Molecular dynamics of post-lithium battery electrolytes : priorities and challenges

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In the current search for alternatives to Li-ion batteries to satisfy rising global demand for energy storage, liquid electrolytes remain at the center of research and development efforts. Their composition varies greatly,[1] [2] as some alkaline and alkaline-earth ion electrodes are incompatible with the carbonate-based solvents traditionally used for Li-ion batteries. Furthermore, even viable systems show deviations from the conduction behavior of Li-ion systems, which resists straightforward experimental explanation.[3] To this end, classical Molecular Dynamics (MD) is an invaluable tool to bridge the gap between experimental observations and atomistic scale phenomena to investigate what causes a system to function or fail. We present our work through two model systems: a glyme-based Ca-ion battery electrolyte and a carbonate-based Na-ion battery electrolyte. First, we discuss the cost-benefit of taking into account physical effects such as polarisability and nuclear quantum effects (i.e. the zero-point energy) based on the physical properties of these electrolytes (e.g. dielectric constant, infrared spectra). In the latter part, we analyze the simulations performed with the previously discussed parameters, using the unconventional tools of graph theory to draw connections between the changing ion transport properties and the modified solvation spheres of cations. Lastly, we project how this kind of analysis may be advantageous to study the enigma that is the build-up of the solid-electrolyte interface, as well as what effects would remain relevant in the case of an applied potential.

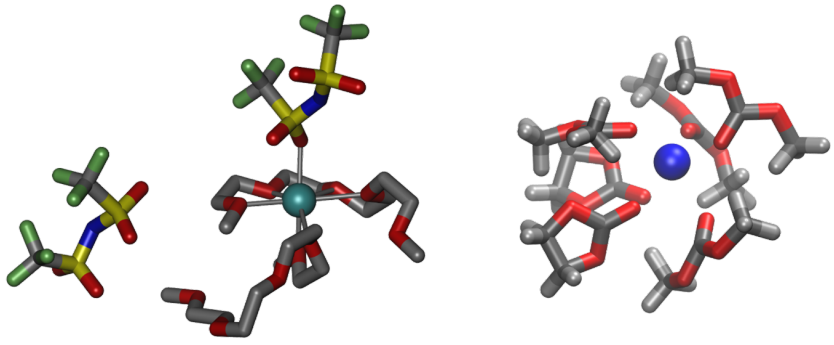

Figure 1: Solvation sphere examples in two different electrolyte systems

Keywords: classical molecular dynamics ; battery electrolytes ; post-lithium batteries ; graph theory.

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