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Structure and mobility of PEO/LiClO4 solid polymer electrolytes

SUSAN FULLERTON, JANNA MARANAS, Penn State — Solid polymer electrolytes [SPEs] for use in rechargeable lithium-ion batteries offer many advantages over traditional liquid electrolytes, including mechanical flexibility and environmental friendliness. The practical limitation is that room temperature conductivity remains insufficient to power a portable device. While it is well-established that ion mobility is driven by polymer dynamics, high conductivity values have also been reported through fully crystalline SPEs. PEO-based SPEs have a rich phase behavior, and can form several crystalline complexes depending on the lithium concentration, temperature, and recrystallization time. We investigate the structure, mobility, conductivity, and thermal properties of both semi-crystalline and amorphous PEO/LiClO4 SPEs. Structure is measured with small-angle neutron scattering, and PEO mobility with quasi-elastic neutron scattering. We observe a decoupling of ionic conductivity and PEO mobility in a semi-crystalline sample. We also determine that PEO hydrogen atoms undergo restricted rotation on a circle. The radius of the circle is consistent with a cylindrical, crystalline structure that persists to some extent in the amorphous phase. The results suggest that directed ion transport via ordered structures is perhaps equally important as polymer mobility for increasing conductivity, provided that the structures percolate over large spatial scales.

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