Probing Speciation of Light Elements in Molten Salts by Electrochemistry, High Temperature Liquid NMR, and Neutron Diffraction

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ABSTRACT:

Light elements such as tritium isotopes and carbon are present in molten salt fuel and play an important role in corrosion and tritium management; they may also prove to have an impact on the molecular structure and long-range order in molten salt ionic liquids. Depending on the fluoroacidity of the melts and the properties of the solvent cations, molten fluoride salts form monomeric and oligomeric species, that in turn can also demonstrate longer-range ordering. The presence of solutes may disrupt the oligomer network and subsequently impact the chemical and thermo-physical properties of the solutes.

Speciation of solutes and the impact of light-element solutes on the solvent structure will be investigated for the following elements: hydrogen (tritium is an activation product in coolants containing Be or Li), beryllium metal (used for redox control), and carbon (present in abundance for graphite-moderated reactors and of importance to corrosion of metal alloys). The relationship between solvent structure and solvation of light elements will be probed; the solvent structure of $2LiF\text{-}BeF_2$ (FLiBe) will be investigated over a range of BeF_2 compositions, representative of a range of fluoroacidity and of oligomer network structure.

The joint application of in-situ electrochemistry, neutron and x-ray diffraction, ab-initio molecular dynamics (AIMD), and liquid NMR to FLiBe is novel. Application of electroanalytical tools for the study of solvated hydrogen, carbon, and beryllium metal is novel. The study of hydrogen is enabled by the discovery of a sufficiently long-lived non-equilibrium state of zero-valence state hydrogen in the FLiBe. Electrochemical study of carbon in molten FLiBe has not been attempted to date. The application of liquid NMR to study FLiBe and light elements (H, Be, C) in FLiBe is novel. If successful this technique will elucidate whether hydrogen forms compounds with carbon or Be, leading to high apparent solubilities of H in FLiBe when C or Be metal are also dissolved in the salt.

The suite of techniques being explored here for a very specific problem would subsequently enable the probing of an entire class of questions pertaining to out-of-equilibrium behavior in molten salt (which is likely to be relevant in nuclear fuel where off-equilibrium species are being produced by fission, activation, and in-salt chemical reactions among intermediate species as well as equilibrium species). The specific studies of hydrogen, carbon and beryllium mental in FLiBe have direct application to tritium management and corrosion management in FHR and MSR reactors.