

Impact of sintering aids on the Li ion conductivity of $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ solid electrolyte

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Abstract: One of the most promising concepts for new energy storage solutions are all solid state batteries (ASSB). Thereby, higher capacities and increased safety are achieved by replacing the flammable, liquid electrolyte in conventional cells with a solid electrolyte. In principle, three classes of materials are considered as Li ion conducting solid electrolytes: polymers, sulfides and oxides. Polymer electrolytes are typically characterized by simple processing and scalable production. However, they offer relatively poor conductivity of $<10^{-4}$ S/cm. By comparison, sulfides exhibit high conductivities of $>10^{-3}$ S/cm, but require processing in inert atmosphere without oxygen and moisture, and high pressure during battery operation. Oxides are characterized by fairly good ionic conductivities $<10^{-3}$ S/cm and straightforward scalability and processability in air. However, in order to achieve the desired conductivity, densification in a sintering step is required under high temperatures of about 1000 °C. A major goal for preparing ASSB composite electrodes is reducing the sintering temperatures below 600 °C to avoid reactions between the solid electrolyte and the cathode material.

In this study, the promising solid electrolyte $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$ (LATP) is used to show how the sintering temperature can be significantly reduced with sintering aids, while still achieving conductivities comparable to the pure material sintered at high temperature. The sintering behaviors are characterized by optical dilatometry and sintering of pressed samples. The microstructure and phase composition are analyzed by SEM and XRD. The Li ion conductivity of sintered bodies are determined by impedance spectroscopy. With Li_2CO_3 as sintering additive the sintering temperature can be reduced to ~800 °C, while achieving high ionic conductivity of up to 3.6×10^{-4} S/cm. These results are an important step towards enabling composite cathodes for future ASSB based on LATP oxide electrolyte.

Keywords: Li-ion conductive ceramic; solid electrolyte; Li-ion batteries; all-solid-state batteries; LATP