Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information

Semi-interpenetrating polymer network electrolyte membrane for enhanced lithium-ion transport and interfacial stability in silicon based lithium-ion battery

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Number of page: 17

Number of figure: 11

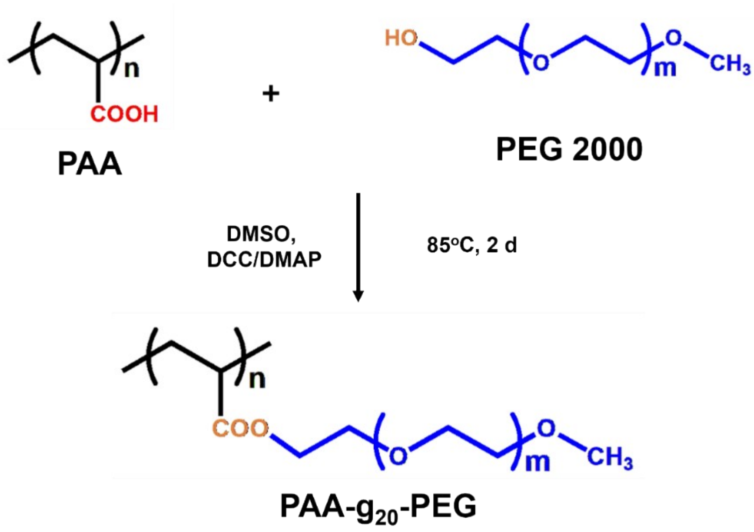
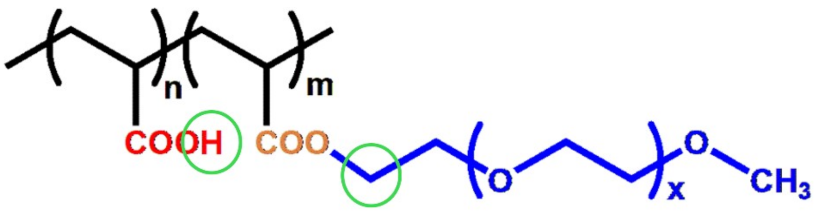


Figure S1. Synthetic scheme of PAA-g-PEG.

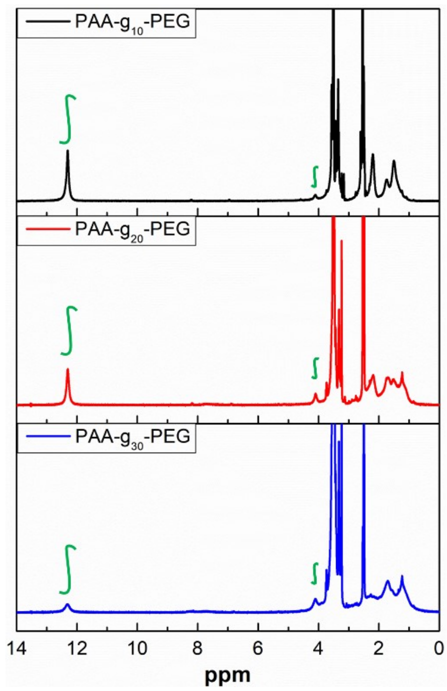
(a)



Grafting degree (%)

$$= \frac{\frac{Integration\ of\ CH_2}{Hydrogen\ number\ of\ CH_2}}{\frac{Integration\ of\ COOH}{Hydrogen\ number\ of\ COOH} + \frac{Integration\ of\ CH_2}{Hydrogen\ number\ of\ CH_2}} \times 100$$

(b)

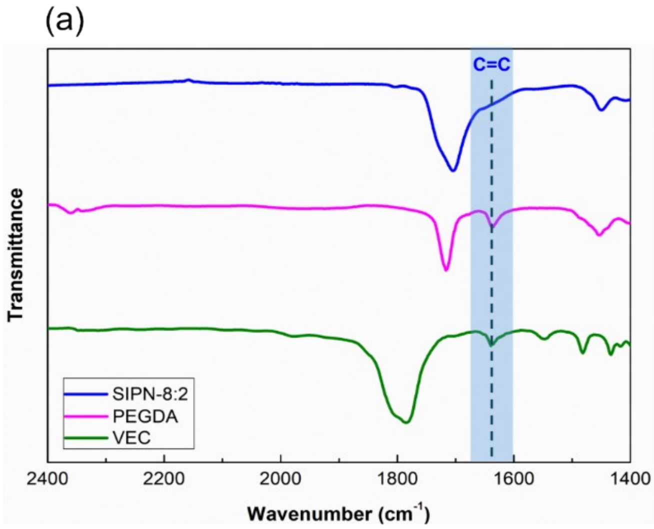


$$\frac{\frac{0.2}{2}}{\frac{1}{1} + \frac{0.2}{2}} \times 100 = 9.09\ \%$$

$$\frac{\frac{0.5}{2}}{\frac{1}{1} + \frac{0.5}{2}} \times 100 = 20\ \%$$

$$\frac{\frac{0.8}{2}}{\frac{1}{1} + \frac{0.8}{2}} \times 100 = 28.57\ \%$$

Figure S2. (a) Chemical structure of PAA-g-PEG and the equation used to calculate the PEG grafting degree; (b) ^{1}H-NMR spectra of PAA-g-PEG polymers with different PEG grafting ratios, calculated based on the integration ratio of PEG methylene protons to PAA backbone protons.



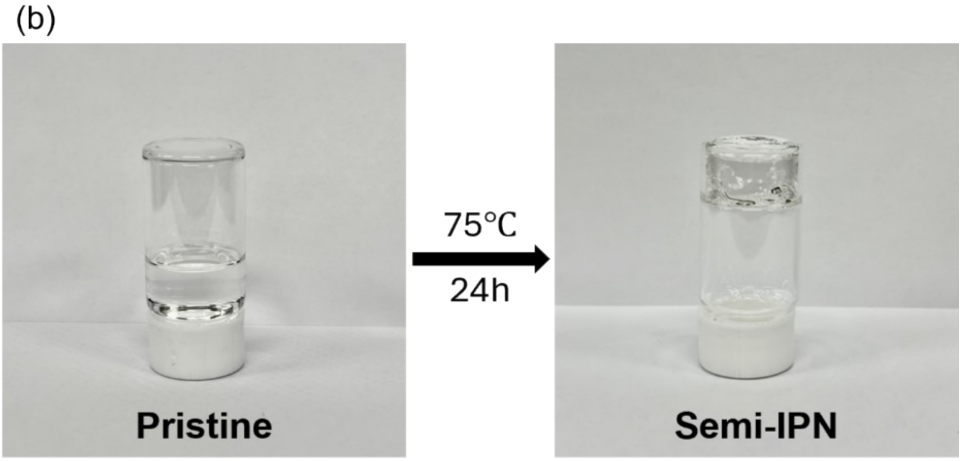


Figure S3. (a) FTIR spectra of PEGDA, VEC, and SIPN-8:2; (b) transformation into a SIPN structure after heat treatment at 75°C for 24 h.

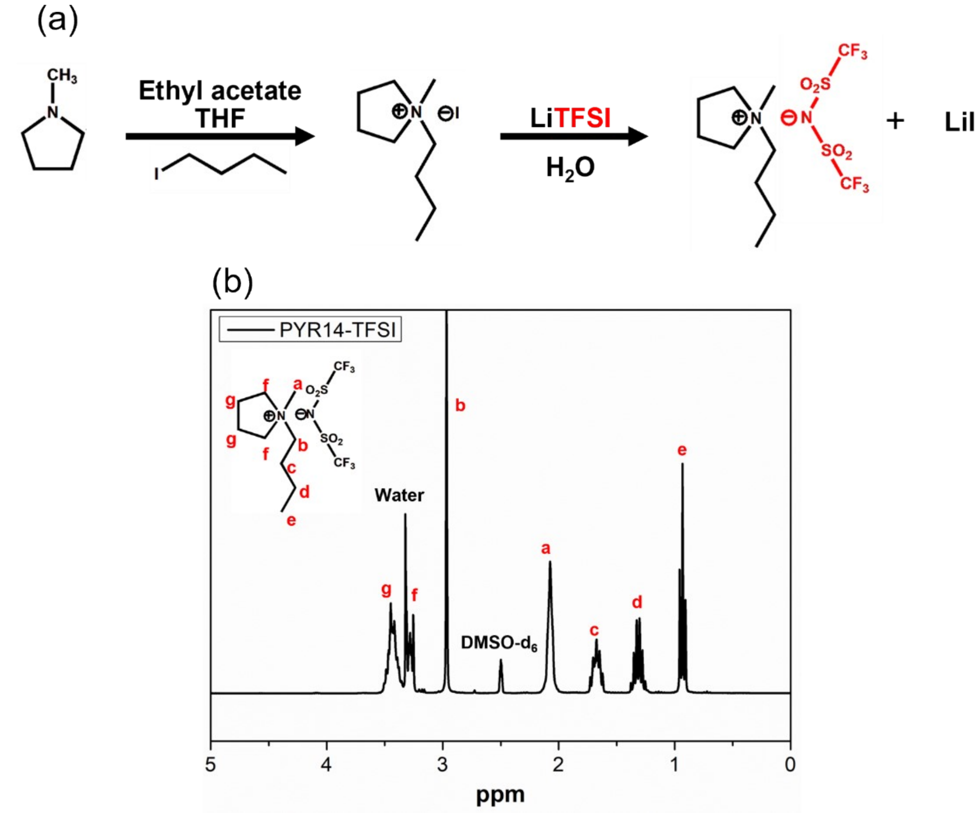


Figure S4. (a) Synthetic scheme of PYR14-TFSI; (b) ^{1}H-NMR of PYR14-TFSI.

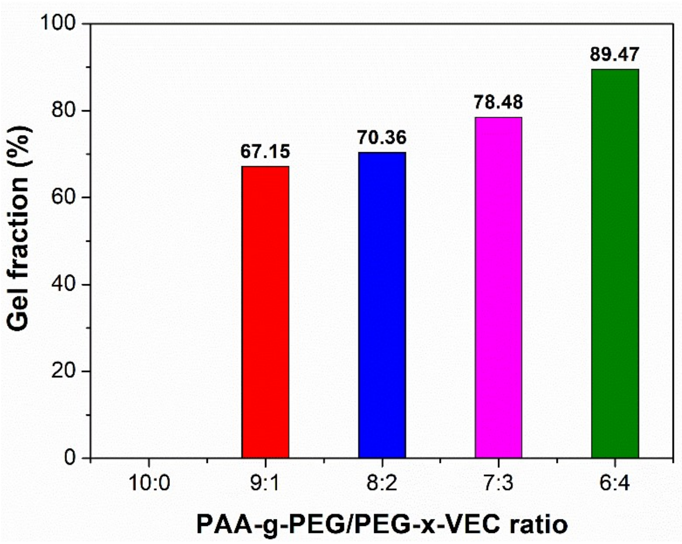


Figure S5. Gel fraction of SIPN membranes with varying PAA-g-PEG/PEG-x-VEC ratios.

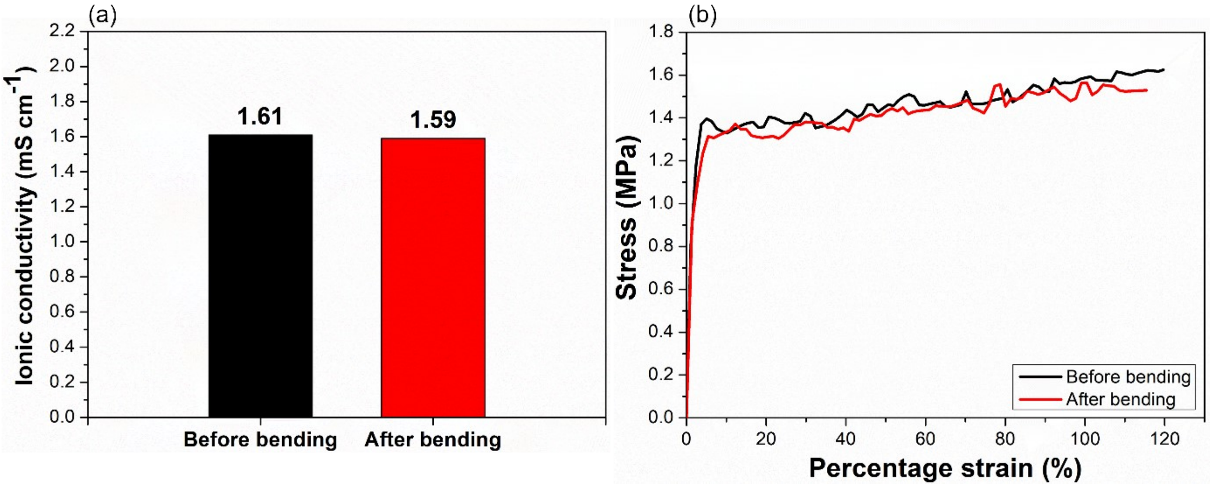


Figure S6. (a) Ionic conductivity and (b) stress–strain curves of the SIPN membrane before and after bending.

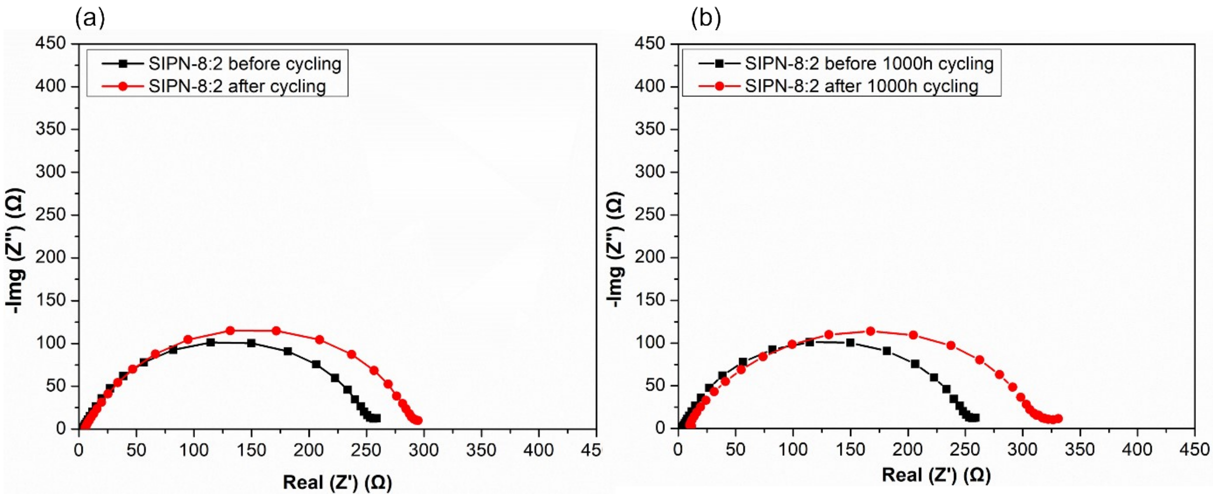


Figure S7. Nyquist plots of SIPN-8:2 cell before and after cycling at (a) various current densities, and (b) at 0.25 mA cm^{-2} current density of for 1000 h cycling.

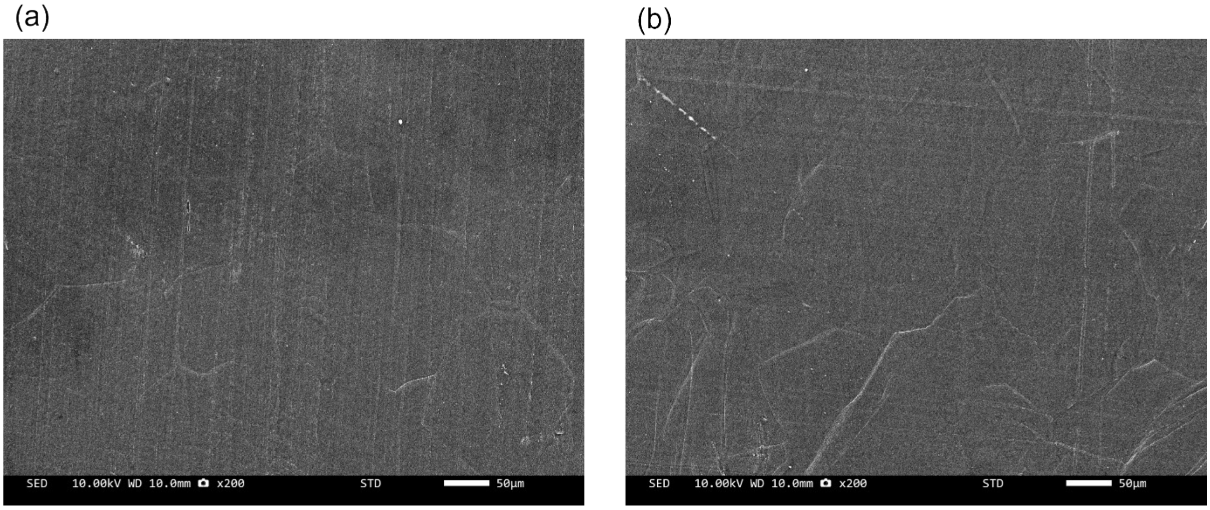


Figure S8. SEM images of lithium metal surfaces (a) before and (b) after cycling in the SIPN-8:2 membrane.

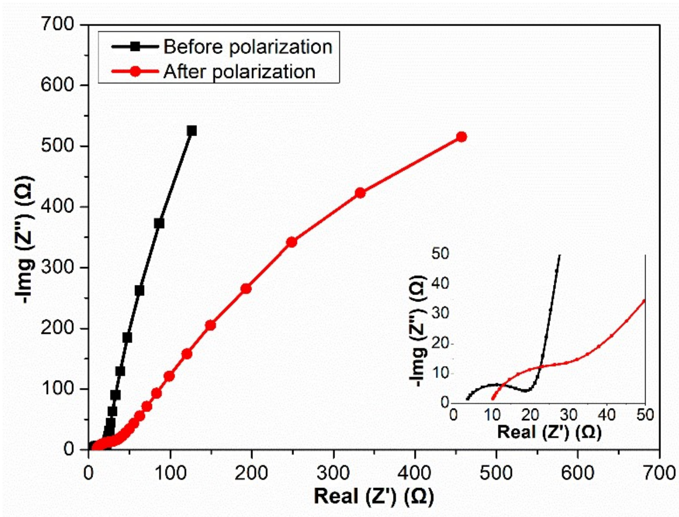


Figure S9. Nyquist plots measured before and after 300 cycles of Si/SIPN membrane/Li half cell.

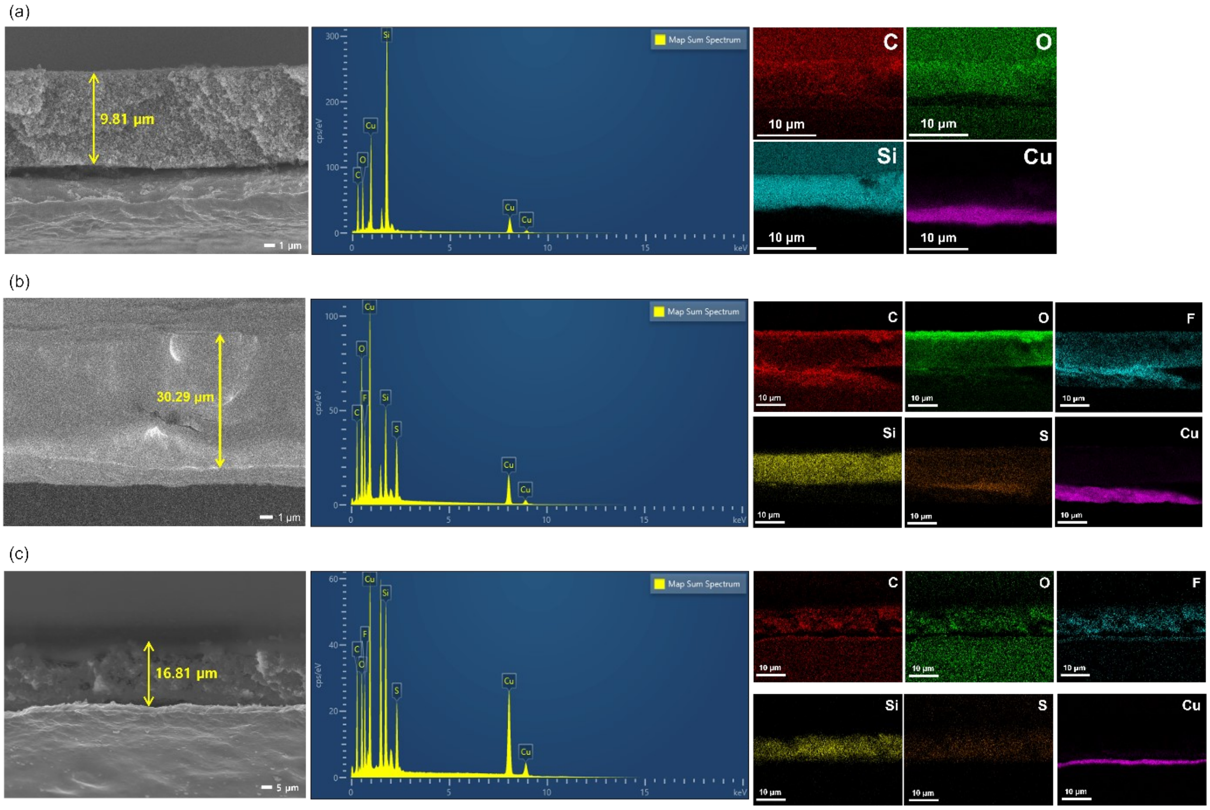


Figure S10. Cross-sectional FESEM images along with EDS mapping of silicon anodes (a) before cycling, and after 300 cycles with (b) liquid electrolyte and (c) the SIPN-8:2 membrane.

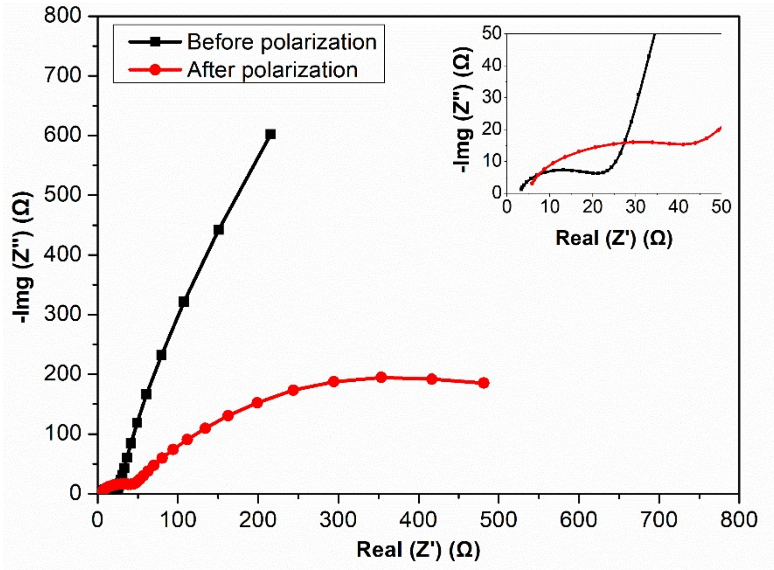


Figure S11. Nyquist plots measured before and after 300 cycles of Si/SIPN membrane/LFP full cell.

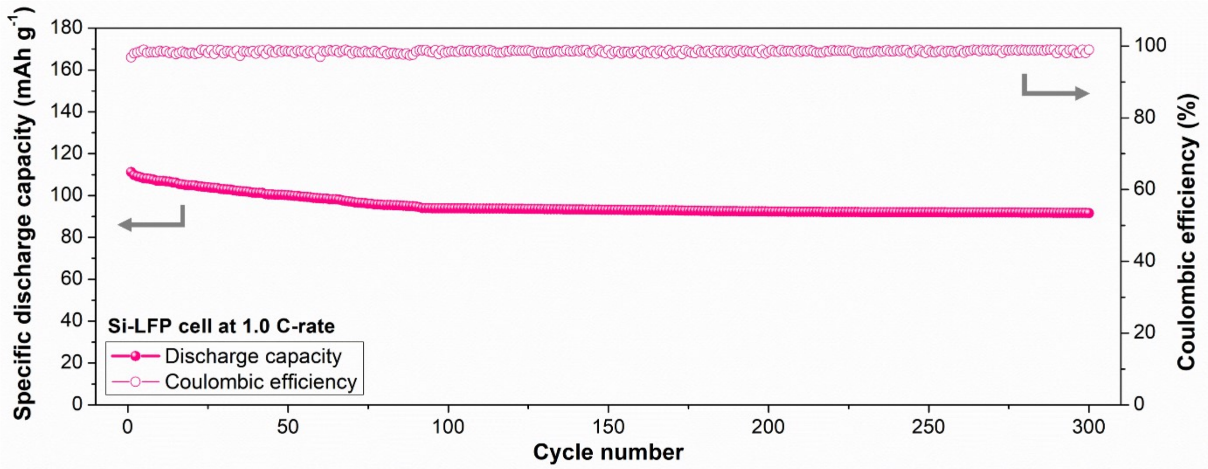


Figure S12. Cycling performance of Si//LFP cell assembled with SIPN-8:2 membrane at 1.0 C-rate.

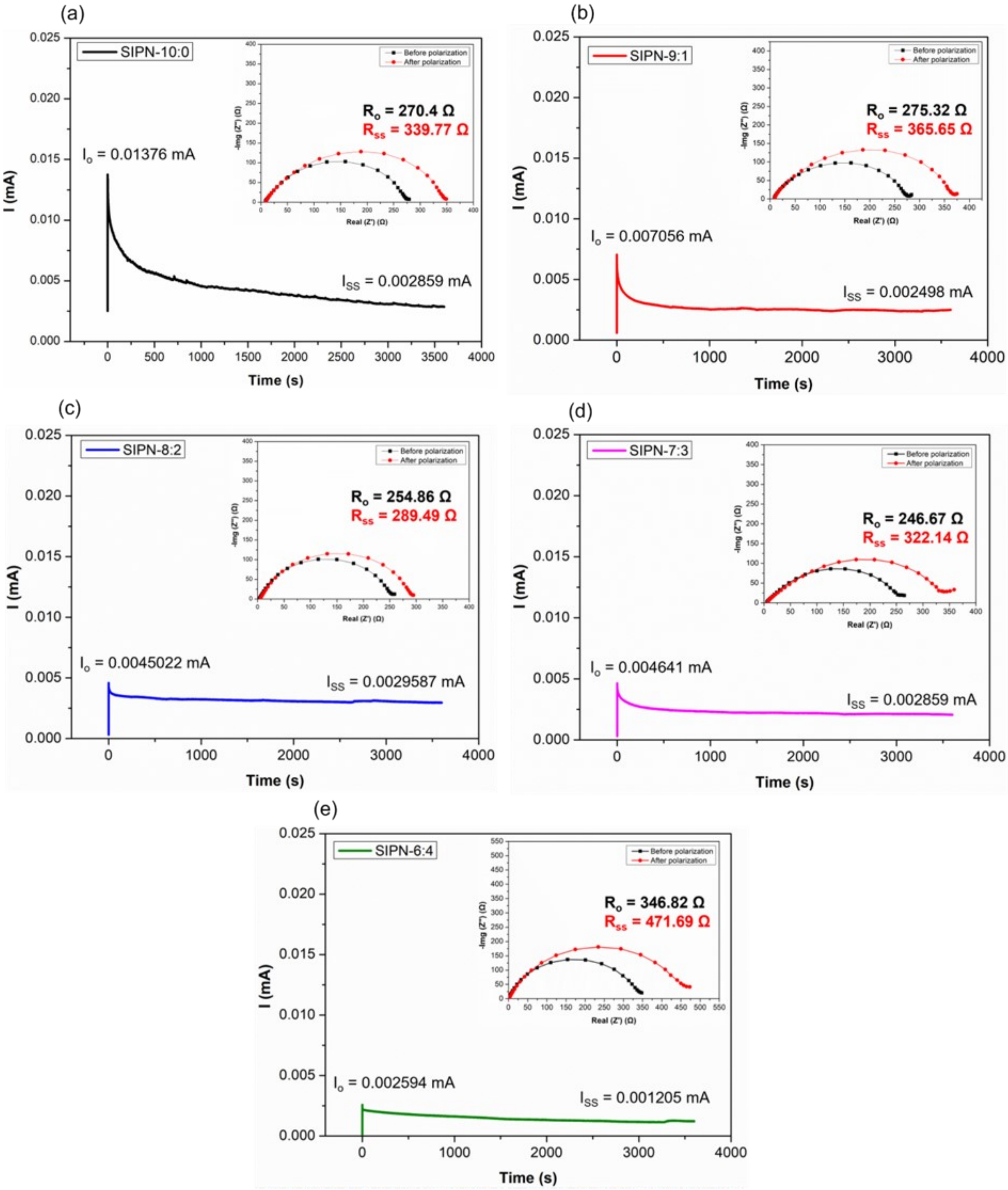


Figure S13. Chronoamperometry curves and interfacial resistance before and after polarization of SIPN membranes with different PAA-g-PEG/PEG-x-VEC ratios.

Table S1. Composition of SIPN membranes

Sample	PAA-g-PEG matrix (wt%)	Crosslinked network	
		PEGDA (wt%)	VEC (wt%)
SIPN-10:0	100	0	0
SIPN-9:1	90	5	5
SIPN-8:2	80	10	10
SIPN-7:3	70	15	15
SIPN-6:4	60	20	20

Table S2. Comparison of ionic conductivity and Li^+ transference number of the SIPN-8:2 membrane with other polymer electrolytes.

Sample	Temperature (°C)	σ (mS cm^{-1})	t_{Li+}	Reference
SIPN-8:2 solid electrolyte	RT	1.61	0.64	This work
PTEC-LiTFSI	RT	0.0112	0.39	[1]
P(EC/EO) *gel electrolyte*	60	0.48	0.66	[2]
BCP/LiTFSI/PEG_{250}	RT	0.6	0.35	[3]
PEO/TDI-TiO_2 *composite*	30	0.1	0.36	[4]
LiBOB/3-methoxysilyl-terminate PP	RT	0.26	0.65	[5]
PEO/IL/glass fiber *composite*	30	0.33	0.33	[6]

Table S3. Comparison of the performance of Si-based cells based on PAA-g-PEG/PEG-x-VEC membranes with those based on other recently reported electrolytes.

Full cell	Current density	Cycle number	Capacity retention (%)	Temperature (℃)	[Ref]
Si/In-situ PDOL-10FEC/LFP	0.5 C	300	76.3%	-	[7]
Si/PPC-garnet/LFP	0.1 C	100	82.6%	-	[8]
Si-PAN/SE/NMC811	0.1 C	100	77.7%	60℃	[9]
Si-Gr/TXE-based quasi-solid-state electrolyte/NCM622	0.5 C	200	86%	RT	[10]
Si/PHP-L15/LFP	1.0 C	100	81%	60℃	[11]
Si@$LiAlO_2$/NCM811@Li_2SiO_x	0.5 C	150	77%	-	[12]
Si/LLZTO/LFP	8 μA cm^{-2}	100	72%	RT	[13]
Si@SiO_2@LPO@C /PEO@LATP/ NMC811	0.5 A g^{-1}	200	44%	-	[14]
Si−C/SE/NCM	0.2 mA cm^{-2}	50	87.7	-	[15]
Si/PAA-g-PEG/PEG-x-VEC/LFP	0.5 C	300	80.6%	RT	This work

Reference

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