Fundamental Electrochemical Properties of Liquid Metals in LiCl-KCl for Separation of Alkali/Alkaline-Earths (Cs, Sr and Ba)

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ABSTRACT:

The proposed research will probe the fundamental thermodynamic and electrochemical properties of liquid metal electrodes for the separation of alkali/alkaline-earth fission products (Cs, Sr, and Ba) in LiCl-KCl electrolytes. Molten salt electrochemical cells are one of the most promising technologies to process used nuclear fuel and help close the nuclear fuel cycle. One key challenge to this approach is the accumulation of fission products in the molten salt electrolyte that changes the electrolyte properties and diminish the performance efficiency of an electrorefiner. This challenge requires advanced separation methods for fission products to minimize the volume of nuclear wastes.

Proposed is a novel liquid metal electrode that selectively dissolves alkali/alkaline-earths (Cs, Sr, and Ba) by leveraging strong atomic interactions between these fission products and liquid metal electrodes (Bi, Pb or Sb). This work specifically aims to: (1) characterize the fundamental atomic interactions between alkali/alkaline-earths (Cs, Sr, and Ba) and candidate liquid metals (Bi, Pb, and Sb); (2) measure the electrode kinetics of alkali/alkaline-earths at liquid metal-electrolyte interfaces; (3) determine the selectivity of liquid electrodes for Cs, Sr, and Ba; and (4) model thermodynamic properties of alkali/alkaline-earths in liquid metal electrodes. Atomic interactions of alkali/alkaline-earths will be directly investigated using an emf cell to obtain thermodynamic properties and supplemented with thermal and structural characterization (DSC and XRD). Electrokinetic properties of electrode-electrolyte interfaces, such as charge transfer, mass transport, and ohmic losses, will be explored using a three-electrode molten salt electrochemical cell and suite of electrochemical techniques. The liquid metal selectivity for alkali/alkaline-earth fission products will be explored using a two-electrode electrolytic cell and supplemented with chemical and structural characterization techniques (SEM/EDS, EPMA, and XPS). Lastly, experimental results will be incorporated into a thermodynamic model that will explain and predict the electrochemical behavior of alkali/alkaline earths in multi-component systems.

This work directly addresses the objective of Topic FC-1.1: to develop innovative methods to separate reusable fractions of used nuclear fuel and manage the resulting wastes by understanding (1) fundamental thermodynamic properties, (2) phase equilibria, and (3) kinetic parameters in molten salt systems. This work will result in accurate thermodynamic and kinetic properties of alkali/alkaline-earth metals in liquid metals and will be leveraged to design liquid metal/molten salt electrochemical cells for separating alkali/alkaline-earth fission products. Ultimately, this work will enable salt recycle in an electrorefiner by electrochemical separation of fission products, and reduce the volume of the nuclear waste by concentrating alkali/alkaline-earths into chloride-free, metallic form which can be easily converted to oxides for storage.