Séminaire LIONS

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Designing Ionomers for Facile Ion Transport

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We synthesize single-ion conducting ionomers with low glass transition temperatures to prepare ion conducting membranes for actuators and lithium battery separators. We use dielectric spectroscopy to determine the number density of conducting ions and their mobility from electrode polarization (using the 1953 Macdonald model) and the number density of ion pairs from measured dielectric constant (using the 1936 Onsager model). This experimental work concludes that the number density of conducting ions is tiny, and we discuss ways to boost that using more polar polymers with weak-binding anions attached to the chain. We use ab initio quantum chemical calculations at 0 K in vacuum to characterize ion interactions and ion solvation by various functional groups on ion-containing polymers. Simple ideas for estimating the ion interactions and solvation at practical temperatures and dielectric constants are presented that indicate the rank ordering observed at 0 K in vacuum should be preserved. Hence, such ab initio calculations are useful for screening the plethora of combinations of polymer-ion, counterion and polar functional groups, to decide which are worthy of synthesis for new ionomers. The results provide estimates of parameters for a simple four-state model for counterions in ion-containing polymers: free ions, isolated ion pairs, triple ions and quadrupoles. We show some examples of how ab initio calculations can be used to understand experimental observations of dielectric constant, glass transition temperature and conductivity of polymerized ionic liquids with either lithium or ionic liquid counterions. In particular, recent calculations provide some important insight as to why poly(ethylene oxide) is able to raise the dielectric constant to boost ion transport.