

## Background

P&T (Partitioning and Transmutation) could allow optimizing the future nuclear fuel cycle. Worldwide there is an increasing interest for advanced spent fuel reprocessing techniques whereby not only U and Pu but in addition the minor actinides Np, Am and Cm and preferably also the long-lived fission products are separated from the to-be-disposed waste. In accelerator driven systems and/or fast reactors the recovered elements could then be transmuted into short-lived or stable fission products. SCK·CEN is testing such an advanced aqueous reprocessing technique on contractual order from IRI (Japanese Institute of Research and Innovation). Their ERIX (Electro Reduction and Ion eXchange) process for MOX (Mixed OXide) fuel from fast breeder reactors consists of anion exchange as main separation method, electrolytic reduction for reducing U(VI) to U(IV) and extraction chromatography for the isolation of the minor actinides Am and Cm.

## Objectives

In a sufficiently concentrated nitric acid solution, U, Pu and Np form anionic nitrate complexes that are adsorbed on an anion exchanger, while Am, Cm and most FP (Fission Products) exist as cations that do nearly not interact. In the ERIX process further partitioning of Am and Cm from the FP is based on their different affinity for the specific chelating agents of the extraction resin.

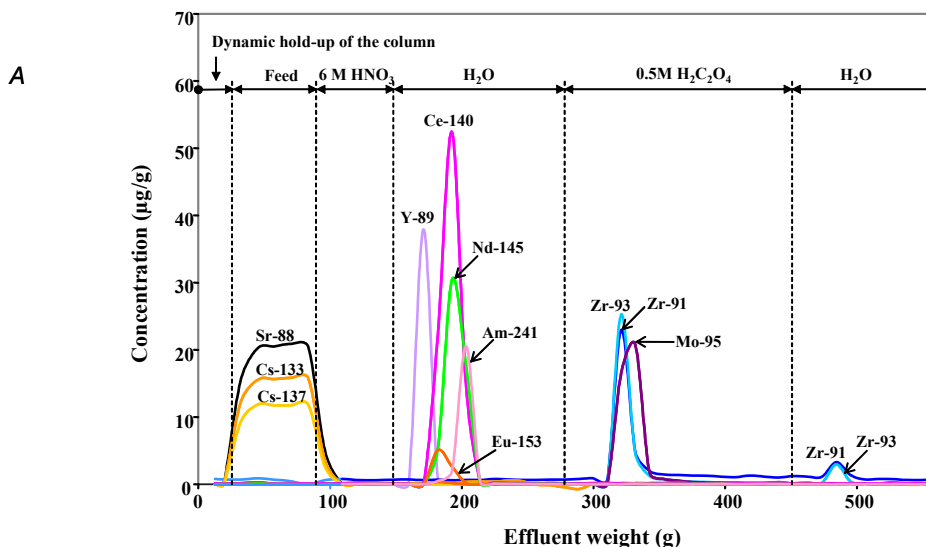
The first step in the partitioning of the waste is the separation of Am and Cm together with the chemically very resembling Ln (Lanthanides) from the FP by extraction chromatography on a CMPO/SiO<sub>2</sub>-P column. Simultaneously, Zr and Mo are also isolated. IRI prepared a novel silica-based extraction resin by impregnating CMPO into a styrene-divinylbenzene copolymer, which was immobilized in 50 µm porous silica particles. Compared to conventional polymeric matrix resins, these new type of silica-based extraction resins have rapid adsorption kinetics and a significantly low pressure drop in a packed column.

After preliminary evaluation of the separation in "cold" conditions, our objective was to examine the separation behaviour of Am and fission products with a real HLW (High-Level Waste) solution.

## Principal results

We studied the partitioning of a U, Pu & Np free HLW solution on CMPO/SiO<sub>2</sub>-P as stationary phase with nitric acid as mobile phase. We noticed that the multi element solution, under the described circumstances, is separated in three different groups of elements as illustrated in the figure below.

The (radio)nuclides in the first group (<sup>133</sup>Cs, <sup>134</sup>Cs & <sup>137</sup>Cs, <sup>88</sup>Sr) show no interaction with the stationary phase and migrate with the mobile phase during the loading process. The majority of both tracers are already detected in the effluent at the end of the loading procedure. Both elements are recuperated quantitatively during the cleaning process with 6 M HNO<sub>3</sub>.



Column packed with CMPO/SiO<sub>2</sub>-P allowed effective separation of MA and Ln from heat-emitting Cs & Sr from Zr & Mo.

Am and the Ln, i.e. Y, Ce, Nd and Eu were completely adsorbed by the CMPO/SiO<sub>2</sub>-P extraction resin and the adsorbed Am and Ln were eluted efficiently by water as mobile phase. The trivalent metal ions such as Am(III) and Ln(III) are adsorbed by CMPO extraction resin as neutral nitrato-complexes. Therefore, the elution effect of Am and Ln is considered to result from the decomposition of the complexes with the decrease of NO<sub>3</sub><sup>-</sup> concentration in the resin bed by supplying water to the column. Note that the Am and Ln showed sharp elution peaks with almost no tailing. This indicates that the silica-based CMPO extraction resin has rapid kinetics.

On the other hand, the Zr and Mo contained in the HLW solution were strongly retained by the CMPO/SiO<sub>2</sub>-P. The adsorbed Mo and most of the Zr were eluted effectively by 0.5 M H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>. A small portion of Zr is finally eluted by water. Zr and Mo are known to form complexes with oxalic acid and the complexes are not adsorbed by the CMPO extraction resin. All of the detected nuclides were quantitatively recovered from the extraction resin.

The results of this hot-cell test demonstrate that successful separation and recovery of Am together with some Ln from HLW can be achieved. Moreover, one has noticed in a previous study that CMPO/SiO<sub>2</sub>-P has relatively excellent resistance against radiation in strong nitric acid medium.

These indicate that the proposed MA-Ln separation process is essentially feasible, though further investigations such as pilot-scale testing and evaluation of the process regarding practical use conditions are necessary.

### Future work

Future work shall focus on the next step in the process: the separation of Am and Cm from the Ln in view of the production of targets for transmutation.

### Main contact persons

Aimé Bruggeman, [aimé.bruggeman@sckcen.be](mailto:aimé.bruggeman@sckcen.be), Patrick Goethals, [patrick.goethals@sckcen.be](mailto:patrick.goethals@sckcen.be)

### Main reference

P. Goethals, L. Vos, D. Penneman, A. Bruggeman, Y. Wei, H. Hoshi, M. Kumagai, "Chromatographic separation of actinides and lanthanides from fission products on CMPO/SiO<sub>2</sub>-P extraction resin.", International Conference on Nuclear and Radiochemistry, 29 August to 3 September 2004, Aachen, Germany.

Y. Wei, H. Hoshi, M. Kumagai, P. Goethals and A. Bruggeman, "A hot test on minor actinides separation from high-level waste by CMPO/SiO<sub>2</sub> extraction resin.", Actinides 2005, Manchester, UK, July 4 – 8, 2005 (Proceedings to be published as a book "Recent Advances in Actinide Science" by the Royal Society of Chemistry).