Synergistically enhancing stable interface with soft gel and garnet-type $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$ bi-functional composite electrolyte of lithium metal batteries

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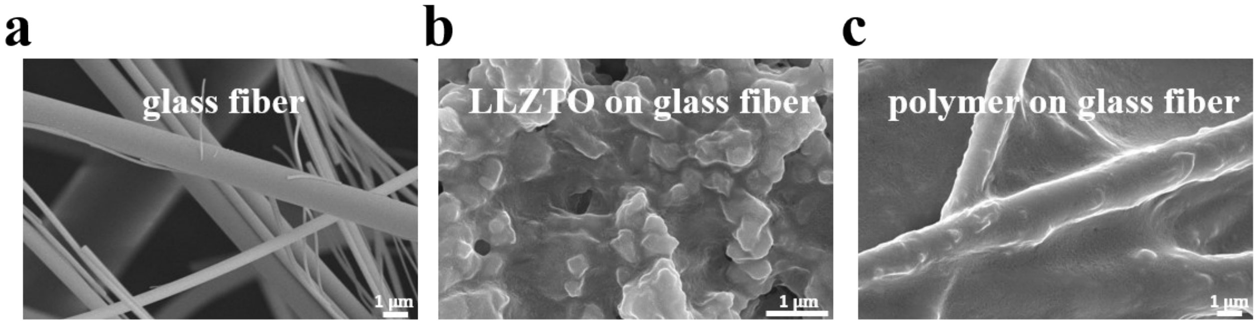

Fig. S1. SEM images of (a) glass fiber; (b) ceramic-rich layer; (c) gel-rich layer.

Fig. S2. Digital photographs of (a) glass fiber; (b) ceramic-rich layer; (c) gel-rich layer.

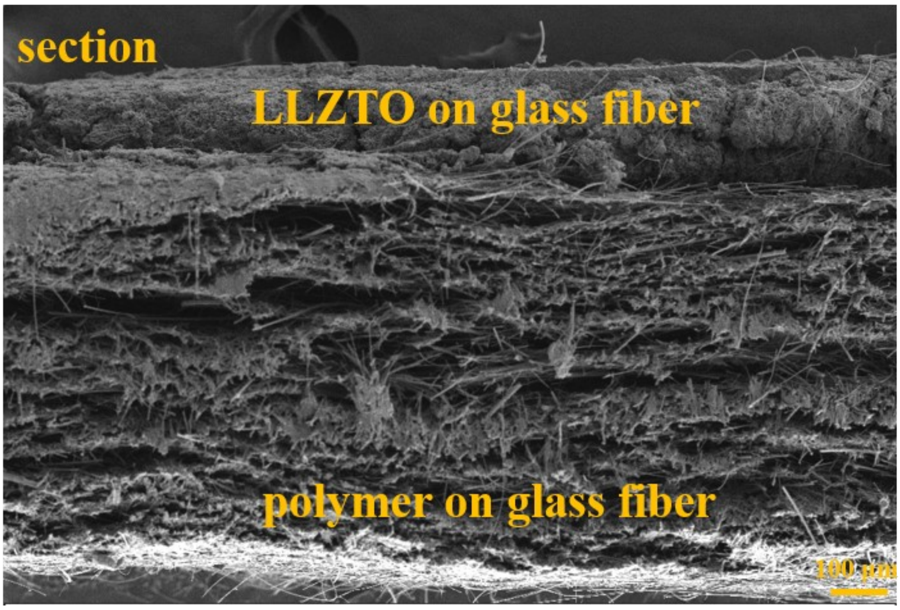

Fig. S3. Cross-section of As-THCE-10%LLZTO.

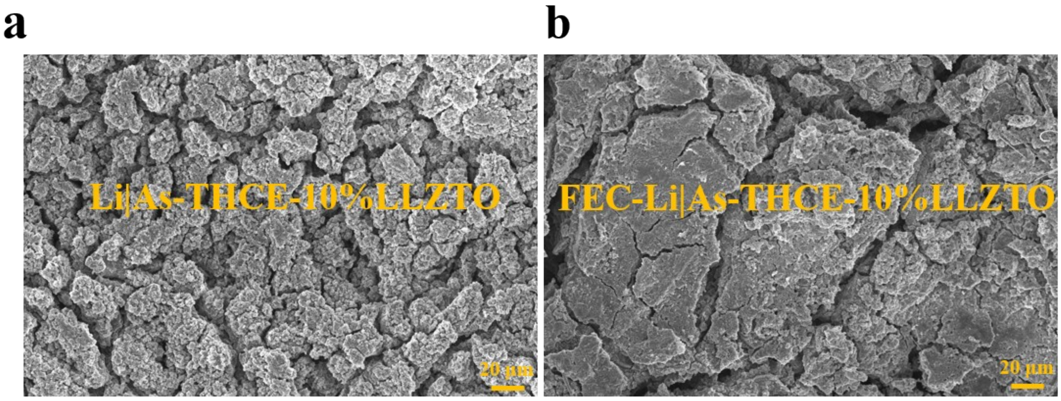

Fig. S4. SEM of images Li metal surface obtained from (a) Li||As-THCE-10%LLZTO||LEP; and (b) FEC-Li||As-THCE-10%LLZTO||LEP cells after 100 cycles.

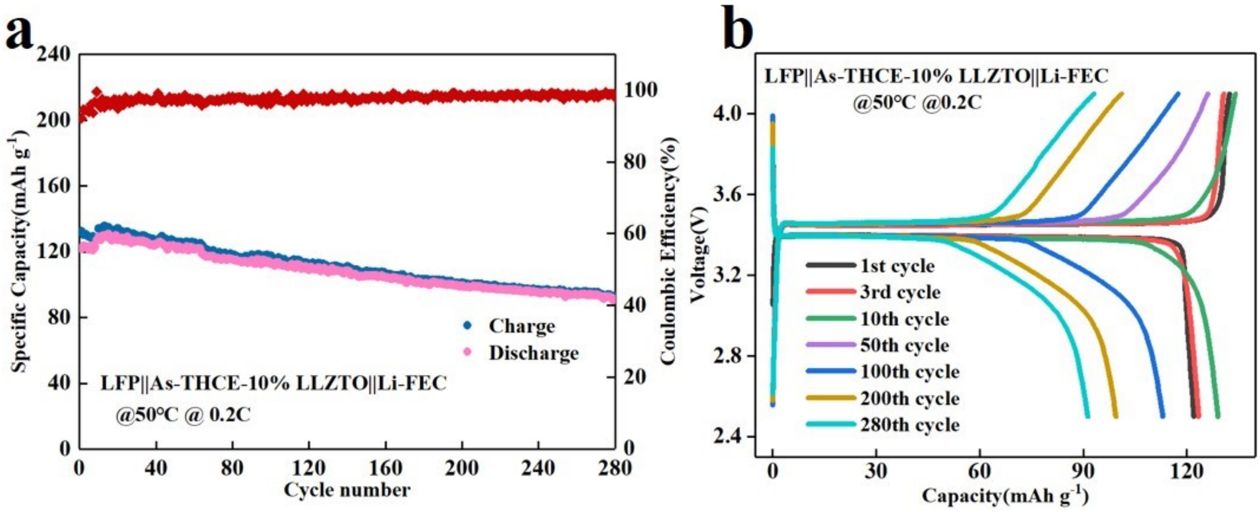

Fig. S5. (a) The cycle performance of LFP||As-THCE-10%LLZTO||Li-FEC cell at 0.2 C at 50 ℃; (b) Charge-discharge curves at 0.2 C of LFP||As-THCE-10%LLZTO||Li-FEC cell at 50 ℃

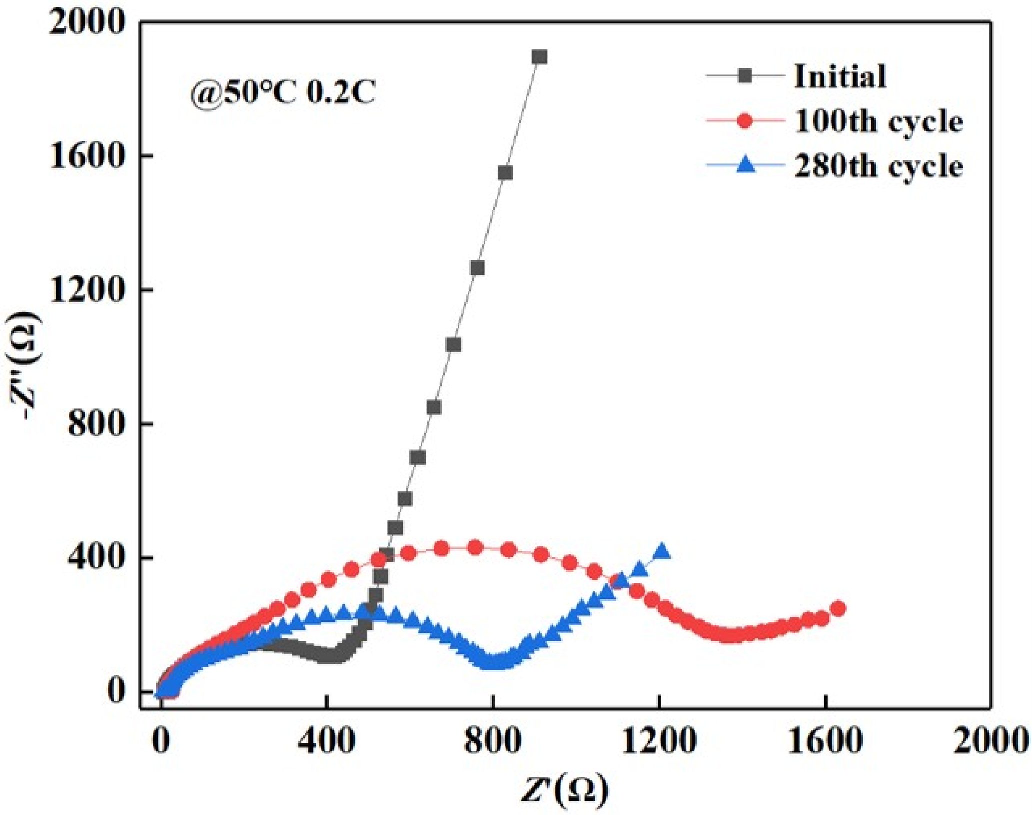

Fig. S6. Nyquist plots of LFP||As-THCE-10%LLZTO||Li-FEC before cycling, after 100 cycles, 280cycles.

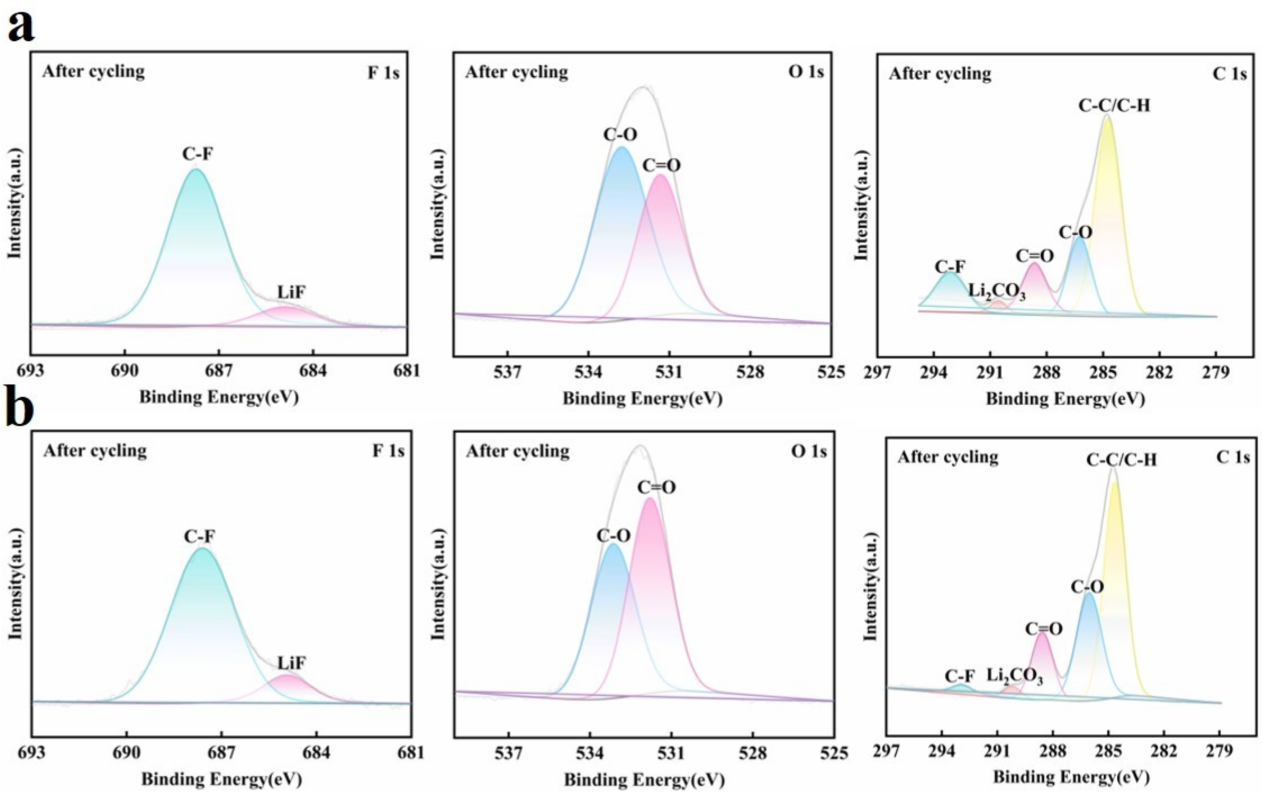

Fig. S7. XPS profiles of LFP obtained from (a) LFP||As-THCE-10%LLZTO||Li and (b) LFP||As-THCE-10%LLZTO||Li-FEC cells.

Tab. S1. Comparison of Ea of different samples

Sample	As-THCE-5%LLZTO	As-THCE-10%LLZTO	As-THCE-15%LLZTO	As-THCE-20%LLZTO
Ea ($kJ \cdot mol^{-1}$)	7.8	6.978	7.976	8.239
Ea (eV)	0.081	0.072	0.083	0.086

Tab. S2. Electrochemical performance comparison of the asymmetric electrolytes.

CPEs	Cell	Rate performance	Cycling stability	Ref.
PEO-LiTFSI-ACN-LLZTO	Li‖LEP	0.1 C,99.1 mAh g^{-1}	82.4%/200th@25 °C	1
TPGDA-$LiPF_6$-EC-DMC-EMC-LLZAO	Li‖NCM	149.1 mAh g^{-1}	83.5%/100th@25 °C	2
PEO-PAN-LiTFSI-LLZTO	Li‖LEP	0.2 C,159.9 mAh g^{-1}	96%/90th	3
TPGDA- $VEImNTF_2$- $LiPF_6$-EC-DMC-EMC-LLZTO	Li‖LEP	0.5 C	300th@25 °C	4
AHPAA-PEO-LiTFSI	Li‖LEP	0.5 C	84%/2000th	5
TMPTMA-HDDA-$N_{1,4,4,4}$TFSI-LiTFSI-DMC-LLZTO	**Li‖LEP**	**2 C, 111.6 mAh g^{-1}**	**84.73%/500th@25 °C**	**This work**

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