Electronic Supplementary Material

Crosslinked ether-based polymer for high performance semi-solid lithium metal battery via in-situ integration

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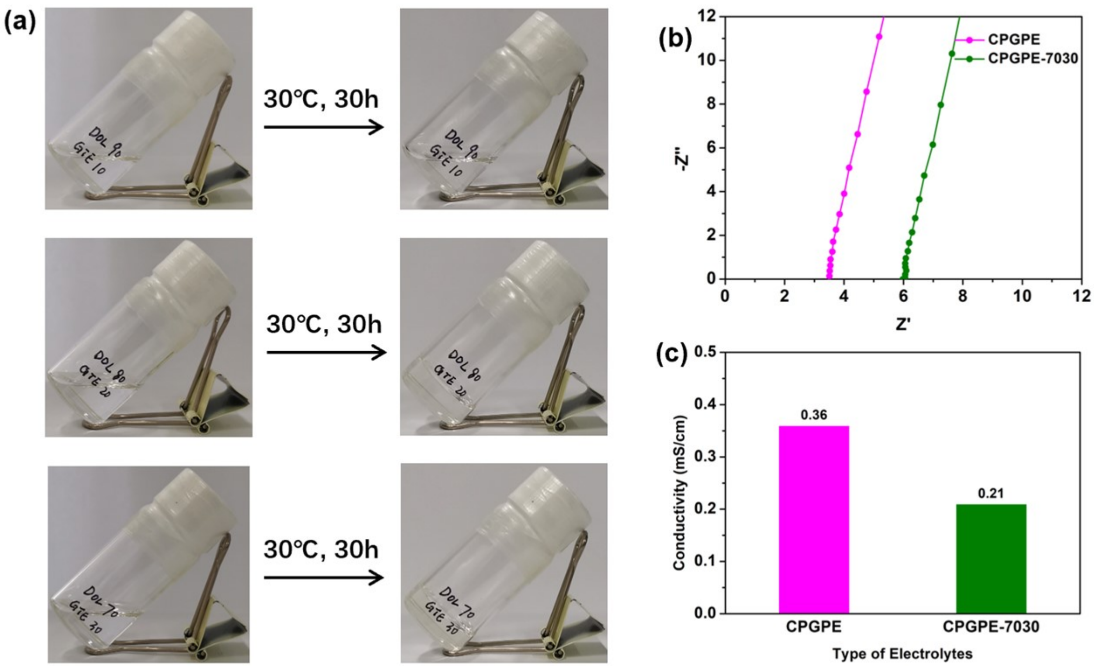


Fig. S1 (a) Curing situation of different volume ratio between DOL and GTE in the precursor solvent formula of 5(DOL(*x*)-GTE(*100-x*)-2HFE-2FEC-1EMC. (b, c) Comparison of the ionic conductivity of different GTE ratio in the electrolyte.

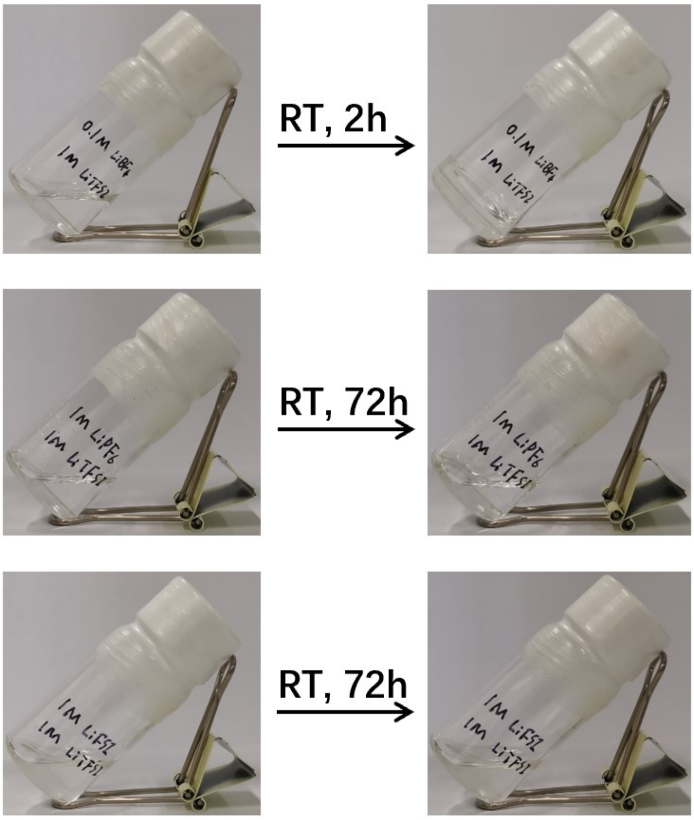


Fig. S2 Effect of initiators ($LiBF_4$, $LiPF_6$, and LiFSI) on the crosslinked polymerization of DOL and GTE (80:20, v/v) with 1M LiTFSI.

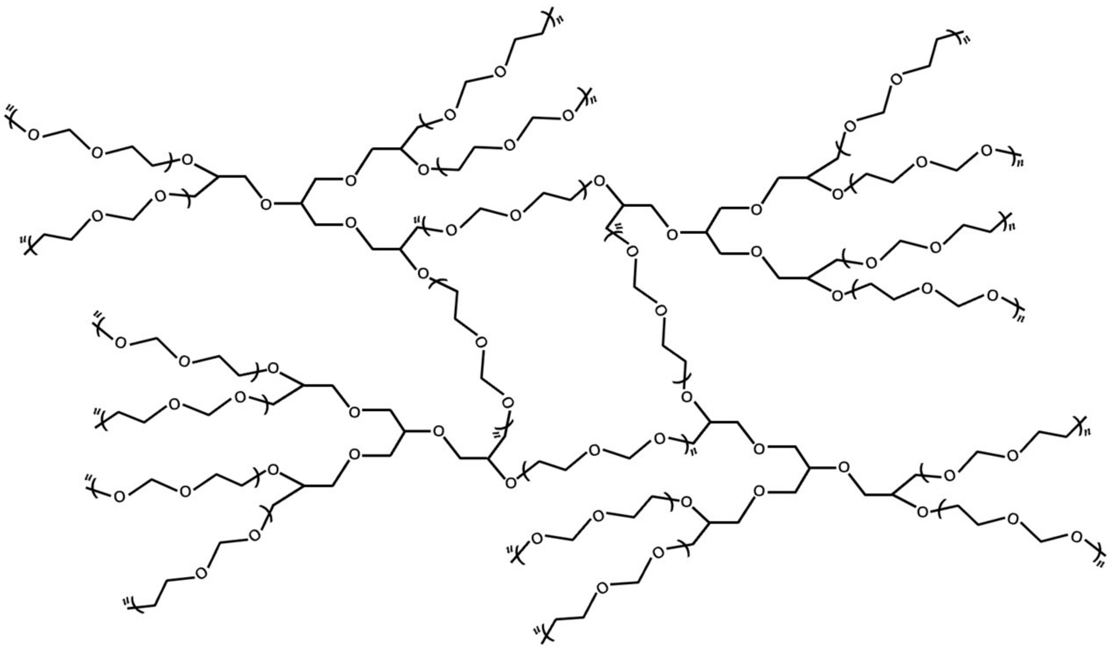


Fig. S3 The possible polymerization product of DOL and GTE.

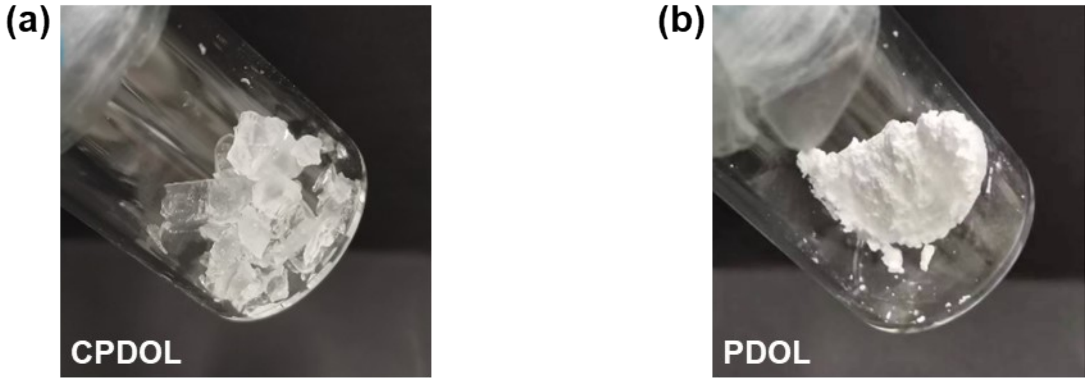


Fig. S4 Purified polymer obtained from repeated ultrasonic cleaning of (a) CPGPE and (b) PGPE with water and ethanol.

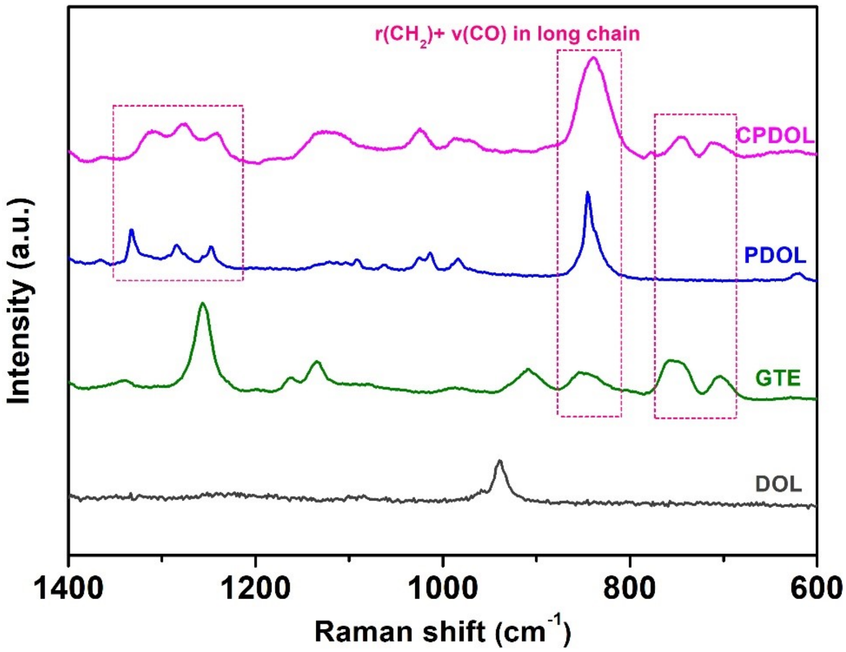


Fig. S5 Raman spectra of DOL, GTE, PDOL, and CPDOL.

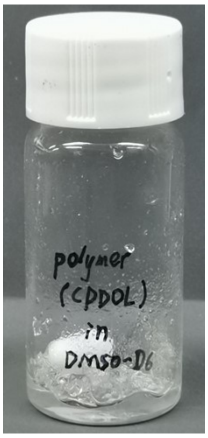


Fig. S6 (a) Optical image of the dissolving state of CPDOL in DMSO-D6.

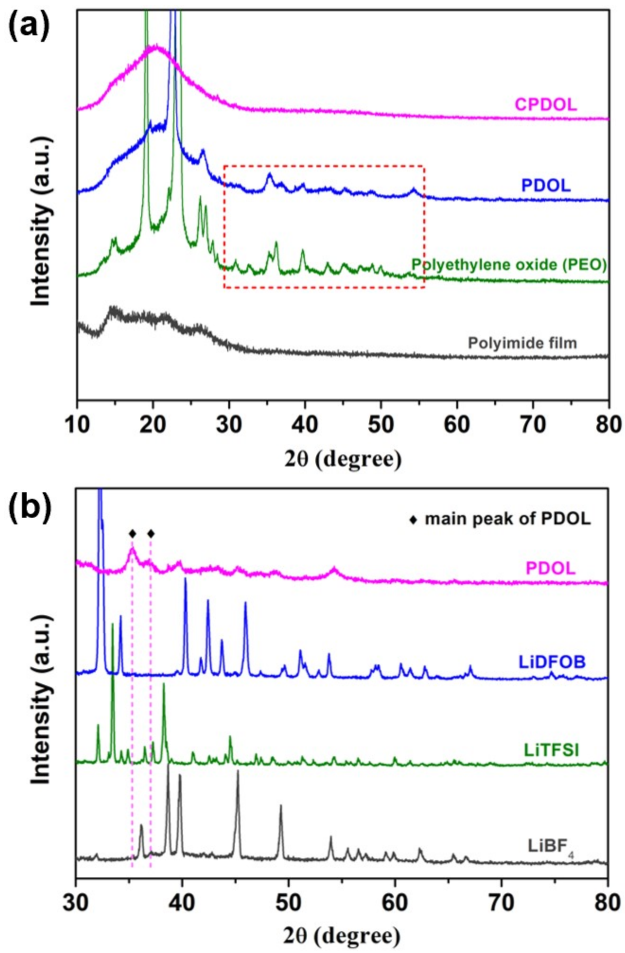


Fig. S7 XRD pattern of (a) CPDOL, PDOL, and PEO, (b) PDOL and various lithium salts used in the electrolyte.

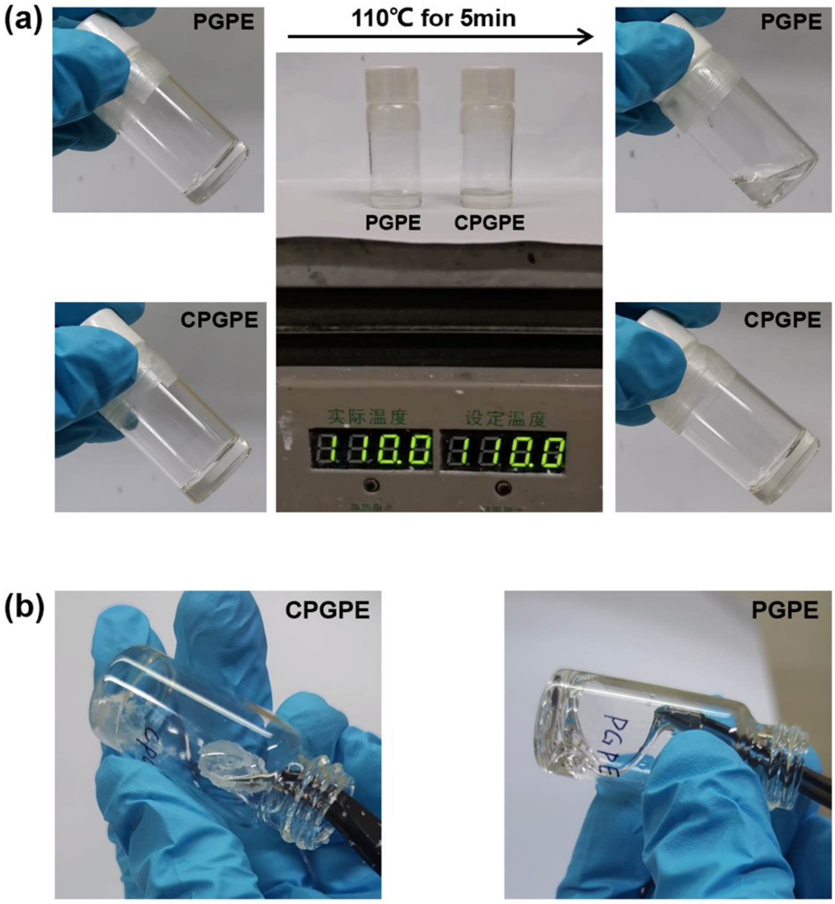


Fig. S8 (a) Optical images of CPGPE and PGPE before and after heated at 110 °C for 5 minutes. (b) Physical characteristic of CPGPE and PGPE.

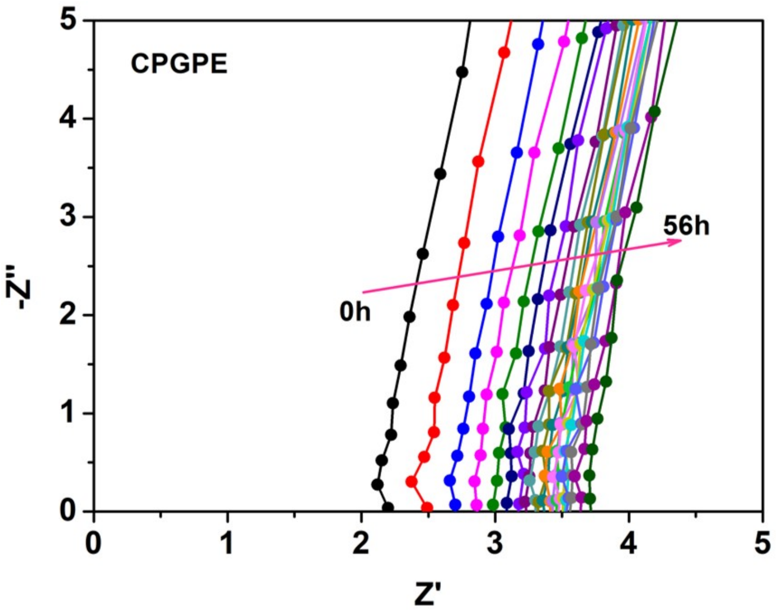


Fig. S9 Change of EIS plot of the SS|CPGPE|SS cell over time.

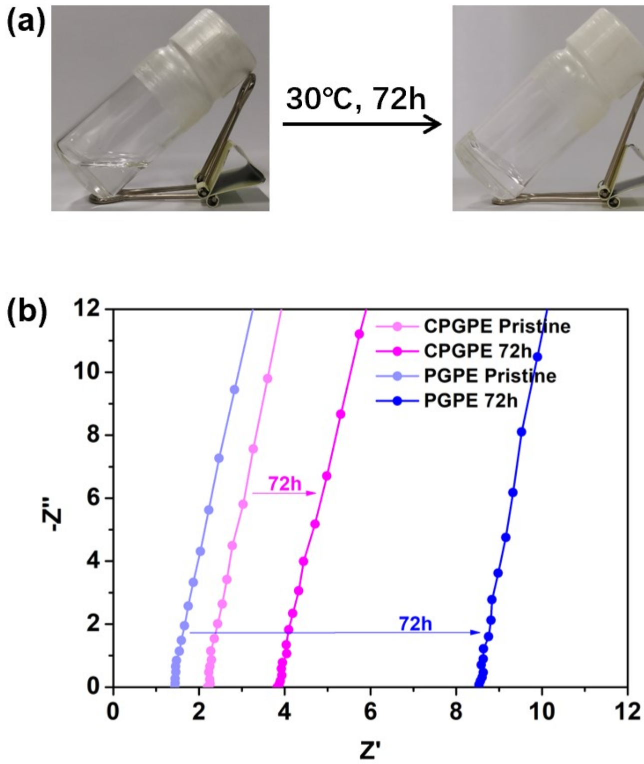


Fig. S10 (a) Optical image of the curing process of PGPE. (b) Comparison of the EIS plot of SS|CPGPE|SS and SS|PGPE|SS cells before and after solidification.

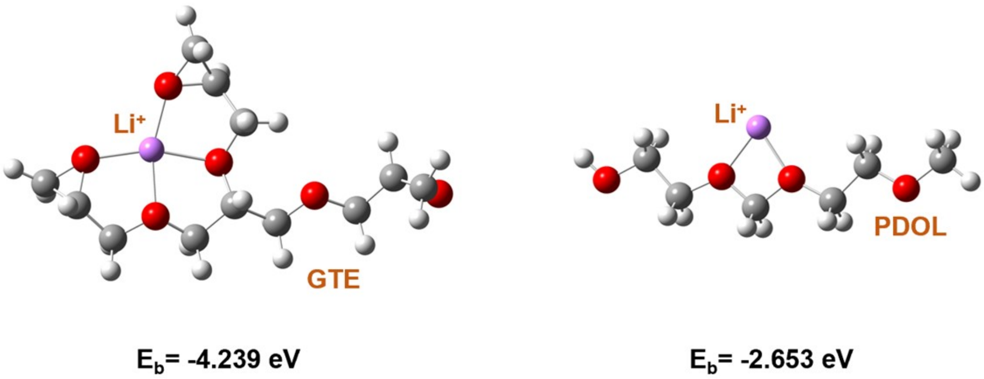


Fig. S11 (a) Schematic diagram of the interaction of GTE and Li^+. (b) Schematic diagram of the interaction of PDOL and Li^+.

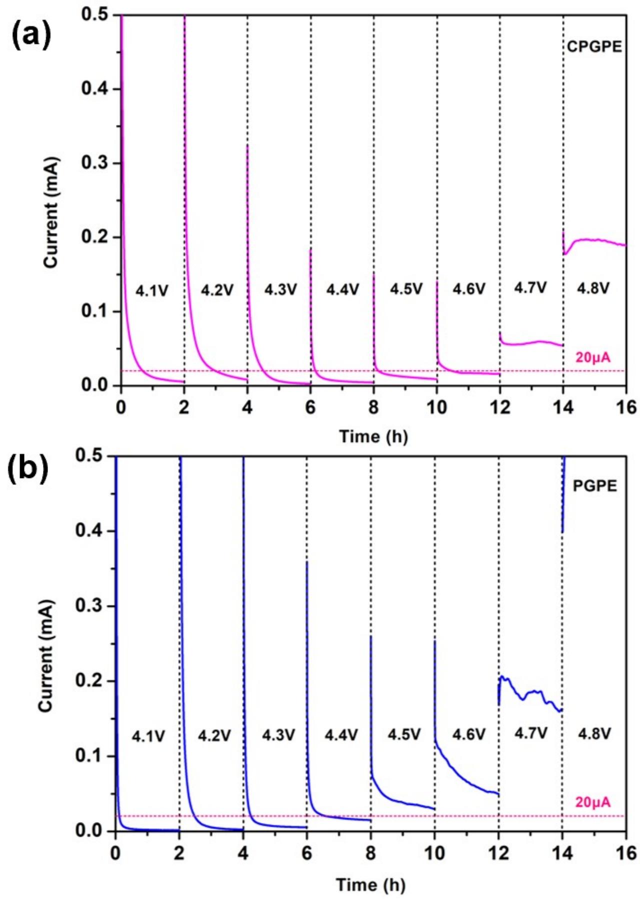


Fig. S12 Electrochemical floating test of the NCM811|CPGPE|Li and NCM811|PGPE|Li batteries.

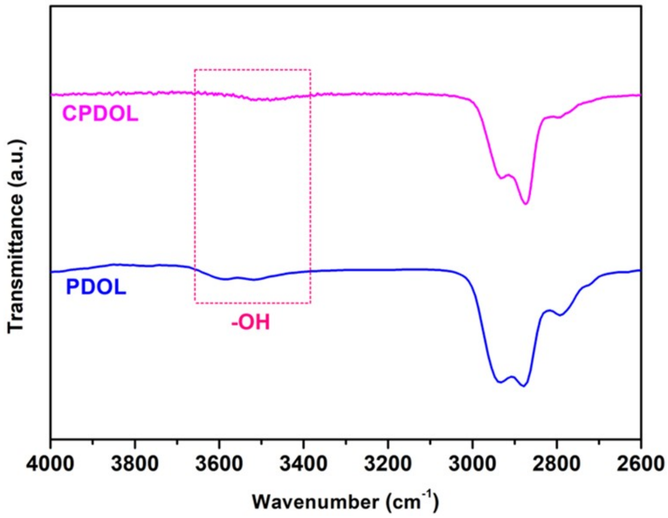


Fig. S13 Comparison of hydroxyl characteristic peaks of CPDOL and PDOL.

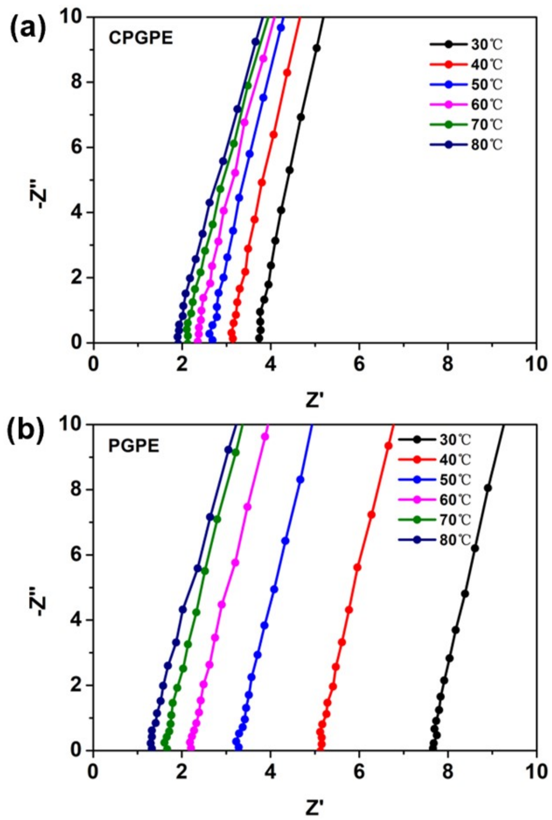


Fig. S14 Change of EIS plot of the (a) SS|CPGPE|SS and SS|PGPE|SS cells over temperature.

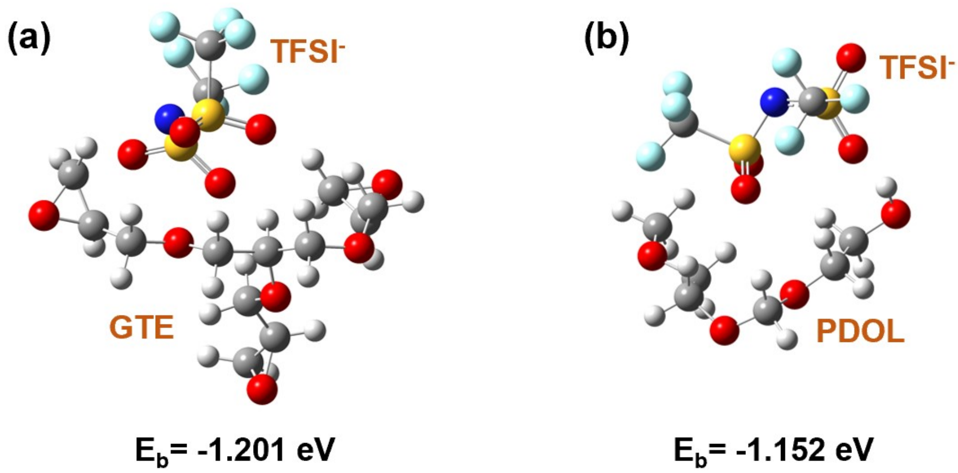
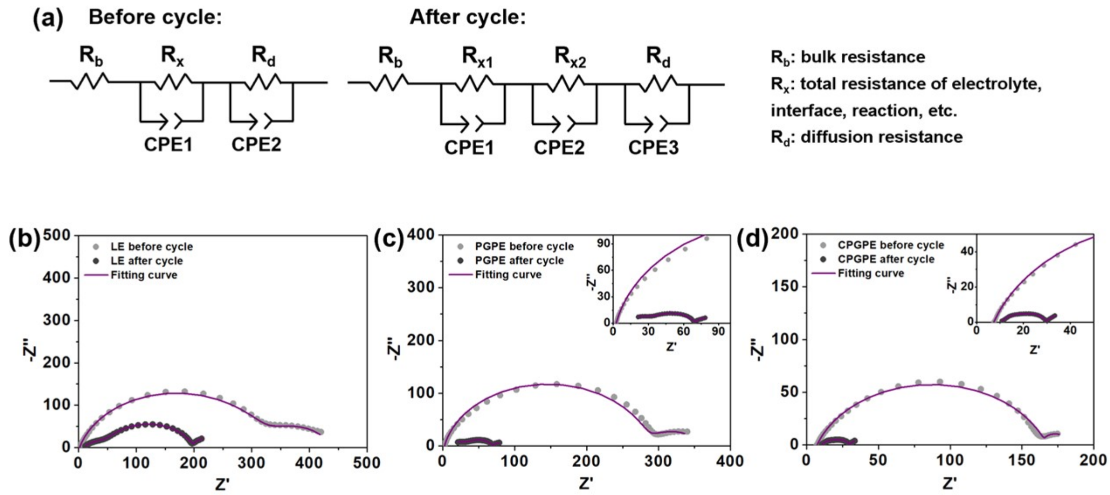


Fig. S15 (a) Schematic diagram of the interaction of GTE and $TFSI^-$. (b) Schematic diagram of the interaction of PDOL and $TFSI^-$.



(e)

	LE	PGPE	CPGPE
R_{x1} (Ω)	39.33	19.93	15.23
R_{x2} (Ω, interface resistance)	151.4	37.01	3.912
$R_{x2}/(R_{x1}+R_{x2})$	79.4%	65%	20.4%

Fig. S16 (a) Equivalent circuits for the EIS plots of the initial and cycled Li||Li cells. EIS plots and its corresponding fitting curves of the initial and cycled (b) Li|LE|Li cell, (c) Li|PGPE|Li cell, and (d) Li|CPGPE|Li cell. (e) The calculated ratio of interface resistance in the total resistance of mid-frequency region for different types of cycled Li||Li cells.

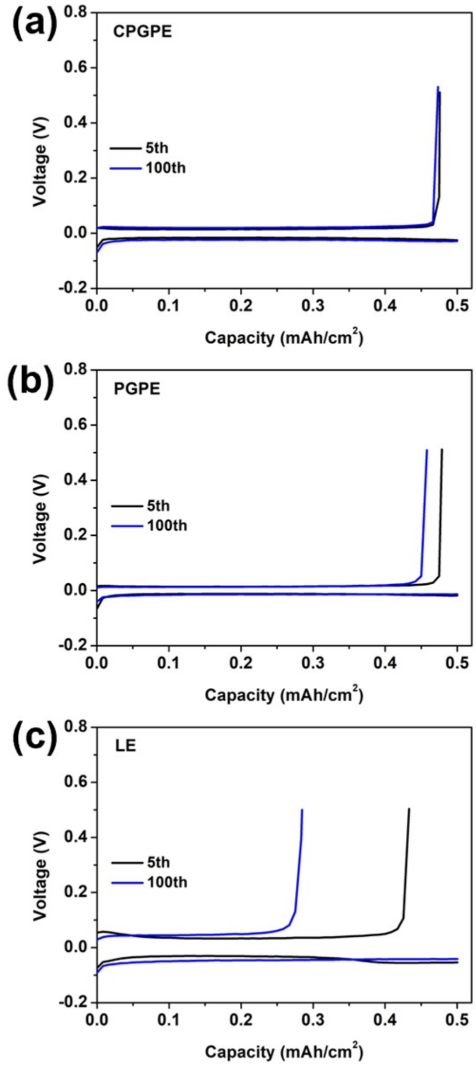


Fig. S17 Charge/discharge curves of the (a) Li|CPGPE|Cu, (b) Li|PGPE|Cu, and (c) Li|LE|Cu cell at different cycle number.

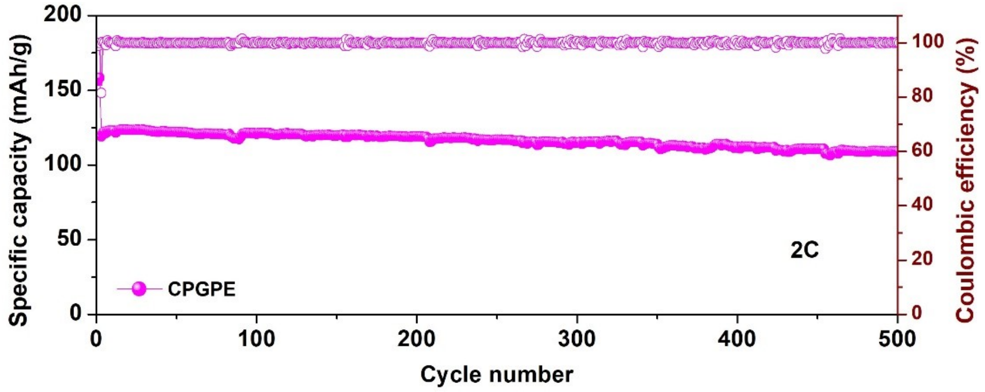


Fig. S18 Cycling performance of the LFP||Li battery assembled with CPGPE at 2C.

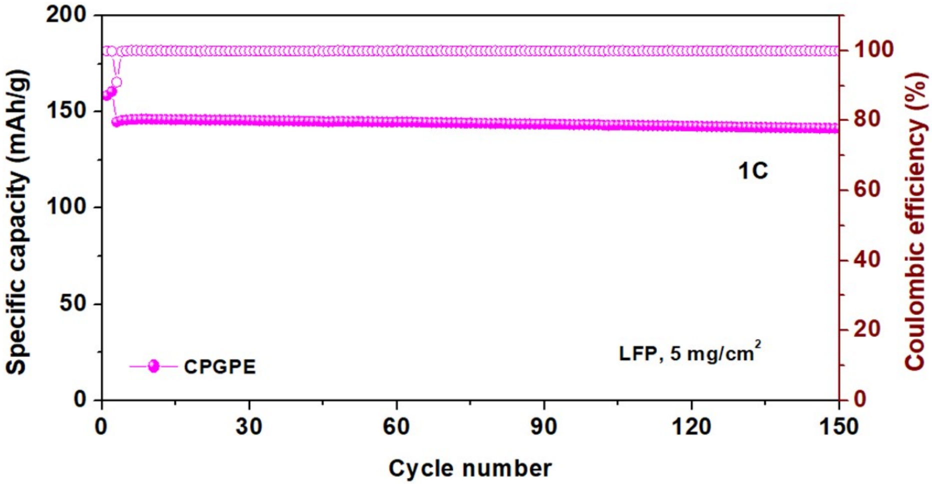


Fig. S19 Cycling performance of the LFP|CPGPE|Li battery with a high active materials loading of 5 mg/cm^2 at 1C.

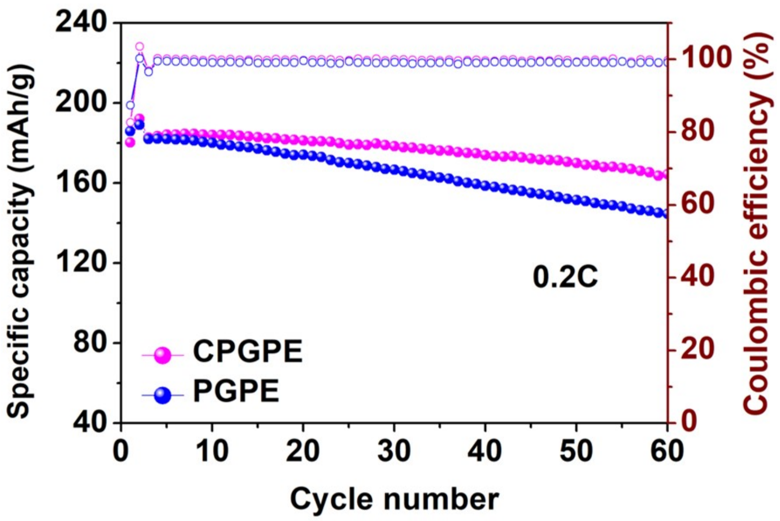


Fig. S20 Cycling performance of the NCM811||Li batteries assembled with CPGPE and PGPE at 0.2C.

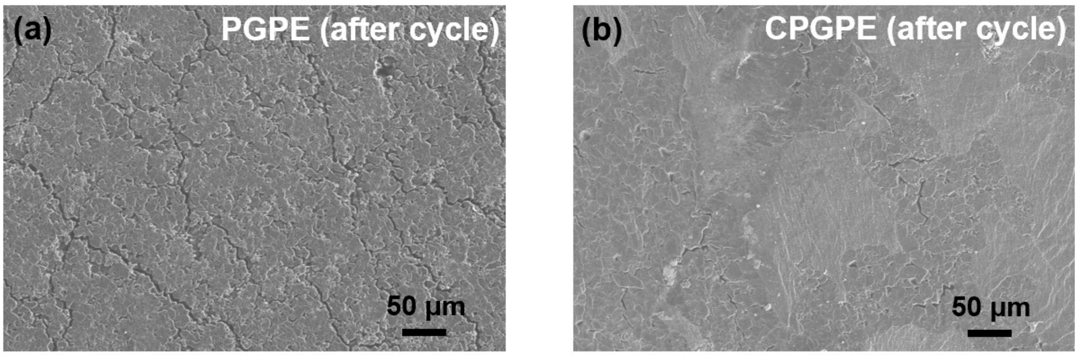


Fig. S21 SEM images of the lithium anode from the NCM811||Li batteries assembled with (a) PGPE and (b) CPGPE operating for 120 cycles.

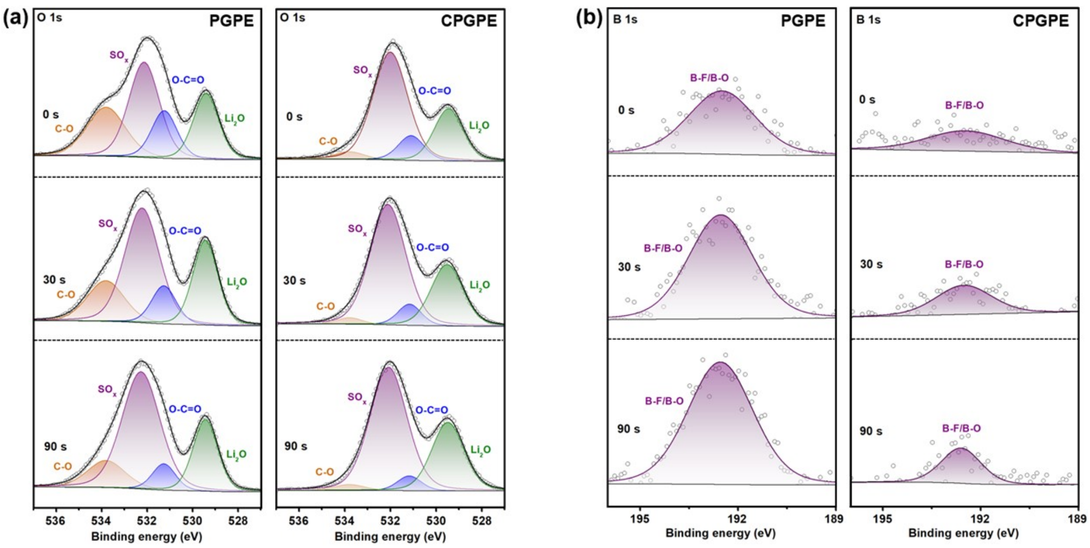


Fig. S22 XPS (a) O 1s spectra and (b) B 1s spectra of the washed lithium anode from the cycled NCM811||Li battery assembled with CPGPE and PGPE.

Table S1 A comparison of the electrochemical performance of this work with the similar PDOL based electrolyte in recent years

Electrolyte materials	**Physical characteristic**	**Cathode materials**	**Active materials loading (mg/cm²)**	**Cycle rate (C)**	**Cycle number@capacity retention**	**Ref.**
Poly(DOL-TTE)	All-solid	LFP	-	0.2	200cycle@80%	1
PTADOL	All-solid	LFP NCM811	3 3	0.5 0.5	300cycle@85.6% 150cycle@81.4%	2
PSiDOL	All-solid	LFP NCM811	2.5	0.5 0.3	160cycle@97% 100cycle@90%	3
PDOL/SN/FEC	Gel	NCM811	3	1	100cycle@70.59%	4
PDOL/SN	Gel	LFP	2	1	1000cycle@83.55%	5
KMSP@PDOL/DME	Gel	LFP	2.5	1	500cycle@90.3%	6
PDOL/DME/FEC	Gel	LFP	4	0.5 2	200cycle@～90% 1000cycle@77.6%	7
PDOL/DOL	Gel	LFP	4.2	1	500cycle@69.28%	8
PDOL/DME	Gel	LFP	10	1	700cycle@～60%	9
PDOL/DOL/$LiNO_3$	Gel	LFP	3	0.5 2	450cycle@72.74(50℃) 500cycle@76.9%	10
PDOL/DOL/PDA/PVDF-HFP	Gel	LFP	2.2	1 2	200cycle@87.13% 800cycle@83.2%	11
This work	**Gel**	**LFP** **LFP** **NCM811**	**2** **2** **2.5**	**1** **2** **0.5**	**1000cycle@88%** **500cycle@91.7%** **165cycle@80.2%**	

References

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