical University Prague	CTU	Czech Rep
Consejo Superior de Investigaciones Cientificas	CSIC	Spain
Univerzita Karlova V Praze (Charles University in Prague)	CUNI	Czech Rep
Instytut Chemii i Techniki Jadrowej	ICHTJ	Poland
Fundacio Privada Institut Catala d'Investigacio Quimica	ICIQ	Spain
Institute of Inorganic Chemistry Academy of Sciences of Czech Republic	IIC	Czech Rep
Politecnico di Milano	POLIMI	Italy
Rijksuniversiteit Groningen	RUG	Netherlands
Universiteit Twente	TWENTE	Netherlands
The University of Edinburgh	UEDIN	UK
Université de Liège	ULG	Belgium
Université Louis Pasteur	ULP	France
Universita degli Studi di Parma	UNIPR	Italy
Université Pierre et Marie Curie – Paris 6	UPMC	France
The University of Reading	UREAD	UK
Nuclear Industrial Companies:		
ALCAN Centre de Recherches de Voreppe	ALCAN	France
Compagnie Générale des Matières Nucléaires-AREVA NC	AREVA NC	France
Electricité de France S.A.	EDF	France
Ustav Jaderneho Vyzkumu Rez A.S. (Nuclear Research Institute Rez plc)	NRI	Czech Rep
Central Research Institute of Electric Power Industry	CRIEPI	Japan
EU Nuclear Research Laboratory		
Commission of the European Communities – Directorate General Joint Research Centre	JRC-ITU	EC
Small and medium size enterprises		
CINC Solutions BV	CINC	Netherlands
Specific exchanges in pyrometallurgy (not in the consortium)		
International Science and Technology Center	ISTC	Russia
Korea Atomic Energy Research Institute	Kaeri	Korea

minor actinides elements in order to decrease the radiotoxic inventories of nuclear waste. However, none of these processes has ever been implemented at the industrial scale but their R&D has sometimes reached demonstration levels at the laboratory scale. Most of the partitioning strategies rely on a three step approach:

- Separation of U (and sometimes also Pu) from spent fuel dissolution liquors;
- An(III) + Ln(III) co-extraction;
- An(III)/Ln(III) separation, the latter step being the most difficult because of the similar chemical properties of 4f and 5f elements.

The processes developed around the world differ from the extracting systems involved in these different steps and the possibility to achieve a selective actinide extraction in a single one:

- For the first step, tributyl phosphate (TBP) is the basis of the PUREX, UREX and COEX<sup>TM</sup> processes, developed in Europe and the US, whereas monoamides support the BAMA process developed in Japan and India (selective U(VI) extraction by branched alkyl monoamide).
- For the second step, malonamides, CMPO and TODGA are used in the DIAMEX, TRUOX and ARTIST 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 US allow all actinides from U to Cm to be co-extracted from the dissolution liquors.
- 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, whereas other soft donor atoms containing ligands, such as polyaminocarboxylates (HEDTA, DTPA) have been tested in France, the US (TALSPEAK) and Japan (SETFICS) to selectively strip the trivalent actinides.

For more than 10 years, several European Projects carried out under the 4th (NEWPART), 5th (PARTNEW, CALIXPART) and 6th (EUROPART) Framework Programmes have widely contributed to the development of these processes.

The main objective of NEWPART was to develop processes for the separation of minor actinides (mainly Am and Cm) from PUREX raffinates without the generation of secondary solid wastes and by minimising the majority of the drawbacks encountered in the previous process studies, such as the precipitation of radionuclides; the difficulties in the stripping of the actinides from the solvent. These minor actinides are present in the high active effluents issuing the reprocessing of the spent fuel at the trivalent state, An(III). Consequently, their separation from these acidic effluents from the other elements (fission products), and particularly the trivalent lanthanides, Ln(III), is a difficult task. Consequently, the proposed principles for process design were to develop molecules (extractants, diluents, aqueous reagents) that can be converted into non radioactive gases at the end of their use in the processes, (CHON principle), with no reduction of the acidity of the HLLW to be processed. These studies have led to the development of the DIAMEX process based on diamide molecules which allows the recovery of both An(III) and Ln(III) from a PUREX raffinate.

Then, in PARTNEW, solvent extraction partitioning processes of americium and curium from lanthanides were proposed (SANEX concept), based on some organic molecules families: diamides, nitrogen polydentate ligands (BTPs), acidic sulphur-bearing ligands. Both basic research and process developments were carried out for the systems studied.

In the same timescale, CALIXPART aimed at developing another molecule family for the direct extraction of An(III) from raffinates: the functionalised calixarenes which make possible to obtain a high intra class selectivity, for example for the alkali cations (separation cesium/sodium) or to discriminate between different lanthanides with respect to their size. In this project, 160 extractants were synthesised and tested.

EUROPART was fruitfully devoted to the synthesis, characterisation and the assessment of extraction properties of more than 100 new ligands from various families. Only a few have successfully passed the screening tests, thus fulfilling the requirements for further process development. Basic and process development studies have also been carried out on several separation systems issued from the former FP5 projects. For the co-extraction of An(III) and Ln(III) from PUREX raffinates, five systems have been studied besides the reference DIAMEX solvent. Among them, only the "TODGA/TBP" mixture was selected to prove the scientific feasibility of the co-extraction of An(III) and Ln(III) from PUREX. The

four other systems have been investigated for “An(III)+Ln(III)” co-extraction, but no real flowsheet development has been carried out. For the separation of An(III) from Ln(III) from DIAMEX product solutions, two SANEX systems have been studied. Only the CyMe4-BTBP/DMDOHEMA system was investigated up to the design of a process flowsheet, but no counter-current test could be performed because preliminary experiments on single stage centrifuges have pointed out kinetic problems (besides, this system still lacks radiolytic stability). For the group actinide separation (An(III–VI)) directly from spent fuel dissolution solutions, in spite of all the efforts carried out by the partners involved to develop viable processes, no system has been developed up to the highly active test even if the “TODGA/TBP” mixture dissolved in kerosene, which was also selected for actinide group separation, was investigated up to batch tests on genuine PUREX high active raffinate.

Concerning the co-conversion of actinides, co-precipitation and sol-gel routes were proposed to convert actinides from partitioning solution into fuels. For the co-precipitation method, a synthesis route of pellets was developed and the parameters optimised. For the internal gelation route, an excellent method to produce uranium-based kernels was assessed but it still needs some optimisations. For the external gelation method, promising results were obtained for neptunium conversion. For the colloidal sol-gel method, promising preliminary results were obtained, offering a great flexibility particularly when handling minor actinides.

## 2.2. Pyrometallurgy

For more than 50 years, pyrometallurgy has been studied as an alternative strategy in spent fuel reprocessing. Indeed, it is considered as the reference route for molten salt reactor fuel reprocessing, the reprocessing of some types of fuels today envisaged for GenIV might not be compatible with current hydrometallurgical processes. A pyrochemical process 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.

The collaborative work started within the FP5 PYROREP project. It was dedicated to the development of different separation processes based on pyrochemistry (high temperature chemistry in molten salts and liquid metals). The planned work including extensive experimental work and documentary concept evaluation studies, focussed on segregation processes by means of electrolysis and/or metal/salt exchange in chloride or fluoride media. Thanks to a comprehensive R&D program, new validated basic data were acquired, assessed and used to establish pyrometallurgical processing flowsheets suitable for use with advanced fuel cycles designed to minimise the radiological impact from nuclear waste. This joint effort consolidated and revive European expertise in this field.

In FP6, EUROPART focussed on basic properties acquisition, process developments, specific waste management and system studies. A very important work was done in thermodynamical data acquisition in molten chloride media, with a comprehensive study of actinides, lanthanides and some other important fission products. The studies carried out on liquid metals confirmed the choice of aluminium as well as for an electrochemical process in molten chloride than for a reductive extraction process in molten fluoride. These two efficient processes have been selected as promising

core processes for the separation of An from Ln as demonstrated by flowsheet calculations and system studies. Real progress was achieved in the decontamination of spent chloride salts coming from electrorefining, the complementary techniques based on zeolite ion-exchange filtration and phosphate precipitation have been selected for their potential to clean up spent salt efficiently. If some specific matrices for salt confinement were identified (sodalite, polylite), a lot of work is still to be done in this field.

## 3. ACSEPT, a comprehensive scientific program

### 3.1. Hydrometallurgy

The hydrometallurgy domain of ACSEPT, DM1, is devoted to the development and assessment of sustainable and non proliferating hydrometallurgical processes that could improve the management of GenIV nuclear spent fuels, either in an actinide heterogeneous recycling strategy or in a homogenous one. DM1 therefore supports laboratory scale demonstrations of the most advanced separation processes to prepare for future demonstrations of fuel dissolution, reprocessing and refabrication at the pilot scale (through close collaboration with the Process Domain, DM3). It address (i) the dissolution of spent nuclear fuels in WP1, (ii) the partitioning of the transuranic elements in WP2 and WP3, and (iii) the refabrication of nuclear fuels using reprocessed materials in WP4.

The work-package dealing with head-end steps addresses challenging dissolution issues for the treatment of advanced (GenIV) nuclear fuels. However, the aqueous matrix (either simulated or genuinely active) used for the development of partitioning processes will essentially be nitric acid. Although current fuels are generally oxides, future fuels may be in oxide, carbide, nitride or metallic form. Future fuels envisaged for the homogeneous recycling have Pu and minor actinide contents respectively of ~20 wt% and 5 wt%. 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 actinides to residues must be minimised to maximise the benefits gained from P&T fuel cycles. The work in this field is divided in two main parts:

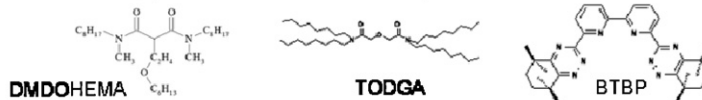
- Conceptual studies of potential chemical treatment options (direct dissolution or pre-treatment) for the dissolution of various types of spent nuclear fuels issuing from current and next generation reactors.
- Experimental and scientific studies of selected potential chemical treatment options (direct dissolution or pre-treatment) for the dissolution of various types of spent nuclear fuels issuing from current and next generation reactors

In the “process development” work-package, ACSEPT aims to develop three routes (two dedicated to minor actinide partitioning; one dedicated to the group separation of actinides), in the continuation of EUROPART, as illustrated in Fig. 3.

The first route is the one cycle direct selective extraction of minor actinides (Am, Cm) from a PUREX raffinate or the alternative two cycle process with direct selective extraction from a first cycle DIAMEX raffinate. This work is based on the acquired knowledge from the SANEX studies undertaken within EUROPART. The current BTBP reference system 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 improving ligand stability vs. radiolysis, An(III) extraction/stripping kinetics and process simplification (3/2/1 steps).

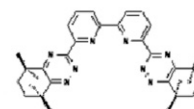
## •Heterogeneous recycling From the less challenging to the more challenging

- “regular” SANEX (on a DIAMEX raffinate), selective actinide extraction
- “innovative” (stripping) SANEX (on a PUREX raffinate), Extraction of An, Ln and selective stripping of An.
- “1 cycle” SANEX (on a PUREX raffinate), selective extraction of An



## •Homogeneous recycling

- GANEX, group actinide extraction
- Two cycles:
- 1<sup>st</sup> cycle : quantitative Uranium extraction,
- 2<sup>nd</sup> cycle : group actinide extraction



BTBP ?

Only the second cycle is under studies within ACSEPT

Fig. 3. Different strategies in process development.

The second route is the selective stripping of minor actinides after their extraction by approved lipophilic ligands (e.g.: malonamides, diglycolamides), in one or two cycle processes and based on selective aqueous complexation of actinides. The baseline clearly comes from achievements of France, USA and Japan as this concept has never been studied before in the previous FPs. It thus represents an alternative route if the An(III) selective extraction cannot be developed.

The third route is the group separation of actinides (GANEX) after a first U separation. All efforts are concentrated on the challenging development of processes for a grouped actinide separation (Np, Pu, Am, Cm. . .) rather than on the separation of uranium that represents lesser difficulties. The TRU actinide group separation has only been scarcely investigated in previous projects, and many challenging technological obstacles (e.g. solvent loading capacity, oxalate substitute) have to be overcome before demonstration tests can successfully be carried out. The group separation process concepts is directly derived from those studied for the one or two cycle selective extraction or stripping of minor actinides, but with higher challenging scientific issues that are due mainly to the presence of plutonium and to a lesser extent due to neptunium in the feed solution.

For each of these routes, the aim is to achieve the following successive milestones: the design and synthesis of promising extracting molecules, the selection of the most promising extracting system, the elaboration of flowsheets for further experimental implementation, cold and/or ‘spiked’ tests of these flowsheets and finally the hot test demonstration at the lab-scale.

To help in these process developments, a “ligand design and assessment” work-package was implemented to benefit from the achievements or conclusions of former Integrated Projects in the field of trivalent actinide separation. Moreover, an exploratory research will be maintained for the design of new extracting molecules, this topic being considered as the driving force of the project to further develop aqueous separation processes. Strong efforts are consequently brought to this field so as to stimulate new idea. The outcome of these studies will be directly the production of sets of promising molecules to be further studied for process implementation. Only the ligands which will have successfully passed the screening tests of this WP will be considered for process development studies in the “process development” work-package (Fig. 4).

Exchange of information between the partners involved in these two WPs will be fostered to identify and solve engineering or technological problems that would occur if implementing the partitioning processes at the industrial scale.

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 end-step to close the loop studies in the “conversion” work-package. The objectives of the research to be carried out in this field are related to the development of methods for the co-conversion of actinide solutions (variously mixed) to polyactinide containing solids for fuel preparation (GenIV, Transmutation). To reach these objectives, different co-conversion processes have been identified, based on co-precipitation and sol-gel routes. In addition, new alternative routes such as co-denitration and co-conversion by impregnating solid extractants, followed by thermal treatment, show also promising assets but have been less studied so far. 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.

The main objectives of ACSEPT in hydrometallurgy is thus to develop aqueous chemical processes in the various fields of spent fuel dissolution treatment, separation process development and fuel refabrication, and to demonstrate by the end of the project their technical feasibility at the laboratory scale on actinide solutions.

DM1 joins together all the major European nuclear research bodies in nuclear hydrometallurgy (e.g.: CEA, JRC-ITU, CIEMAT, FZJ, KIT-INE, and NNL), as well as the Universities or research bodies acting in support to nuclear research and development programs (e.g.: CHALMERS, ICHTJ, ULG, POLIMI, CTU, CNRS, CUNI, UREAD, ICIQ, CSIC, IIC, UNIPR and TWENTE).

### 3.2. Pyrometallurgy

To make advances beyond the current state of the art in pyrochemical separation processes, the pyrometallurgical domain of ACSEPT, DM2, is built on considering a process approach based on system studies. The four Work-Packages of DM2 represent the four main steps of a process bloc diagram: head-end steps, core process development, salt treatment for recycling, and waste conditioning. All the results will be integrated in the Process Domain DM3 in order to orient ‘online’ the R&D studies and to propose, at the end

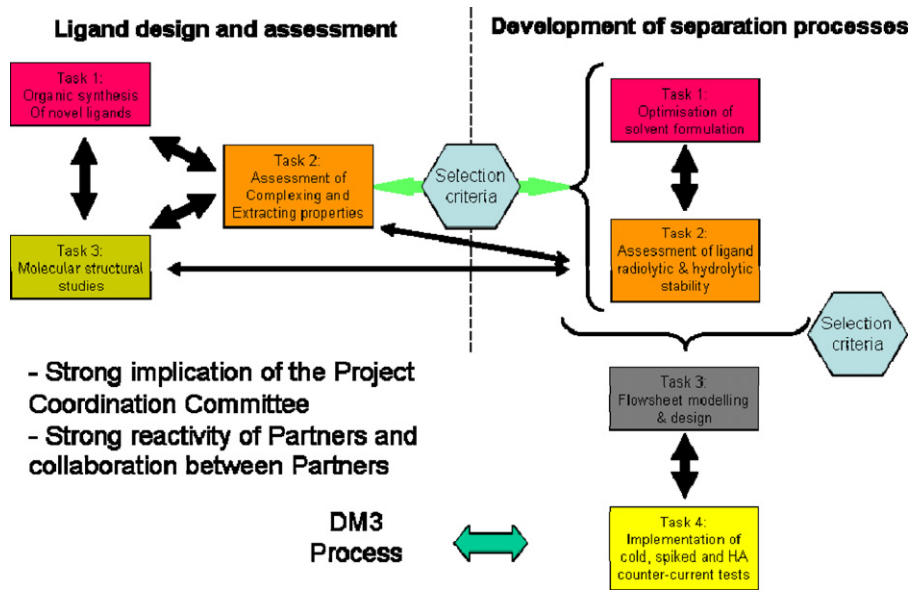


Fig. 4. Connections between “process development” and “ligand design and assessment” WPs.

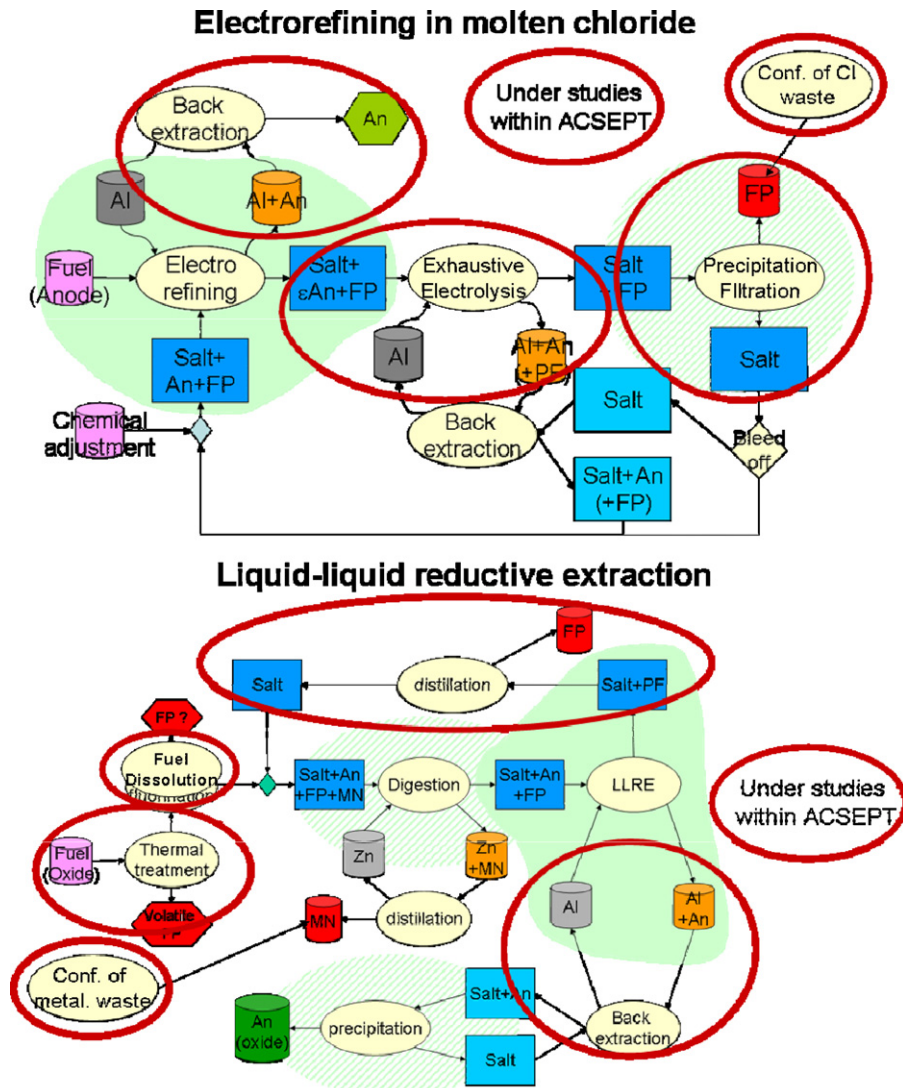


Fig. 5. The work to be done in ACCEPT in DM is orient by the validation of process flowsheets.

of the contract, validated flowsheets (Fig. 5). In addition, process modelling, materials and engineering studies will be performed.

In each WP, the technological blocks identified in EUROPART as key scientific points for the development of the two reference pyrochemical separation processes will be optimised and validated on one side or will be the scope of development studies if needed on the other side.

Head-end steps will be developed in fluoride and optimised in chloride in the first work-package. The studies concern mainly the electrochemical dissolution of oxide fuel in fluoride and the direct reduction of oxide to metal, both in chloride and fluoride media. The behaviour of fission products during the head-end thermal treatment is also investigated.

In the “process development” work-package, some ancillary steps of the two promising core processes must be assessed. The work in ACSEPT focuses on the exhaustive electrolysis step in chloride and on the 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. This is important because, first, the two reference processes show a common weakness which is the still undemonstrated viability of the An recovery step, and second because fluoride ions would be more compatible with the current industrialised vitrification technology than chloride ions thus making waste management much easier. This work includes development of specific set-ups for electrochemical experiments of An and Ln, investigation of basic data and material stability, and electrodeposition experiments onto different cathodes.

Progress is still expected in the work-package on “salt treatment for recycling”. Considering the main conclusions for the salt treatment processes investigated in EUROPART, the first task addresses some of the main development issues arising for the treatment and recycle of spent chloride salt. These include the practical application of a zeolite-based column for salt clean-up, as well as techniques that address the poor performance of current clean-up techniques e.g. electrochemical methods or the use of a closed aqueous ion-exchange loop for salt decontamination. Alternatives to the phosphate and carbonate precipitation processes will be investigated for chloride salt decontamination. Specifically, the use of gaseous reagents is to be studied in order to prevent contamination of the salt with solid reagents, as well as minimising the potential increase in salt inventory. The decontamination and potential recycle of spent fluoride salt has not been investigated in any detail prior to this programme. A task is therefore arranged to address this shortfall in technical knowledge, applied to the reference core process in molten fluoride. The most promising process seems to be based on distillation. In addition, an understanding of the phases and speciation of the FP present in the melt is essential in order to be able to understand and optimise the various salt treatment processes.

The “waste conditioning” work package foresees experimental activities about synthesis and characterisation of matrices suitable for conditioning chloride, fluoride, and metallic waste coming from pyro-processes, with the aim to minimise the waste volume and to comply with the criteria for storage and disposal. First, a joint research programme about synthesis and characterisation of sodalite will be conducted in order to assess the potentialities of this material as confinement matrix. As an alternative, studies will be carried out to optimise conditions to obtain the chloroapatite phase as agent for incorporating alkaline, alkaline-earth and rare earth elements. The immobilisation of noble fission products (Pd, Mo, Ru, Rh, Tc, etc) will be studied through the formation of solid solutions in Cu/Ni, Cu/Sn, and Al based alloys. Small ingots prepared inside furnaces under controlled anoxic atmospheres will be characterised by SEM, EPMA, XRD, and TGA analyses. The experimental activities will be completed by studies on alloy corrosion in water,

aimed at identifying the key parameters (such as corrosion potential) to be taken into account for description of metal behaviour under water.

DM2 joins together all the major European players in pyrometallurgy. (CEA, ITU, NEXIA, CIEMAT, ENEA, NRI as nuclear research bodies; POLIMI and UPMC as universities; CNRS as a national research institution, EDF as a potential end-user), but also non European research bodies (CRIEPI and ANSTO). Links also exist with related ISTC projects and KAERI.

### 3.3. Process integration

The Process Domain of ACSEPT, DM3 is organised to carry out detailed engineering and systems studies on aqueous and pyrochemical separation processes, thus paving the way to a future demonstration at a pilot level. Divided into three Work-Packages, this domain tackles any challenging problem in order to meet the request from industry to include the early engineering integration methodology and perspective so as to insure a minimisation of the cost of the final processes, a better integration of project goals and constraints. This should help maximising the overall chance of success of the whole project.

To reach its objectives, this domain creates constructive feedback between the different domains in order to help achieve optimisation and rationalisation of the research efforts, and on early identification of the engineering constraints at a systems level. It aims to also foster possibilities of partners to collaborate between domains. It will finally seek to generate sufficient scientific and engineering data to support the design concept of a technology and engineering demonstrators in a future project. Hence one of the main goals is to have established an understanding of some of the key engineering factors and engineering technological challenges by the end of the project. This will enable an effective assessment of the technical feasibility at larger scale of any retained process option.

P&T requires a coherent approach, where all necessary steps should be compatible and integrated with one another, thus building a chain, representing a comprehensive P&T scheme and an adequate closing of the nuclear fuel cycle. This is schematically represented in the figure below, which shows that irradiated fuels and targets should be compatible with subsequent dissolution (head-end steps) and separation processes, which at their turn should provide suitable precursors for fabrication of fuels and targets for transmutation.

Part of DM3 work aims at reinforcing integration between P&T related activities in order to develop strong links with other European projects. Mainly two technical actions are considered here:

- a study of the feasibility of a minor actinide-bearing pin fabrication in the framework of the MARIOS programme to be implemented in FP7-FAIRFUELS,
- a study on the reprocessing capabilities of different types of minor actinide bearing targets.

In DM3, 15 partners, from industry (ALCAN, AREVA NC, EDF, CINC), nuclear research bodies (CEA, CIEMAT, ENEA, JRC-ITU, NEXIA, NRG, CRIEPI) and universities (CTU, ICIQ, RUG, UEDIN) join their skills and resources to go further in these developments with specific efforts to disseminate their results. The goal is also to educate all the consortium members on the importance of early integration so they can incorporate as much as possible the process viewpoint in their own field of research when exploring new ideas.



#### 4. Training and education: a key issue

ACSEPT seeks 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, teaching establishments, young researchers thus providing the necessary competence and expertise for a sustainable development of nuclear energy.

This Domain seeks 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 which will support co-operative work among the members of the ACSEPT nuclear community. Of course strong links are established with the on-going ACTINET I3 direct or derived actions. The focus will be on complementarities rather than duplication so as to optimise EC funds.

The main goal of education and training programme in ACSEPT is to enhance the knowledge in separation sciences within the consortium. 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 rely on the mobility of personnel. Actually, ACSEPT supports the funding of post-doctoral students and helps students to attend specific ACSEPT training sessions or summer schools.

#### 5. First achievements of ACSEPT

After two years of work, significant progresses were achieved in hydrometallurgy. In pyrometallurgy, the work progresses following the work-plan. Important efforts were also devoted to cross-cutting activities, training and education.

In hydrometallurgy, one hot test was performed at ITU to validate the SANEX concept and a second hot-test was performed at CEA in April 2009 to validate the innovative stripping SANEX concept. In this concept, An and Ln are together extracted from a PUREX raffinate but only the An are then selectively back-extracted from the organic phase to an aqueous phase, combining DIAMEX and SANEX processes. Therefore, the efforts could be mainly put on the development of the much more challenging one cycle SANEX (where An would be selectively extracted from a PUREX raffinate) and GANEX processes on the three remaining years of the project. Around 40 new molecules were synthesised. Most of them were tested for extraction, but had unfortunately no improved extracting properties. Therefore, no molecule was then selected for further process development. In order to encourage new ideas in this field, a specific brainstorming meeting was organised in January 2009, joining all the teams involved in organic synthesis around the Hydro Domain Coordination Team. At this occasion, the constraints of a process development and define the requirements of each route to be studied were deeply described. The brainstorming led to the proposition of some new molecules, to be synthesised and tested in the following months. This meeting was highly appreciated by the attendees, contributing to the effort of training and cross-fertilisation.

In pyrometallurgy, the work-program is on the longer term and no major milestones were planned in the first year. One hundred grams of sodalite were successfully synthesised and are now ready for pelletisation prior to leaching tests.

In DM3, in collaboration with DM4, a meeting devoted to cross-fertilisation and integration of the different ACEPT communities was organised in Prague in September 2008. One session was aimed at explaining how a system study is performed and what the needs

to build a relevant process flowsheet are. A second one was a cross-fertilisation session proposed by ALCAN in pyrometallurgy to present how to develop an industrial process in pyro. Two additional courses were given, bases in solvent extraction and bases in pyrometallurgy, contributing to the training and the integration of all the ACSEPT beneficiaries.

Two websites were developed; one restricted to ACSEPT members for collaborative work and a public one for external communication. On the restricted website, all the documents related to the project are directly uploaded by their authors and validated by the coordinator. These documents are then available in real time to all the community. The public website for communication was visited around 1000 times from July 2008, from more than 45 different countries. It is well referenced on Google, fostering the visibility of the project.

The first ACSEPT post-doctoral student was appointed in January 2009. Formerly PhD student in Juelich, he is working at the University Louis Pasteur in Strasbourg, in collaboration, among others, with Chalmers on molecular modelling. The second post-doctoral student was appointed in September 2009. He shared his time between CNRS-Toulouse and JRC-ITU to work on plutonium chemistry in molten salts.

Last but not least, two important events were organised through ACSEPT:

- An international training meeting was organised in March 2009 in Madrid on pyrometallurgy. Joining ACSEPT partners, ISTC members and KAERI representatives, this meeting joined more than 60 people on three days to exchange on their experiences. Formal exchanges with both bodies were established. The participation of ACSEPT students to this training session was supported by DM4 funding.
- An international workshop, joining more than 100 people was organised from 31st of March to 2nd of April 2010 in Lisbon. Among the 35 oral contributions, 22 were given by young scientist, with the opportunity to exchange with acknowledged international experts in the field of partitioning, transmutation and waste management.

#### 6. Conclusions

With the technical advances expected within ACSEPT on advanced closed fuel cycles it should be possible to propose options to Governments, European utilities as well as technology providers. A technically feasible strategy for the recycling of actinides will certainly produce positive arguments in the sense that:

- European decision makers and public opinion could be convinced that some sound technical solutions for a better nuclear waste management are now available,
- One of the identified major drawback of the nuclear energy production, i.e. the potential dissemination of hazardous radionuclides within the bio-sphere in the far future, is considerably reduced, thus paving the way towards a sustainable energy supply.

In addition, the training and education programme will significantly help developing nuclear skills in Europe.

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