Effect of Multiple Uranium Complexes on Chloride Fast Reactor Molten Salt Properties

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ABSTRACT:

It has been reported by this NEUP team and others that multivalent transition metal ions in a molten salt can exhibit multiple coordination states that are dependent on temperature and composition, and these can significantly affect the prediction of molten salt properties. The tendency toward such disordered structures is expected from uranium complexes in the melt due to the multiple valence states for uranium and complex halide phase equilibria. This project will use knowledge gained in measuring the local properties of uranium ion complexes of UCl_3 and UCl_4 in NaCl molten salts to better understand how uranium complexes coordinate in the melt, and the implication for molten salt properties. The effect of CsCl as a fission product in the melt on local uranium properties will also be studied. Local chemistry, valence and coordination will be measured using high-energy resolution fluorescence detection (HERFD) X-ray absorption near edge structure (XANES) spectroscopy, which has recently been demonstrated to overcome issues associated with the interference and distortion of the extended X-ray absorption fine structure (EXAFS) spectra that can result in anomalous coordination numbers. Results will be validated by Raman spectroscopy and with Universal Structure Predictor: Evolutionary Xtallography (USPEX) with First-Principles Molecular Dynamics (FPMD) simulations. A CALPHAD modified quasichemical model in the quadruplet approximation to handle multiple uranium complexes will be created to accurately predict thermodynamic properties of molten salts. This effort builds upon previous work under DOE-NEUP Project CFA-18-15065 where we successfully characterized multiple Zr and U complexes in molten salts to pursue a new technical scope to investigate the influence of multiple uranium complexes on molten salt properties. We expect this project to further the understanding of the effect of multiple uranium complexes on a molten salt's molecular structure and properties. This project will interact closely with the Molten Salt Thermodynamic Database at Oak Ridge National Laboratory and other molten salt databases, as well as team efforts on synchrotron x-ray spectroscopy and molten salt research by the team in current and past NEUP projects and DOE Energy Frontiers Research Centers.