Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information：

Advancing Anode-Less Lithium Metal Batteries: ZnF_2 Modification and In-Situ Structural Regulation for Enhanced Performance

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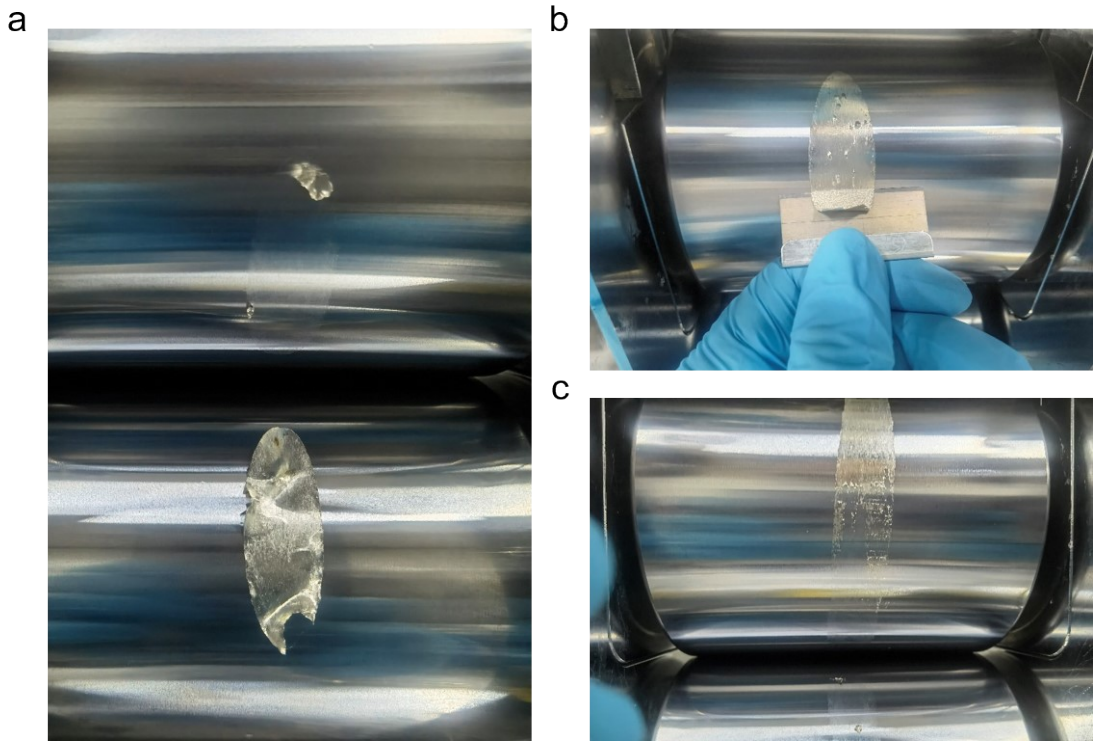

Figure. S1 (a-c) The tape-break and sticky properties of Li foil.

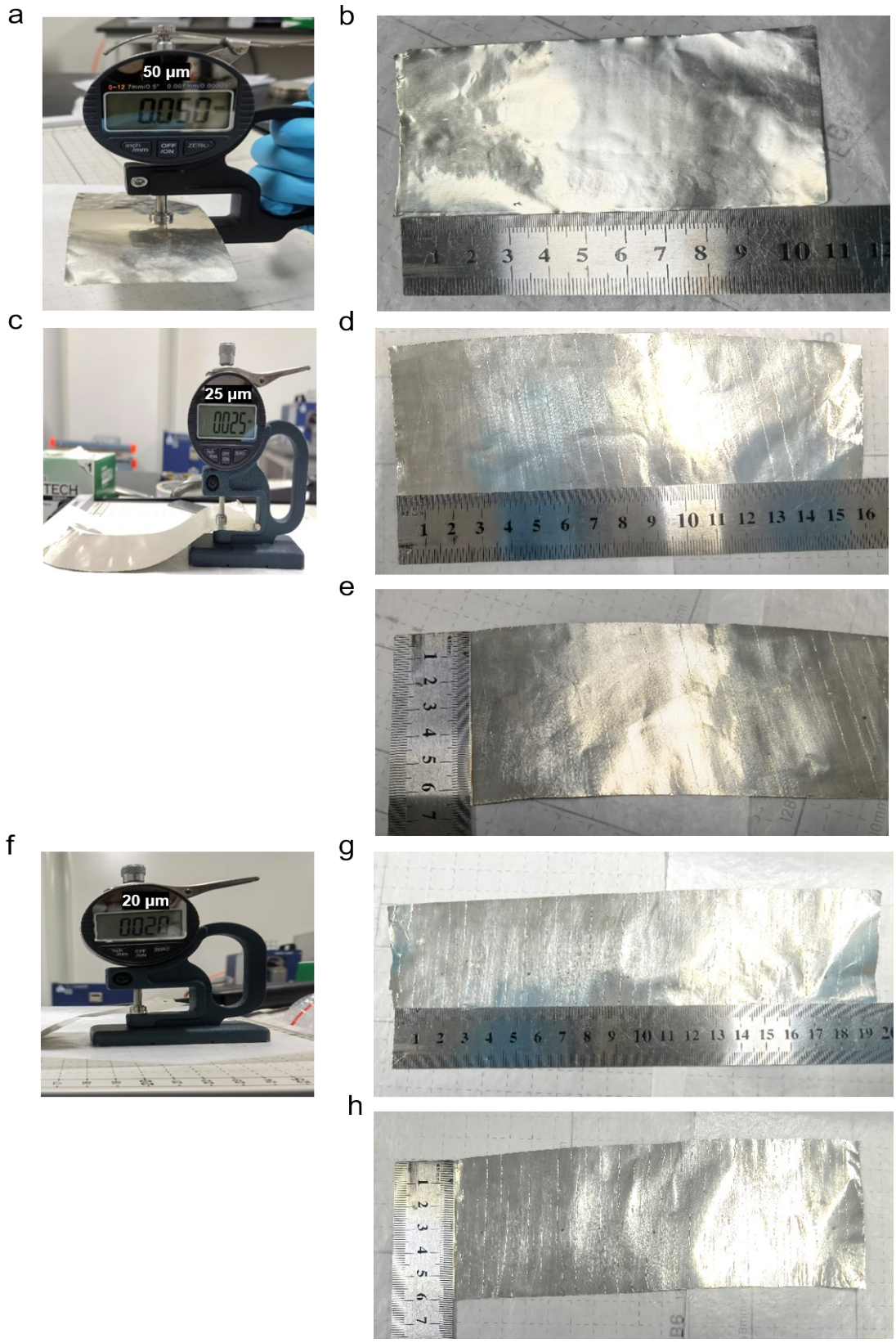

Figure S2. (a, b) Digital photographs of the as-prepared 50 µm-thick LiF@LiZn10/Li foil. (c-e) Digital photographs of the as-prepared 25 µm-thick LiF@LiZn10/Li foil. (f-h) Digital photographs of the as-prepared 20 µm-thick LiF@LiZn10/Li foil.

Figure S3. Digital photograph of the LiF@LiZn10/Li foil with 5 µm-thickness.

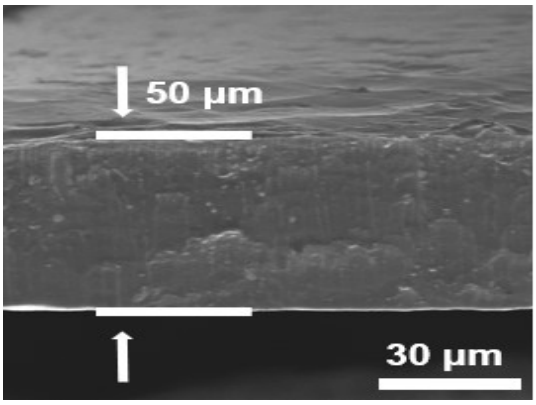

Figure S4. SEM images of the LiF@LiZn10/Li foil with 50 µm.

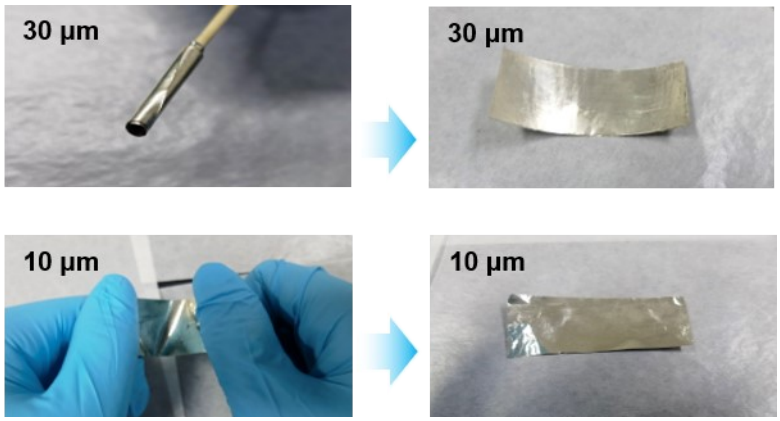

Figure S5. Digital photographs of the 30 µm-thick LiF@LiZn10/Li foil during the flexibility test and the 10 µm-thick LiF@LiZn10/Li foil during the bending-stretching test.

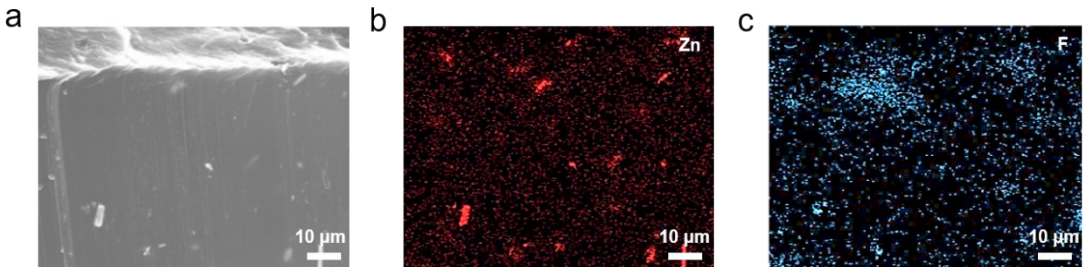

Figure S6. (a) SEM images on the cross-section of the LiF@LiZn10/Li foil. EDS illustrate the distribution of (b) the Zn element and (c) the F element on the cross-section of the LiF@LiZn10/Li foil.

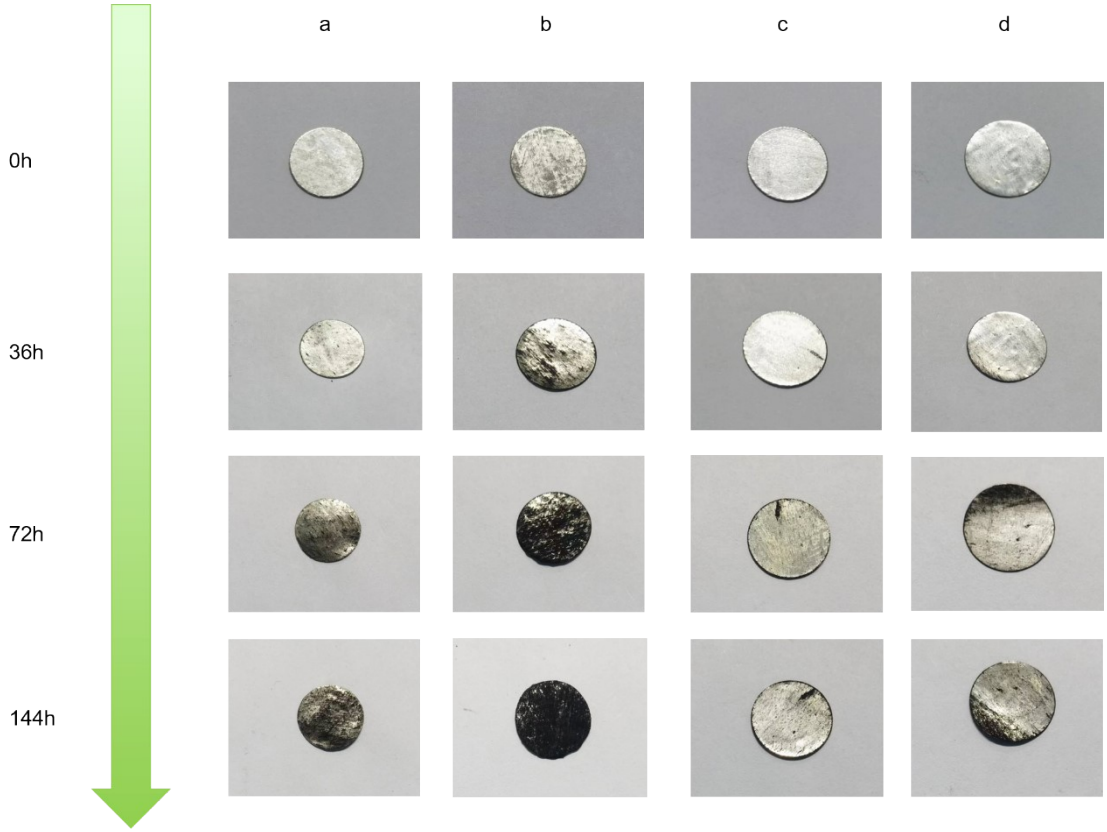

Figure S7. The surface change of (a) bare Li electrode exposed in a controlled environment with a relative humidity (RH) of 10%. (b) bare Li electrode exposed in a controlled environment with a relative humidity (RH) of 20%. (c) the LiF@LiZn10/Li foil electrodes exposed in a controlled environment with a relative humidity (RH) of 10%. (d) the LiF@LiZn10/Li foil electrode exposed in a controlled environment with a relative humidity (RH) of 20%.

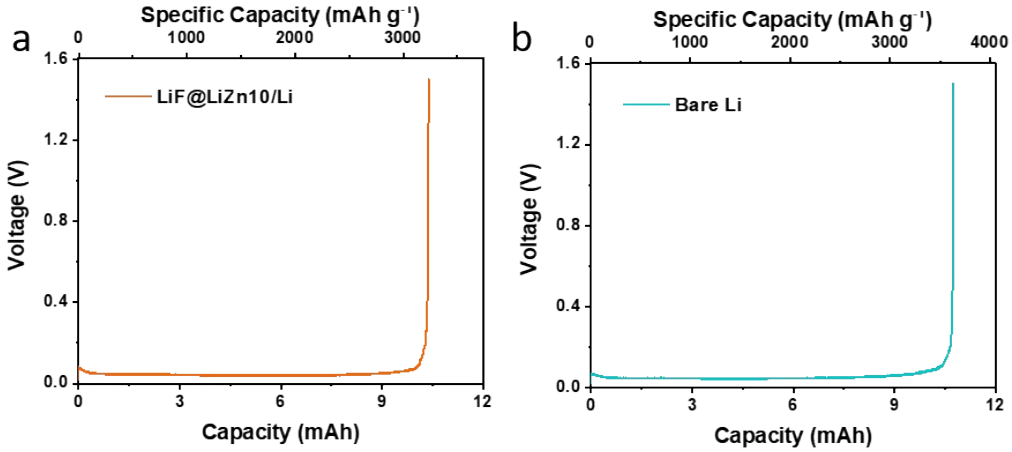

Figure S8. Delithiation curve of (a) bare Li electrode and (b) the LiF@LiZn10/Li electrode.

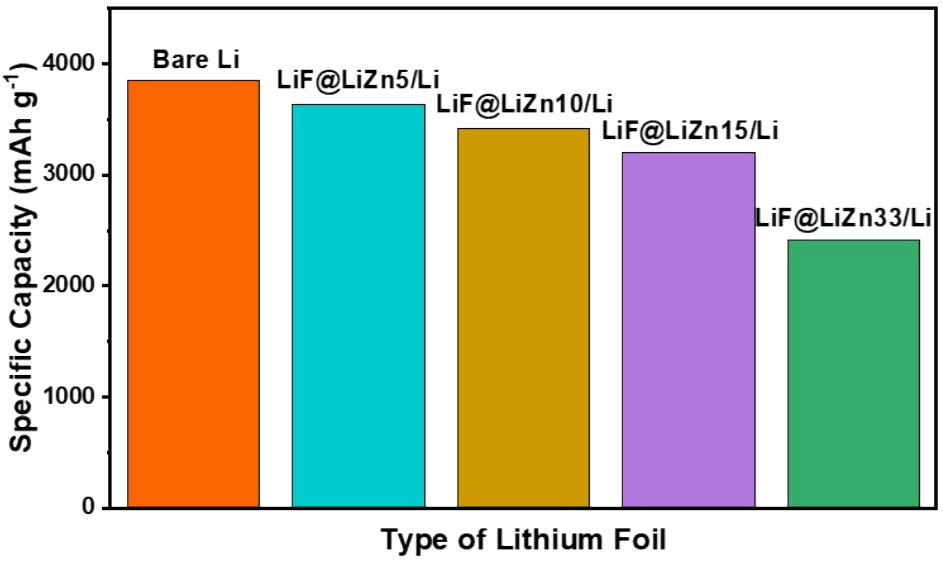

Figure S9. (a) The theoretical specific capacity of different types of lithium foil.

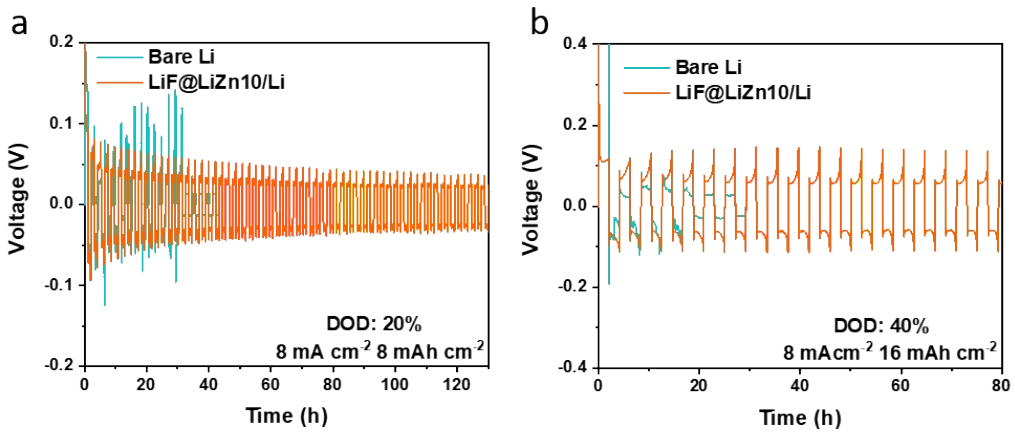

Figure S10. Cyclic performance of bare Li and the LiF@LiZn10/Li symmetric cells at a current density of 8 mA cm^{-2} with an areal capacity of (a) 8 mA h cm^{-2} and (b) 16 mA h cm^{-2}.

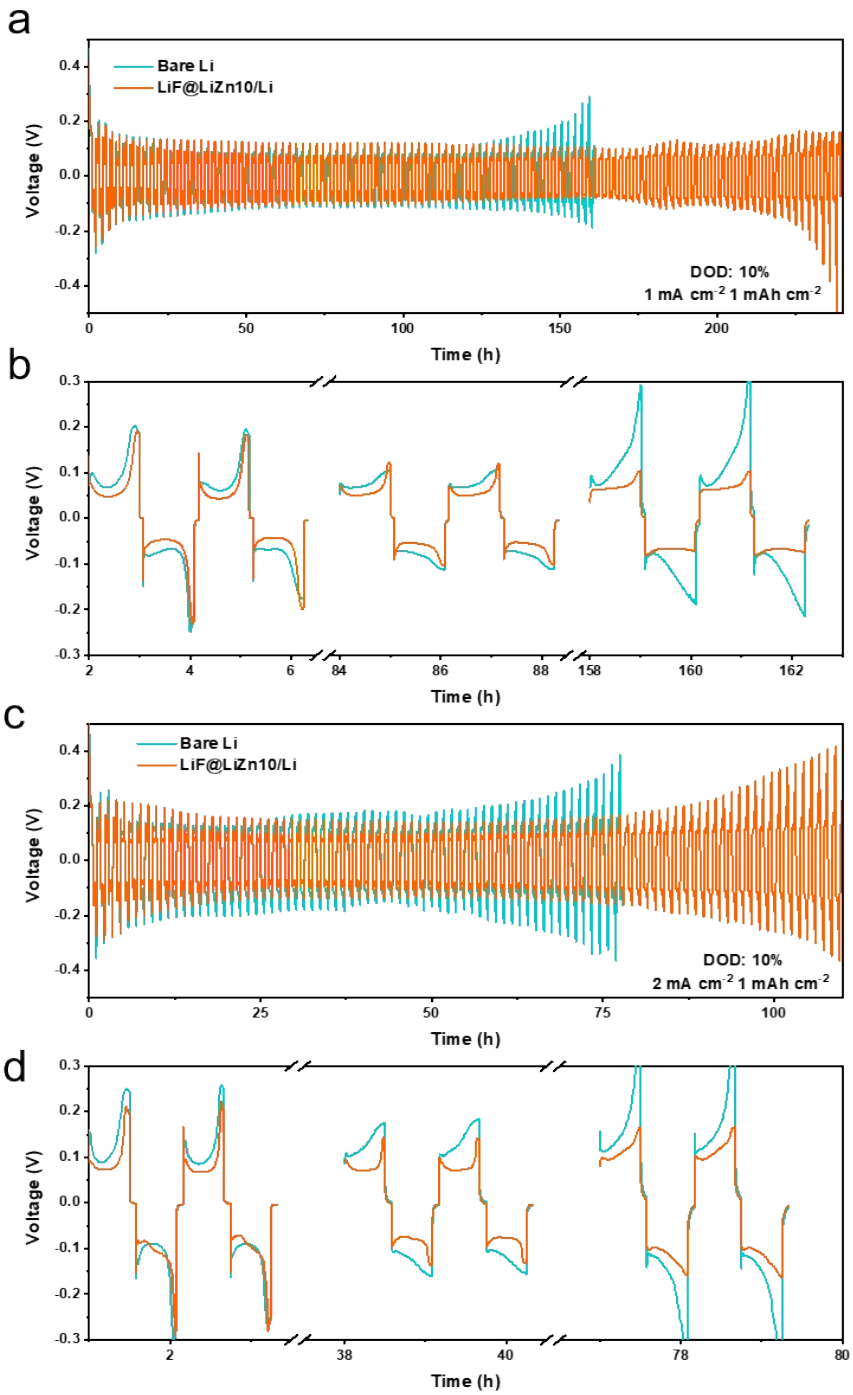

Figure S11. Cyclic performance of (a, b) the bare Li and the LiF@LiZn10/Li symmetric cells at 1 mA cm^{-2} and 1 mA h cm^{-2} in a carbonate ester electrolyte. (c, d) the bare Li and the LiF@LiZn10/Li symmetric cells at 2 mA cm^{-2} and 1 mA h cm^{-2} in a carbonate ester electrolyte.

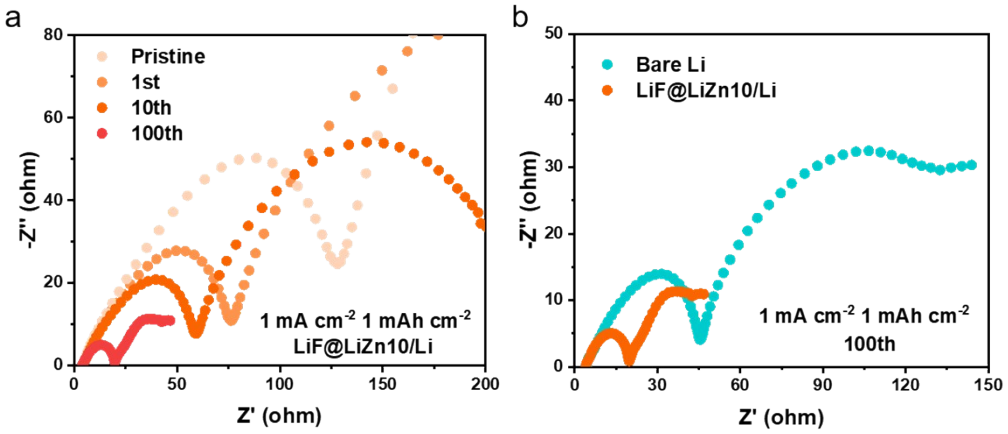

Figure S12. Nyquist plots of (a) the bare Li electrode and the LiF@LiZn10/Li foil electrode in symmetric cells. (b) the LiF@LiZn10/Li symmetric cells at 1 mA cm^{-2} and 1 mA h cm^{-2} after different cycles.

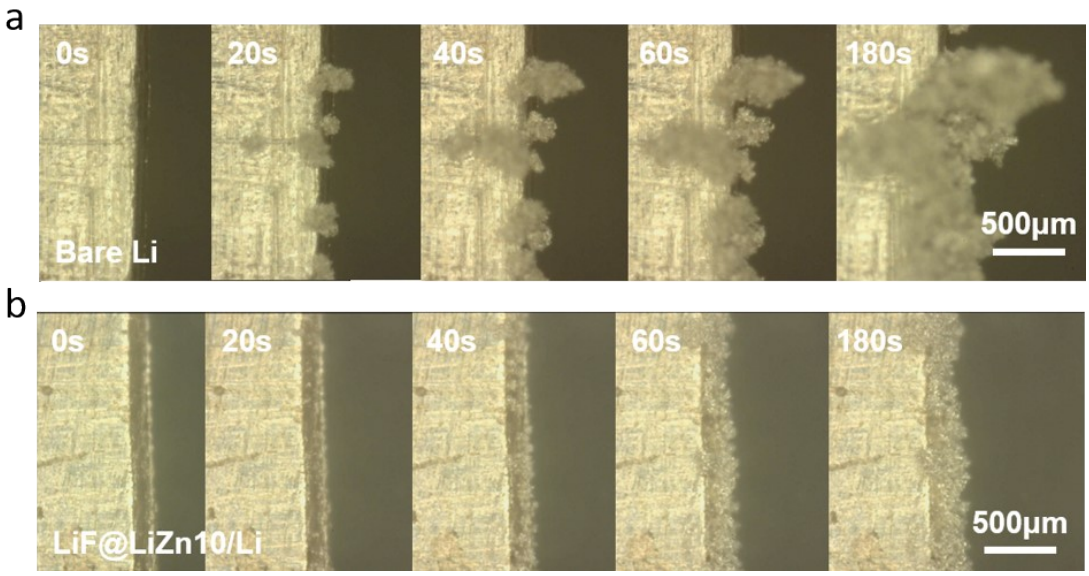

Figure S13. Operando optical microscopy images of (a) bare Li symmetric cells and (b) the LiF@LiZn10/Li symmetric cells.

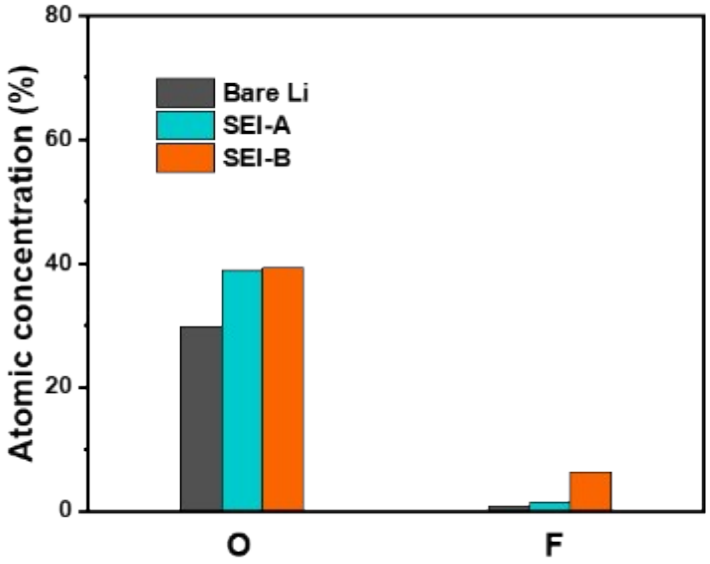

Figure S14. Atomic concentrations of O and F at the bare Li, SEI-A, and SEI-B according to XPS analysis.

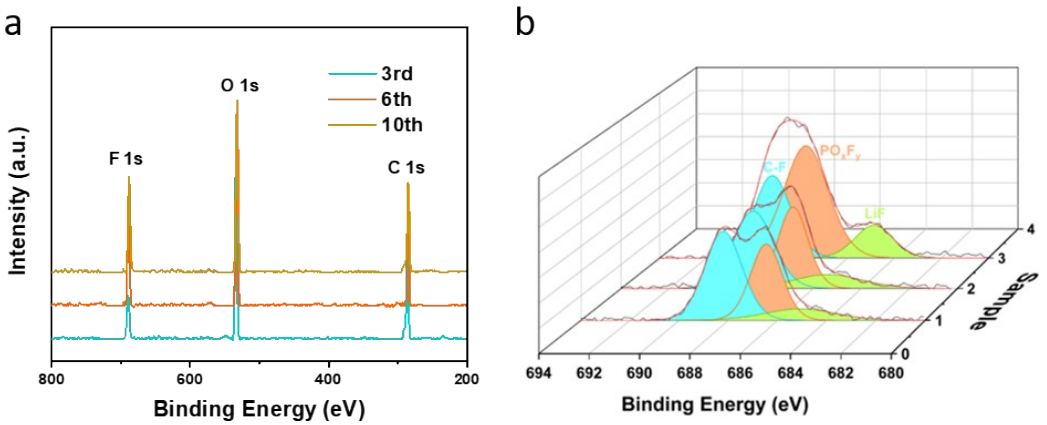

Figure S15. (a) XPS survey spectra of the LiF@LiZn10/Li symmetric cells after 3, 10, and 30 cycles. (b) High-resolution XPS spectra of F 1s on the LiF@LiZn10/Li surface after 3, 10, and 30 cycles.

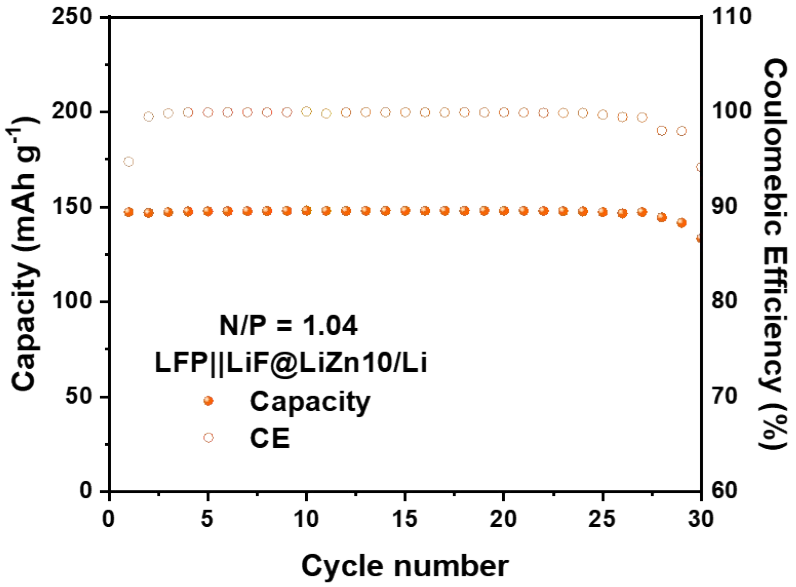

Figure S16. Cycling performance of LFP||LiF@LiZn10/Li full cells with 20 μm thicknesses of the LiF@LiZn10/Li electrode.

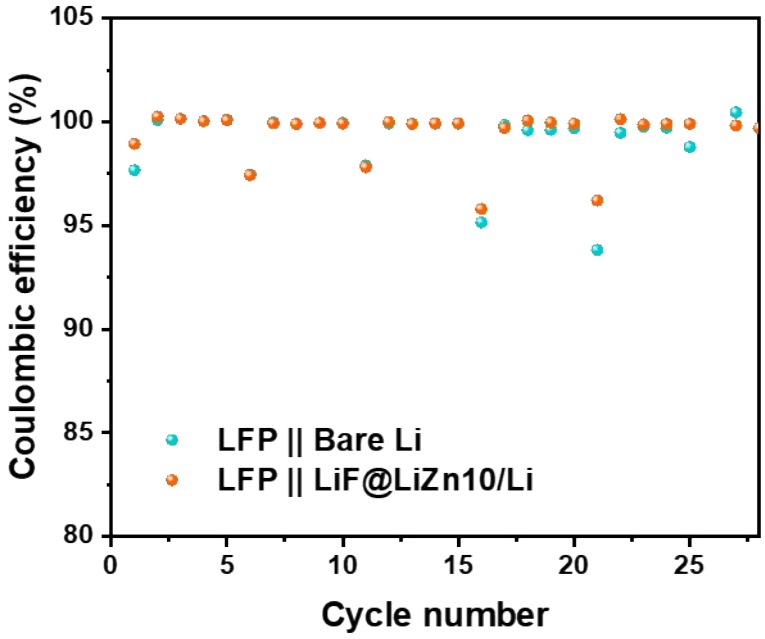

Figure S17. The Coulombic efficiency of the $LiFePO_4$||bare Li and $LiFePO_4$||LiF@LiZn10/Li full cells during rate capability.

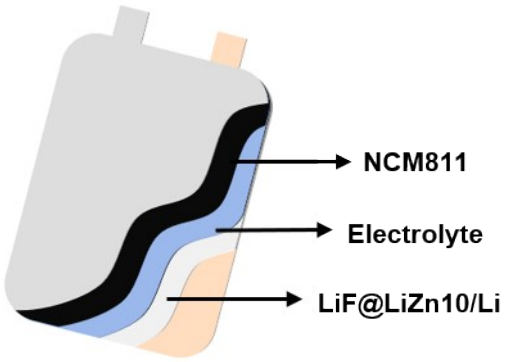

Figure S18. Schematic illustration of the NCM811||LiF@LiZn10/Li pouch cell with 0.13 Ah.

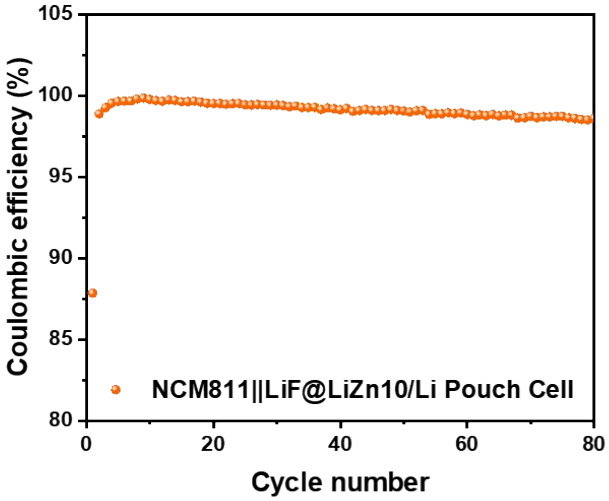

Figure S19. The Coulombic efficiency of the pouch battery during cycling.

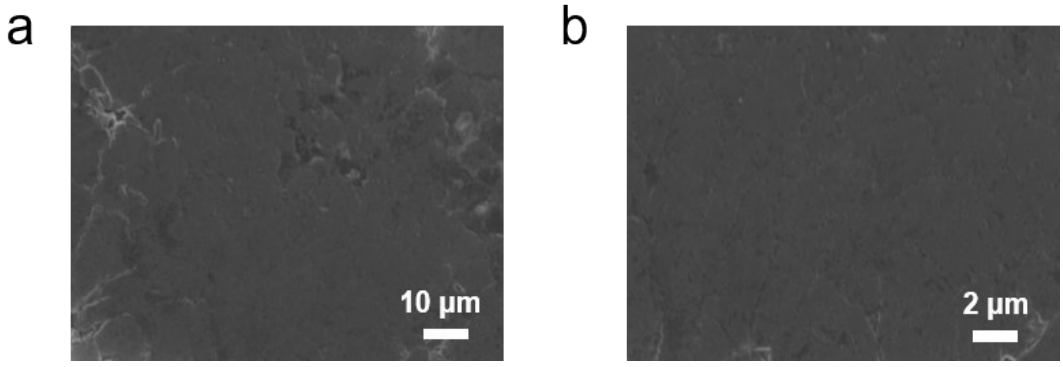

Figure S20. (a, b) SEM images of the surface of the LiF@LiZn10/Li foil composite electrode in pouch battery after 80 cycles.

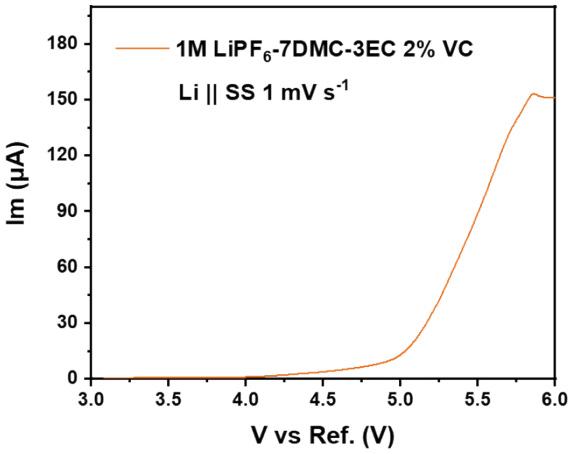

Figure S21. The LSV curve of the Li||SS (Stainless Steel) cell.

Table S1. Calculation of the theoretical specific capacity of the LiF@LiZn10/Li composite electrode

Mass ratio (ZnF_2)	Mass ratio (Li)	Ar (ZnF_2) / g mol^{-1}	Ar (Li) / g mol^{-1}	Specific capacity (Active Li) / g mol^{-1}
0	1	103.38	6.94	3860
5%	95%			3641.14
10%	90%			3422.28
15%	85%			3203.42
33%	67%			2415.20

Table S2. Parameters of the NCM811||LiF@LiZn10/Li pouch cell.

Parameters	Numerical value
Capacity	0.1302 Ah
Cathode	0.8692 g
Anode	0.1462 g
Electrolyte	0.4140 g
Mass	1.4294 g