

Supporting information

Electrolyte-Solvent-Modified Alternating Copolymer as a Single-Ion Solid Polymer Electrolyte for High-Performance Lithium Metal Batteries

Chen Cao,[†] Yu Li,^{,†,‡,§} Shaoshan Chen,[†] Cong Peng,[†] Zeyu Li,[†] Lin Tang,[†] Yiyu Feng,^{†,‡,§} and Wei Feng^{*,†,‡,§,¶}*

[†]School of Materials Science and Engineering, Tianjin University, Tianjin 300354, China.

[‡]Key Laboratory of Advanced Ceramics and Machining Technology, Ministry of Education, Tianjin 300354, China.

[§]Tianjin Key Laboratory of Composite and Functional Materials, Tianjin 300354, China.

[¶]Collaborative Innovation Center of Chemical Science and Engineering, Tianjin 300354, China.

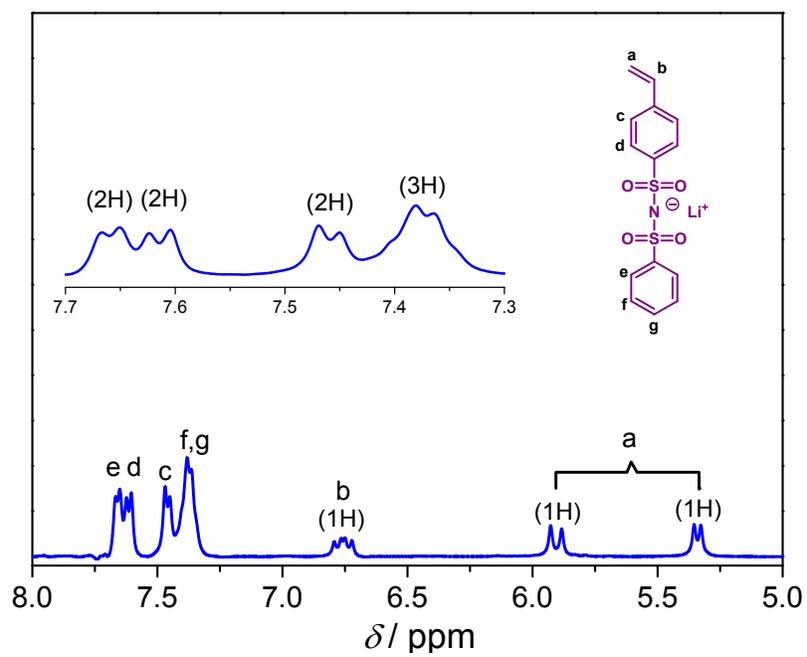


Figure S1. The ^1H NMR spectrum of SSPSiLi.

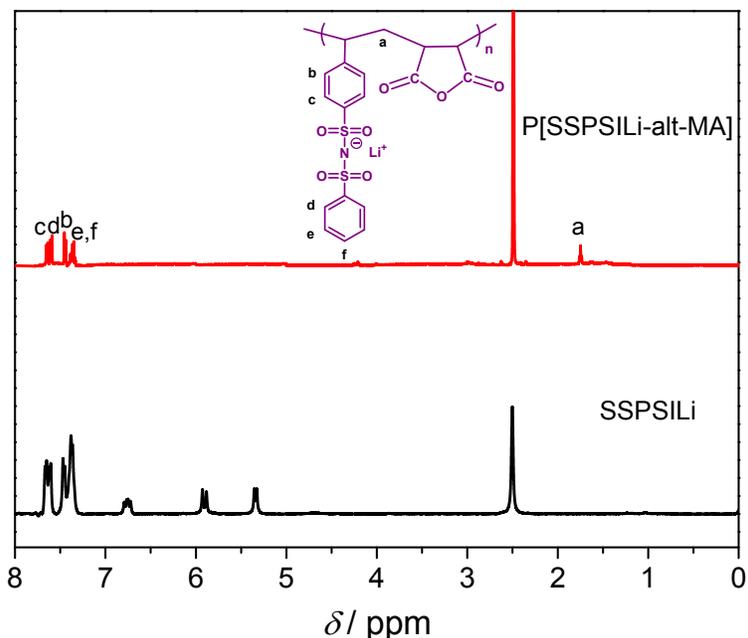


Figure S2. The comparison of the ^1H NMR spectra of $\text{Li}[\text{SSPSI-}i\text{alt-MA}]$ and SSPSILi . Besides the huge peak derived from DMSO solvent in the range from 2 to 3 ppm, several peaks located in the range from 7.68 to 7.29 ppm correspond to different chemical environments of hydrogen nuclei of aromatic rings. The disappearance the consecutive three peaks appearing at 6.73, 5.87, and 5.33 ppm and the emergence in the range from 0 to 2 ppm demonstrate the breakage of $\text{C}=\text{C}$ bonds and the successful polymerization.

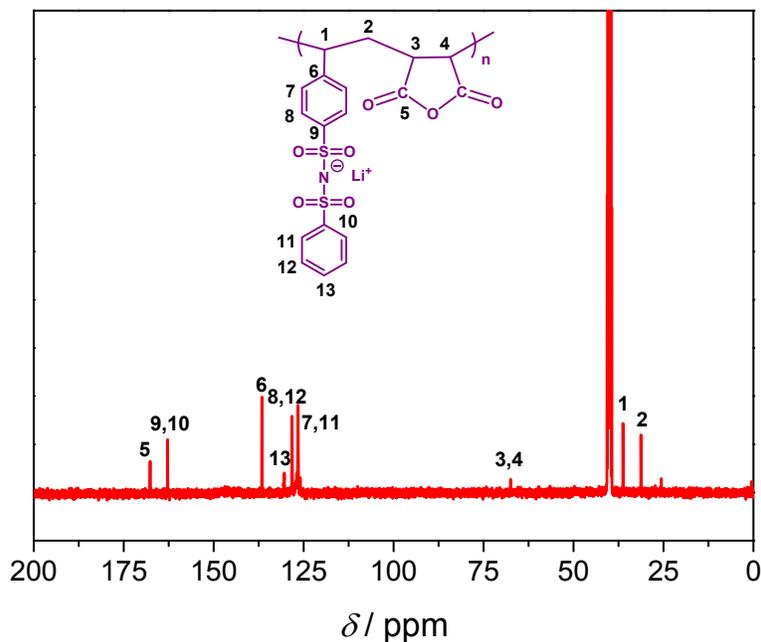


Figure S3. The ^{13}C NMR spectrum of Li[SSPSI-*alt*-MA].

Due to the failure of distinguishing the peaks of anhydride, the alternating structure of the polymer cannot be confirmed by computing the area ratio of peaks belonging to the aromatic rings and anhydride. Fortunately, according to the previous works reported by N. T. Hieu Ha,^[1] the alternating structure of the polymer can be determined by the quaternary ^{13}C NMR spectroscopy. In the ^{13}C NMR spectrum of Li[SSPSI-*alt*-MA] shown in the Fig. S3, the peak attributed to the quaternary carbon appear at 136.7 ppm contained in the range of 140.5-136.5 ppm, indicating the alternating structure of Li[SSPSI-*alt*-MA].

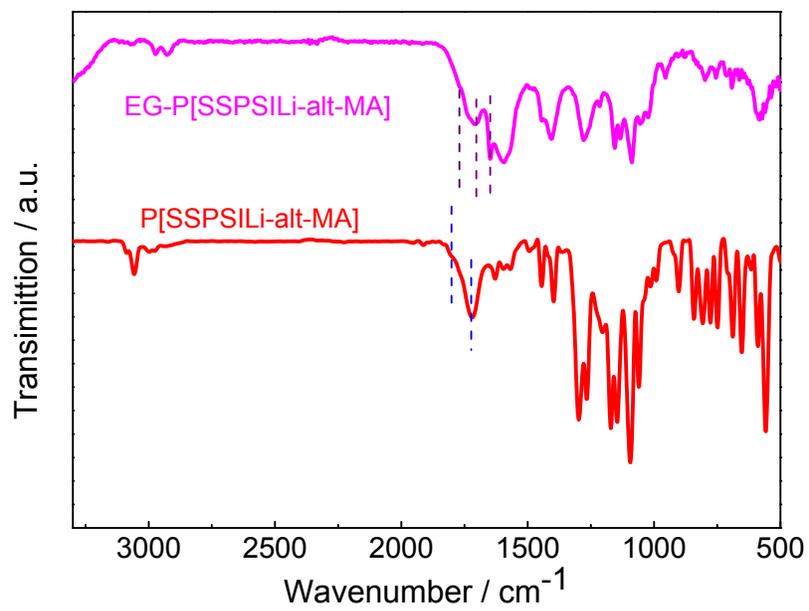


Figure S4. The FT-IR spectra of EG-Li[SSPSI-*alt*-MA] and Li[SSPSI-*alt*-MA].

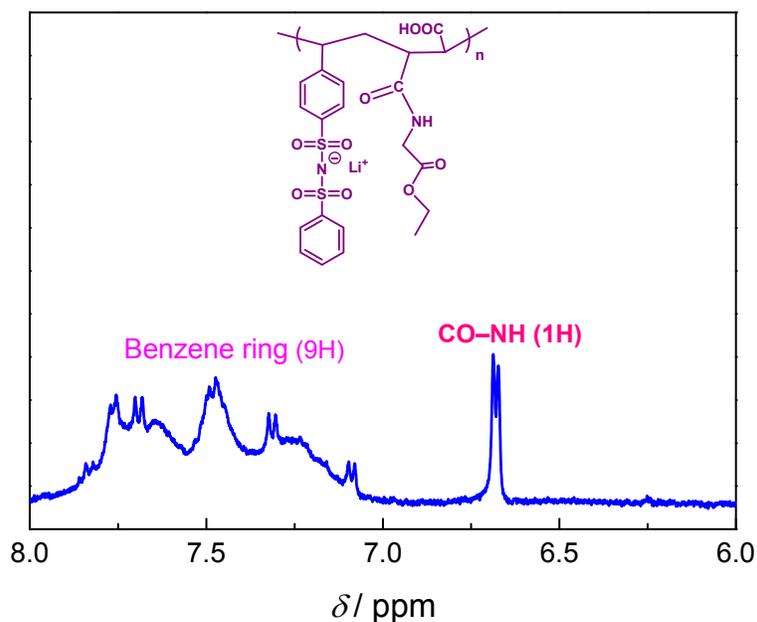


Figure S5. The ^1H NMR spectrum of EG-Li[SSPSI-*alt*-MA].

In the IR spectrum of spectrum of Li[SSPSI-*alt*-MA], the characteristic absorption peaks of the anhydride group appear at 1794 and 1720 cm^{-1} pertaining to symmetrical and asymmetrical stretching of the C=O groups, respectively. However, when ethyl glycine reacted with maleic anhydride to form the carboxylic acid and amide, the chemical environment of carbonyl group was changed. Therefore, the absorption peak of the C=O groups in EG-Li[SSPSI-*alt*-MA] moves to 1710 and 1647 cm^{-1} , corresponding to carboxylic acid and amide, respectively. The absorption peak at 1750 cm^{-1} belongs to the stretching vibration of the ester group in EG-Li[SSPSI-*alt*-MA]. As shown in Fig. S5, the ratio of hydrogen nuclei in benzene ring to that in amide is close to 9, indicating that the ring-opening efficiency of ethyl glycine on maleic anhydride is close to 100%.

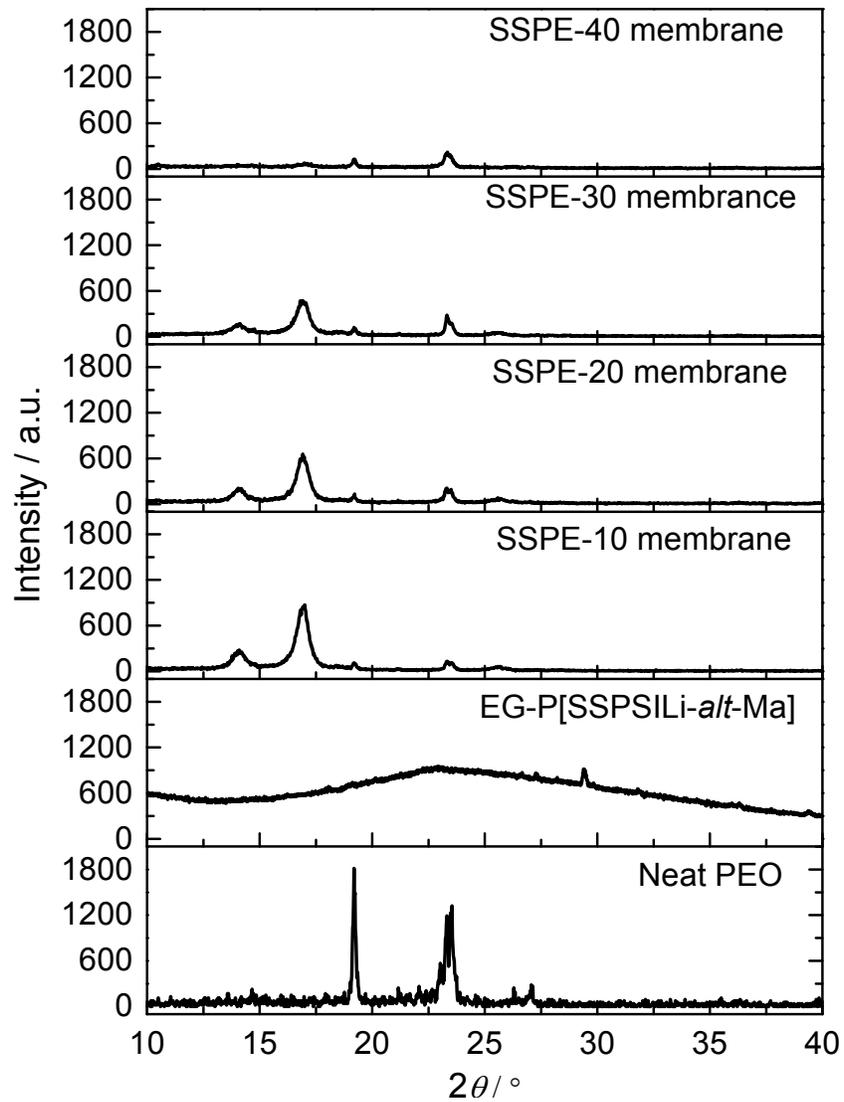


Figure S6. The XRD of SSPE membranes, neat EG-P[SSPSILi-*alt*-MA] salt and neat PEO membrane.

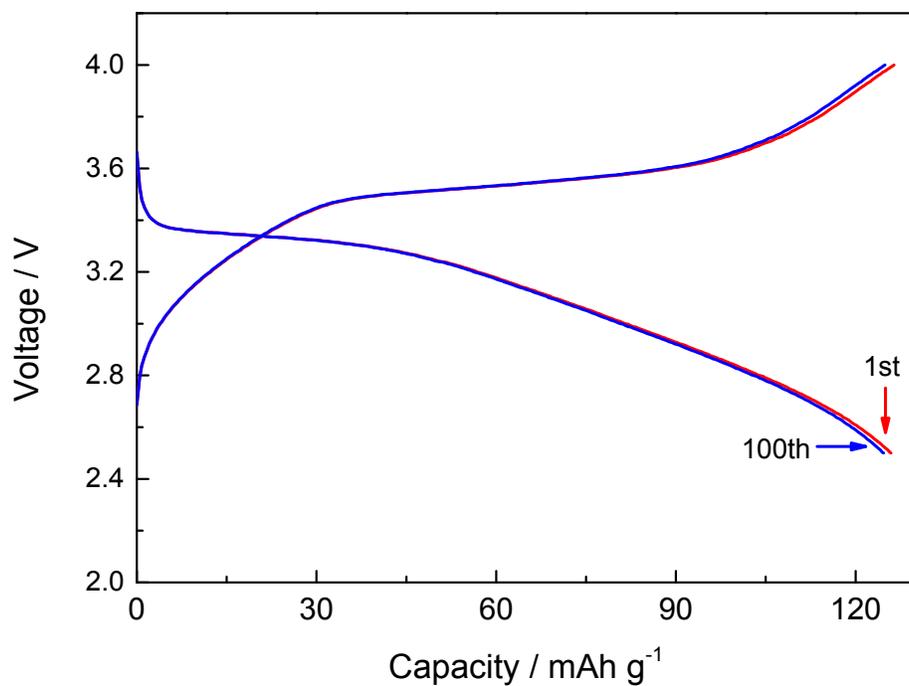


Figure S7. Galvanostatic charge/discharge profiles at the first and 100th cycle of the LFP cell with the SSPE-30 membrane at 0.1 C.

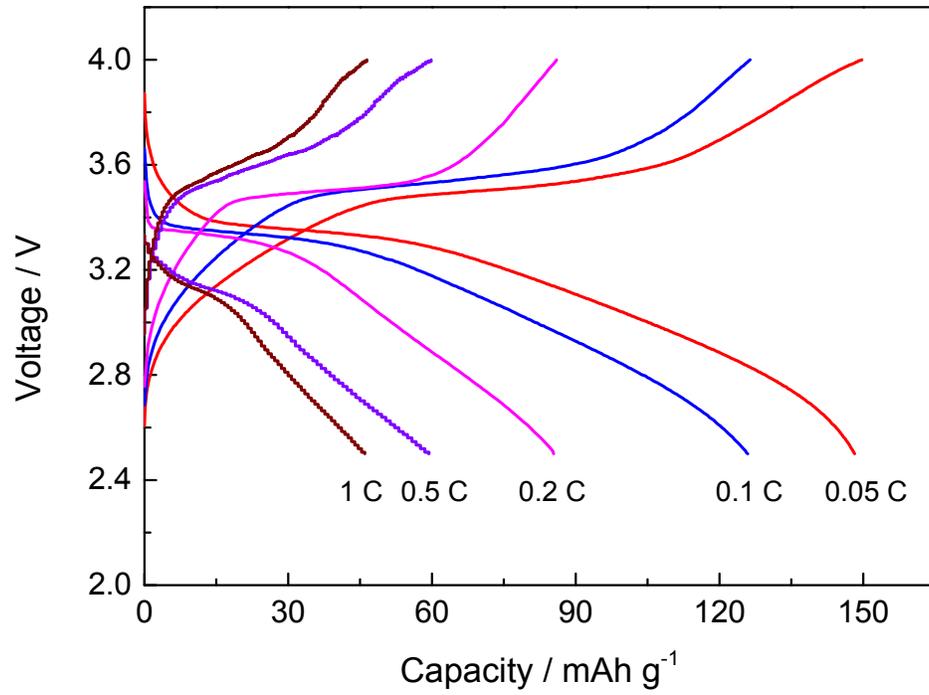


Figure S8. Galvanostatic charge/discharge profiles of the LFP cell with the SSPE-30 membrane at different rates.

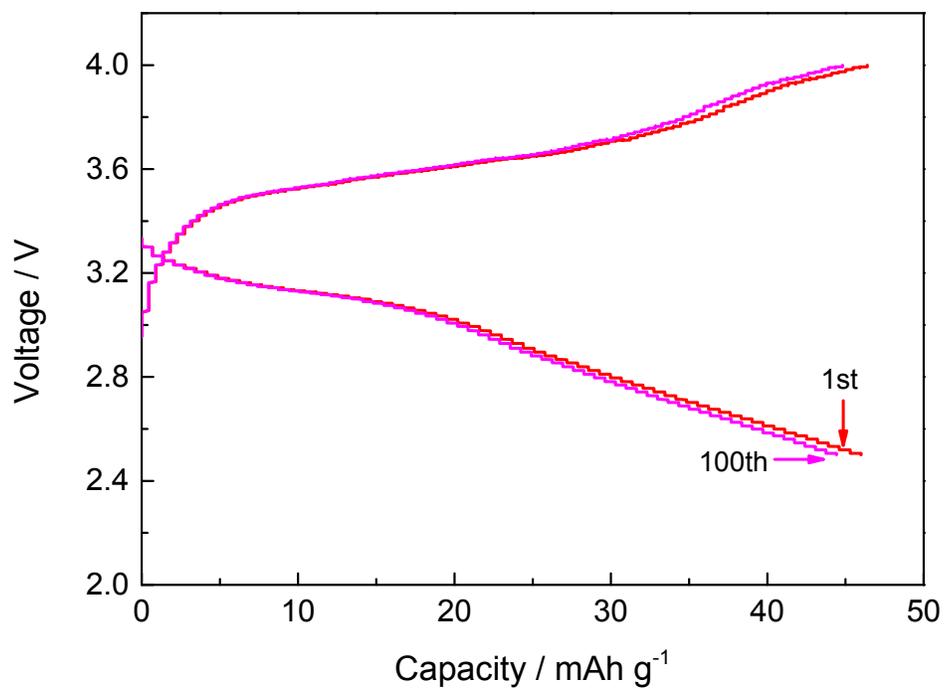


Figure S9. Galvanostatic charge/discharge profiles at the first and 100th cycle of the LFP cell with the SSPE-30 membrane at 1 C.

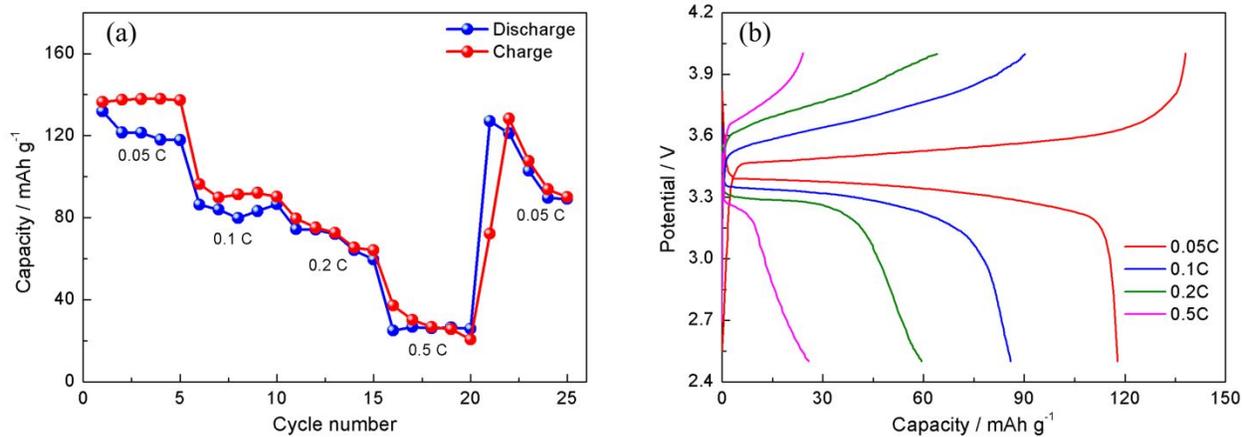


Figure S10 (a) Rate performance and (b) the typical galvanostatic charge/discharge profiles of the Li/PEO-LiTFSI/LFP cell and 80 °C, in which the EO/Li⁺ ratio of PEO-LiTFSI membrane is the same with the value of SSPE-30 membrane.



Figure S11. The open circuit voltage of Li/SSPE-30membrane/LFP flexible battery.



Figure S12. The LED lamp lighted by Li/SSPE-30membrane/LFP flexible battery in the dark.

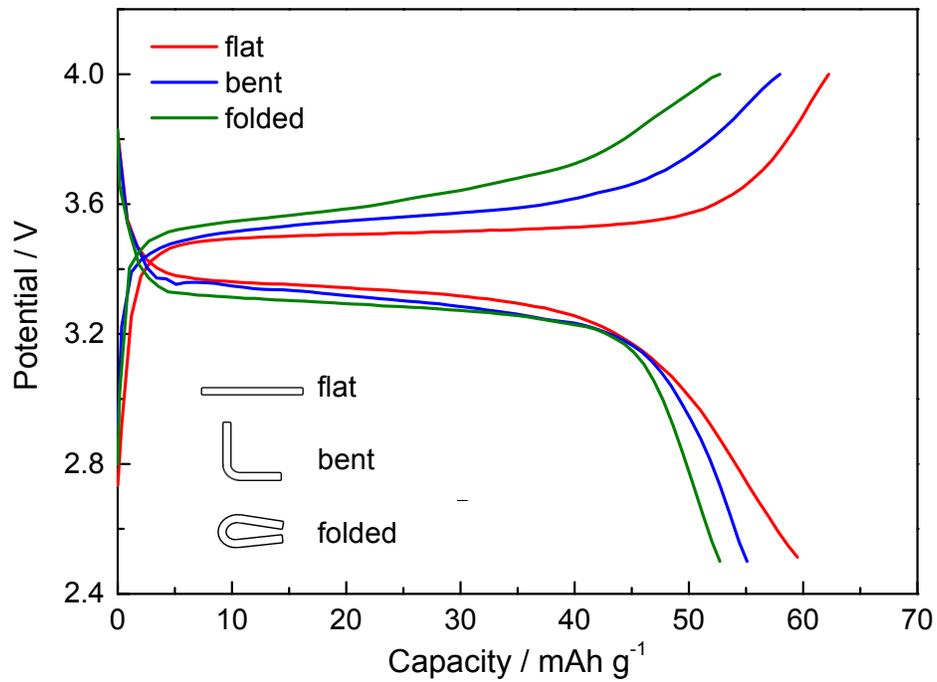


Figure S13 The galvanostatic charge/discharge profiles of fabricated flexible battery at 0.05 C in flat, bent and folded states of ambient condition, respectively.

Table. S1 Lithium ion transference numbers (t_{Li^+}) of the SSPE membranes with different contents of EG-P[SSPSILi-*alt*-MA] at 80 °C.

EG-P[SSPSILi- <i>alt</i> -MA] wt%	I_0 (μA)	I_S (μA)	R_0 (Ω)	R_S (Ω)	t_{Li^+}
40%	0.404	0.312	8960	9300	0.74
30%	0.904	0.827	2900	3460	0.93
20%	2.01	1.86	3790	3820	0.89
10%	2.22	1.844	1920	2040	0.81

Movie S1 Lighting a LED lamp through the fabricated Li/SSPE-30membrane/LFP flexible battery in the dark circumstance.

Movie S2 The stable operation of the fabricated Li/SSPE-30membrane/LFP flexible battery at different bent states.

Reference

[1] Hieu-Ha, N. T. Determination of triad sequence distribution of copolymers of maleic anhydride and its derivatives with donor monomers by ^{13}C NMR. Spectroscopy. *Polymer*, **1999**, *40*, 1081.