GENIORS

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Exploitation plan based on the conclusions of the interactions with stakeholders/end-users

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Summary

exploitation plan

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1. SUMMARY OF EXPLOITABLE RESULTS OBTAINED

DM1

First, progress was made on fission product behaviour and in particular ruthenium. In first approximation, the ruthenium extraction properties of a given solvent are strongly related to its ability to extract water. Further experiments demonstrated that nitrite (so far neglected) is also important for this system because it strongly interacts with ruthenium complexes in both aqueous and organic phases. It was also possible to optimize conditions for effective fission product masking/ scrubbing, with the determination of conditions suitable for scrubbing fission and corrosion products such as Tc, Sr, Zr, Mo, Pd from TODGA and mTDDGA solvents. In particular, replacing TODGA by mTDDGA in EURO-GANEX flowsheet offers several advantages with respect to the routing of non-Ln fission products while maintaining excellent extraction properties for TRU and Ln: The studies showed that Distribution ratios for Sr, Zr and Mo are significantly lower compared to those determined the reference solvent. This in turn will allow to simplify the scrubbing section of a EURO-GANEX process.

Radiolytic damages on the chemical systems can reduce the process performances but also impact the safety of the processes. Both the aqueous and organic phases were studied. Although most of the aqueous phases containing stripping agents are intended as once-through reagents, their degradation could lead mass transference and accumulation into the recycled organic phases, as well as condition their resistance to radiation. For the organic phase, in the case of PTD, several studies were performed to assess its hydrolytic and radiolytic stability under the harsh conditions of the partitioning i-SANEX and GANEX processes. All results point out towards the TODGA-PTD system is able to keep the perform during An/Ln separation up to 200 kGy.

While the degradation of compounds by hydrolysis and radiolysis under normal process operating conditions has already been the subject of numerous studies, other important aspects must also be considered such us the intrinsic reactivity of pure compounds and the reactivity of organic molecules under severe conditions than those of the separating operations, The thermal stabilities studies of PTD, PTT, PT EtHex, and azide reagents, neither TODGA extractant (pure or in TPH) in contact with nitric acid did not show additional risk of spontaneous decomposition during the synthesis process or the operation. It confirms the interest and the relevance of such molecules, in particular PTD. Addressing EURO-GANEX development, an analytical method for degradation products quantification was developed and successfully tested. CyMe4-BTBP and PTD irradiations were completed. Radiolysis modelling work was completed.

The stripping solvent based PTD has been extensively studied and its applicability to the i-SANEX and EURO-GANEX processes was demonstrated. The present work contributes to a better understanding of the selective complexation mechanism involved in the i-SANEX extracting system. The selectivity of the innovative PTD-based system can arise from the formation of intermediary mixed species during the extraction procedure. The results were so promising that the system composed of PTD + AHA + TODGA and DMDOHEMA was selected as new EURO-GANEX reference system by the European scientific community of GENIORS project subject to future flowsheet tests. Distribution data were collected for PTD-based EURO-GANEX systems and for PTEH. Me-TDDGA was tested as an alternative to mTDDGA. Solvent clean-up and recycling options were reviewed.

The impact of experimental parameters on the dissolution of U-Ln solid solution samples was investigated and explained on a microscopic level. The hydrothermal synthesis of uranium oxide was optimized. Powders were prepared, and pellets were sintered and characterized.

All these results will be directly used to increase the process safety or to develop models that will feed process simulation codes.

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DM2

For reprocessing MOX fuel, It is important that the chemical system can manage high Pu loading. The reference molecule, TODGA, alone shows poor loading leading to adding a modifier such as DMDOHEMA. A new molecule, m-TDDGA is now studied, showing a relevant loading. This molecule was studied in depth in the Project. All the work done until now confirms the relevance of the reference chemical system based on PTD + (TODGA-DMDOHEMA), with a possible substitution of (TODGA-DMDOHEMA) by m-TDDGA for the EURO-GANEX flowsheet.

A brainstorming led to the definition of a procedure for comparing radiolytic studies in irradiation loops. A benchmark of irradiation loops was carried out, allowing in the future a better exploitation of irradiation experiments. The results obtained show the good capability of EURO-GANEX and CHALMEX processes and especially the radiolytic stability of main reagents has been confirmed during irradiation tests on an experimental loop.

Kinetics studies and hydrodynamics modelling will contribute to improve the process simulation codes and the development of online monitoring techniques is key to improve process performance and safety (WP5). By integrating the knowledge gain in DM1, simplification options of the chemical systems of the EURO-GANEX and EURO-EXAM processes have been proposed and are under testing (WP6). In order improve the links between reprocessing and fuel fabrication (integrated approach of the fuel cycle), studies are carried out to identify possible issues at the interface of fuel dissolution and separation, and separation and conversion/fabrication. In particular, a key issue is whether complexing agents or their degradation products (such as acetic acid) will enhance the solubility of actinide ions and reduce the efficiency of the oxalate co-precipitation process (WP7).

New work was carried out on MOX fuel dissolution. Eighteen different actinide samples were synthetized in order to be able to study dissolution in function of morphology and Pu amount. The characterization tools employed in this study were used to describe the plutonium content and the morphology of the different powders. It appears that the specific surface area decreases and the crystallite size increases while the calcination temperature increases too, in agreement with literature data. The work has to be continued.

Another important work addresses MOX fuel fabrication, with innovative techniques, without the use of powders. In this frame, investigations on the synthesis of simulated MOX fuel particles, potentially bearing MAs and blanket materials, were carried out using the sol-gel route via internal gelation.

The sol-gel route via internal gelation was applied for the production of and Ce-doped uranium dioxide microspheres. Trivalent and tetravalent Ce precursors were used and the influence of the precursors' oxidation state on the fabrication process and the final product was studied.

The successful introduction of the dopant into the $3UO_3 \cdot 2NH_3 \cdot 4H_2O$ matrix of the dried gels, independent of the dopant and the oxidation state of the precursor, was demonstrated for Ce contents up to 30 mol.%. the work has to be continued to confirm the interest of this method.

For the conversion of actinides to oxides, the presence of complexing agents in the solution to process was studied and no effect on conversion performances of actinides to oxides was observed.

DM3

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Now that reference processes have been clearly identified in previous projects, it is possible to start working on concept plant design. As part of the design process, a gPROMs model of the plant was constructed. FISPIN modelling data was used to generate a representative spent fuel inventory that acted as an input to the gPROMs model (WP8). To better integrate the results of GENIORS in the international context, a study undertook a comparative assessment of the different recycling needs and options considered, and identified the facilities that could integrate the reprocessing options at EU level. Holistic impacts of flowsheet deployment can be assessed using the Sim Plant tool to show changes to plant footprint and lifetime cost variances.

An important part of GENIORS is the system integration studies in order to position GENIORS's work in the global context. Based on three reactor's concepts (ASTRID, ALFRED and LWR) a total of six fuel cycles have been described, bearing in mind that the exact parameters of these cycles will result from further engineering studies. ASTRID and ALFRED consider the same two fuel cycles: MOX fuels with actinides recycled in the reactor and MOX fuels with actinides treated in MYRRHA. In addition, a fuel cycle of a light-water reactor and a fuel cycle coupling to MYRRHA have also been considered so as to point out the differences between fast reactors and LWRs.

At the industrial scale, it will be important to have online monitoring to ensure process performances, identify loss of performance as soon as possible, and reinforce the process safety. In this frame, electrochemical microelectrode array sensor systems, capable of electrochemical measurements rich in diagnostic information, were successfully developed. These sensors produce information enabling quantitative electrochemical analysis in the harsh environment presented by the acid systems required for EURO-GANEX liquid-liquid extraction, and which are unaffected by initial irradiation tests. The work will continue to assess the performances on relevant species.

Safety studies are part of the system integration. A safety and environmental hazards review of EURO-GANEX made prioritized recommendations for the progression of the Euro-GANEX process towards full industrialization. The recommendations also include research topics that would allow for elimination or mitigation of the hazards identified. Twenty recommendations were made to improve knowledge and investigate hazards with a view to elimination or mitigation were defined.

Utilising the Concept Design, a safety review of the EURO-GANEX process was carried out and then reviewed by the consortium partners at the Antwerp Winter Meeting in November 2018, allowing for a wide variety of experience and skills to participate. A methodology for density laws was defined for EURO-GANEX chemicals. It will allow the production of an in-depth criticality review for the process as further data is provided by other areas of the project (WP9). The Stakeholder and Clustering Events were jointly organised by CEA and SCK•CEN. Active participation (presentations, discussions) was received from all GENIORS partners and also beyond the GENIORS community or beneficiaries. Safety Studies have been concluded through the production of the major hazard and criticality review of the EURO-GANEX which has shown no significant reasons the flowsheet could not be deployed at an industrial scale and informs areas of required future development. All the collected information and methods can now be used to develop the processes at higher TRL up to the pilot scale with a high level of confidence both in terms of performance and safety.

2. EXPLOITATION PLAN

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Due to Covid situation and in particular the end of the project in the middle of the third wave did not allow the interactions with stakeholders and end-users as planned. It is an internal analysis. Today perspectives for industrial MOX recycling seemed to be postponed. However, results of GENIORS open many perspectives in spent fuel recycling process development. Below, the exploitation opportunities issued from the technical WPs 1-9 are presented.

WP1: the new acquired data will feed process simulation codes in order to better design the scrubbing part of the process flowsheets. Also, these data will be use to define recovery processes of potentially valuable elements such as ruthenium, to be developed in the proposal under building on the topic EURATOM 2021-NRT-01.03

WP2: all the results on degradation studies can be used for safety case studies (impact of gaseous fission products, impact on fissile material retention…), for improving the stability of molecules and for defining/optimising the washing part of the process flowsheets.

WP3: distribution data are of paramount importance for defining extraction models and for feeding simulation process codes. In particular, CEA, NNL and KIT who are developing simulation codes will use them. In addition, some results have been used to define the work to be done in PATRICIA on an americium extraction process.

WP4: a better understanding of solid/liquid interface will allow an optimisation of the dissolution processes, in particular by optimising/limiting the use of highly reactive reagents as well as the operating temperature. At the conversion, it helps at understanding the microstructure formation mechanisms. This could improve the performance of the fuel. This work will be useful in the NRT01-03 proposal.

WP5/WP6: all the work done in these 2 WPs allow the optimisation of process flow sheets, relying on improved thermodynamics and kinetics data. Relevant pilot experiments can be proposed. An additional work performed in the frame of an I-NERI project together with US national labs (INL, ORNL) allowed the benchmarking of irradiation loops. Now irradiation results can be compared with a higher degree of confidence.

WP7: following the work done on conversion, we are now confident on the low impact of impurity coming from stripping agent in the aqueous phase on the quality of the actinide materials. Processes were proposed to reduce the carbon content and they are efficient.

WP8: methods and tools were developed to make the best use of experimental results for upscaling a process to a workshop or a plant thanks to the Simplant tool. With such pictures, a technical-economic estimation of a plant is possible.

WP9: no showstoppers have been identified in terms of safety on the reference separation processes. The development of pilot facilities can be done with more confidence. A clear methodology was also tested to support further developments.

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