SYNERGISTIC SYSTEMS USING POLYPYRIDINES AND CARBOXYLIC ACIDS FOR THE AN(III) / LN(III) SEPARATION

P.Y. Cordier, C. Madic

CEA-Valrhô DCC/DRDD/SEMP, Marcoule BP 171 30207 Bagnols-sur-Cèze France

J.O. Liljenzin

Department of Chemistry University of Reading Box 224, Whiteknights Berkshire RG6 2AD UK

P. B. Iveson, M. J. Hudson

Department of Nuclear Chemistry Chalmers University of Technology 41296 Göteborg Sweden

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. To achieve this separation, synergistic systems were tested, with 2-bromodecanoic as a cationic exchanger and different nitrogen bearing ligands.

The groups of ligands tested are:

- 2,2':6',2"-terpyridine (Tpy) and alkylated derivatives.
- 2,4,6-tri-(pyrid-2-yl)-1,3,5-triazine (TPTZ) and alkylated derivatives.
- Alkylated Tetra and Penta pyridine.
- 2-amino-4,6-di-(pyrid-4-yl)-1,3,5 triazine (ADPTZ) and alkylated derivatives.

All these ligands were tested in synergistic systems with 2-bromo-decanoic acid. The main results we can draw about these experiments are:

- The basicity of the molecule is a key point for the extraction performances at low pH of the aqueous phase. Only molecules with a low pKa (TPTZ) can extract M(III) from such a media.
- The number of nitrogen directly in contact with the metal has a positive effect on the separation factor SF_{Am/Eu}, and sometimes on the distribution ratio D_{M(III)}. Tetra and Penta Pyridine offers SF_{Am/Eu} around 20, while Tpy is around 10.
- The electronic configuration of the binding site appears to be the most important factor for the extraction. The change in position and number of nitrogens in the heteroaromatic cycle leads to very important modifications of the properties ($D_{M(III)}$ and $SF_{Am/Eu}$).
- The lipophilic character of the molecule has a secondary effect on the extraction, less important than the electronic property of the binding site. In some case (TPTZ) the alkylation of the basic molecules leads to en enhancement in the extraction properties. In some other case (Tpy), this leads to a decrease of the M(III) distribution ratio. This phenomenon can be due to steric hindrance, or to a small modification of the electronic configuration of the binding site.

The results gathered by this comparative study lead to a better knowledge of the properties required to design a selective extracting system from high acidity aqueous phase: the ligand should have a low basicity, a tri or quaterdendate binding site, and as many nitrogen atoms as possible inside the aromatic rings.