
Evaluating hydroxypyridinone-based ligands for actinide and fission products recovery in used fuels

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ABSTRACT:

High level waste, such as Plutonium Uranium Redox Extraction (PUREX) raffinate, generates much heat and remains highly radiotoxic for thousands of years, mainly due to long-lived fission products (some of which are lanthanides, Ln) and minor actinides (minor An, mostly Np, Am, and Cm). Among the many radionuclides existing in used nuclear fuel (UNF), separating trivalent Ln and minor An from each other is of particular interest. The separation of Am^{3+} and Cm^{3+} from Ln^{3+} is very challenging because they exhibit the same oxidation state, comparable charge densities, and similar hydrated ionic radii. Discrimination primarily relies on the fact that Ln^{3+} ions are slightly harder than An^{3+} ions. One approach for effecting this separation is the use of water-soluble soft donor chelators to hold back Am^{3+} and Cm^{3+} in the aqueous phase, while Ln^{3+} are extracted into an organic phase by hard donor extractants. Our team has developed water-soluble hydroxypyridinone-based (HOPO-) chelators for complexation of An and Ln elements for fundamental chemistry studies, and various medical applications. We determined that some of these chelators present a unique set of properties highly suitable for purification technologies: (i) unprecedented charge-based selectivity; (ii) formation of highly stable complexes even at high acidity (up to 10 M H_{aq}^{+}); (iii) ability to change the oxidation state of metal ions (Sn, Ce, Pu, and Bk) without adding redox active species; (iv) simple and controllable speciation over a broad pH range; and (v) fast complexation kinetics. These properties are un-equalled by other chelating structures, namely the polyaminocarboxylates, and could be the cornerstone of new nuclear fuel treatment strategies. However, as these new chelators would be expected to operate under the extreme conditions of a multi-component (alpha, beta, and gamma) radiation field, their radiolytic robustness, metal complexes, and subsequent radiation-induced changes to coordination environment and oxidation state distribution must be investigated to ascertain the impact on potential process performance. The goal of this project is to investigate the applicability of leveraging the unique An and Ln coordination properties of HOPO aqueous chelators for novel chemical separation approaches under advanced solvent extraction system conditions. A combination of speciation and spectroscopic measurements with liquid-liquid extraction and radiolysis experiments will provide a clear phenomenological understanding of Ln/An complexation with HOPO-based ligands, necessary for their determination as new reagents for UNF.