Project 90252

Next Generation Extractants for Cesium Separation from High-Level Waste: From Fundamental Concepts to Site Implementation

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RESULTS TO DATE: Solvent extraction with a lipophilic calix[4]arenebiscrown-6 ligand is currently the selected technology for removal of radioactive cesium-137 from DOE nuclear wastes. In this collaborative DOE-EMSP project, related "second-generation" extractants are being synthesized at Texas Tech University (TTU) and their alkali metal cation compexation and separation abilities evaluated at TTU and Oak Ridge National Laboratory (ORNL). The novel feature of the "second-generation" calix[4]arenecrown extractants is incorporation of a proton-ionizable group into the ligand structure. This modification markedly enhances the efficiency with which metal ions can be extracted from an aqueous phase into an organic diluent, since concomitant extraction of a hydrophilic anion from the aqueous solution into the hydrophobic organic phase is avoided. During Year 1 of this EMSP project, we established synthetic routes to new, lipophilic, proton-ionizable calix[4]arenebiscrown-6 molecules at TTU and prepared them in sufficient quantities that their efficiency and selectivity in alkali metal cation extraction could be evaluated at ORNL using radiotracer techniques. In Year 2, we have prepared a series of related lipophilic, proton-ionizable calix[4]arenecrown-6 molecules to determine if even higher cesium ion selectivities can be obtained when the extractant has only a single crown ether unit. Evaluation of the these ligands at ORNL has revealed their greater solubility in low polarity organic diluents than that of analogous lipophilic, proton-ionizable calix[4]arenebiscrown-6 ligands. In Year 3, we will continue the synthesis of lipophilic, proton-ionizable calix[4]arenecrown-6-type compounds for evaluation of the influence of varying the acidity of the proton-ionizable group, as well as its positioning with respect to the crown ether ring in which the metal ion is complexed. Special emphasis will be placed on developing practical preparative routes to promising extractants.