A $N,N'$-di-$n$-OKTYL-TODGA-dicarbollide Extractant for Actinide(III) Separation from Acidic Waste Solutions

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Abstract

Chemical partitioning of minor actinides from other fission products, in particular from lanthanides, is many-faceted challenge for existing nuclear fuel treatments. In recent years, several HLLW partitioning processes such as TRUEX, TRPO, DIDPA, DIAMEX, TODGA/TBP utilizing variety of extractants have been developed for actinide separations. However, none of them provided any substantial actinide(III)/lanthanide(III) selectivity. Processes such as SANEX or ALINA have been proposed for subsequent selective separation of actinides(III) from lanthanides(III).

Recently, the extractants based on two cobalt bis(dicarbollide) (1) ions covalently bound to organic, diglycolyl acid platform (with different alkyl/aryl substituents on the amide nitrogen) by amidic bonds, have been developed for use in nuclear treatment. In this work, the extractant $N,N'$-di-$n$-oktyl-TODGA-COSAN of the formula $\left\{\left(\left(N,N'-\left(\left(8-(OCH_2CH_2)\right)_{1,2}-C_2B_{11}H_{10}\right)_{1,2'}-C_2B_{11}H_{10}\right)_{3,3'}-Co\right)(N,N'-n-C_{18}H_{37})NCOCH_2\right\}_2O\}_2Na_2$ was evaluated for actinides(III) separation from acidic aqueous waste solutions. The extractant provided sufficient solubility in an ecological low polar solvent mixture of hexyl methyl ketone and $n$-dodecane and exhibited good hydrolytical stability. The $N,N'$-di-$n$-oktyl-TODGA-COSAN enabled good separation of actinides(III) and lanthanides(III) from bulk of the fission products present in the model PUREX feed. The co-extraction of some undesirable elements as Zr, Mo, Pd was suppressed by using oxalic acid and HEDTA in the feed solution. The $N,N'$-di-$n$-oktyl-TODGA-COSAN, similarly as other TODGA-like compounds, did not show any substantial actinide(III)/lanthanide(III) selectivity. Therefore, for selective separations of actinides(III) from lanthanides(III) the hydrophilic complexing agent of diethylene-triamine-$N,N,N',N''$-pentaacetic acid (DTPA) with sodium nitrate alone or in combination with different carboxylic buffers have been tested. It was found that Am(III) could be selectively stripped from loaded organic phase by using the solution of 0.05 M DTPA buffered with 1 M citric acid and 1 M NaNO₃ at pH 3.0 ($SF_{Eu/Am} = 13$). A high sensitivity of trivalent metal extraction to pH changes at the stripping step might be mentioned here as a possible drawback of the system.