

Supplementary Information for

**In situ generation of soft-tough asymmetric composite  
electrolyte for dendrite-free lithium metal batteries**

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## Supplementary Figures

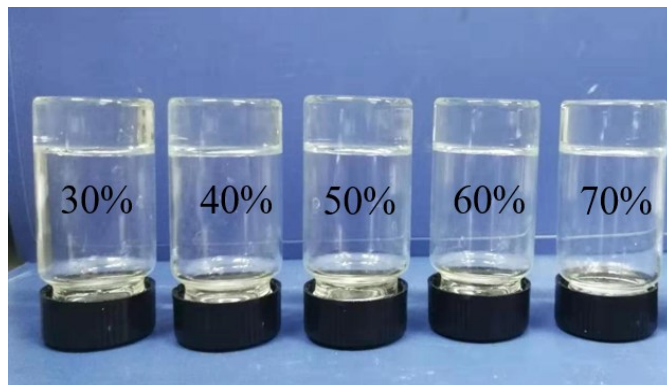


Fig. S1 Photo of TPGDA with varied amounts of LE after the polymerization process.

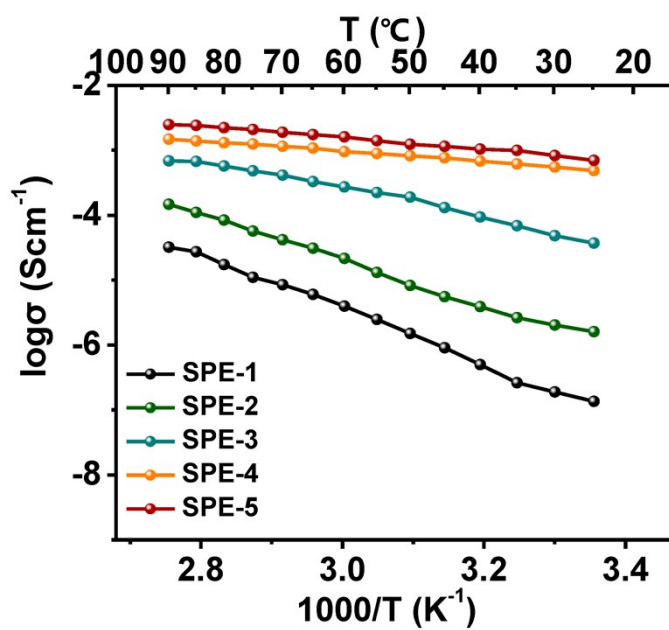
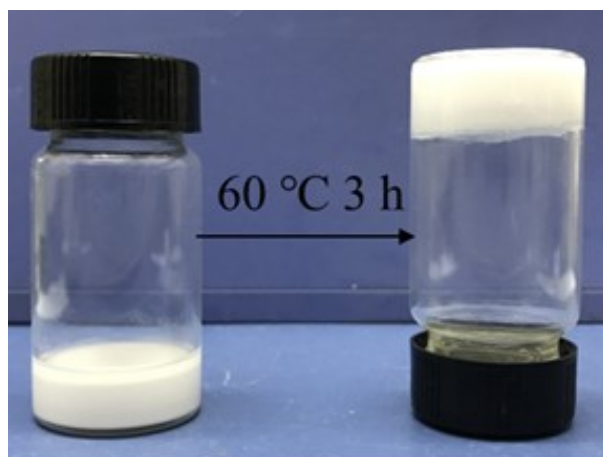
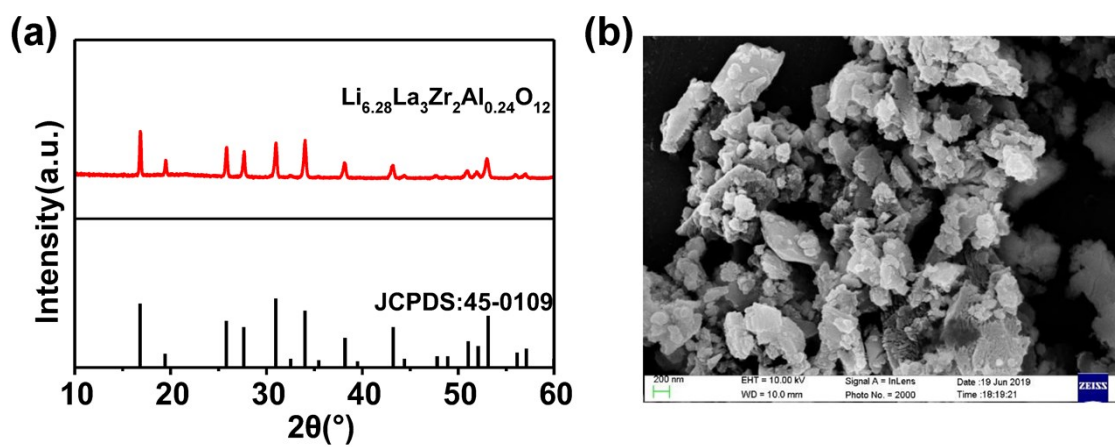


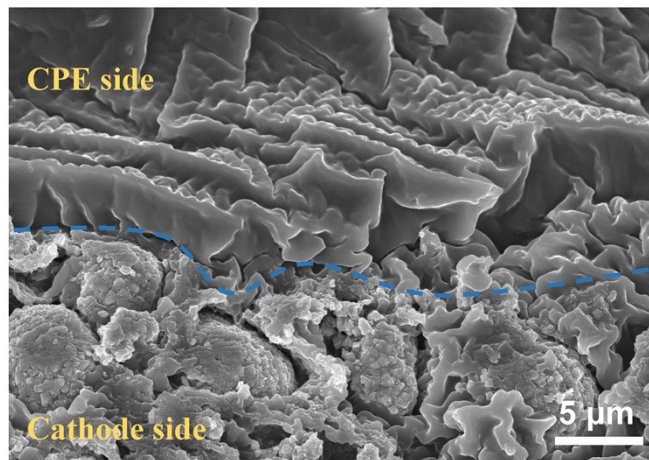
Fig. S2 The ionic conductivity of the SPE with different content of LE.



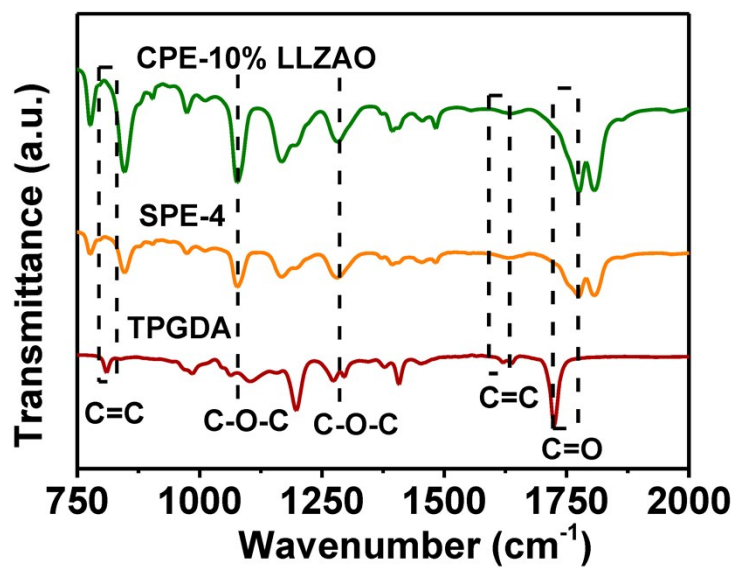
**Fig. S3** The typical image of composite electrolyte precursor solution before and after heating at 60 °C for 3 h.



**Fig. S4** (a) XRD pattern and (b) SEM image of the LLZAO powders.



**Fig. S5** SEM cross-section image of the CPE and NCM cathode layer interface.



**Fig. S6** FTIR adsorption spectra of TPGDA, SPE-4, and CPE-10%LLZAO.

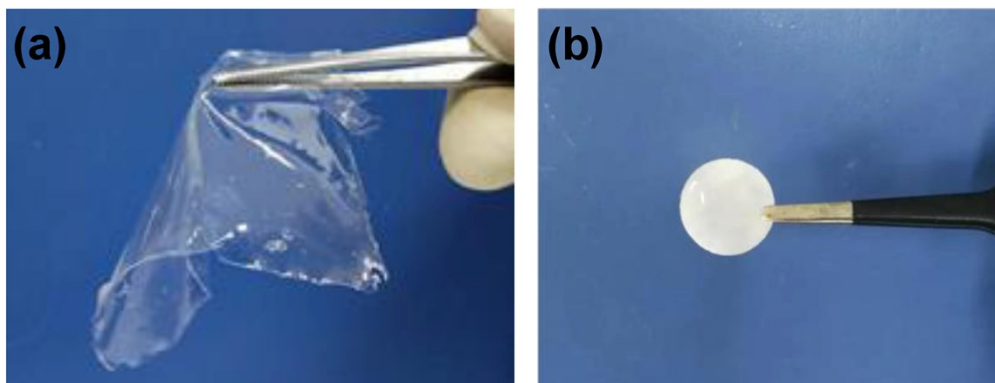


Fig. S7 The digital images of (a) TPGDA polymer electrolyte (b) CPE-10%LLZAO electrolyte.

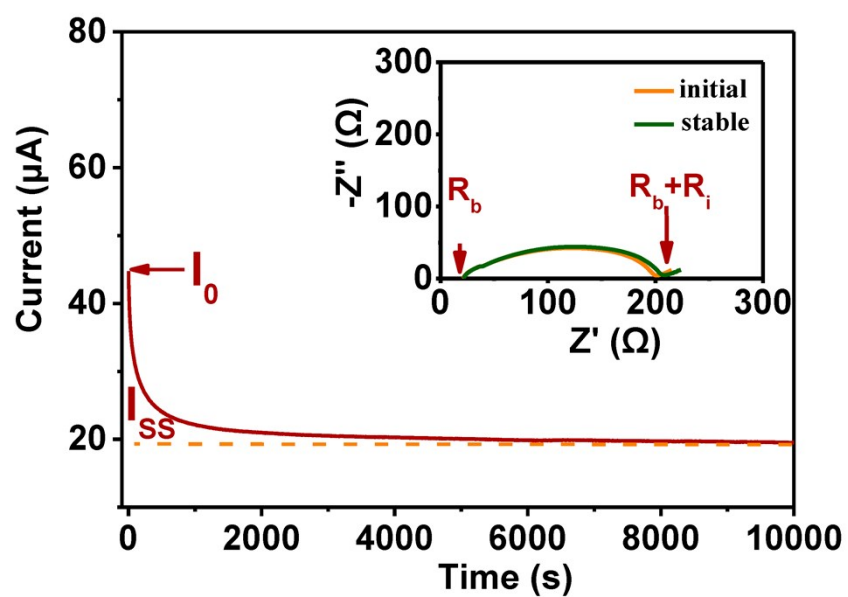
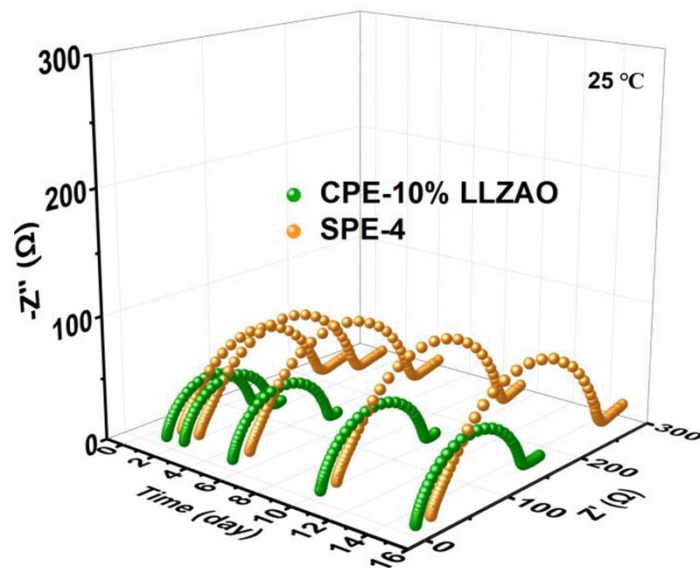
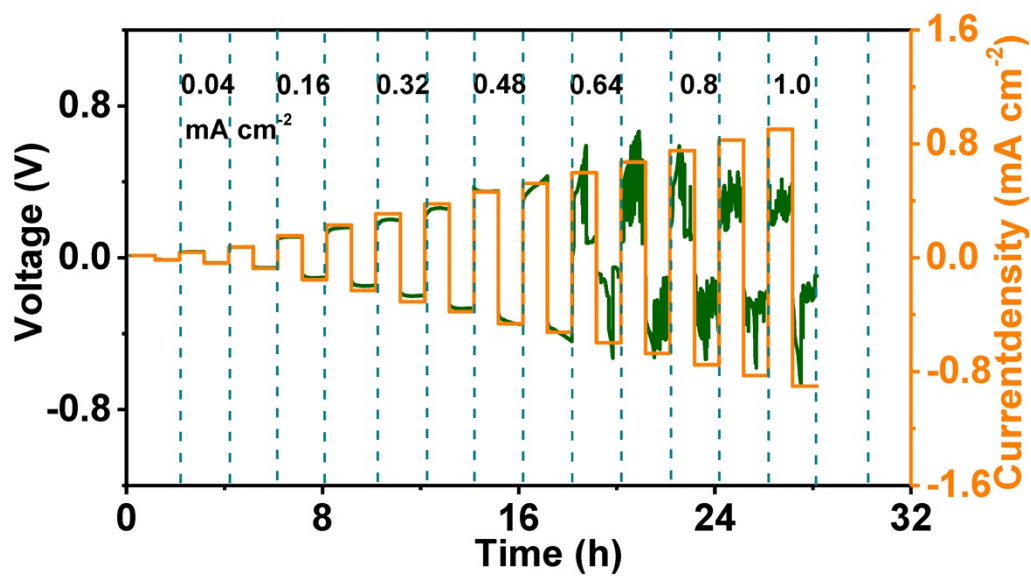


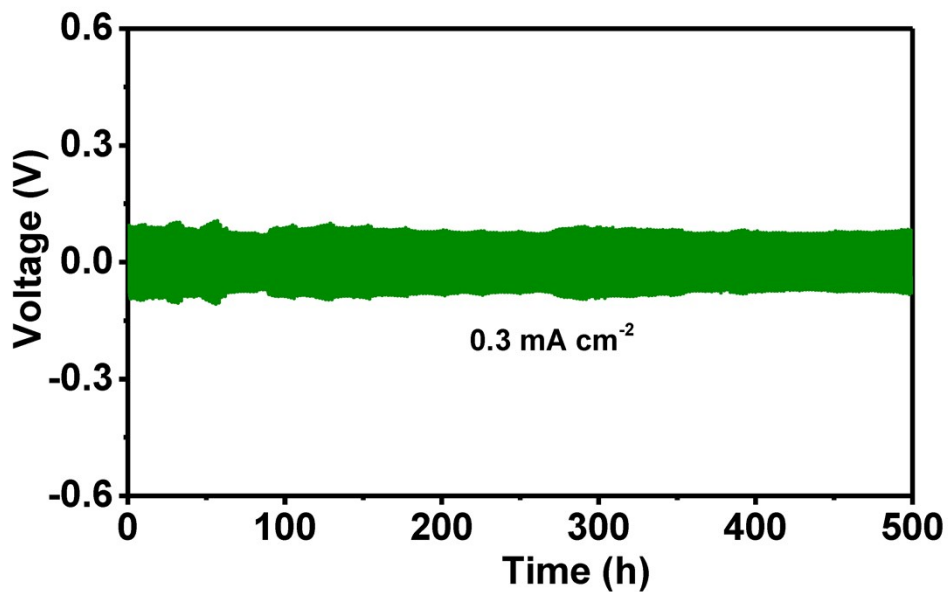
Fig. S8 Current variation with time of a symmetrical Li/SPE-4/Li cell.



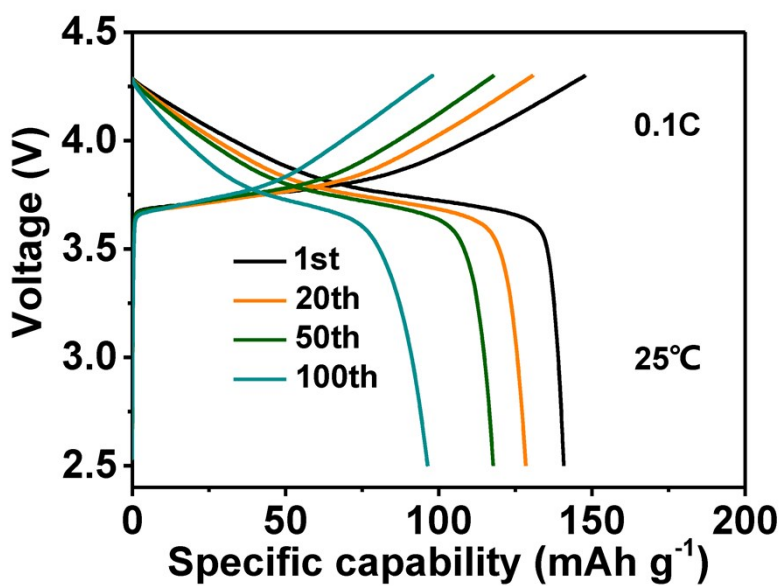
**Fig. S9** Impedance response with time evolution of the Li/CPE-10%LLZAO/Li and Li/SPE-4/Li.



**Fig. S10** Direct current cycling of Li/CPE-10%LLZAO/Li symmetrical cell, stepping the current density from 0.02 to 1.0  $\text{mA cm}^{-2}$ .



**Fig. S11** Galvanostatic cycling data of Li/CPE-10%LLZAO/Li symmetrical cells at a current density of  $0.3 \text{ mA cm}^{-2}$ .



**Fig. S12** charge and discharge voltage profiles of SPE-4 based NCM/Li at different cycles with 0.1C.