Analysis of composite electrolytes for stability with Li metal anodes

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Development of solid electrolytes for electrochemical energy storage aims at elimination of safety concerns associated with flammability of solvents used in liquid electrolytes. Many systems have been developed based on the glass, ceramic and polymer electrolytes [1]. The research has been primarily driven towards improvements in ionic conductivity. The additional requirement for a solid electrolyte is its ability to block lithium dendrites which imposes certain restrictions on mechanical properties of such system. While ceramics and glass electrolytes are certainly stiff enough to prevent dendrites from forming on the surface, they are highly brittle and develop crack easily under loading. Polymer electrolytes alone may not be sufficient to block lithium dendrites due to their low stiffness. In order to resolve the above issues, composite polymer electrolytes (CPE) have been studied as a combination that integrates high stiffness of ceramics with incompressibility of polymers to form an electrolyte with suitable mechanical properties as well as improved conductivity.

The presentation will focus on the modeling approaches to guide the development of such composites. Analysis of the systems with dispersed particles will be presented first with the methods for microstructure development (Fig.1). Analytical as well as numerical methods for computation of effective properties will be discussed. The results will be discussed in terms of the influence of particle size, particle size distribution, and critical volume fraction required for both mechanical stability and improved conductivity.

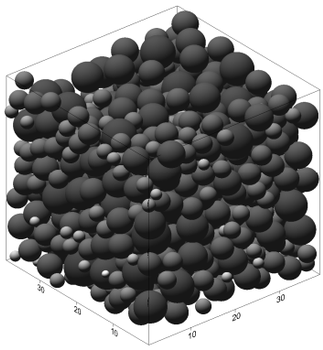

Fig.1. Representative volume element showing the microstructure with Gaussian particle size distribution at 52% volume fraction.

As has been determined rather high volumetric loadings of filler are required to reach the stiffness values required for mechanical stability with lithium metal [2]. In addition, interfacial resistance may hinder achievement of modeling predicted conductivities in dispersed systems. In order to eliminate the above concerns, a system consisting of sintered ceramics filled with polymer is considered (Fig. 2).

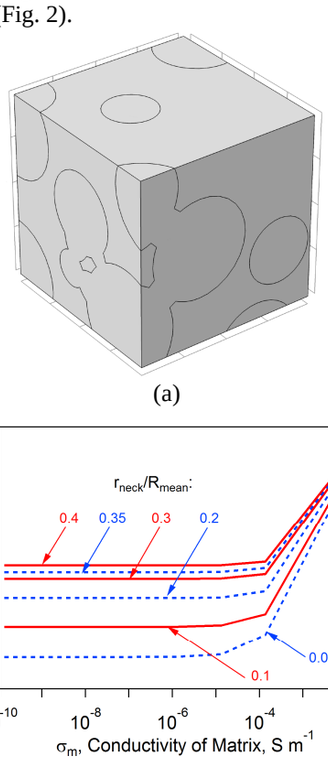

Fig.2. Sintered CPE: (a) microstructure with inter-particle sintering connections; (b) effective conductivity

The computations were based on garnet lithium lanthanum zirconate ceramic sintered bed filled with polymer. A typical polyethylene oxide with lithium salt (LiTFSI) was considered as a baseline. By varying the polymer mechanical properties and conductivity, as well as size of inter-particle sintering necks, conditions necessary for mechanical stability were determined [3]. Combination of mechanical properties with conductivity allows development of optimized microstructure. Comparison with the experimental work will be presented.

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