Modified DTPA ligand systems for simplified trivalent actinide-lanthanide separations based on the TALSPEAK process

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The necessity to reprocess spent nuclear fuel has arisen from increasing volumes of highly active and long-lived radionuclides associated with nuclear fission alongside increasing environmental impact concerns. In particular, one major challenge is the separation of the long-lived trivalent minor actinides (here, Am^{III} and Cm^{III}) from the trivalent lanthanides in order to allow the future transmutation of the actinides into shorter lived radionuclides for the purpose of reducing the long-term radiotoxicity of the waste and the volume build up in storage. There are currently a number of different processes under development world-wide, all of which rely on the coordination chemistry and selective extraction of the minor actinides from the trivalent lanthanides using solvent partitioning. Of these, The TALSPEAK process (Trivalent Actinide Lanthanide Separation by Phosphorus Reagent Extraction from Aqueous Komplexations) has showed great promise in separating the trivalent lanthanides from the minor actinides. The process uses DTPA (diethylenetriamine pentaacetic acid) as the chelating holdback reagent, HDEHP (bis-2-ethylhexyl phosphoric acid) as the extracting ligand and lactic acid as the buffer to prevent precipitation of DTPA that occurs at below pH 3.6. However, despite the considerable separation factors obtainable, and the fact the process exhibits promising radiolysis resistance, the process operates at sub optimal pH values. To overcome these limitations, we have developed a family of DTPA-amino acid conjugates that operate under TALSPEAK-like conditions over a much lower pH range of 1.5–2.5. Additionally, these ligands show comparable selectivity to the TALSPEAK process and are also relatively resistant to radiolysis. We will discuss the coordination chemistry of these ligands with the lanthanides and minor actinides using a combination of luminesence and NMR spectroscopies.