Title: Development of DTPA-amino acid conjugates for successful trivalent actinide-lanthanide separations

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Abstract:

Solvent extraction techniques allow the separation of two or more species using two immiscible liquids and a complexing agent to selectively transfer one species between phases. Multi-dentate ligands are instrumental to extraction and separations chemistry associated with nuclear fuel reprocessing but their solubility at low pH can be problematic. Specifically, the TALSPEAK (Trivalent Actinide Lanthanide Separation by Phosphorus reagent Extraction from Aqueous Komplexations) process utilities DTPA to facilitate the separation of minor actinides, MA^{3+} (Am^{3+} and Cm^{3+}), from Ln^{3+} and Y^{3+}, allowing the MA^{3+} to be reprocessed further by transmutation. The process involves the preferential extraction of the major component (Ln^{3+}) into the organic phase, while the DTPA-MA^{3+} complexes remain in the aqueous phase. The process requires the use of lactic acid as a buffer to maintain pH 3.6 in order to prevent the precipitation of DTPA complexes at low pH, commonly experienced during the processing cycle. To overcome these solubility problems and increase selectivity, a series of DTPA-amino acid conjugates have been synthesized which produce an internal buffer of pH 1.5–2.5, thereby removing the need for lactic acid and increasing the solubility of the subsequent complexants formed. The new ligands simplify the current state-of-the-art TALSPEAK process, allowing it to be carried out at lower pH, increasing efficiency as more acidic conditions have been shown to increase separation factor (SF).

The synthesis, coordination chemistry, photophysical properties and separation behavior of these amino-acid derived ligands is presented and discussed, as well as investigations into their stability towards ionising radiation.