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Insights in the behavior of an emerging lipophilic PyTri ligand for SANEX-like processes

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Continuous efforts in research and development are resulting in new spent nuclear fuel (SNF) management strategies and technologies, that might be the pathway towards the sustainability and social acceptance of nuclear energy. Multi recycling of actinides is one of these paths, reducing the long-term heat load and radiotoxicity of SNF. In the last decades, many solvent extraction systems have been proposed for the hydrometallurgical separation of actinides. Owing to the outstanding properties of the hydrophilic counterparts, the lipophilic PyTri Ethyl-Hexyl (PTEH) ligand has been recently developed for the selective extraction of Am(III) and Cm(III) in SANEX (Selective ActiNide EXtraction) like processes [1, 2]. Endowed with a satisfactory solubility in kerosene/1-octanol mixtures, PTEH exhibits a pronounced selectivity (SFAm/Eu ~ 100, SFAm/Ln ~ 60-340) and an extraction kinetics faster than the reference compound CyMe4-BTBP for SANEX-like processes. The extracting performance of the PTEH-based organic solvent was fully studied, also in presence of macro-concentrations of stable lanthanides (about 2.4 g/L) and in the temperature range of interest (20-50 °C) for the industrial application. In addition, qualitative and quantitative investigations on PTEH solutions irradiated up to 300 kGy by means of a ⁶⁰Co source or aged for several weeks demonstrated the remarkable radiolytic stability of the organic solvent. Furthermore, being crucial for the development of such class of ligands, the complexation and extraction mechanisms were investigated by different techniques. The first insight into the metal/ligand stoichiometry in the species formed upon complexation with PTEH was obtained by electrospray ionization mass spectrometry. Then, ligand stability constants for Cm(III) and Eu(III) were derived by Time Resolved Laser Fluorescence Spectroscopy, confirming the observed An(III)/Ln(III) selectivity. Finally, relevant insights into the binding of Lu(III) and Am(III) ions with PTEH were obtained by NMR investigations, indicative of a different bond nature. The achieved results highlight the noteworthy properties of PTEH-based solvents, confirming their potential to be used in SANEX-like processes and enable to acquire relevant information for further developing PyTri ligands.

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[1] E. Macerata et al., J. Am. Chem Soc., 138, 7232-7235 (2016).

[2] A. Ossola et al., J Radioanal Nucl Chem., 318, 2013-2022 (2018).

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