

Supporting Information

All-Solid-State Interpenetrating Network Polymer Electrolytes for Long Cycle Life of Lithium Metal Batteries

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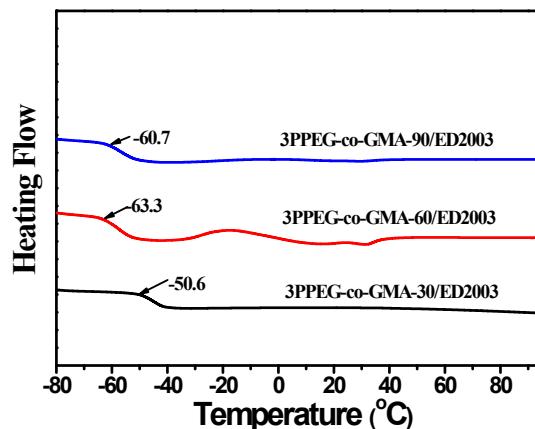


Fig. S1 DSC thermograms of cross-linking solid state polymer based on 3PPEG-co-GMA-x and ED2003 recorded under nitrogen during the second heating scans at a scan rate of $10\text{ }^{\circ}\text{C min}^{-1}$, the molar ratio of the epoxy and amino is fixed 2:1.

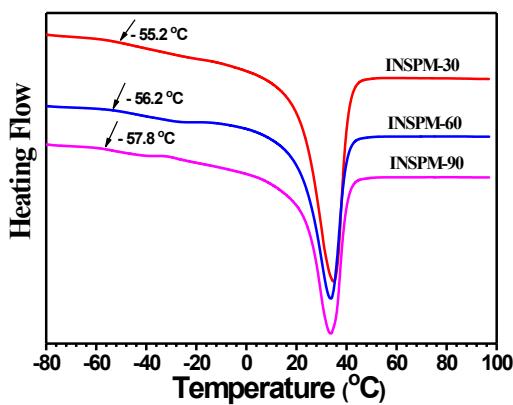


Fig. S2 DSC thermograms of interpenetrating network solid state polymer membrane (INSPM-x) recorded under nitrogen during the second heating scans at a scan rate of $10\text{ }^{\circ}\text{C min}^{-1}$.

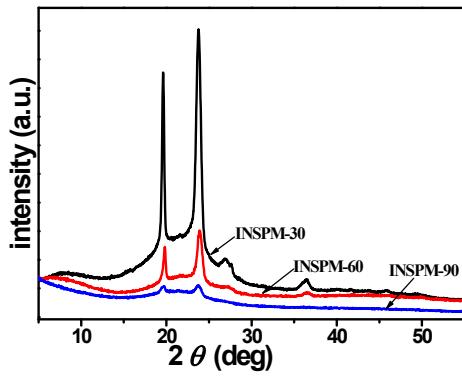


Fig. S3 XRD spectra of interpenetrating network solid polymer membrane (INSPM-x).

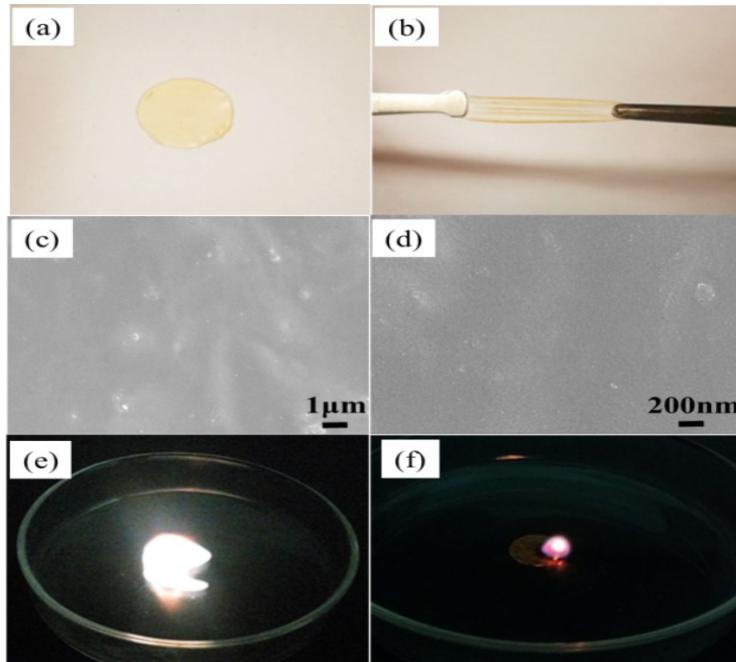


Fig. S4 Photographs (a) and (b), SEM images (c) and (d) of INSPM-60-LiTFSI, photographs demonstrating flammable behavior of liquid electrolyte (e) and INSPM-60-LiTFSI (f).

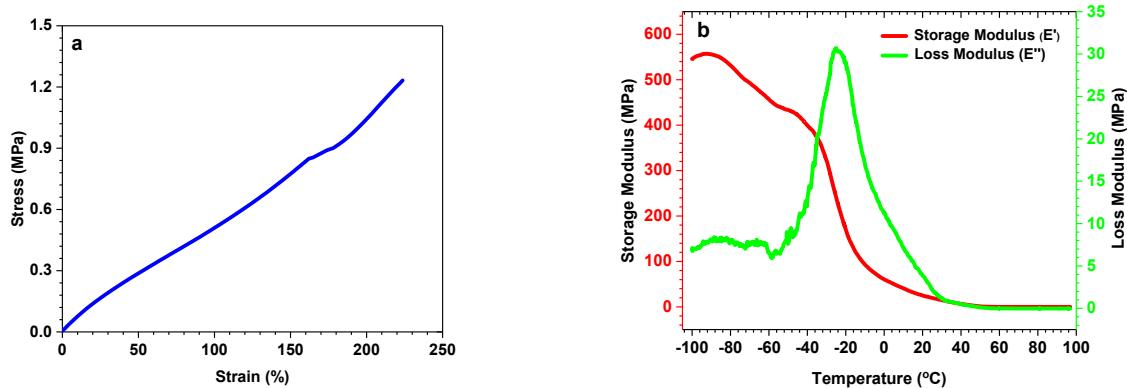


Fig. S5 (a) Stress-strain measurement of INSPM-60 at 25°C with force ramp rate of 0.2 N/min and (b) storage and loss modulus measurement of INSPM-60 at a heating rate of 3 K/min .

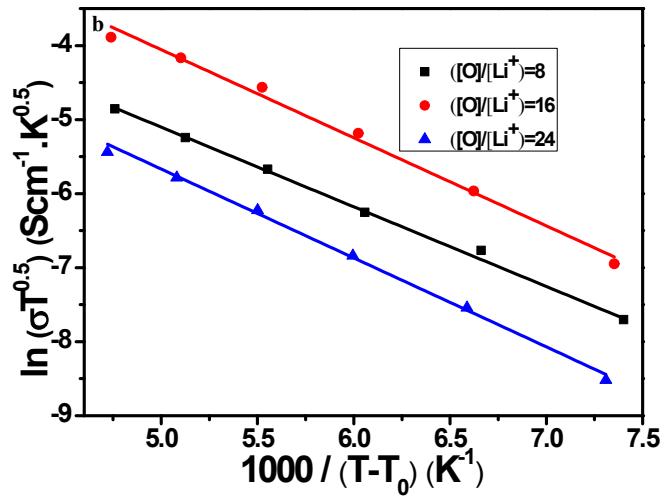


Fig. S6 VTF fitting curves of INSPM-60-LiTFSI with various $[O]/[Li^+]$.

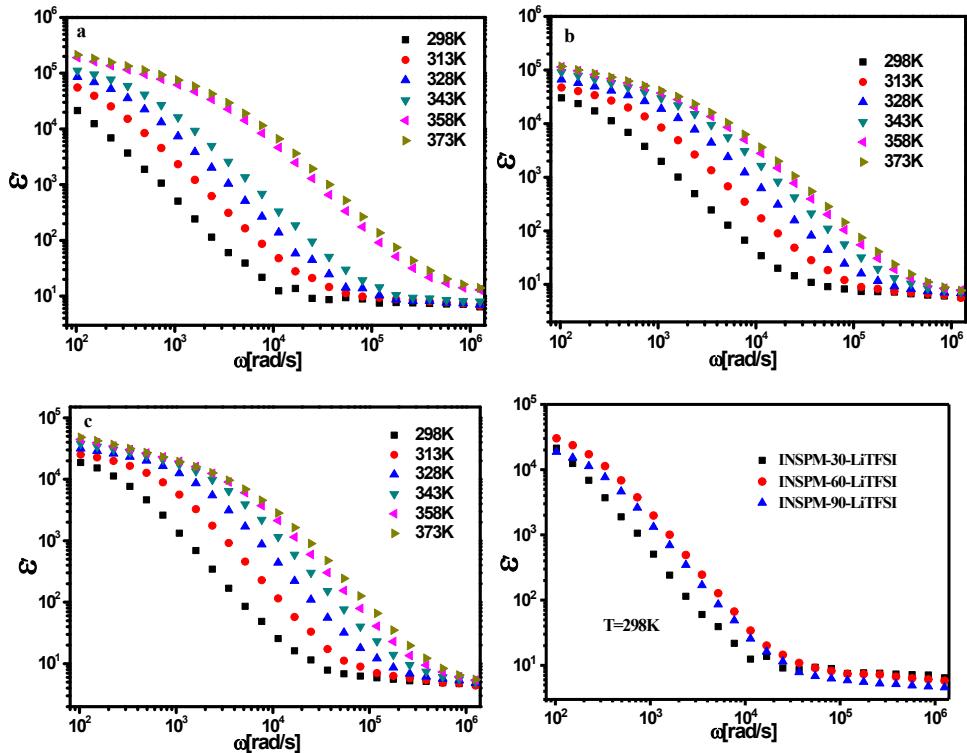


Fig. S7 Dielectric permittivity (ϵ') spectra as functions of angular frequency ω of (a) INSPM-30-LiTFSI, (b) INSPM-60-LiTFSI, (c) INSPM-90-LiTFSI at different temperature and (d) a comparison of the three samples at 298 K.

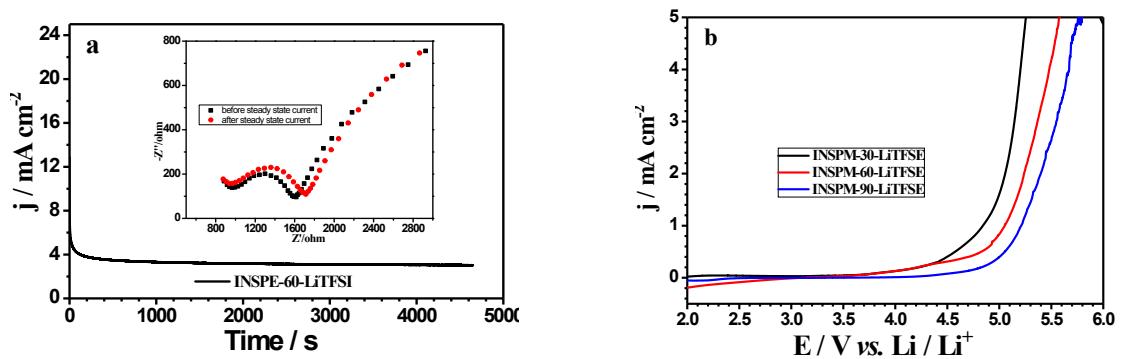


Fig. S8 (a) Chronoamperometry profile of INSPM-60-LiTFSI; (b) linear sweep voltammetry curves of the cell based on INSPM-x-LiTFSI electrolyte at 60 °C, $[\text{O}] / [\text{Li}] = 16$.

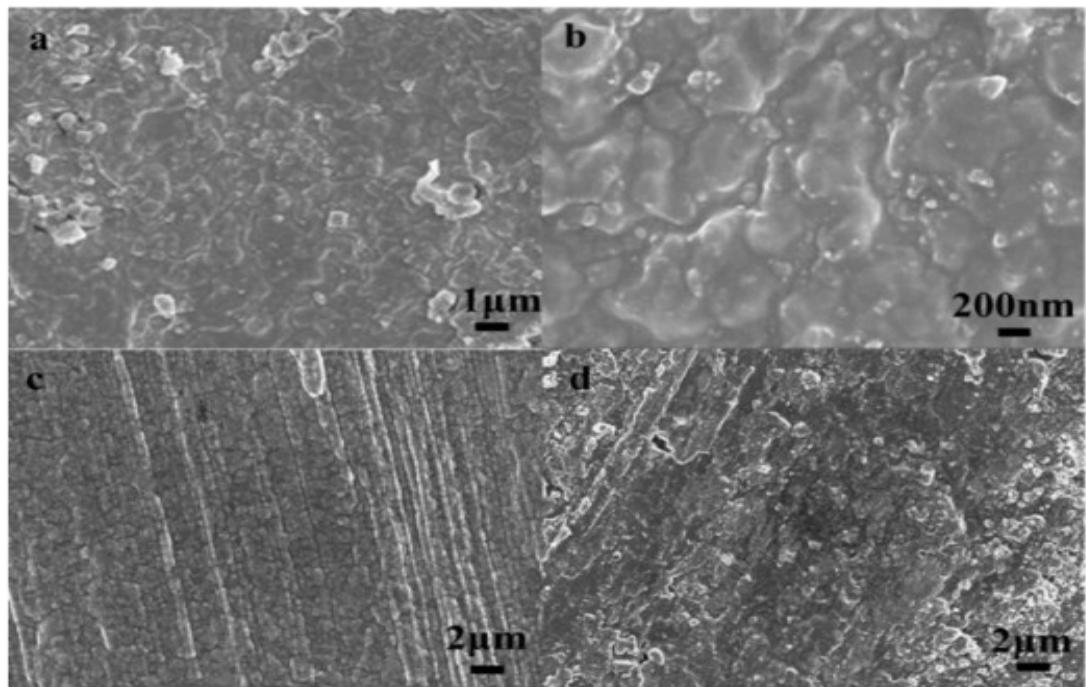


Fig. S9 SEM images of the INSPM-60-LiTFSI membrane (a, b) and Li electrode (c, d) in a $\text{Li} \parallel \text{INSPM-60-LiTFSI} \parallel \text{Li}$ cell before (a, c) and after (b, d) 2200 cycles.

Table S1 Thermal property, ionic conductivity, and VTF fitting parameters of the INSPM-60/LiTFSI electrolytes

[O]/ [Li ⁺]	T _g , °C	T _m (°C)	ΔH _m (J/g)	E _a (kJ/mol)	A (Scm ⁻¹ K ^{0.5})	t ₊	σ/S cm ⁻¹ 25°C	σ/S cm ⁻¹ 100°C
8	-54.9	23.3	-40.4	8.95	0.28	-	2.1×10 ⁻⁵	3.3×10 ⁻⁴
16	-55.4	26.4	-50.5	9.89	1.89	0.37	5.6×10 ⁻⁵	1.1×10 ⁻³
24	-56.9	27.7	-67.9	9.99	0.34	-	9.5×10 ⁻⁶	1.8×10 ⁻⁴

A is a pre-factor that is often related to the number and mobility of charge carriers, *E_a* is the activation energy

Table S2 Comparison of the electrochemical performance based on solid-sate electrolytes.

Reference	Ionic conductivity (S cm ⁻¹)	The initial discharge capacity (mAh g ⁻¹)	Test condition	Electrolyte
1	2.5 × 10 ⁻⁵	120-130 / 0.05C	30 °C	PEC-LiTFSI
2	2.2 × 10 ⁻⁴	141 / 0.5C	25 °C	ipn-PEA/LiPF ₆
3	~1.4 × 10 ⁻³	~140 / 0.2C	25 °C	PEM/SCN/LiTFSI
4	~10 ⁻⁵	160 / 0.1C	70 °C	TBP-3/LiTFSI
5	1.1×10 ⁻⁴	137.1 / 0.5C	25 °C	SPEM/LiTFSI
6	4.0×10 ⁻⁴	/	25 °C	PTL-3/LiTFSI
This work	5.6×10 ⁻⁵	156.2 / 0.1C	60 °C	INSPM-60/LiTFSI

All the batteries were assembled with lithium metal as anode and LiFePO₄ as cathode.

Supplementary References

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