Synthesizing computational and experimental strategies for the design and analysis of novel fuel-cell and battery electrolytes driven by the Grotthuss mechanism

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Candidate systems for next-generation electrolyte materials, such as deep eutectic solvents and ionic liquids, often suffer from high viscosities, which suppresses rates of charge transport and limits their performance characteristics. A strategy for circumventing this problem is to leverage the structural or Grotthuss mechanism of proton transport through hydrogen-bond networks, in which charge transport occurs through a series of proton hops from donor to acceptor. Liquids that support the Grotthuss mechanism can exhibit high proton diffusion rates even when rates of vehicular charge transport are poor. In this talk, I will describe a project aimed at leveraging computational design and simulation approaches, in combination with experimental synthesis and characterization to develop novel classes of battery electrolytes based on the structural/Grotthuss diffusion mechanism. The workflow involves identification of a suitable proton-carrier liquid, a proton source, and a redox-active species capable of undergoing proton-coupled electron transfer (PCET) reactions. I will discuss the computational methods, which involve a combination of ab initio molecular dynamics and machine learning simulation techniques combined with path integrals for treating nuclear quantum effects, candidate chemical species selection for each of the components, protocols for combining these components into a high-performance electrolyte, current results, and next steps in the evolution of the project. Machine learning models include equivariant transformer networks, which allow for rapid training on relatively small data sets. This work will serve to illustrate both the power of modern computational approaches in the design of electrochemical systems but also to broaden the perspective on what constitutes a "breakthrough" electrolyte.