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Recycling americium from spent fuels is an important option considered for the future nuclear fuel cycle as americium is the main contributor to the long-term radiotoxicity and heat power of final waste. Removing Am would allow to significantly reduce the surface necessary for a geological repository of vitrified waste. In this context, the liquid-liquid extraction process "Euro-EXAm" is under development, to allow the recovery of Am alone from a PUREX raffinate (a nitric acid solution of spent fuel dissolution liquor already cleared from U, Np and Pu). The challenge here is to recover americium (III) with high selectivity towards curium and lanthanide (Ln) trivalent cations which have very similar physico-chemical properties. In this new separation system, two main steps would be necessary (Figure 1):

- first all lanthanide cations are co-extracted together with americium and curium in the organic phase, using a solvent containing 0.2 mol/L of TODGA (*N,N,N',N'*-tetraoctyldiglycolamide*)* extractant in TPH (Hydrogenated TetraPropylene) with 5%$_{vol.}$ octanol,
- then americium is stripped in a second step with selectivity towards curium and lanthanides. The water soluble ligand TPAEN (*N,N,N',N'*-tetrakis[(6-carboxypyridin-2-yl)methyl]-ethylenediamine) was tested to selectively strip Am from a loaded organic phase. Used in combination with TODGA in the organic phase, TPAEN shows a quite high Cm/Am selectivity ($SF_{Cm/Am}$ = 4 at 0.1M HNO_3), which allows to selectively strip Am while Cm and Ln remain extracted in the organic phase.

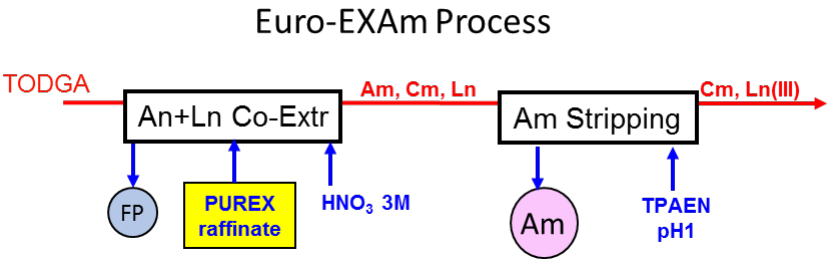

Figure 1: Simplified scheme of the Euro-EXAm process (FP = fission products, An = Am and Cm, Ln = lanthanides)

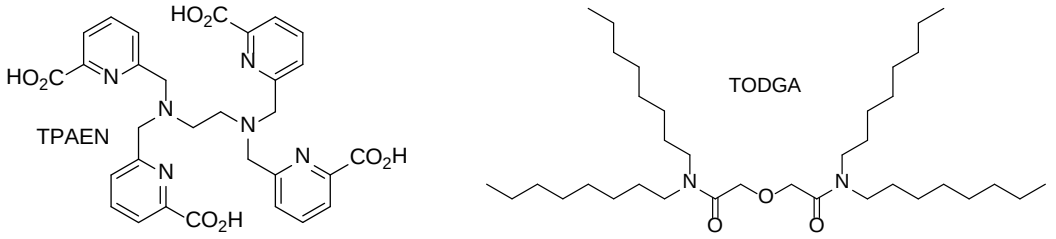

Figure 2: Structure of ligands used in the Euro-EXAm process

Batch extraction data were acquired to evaluate the best conditions to develop a liquid-liquid separation flowsheet with this promising TODGA/TPAEN separation system. It was demonstrated that TPAEN has a strong complexing capacity allowing the back extraction of Am in macro concentration (1 – 2 mM) even with low ligand concentrations (around 1.5 equivalent of ligand is enough). TPAEN has a higher affinity for light lanthanides and the selectivity between La and Am might become low depending on experimental conditions. The influence of parameters such as temperature, pH, ligand and cations concentrations was studied. Kinetics experiments in batch contactors were also performed to characterize times necessary to reach equilibrium. This set of experimental work data allowed the elaboration of a thermodynamical model which was implemented in the PAREX simulation code in order to propose a flowsheet. The feasibility of this process will be evaluated during spiked tests in centrifugal contactors starting from a surrogate feed solution spiked with traces amounts of Am and Cm. A final hot test will finally be performed at ITU from a genuine PUREX raffinate.

This work is the result of collaborations in the framework of the SACSESS European Project.

Keywords: americium, curium, liquid-liquid extraction, TPAEN

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