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Optimization of Lithium Content and Sintering additives in Tape Cast Lithium Garnet Electrolyte Sheets

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The lithium garnet (LLZO) remains one of the most promising electrolyte materials for producing solid state batteries. Tape casting is one method for producing the thin ceramic electrolyte layer required for a high-performance solid state battery. Processing of LLZO is complicated by several factors including lithium volatilization, abnormal grain growth and phase instability. Many of these issues are exacerbated in tape cast sheets by the sheets' higher surface area to volume ratio, when compared to pellet processing methods. We report on an environmentally friendly aqueous tape casting process for LLZO using methylcellulose as a binder and compare final sheet properties to other, solvent-based, options. Solids loading, binder content, dispersant composition, and wetting agent type are varied to improve ease of casting and enhance properties of the as-cast tape. Lithium content in sintered LLZO sheets is controlled by the inclusion of excess Li₂CO₃ as lithium source in the ceramic tape itself and by a novel method of producing a lithiumsaturated furnace atmosphere. Sintering time and temperature are optimized, and rapid thermal processing is investigated as a method of reducing lithium loss. AI_2O_3 and MgO are examined for use as sintering additives. Characterization of LLZO sheets is performed with XRD, EIS and SEM. Li₂CO₃ and sintering additive levels are optimized for ionic conductivity and density. The optimization effort improved the conductivity and density of $100\mu m$ thick sintered sheets to greater than $3x10^{-4}$ S/cm conductivity and 90% density.