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SACSESS Safety of actinide separation processes

Water soluble BTBP ligand – a highly efficient ligand for the separation of Am(III) and Cm(III)

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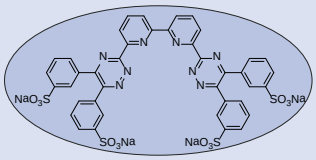
Introduction

- Pu and Am are mainly responsible for long-term heat load and radiotoxicity of nuclear waste, whereas Cm has no significant contribution.
- Selective separation of Pu and Am from nuclear waste is an important part of the partitioning and transmutation (P&T) strategy.
- Separating only Am from PUREX raffinates is desirable as the neutron dose rates & heat load of the short lived Cm isotopes complicates fuel fabrication.
- The ionic radii of Am(III) and Cm(III) differ by only 1 pm, so separating Am(III) and Cm(III) is extremely difficult.[1]
- Processes developed so far need pH 2-3, buffer, and auxiliary ligands.[2]

Why this system?

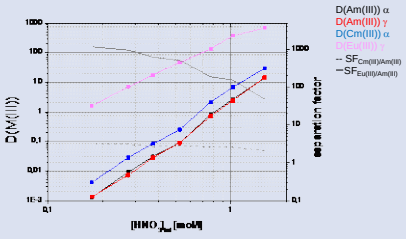
- BTBP ligands show a slight preference for Am(III) over Cm(III), Diglycolamide ligands like TODGA show inverse selectivity
  → separation factors multiply.
- System does not require buffer.



2,6-Bis(5,6-(di(sulfophenyl)-1,2,4-triazin-3-yl)-1,4-bipyridine **(SO_3-Ph-BTBP)**.

Extraction Experiments

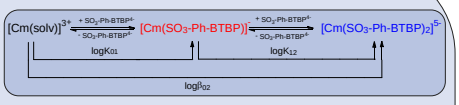
- Cm(III) and Ln(III) are extracted into the organic phase by TODGA
- Am(III) is stripped into the aqueous phase by SO_3-Ph-BTBP
- Obtained separation factors in 0.5 M nitric acid:
  $SF_{Cm(III)/Am(III)}$ = 2.6
  $SF_{Eu(III)/Am(III)}$ = 515



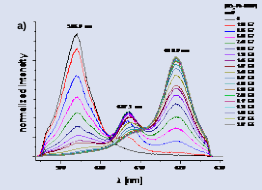
Organic phase 0.2 M TODGA + 5% vol. 1-octanol in TPH
Aqueous phase An(III) + Ln(III) in HNO_3 with 20 mmol/l SO_3-Ph-BTBP

TRLFS Experiments

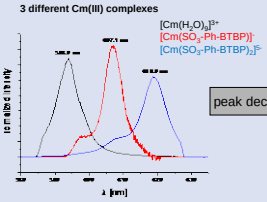
TRLFS was used to study the complexation of Cm(III) by SO_3-Ph-BTBP.
Conditional complexation constants were obtained in different media for the reaction:



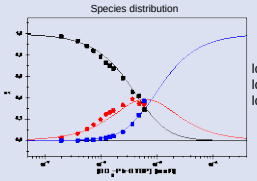
Fluorescence spectra upon increasing SO_3-Ph-BTBP concentration in a) 10^{-3} M $HClO_4$, b) 0.5 M HNO_3



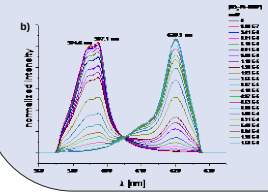
3 different Cm(III) complexes



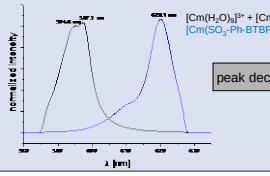
peak deconvolution



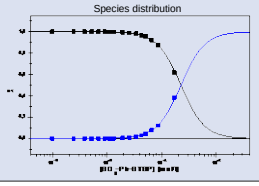
$logK_{01}$ = 5.3
$logK_{12}$ = 5.1
$log\beta_{02}$ = 10.4



2 different Cm(III) complexes



peak deconvolution



$log\beta_{02}$ = 7.3

Conclusion

- The SO_3-Ph-BTBP/TODGA system shows good performance for the separation of Am(III) from Cm(III) + Ln(III) under various conditions.
- $SF_{Cm(III)/Am(III)}$ = 3.2 - 2, $SF_{Eu(III)/Am(III)}$ = 1200 - 50 in 0.1 M - 1.5 M nitric acid.
- No buffer or auxiliary ligands required.
- TRLFS experiments show formation of 1:1 and 1:2 complexes in 10^{-3} M $HClO_4$, in 0.5 M HNO_3 only the 1:2 complex is formed.
- $log\beta_{02}$ value in 10^{-3} M $HClO_4$ = 10.4, $log\beta_{02}$ in 0.5 M HNO_3 = 7.3 which is 3.1 orders of magnitude lower than in 10^{-3} M $HClO_4$ → large effect of medium (pH, ionic strength, anion) on speciation and $log\beta_{02}$ value.
- $[Cm(SO_3\text{-Ph-BTBP})_2]^{5-}$ complex observed in aqueous phase of extraction experiments.

Supported by the European Atomic Energy Community Seventh Framework under grand agreement No 323282

KIT – University of the State of Baden-Wuerttemberg and
National Research Center of the Helmholtz Association

[1] G. T. Seaborg, *Radiochim. Acta*, **1993**, *61*, 115-122.
[2] Bollesteros, M.-J. et al., *Procedia Chem.* **2012**, *7*, 178-183.

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