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Supporting information

3D Polyacrylonitrile nanofibers and flexible Polydimethylsiloxane macromolecule combinedly all-solid-state composite electrolyte for efficient lithium metal batteries

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Abstract

All-solid-state polymer electrolytes have received widespread attention due to their superior safety over liquid electrolytes that are prone to leaks. However, poor ionic conductivity and uncontrollable lithium dendrite growth have greatly limited the quick development of polymer electrolytes. Hence, we report a composite polymer electrolyte combining the polyacrylonitrile (PAN) electrospun fiber membrane, flexible polydimethylsiloxane (PDMS) macromolecule and polyethylene oxide (PEO) polymer. The introduction of PDMS with a highly flexible molecular chain, ultra-low glass transition energy and high free volume can help optimize lithium ion migration paths and improve the interface compatibility between the electrolyte and the electrode. In addition, the nano-network structure of the PAN nanofiber membrane can promote the interaction between adjacent polymer molecular chains and improve the mechanical property of the composite electrolyte to suppress the lithium dendrites growth. The synergistic effect of the PDMS and PAN electrospun nanofiber membranes endows the composite electrolyte superior ionic conductivity and excellent electrochemical stability with lithium metal. The interface impedance of the Li/Li symmetric battery with the composite electrolyte after 15 days of continuous standing has no significant change compared with the initial state, and the battery can maintain stable cycling for 1200 h without short circuit under the dynamic current of 0.3 mA cm^{-2}. The obtained composite polymer electrolyte has potential application prospects in the field of high-energy lithium metal batteries.

Keywords

Polydimethylsiloxane; Nano-network structure; Composite polymer electrolyte; Superior ionic conductivity; Excellent interface compatibility; All-solid-state battery

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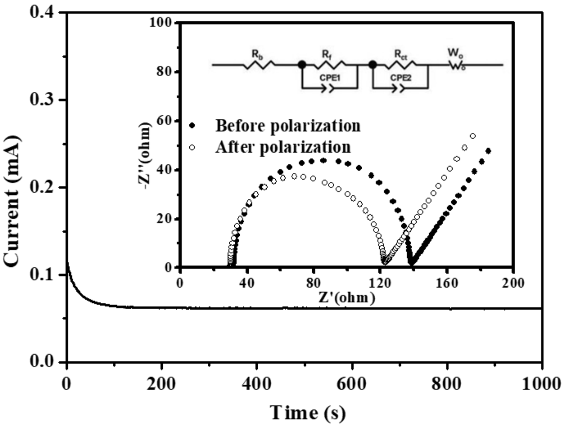

Fig. S1 Chronoamperometry profile of Li/Li symmetric battery with PEO-LiTFSI-PAN/PDMS-0 composite electrolytes, inset: the Nyquist impedance spectra of the battery before and after polarization.

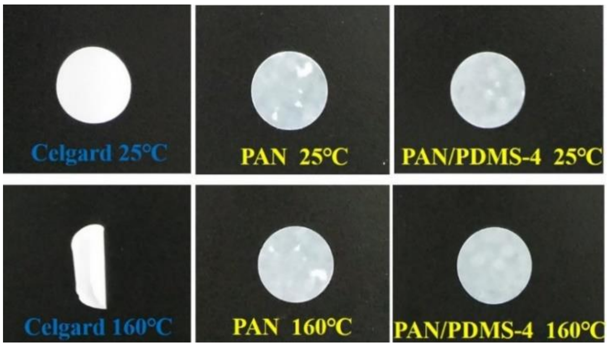

Fig. S2 Optical photos of the commercial Celgard separators (left), PEO-LiTFSI-PAN/PDMS-0 composite electrolyte (middle) and PEO-LiTFSI-PAN/PDMS-4 composite electrolyte (right) before and after heating at 160 °C for 1 h.

Table S1. The characterization data for the phase behavior of all-solid-state electrolytes.

Electrolytes	T_m/℃	Δ H_m/J g^{-1}	χ_c/%
PEO-LiTFSI	58.3	80.7	39.8
PEO-LiTFSI-PAN/PDMS-0	47.3	17.2	10.0
PEO-LiTFSI-PAN/PDMS-2	43.2	13.9	9.7
PEO-LiTFSI-PAN/PDMS-4	41.0	6.2	5.1
PEO-LiTFSI-PAN/PDMS-6	57.2	19.3	17.9

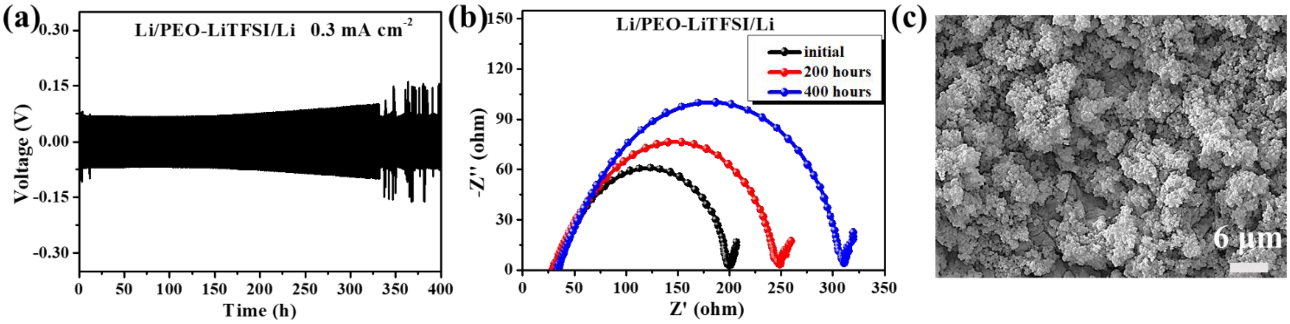

Fig. S3 Electrochemical performance of the Li/PEO-LiTFSI/Li symmetric battery under 0.3 mA cm^{-2} and 60 °C. **(a)** Galvanostatic plating/stripping profiles. **(b)** Electrochemical impedance spectra before and after different cycle time. **(c)** SEM image of lithium metal anode obtained from the battery after cycling.