Reinforced Cathode-Garnet Interface for High-Capacity All-Solid-State Batteries

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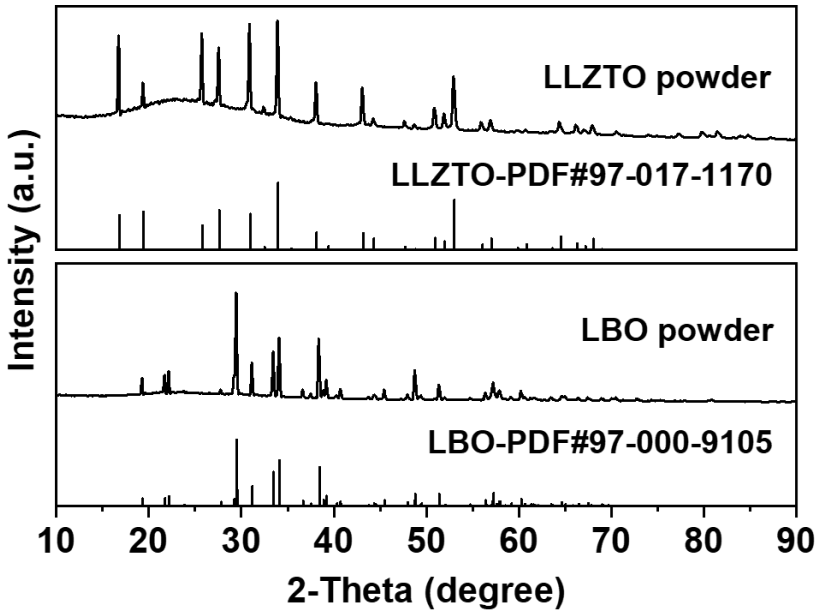

Figure S1. XRD patterns of the as-synthesized LLZTO and LBO powder.

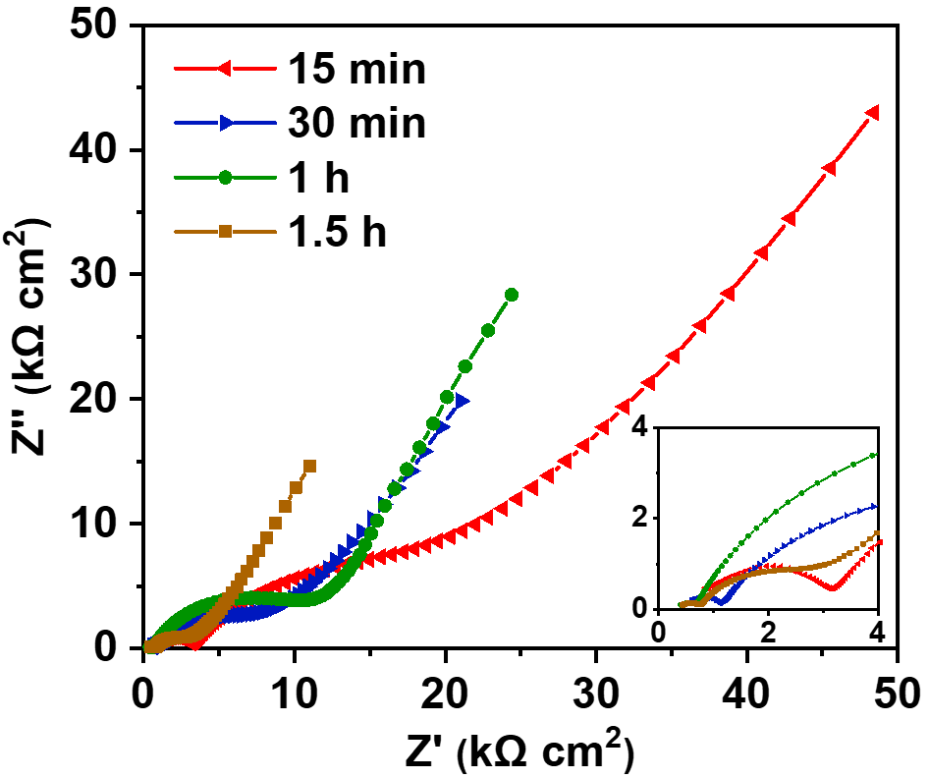

Figure S2. EIS spectra of the LLZTO pellets with various dwell time.

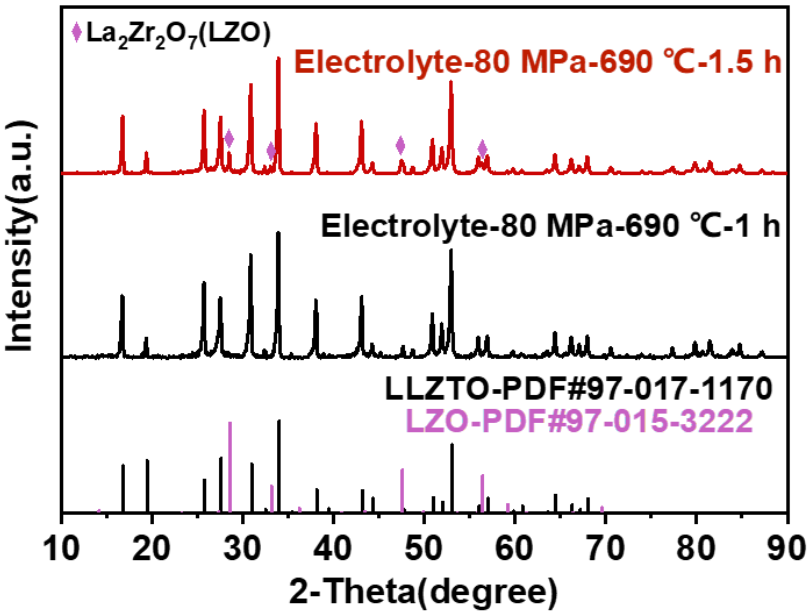

Figure S3. XRD patterns of the LLZTO pellets with the dwell time of 1 hour and 1.5 hours.

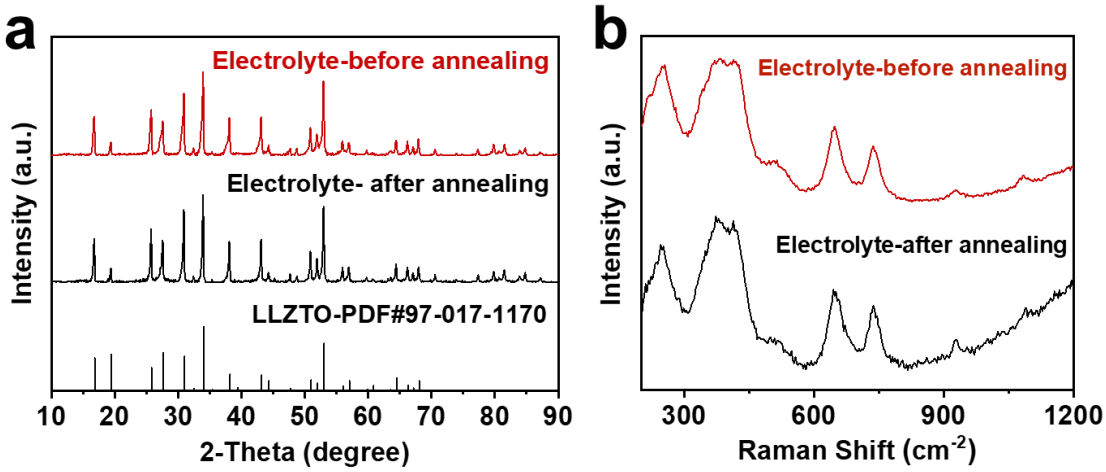

Figure S4. (a) XRD, (b) Raman patterns of the electrolyte side of the LCO-LLZTO/LLZTO half cell before and after annealing.

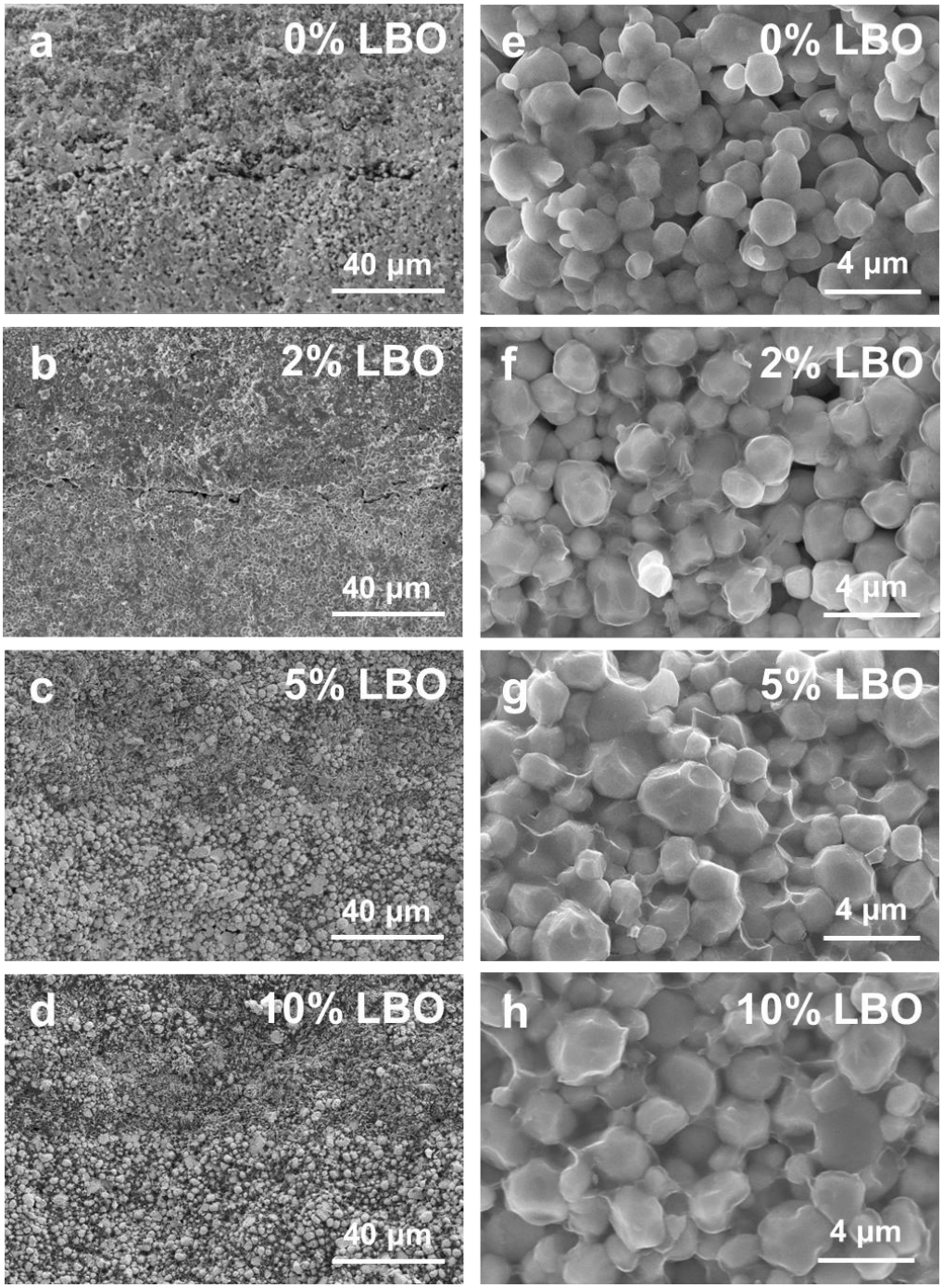

Figure S5. SEM images of the LCO-LLZTO/LLZTO half cell. The cross section with (a) 0%, (b) 2%, (c) 5%, (d) 10% LBO and the contact state of particles inside the composite cathode with (e) 0%, (f) 2%, (g) 5%, (h) 10% LBO at high magnification.

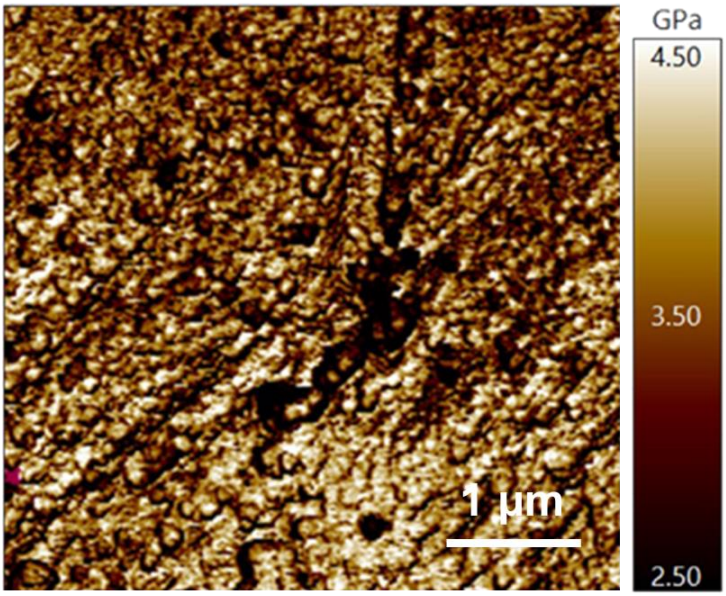

Figure S6. Young's modulus mapping of the LLZTO pellet without LBO.

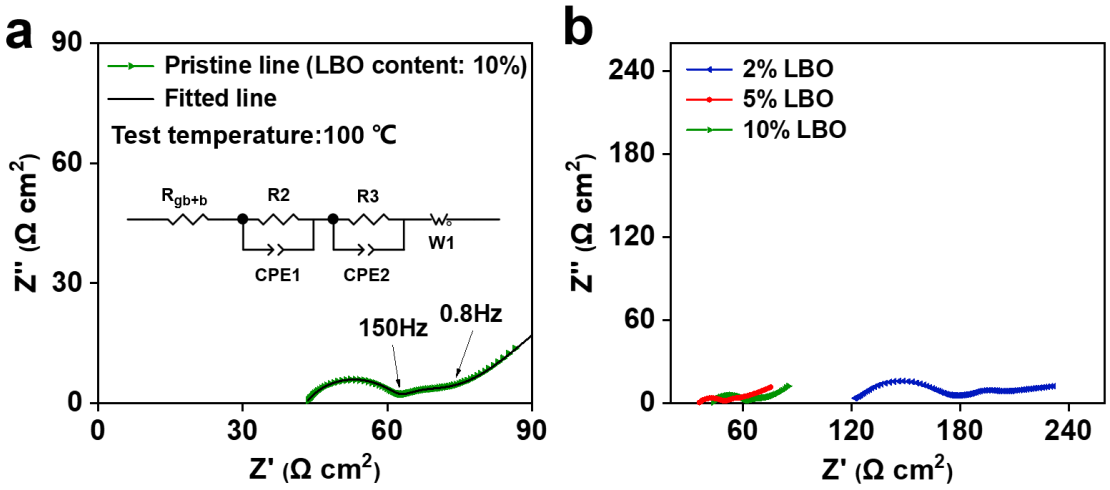

Figure S7. (a) A typical EIS spectrum model of the Li | LLZTO | LCO-LLZTO full cell. (b) EIS spectra of the full cells with different LBO content.

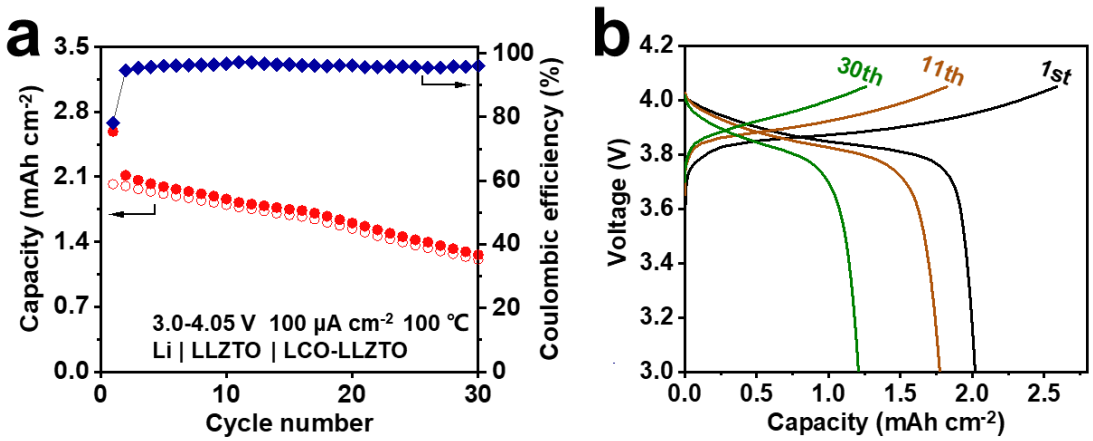

Figure S8. (a) Cycling performance of the full cell with an areal capacity of 2.02 mAh cm^{-2}, (b) The corresponding charge-discharge profiles.

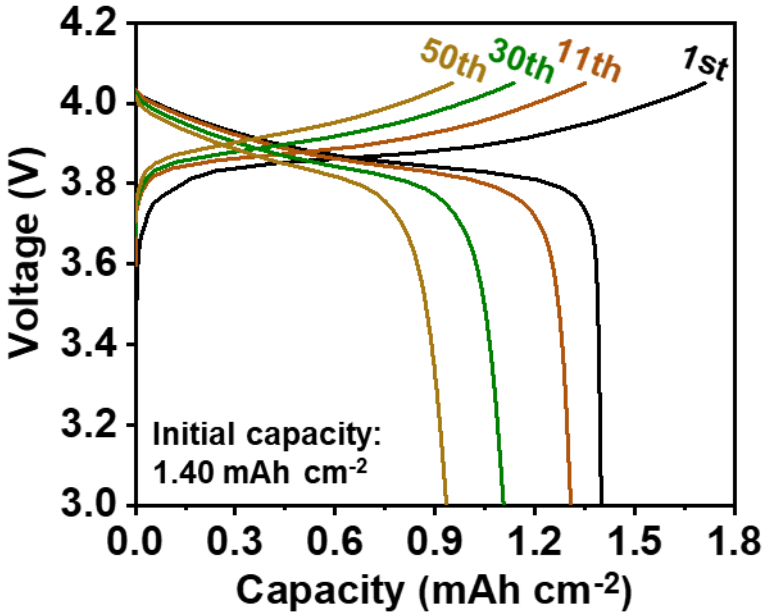

Figure S9. The charge-discharge profiles of the full cell with an areal capacity of 1.40 mAh cm^{-2}.

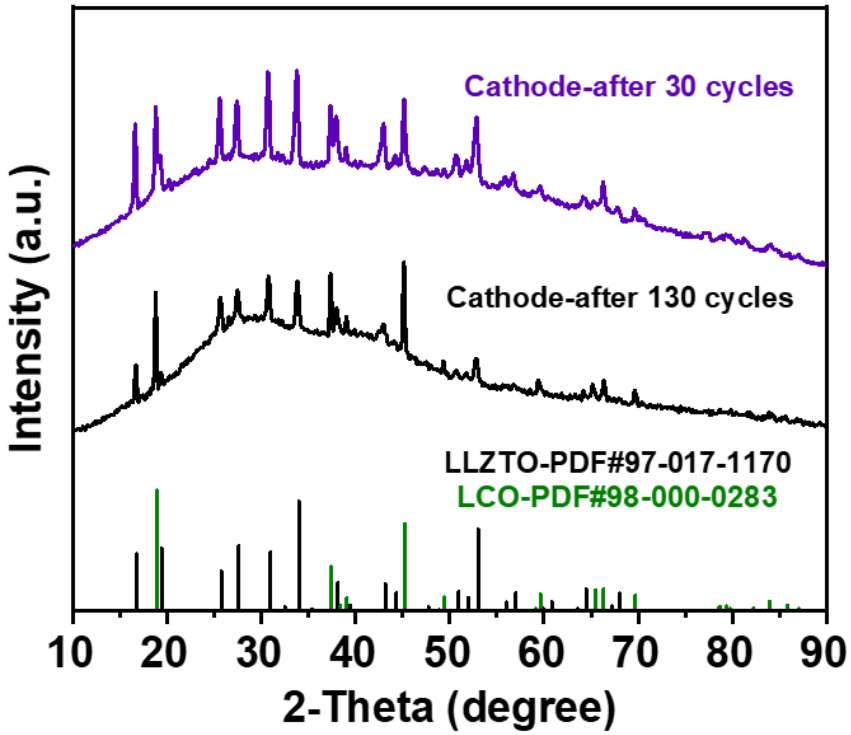

Figure S10. XRD patterns of the composite cathode side of the full cells with different cycle numbers.

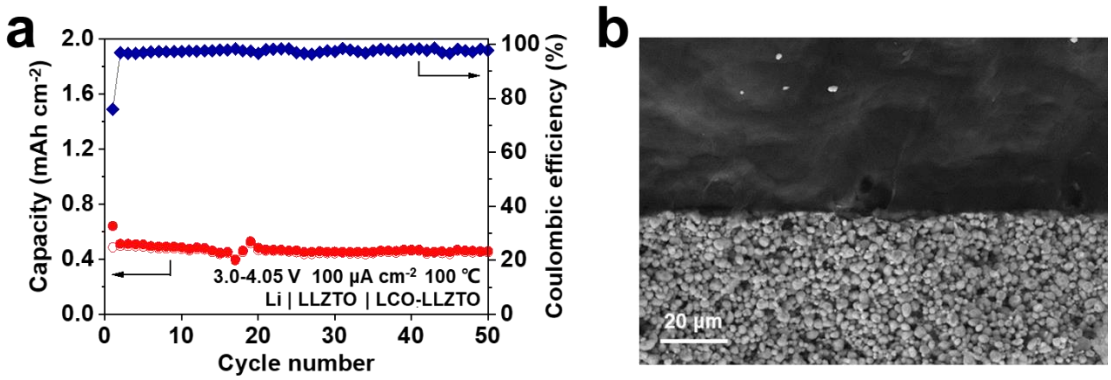

Figure S11. (a) Cycling performance of the full cell with an areal capacity of 0.49 mAh cm^{-2}, (b) SEM image of Li/LLZTO interface after 50 cycles.

Table S1. The room temperature Li-ion conductivity of LLZTO pellets with various dwell time.

Dwell time	Diameter (mm)	Thickness (mm)	Impedance (Ω cm^2)	Ionic conductivity (S cm^{-1})
15 min	9.95	1.05	3144	3.34×10^{-5}
30 min	9.93	1.00	1168	8.56×10^{-5}
1 h	9.98	1.02	655	1.56×10^{-4}
1.5 h	9.96	1.00	753	1.33×10^{-4}

Table S2. Fitted data of resistance components for full cells with different LBO content.

LBO content	R_{LLZTO} (Ω cm^2)	$R_{LCO\text{-}LLZTO/LLZTO}$ (Ω cm^2)	$R_{Li/LLZTO}$ (Ω cm^2)
2%	92.6	5.6	34.6
5%	35.2	3.8	11.0
10%	43.1	4.4	18.1