

Supplementary Information

Design of multi-functional gel polymer electrolyte with 3D compact stacked polymer micro-sphere matrix for high- performance lithium metal batteries

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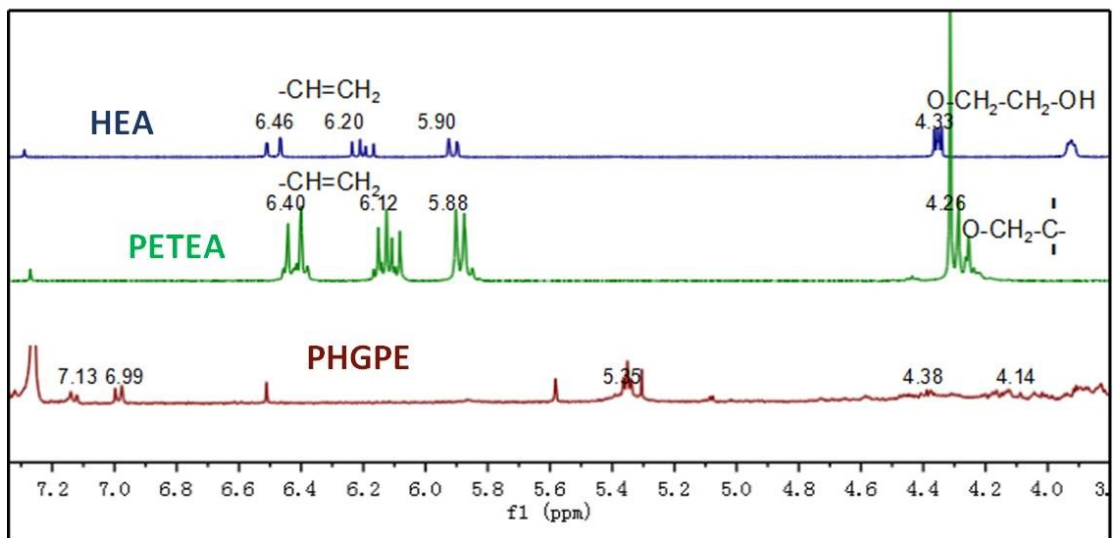


Fig. S1. ¹H NMR spectra of HEA, PETEA and PHGPE

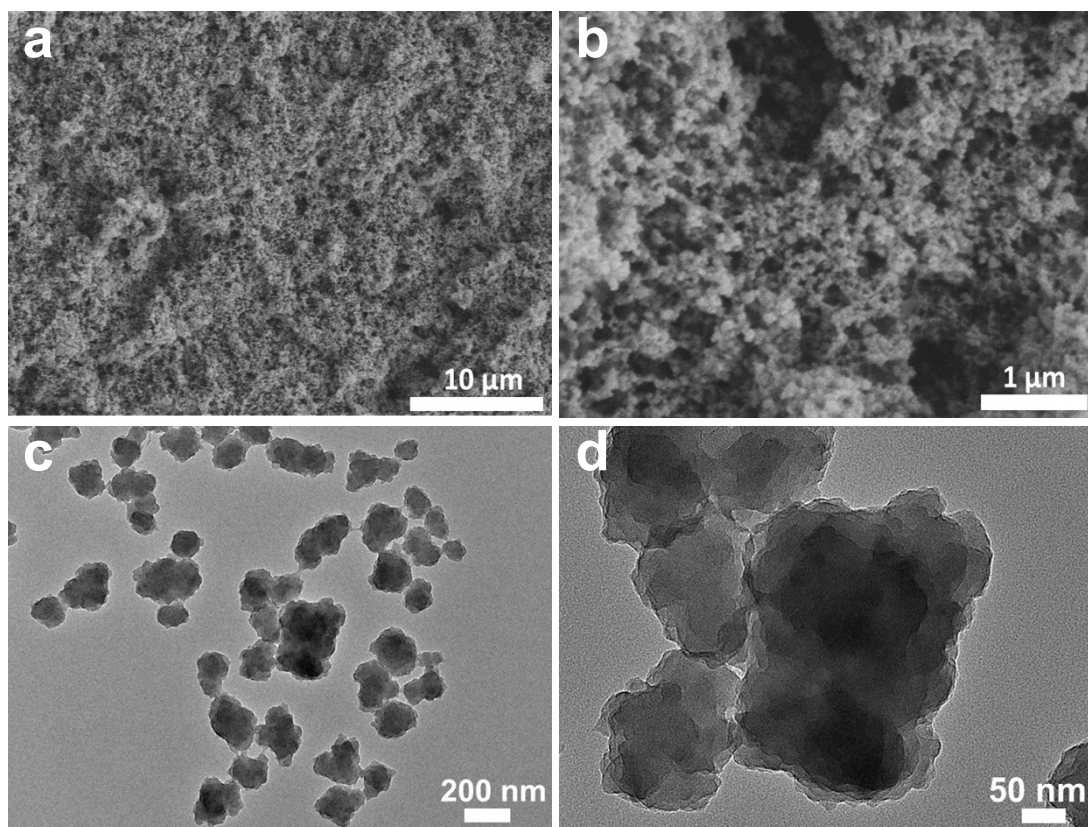


Fig. S2. (a, b) SEM images of PGPE matrix. (c, d) TEM images of PGPE matrix.

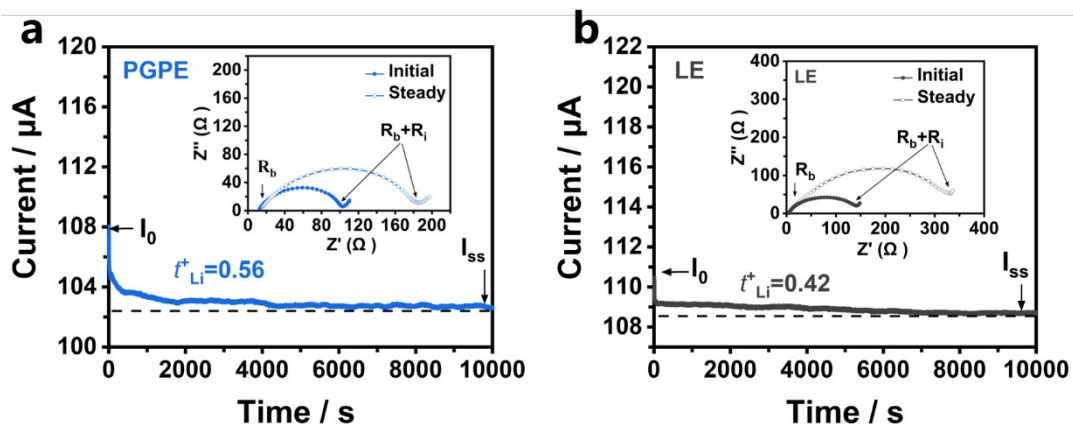


Fig. S3. The chronoamperometry profiles of (a) Li||PGPE||Li and (b) Li||LE||Li cells under a polarization voltage of 10 mV. The corresponding EISs before and after polarization are shown in the insets.

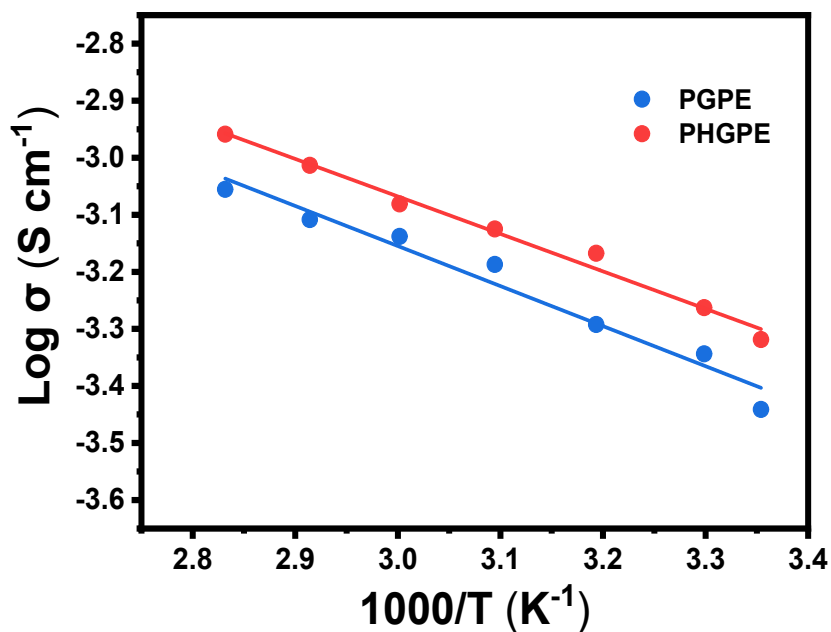


Fig. S4. Ionic conductivities of PGPE and PHGPE as a function of temperature.

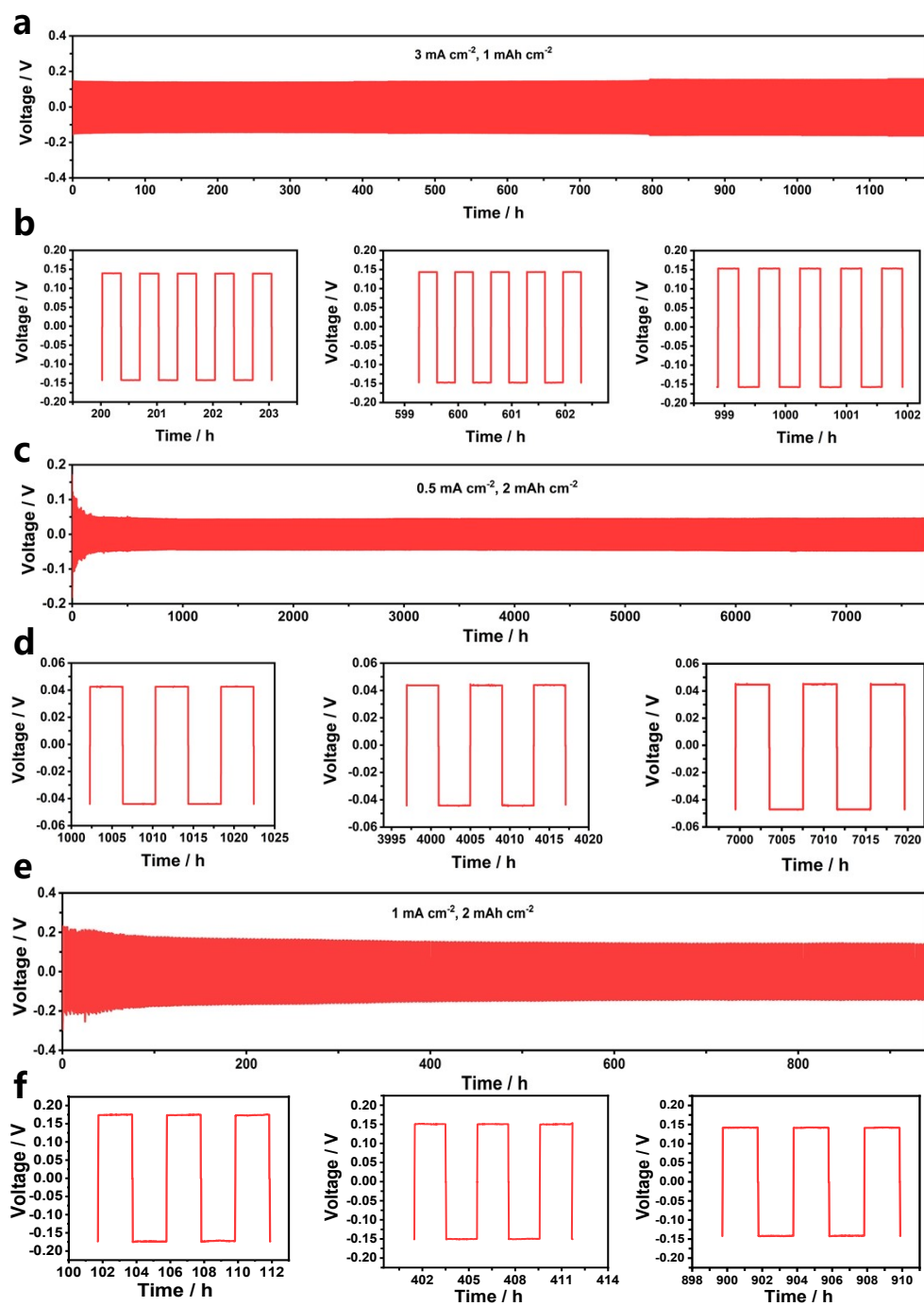


Fig. S5. Cycling performance of Li||PHGPE||Li symmetric cells at (a-b) 3 mA cm^{-2} , 1 mAh cm^{-2} , (c-d) 0.5 mA cm^{-2} , 2 mAh cm^{-2} , (e-f) 1 mA cm^{-2} , 2 mAh cm^{-2} .

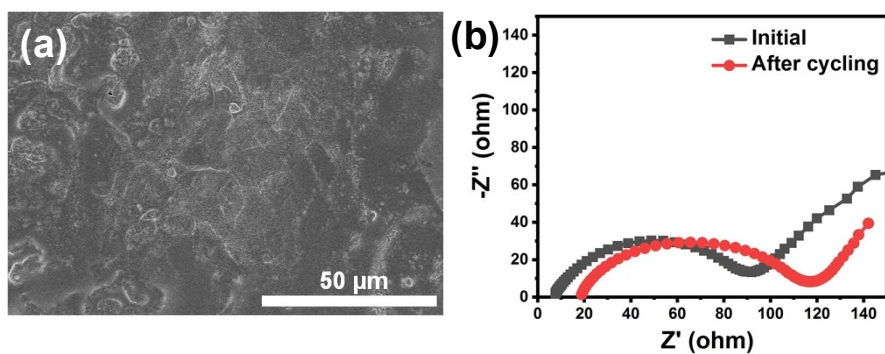


Fig. S6. (a) SEM image of Lithium metal in PHGPE based symmetric cell after 960 h cycling with a cycling capacity of 2 mAh cm^{-2} at a current rate of 1 mA cm^{-2} : (b) Nyquist plots of Li||PHGPE||Li symmetric cell before and after cycling.

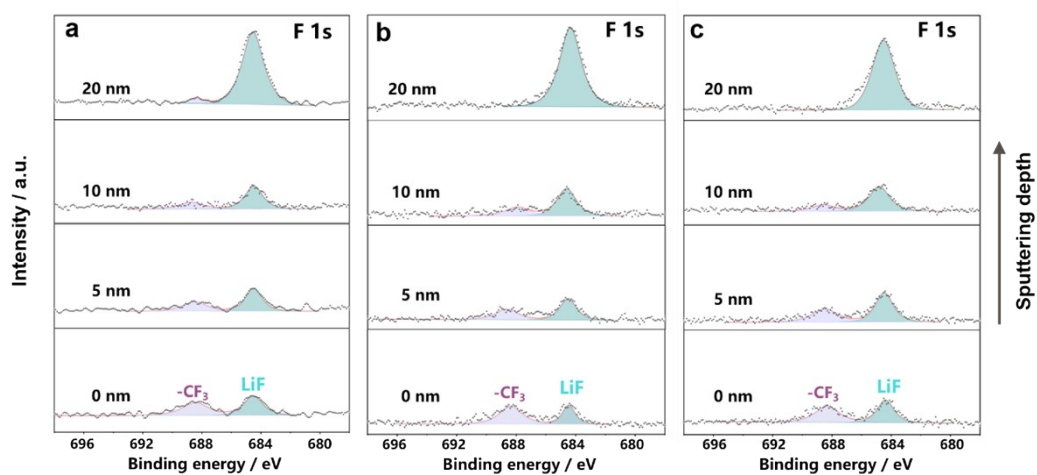


Fig. S7. F-1s HRXPS spectra of (a) LE, (b) PGPE and (c) PHGPE at different sputtering depths after 10 cycles of symmetric Li-ion battery. C-F: 688.7 eV, LiF: 684.5 eV.

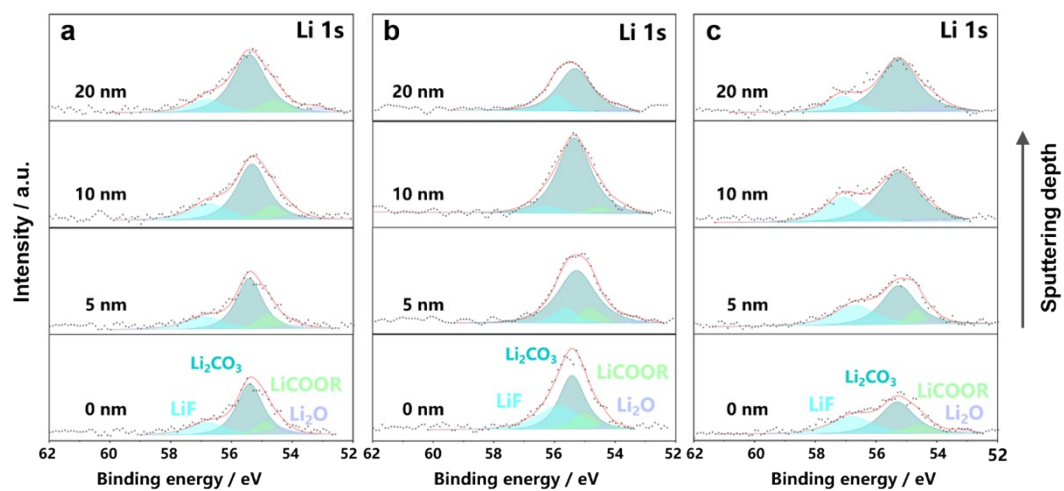


Fig. S8. Li-1s HRXPS spectra of (a) LE, (b) PGPE, (c) PHGPE at different sputtering depths after 10 cycles of symmetric Li-ion battery. LiF: 56.88 eV, Li₂CO₃: 55.38 eV, LiCOOR: 54.88 eV, Li₂O: 54.28 eV.

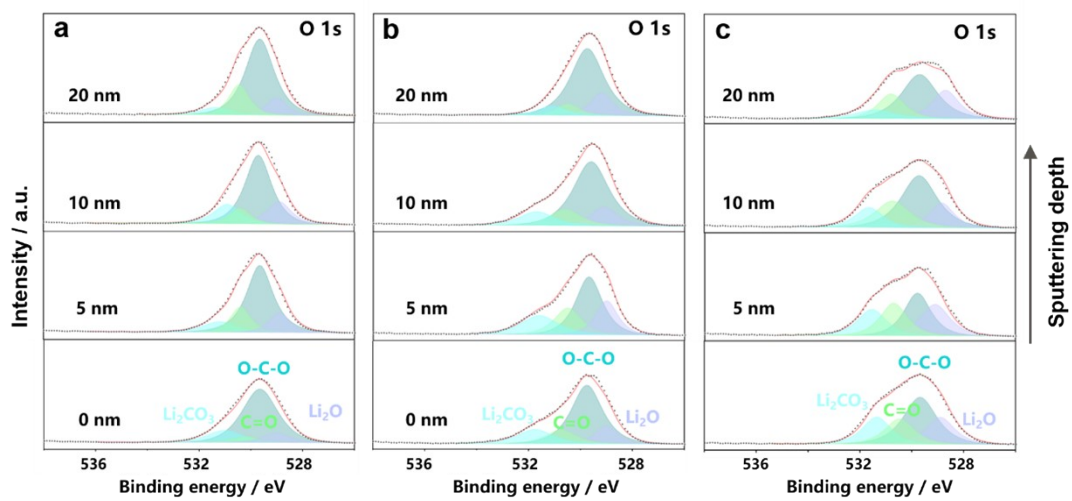


Fig. S9. O-1s HRXPS spectra of (a) LE, (b) PGPE, (c) PHGPE at different sputtering depths after 10 cycles of symmetric Li-ion battery.

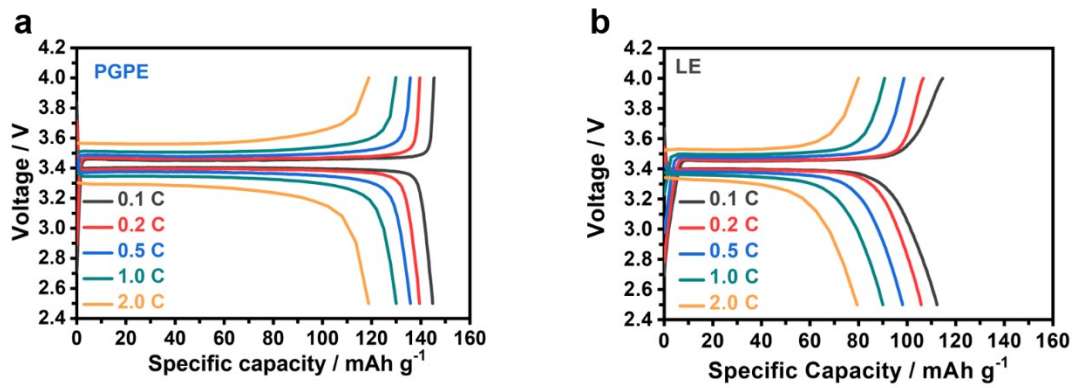


Fig. S10. Capacity voltage distribution of Li||electrolyte||LFP at different multipliers, (a) PGPE; (b) LE.



Fig. S11. Digital images of Li foil for nickel tabs

Table S1. The comparison of Li||Li symmetrical cell cycling performances between this work and other reported literatures.

Electrolyte	Current density / areal capacity (mA cm ⁻² / mAh cm ⁻²)	Plating/ stripping (hours)	Ref.
SPE1-PI-ZIF8	0.1/0.1	800 h	1
	0.3/0.3	300 h	
Li/CF composites	0.25/0.5	750 h	2
	0.5/0.5	560 h	
PEO-SCN-LiTFSI	0.05/0.05	600 h	3
Al ₂ O ₃ -GPE	1/1	1000 h	4
CSE-B- 71515	0.2/0.2	333 h	5
PDADMA-FSI	0.1/0.1	225 h	6
IPN9-10PPC	1.5/1.5	300 h	7
PIL-IPN	0.5/1.5	1800 h	8
POSS-4PEG2K	0.3/0.9	2600 h	9
GPR ₅ -LiTFSI	0.1/0.1	600 h	10
PVDF film	0.5/0.5	1400 h	11
	1/1	650 h	
	1/2	370 h	
Li ₃ PO ₄ /PVA layer	1/2	1000 h	12
PU-LiF	1/1	1000 h	13
	1/2	800 h	
PHGPE	1/1	6000 h	This work
	0.5/2	7700 h	
	1/2	7700 h	

Table S2. The specifications of the Li||LFP pouch cell.

Cell components	Specification	Parameters
Cathode (LFP)	Areal mass loading (mg cm^{-2})	11.71
	The area of cathode ($\text{mm} \times \text{mm}$)	50×83
	The content of LFP (wt.%)	96
	Folds	12
Anode (Lithium foil)	Thickness (μm)	60
	The area of lithium foil (mm^2)	55×88
	Folds	13
Electrolyte	1.0 M LiPF_6 in EC : DMC=1 : 1 V/V (contain 1.5% of the mass fraction with PHGPE)	
	The mass of electrolyte (g): capacity of pouch cell (Ah)	2:1
Pouch cell	Specific capacity (Ah)	1

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