Investigation of technetium-99 speciation in molten salts

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ABSTRACT:

Molten salt reactors (MSRs) are one of the most promising emerging nuclear reactor technologies. In MSRs, the fissile nuclear fuel is dissolved in a molten chloride or fluoride salt that also acts as a coolant. As the reactor operates, the fuel salt composition will change due to fission product ingrowth. Removal of fission products is challenging, therefore understanding the speciation of fission products in MS is key to designing effective separation strategies.

Significant research is already being conducted on the speciation of uranium fuel and fission products in molten salts (MS). However, comparable studies on the very important fission product, technetium-99, Tc, in MS are lacking, and that is the gap that this project intends to fill. Tc-99 is a high yield by-product of uranium fission and is one of the most problematic fission products.

This project combines accumulated decades of experience in inorganic and Tc chemistry at Hunter and Lehman Colleges with profound experience and knowledge in molten salts, radiation chemistry and advanced X-ray Absorption Fine Structure (XAFS) spectroscopy methods and state-of-the-art analysis methods at BNL. This project will apply that powerful infrastructure to understand the speciation of Tc in MS.

Objective 1: **Elucidate the effects of molten salts' strong cation-anion interactions on Tc speciation using low-melting eutectic LiCl-KCl.** In this objective, we will examine Tc (and its surrogate, Re) speciation in molten LiCl-KCl eutectic, while developing experimental protocols for synchrotron and radiolysis experiments on radioactive materials at 400-450 °C. LiCl-KCl eutectic is used for the electrolytic recovery of uranium and other elements from used nuclear fuel. The goal is to understand how the cation-anion interactions intrinsic to the salt matrix control the solute (Tc^{n+}) – solvent (salt) interactions responsible for the behavior of Tc species in this system.

Objective 2: **Interpret the speciation and redox chemistry of Tc in complex, tunable salt systems.** Mixtures of monovalent and multivalent cations with tunable Lewis basicity represent practical systems for molten salt reactors. We will use two salt mixture systems and vary their compositions to explore how the speciation of Tc (and Re) complexes can be controlled by tuning mixture composition: $MgCl_2$-KCl and $MgCl_2$-NaCl, over temperatures up to 700 °C. These specific systems are important to study because they are, in fact, considered for planned fuel salt formulations.

Understanding the chemistry that Tc undergoes in the reactor starts with understanding its speciation under reactor conditions, and the work in this proposal establishes the methodologies to address this challenge while exploring operationally relevant salt systems. The outcome/impact of this project is that understanding the speciation of Tc in MS will lead to better approaches to manage Tc in reactor fuel salt and separate Tc from used MS fuel.