Understanding Molten Salt Chemistry Relevant to Advanced Molten Salt Reactors through Complementary Synthesis, Spectroscopy, and Modeling

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ABSTRACT:

The goal of the proposed research is to understand molten salt chemistry relevant to advanced molten salt reactors through complementary synthesis, spectroscopy, and modeling. In synthesis, we will prepare high purity halide melts, instead of the more commonly studied fluorides. In characterization, we will employ high-temperature sample cells to enable structural and spectroscopic studies of the melts by *in situ* x-ray and neutron scattering, x-ray absorption spectroscopy, UV-vis, and IR/Raman. In modeling, we will leverage both ab initio molecular dynamics and classical molecular dynamics based on polarizable-ion models, coupled with advanced hardware such as graphics processing unit, to interpret the experimental data and understand the underlying mechanisms. Through complementary synthetic, spectroscopic, and computational efforts, we aim to achieve atomistic and molecular-level understanding of liquid structure, coordination geometry, chemical bonding, and reactivity of novel molten salt melts relevant to advanced molten reactor designs. Our proposed work will synthesize and purify UCl_3/UCl_4 in NaCl, LiCl, and $MgCl_2$; validate experimental methodology by comparing physical (e.g. viscosity, density, heat capacity) and spectroscopic (UV-vis) data with literature sources; apply x-ray absorption fine structure spectroscopy, analysis of the pair distribution function, and neutron diffraction to investigate the uranium coordination environment and its influence on NaCl, LiCl, and $MgCl_2$ structure; computationally investigate the structure, excited states, and chemical reactivity of UCl_3/UCl_4 in NaCl, LiCl, and $MgCl_2$ through molecular dynamics, time-dependent density functional theory, and ab initio molecular dynamics studies; and synthesize, characterize, and simulate UCl_3/UCl_4 in $NaCl/MgCl_2$ and $LiCl/MgCl_2$ eutectic mixtures. This proposed research directly addresses the priority direction "Understanding the Structure, Dynamics, and Chemical Properties of Molten Salts" identified in the recently published DOE Office of Nuclear Energy Molten Salt Chemistry Workshop report.