

## Supplementary Information

### **Propanesultone-based polymer electrolyte for high-energy solid-state lithium battery with lithium-rich layered oxides**

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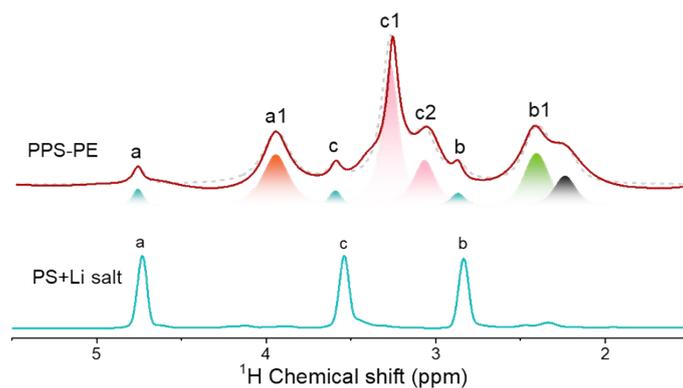
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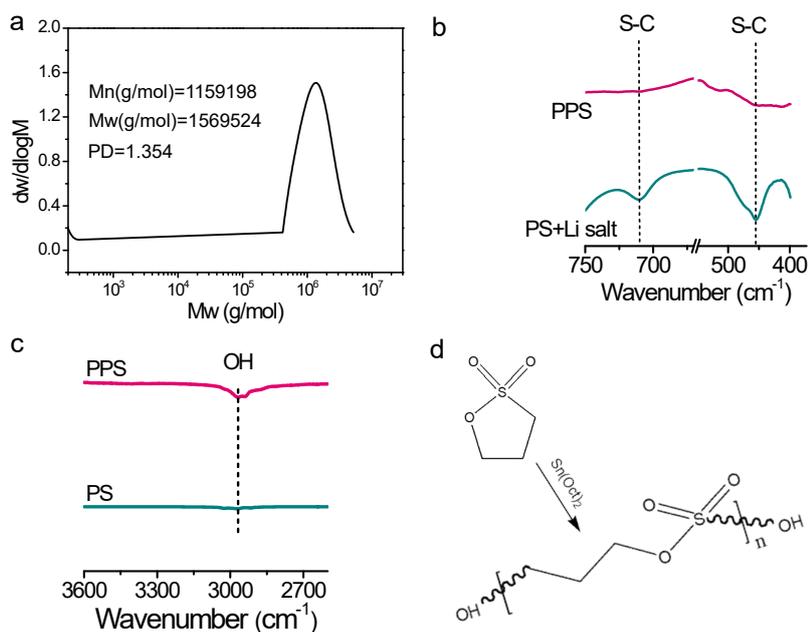
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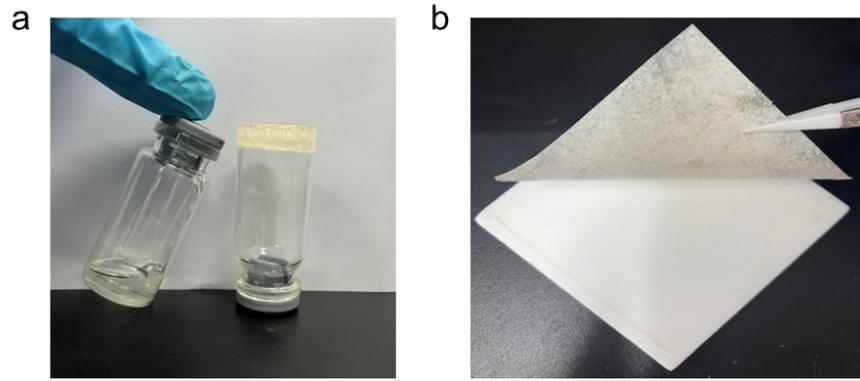
<sup>1</sup> These authors contributed equally to this paper.



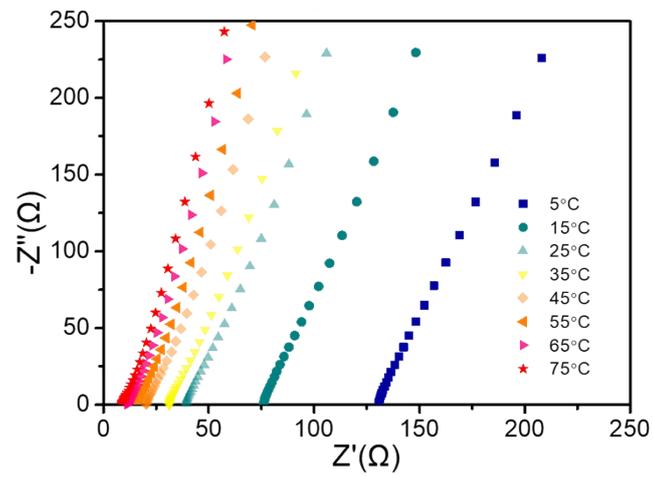
**Fig. S1** The  $^1\text{H}$  spectra and simulations for percent conversion of PPS-PE from PS monomer. The weak peaks in the range of 2.2~2.5 ppm can be indexed to the impurities.



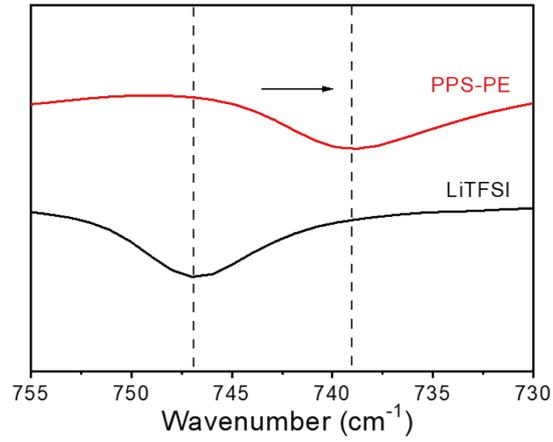
**Fig. S2** a, The GPC test of PPS-PE. b, c, The S-C and -OH bonds in FT-IR spectra of PS+Li salt precursor and PPS-PE. d, The chain-like structure of PPS-PE form PS monomer by  $\text{Sn}(\text{Oct})_2$  catalyzation.



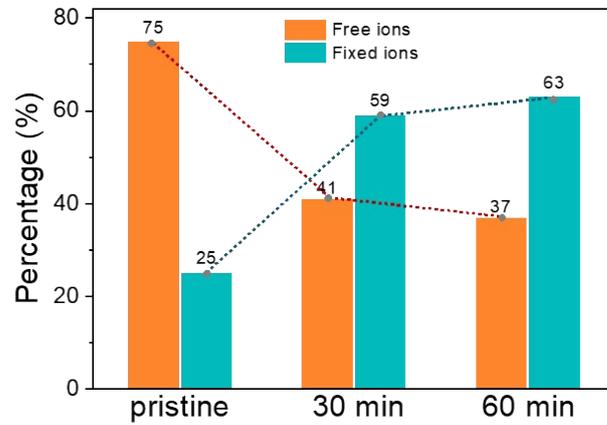
**Fig. S3** a, The photos of PPS-PE before and after polymerization. b, The flexible PPS-PE film.



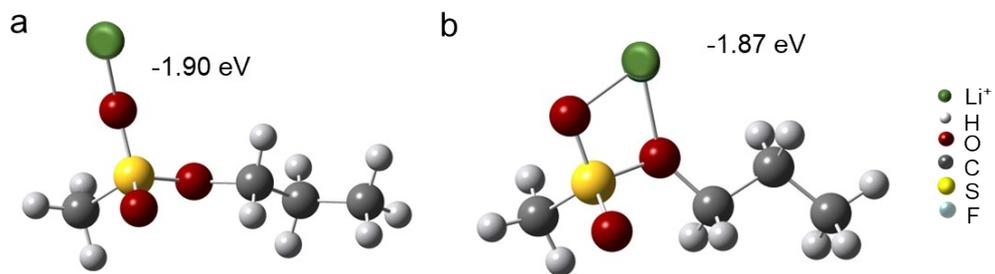
**Fig. S4** The EIS of SS/PPS-PE/SS symmetric cell at different temperatures.



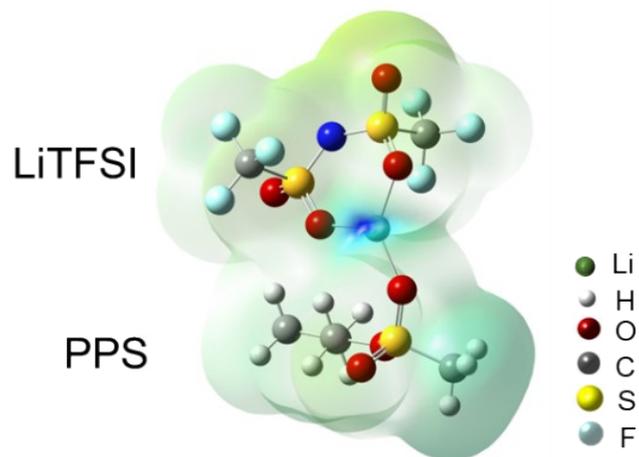
**Fig. S5** The FT-IR spectra of Li salt and PPS-PE.



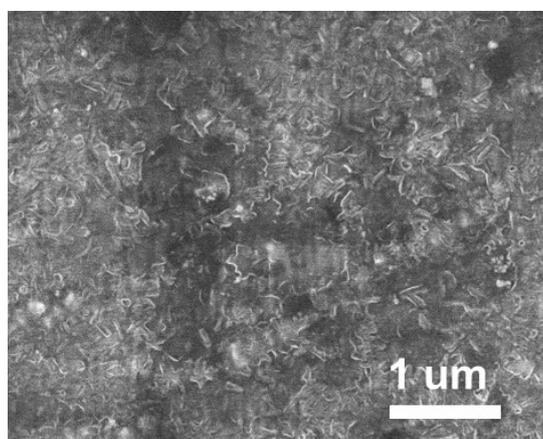
**Fig. S6** The changes of area ratios of free and fixed ions at different states.



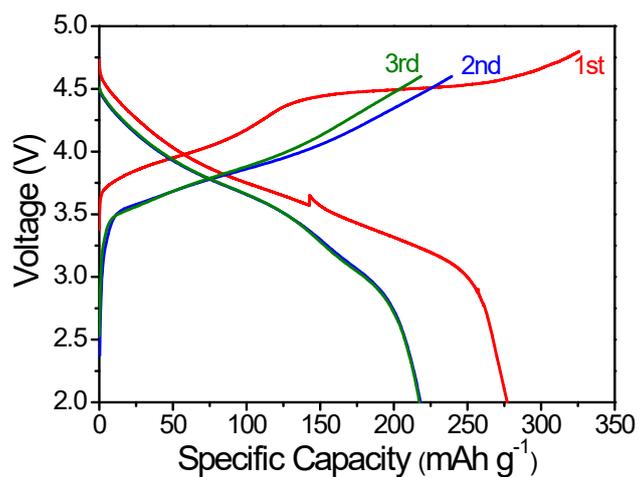
**Fig. S7** The configurations and binding energies between the free Li ion and a, the O atom on S=O group, b, the O atoms on S=O and S-O groups.



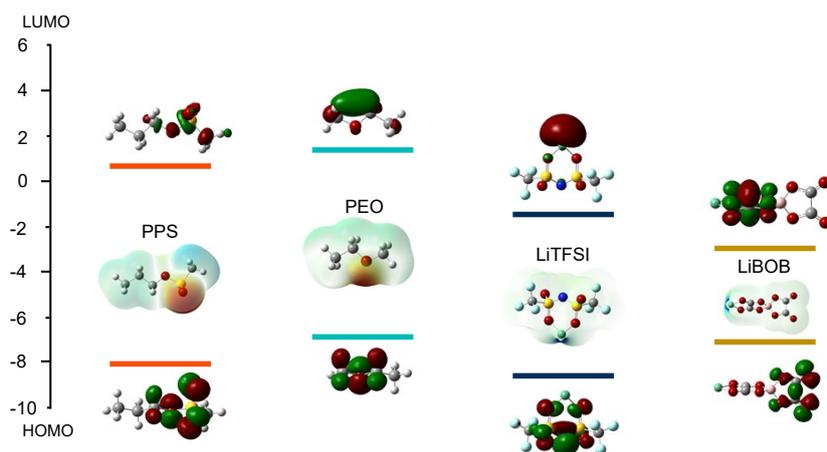
**Fig. S8** The electrostatic potentials with the interaction between LiTFSI and PPS.



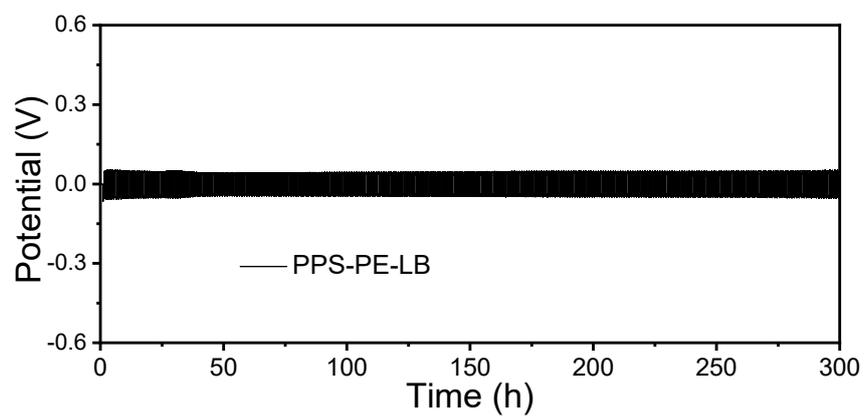
**Fig. S9** The SEM image of Li anode surface after test of battery with PPS-PE.



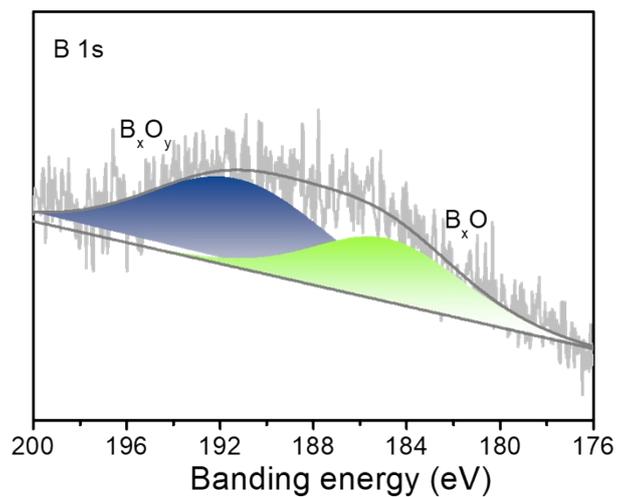
**Fig. S10** The initial charge/discharge curves of liquid electrolyte based battery with LLOs cathode material.



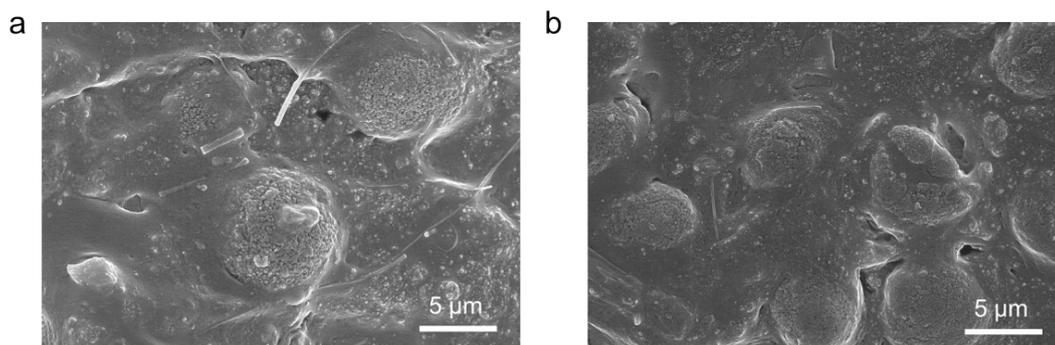
**Fig. S11** The HOMO and LUMO energy levels of PPS, PEO, LiTFSI, LiBOB and the interactions.



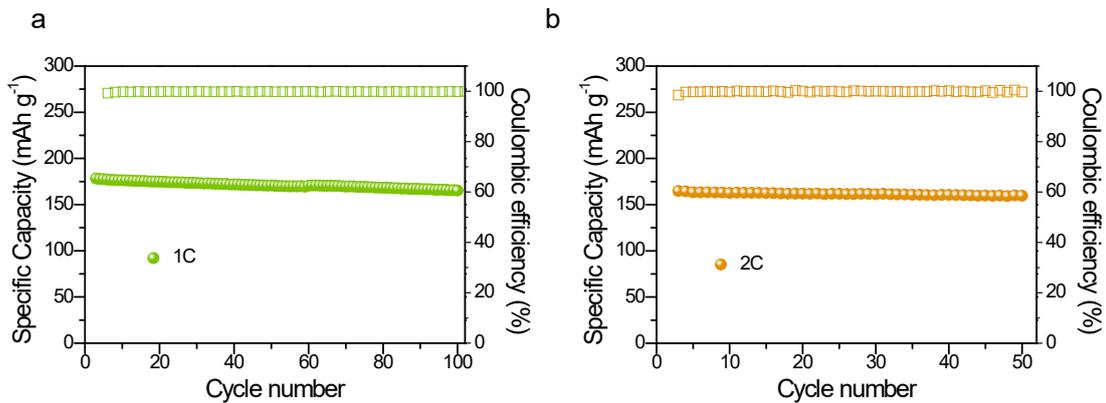
**Fig. S12** The potential profiles of Li plating/stripping in Li/PPS-PE-LB/Li symmetric battery with current density of  $0.1 \text{ mA cm}^{-2}$ .



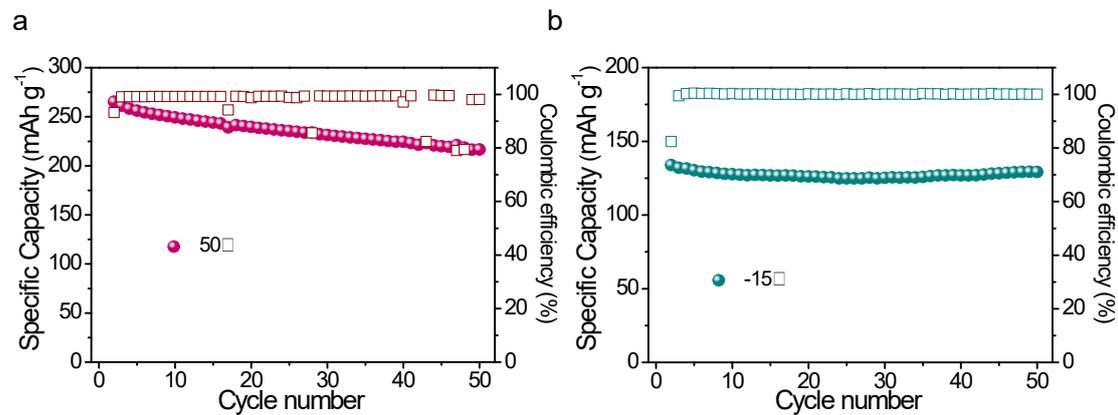
**Fig. S13** The XPS spectra of B element on the cathode side after the cycling of LLOs/PPS-PE/Li battery.



**Fig. S14** The SEM images of composite cathodes in (a) LLOs/PPS-PE/Li and (b) LLOs/PPS-PE-LB/Li batteries after cycling.



**Fig. S15** The cycling stabilities of LLOs/PPS-PE-LB/Li batteries at current density of (a) 1C and (b) 2C.



**Fig. S16** The cycling stabilities of LLOs/PPS-PE-LB/Li batteries at (a) 50 °C and (b) -15 °C.



**Fig. S17** The optical photographs for the LLOs/PPS-PE-LB/Li soft package battery that lit up a logo at a, normal, b, wrinkled and c, cutting states.

**Table S1** The relative area for each peak of PS monomer and PPS-PE from **Fig. S1**.

Type of peak	PS monomer			PPS-PE				Impurity
	a	c	b	a1	c1	c2	b1	
Integral area (e9)	2.36	2.36	2.35	16.69	28.71	12.61	14.20	7.55
Percent conversion	$(72.21/86.83) / (1-7.55/86.83) = 91.08\%$							

**Table S2** The comparisons of charge voltages of cathode materials in the solid-state batteries, and the ions transference number of polymer electrolytes, which including the  $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ ,  $\text{LiCoO}_2$ ,  $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$  and Li-rich cathode materials.

Composition	Anode/ Cathode	The Li <sup>+</sup> transference number	Current density	Voltage Range (V)	Initial discharge capacity (mAh/g)	Cycles	Final discharge capacity (mAh/g)	Ref.
PEGDA+BA+SN/LiTFSI	Li/LiNi <sub>0.83</sub> Mn <sub>0.06</sub> Co <sub>0.11</sub> O <sub>2</sub>	0.75	1.1C	2.7-4.5V	~170	100	140	23
PEGDME/LiTFSI+LiFSI	Li/LiNi <sub>0.5</sub> Mn <sub>0.3</sub> Co <sub>0.2</sub> O <sub>2</sub>	-	0.2C	2.5-4.5V	167	100	97	24
VEC+TFEMA/LiTFSI	Li/LiNi <sub>0.83</sub> Co <sub>0.11</sub> Mn <sub>0.06</sub> O <sub>2</sub>	0.44	0.1C (1C=275 mA g <sup>-1</sup> )	3.0-4.5V	218	300	~153	25
PME+LiPVFM+NMP/ LiTFSI+SN	Li/LiCoO <sub>2</sub>	0.62	0.5C	2.8-4.5V	196.3	50	180.4	26
VEC/LiTFSI	Li/LiCoO <sub>2</sub>	0.4	0.5C (1C=160 mA g <sup>-1</sup> )	3.0-4.5V	154.6	100	104.9	27
Uio-66-NH- MET+PEGDA+PETMP/ LiTFSI	Li/LiCoO <sub>2</sub>	0.44	0.5C (1C=148 mA g <sup>-1</sup> )	3.0-4.5V	136	100	108.8	28
PEGDA+NML+UPyMA/ LiTFSI	Li/LiCoO <sub>2</sub>	0.66	0.5C (1 C =180 mA g <sup>-1</sup> )	3.0-4.6V	188.5	1000	150.8	29
CUMA/SN+LiTFSI +LiDFOB	Li/LiCoO <sub>2</sub>	0.62	0.5C	3.0-4.6V	~220	100	~181	30
FEC/LiDFOB	Li/LiNi <sub>0.5</sub> Mn <sub>1.5</sub> O <sub>4</sub>	-	0.2C (1C=148 mA g <sup>-1</sup> )	3.0-4.9V	~110	20	~90	31
PMHS+PEO+DMAA+ PS+THF/LiTFSI/Speiers	Li/LiNi <sub>0.5</sub> Mn <sub>1.5</sub> O <sub>4</sub>	0.25	0.1C (1C=170 mA g <sup>-1</sup> )	3.5-4.9V	131.5	50	126.3	32
PEGDA+NML+UPyMA/ LiTFSI	Li/Li <sub>1.2</sub> Ni <sub>0.13</sub> Co <sub>0.13</sub> Mn <sub>0.54</sub> O <sub>2</sub>	0.66	0.2 C	2.1-4.9V	225	60	~220	29
<b>PS/LiTFSI</b>	<b>Li/Li<sub>1.13</sub>Mn<sub>0.517</sub>Ni<sub>0.256</sub>Co<sub>0.097</sub>O<sub>2</sub></b>	<b>0.78</b>	<b>0.1C 0.5C (1C=200 mA g<sup>-1</sup>)</b>	<b>2.0-4.8V 2.0-4.6V</b>	<b>270 208</b>	<b>100</b>	<b>192</b>	<b>This work</b>