

# Polymer Electrolytes Containing Solvate Ionic Liquids

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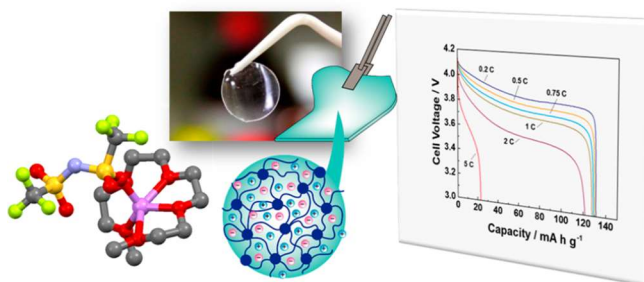
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We report ionic transport, electrochemical properties and battery performance of polymer electrolytes composed of ABA-triblock copolymers and Li-glyme solvate ionic liquids (SILs), which consist of the  $[\text{Li}(\text{glyme})]^+$  complex cation and bis(trifluoromethanesulfonyl)amide ( $[\text{TfSA}]^-$ ) anion, to simultaneously achieve high ionic conductivity, thermal stability, and a wide potential window. Three different block copolymers, consisting of a SIL incompatible A segment (polystyrene, PSt) and SIL compatible B segments (poly(methyl methacrylate) (PMMA), poly(ethylene oxide) (PEO), and poly(butyl acrylate) (PBA)) were used as the polymer matrix. The SILs were solidified with the copolymers through physical cross-linking by the self-assembly of the PSt segment. The thermal and electrochemical properties of the polymer electrolytes were significantly affected by the stability of the  $[\text{Li}(\text{glyme})]^+$  complex in the block copolymer B segments. Pulsed-field gradient NMR measurements of the polymer electrolytes and molecular dynamics simulation indicate that the  $[\text{Li}(\text{glyme})]^+$  complex cation is unstable in the PEO matrix because of the competitive coordination of the PEO chain and glyme with  $\text{Li}^+$ . On the other hand, the complex structure of  $[\text{Li}(\text{glyme})]^+$  is stable in the PMMA- and PBA-based polymer electrolytes because of the weak interaction between  $\text{Li}^+$  and the polymer chains. By use of the PMMA- and PBA-based polymer electrolytes, 4-V class Li batteries with a  $\text{LiCoO}_2$  cathode and a Li metal anode could be operated stably at 60 °C; in contrast, this was not possible using the PEO-based electrolyte.



**Figure.** Schematic illustration for polymer electrolytes containing solvate ionic liquids.

## References:

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