A modified trilayer membrane for suppressing Li dendrite growth in all-solid-state lithium-metal batteries

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ABSTRACT

Mechanically robust trilayer HSE membrane modified with an optimized amount of functionalized vapor-grown carbon fiber (f-VGCF) is prepared using a solution-casting method. This trilayer membrane had the structure SPE0 (PVDF-HFP/LiTFSI/SN)|SPE2 (PVDF-HFP/Al-doped-LLZO/LiTFSI/SN)|SPE1 (PVDF-HFP/LiTFSI/SN/f-VGCF). The SPE0 (unmodified) and SPE1 (modified) layers faced the cathode and anode sides, respectively; SPE2 is the same as SPE0 but loaded with needle-like $Li_{6.25}Al_{0.25}La_3Zr_2O_{12}$ filler. The modified trilayer HSE membrane possessed an ionic conductivity (4.72×10^{-4} S cm^{-1}) and a Li transference number (0.52) higher than unmodified trilayer HSE membrane (3.68×10^{-4} S cm^{-1} and 0.41, respectively). A Li/SPE1-SPE2-SPE1/Li symmetric cell displayed extremely stable Li plating/stripping performance when tested sequentially at 0.1, 0.2, 0.5, and 1 mA cm^{-2} for 800 h (200 h for each current density). A 2032 coin cell incorporating NCM622/SPE0-SPE2-SPE1/Li delivered a desirable capacity output with excellent cycling stability (85.04% capacity retention after 627 cycles with an average coulombic efficiency of 99.17% at a rate of 0.5C at room temperature). A pouch cell based on NCM622/SPE0-SPE2-SPE1/Li also achieved excellent capacity retention of 95.37% after 100 cycles with an average coulombic efficiency of 99.81% at 0.2C at room temperature. Thus, ASSLMBs incorporating such modified trilayer HSE membranes have potential for practical applications.

KEYWORDS

All-solid-state lithium-metal batteries; Modified trilayer HSE membrane; f-VGCF; NCM622 cathode material; Needle-like filler

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