Ultrafast-sintered self-standing LLZO membranes for high energy density Li-garnet solid-state batteries

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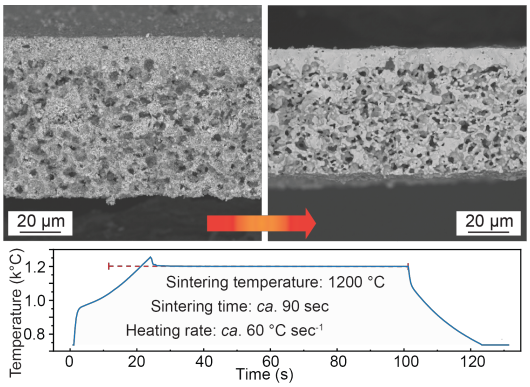
Towards building non-flammable and temperature-tolerant Li-ion batteries with high energy density, $Li_7La_3Zr_2O_{12}$ (LLZO) has recently attracted considerable attention as a compelling solid-state Li-ion electrolyte (SSE) due to its high thermal stability, Li-ion conductivity of up to 1 mS cm^{-1} (RT), and a wide electrochemical operation window of 0–6 V *vs*. Li^+/Li.[1, 2] Additionally, unlike other Li-ion soft solid conductors such as those based on sulfides, LLZO SSE can be manufactured with a bilayer dense-porous microstructure, which prevents the issues of dynamic volume change of the Li anode and the formation of voids at the Li/SSE interface during cycling and thus eliminates the need for external pressure.[3, 4] Here we report a facile, ultrafast sintering methodology for the fabrication of LLZO solid-state electrolyte in the form of self-standing bilayer dense-porous LLZO membranes. The thickness of the produced dense and porous layers was *ca*. 8 µm and 55 µm, which hypothetically allows to achieve high gravimetric and volumetric energy densities of Li-garnet batteries of 218 Wh kg^{-1} and 769 Wh L^{-1} (in combination with an NMC811/LiFSI-Pyr_{13}FSI cathode). Electrochemical measurements confirmed that produced LLZO membranes possess high critical current density up to 1.7 mA cm^{-2} and cycling stability of over 160 cycles at a current density of 0.4 mA cm^{-2}.



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