The influence of the hybrid polymer/ceramic interface on the Li-ion transport in composite solid state electrolytes

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Solid state batteries (SSB) are considered the next step in battery technology. Advanced features of SSBs over the state-of-the-art Li-ion batteries such as enhanced intrinsic battery safety and increased energy density are of particular interest. Currently vast number of SSB related research focus on the investigations of suitable ceramic, glassy and polymer-based electrolyte materials as solid state alternatives for the liquid electrolyte. Nevertheless, ceramic-type solid state electrolytes such as $Li_7La_3Zr_2O_{12}$ (LLZO) offer good electrochemical stability against metallic lithium negative electrode and high room temperature ionic conductivity, but its processing as a thin layer into the battery cell is rather difficult. Furthermore, the rigid solid/solid interfaces in the battery cell have crucial influence on the electrochemical performance. The contact problem between the electrolyte and electrode layers could be solved if polymer composite functional materials are used. However, the ionic transport across the formed polymer/ceramic interface must be sufficient.

Our present contribution is concerned with the preparation and characterization of a solid-state composite electrolyte based on the cubic $Li_7La_3Zr_2O_{12}$ (LLZO) and poly(ethylene oxide) (PEO). LLZO powder is synthesized using a wet-chemistry synthesis route developed in our laboratory. As-synthesized LLZO powder is embedded into polymer PEO electrolyte using different LLZO volume fractions. Free standing, flexible composite electrolyte membranes are produced via tape casting technique. We investigate the temperature dependent Li-ion conductivities of the pristine components and the synthesized polymer/ceramic composites. A model system for the EIS analyses consisting of symmetrical polymer/LLZO/polymer layers is proposed, with the aim to identify and quantify the lithium ion transition resistance across the ceramic/polymer interface. According to our investigations the ionic movement across the ceramic/polymer interface needs high activation energy and above polymer's melting temperature lithium ions move rather through the individual phases.

References:

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