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Multilayer hybrid solid-state electrolyte membrane for the high rate and long-life cycle performance of lithium-metal batteries

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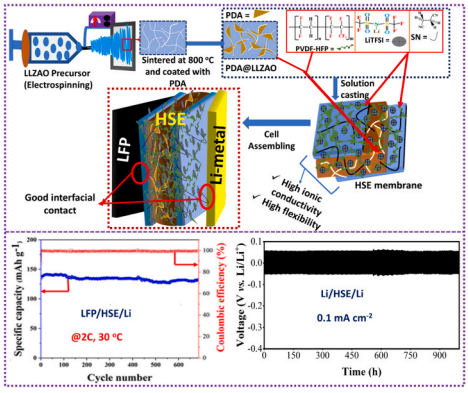
HIGHLIGHTS

- Interconnected LLZAO ceramic filler modified with PDA (PDA@LLZAO) was used to build HSE.
- HSE membrane exhibited a maximum ionic conductivity of 8.72 $\times 10^{-4}$ S cm^{-1} at 70 °C.
- $LiFePO_4$/HSE/Li coin-type cell using 10 mL of liquid electrolyte showed long-term cycling stability.
- HSE membrane displayed comparatively lower total exothermic heat generation.

GRAPHICAL ABSTRACT



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ABSTRACT

Hybrid solid-state electrolytes (HSEs) can be used to increase the electrochemical performance of lithium-metal batteries (LMBs), while also suppressing dendrite formation and preventing flammable behavior and electrolyte leakage, which are frequently present in conventional organic-liquid electrolytes. Notably, multilayer HSE membranes have received increasing emphasis since they can significantly ameliorate the interface contact toward electrodes and the mechanical strength. In this current work, we fabricated multilayer HSE membranes via a solution-casting technique that incorporated poly(vinylidene fluoride–co–hexafluoropropylene) (PVDF-HFP), polydopamine-modified $Li_{6.28}La_3Zr_2Al_{0.24}O_{12}$ (PDA@LLZAO) filler, succinonitrile (SN) and lithium bis(trifluoromethane sulfonyl)imide (LiTFSI). The resulting HSE membrane exhibited a high ionic conductivity (2.49 $\times 10^{-4}$ S cm^{-1} at 30 °C), transference number of 0.65, and excellent electrochemical window (4.80 V). The

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