

Atomistic Simulations of Ionic Liquid and Polymer Electrolytes: From Bulk Phases to Interfacial Behavior

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Abstract:

Ionic liquids and polymers are candidate electrolytes for high-energy density, rechargeable batteries. We present an extensive computational analysis with experimental comparisons of bulk and interfacial properties of three ionic liquid electrolytes ([pyr14][TFSI], [pyr13][FSI], and [EMIM][BF₄]) as a function of Li-salt doping. We investigated the bulk electrolyte using quantum chemistry and *ab initio* molecular dynamics to elucidate the solvation structure of Li⁺. MD simulations using polarizable force fields were performed, from which we obtained an array of thermodynamic and transport properties. We computed the electrochemical window of the electrolytes across a range of Li⁺-doping levels, including the effect of the liquid environment. In addition, we considered these Li-doped electrolytes at ideal electrified interfaces to evaluate the differential capacitance and the equilibrium Li⁺ distribution in the double layer. Finally, we investigated polymers as candidate structural electrolytes. We report results for ionic conductivity, glass transition temperature and bulk modulus for representative polymer systems.