

Supplementary materials for

**Crosslinked gel polymer electrolyte based on multiply-epoxy groups enable conductivity and electrochemical performance for Li-Ion Batteries**

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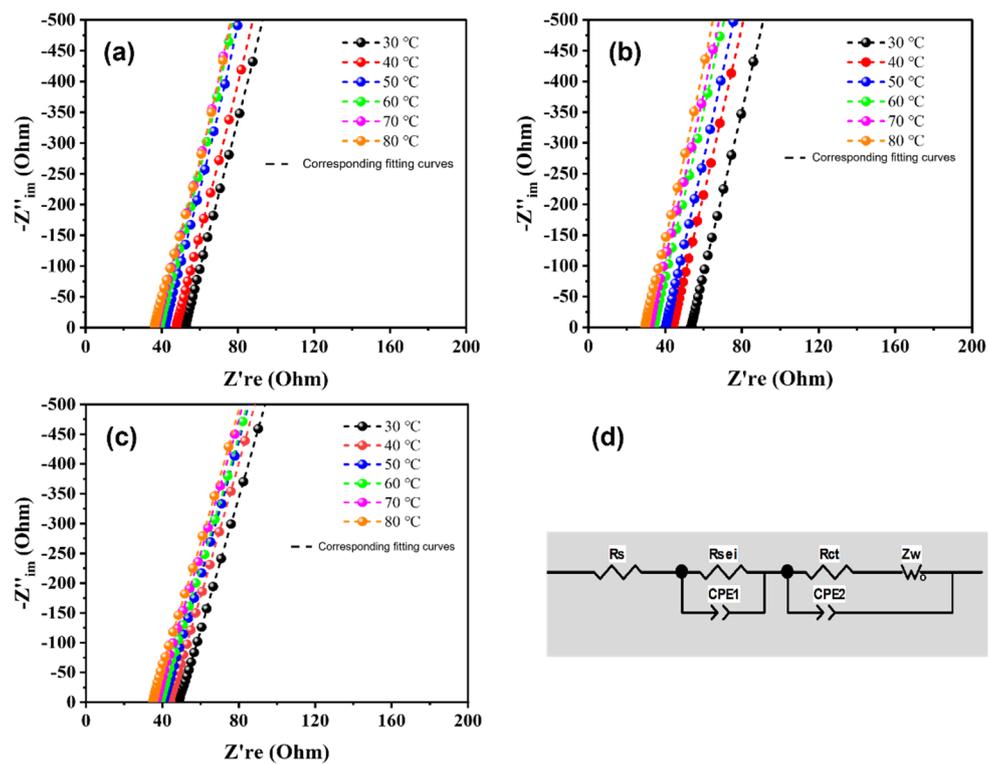


Figure S1. Nyquist plots of as-prepared C-GPE-1(a), C-GPE-2(b), and C-GPE-3(c), and corresponding an equivalent circuit(d).

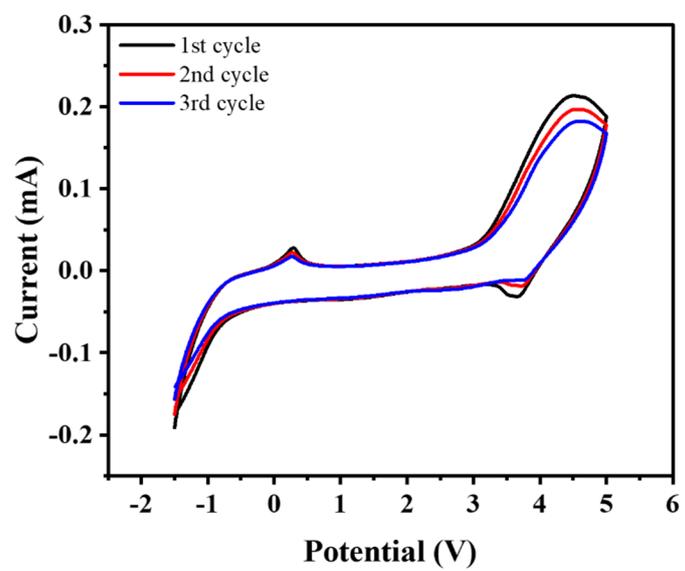


Figure S2. CV profiles of as-prepared C-GPE-4.

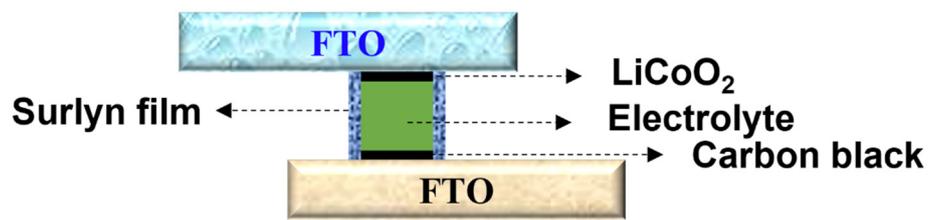


Figure S3. The schematic illustration of an asymmetric dummy cell.

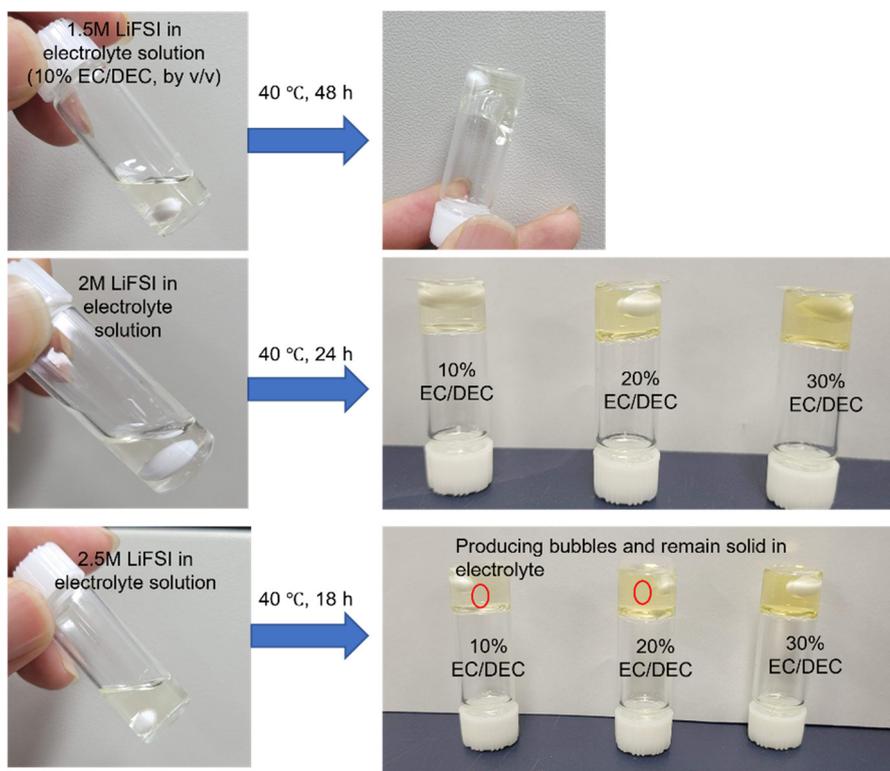


Figure S4. The digital photos of in-situ polymeric C-GPEs with varies concentrated LiFSI.

Table S1. Comparison of electrochemical properties of polymer electrolyte with epoxy group from reported works.

Electrolyte components	Ionic conductivity (S/cm at 30 °C/ RT <sup>a</sup> )		LSV (V, Li/Li <sup>+</sup> )	Battery performances	Ref.
TMPEG, NPEG, LiTFSI	1.10×10 <sup>-4</sup>		5.4	LFP, 144.8 mAh/g, 0.2C	[1]
PEE, LiTFSI, LiDFOB	2.36×10 <sup>-3</sup>		4.5	NMC622, 160 mAh/g, 0.5C	[2]
NGDE, LiFSI	1.57×10 <sup>-3</sup>		4.0	LFP, 131 mAh/g, 0.2C	[3]
PEGDGE, LiTFSI, LiBF <sub>4</sub>	1.10×10 <sup>-4</sup>		5.5	LFP, 160.2 mAh/g, 0.05C	[4]
GMA, OE, LiClO <sub>4</sub>	HPGO-70	2.26×10 <sup>-6</sup>	4.0-4.5	Li, 99 mAh/g, 0.1C	[5]
	PGO-70	2.58×10 <sup>-7</sup>			
PGTE, LiDFOB	4.16×10 <sup>-4</sup> at 60 °C		5.0	LFP, 135.2 mAh/g, 0.2C	[6]
C-GPE-2, LiFSI	0.23×10 <sup>-3</sup>		5.19	LFP, 161.3 mAh/g, 0.1C	<b>This work</b>

**Note:** 1) <sup>a</sup> Room temperature (RT)

2) LiFePO<sub>4</sub> (LFP), Trimethylolpropane triglycidyl ether (TMPEG), Poly (ethylene glycol) diamine (NPEG), Lithium bis(trifluoromethane)sulfonilimide (LiTFSI), Pentaerythritol glycidyl ether (PEE), LiNi<sub>0.6</sub>Mn<sub>0.2</sub>Co<sub>0.2</sub>O<sub>2</sub> (NMC622), Lithium difluoro(oxalate)borate (LiDFOB), Neopentyl glycol diglycidyl ether (NGDE), Poly(ethylene glycol) diglycidyl ether (PEGDGE), Lithium tetrafluoroborate (LiBF<sub>4</sub>), Glycidyl methacrylate (GMA), Oligo (ethylene oxide) methyl ether methacrylate (OE), Lithium perchlorate (LiClO<sub>4</sub>), Polymerized glycerin triglycidyl ether (PGTE)

Table S2. The composition of the as-prepared crosslinked-gel polymer electrolyte in this work

<b>Crosslinked-Gel Polymer electrolytes (C-GPEs) names</b>	<b>ESO (mL)</b>	<b>LiFSI (g)</b>	<b>EC/DEC (mL)</b>
<b>C-GPE-1</b>	2	0.748	0.2
<b>C-GPE-2</b>			0.4
<b>C-GPE-3</b>			0.6

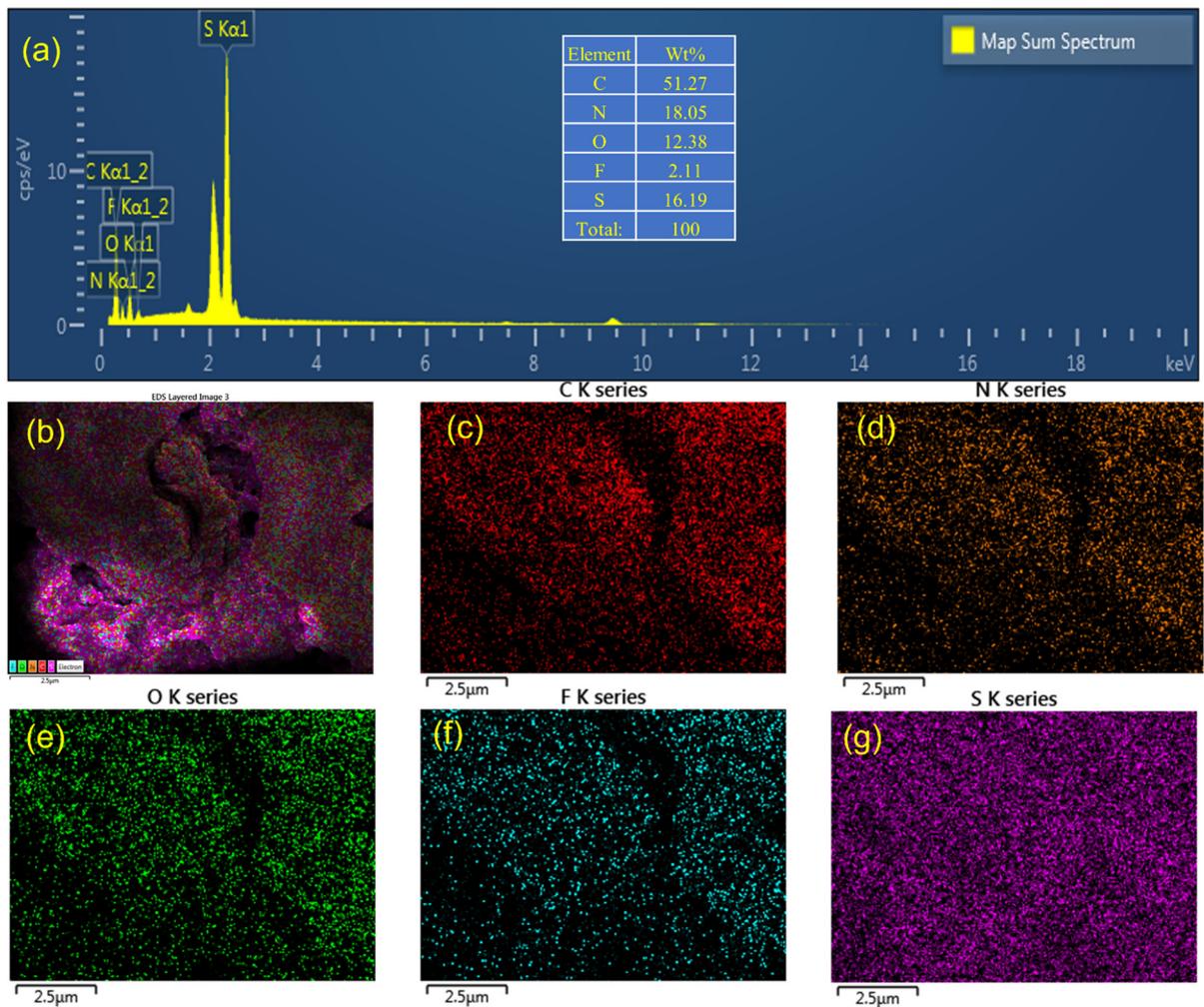


Figure S5: (a-b) Selected area of graphite anode of C-GPE-2 electrolyte based LIB (inset shows the wt% of elemental C, N, O, F, and S), and (b-g) EDS elemental mapping of C, N, O, F, and S, respectively. The graphite anode was recovered after 300 CD cycles.



## Reference

1. Chen, B.; Xu, Q.; Huang, Z.; Zhao, Y.; Chen, S.; Xu, X. One-pot preparation of new copolymer electrolytes with tunable network structure for all-solid-state lithium battery. *J. Power Sources* 2016, 331, 322-331. doi:10.1016/j.jpowsour.2016.09.063.
2. Zhu, J.; Zhang, J.; Zhao, R.; Zhao, Y.; Liu, J.; Xu, N.; Wan, X.; Li, C.; Ma, Y.; Zhang, H.; Chen, Y. In situ 3D crosslinked gel polymer electrolyte for ultra-long cycling, high-voltage, and high-safety lithium metal batteries. *Energy Storage Materials* 2023, 57, 92-101. doi:10.1016/j.ensm.2023.02.012.
3. Zhang, W.; Ryu, T.; Yoon, S.; Jin, L.; Jang, G.; Bae, W.; Kim, W.; Ahmed, F.; Jang, H. Synthesis and Characterization of Gel Polymer Electrolyte Based on Epoxy Group via Cationic Ring-Open Polymerization for Lithium-Ion Battery. *Membranes* 2022, 12, 439. doi:10.3390/membranes12040439.
4. Nair, J.R.; Shaji, I.; Ehteshami, N.; Thum, A.; Diddens, D.; Heuer, A.; Winter, M. Solid Polymer Electrolytes for Lithium Metal Battery via Thermally Induced Cationic Ring-Opening Polymerization (CROP) with an Insight into the Reaction Mechanism. *Chem. Mater.* 2019, 31, 3118-3133. doi:10.1021/acs.chemmater.8b04172.
5. Yao, W.; Zhang, Q.; Qi, F.; Zhang, J.; Liu, K.; Li, J.; Chen, W.; Du, Y.; Jin, Y.; Liang, Y.; Liu, N. Epoxy containing solid polymer electrolyte for lithium ion battery. *Electrochim. Acta* 2019, 318, 302-313. doi:10.1016/j.electacta.2019.06.069.
6. Ma, Y.; Sun, Q.; Wang, S.; Zhou, Y.; Song, D.; Zhang, H.; Shi, X.; Zhang, L. Li salt initiated in-situ polymerized solid polymer electrolyte: new insights via in-situ electrochemical impedance spectroscopy. *Chem. Eng. J.* 2022, 429, 132483. doi:10.1016/j.cej.2021.132483.