

Chemical Engineering Doctoral Defense

Development of Liquid Electrolytes for Extreme Temperatures: Design, Optimization, and Impact of Molecular Interactions

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Abstract

Electrolytes play a critical role in electrochemical devices and applications, and therefore design and development of electrolytes with tailored properties are much desired to accommodate variety of operation requirements. Extreme temperatures are considered as one of the challenging environmental conditions, especially for devices rely on liquid state electrolytes, rendering failure of operations once the electrolyte systems undergo phase transitions. This work focuses on development of low-temperature iodide-containing liquid electrolyte systems, specifically designed for the molecular electronic transducer (MET) sensors in space applications. Utilizing ionic liquids, molecular liquids, and salts, multiple low-temperature liquid electrolytes were designed with enhancements in thermal, transport, and electrochemical properties. Effects of intermolecular interactions were further investigated, revealing correlations between optimization of microscopic dynamics and improvements of macroscopic characteristics. As a result, three low-temperature electrolyte systems were reported, and the liquidus range of these systems have been extended with the lowest down to $-152\text{ }^{\circ}\text{C}$, marking the lowest liquidus temperature of electrolytes to our best knowledge. Moreover, transport properties of designed systems were characterized from 25 to $-75\text{ }^{\circ}\text{C}$. Effects of selected cosolvent/solvent on evolution of transport properties were observed, revealing interplay between two governing mechanisms, ion disassociation and ion mobility, and their dominance at different temperatures. Experimental spectroscopy characterization techniques validated the hypothesized intermolecular interactions between solvent-cation and solvent-anion, complimented by computational simulation results on the complex dynamics between constituent ions and molecules.

To support MET sensing technology, the essential iodide/triiodide redox were investigated in developed electrolytes. Effects of different molecular solvents on electrochemical kinetics were elucidated, and steady performances were validated under a properly controlled electrochemical window. Optimized electrolytes were tested in the MET sensor prototypes and showcased adequate functionality from calibration. The MET sensor prototype has also successfully detected real-time earthquake with low noise floor during long term testing at ASU seismology facility. The presented work demonstrates a facile design strategy for task-specific electrolyte development, which is anticipated to be further expanded for broader applications in the future.



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