ACSEPT: a new FP7-Euratom Collaborative Project in the field of partitioning processes for advanced fuel cycles

Stéphane Bourg, Emmanuel Touron, Concha Caravaca, Christian Ekberg, Emmanuel Gaubert, Clément Hill

CEA/DEN/MAR/DRCP, Bat 181, CEA Marcoule, BP 17171, 30207 Bagnols/Cèze Cedex
stephane.bourg@cea.fr

***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, thus contributing to the sustainability of nuclear energy. Consistently with potentially viable recycling strategies, the Collaborative Project ACSEPT will provide a structured R&D framework to develop chemical separation processes compatible with fuel fabrication techniques, with a view to their future demonstration at the pilot level. A training and education programme will also be implemented to share the knowledge among communities and generations so as to maintain the nuclear expertise at the fore-front of Europe. The challenging objectives of ACSEPT will be addressed by a multi-disciplinary consortium composed of European universities, nuclear research bodies and major industrial players. This consortium will generate fundamental improvements for the future design of a potential Advanced Processing Pilot Unit.*

INTRODUCTION

Presently, the European nuclear fleet leads to the annual production of approximately 2500 t/y of spent fuel, containing 25 t of Plutonium, 3,5 t of minor actinides (MA, namely Np, Am, Cm) and 3 t of long-lived fission products (LLFPs). These MA and LLFPs stocks need to be managed in an appropriate way. Some European countries have chosen the strategy of closed fuel cycle, currently involving *(i)* the reprocessing of spent nuclear fuels to recover uranium and plutonium and the recycling of plutonium in Light Water Reactors *(ii)* the vitrification of long lived radionuclides (the final waste being expected to be disposed of in deep geological repositories). Some others have chosen the direct geological disposal of nuclear spent fuel. Both strategies are today the envisaged solutions depending on national fuel cycle options and waste management policies. Required time scale for the geological disposal exceeds our accumulated technological knowledge and this raises problems of public acceptance.

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 technological and manageable time scales.

Despite the diversity between European Member States concerning nuclear power and envisaged fuel cycle policy ranging from the once through without reprocessing to the double strata fuel cycle ending with the ADS as the ultimate burner or Gen IV fast critical reactors multi-recycling all transuranic (TRUs), P&T requires an integrated effort at the European level and even worldwide.

When considering sustainable energy development worldwide, P&T should now be seen as fully integrated in a more global approach based on advanced fuel cycles associated to fast neutron reactors. These advanced nuclear systems should play a key role for not only minimising the production of long lived radioactive waste but also for optimising the use of natural resources and for increasing resistance to proliferation.

In parallel with the development of fast neutron reactors, a separation and treatment strategy needs to be implemented, aiming at a closed fuel cycle that includes the recycling of actinides. This strategy will in particular have to comply with the worldwide policy of safeguarding and proliferation resistance that favours recycling modes with a co-management of actinides. It would also permit the transition from the currently practiced mono-recycling of Plutonium in Light Water Reactors (LWR) to actinides (U, Pu, MA) recycling in Gen IV reactors, thereby allowing minimization of radiotoxicity in the ultimate waste. Even when considering the phase out of nuclear energy, the combination of P&T and dedicated burner such as ADS technologies, but this time at regional scale, would allow meeting the objectives of minimising the long lived waste to be ultimately disposed of.

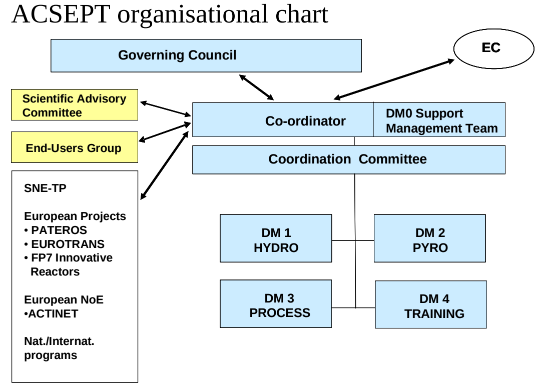
Considering this evolution towards closed fuel cycles, it has been considered as a priority to strengthen the links and synergies with Transmutation as well as with geological disposal or interim storage activities. This evolution towards more integration should thus materialize and be consistent with 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).
Despite the different national strategies envisaged for managing nuclear wastes, the options for actinide recycling implementation show a significant common trunk which allows to draw a consensual European roadmap for research and development activities as well as for future pilot-scale fuel cycle facilities.
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 actinides 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 group recovery of trans-uranium elements (TRUs) *vs.* minor actinides (MA) selective separation should also be undertaken by 2012. Priorities should also be given following the review of ADS *vs.* critical fast systems potentialities and their different coolants. All these reviews should lead to take decisions on demonstration facilities to be built at a time horizon 2015-2020.

OBJECTIVES OF ACSEPT

In line with these timescales, the ACSEPT collaborative project (2008-2012) will provide a structured R&D framework with the ambitious objective to develop chemical separation processes compatible with fuel fabrication techniques, in view of their future demonstration at the pilot level. ACSEPT will develop elementary building blocks in terms of separation processes so as to offer technical solutions to the various fuel cycle options envisaged today. Flexibility will thus be kept as a main driving force while waiting for the consolidation of these options.

ACSEPT is structured in a very clear way so as to optimize the production, dissemination and potential exploitation of the generated knowledge. Each technical issue is clearly identified to form a domain.

ACSEPT organisational chart



The challenging and ambitious objectives of ACSEPT will be addressed by a multidisciplinary consortium composed of European universities, nuclear research bodies and major industrial players.

TECHNICAL PROGRAMME

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,
- and 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

As regard to partitioning processes, many countries have developed for the past 4 decades hydrometallurgical processes to recover TRU 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 have 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 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 4*f* and 5*f* elements.

The processes developed around the world differ from the extracting systems involved in these different steps and the possibility to shrink two of these steps in a single one.

For the first step, TBP is the basis of the PUREX, UREX and COEX™ processes, developed in Europe and the US, whereas monoamides support the BAMA process developed in Japan, India and France. As these processes are well advanced in these countries, ACSEPT does not intend to put any resources on this topic.

- For the second step, malonamides, CMPO and TODGA are used in the DIAMEX, TRUEX 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. 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. 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, 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.

Current BTBP reference system developed during IP EUROPART for An(III) selective extraction does not fulfill 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 area: ligand stability vs. radiolysis, An(III) extraction/stripping kinetics and process simplification (3/2/1 steps).

In parallel, a new approach will be investigated: the selective stripping of trivalent minor actinides after their extraction by approved lipophilic ligands (e.g.: malonamides, DGA). 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 TRU actinide group separation (i*.e.:* Np, Pu, Am-Cf) from spent 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 flowsheets 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 end-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 actinide solutions (variously mixed) to polyactinide containing solids for fuel preparation (GenIV, Transmutation). 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 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. Furthermore, as coprecipitation 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 beds, from these selected technologies.

The main objectives of DM 1 will thus be to develop aqueous chemical processes in the various fields of spent 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 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. 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 processes 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 flowsheets 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 GenIV 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 "paper" studies to evaluate, 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 t0+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, 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 which 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.

CONCLUSIONS

ACSEPT will provide the sound basis and fundamental improvements for future demonstrations of fuel treatment in strong connection with fuel fabrication techniques. The timelines of this four-year R&D project (2008-2012) should allow to offer technical solutions in terms of process separation that may be reviewed by Governments, European utilities as well as Technology Providers at that time horizon. By showing a technically feasible recycling of actinides strategy, ACSEPT will certainly produce positive arguments in the sense that :

- European decision makers and more globally public opinion could be convinced that some sound technical solutions for a better management of the nuclear wastes are now technologically feasible,
- One of the identified major difficulties for nuclear energy production, i.e. to guarantee the non-dissemination of hazardous radionuclides within the bio-sphere in the far future, is on the way to be solved, thus paving the way towards sustainability.

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