# Modeling Solute Thermokinetics in LiCl-KCl Molten Salt for Nuclear Waste Separation

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**ABSTRACT**

Recovery of actinides is an integral part of a closed nuclear fuel cycle. Pyrometallurgical nuclear fuel recycling processes have been developed in the past for recovering actinides from spent metallic and nitride fuels. The process is essentially to dissolve the spent fuel in a molten salt and then extract just the actinides for reuse in a reactor. Extraction is typically done through electrorefining, which involves electrochemical reduction of the dissolved actinides and plating onto a cathode. Knowledge of a number of basic thermokinetic properties of salts and salt-fuel mixtures is necessary for optimizing present and developing new approaches for pyrometallurgical waste processing. The properties of salt-fuel mixtures are presently being studied, but there are so many solutes and varying concentrations that direct experimental investigation is prohibitively time consuming and expensive (particularly for radioactive elements like Pu). Therefore, there is a need to reduce the number of required experiments through modeling of salt and salt-fuel mixture properties.

This project will develop first-principles-based molecular modeling and simulation approaches to predict fundamental thermokinetic properties of dissolved actinides and fission products in molten salts. The focus of the proposed work is on property changes with higher concentrations (up to 5 mol%) of dissolved fuel components, where there is still very limited experimental data. The properties predicted with the modeling will be density, which is used to assess the amount of dissolved material in the salt; diffusion coefficients, which can control rates of material transport during separation; and solute activity, which determines total solubility and reduction potentials used during electrorefining. The work will focus on La, Sr, and U, which are chosen to include the important distinct categories of lanthanides, alkali earths, and actinides, respectively.

Studies will be performed using LiCl–KCl salt at the eutectic composition (58 mol% LiCl, 42 mol% KCl), which is used for treating spent EBR-II fuel. The same process being used for EBRII fuel is currently being studied for widespread international implementation. The methods will focus on first-principles and first-principles derived interatomic potential based simulations, primarily using molecular dynamics. Results will be validated against existing literature and parallel ongoing experimental efforts.

The simulation results will be of value for interpreting experimental results, validating analytical models, and for optimizing waste separation by potentially developing new salt configurations and operating conditions.