Environmental Management Science Program

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Potential-Modulated Intercalation of Alkali Cations into Metal Hexacyanoferrate Coated Electrodes

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Research Objective

This program is studying potential-driven cation intercalation and deintercalation in metal hexacyanoferrate compounds, with the eventual goal of creating materials with high selectivity for cesium separations and long cycle lifetimes. The separation of radiocesium from other benign cations has important implications for the cost of processing a variety of cesium contaminated DOE wasteforms.

Research Progress and Implications

This report summarizes results after nine months of work. Much of the initial efforts have been directed towards quantitatively characterizing the selectivity of nickel hexacyanoferrate derivatized electrodes for intercalating cesium preferentially over other alkali metal cations. Using energy dispersive xray spectroscopy (ex-situ, but non-destructive) and ICP analysis (ex-situ and destructive), we have demonstrated that the nickel hexacyanoferrate lattice has a strong preference for intercalated cesium over sodium. For example, when ions are reversibly loaded into a nickel hexacyanoferrate thin film from a solution containing 0.9999 M Na^+ and 0.0001 M Cs^+, the film intercalates 40% as much Cs^+ as when loaded from pure 1 M Cs^+ containing electrolyte (all electrolytes use nitrates as the common anion). We have also shown that, contrary to the common assumptions found in the literature, a significant fraction of the thin film is not active initially. A new near infrared laser has been purchased and is being added to our Raman spectroscopy facilities to allow in-situ studies of the intercalation processes.

Planned Activities

We are just beginning to look at other metal hexacyanoferrate compounds, the use of chemometric methods to quantify the Cs content of the thin films more readily using electrochemical methods alone, and also to use quartz crystal microbalance studies to measure selectivity in the form of weight gain. Raman spectroscopy at longer wavelengths than currently achieveable will also come on line soon. These studies should become routine in our laboratory in the next 6-8 months. These new in-situ characterization techniques are expected to lead to a better understanding of the the processing-structure-property relationships that control the ion intercalation behavior in these materials.