Anisotropic Ion Transport Properties in Thin-Film Solid Polymer Electrolytes

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With the advent of stretchable electronics, including implantable medical devices and portable electronics, there is a strong desire to develop thin, flexible batteries for these applications. In particular, much research is being devoted to designing sustainable solid polymer electrolytes, which offer distinct advantages over conventional organic liquid electrolytes, such as enhanced flexibility and strength. The relationship between polymer microstructural changes from extreme confinement and ion transport is still elusive. Although anisotropy was previously observed in stretched bulk polymer samples, it has not yet been observed in thin films. Thus, this study investigates the effects of nanoconfinement on ionic conductivity in poly(ethylene oxide) (PEO) thin films through electrochemical characterization methods. Impedance spectroscopy was performed in both the in-plane and through-plane directions at temperatures between 25 °C and 60 °C to quantify conductive anisotropy. Using resistance values derived from equivalent circuit modeling, the average in-plane conductivity at 25 °C was determined to be 10^-7.8 S/cm. This value was about one order of magnitude greater than the through-plane conductivity at room temperature, which was estimated to be 10^-8.8 S/cm. Results also support that ionic conductivity is dependent on temperature, exhibiting Arrhenius-like behavior in both configurations. These measurements dispute the common notion that crystallinity is detrimental to conductivity in solid electrolytes, and instead suggest that charge transport pathways within the polymer film facilitate ion movement. The observed anisotropy holds significant implications for understanding similar ion conduction mechanisms in next-generation polymer-ceramic composite electrolytes.