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## **Processing, Mechanics, and Electrode Structure for All-Solid State LLZO Batteries**

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LLZO ( $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ ) and related garnet materials are primary candidates for the electrolyte and electrode scaffold of all-solid state lithium batteries. LLZO is a ceramic oxide with a wide voltage stability window, good conductivity at room temperature, and chemical compatibility with a wide variety of electrode active materials. Implementation of LLZO in full cells with high energy density is challenging, however, due to Li loss during sintering, reaction with transition metal cathodes at elevated processing temperatures, high volumetric density that limits cell energy density, brittle mechanical failure, and lithium intrusion into the electrolyte during cell operation at high current density. This presentation will summarize our efforts to address these issues through advancements in processing, mechanical properties, electrode structure, and composite electrode material selection.

To enable high energy density, it is desirable to utilize a thin ( $<100\ \mu\text{m}$ ) LLZO electrolyte layer. High conductivity of the thin electrolyte is maintained by adding sacrificial lithium carbonate, which compensates for lithium evaporation during sintering. Adding MgO prevents abnormal grain growth and maintains the desired fine grain structure, leading to a  $\sim 70\%$  increase in fracture strength. Preparing a thin, flexible green ceramic sheet via tapecasting allows the electrolyte to be laminated to a porous LLZO electrode scaffold, or shaped via texturing prior to sintering. Scaffolds are prepared by high shear compaction, phase inversion, freeze tape casting, and other processing techniques based on scalable tape casting. In the case of texturing, surface features are impressed into the green LLZO tape and the additional surface area of the sintered LLZO surface decreases interfacial resistance and increases critical current density from  $0.2$  to  $0.5\ \text{mA}/\text{cm}^2$ .

Utilizing these optimized ceramic fabrication processes, solid state cells are prepared with  $\sim 80\ \mu\text{m}$  thick LLZO electrolyte, no added liquid electrolyte, and no exogenous pressure. A catholyte comprising organic ionic plastic crystal and a mixture of lithium salts is melt-processed to achieve good wetting and interfacial contact between the rigid LLZO and NMC cathode active material. This cell design achieves  $\sim 180\ \text{mAh}/\text{g}$  and stable cycling for  $>100$  cycles at room temperature. Efforts to scale up the cell size and improve rate capability will also be discussed.