

## Electronic Supplementary Information

### Highly Stretchable, Nonflammable and Notch-insensitive Intrinsic Self-healing Solid-state Polymer Electrolyte for Stable and Safety Flexible Lithium Batteries

Cheng Wang,<sup>‡a</sup> RuiJing Li,<sup>‡a</sup> Peng Chen,<sup>a</sup> Yongsheng Fu,<sup>a</sup> XinYan Ma,<sup>a</sup> Tao Shen,<sup>a</sup> Baojing Zhou,<sup>a</sup> Ke Chen,<sup>a</sup> JiaJun Fu<sup>\*a</sup>, Xiaofang Bao,<sup>a</sup> Wuwei Yan,<sup>b</sup> Yong Yang<sup>\*a</sup>

<sup>a</sup>School of Chemical Engineering, Nanjing University of Science and Technology, Nanjing 210094, PR China

<sup>b</sup>Shenzhen BTR Nanotechnology Co., Ltd., Shenzhen 518106, PR China.

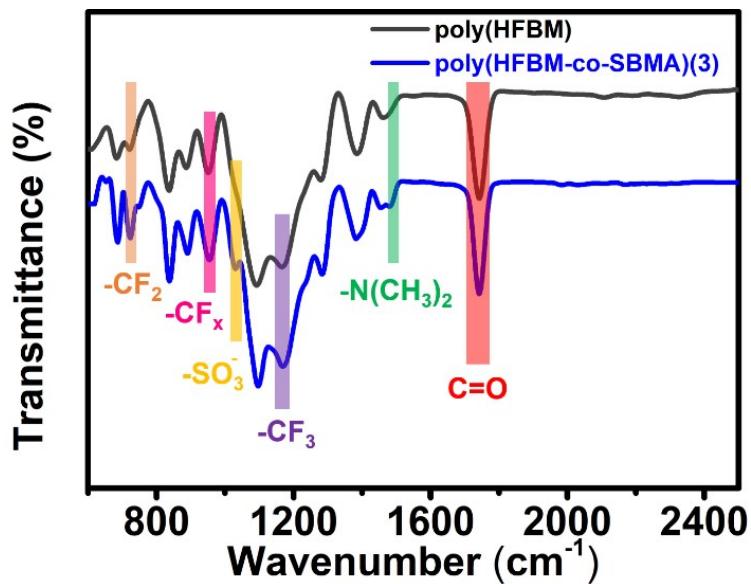
<sup>‡</sup>These authors contributed equally to this work.

Corresponding author: E-mail address: fujajun668@gmail.com; yychem@njust.edu.cn

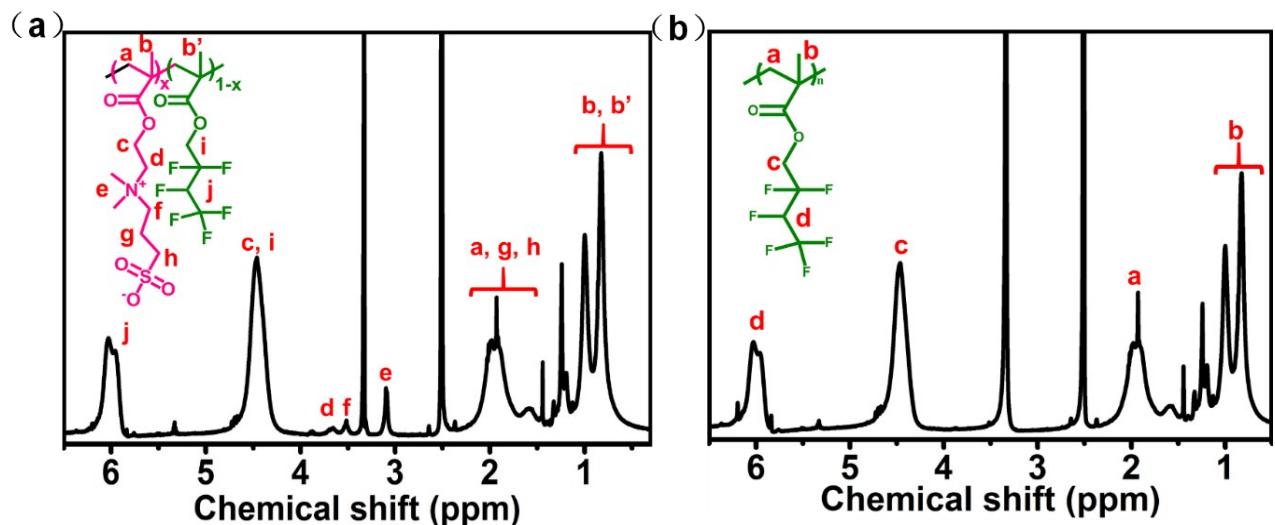
## **Table of Contents**

1. Supplementary Figures and Table.....	3
2. Supplementary Video.....	10

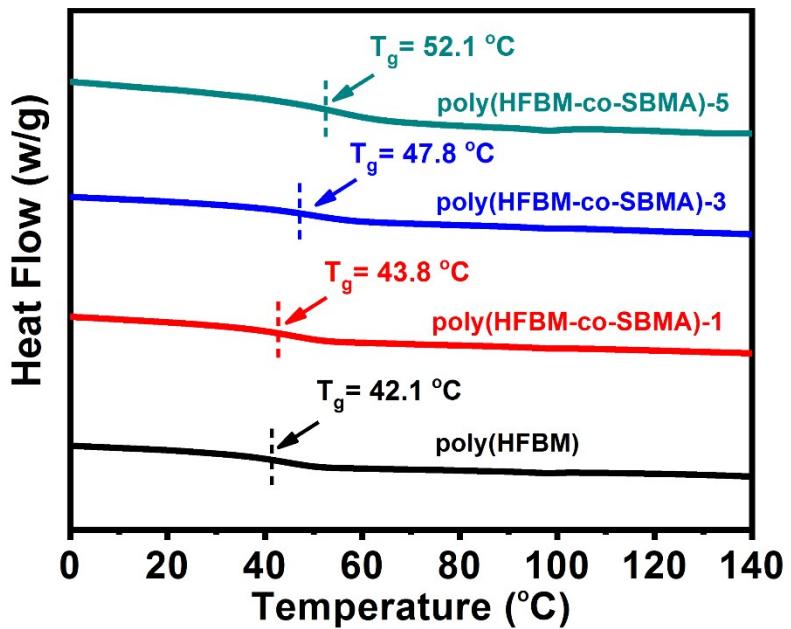
## 1. Supplementary Figures and Table



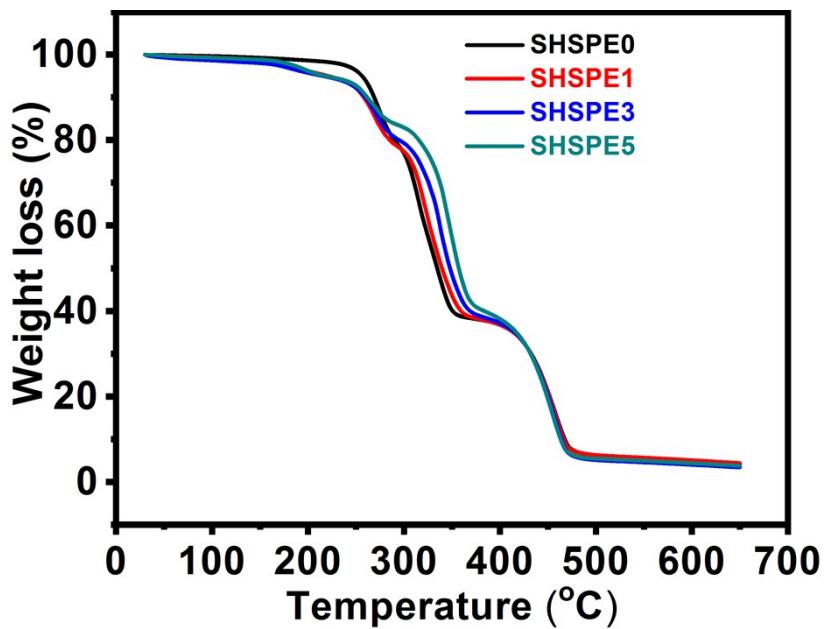
**Fig. S1** FTIR spectra of poly(HFBM) and poly(HFBM-co-SBMA)-3.



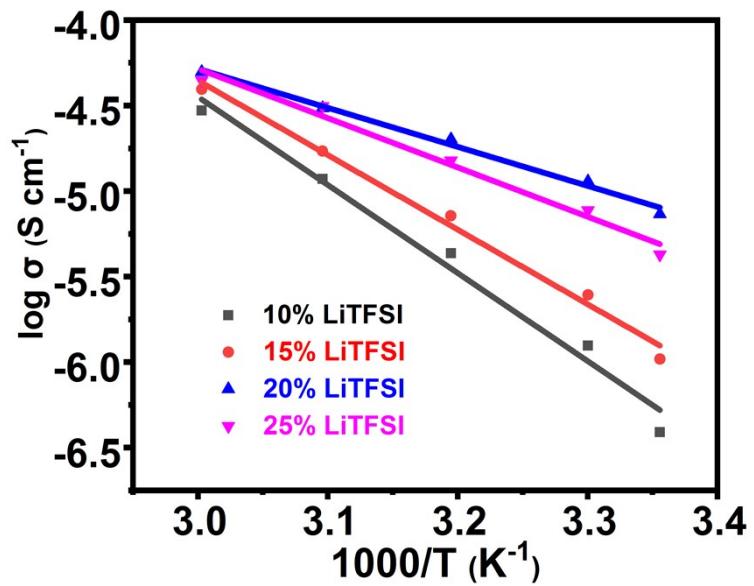
**Fig. S2**  $^1\text{H}$  NMR spectra of poly(HFBM-co-SBMA)-3 (a, DMSO- $\text{d}_6$ , 500 MHz) and poly(HFBM) (b, DMSO- $\text{d}_6$ , 500 MHz).



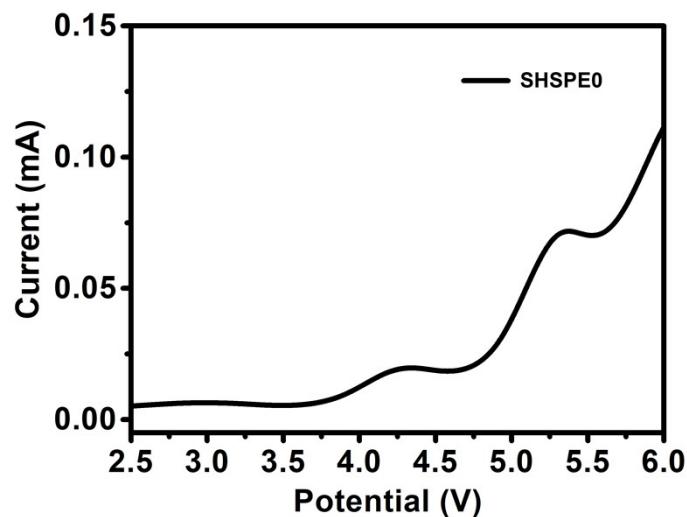
**Fig. S3** DSC curve of the samples.



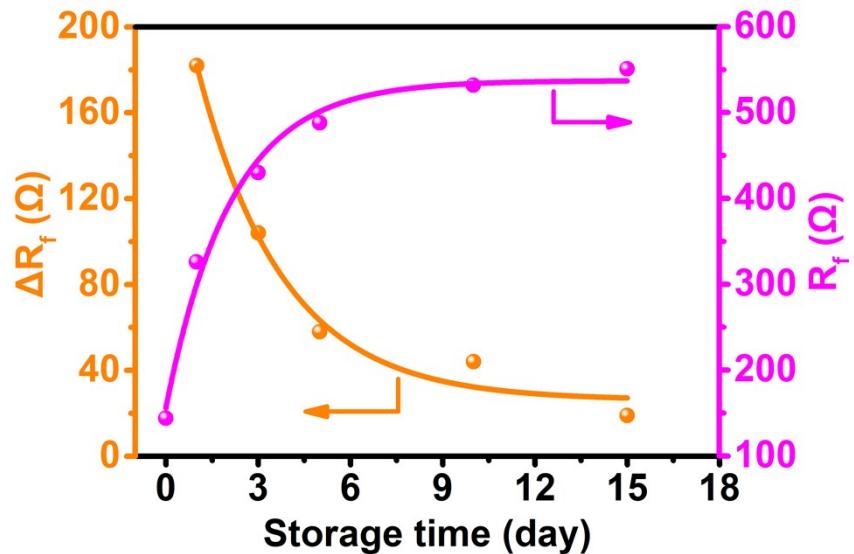
**Fig. S4** TG curves of SHSPEs.



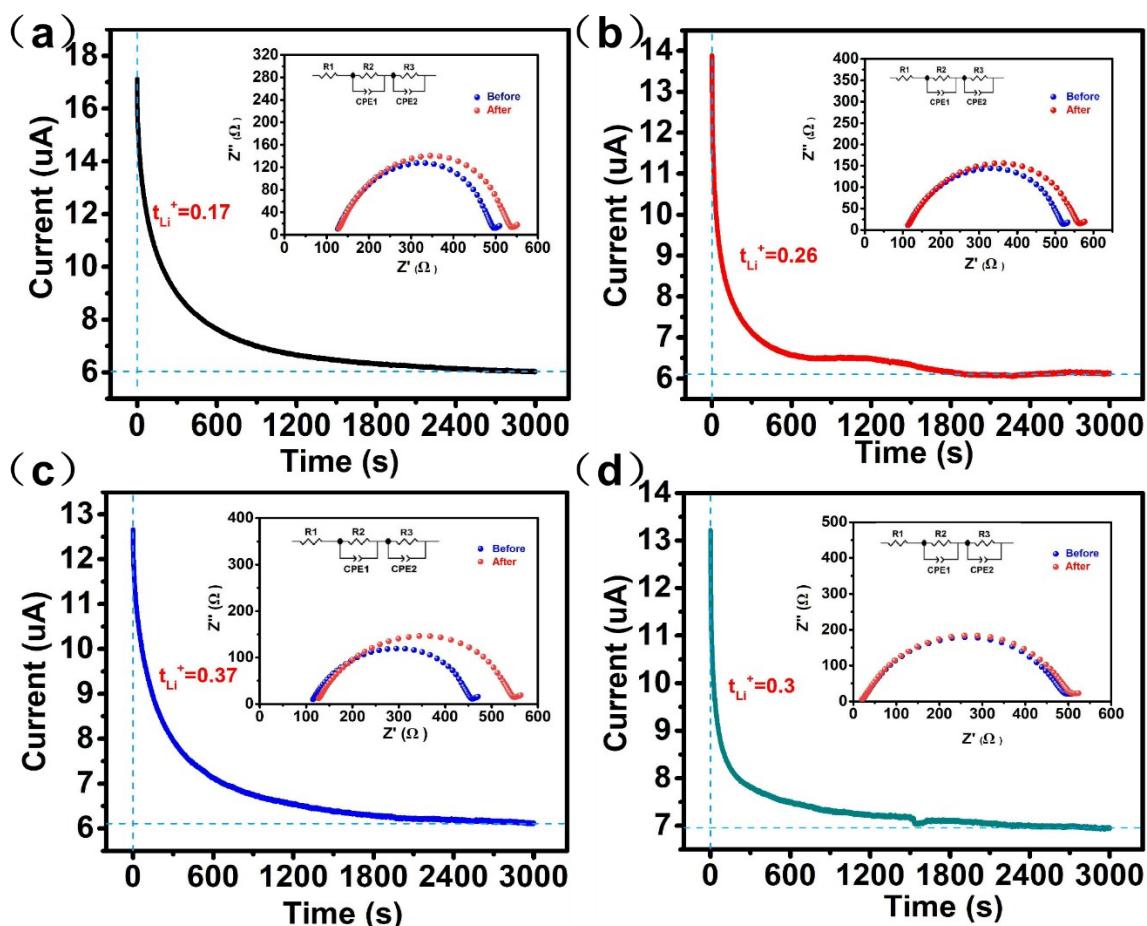
**Fig. S5** Temperature dependence of the ionic conductivity for SHSPE3 with different weight ratios of LiTFSI.



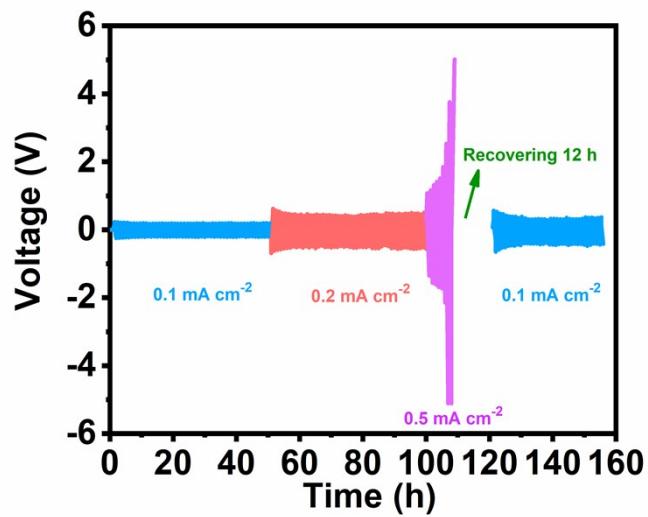
**Fig. S6** The linear sweep voltammetry plot of SHSPE0.



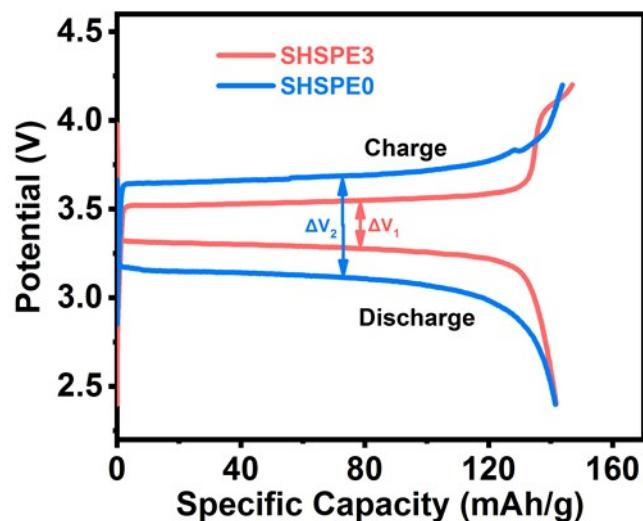
**Fig. S7** The change of the interfacial resistance for Li/SHSPE3/Li symmetrical cell over different storage time.



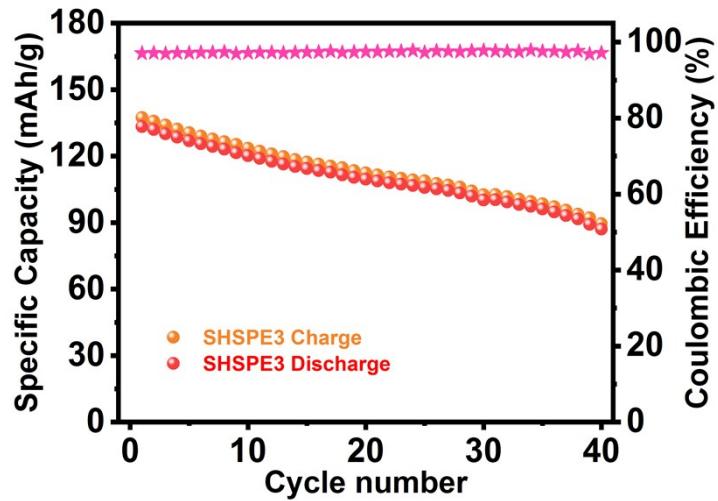
**Fig. S8** Chronoamperometry profile and the AC impedance spectrum (inset) before and after polarization of (a) Li/SHSPE0/Li, (b) Li/SHSPE1/Li, (c) Li/SHSPE3/Li and (d) Li/SHSPE5/Li.



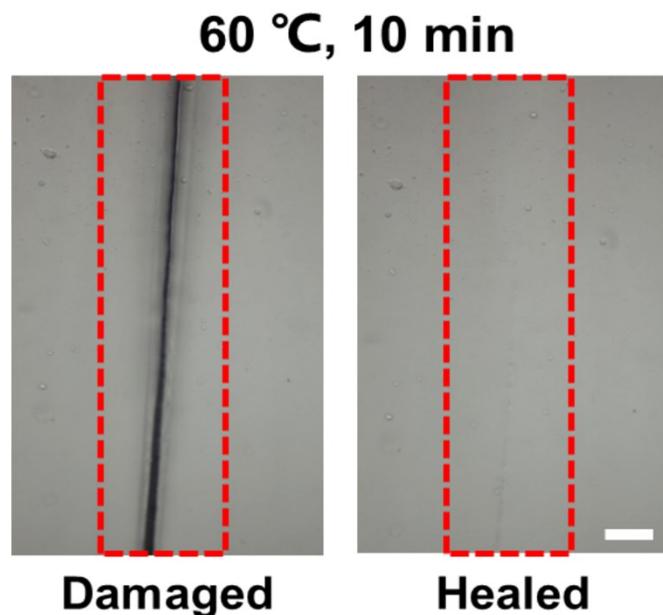
**Fig. S9** The cycling performance of Li/SHSPE3/Li symmetric cell at different current density.



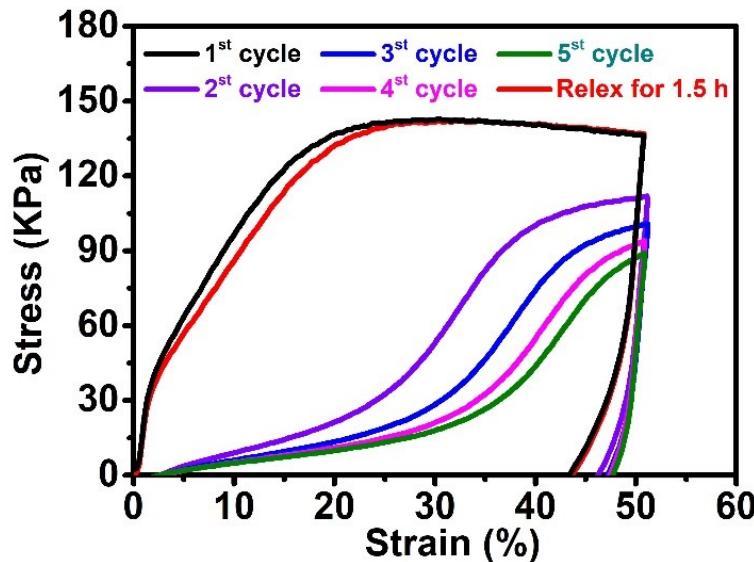
**Fig. S10** The initial charge/discharge profiles of SHSPE0 and SHSPE3 based LiFePO<sub>4</sub>/Li batteries at 0.2 C and 60 °C.



**Fig. S11** Long-term cycling performance of Li/SHSPE3/LiFePO<sub>4</sub> at 0.2 C and 60 °C (The loading of LiFePO<sub>4</sub> is 12 mg cm<sup>-2</sup>).



**Fig. S12** Images of the damaged and healed SHSPE3 membrane at 60 °C for 10 min (Scale bar: 100 μm).



**Fig. S13** Five successive loading-unloading cycles of SHSPE3.

**Table S1** The self-healing property of SHSPE3.

Healing time (h)	Toughness <sup>a</sup> (MJ/m <sup>3</sup> )	Self-healing efficiency <sup>b</sup> (%)
Original film	1.35 ± 0.03	/
0.5	0.28 ± 0.11	20.7
1.5	0.80 ± 0.08	59.3
3	1.03 ± 0.09	76.3
5	1.32 ± 0.05	97.8

a: Toughness was calculated by manually integrating the area under the stress-strain curve.

b: Self-healing efficiency was calculated from the ratio of toughness of healed films to that original film.

## 2. Supplementary Movie

The file contains supplementary video S1. In order to simulate the bending/folding of the soft-pack batteries during the actual use, SHSPE3 was compressed to 50% of the original length and relaxed to its initial state.