Connecting Advanced High-Temperature X-ray and Raman Spectroscopy Structure/Dynamics Insights to High-Throughput Property Measurements

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ABSTRACT:

Molten salt reactors (MSRs) are nuclear fission reactors that provide many unique benefits. In the Generation IV International Forum it was selected as one of six next-generation reactors. In MSRs the primary/secondary coolant and/or the fuel itself is a molten salt mixture. Molten salts have high volumetric heat capacities, can be operated at near atmospheric pressures, easily dissolve fissile and/or fertile materials including some of the fission products, and have higher boiling points than traditional coolants allowing higher temperature (> 700 C) operation resulting in greater efficiencies. When used as fuels, molten salts allow safer operation since they have high coefficients of thermal expansion resulting in negative temperature coefficients of reactivity, and simpler fuel cycles by enabling online fission-product removal and eliminating solid fuel handling. In spite of the many benefits of using molten salts in nuclear reactors, there remain significant challenges that need to be overcome. The Molten Salt Chemistry Workshop held at ORNL in April 2017 highlighted some of the major remaining challenges in MSRs and identified six future research directions (FRDs). This proposal primarily addresses one FRD, "Understanding structure, dynamics, and chemical properties", and will also contribute to another FRD, "Understanding, predicting, and optimizing physical properties".

This research will focus on model MSR coolant and carrier salts containing fissile/fertile fuel halides: LiF-NaF-KF (m% 46.5-11.5-42 - FLiNaK) and NaF-ZrF_4-UF_4 (m% 53-41-6). The latter salt was used as a fuel and primary coolant in the Aircraft Reactor Experiments (ARE) [2] and the former salt was used as a heat transfer coolant in very high temperature thermal spectrum reactor (VHTR) [3] and is also used for solar energy storage applications. Although many experimental studies and measurements have been made on these salt systems including thermophysical properties and in some instances phase equilibria, there is no study that tries to systematically understand the relationship between speciation and their thermophysical properties. In this work, thermally activated thermophysical properties (viscosity, surface tension, electrical conductivity, and vapor pressure of chemical species) will be measured as a function of temperature while corresponding speciation information are also simultaneously determined from their structure and dynamics data. The structure and dynamics data at high temperatures will be obtained from novel applications of high-brightness x-ray scattering experimental techniques and Raman spectroscopy. From the various structure dynamic measurements and combined with modeling one can determine with relatively high confidence the speciation information: oxidation state, coordination numbers, the size and nature of the species, the bond lengths and bond energies associated with the species, and nearest and next nearest anion species surrounding the cations. It is expected that the speciation in NaF-ZrF_4-UF_4 and FLiNaK will be very different. The former will have more covalent characteristics while the latter will be essentially ionic. By studying the variation of the species in the salt with temperature, the thermally-activated properties will be related to speciation and Lewis melt basicity. The work will provide a methodology for connecting advanced high-temperature x-ray and Raman spectroscopy structure-dynamics data to thermophysical properties.