283c Lithium Ion Transport in Rubber-like Poly(Ethyleneimine) Electrolytes

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Polymer gel electrolytes represent a promising "leak free" alternative to liquid electrolytes for rechargeable lithium ion batteries. For optimal function, the gel system should remain amorphous over a broad temperature range (-20 °C to 60 °C), should not phase separate at low or high temperature, must have adequate mechanical properties, and must not undergo oxidation at or below a potential of ~4 V. In addition, the electrolyte material should have an ionic conductivity of at least 10^{-3} S/cm (preferably higher) over the temperature range of operation, which is usually the limiting factor in material performance. Several promising gel electrolyte systems have been examined in recent years, frequently based upon poly(ethylene oxide) (PEO) or poly(vinylidene fluoride)/poly(hexafluoropropylene) copolymers (PVDF-HFP). Although promising results have been obtained, improvements in ionic conductivity would permit development of more versatile electrolytes.

Gel electrolytes generally consist of a matrix polymer, a solvent, a lithium salt, and sometimes crosslinkers and/or fillers. Microporosity may also be introduced to enhance Li ion transport. Identifying the physical and chemical factors governing ion transport is often complicated by the number of components present and the often ill-defined microstructure of the gels. Characterization of well-defined gel electrolytes is therefore a priority. Our group has been characterizing Li ion transport in a less frequently studied gel system, poly(ethyleneimine) (PEI). Using a crosslinker and a high boiling solvent, we are able to prepare completely amorphous, single-phase PEI gels with rubber-like mechanical behavior and very high Li salt content. Li ion conduction in our materials is characterized over a broad temperature range using broadband dielectric spectrometry (BDS). We are establishing how ionic conductivity is affected by crosslink density, solvent content, and temperature, and characterizing mechanical properties by dynamic mechanical analysis and swelling in good solvents. Our presentation will summarize our findings and compare the advantages/limitations of the PEI electrolytes with respect to the more frequently studied PEO and PVDF-HFP systems.