

**SOLID POLYMER ELECTROLYTES FOR LITHIUM BATTERIES:
WETTING, STRUCTURE AND ADDITIVE EFFECTS**

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Solid polymer electrolytes (**SPEs**) are a potential disruptive advance in lithium battery technology. In contrast to other electrolyte systems, liquids, gelled polymers, and inorganic solids, the benefits of SPEs include relatively low volatility (increased safety), compatibility with metallic lithium (increased energy density), and relatively low-cost materials and manufacturing. The major challenge inhibiting large-scale commercialization is sluggish ionic conductivity, at least an order of magnitude below the desired $10^{-4} \text{ S cm}^{-1}$ at room temperature. Paradoxically, the feature that provides the requisite mechanical properties leading to higher cell energy density and lower cost is the same that reduces ion transport; restricted polymer chain motion. Thus, the crux of the development challenge is how to decouple mechanical properties from conductivity. Three distinct copolymer systems were studied here in an attempt to identify possible strategies to do just that: **(1)** ISO: A 3D continuous network forming block copolymer (**BCP**), where phase separated mechanical blocks and conductive blocks could independently enhance desired properties. Through this system it was discovered that preferential wetting behavior of the BCP must be engineered by electrode surface modification to prevent unwanted conductivity drops related to nonconducting blocks at the electrode surface. This should generally apply to all BCP, structure forming SPEs, opening a clear pathway to more accurately measure conductivity. **(2)** PLiMTFSI-co-PHEA-/PEG: A facile and scalable UV polymerized SPE with a single ion conductor (**SIC**), where crosslinking provides the mechanical strength, and the SIC provides the targeted conductivity. The final result was an easily modifiable platform that showed modest function during cycling and voltammetry. **(3)** Cellulose reinforced EO-co-EPI: Establishing performance in (non-hydrated) dry electrolytes revealed a future project to surface modify cellulose nanocrystals to improve stiffness and conductivity simultaneously.

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