

Abstract/Inhaltsangabe

The recycling of minor actinides (Am, Cm) from spent nuclear fuel is the goal of the “Partitioning and Transmutation”-strategy (P&T). However high neutron dose rates and heat load due to short lived Cm Isotopes are some of the main problems while handling minor actinides as nuclear fuels. Therefore various processes for the separation of Am(III) from Cm(III), especially from PUREX/COEX raffinates, have been developed and tested.

Even though some processes like the French EXAm process have been developed and tested successfully there are drawbacks like a limited pH range (pH = 2-3) and the requirement of ligands like HEDHP to prevent Ln(III) stripping at low acidic strength.¹ To address these problems we developed a new water soluble BTBP-ligand (SO₃-Ph-BTBP) for the separation of Am(III) from Cm(III) which shows a slight preference for Am(III) over Cm(III).² When an aqueous phase containing SO₃-Ph-BTBP is brought in contact with a TODGA organic phase loaded with Am(III), Cm(III) and Ln(III) the slight preference of SO₃-Ph-BTBP for Am(III) over Cm(III) is multiplied with the inverse selectivity of TODGA yielding a separation factor $SF_{\text{Cm(III)/Am(III)}} \approx 2.5$. In contrast to the EXAm process no buffer or additional ligands are needed for the Am(III)/Cm(III) separation.

The selectivity of the SO₃-Ph-BTBP/TODGA system was studied in liquid-liquid extraction experiments and distribution ratios of Am(III), Cm(III) and Eu(III) were determined as a function of nitric acid and SO₃-Ph-BTBP concentration. The extraction experiments showed that the complexation of An(III)/Ln(III) is not affected by slow kinetics. To investigate the molecular reason for the high selectivity of SO₃-Ph-BTBP for An(III) over Ln(III) TRLFS studies were carried out. Conditional complex formation constants were determined at pH = 3 and in 0.5 M HNO₃ yielding values for Cm(III) ($\log\beta_{\text{pH}=3} = 10.4$, $\log\beta_{0.5 \text{ M HNO}_3} = 7.3$) that are two orders of magnitude higher than for Eu(III) ($\log\beta_{\text{pH}=3} = 8.4$, $\log\beta_{0.5 \text{ M HNO}_3} = 5.4$). These results are in excellent agreement with the separation factor determined in the extraction experiments.

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1 Bollesteros, M.-J. et al., *Procedia Chem.* **2012**, 7, 178-183.

2 Müllich, U. et al., EP2377861, 2011-10-19.