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Electrochemical separation of actinides from molten LiCl-KCl on solid Al cathodes

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To comply with the sustainability goals defined for innovative reactor systems, a waste minimization through recycling of all actinides is required and can be realised using a so-called Partitioning and Transmutation (P&T) concept inherent to the reactor systems. It is obvious that the corresponding fuel cycles will play a central role because an efficient and selective recovery of the key elements is mandatory.

One of the major goals is to develop and implement clean waste technologies using a global actinide management. For this purpose, advanced fuel cycle processes with full actinide fuel technologies and ultimate waste forms with a minimal content in actinides (< 0.1%) are required. This necessitates that Am and Cm can be selectively separated from lanthanide fission products, certainly the most difficult and challenging task in advanced reprocessing of spent nuclear fuel due to the very similar chemical behaviour of these elements.

The separation can be derived from aqueous or pyrochemical partitioning processes. Both have advantages and disadvantages and should be applied in a complementary way. In Europe, significant progress was made for both routes in international collaborations in the frame of the European research projects PARTNEW, PYROREP, EUROPART and ACSEPT.

Pyrochemical processes rely on refining techniques in high temperature (around 500°C) molten salt (e.g. LiCl/KCl eutectic). In nuclear technology, these processes are mostly based on electrorefining or on extraction from the molten salt phase into liquid metal. In the electrorefining process the fuel is anodically dissolved, releasing actinides and lanthanides mainly as MCl_3 species into the molten salt electrolyte. The separation is based on electrodeposition of the actinides on a cathode leaving the fission products in the bulk. The choice of cathode material onto which the actinides are deposited during electrolysis is essential. Molten Cd and Bi have been widely used because they are reactive electrodes, i.e. they form stable alloys with the actinides. This alloying inhibits the re-dissolution of the electrodeposited trivalent actinides by a comproportionation reaction with not deposited An(III) in the salt melt and thus allows an efficient recovery of An metal. The redox potentials on solid cathodes show a difference in the reduction potential between actinides and lanthanides which is about 3 times higher than for liquid cathodes, improving obviously a separation of these two element groups. Solid Al cathodes unite both advantageous properties, i.e. the large difference in the electrodeposition potential and the reactivity to form alloys. Therefore we use Al electrodes for our electrochemical partitioning studies.

In view of a grouped actinide recycling, foreseen in the fuel cycles of new generation reactors, electrorefining experiments using a fuel with the overall composition $U_{61}Pu_{22}Zr_{10}Am_2Ln_{5}$ were carried out. The metallic alloy was anodically dissolved in a LiCl-KCl eutectic and the actinides were co-deposited on Al cathodes as alloys, leaving more than 98% of the lanthanides in the salt phase. The results of a multiple-run electrorefining experiment without changing the salt bath, are very promising in view of a large-scale development of pyroreprocessing in advanced nuclear fuel cycles. They demonstrate that a grouped actinide recovery better than 99.9% are possible and therefore represent an important step towards achieving the sustainability goals of future reactor systems.

Primary author: Dr NOURRY, C. (European Commission, JRC, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany)

Co-authors: Dr GLATZ, J.-P. (European Commission, JRC, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany); Dr SOUČEK, P. (European Commission, JRC, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany); Dr MALMBECK, R. (European Commission, JRC, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany)

Presenter: Dr GLATZ, J.-P. (European Commission, JRC, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany)

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