Optimizing Garnet-Type LLZO Electrolytes Through Solid-State Processing for Solid-State Batteries

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Abstract:

All Solid-State Batteries (ASSBs) offer significant advantages for electric vehicle and grid storage applications including enhanced safety and higher energy density compared to conventional lithium-ion batteries. Garnet-type Lithium Lanthanum Zirconium Oxide, LLZO, ($Li_7La_3Zr_2O_{12}$) is a ceramic oxide solid electrolyte candidate due to its wide electrochemical stability window (>5 V), chemical compatibility with lithium metal, and mechanical rigidity. However, processing-related microstructural variations often introduce additional grain boundary resistance, which reduces ionic transport through the material affecting the net ionic conductivity of the electrolyte. The current work systematically investigates the influence of solid-state synthesis processes on the morphology and ionic conductivity of cubic LLZO. A modified synthesis route incorporating optimized wet ball milling and subsequent thermal treatment was conducted to improve chemical reactivity, phase stabilization, densification. X-ray diffraction (XRD) pattern confirmed the formation of cubic garnet phase of LLZO. Microstructural analysis revealed particle size reduction following ball milling, indicating effective mechanical refinement and improved powder homogeneity prior to pelletization.. Electrochemical Impedance Spectroscopy (EIS), in the frequency range of 0.1 Hz–1 MHz, enabled analysis of bulk and grain boundary resistance through Nyquist plots. These findings highlight the importance of synthesis process in modifying the structural and electrochemical properties of LLZO electrolyte. They also corelates the processing–structure–transport relationships and provide practical insight for integrating LLZO electrolytes into scalable, high performance solid-state battery systems for energy storage applications.

Keywords:

All-solid-state batteries, lithium metal batteries, solid-state electrolytes, high-energy-density storage, processing optimization.