All-Solid-State Li-ion Batteries Based on Garnet-Type Fast Li-ion Conductor $Li_7La_3Zr_2O_{12}$

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Solid oxide electrolytes for Li-ion batteries offer many advantages in comparison to their liquid electrolyte battery counterparts, such as a higher safety, a much higher electrochemical and thermal stability window, and higher chemical stability with a variety of different electrode materials. They have therefore the potential to enable higher energy densities of Li-ion batteries in the future. The Li-stuffed garnet-type oxide $Li_7La_3Zr_2O_{12}$ (LLZO) is the most promising solid oxide electrolyte at the moment because it offers high Li-ion conductivity and a large electrochemical stability window over a wide potential range. LLZO is mostly synthesized by standard solid-state reactions, requiring relatively high temperatures. Here a novel modified sol-gel combustion method is presented that greatly simplifies processing and lowers processing temperatures of LLZO powder with composition $Li_{6.25}Al_{0.25}La_3Zr_2O_{12}$. Electrolyte pellets pressed from this powder show high relative densities and ionic conductivities after sintering. All-solid-state batteries based on LLZO were so far only tested with cathode materials, and reasonable electrochemical activities were only obtained for cathodes deposited by thin film deposition. Here, the anode material $Li_4Ti_5O_{12}$ is deposited directly on sintered electrolyte pellets using drop casting of a composite electrode slurry. The important role of the solid electrolyte–electrode interface is investigated, and it is shown how to decrease resistance values and to improve charge–discharge capacities by interface-modification. It is revealed how a more intimate embedding of the electrode into the electrolyte by means of a porous interface enhances delivered capacities and cycling properties of all-solid-state Li-ion batteries based on garnet-type LLZO electrolytes.