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Direct Dissolution and Electrochemical Studies of f-elements in Ionic Liquid

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The unique and tunable properties of the second generation moisture-stable ionic liquids (ILs) have generated interest in their applications to the nuclear fuel cycle. Generally, ILs have low vapor pressure, chemical and radiation stability, good conductivity, and importantly, wide electrochemical windows. This beneficial electrochemical property allows for the ability to reach the negative potentials that are required for the reduction of lanthanides and actinides to metal species. Reduction of actinides and lanthanides to the metal has been largely unavailable in aqueous systems without the aid of mercury electrodes. Ionic liquids could therefore be envisioned as a system into which the used fuel once dissolved could be used to recover the uranium and separate other fission products by applying different potentials. However, the solubility of f-elements in ILs has not been extensively explored. Therefore, a more fundamental understanding of f-element chemistry and electrochemistry in ionic liquids is necessary to evaluate the system.

Exploring methods to enhance and control f-element solubility in ILs is presented. In addition, investigation of the conditions for electrodeposition of uranium from the IL and analysis of the deposited species were conducted. A direct dissolution method which uses the protic form of the anion common to an ionic liquid can provide a pathway for increased solubility of f-elements in ILs. The inclusion of water also enhances the direct dissolution process within the IL. The direct dissolution of lanthanide carbonates and uranium carbonate has been achieved in the IL trimethyl-n-butylammonium bis(trifluoromethanesulfonyl)imide ([Me₃NnBu][TFSI]) using the acid bis(trifluoromethanesulfonyl)amide (HTFSI). In addition to the carbonates, direct dissolution of U₃O₈ was performed. Analysis included UV-Visible and IR spectroscopy to characterize the soluble species and investigate possible complexation with the TFSI ligand. Cyclic voltammetry has been performed at common working electrodes such as Pt, Au, and glassy carbon (GC) to investigate the pathway from the soluble species to reduction. Potential mediated deposition of species from the [Me₃NnBu][TFSI] has been achieved and analysis of the deposits was conducted using scanning electron microscopy (SEM) and energy dispersive x-ray emission spectroscopy (EDX). The results indicate that while deposition from the IL is possible despite residual water, the resulting deposited species tend to be oxides. Further studies on electrochemical parameters and utilizing simple methods to remove water are being investigated in an attempt to promote deposition to the metal from IL solutions.

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