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Complexation And Extraction Studies Of High Valency Actinides By Salicylideneimine-based Ligands

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Increased knowledge of actinide coordination chemistry and the development of advanced actinide separation processes are essential to reducing the radiotoxicity of used nuclear fuel. Commercial separation techniques for nuclear fuel (i.e. PUREX) selectively remove U(VI) and Pu(IV) from the other components, while the minor actinides, e.g. Np and Am, are not extracted. However, these four mid-actinides (U, Np, Pu, Am) all have highly accessible oxidation states (+V, +VI) at which they exist as linear dioxo actinyl ions [AnO2]n+. The An=O bonds are incredibly stable, permitting ligand coordination in the equatorial plane of the metal ion center and affording a geometry for unique bonding characteristics. Tetradentate Schiff bases can be prepared with synthetic ease and have been used extensively in the area of metal ion coordination chemistry. These ligands have rather planar structures that chelate around the equatorial plane of actinyl ions through their N(2)O(2) binding site, presenting the possibility that this distinct coordination environment may facilitate the selective solvent extraction or aqueous retention of pentavalent and hexavalent actinides. We have synthesized multiple organic and water soluble Schiff base ligands, of the salen and salophen type, and have conducted complexation and solvent extraction experiments with UO22+ and NpO2+, as representative actinyl ions. Preliminary results indicate that salen-based ligands show promise for extracting uranium from nitrate media, while a water soluble salen and a lipophilic di-t-butyl-salophen are the most promising in providing crystallized structures with UO22+ and NpO2+. Ongoing studies of these ligands in our laboratories will help to further define possible methods for used nuclear fuel separations.

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