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Pyrochemical Separation of Lanthanide and Actinide Metals[†]

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A highly efficient group separation of lanthanide and actinide metals can be effected by molten salt extraction of the lanthanide fraction(s) from a solution of actinide and lanthanide metals dissolved in molten copper–magnesium alloy. The extraction salt used for this separation is 50 m/o $MgCl_2$, 30 m/o NaCl and 20 m/o KCl, and the composition of the solvent metal alloy is approximately 20 to 30 mass-percent magnesium in copper. At 800 °C, the distribution coefficient (K_d) for +3 lanthanide chlorides varies from 150 to 70 while the distribution coefficient for the +3 actinides varies from 4.5 to 6.0. At 20 mass percent magnesium, the ratio of K_d is about 30 between Cerium (150) and Americium (4.5). With these large differences in K_d, a simple batch extraction can be used to separate Cerium from Americium. The lanthanide elements are extracted into the molten salt phase as the +3 chloride salt and the equivalent mass of magnesium metal is deposited in the immiscible metal alloy phase. After separation of phases which can be performed mechanically after cooling, or by decantation at elevated temperature, the pure lanthanide metal can be recovered from the salt phase by reduction with calcium metal at 800 °C. The lanthanide metal is formed as a high-density-powder bottom-phase, which can be converted to a molten liquid by the addition of zinc. After retorting, the pure lanthanide metals are recovered as metallic sponge.

[†] This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

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Actinide Partitioning by HDEHP Solvent Extraction. A Verification of the Process Scheme by Countercurrent Experiments in Hot Cells

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Three HAW partitioning processes have been selected and investigated on a laboratory scale at the JRC-Ispra Establishment (CEC). The purpose of these processes was to demonstrate the chemical feasibility of the actinide separation from HAW in order to reduce the potential long term hazard of vitrified HAW.

Two of these processes (HDEHP and TBP processes) are based on liquid–liquid extraction of actinides from original and concentrated HAW solutions, respectively.

To verify on a larger fully active scale these two process flow sheets, a fruitful collaboration was established between the JRC (Ispra Establishment) and the CEA (CEN-Fontenay aux Roses, France).

For this purpose, on the basis of indications resulting from the previous laboratory investigations at the JRC, continuous countercurrent experiments with mixer–settler batteries were performed in the hot cells of CENFAR laboratories. As a feed solution a fully active HAW raffinate prepared by treatment of high burnt UO_2 fuel specimens (33 GWd/tU) was used.

The present paper deals with the verification of option 1 of the HDEHP process flow sheet and illustrates related experimental results. By means of M–S batteries continuous countercurrent operating conditions were thus applied to the most relevant steps of the HDEHP 1 process scheme, namely:

actinide (U, Pu, Np, Am, Cm) extraction from HAW;

trivalent actinides (Am, Cm) back-extraction from loaded HDEHP, with the aim of verifying the performances and hydraulic behaviour of these two specific process steps under dynamic conditions.

For the remaining process steps (RE and Pu back-extractions, exhausted solvent clean-up) experiments were carried out batch wise.

The reduction of the HAW acidity (denitration) was successfully operated on fully active and rather large (42 litres of HAW) scale according to the experimental conditions indicated by additional exploratory experiments, performed at Ispra.