

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

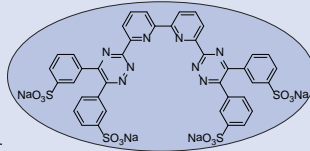
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Introduction

- 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.

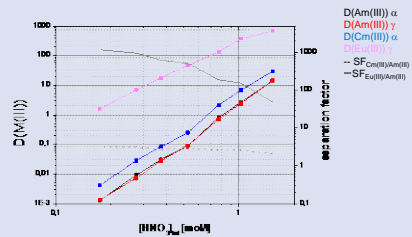


Extraction Experiments

- Cm(III) and Ln(III) are extracted into the organic phase by TODGA
- Am(III) is stripped into the aqueous phase by SO₃-Ph-BTBP
- Obtained separation factors in 0.5 M nitric acid:

$$SF_{Cm(III)/Am(III)} = 2.6$$

$$SF_{Eu(III)/Am(III)} = 515$$

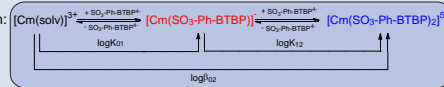


Organic phase 0.2 M TODGA + 5% vol. 1-octanol in TPH
 Aqueous phase Am(III) + Ln(III) in HNO₃ with 20 mmol/l SO₃-Ph-BTBP

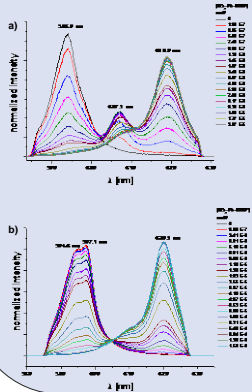
TRLFS Experiments

TRLFS was used to study the complexation of Cm(III) by SO₃-Ph-BTBP.

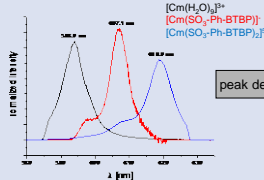
Conditional complexation constants were obtained in different media for the reaction:



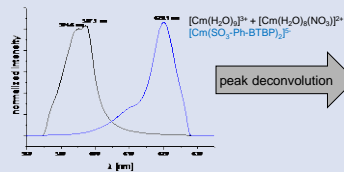
Fluorescence spectra upon increasing SO₃-Ph-BTBP concentration in a) 10⁻³ M HClO₄, b) 0.5 M HNO₃



3 different Cm(III) complexes



2 different Cm(III) complexes



Conclusion

- The SO₃-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 HClO₄, in 0.5 M HNO₃ only the 1:2 complex is formed.
- logβ₀₂ value in 10⁻³ M HClO₄ = 10.4, logβ₀₂ in 0.5 M HNO₃ = 7.3 which is 3.1 orders of magnitude lower than in 10⁻³ M HClO₄ → large effect of medium (pH, ionic strength, anion) on speciation and logβ₀₂ value.
- [Cm(SO₃-Ph-BTBP)₂]²⁻ complex observed in aqueous phase of extraction experiments.

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