Recovery of Rare-Earth Elements (Nd, Gd, Sm) in Oxide Wasteform Using Liquid Metals (Bi, Sn)

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ABSTRACT:

The proposed research investigates a new approach for recovering rare-earth fission products (Nd, Gd, and Sm) from molten chlorides (LiCl-KCl eutectic) using liquid metals (Bi and Sn). In the electrorefining process for separating uranium and transuranic metals from used nuclear fuel, rare-earth fission products accumulate in the molten salt as dissolved ions, which must be removed to recycle the process salt and minimize nuclear waste generation. One key challenge in the recovery of rare-earth fission products is a low recovery yield due to the presence of side reaction pathways: the partial reduction (e.g., $Nd^{3+} + e^- \rightarrow Nd^{2+}$) from multivalent states of rare-earth ions in the salt and the back dissolution (e.g., $Nd + 2NdCl_3 \rightarrow 3NdCl_2$) due to the high reactivity of rare-earth metals.

In order to achieve a high recovery yield of rare-earth fission products, a liquid metal electrode is proposed that will eliminate the side reaction pathways by leveraging the strong chemical interactions between rare-earth elements and liquid metals to facilitate a direct one-step reduction of the rare-earths into the liquid metals (e.g., $Nd^{3+} + 3e^- \rightarrow Nd(\text{in Bi})$). Ultimately, the reduced rare-earth metals will be converted to rare-earth oxides by selective oxidation so that the chloride-free rare-earth oxides could be incorporated into conventional glass or ceramic wasteforms at optimal loadings. This work specifically aims to develop an efficient rare-earth recovery process by determining (1) the thermodynamic and electrochemical properties of rare-earth metals (Nd, Gd, and Sm) in liquid metals (Bi and Sn) in molten LiCl-KCl; (2) predictive thermodynamic models of multi-component alloys to identify optimal liquid metal compositions for maximum recovery yield using computational modeling (e.g., high throughput CALPHAD modeling and high throughput first-principles calculations); and (3) the overall rare-earth recovery efficiency from the electrochemical separation into liquid metal electrodes to the conversion process into rare-earth oxides. On a fundamental level, this work will contribute new thermodynamic and electrochemical data for rare-earth species in liquid metals to accelerate the development of rare-earth recovery process, filling a critical knowledge gap in understanding their physical and chemical behavior in liquid alloys and molten salts.

Advanced separation of rare-earth fission products with improved control of chemical selectivity and high recovery yield in this work will (1) enable the recycling of molten salts by electrochemically separating rare-earth fission products into liquid metals at high recovery efficiencies; (2) develop chloride-free rare-earth oxides that can be incorporated into conventional glass or ceramic wasteforms; and (3) minimize the generation of additional waste streams by reusing the liquid metals.