Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Metal organic framework optimized hybrid solid polymer electrolytes with high lithium-ion transference number and excellent electrochemical stability

Dong Han[a], Zongchen Zhao[a], Wei Wang[a], Hongjie Wang[a], Huanlei Wang[a], Lili Zheng[b], Jing Shi[a]*, Xichao Li[b]*

[a] School of Materials Science and Engineering, Ocean University of China, Qingdao 266100, People's Republic of China.

[b] School of Mechanical & Electrical Engineering, Qingdao University, Qingdao, 266071, China.

*Co-corresponding author Tel and email. Tel: +0532-66781690; E-mail: shijing@ouc.edu.cn (Jing Shi); lixichao2015@163.com (Xichao Li)

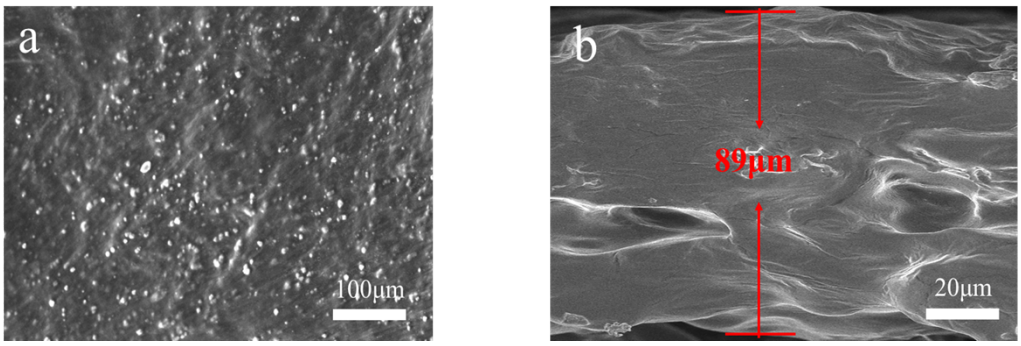

Figure.S1. SEM image of (a) PEM15 electrolyte and (b) cross sectional of PEM10 electrolyte.

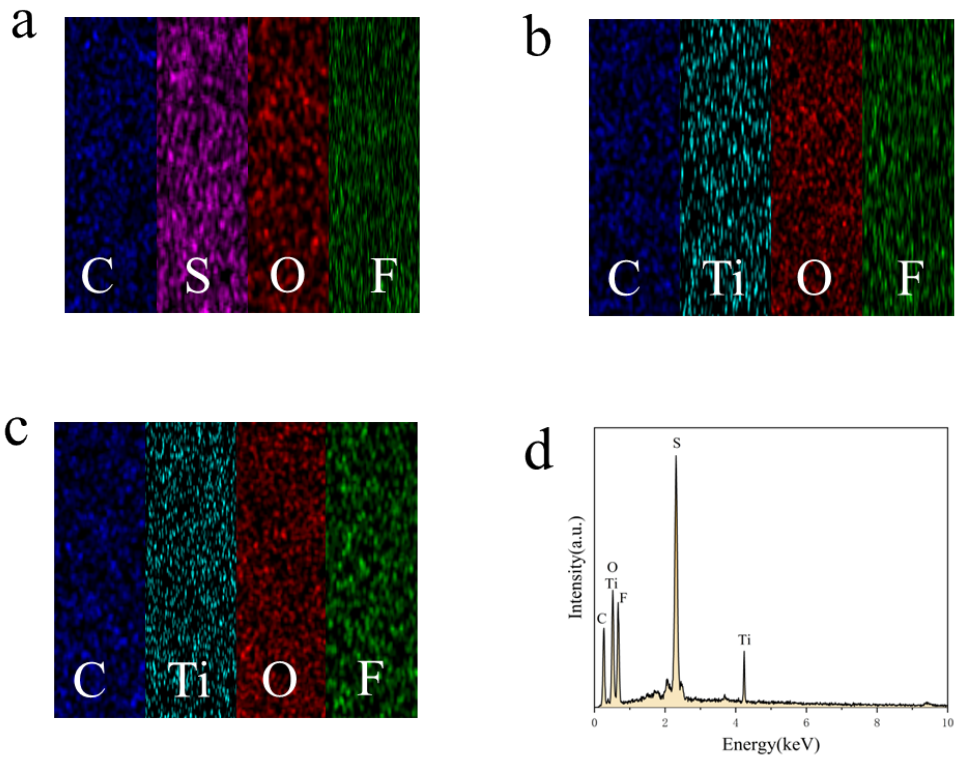

Figure.S2. EDX mapping of (a) PE, (b) PEM5 and (c) PEM15. (d) EDX spectra of PEM10 electrolyte.

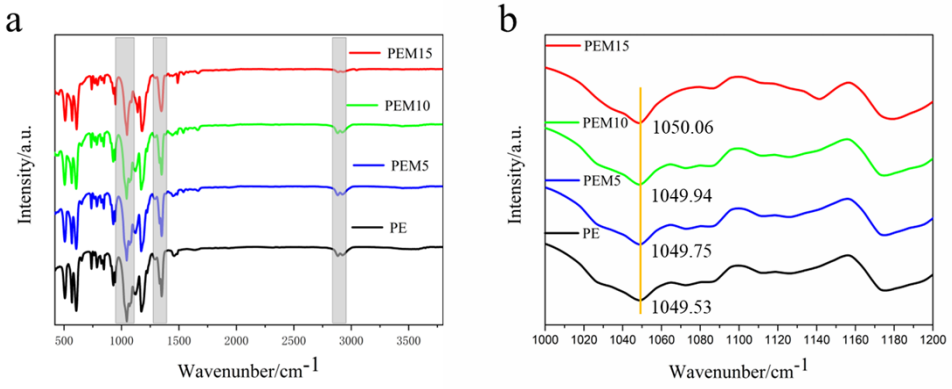

Figure.S3. FTIR spectrum of PE, PEM5, PEM10 and PEM15 electrolyte.

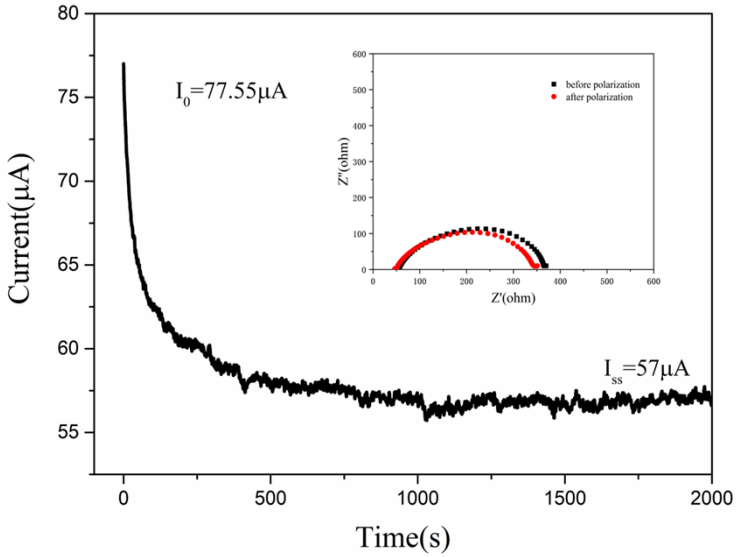

Figure.S4. The chronoamperometry curve of Li/PE/Li cell under a potential step of 10 mV at 50 ℃.

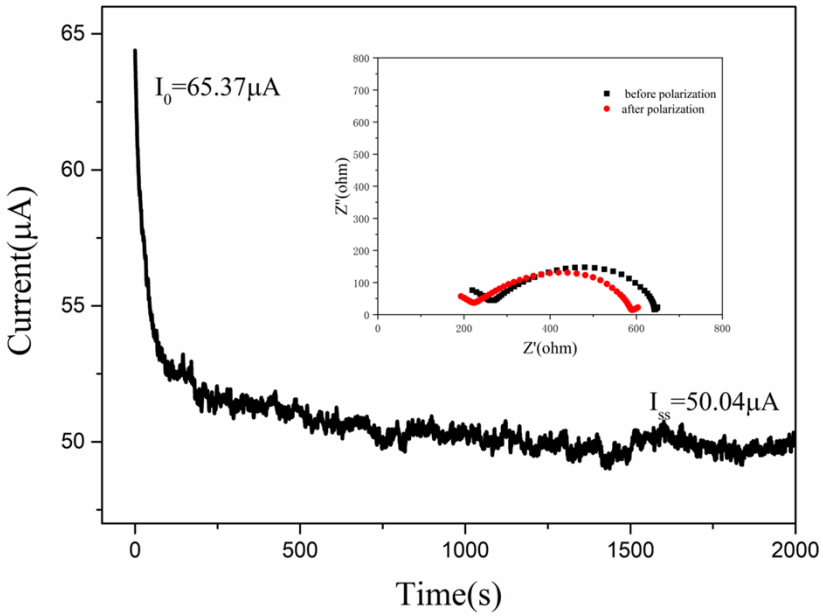

Figure.S5. The chronoamperometry curve of Li/PEM5/Li cell under a potential step of 10 mV at 50 ℃.

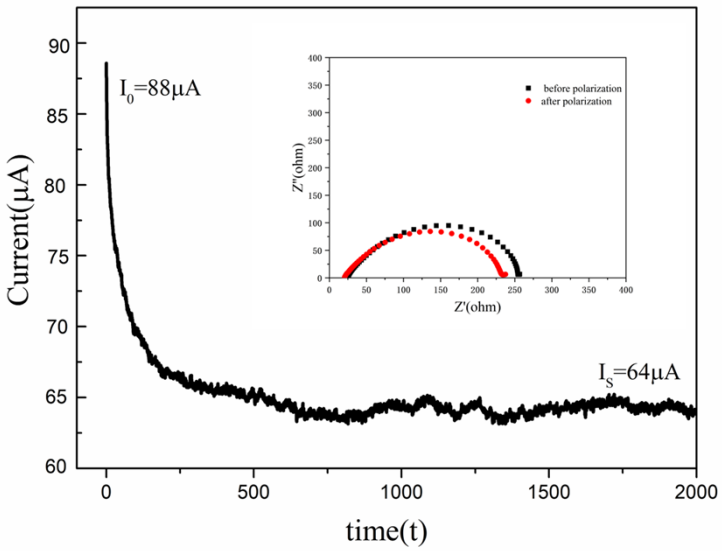

Figure.S6. The chronoamperometry curve of Li/PEM15/Li cell under a potential step of 10 mV at 50 ℃.

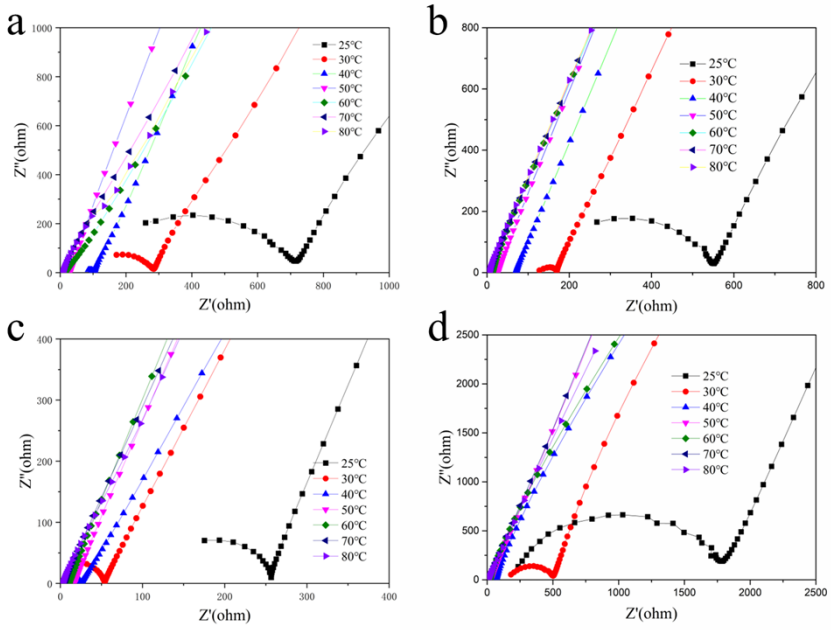

Figure.S7. EIS spectra at 25 ℃, 30 ℃, 40 ℃, 50 ℃, 60 ℃, 70 ℃ and 80 ℃ of electrolytes. (a) PE; (b) PEM5; (c) PEM15; (d) PEM5, [EO]: [Li]=16:1.

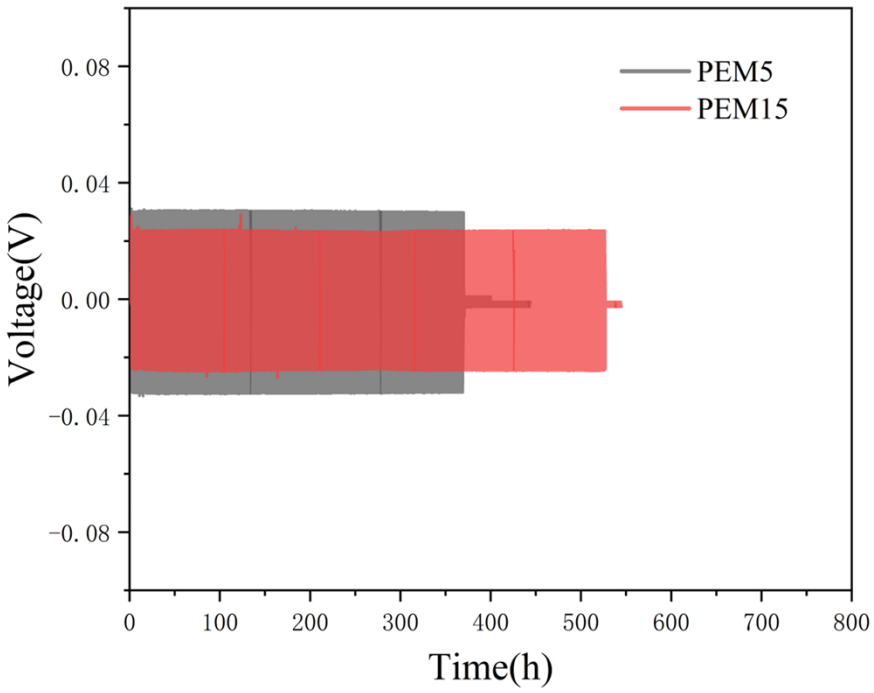

Figure.S8. Long-term cycling profiles of Li/PEM5/Li and Li/PEM15/Li batteries with 0.1 mA cm^{-2} at 50 °C.

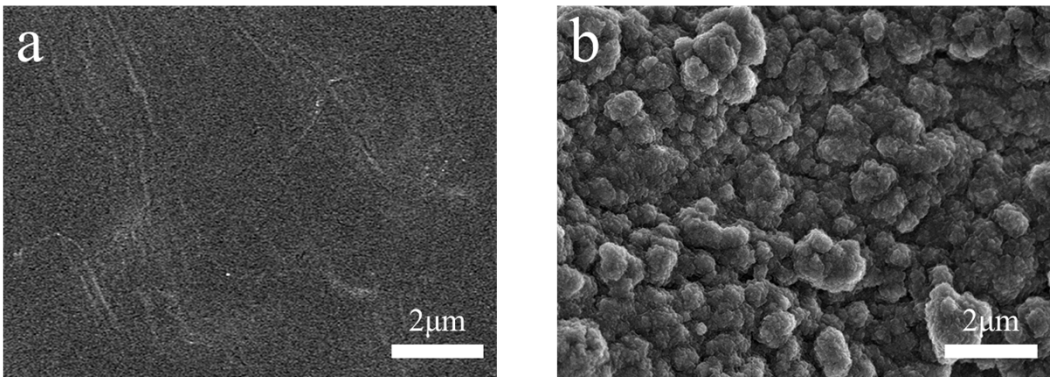

Figure.S9. SEM images of Li electrode after cycling (a) Li/PEM10/Li symmetric cell for 600 h and (b) Li/PE/Li symmetric cell for 300h.

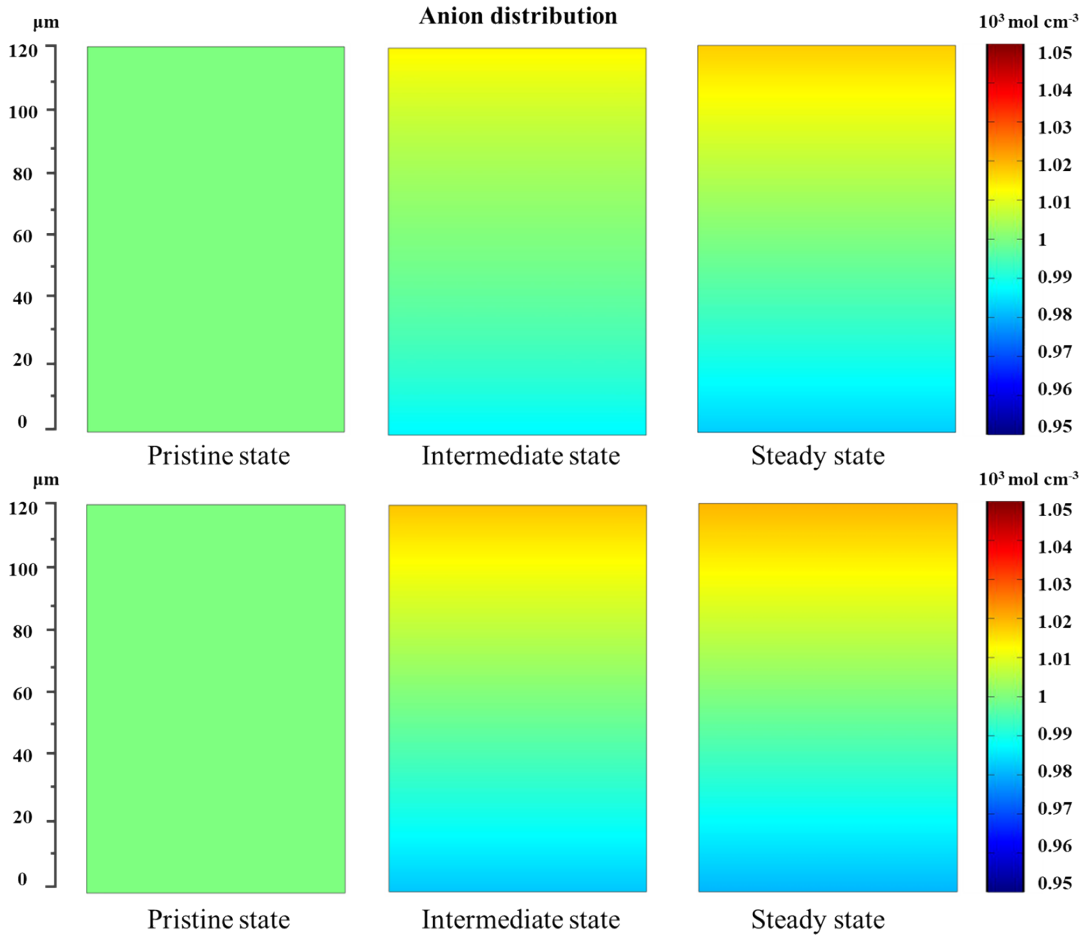

Figure.S10. Diffusion behaviors of anion in (a) PEM10 and (b) PE electrolytes at pristine state, intermediate state, and steady state during charging from the COMSOL simulations.

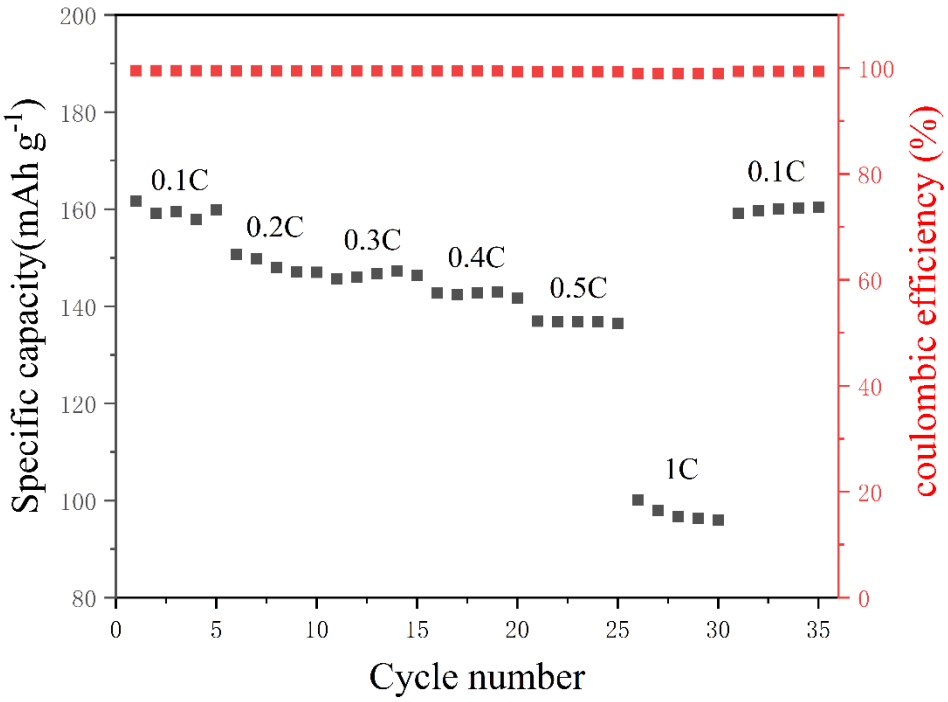

Figure.S11. Rate capacities and coulombic efficiency of $LiFePO_4$/PEM10/Li cell at 50 °C.

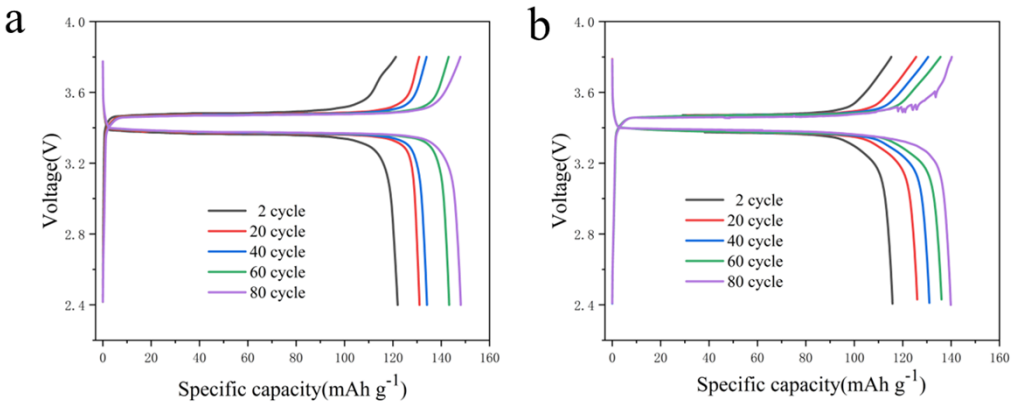

Figure.S12. Corresponding charge-discharge profiles of (a) $LiFePO_4$/PEM5/Li and (b) $LiFePO_4$/PEM15/Li cell.

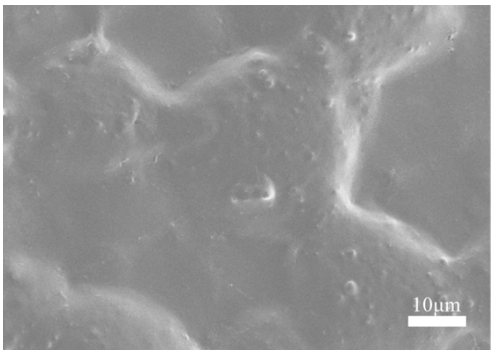

Figure.S13. SEM images of PEM10 electrolyte membrane after Cycling test of $LiFePO_4$/PEM10/Li cell.

electrolyte	[EO]: [Li]	25 ℃	30 ℃	40 ℃	50 ℃	60 ℃	70 ℃	80 ℃
PE	6:1	668.21	239.67	101.24	29.35	15.27	9.25	6.67
PEM5	6:1	459.41	166.29	69.34	25.52	10.78	6.94	3.09
PEM10	6:1	76.08	54.45	22.01	5.37	3.17	2.09	1.73
PEM15	6:1	142.72	87.53	50.43	17.02	10.29	4.91	2.54
PEM5	16:1	1681.5	348.26	70.53	29.32	21.01	10.84	6.11

Table S1. Comparison of the impedance value (Ω) of PE, PEM5, PEM10, and PEM15 electrolyte at 25 ℃, 30 ℃, 40 ℃, 50 ℃, 60 ℃, 70 ℃ and 80 ℃

electrolyte	[EO]: [Li]	25 ℃	50 ℃	80 ℃
PE	6:1	8.93×10^{-6}	2.03×10^{-4}	8.9×10^{-4}
PEM5	6:1	1.3×10^{-5}	2.34×10^{-4}	1.92×10^{-3}
PEM10	6:1	7.8×10^{-5}	1.43×10^{-3}	3.4×10^{-3}
PEM15	6:1	4.18×10^{-5}	3.51×10^{-4}	2.3×10^{-3}
PEM5	16:1	3.55×10^{-6}	2.03×10^{-4}	9.76×10^{-4}

Table S2. Comparison of the ionic conductivity (S cm^{-1}) of PE, PEM5, PEM10, PEM15 and PEM5 [EO]: [Li]=16:1 electrolyte at 25 ℃, 50 ℃ and 80 ℃.

	PE	PEM5	PEM10	PEM15
extension values (%)	275%	450%	555%	492%

Table S3. Maximum extension values (%) of PE, PEM5, PEM10, PEM15 electrolytes.