Supporting information

In-Situ Polymerizable Deep Eutectic Solvent Electrolyte based on TFEA-co-MBA for High-Safety and High-Voltage Lithium Metal Batteries

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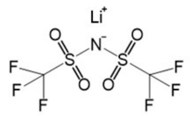
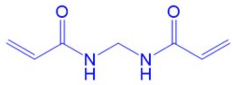
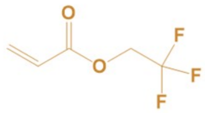
LiTFSI	MBA	TFEA	SN
			
286 °C	> 300 °C	-45.8 °C	58.1 °C

Fig. S1. Chemical structural formulae and T_m of each monomer.

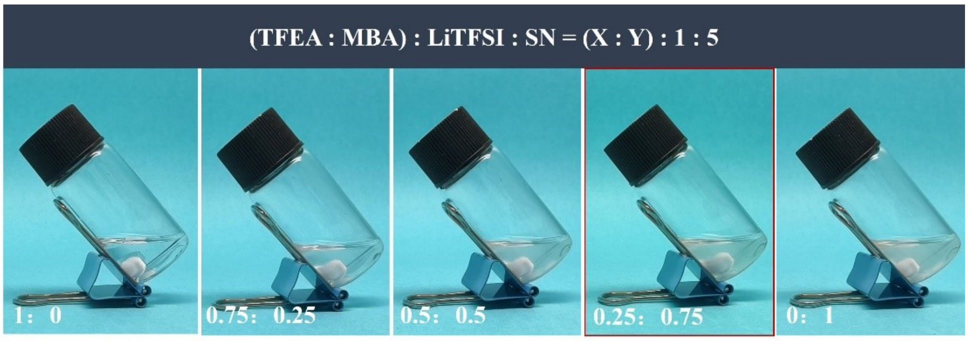


Fig. S2. Optical photographs of PDES monomers formed with different ratios of monomers.

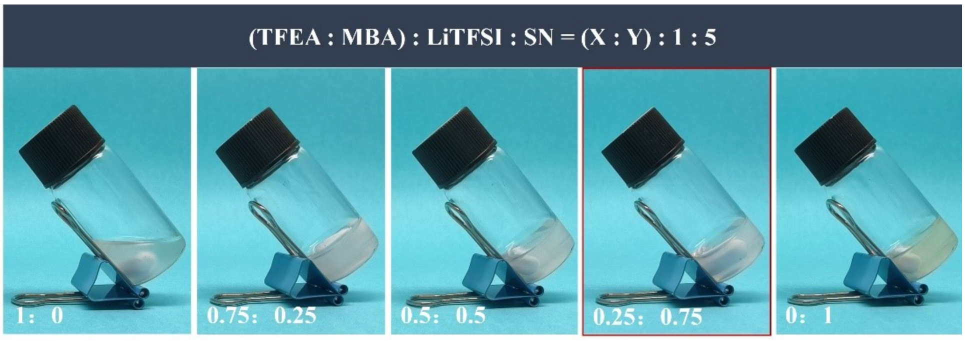


Fig. S3. Optical photographs of poly(PDES) electrolytes formed with different ratios of monomers.

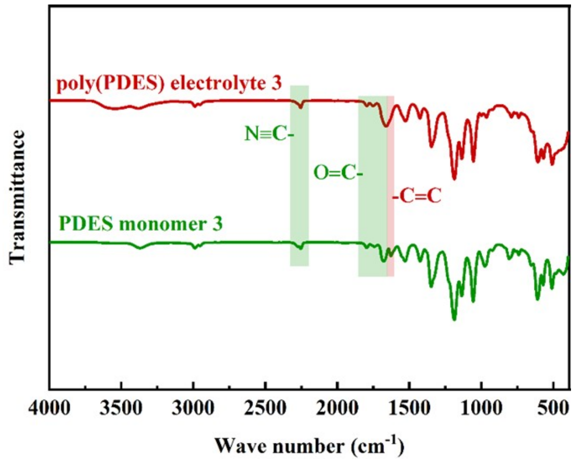


Fig. S4. FTIR spectrum of PDES monomer 3 before and after polymerization.

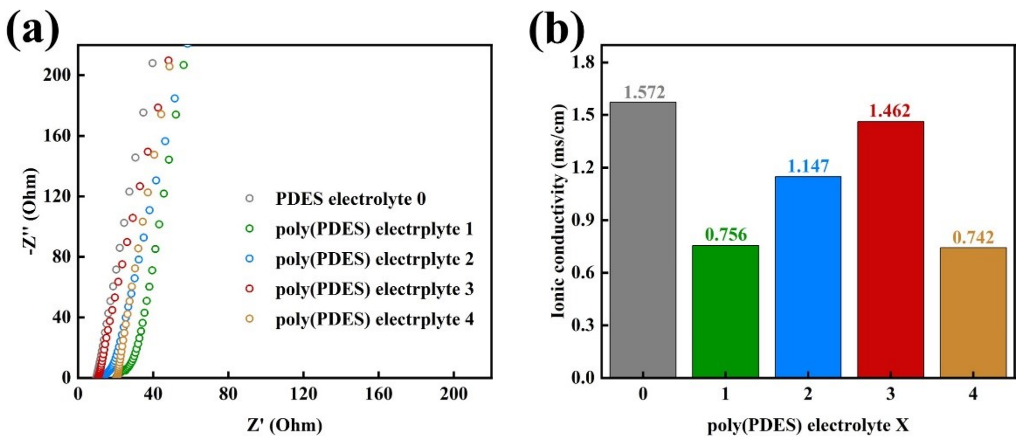


Fig. S5. EIS and Ionic conductivity of PDES electrolyte 0 and poly(PDES) electrolyte X.

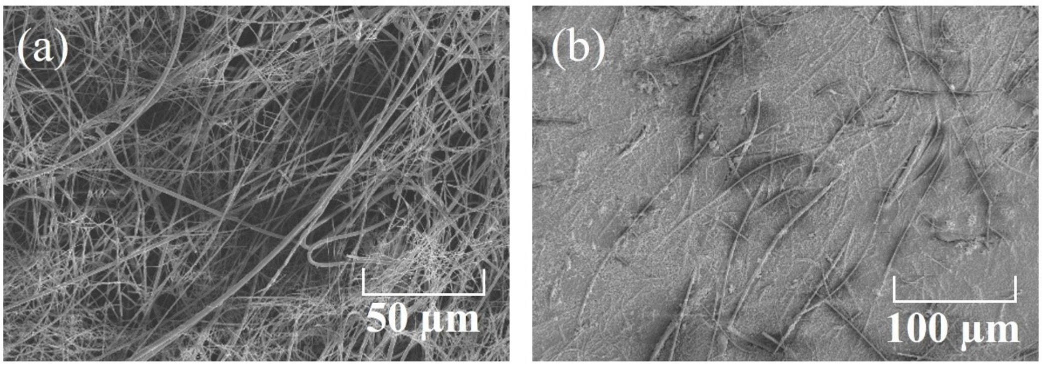


Fig. S6. The SEM images of (a) GF/A and (b) the poly(PDES) electrolyte 3 @ GF/A.

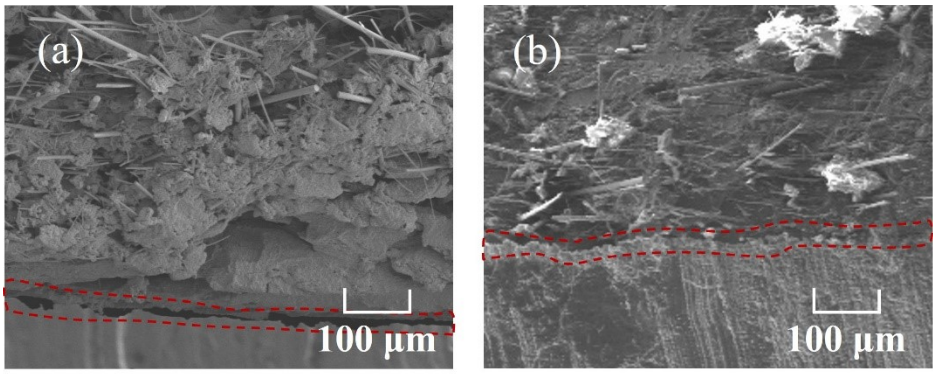


Fig. S7. The cross-sectional image showing the (a) ex-situ and (b) in-situ interface between the LFP cathode and poly(PDES) electrolyte 3.

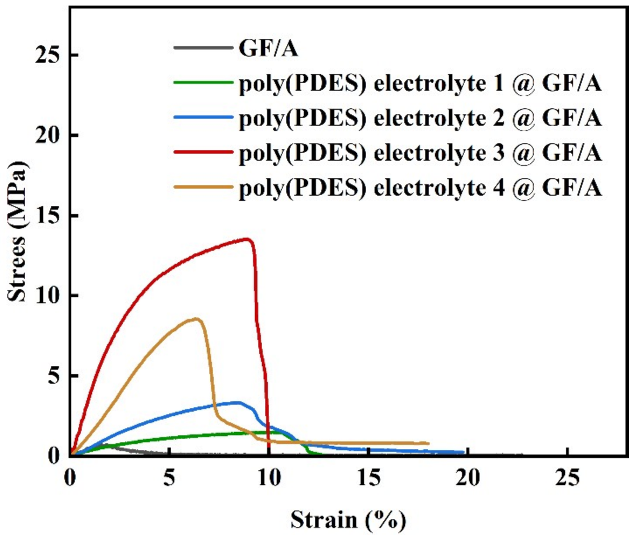


Fig. S8. Stress-stain curves for GF/A and poly(PDES) electrolyte X @ GF/A.

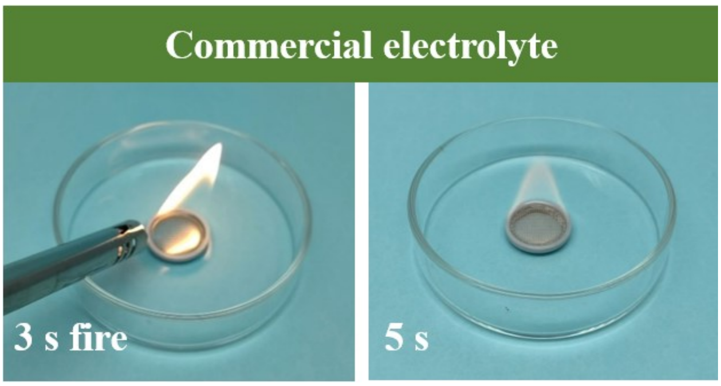


Fig. S9. Optical images of the ignition of the commercial electrolyte.

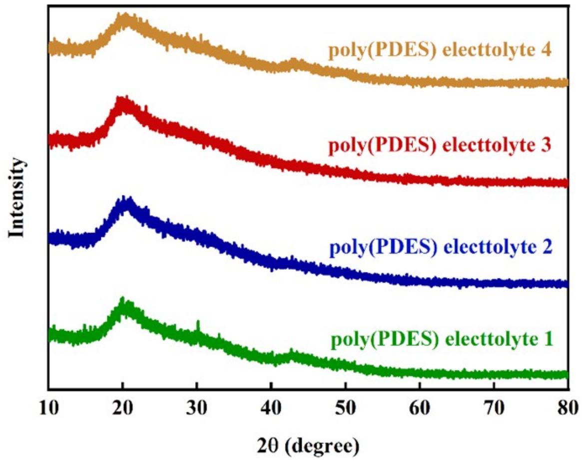


Fig. S10. XRD patterns of different poly(PDES) electrolyte X.

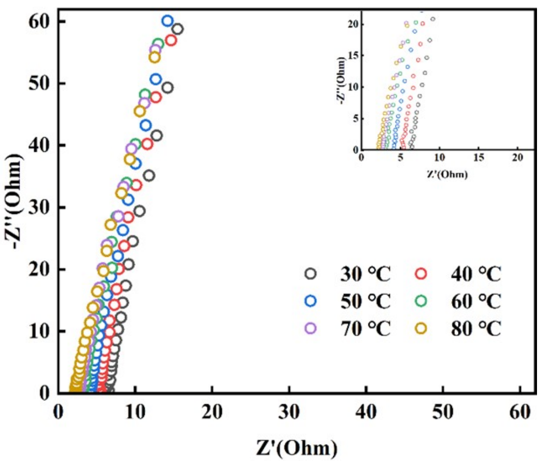


Fig. S11.EIS of poly(PDES) electrolyte 3 at different temperature.

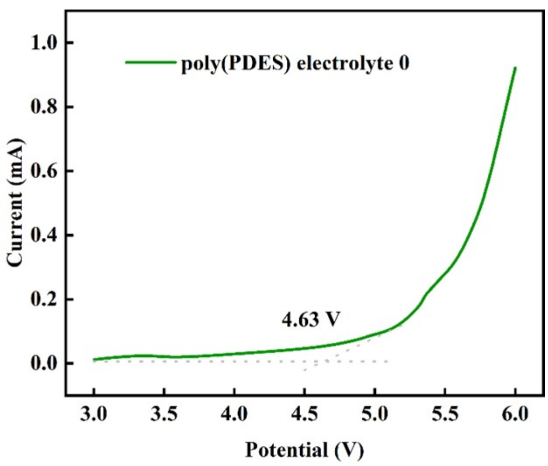


Fig. S12. LSV curves of the PDES electrolyte 0 at a scan rate of 10 mV s^{-1}.

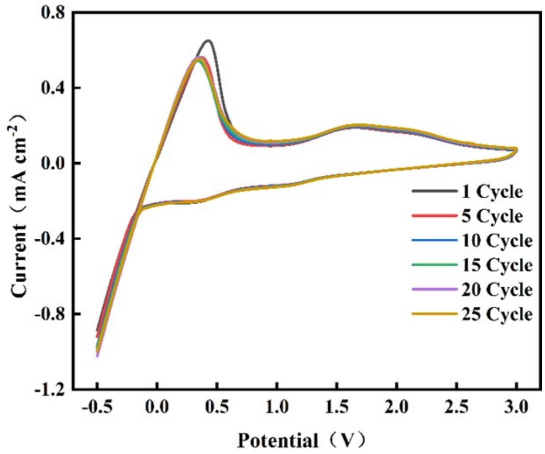


Fig. S13. CV curves of lithium plating/stripping with the Li || poly(PDES) electrolyte 3 || SS battery at a scan rate of 10 mV s^{-1}.

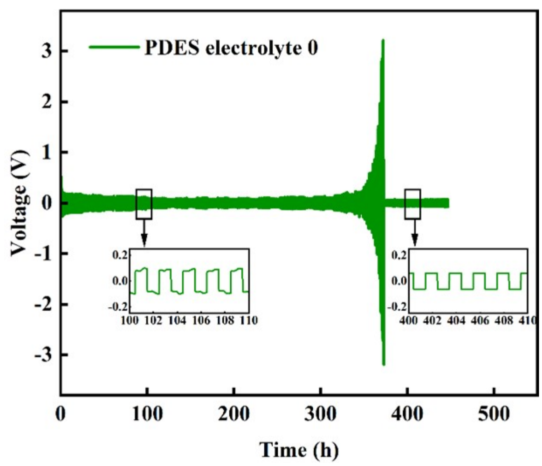


Fig. S14. Galvanostatic cycling curve of a Li || PDES electrolyte 0 ||Li symmetric battery at a current density of 0.1 mA cm^{-2}.

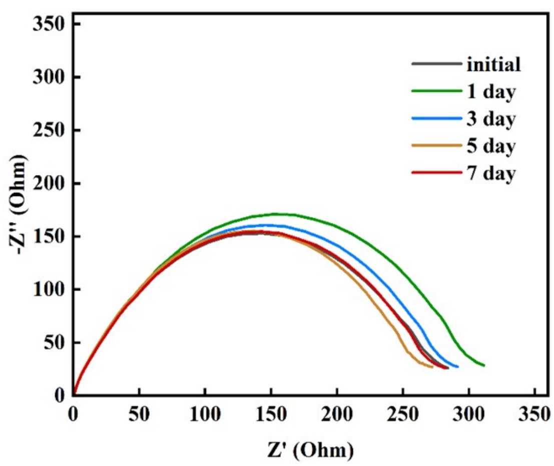


Fig. S15. EIS of Li || poly(PDES) electrolyte 3 || Li symmetric battery after long time standing.

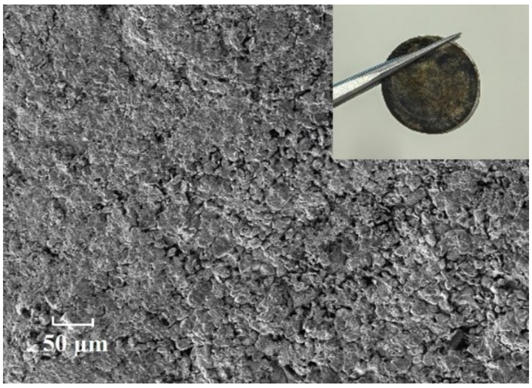


Fig. S16. SEM images of lithium metal anode in the PDES electrolyte 0 after plating/stripping for 400 h. Insert shows the photo of lithium metal anode after 400 h.

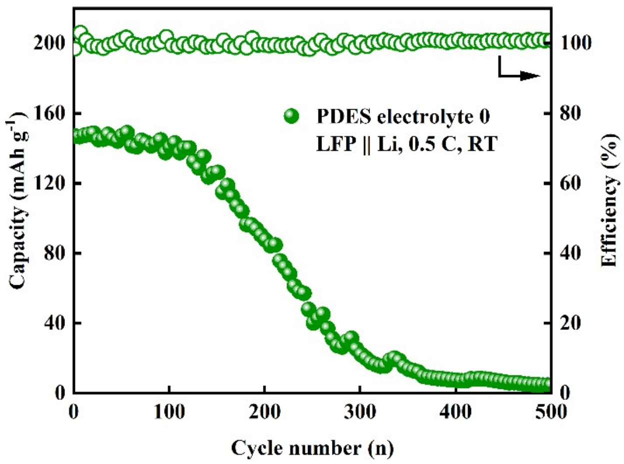


Fig. S17. The cycling performance and corresponding Coulombic efficiency of the Li || PDES electrolyte 0 || LFP full batteries at room temperature.

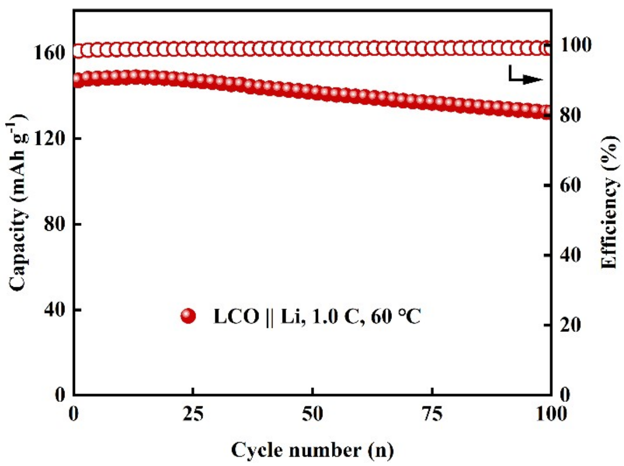


Fig. S18. The cycling performance and corresponding Coulombic efficiency of the Li || poly(PDES) electrolyte 3 || LCO full batteries at 60 °C.

Table S1. Comparative analysis of the mechanical properties of poly (PDES) electrolyte X @ GF/A.

poly(PDES) electrolyte *X* @GF/A	Stress (MPa)	Strain (%)	Young's modulus (MPa)
GF/A	0.6	1.7	60.8
1	1.5	10.6	30.1
2	3.3	9.2	62.2
3	13.8	9.0	265.2
4	8.6	6.5	162.8

Table S2. Comparative analysis of the performance of different electrolytes.

Additional solvents (Y/N)	Ionic conductivity (mS cm^{-1})	Electrochemical window (V)	High voltage batteries (V)	Mechanical stress (MPa)	Ref.
N	1.462, RT	5.36	4.6	13.8	this work
Y	0.19, 60 ℃	5.0	4.4	1.7	[1]
N	1.4, RT	5.2	4.5	N/A	[2]
Y	0.32, RT	5.0	4.0	50.79	[3]
Y	0.11, 30 ℃	5.4	4.0	0.75	[4]
N	0.13, RT	4.76	4.2	N/A	[5]
Y	0.87, RT	5.1	4.4	6.3	[6]
Y	0.241, RT	4.9	4.3	N/A	[7]
N	0.358, RT	5.4	4.3	7.54	[8]
Y	0.158, 30 ℃	5.2	4.3	N/A	[9]

References

1 S. E. Abraham, R. Murugan, T. Panneerselvam and S. O. Veedu, *J. Alloy. Compd.*, 2025, 178636.

2 S. Wu, C. Wang, S. Li, L. Lin, Q. Tong, M. Zhu and J. Weng, *ACS Appl. Mater. Interfaces*, 2024, **16**, 28482-28492.

3 Y. Zhang, Z. Chen, J. Wang, S. Fan, T. Zhang, C. Zhang, Y. Zhang and Q. Chi, *Small*, 2024, **20**.

4 H. T. T. Nguyen, D. H. Nguyen, Q. Zhang, Y. Lee, J. Jan, C. Chiu and H. Teng, *J. Mater. Chem. A*, 2021, **9**, 25408-25417.

5 C. Zhang, Z. Niu, J. Bae, L. Zhang, Y. Zhao and G. Yu, *Energy Environ. Sci.*, 2021, **14**, 931-939.

6 J. Kim, J. Sun, J. Jang, D. Park, S. Ahn, W. C. Kim, K. Min and K. Park, *Energy Storage Mater.*, 2024, **64**.

7 X. Li, Y. Wang, Q. Zhou, H. Kuai, C. Ji and X. Xiong, *J. Mater. Chem. A*, 2024, **12**, 7645-7653.

8 D. Xu, D. Zhao, X. Niu, T. Wang and Z. Yang, *Chem. Eng. J.*, 2024, **490**, 151780.

9 W. Liu, L. Meng, X. Liu, L. Gao, X. Wang, J. Kang, J. Ju, N. Deng, B. Cheng and W. Kang, *J. Energy Chem.*, 2023, **76**, 503-515.