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Screening of modified diglycolamide diastereomers to increase Am(III) selectivity during the partitioning of PUREX raffinate solution

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Multiple strategies have been developed to separate lanthanides (Ln) and actinides (An) from Spent Nuclear Fuel (SNF) and PUREX raffinate through solvent extraction. Current research is focusing on the development of new ligands to separate Am(III) from Cm(III) and other fission Ln(III). Diglycolamides (DGA) such as N,N,N',N'-tetra-n-octyl diglycolamide (TODGA) are promising extractants for this task. Recently, methyl substitutions in the backbone of TODGA with different orientation were studied, and interesting effects on the selectivity for Am(III) over Cm(III) and other Ln(III) were observed.^[1] The orientation on the alkyl groups has shown different behavior reflected on the affinity for Am(III) and efficiency of the system. Here, we study the selectivity and affinity of two related extractants (TODGA and N,N,N',N'-tetra-n-decyl diglycolamide, TD-DGA) with dipropyl and ethyl-methyl substitutions in the backbone, and their respective diastereomers (syn or anti 0.1 M in n-dodecane). An inverse selectivity is observed for Am(III) over Cm(III) and other Ln(III) at high nitric acid concentration (7 - 10 M). The steric hindrance gives low distribution ratios D < 1.5 for trivalent metal ions. Pu (IV) was also tested for the interest in the complexation behavior with tetravalent ions. Pu(IV) shows high distribution values at the same HNO-sub>3</sub> concentration (D > 10). The separation factors are inversely proportional to the HNO₃ concentration. The syn-ethyl-methyl substitutions for both analogues showed the highest D values for Am(III) among the other ligands, giving a promising molecule for further studies. The results of the extraction studies will be presented and discussed in the poster.

 Wilden, A.; Kowalski, P. M.; Klaß, L.; Kraus, B.; Kreft, F.; Modolo, G.; Li, Y.; Rothe, J.; Dardenne, K.; Geist, A.; Leoncini, A.; Huskens, J.; Verboom, W. Unprecedented Inversion of Selectivity and Extraordinary Difference in the Complexation of Trivalent f-Elements by Diastereomers of a Methylated Diglycolamide. *Chem. Eur. J.* 2019, 25, 21, 5507-5513. DOI:10.1002/chem.201806161

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