Designing polymer-based electrolytes with high lithium ion transference number and conductivity

Bryan D. McCloskey
Chemical and Biomolecular Engineering, UC, Berkeley
Energy Storage and Distributed Resources Division, LBNL
bmcclosk@berkeley.edu

Single ion conducting polymer electrolytes have been studied for many years as a promising means to create a high lithium ion transference number electrolyte. A high transference number electrolyte would eliminate concentration polarization, the buildup of concentration gradients within the battery electrolyte during operation, thereby allowing rapid charging and increasing the accessible energy density. Current state of the art polymer electrolytes still suffer from low conductivity at moderate to room temperature. In this presentation, we discuss the creation of a new single-ion conducting polysulfone-co-poly(ethylene glycol) copolymer (PSf-co-PEG) and leverage its large accessible miscible composition window to probe the established understanding of ion conduction. We demonstrate the inherent tradeoff between segmental motion and ion content, clearly illustrating the challenges facing polymer electrolyte design. We further demonstrate the existence of the compensation effect, a positive correlation between the prefactor and apparent activation energy, within the Vogel-Tammann-Fulcher equation. The compensation effect has significant implications for any polymer electrolyte system, as its existence predicts that the maximum in conductivity for any given system will occur as the prefactor tends to low values. We show that blending of a small molecule diluent is sufficient to break the apparent trend in the PSf-co-PEG system, indicating a clear path to an electrolyte with optimal transport properties. When this strategy is fully extended to electrolytes in which single-ion conducting polymers are entirely dissolved in nonaqueous solvents, an unprecedented combination of liquid-like conductivities (1.3 mS/cm at 25 ^{o}C) and Li^{+} transference numbers (>0.95) is obtained for certain electrolyte compositions.