
Characterizing multivalent states of fission products (Sm, Eu, and Te) in molten salts for their recovery and stabilization

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ABSTRACT:

This project investigates samarium (Sm), europium (Eu), and tellurium (Te) fission product elements in molten salts by elucidating their multivalent states as governed by the redox conditions of molten salts. Understanding these multivalent states is important because they significantly influence the chemical behavior, solubility, and partitioning of these elements which are critical factors for optimizing separation processes and improving the efficiency of nuclear fuel recycling. Specifically, the neutron absorption of Sm, dissolution of structural materials by Eu, and the formation of brittle intermetallic Te compounds compromise the efficiency and integrity of molten salt reactors and nuclear fuel recycling processes. Building upon the electrochemical, spectroscopic, and thermodynamic properties of multivalent fission products in molten salts, this work will demonstrate the recovery of stable Sm and Eu rare-earth fission products using liquid metal cathodes (Bi, Sn, and Cd) and elucidate the redox condition(s) necessary to stabilize Te fission products in molten salts to mitigate materials degradation.

To achieve the above objectives, the project team will (1) determine multivalent transitions of fission products (e.g., $\text{Sm}^{3+} \rightarrow \text{Sm}^{2+} \rightarrow \text{Sm}^0$ (in liquid Bi)) in molten salts (LiCl-KCl and NaCl- UCl_3) using electrochemical techniques over a range of potentials at 450-600 °C; (2) validate the change in fission product oxidation state during electrolysis by *in situ* spectroscopic measurements and determine the speciation and coordination of each valence state in molten salts; (3) demonstrate electrochemical recovery of Sm and Eu ions into liquid metal cathodes in complement with thermodynamic property measurements of liquid alloys (e.g., Sm-Bi); and (4) comprehensively monitor the concentrations of multivalent fission products (Sm, Eu, and Te) in an electrorefiner at Idaho National Laboratory (INL) to optimize separation processes for these fission products to inform future fuel cycle strategies.

By elucidating the fundamental properties of multivalent fission products, the project aims to improve the reliability and efficiency of molten salt reactors and pyroprocessing systems. The findings will be integrated into INL's proprietary mass tracking and process simulation tool, the Modeling and Simulation Tool for Electrochemical Recycling Systems (MASTERS), to enhance waste management and operational practices. This research supports DOE NEUP's mission to advance nuclear technologies by contributing to sustainable fuel cycles, improving reactor performance, and extending the lifespans of structural materials.