NEW MOLECULES FOR AN(III)/LN(III) SEPARATION BY LIQUID LIQUID EXTRACTION

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Abstract

The separation of trivalent actinides, americium and curium, from actinides and lanthanides mixtures is an important step in an advanced partitioning process for wastes issued from the reprocessing of spent nuclear fuel. Our goal is to allow the An(III) selective extraction from concentrated nitric acid solutions (up to 1 mol/l), using if possible potentially incinerable molecules, containing only C, H, O and N.

We present here a new family of An(III) selective extractants, the 2,6-bis-(5,6-di-n-alkyl-1,2,4-triazin-3-yl)-pyridine named thereafter BTP (Bis Triazin Pyridin). These molecules were designed by Z. KOLARIK at the KFZ, member of the NEWPART contract, and seem able to fulfil this goal.

These molecules exhibit very strong extracting properties, allowing Am(III) to be extracted from nitric solutions ($D_{Am(III)} > 2$ for $[HNO_3] = 1$ mol/L), with a very good separation factor over Eu(III) ($FS_{Am/Eu} > 100$).

The first studies conducted on this family of molecules show that:

- The kinetics of the extraction is rather slow (time to reach equilibrium is around 1 hour, in batch test tube conditions), and depends on the nature of the alkyl substituents (equilibrium is faster for *n*-Propyl BTP than for *n*-Butyl-BTP). The kinetics do not seem to be affected by the concentration of the extractant, the concentration of nitric acid or nitrate ions in a wide range.

- The extraction and separation performances are influenced by the nature of the diluent. A mixture of non polar diluent (hydrogenated tetrapropene) and polar one (*n*-octanol), in a volume ratio (80/20) is needed to solubilise the BTP, and ensure the best performances. Dissociating diluent (Nitrophenyl-Octylether, Nitrobenzene) gives good extraction, thus suggesting a solvatation extraction mechanism, but lower separation factor. Aromatic diluent such as Tertiary-butyl-benzene gives poor extraction and separation performances, but a fast kinetic.
- The BTP appears to have a very low basicity, compare to other nitrogen bearing ligands. A relative scale of pKa was established, in methanol/water media, and shows that the pKa of BTP was lower from 1 to 2 units than the ones of Terpyridine or tripyridiltriazine (TPTZ). This property can explain the extraction performances at high nitric acid concentration, the competition H^+ / M(III) complexation shifting towards the M(III) side.

These results show that it should now be possible to design a process for the selective extraction of An(III) over Ln(III) from high-level liquid wastes, using only disposable molecules and diluent. Such a process will be tested in hot conditions at the Transuranium Elements Institute (TUI, NEWPART member), on a PUREX / DIAMEX raffinate very soon.