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## Optimization of the transport and mechanical properties of polysiloxane/polyether hybrid polymer electrolytes

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**ABSTRACT:** In this study, the thermo-mechanical properties of networked, polysiloxane/polyether-based, hybrid polymer electrolytes are optimized with the aim of enabling room-temperature operation in lithium metal-polymer batteries. The structural parameters of the electrolytes (polyether chain length, cross-linking and salt concentration) are varied in order to get the best tradeoff between conductivity and mechanical stability. The optimized material has a conductivity close to  $1.5 \cdot 10^{-4} \text{ S cm}^{-1}$  at room temperature and a shear storage modulus of 50 kPa up to 100 °C. The effect of TiO<sub>2</sub> nano-particles is also studied with the results showing an overall ambiguous effect on the materials properties. Finally, one of the materials with the highest conductivity is used as electrolyte in a Li/LiFePO<sub>4</sub> cell. This cell has good rate capability and cyclability due to the high conductivity of the electrolyte. However, the high conductivity is reached at expense of the mechanical stability and the resulting electrolyte proves to be too weak to work as an efficient barrier against lithium dendrite growth.

**KEYWORDS** Composite polymer electrolytes, inorganic-organic hybrids, lithium metal batteries

### 1. Introduction

Solid polymer electrolytes (SPEs) have many advantages over commercial liquid electrolytes. These include improved safety, low cost, mechanical stability and easy processability [1]. However, the use of polymer electrolytes has been limited by their low ionic conductivity. Complexes of lithium salts with poly(ethylene oxide) (PEO), which are the most extensively studied SPEs, have conductivities lower than  $10^{-5} \text{ S} \cdot \text{cm}^{-1}$  at room temperature [2]. An ionic conductivity higher than  $10^{-4} \text{ S} \cdot \text{cm}^{-1}$  is required for a commercial application [3]. In PEO-based polymer electrolytes, the conductivity is limited by polymer segmental motion [4], and by the partial crystallization of PEO complexes. Consequently, most of the research on polymer electrolytes attempted to suppress the crystallinity and lower the glass transition temperature, although this approach did mostly result in a deterioration of the mechanical properties.

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