Fast self-healing solid polymer electrolyte with high ionic conductivity for Lithium metal batteries

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Experimental details

Materials. Thermoplastic polyurethane (TPU, yantaiwanhua, 1190A): 10 wt%, dissolving in anhydrous DMSO (Mw=106, Aldrich); amino terminated poly(dimethylsiloxane) (N-PDMS, Mw=1,500, Aldrich); Isophorone diisocyanate (IPDI, Aldrich); Chloroform (AR, Aldrich);

Liquid electrolytes: 2M $LiClO_4$ (or LiTFSI or $LiBF_4$) in diethyl carbonate/dimethyl carbonate/ethylene carbonate with a ratio of 1:1:1(v/v/v)**.**

Synthesis. The self-healable solid polymer electrolytes (SHSPE) were prepared through the condensation of N-PDMS, TPU and IPDI in hot chloroform solutions (~80 °C) with different liquid electrolytes. In the condensation reaction, the mass ratio of N-PDMS, TPU and IPDI is 20:1:1. The volume ratio of the liquid electrolytes and the chloroform is 1:5. After being condensational polymerized for about 12 h, the above solution was cooled down naturally. Then the SHSPEs were obtained by drop-casting the above cooled solution on the glass tablets with clean surfaces and kept in an oven (~ 100 °C) for 1h in the super dry room (the dew point temperature < -70 °C) for the later tests.

Materials Characterization. The morphology of the films was observed by scanning electron microscopy (SEM JEOL 6701F). The structure of the films was studied by Fourier transform infrared spectroscopy (FT-IR, 400-4000 cm^{-1}) and powder X-ray diffraction (XRD). The thermal stability of the polymer film was studied by thermogravimetric analysis (TGA, 30 to 750 ^oC at 10 ^oC min^{-1}) and differential scanning calorimetry (DSC, -160 to 100 ^oC at 10 ^oC min^{-1}).

Electrochemical Tests. The ionic conductivity and activation energy of the polymer membrane were calculated from the electrochemical impedance spectroscopy (EIS) of the stainless-steel (SS)/SHSPE/SS model in the frequency range of 0.1~1 M Hz. According to the Eq. (C): $\sigma = h/SR$, the σ of the SHSPE can be obtained. h (cm) is thickness of the SHSPE, S (cm^2) is the contact area (Φ = 1.25 cm) between the stainless steel and the SHSPE, R (Ω) is the impedance measured by EIS). According to the Arrhenius Eq. (D): $\sigma = A \exp(-Ea/RT)$, (A: frequency factor, R: molar gas constant, T: absolute temperature) Ea is obtained. The electrochemical stability of SHSPE was determined by linear sweep voltammetry (LSV) with SS/SHSPE/Li as the model.

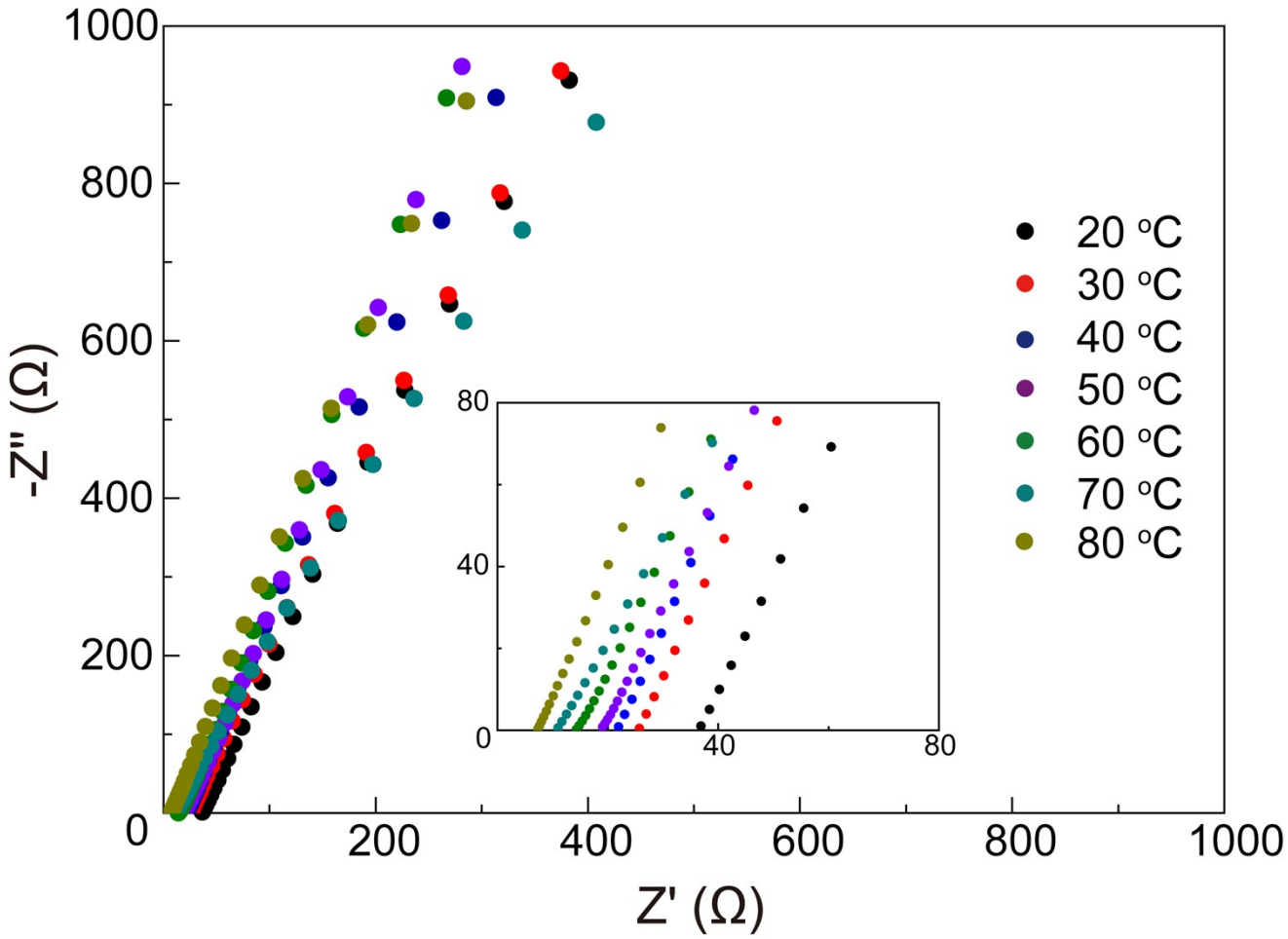

Figure S1 EIS measurements of the SHSPE ranging from 20 to 80 °C.

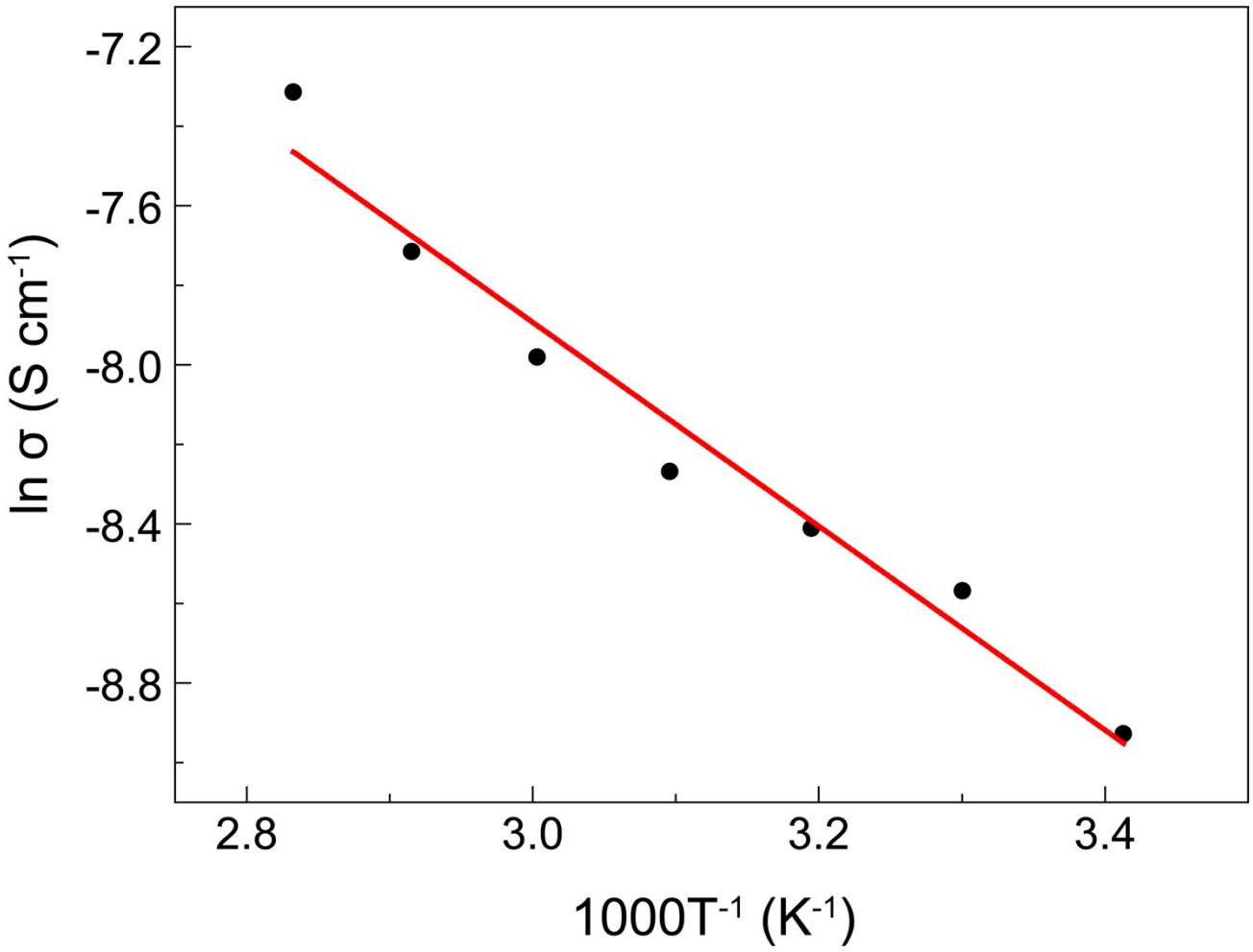

Figure S2 Log plot and linear fit of SHSPE σ *vs.* temperature ranging from 20 °C to 80 °C.