A Novel Polymer Electrolyte with High Elasticity and High Performances for Lithium Metal Batteries

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Experimental

Firstly, DOL and 1,2-dimethoxyethane (DME) were mixed in 1:1 (v/v). Then, TPU was added into the above solution. Finally, after the completely dissolving of TUP, LiFSI (0.5 mol L⁻¹) was added to the above solution. The polymer electrolyte was labeled as PLEI. The preparation process is display in Figure S1. Here, LiFSI can not only trigger DOL to initiate ring-opening polymerization reaction, but also provide ion transport for the polymer electrolyte.



Figure S1. Diagram of the preparation process of the polymer electrolyte



Figure S2. Mechanical properties of the polymer electrolyte

The pattern of PLEI was tested using X-ray diffractometer (XRD, Ultima, IV) with Cu Ka radiation ($2\theta = 10^{\circ} \sim 60^{\circ}$, 4 ° min⁻¹). The scanning electron microscope (SEM, Bahens, S-3400N) was used to analyze the morphology of PLEI. The Fourier-transform infrared spectroscopy (FTIR, Nicolet iS 50 ART) and Nuclear Magnetic Resonance (NMR, Ascend 400 MHz with DMSO) were used to analyze the chemical structures of PLEI.

The ionic conductivity of PLEI was tested using the AC impedance method (CHI-760E) and the symmetric cell (stainless-steel/PLEI/stainless-steel) at different temperatures. The linear sweep voltammetry (LSV) measurement was applied to investigate the electrochemical stability of PLEI with a scan rate of 0.1 mV s⁻¹ for Li|PLEI|stainless-steel cell. AC impedance and potentiostatic DC polarization were used to measure the lithium ions transference number of PLEI. Lithium metal was used as the working electrode and counter electrode. In the potentiostatic DC polarization test, the polarization voltage was 10 mV. In the AC impedance test, the amplitude was 5 mV, and the impedance frequency was 0.01-10⁶ Hz. Then, the stability of PLEI with lithium metal was evaluated in the symmetric cell (Li/PLEI/Li). The Xinwei battery-testing equipment (Shenzhen, China) was employed to evaluate the electrochemical performances of the Li/PLEI/LiFePO₄ batteries within a voltage range of 3 to 4 V.



Figure S3. FTIR curves of PLEI and the pristine polymers



Figure S4. a) ¹H NMR spectrum of DOL and PLEI, and b) ¹³C NMR spectrum of DOL and

PLEI



Figure S5. The potential profile of Li/electrolyte/Li symmetric cell (0.5 mA cm⁻², 25 °C)



Figure S6. SEM images of a) fresh metal lithium, b) lithium surface in Li/LE/Li symmetric

cell after cycling, and c) lithium surface in Li/PLEI/Li symmetric cell after cycling

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