

Electronic Supplementary Material

Fabricating ion-conducting channel in SU-8 matrix for high-performance patternable polymer electrolytes

Tianzhao Li[§], Xuelei Pan[§], Zhongzhuo Yang, Fang Liu, Kesong Yu, Lin Xu (✉), and Liqiang Mai (✉)

State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China

[§] Tianzhao Li and Xuelei Pan contributed equally to this work.

Supporting information to <https://doi.org/10.1007/s12274-022-4751-2>

Experimental section

Materials

SU-8 2014 (Micro Chem), poly(ethylene oxide) (PEO, Mw=600,000, Aladin), bis(trifluoromethane)sulfonimide lithium salt (LiTFSI, >99.9%, Aladin), anhydrous Acetonitrile (ACN, >99.8%, Aladin), propylene glycol monomethyl ether acetate, (PGMEA, >99.7%, Jianghuamem).

Preparation of Electrode

Amorphous silicon (a-Si) was deposited by DC magnetron sputtering (PD-280, Wuhan PDVACUUM) onto copper foils with a thickness of 50 μm . PSU-8 was spin-coated and fabricated onto a-Si films with 1.44 μm thick. LiFePO_4 (LFP) is chosen as the cathode material. LFP, Super P and polyvinylidene fluoride (PVDF) are mixed evenly in N-methyl-2-pyrrolidone (NMP) at a mass ratio of 8:1:1 to form a uniform slurry, and then coated on a clean aluminum foil. Then the mixture is dried at 70 $^{\circ}\text{C}$ in a vacuum oven for 12 h.

Address correspondence to Liqiang Mai, mlq518@whut.edu.cn; Lin Xu, linxu@whut.edu.cn

The measurement of ionic conductivity

The bulk resistance (R_b) of polymer electrolytes was obtained from the EIS. The ionic conductivity was calculated from Equation (S1):

$$\sigma = \frac{L}{R_b S} \quad (1)$$

where R_b is the bulk resistance of polymer electrolytes, L is the thickness of the polymer electrolytes and S is the area of polymer electrolytes.

The computational formula of the activation energies (E_a)

The E_a of polymer electrolytes in Figure 3b was calculated by Arrhenius relation from Equation (S2):

$$\sigma = A \exp\left(-\frac{E_a}{kT}\right) \quad (2)$$

Where σ is ionic conductivity of polymer electrolytes, A is pre-exponential factor, E_a is activation energy, k is Boltzmann constant, and T is temperature of testing process.

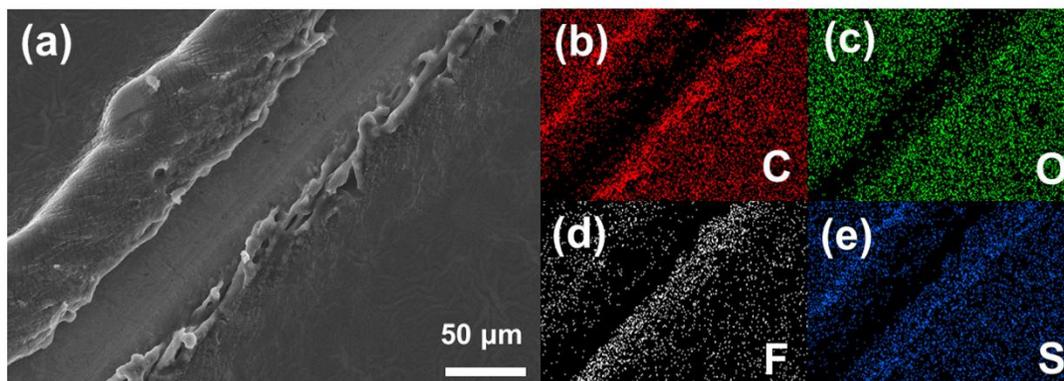


Figure S1 SEM image (a) and Energy Dispersive Spectrometer image (b-e) of PSU-8.

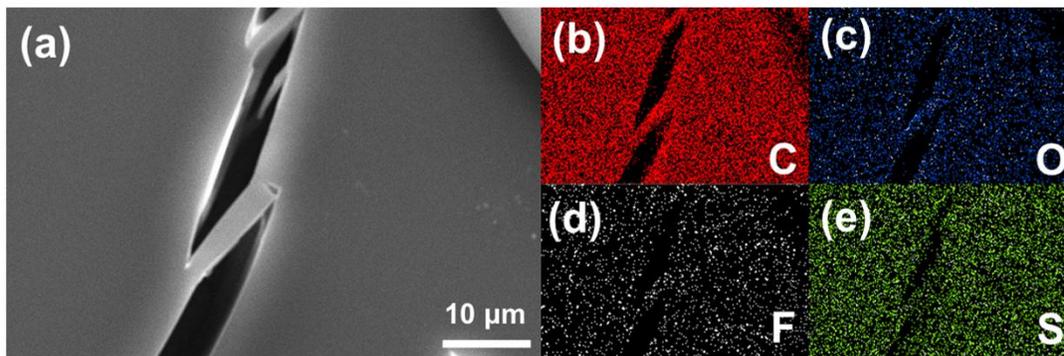


Figure S2 SEM image (a) and Energy Dispersive Spectrometer image (b-e) of SU-8.

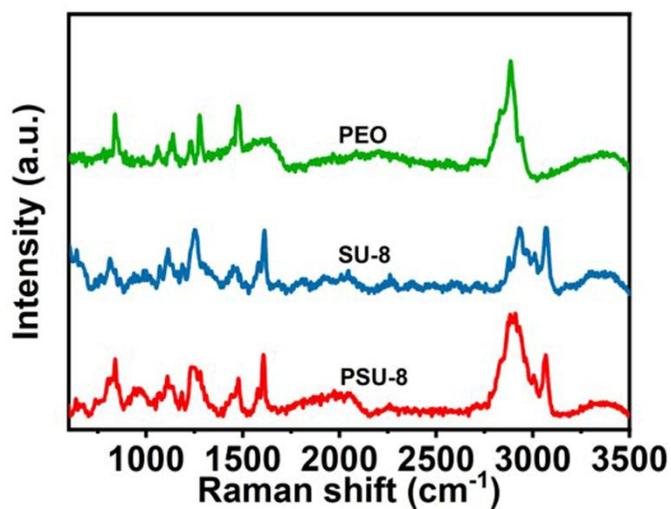


Figure S3 Raman spectra of PEO, SU-8 and PSU-8.

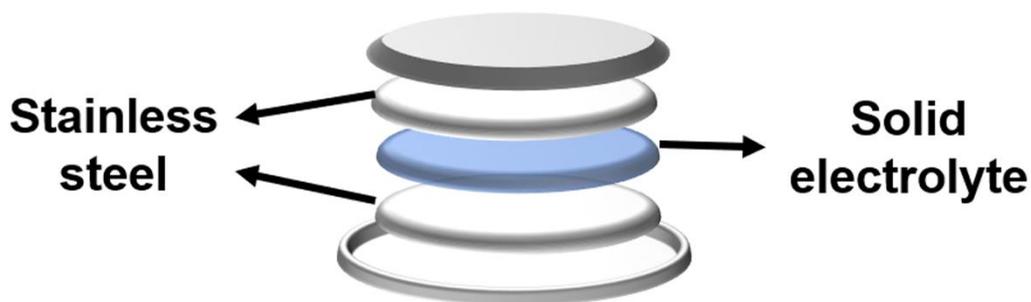
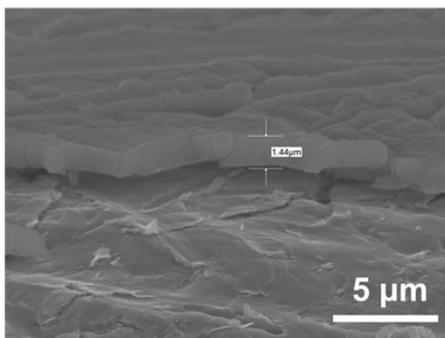


Figure S4 Schematic illustration of SS|PSU-8|SS half-cells.

(a)



(b)

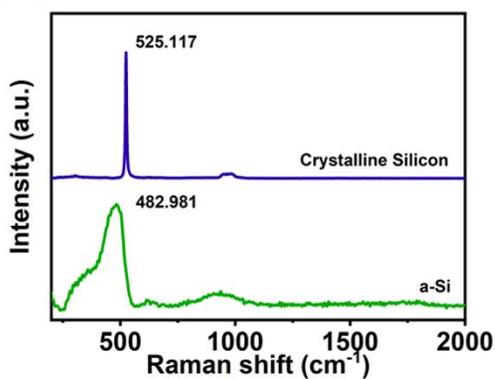


Figure S5 (a) Cross-sectional SEM image of a-Si film. (b) Raman spectra of crystalline silicon and a-Si.

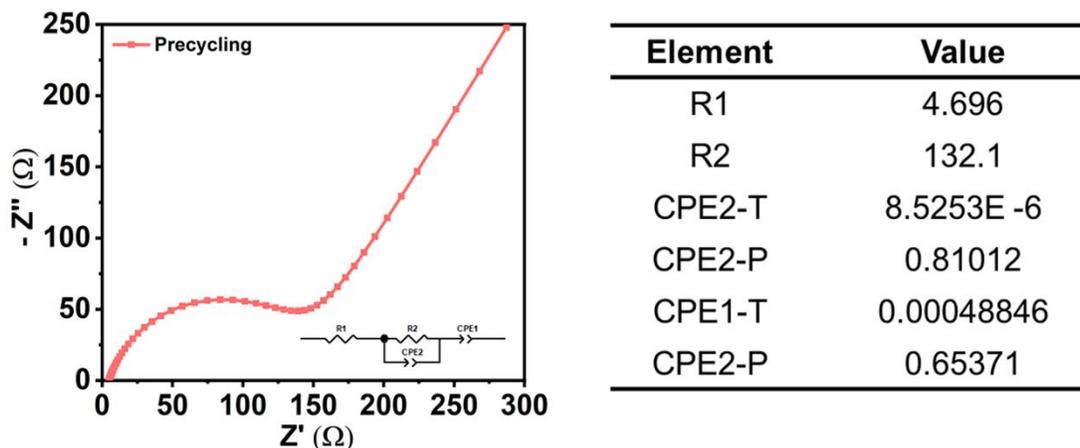


Figure S6 The fit curve of impedance spectroscopy and fitted values of a-Si|PSU-8|Li half-cells for precycling.

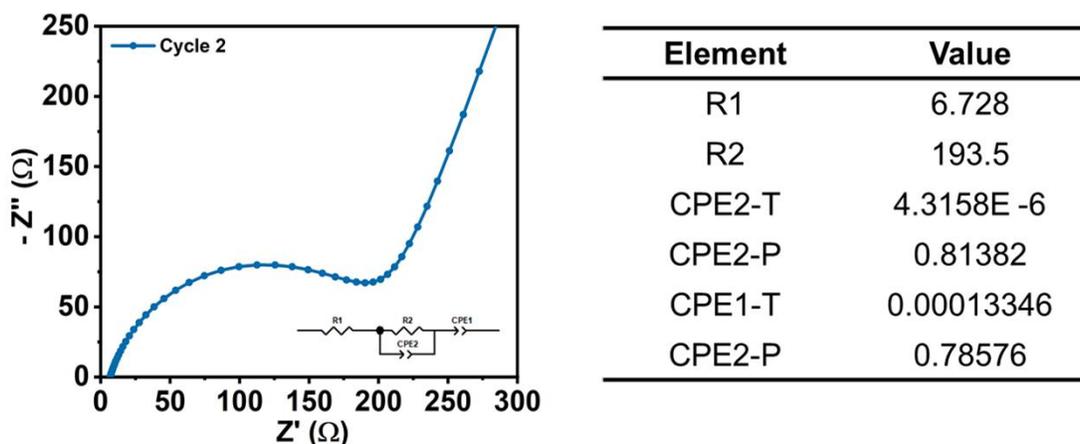


Figure S7 The fit curve of impedance spectroscopy and fitted values of a-Si|PSU-8|Li half-cells for the second cycle.

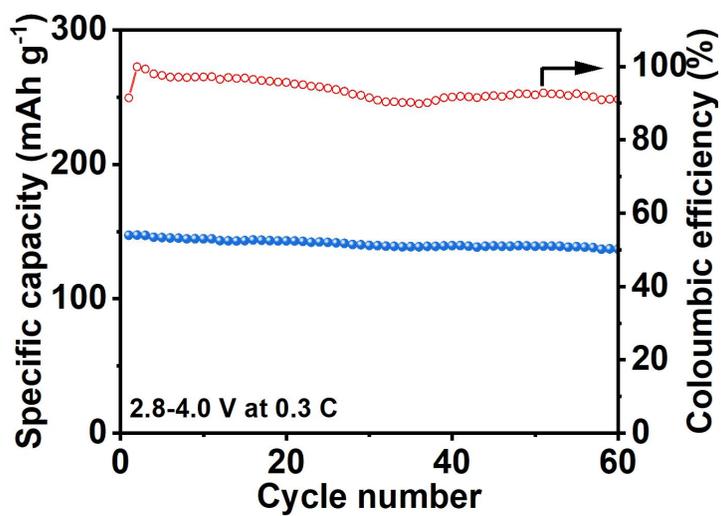


Figure S8 The long-term cycle performance of the $\text{LiFePO}_4|\text{PSU-8}|\text{Li}$ battery at a rate of 0.3 C.

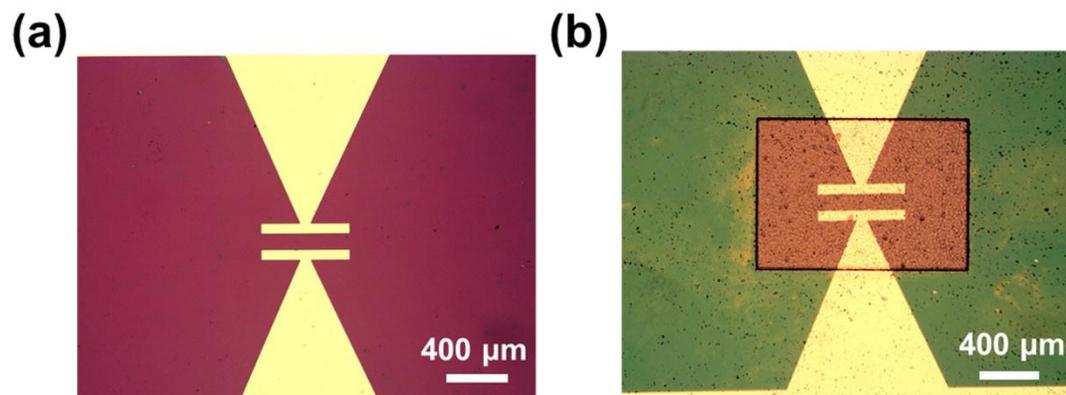


Figure S9 (a) Optical image of Au electrodes. (b) Optical image of Au electrodes with PSU-8.

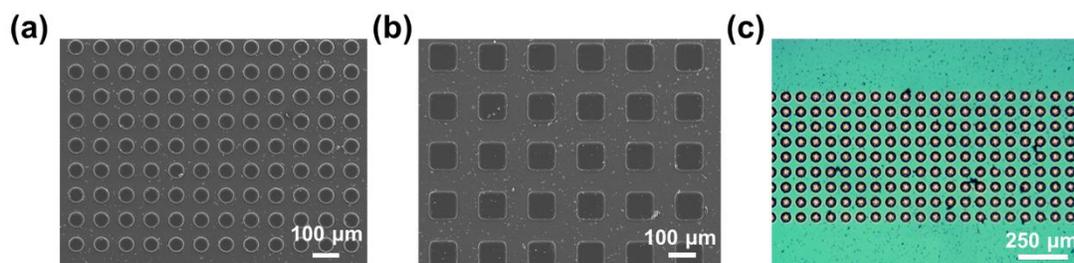


Figure S10 (a) SEM image of PSU-8 with features: circular, 100 μm in diameter. (b) SEM image of PSU-8 with features: square, 100 μm in length. (c) Optical image of PSU-8 with features: circular, 50 μm in diameter.

Table S1 The comparisons of this work and previous reports in different polymer electrolytes.

Polymer electrolytes	Ionic conductivity (S cm ⁻¹)	Elastic Modulus	Ref.
PSU-8	2.9×10^{-4}	1.52 GPa	This work
Crosslinked PEO/ SiO ₂ aerogel network/ LiTFSI/SCN	$\sim 6 \times 10^{-4}$	0.43 GPa	S1
SU-8/LiClO ₄	5.2×10^{-5}	4.2 GPa	S2
Poly (propylene oxide)/ PEO-tethered SiO ₂ /LiTFSI/PC	$\sim 5 \times 10^{-3}$	~ 1.0 MPa	S3
PEO/PEG/Octa-POSS/ LiTFSI	$\sim 1.3 \times 10^{-5}$ (30 °C)	~ 2.98 MPa	S4
PEO / Vermiculite sheets/LiTFSI	1.2×10^{-3} (60 °C)	35.4 MPa	S5

Supplementary References

- [1] Lin, D.; Yuen, P. Y.; Liu, Y.; Liu, W.; Liu, N.; Dauskardt, R. H.; Cui, Y. A silica-aerogel-reinforced composite polymer electrolyte with high ionic conductivity and high modulus. *Adv. Mater.* **2018**, *30*, 1802661.
- [2] Choi, C. S.; Lau, J.; Hur, J.; Smith, L.; Wang, C.; Dunn, B. Synthesis and properties of a photopatternable lithium-ion conducting solid electrolyte. *Adv. Mater.* **2018**, *30*, 1703772.
- [3] Choudhury, S.; Mangal, R.; Agrawal, A.; Archer, L. A. A highly reversible room-temperature lithium metal battery based on crosslinked hairy nanoparticles. *Nat. Commun.* **2015**, *6*, 10101.
- [4] Zheng, Y.; Li, X.; Fullerton, W. R.; Li, C. Y. Decoupling the modulus and toughness effects of solid polymer electrolytes in all-solid-state lithium batteries. *ACS Appl. Energy Mater.* **2021**, *4*, 14093-14101.
- [5] Tang, W.; Tang, S.; Zhang, C.; Ma, Q.; Xiang, Q.; Yang, Y. W.; Luo, J. Simultaneously enhancing the thermal stability, mechanical modulus, and electrochemical performance of solid polymer electrolytes by incorporating 2d sheets. *Adv. Energy Mater.* **2018**, *8*, 1800866.