Supplemental Information

Experimental Procedures

Preparation of LA foil

Firstly, pieces of high-purity Li metal foil were put in a stainless-steel crucible and heated to ~200℃, after which some high-purity Ag (99.99%) was added into the molten Li. The mixture was thoroughly stirred to accelerate the dissolving, after which the surface "rubbish layer" caused by residual O_2 and organic solvents in glove box was removed to obtain a fresh surface. Then the LA ingot was cooled down to room temperature. The above operations were all conducted in the Ar-glove box (H_2O≤0.01 ppm and O_2≤0.01ppm). Lastly, the Li-Ag block was pressed using a rolling machine into foils with a thickness of micrometers in dry room. The specific melting process was demonstrated in Supplemental video 4.

Electrode fabrication

LA foil with different thicknesses was cut into φ12(mm) disks for coin cell fabrication. Graphite anodes were prepared by mixing graphite powder (MTI) with polyvinylidene fluoride (MTI) and carbon black (TIMCAL) at a weight ratio of 90:5:5 using N-methyl-2-pyrrolidone (NMP) solvent. The slurry was blade-coated on copper foil (MTI), dried in an 80 °C vacuum oven and calendared before use. The areal mass loading of graphite was ~11.0 mg cm^{-2}. The cathode used in this work was commercial LFP (3 mAh cm^{-2}) and NCM811 (8 mAh cm^{-2}, double side).

Characterization

The morphologies and structures of Li-Ag foil were determined by a field emission scanning electron microscopy (SEM, FEI, QUANTA 250FEG) under an accelerate voltage of 10 kV. XRD measurements were carried out on a Bruker D8-Advance powder X-ray diffractometer operating at 40 kV and 30 mA, using Cu Kα radiation (λ = 0.15405 nm). Lattice spacing was performed on the transmission electron microscope (TEM, FEI Talos F200). XPS measurements was carried out on the American Thermo Fisher Scientific ESCALAB 250Xi, the sputtering space is 0.46 nm S^{-1}. A Perkein Elemer, Simutaneous Themal Analyzer STA 8000 was used for thermogravimetric analysis at a heating rate of 5 °C min^{-1}. A continuous stiffness

measurement using a Berkovich indenter was conducted.

Electrochemical cycling tests

1M lithium hexafluorophosphate (LiPF6) in a 1:1 vol/vol mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) with 10 wt% fluorinated ethylene carbonate (FEC) and 1 wt% vinylene carbonate (VC) was selected as electrolyte in Li (LA) metal cell. And 40 μL 1M lithium hexafluorophosphate (LiPF6) in in a 1:1 vol/vol mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) with 2 wt% vinylene carbonate (VC) was used in graphite coin cells.

All the coin cells were assembled in the CR2025-type coin cells, and the electrochemical performance test of cells were carried out by Neware test system (CT-4008, Neware) at 25 ℃. The Li-Ag||Li half-cells were delithiated to 1.5 V to detect its actual area capacity. The LFP||Li-Ag full cells were cycled between 2.0 V to 3.8 V at 0.3C. The electrochemical test on NCM//Li-Ag and NCM//Li pouch cells were carried out at 0.1C rate between 2.5~4.2 V. The graphite full cell was tested between 2.5 V and 3.8 V at 0.3C after four 0.1C formation cycles.

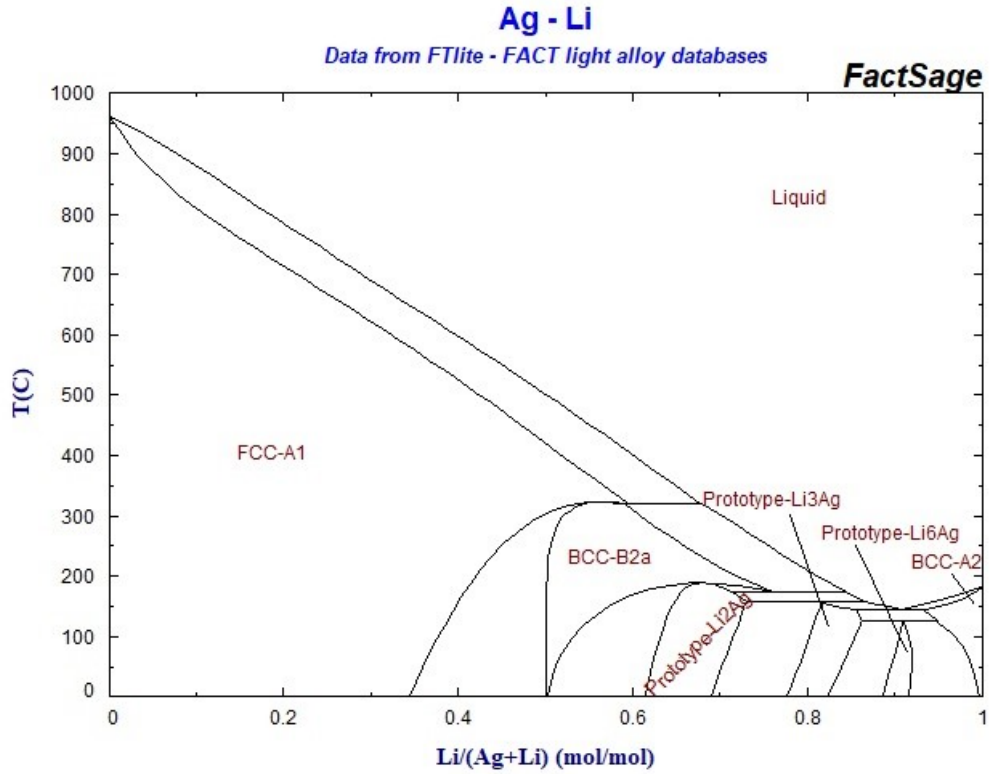

Fig. S1 Phase diagram of the Li-Ag.

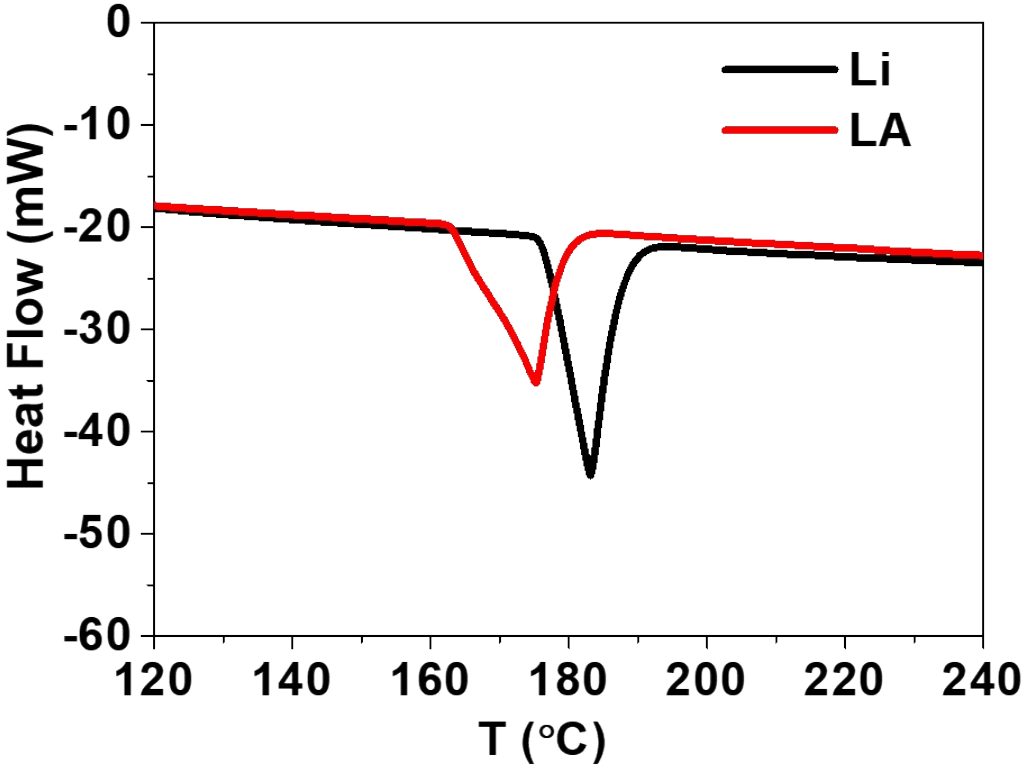

Fig. S2 Differential scanning calorimetry (DSC) results of the Li and LA51.

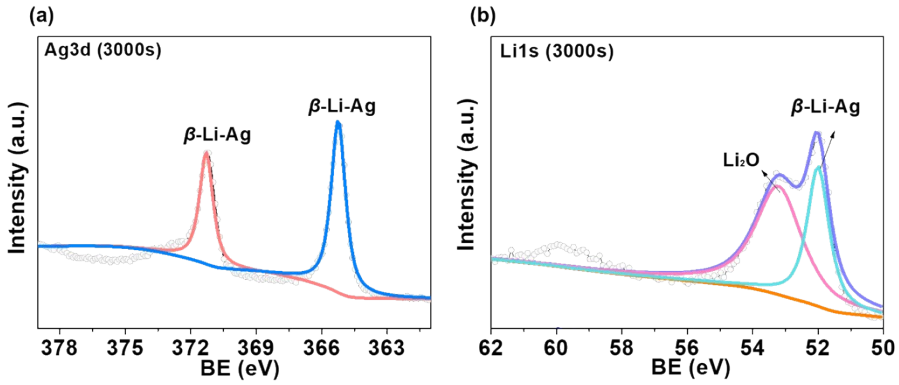

Fig. S3 Peaks of the Ag3d and Li1s of LA51 matrix.

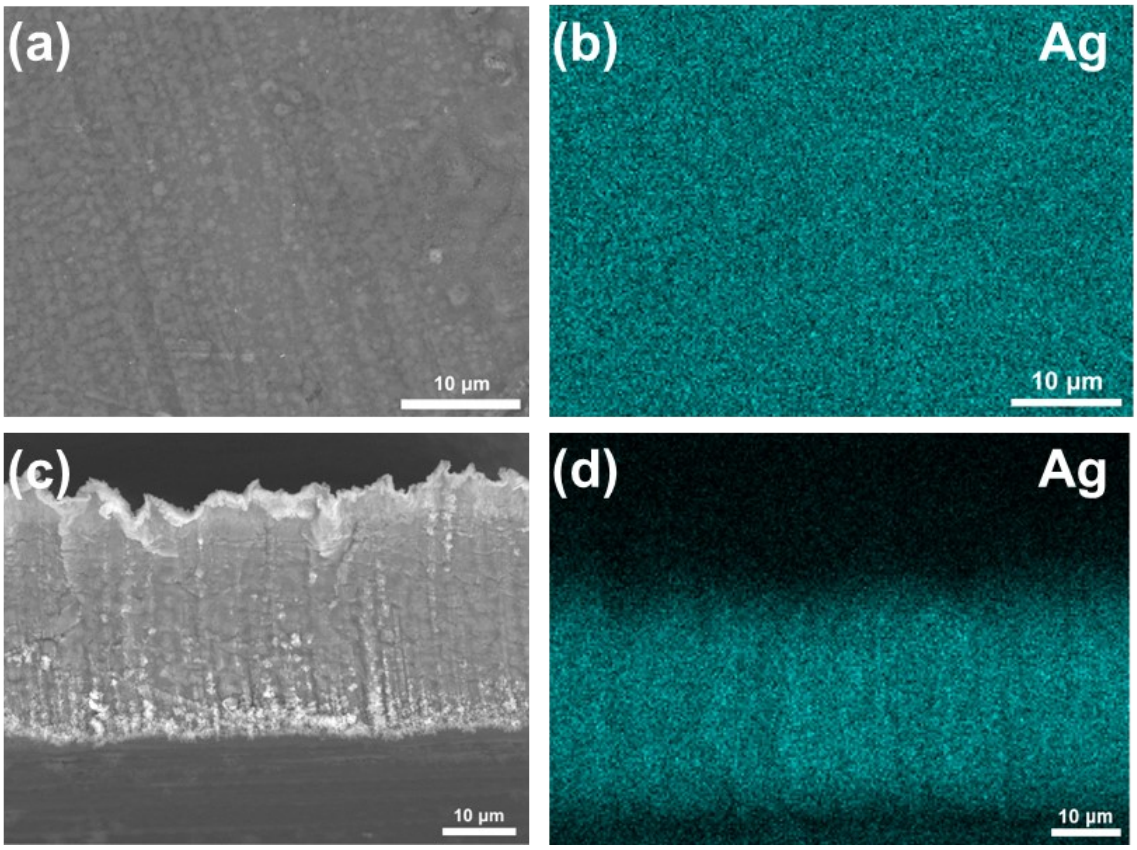

Fig. S4 Ag mapping on the surface (a-b) and cross section (c-d) of LA foil.

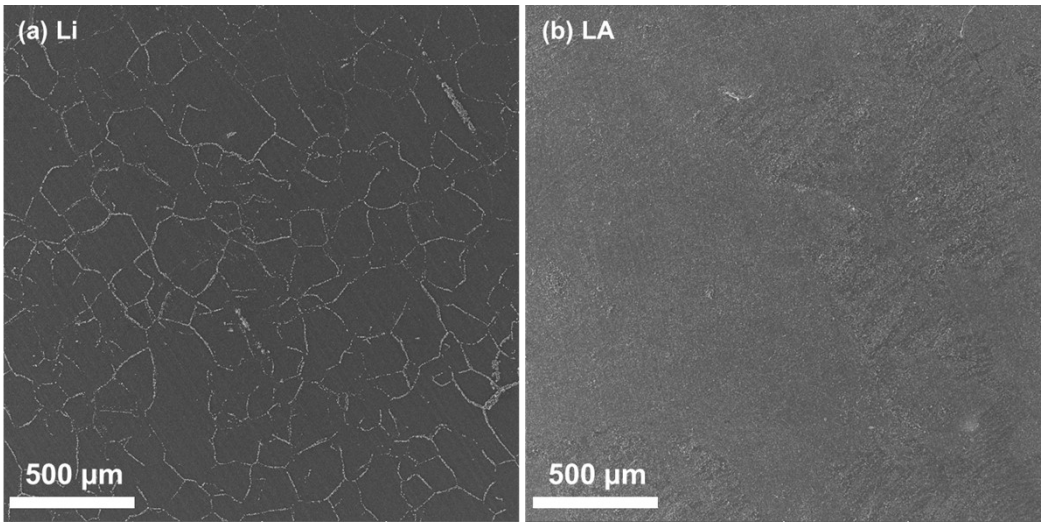

Fig. S5 SEM image of air-corroded surface of Li_{BCC} and LA_{BCC}.

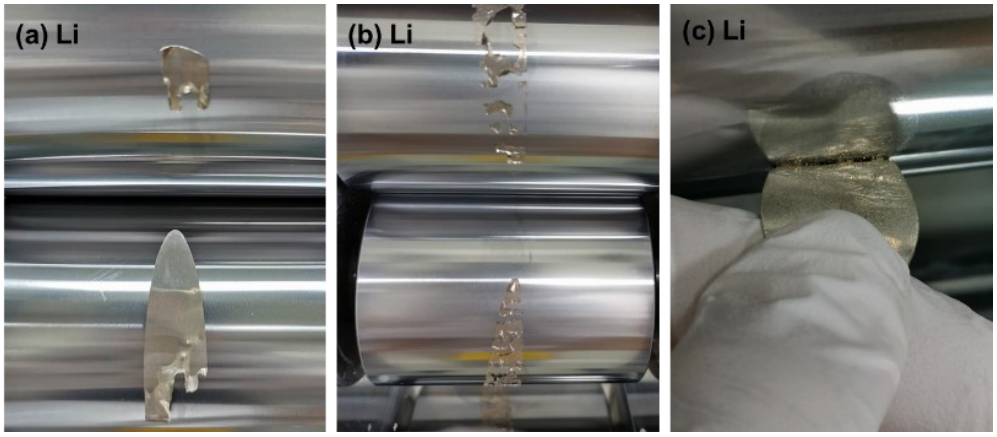

Fig. S6 (a-c) The tape-break and sticky property of Li foil.

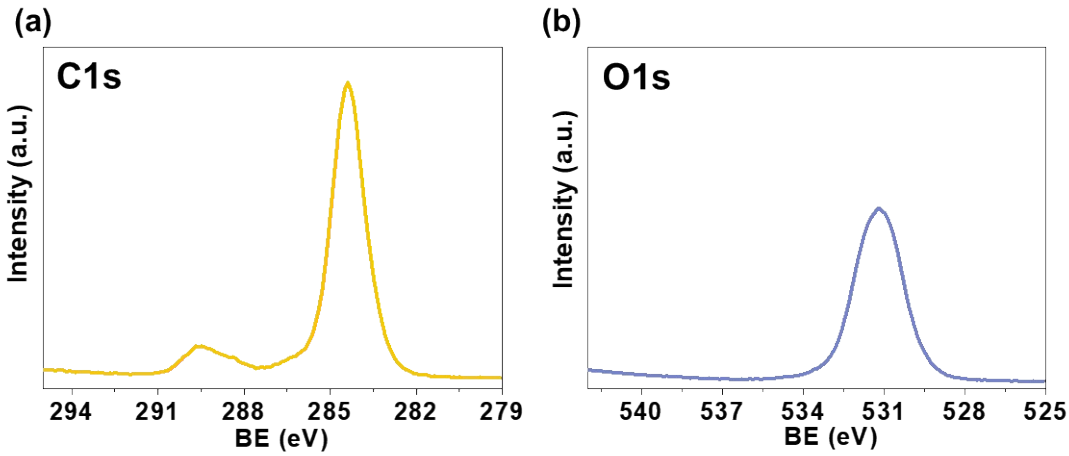

Fig. S7 (a-b) The C1s and O1s peak of the top surface of the LA foil.

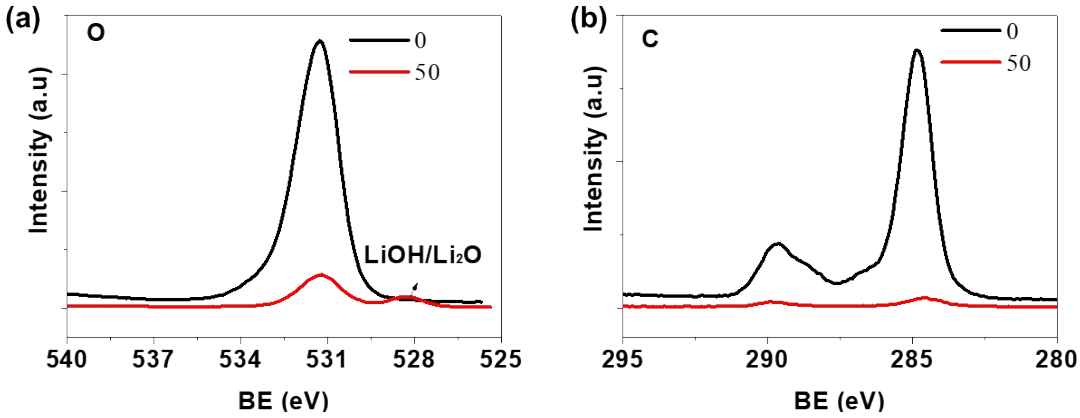

Fig. S8 (a-b) O1s (a) and C1s (b) signals of LA foil from the uppermost surface to the depth of 50 nm.

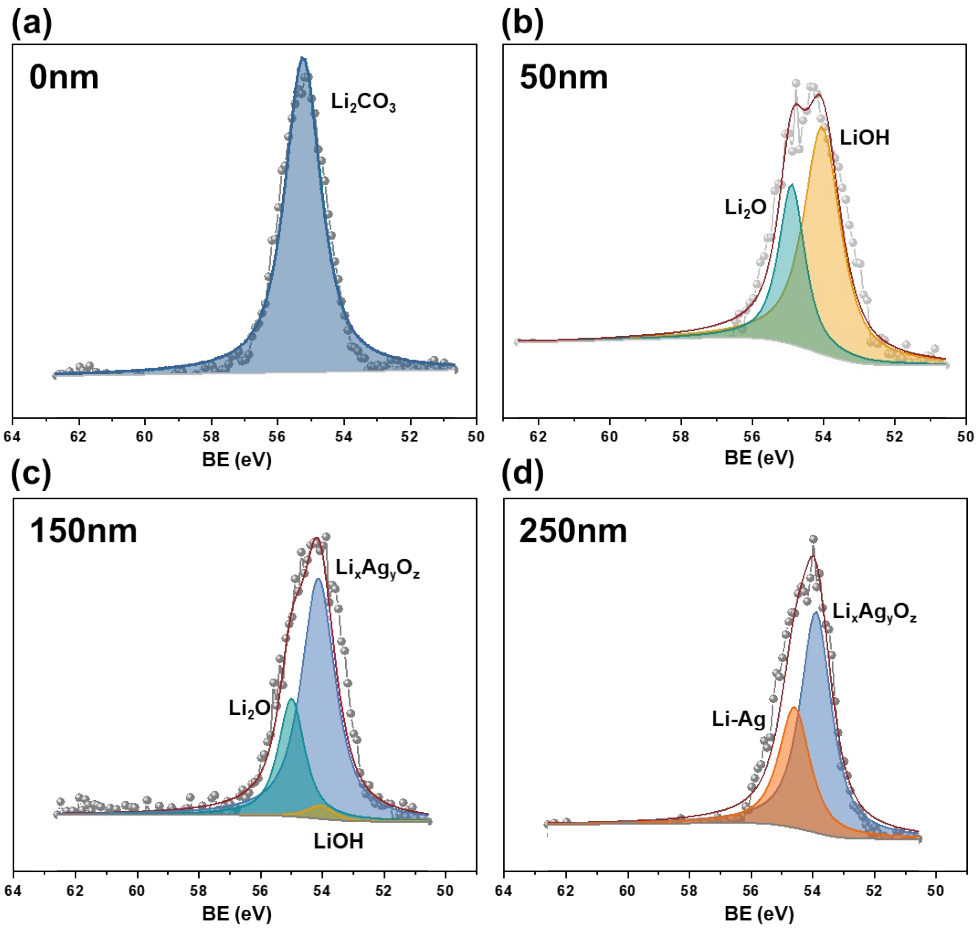

Fig. S9 Li1s spectra at 0nm, 50nm, 150nm and 250nm etching depth.

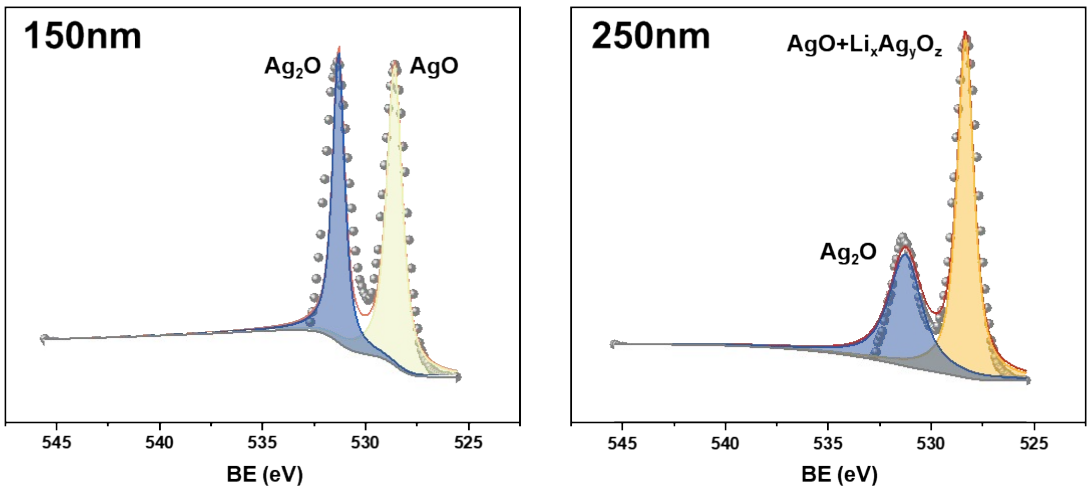

Fig. S10 XPS results of O1s peak at 150nm and 250nm etching depth.

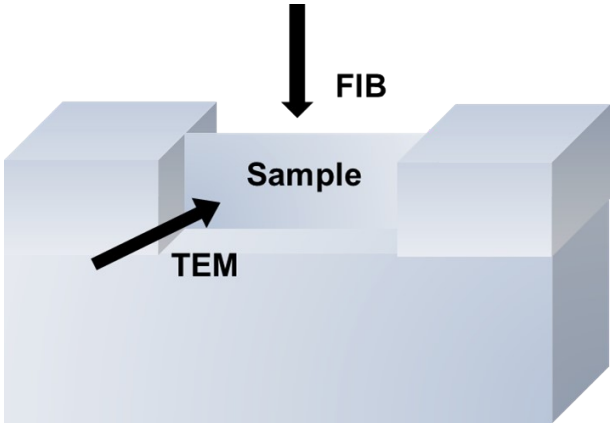

Fig. S11 Schematic graphic of TEM operation

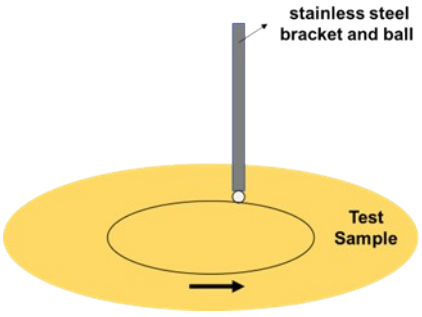

Fig. S12 Schematic diagram of friction test.

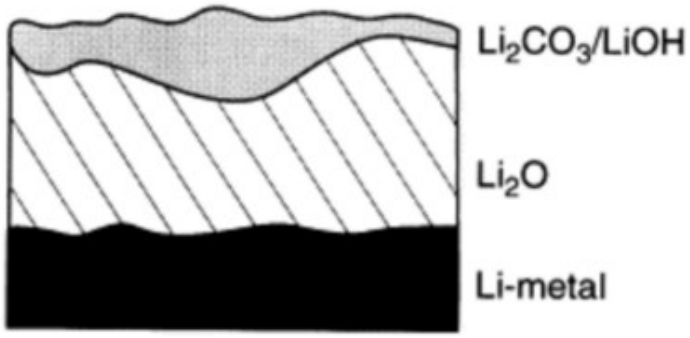

Fig. S13 Schematic diagram of the oxidized film on the Li foil.

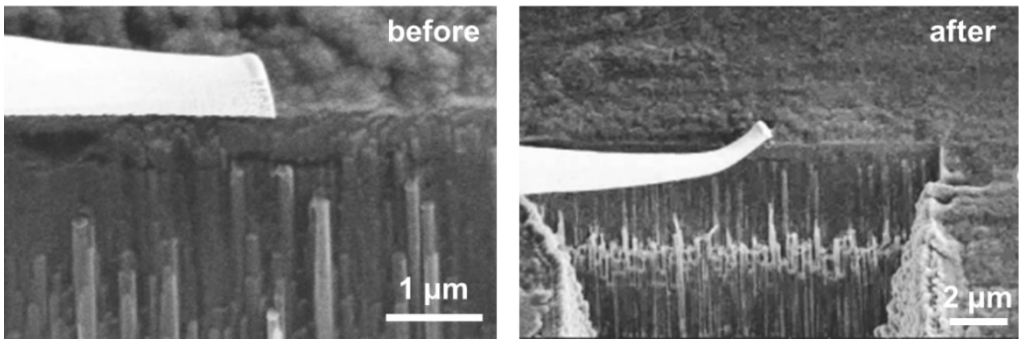

Fig. S14 The morphology of the tungsten needle before and after piercing the LA foil.

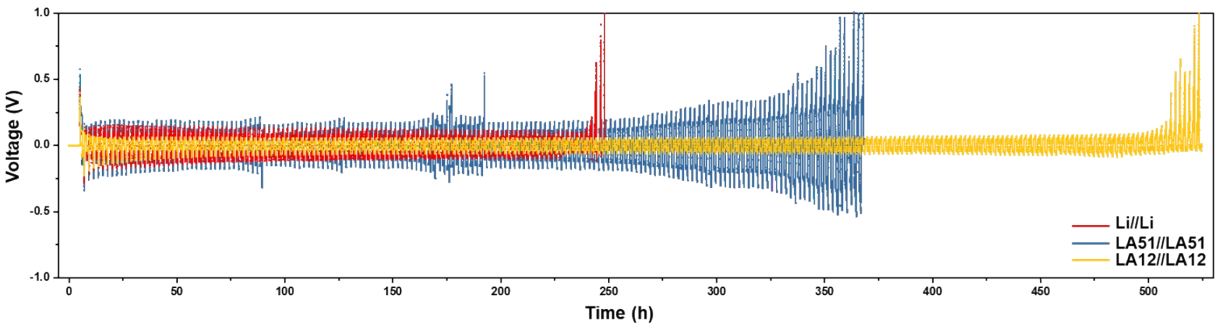

Fig. S15 Symmetric cell performance of Li, LA51 and LA12.

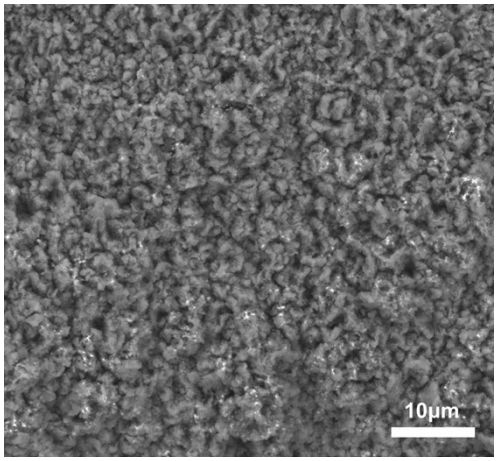

Fig. S16 Surface morphology of LA12 after extracting 80% Li.

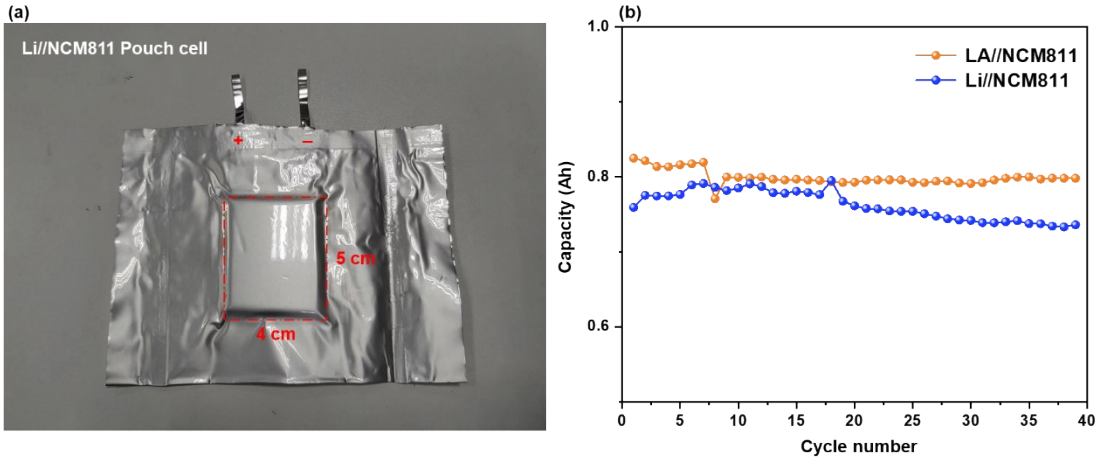

Fig. S17 (a) Prepared Li//NCM811 pouch cell. (b)Cycling performance of Li//NCM811 and LA//NCM811 pouch-cell in initial 40 cycles.

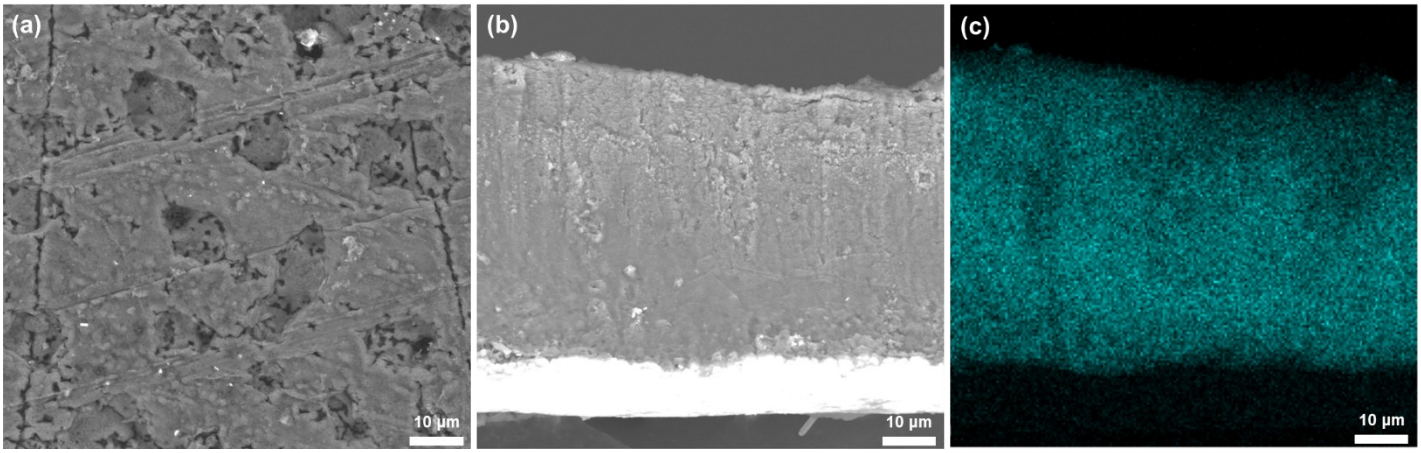

Fig. S18 (a-b) SEM image of cycled LA surface (a) and cross section (b). (c) Ag distribution of cycled LA cross section.

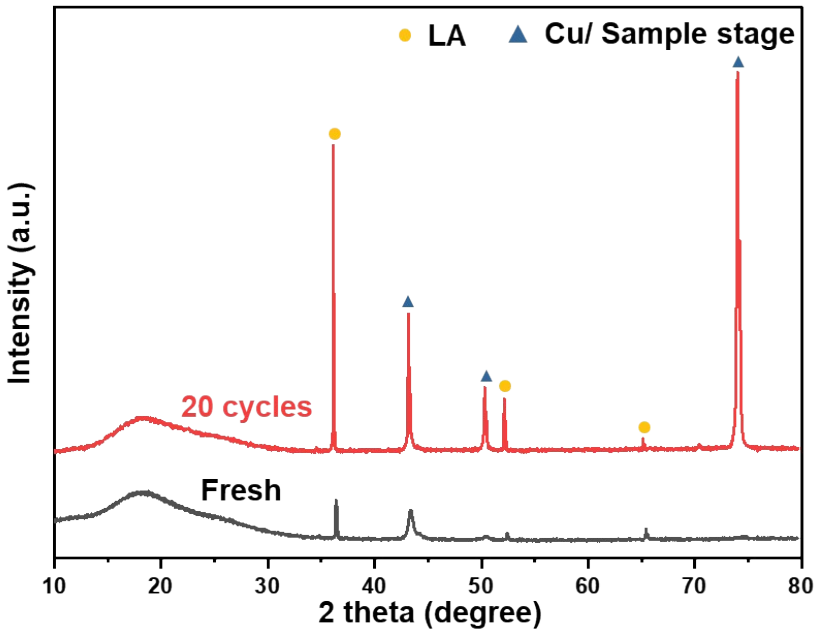

Fig. S19 XRD results of fresh and cycled LA

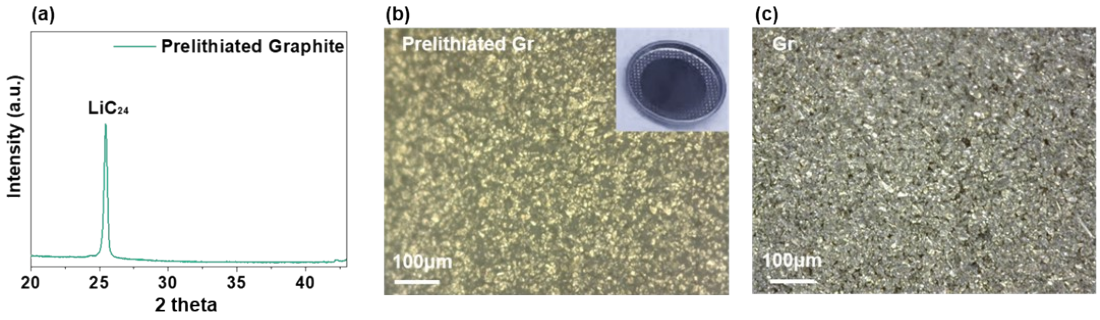

Fig. S20 (a) XRD characterization of the prelithiated Gr after rest. (b-c) Optical photos of the prelithiated-Gr (b) and pure Gr (c).

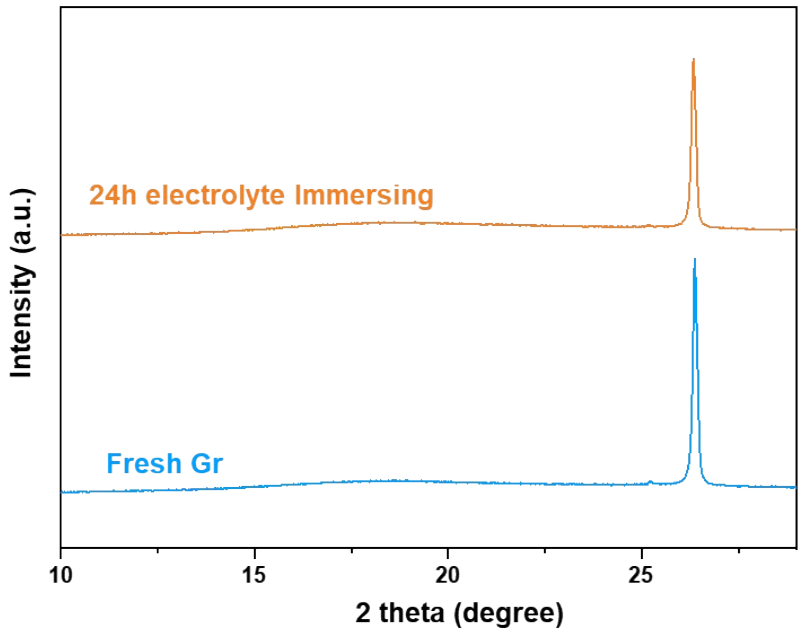

Fig. S21 XRD results of graphite after immersing into electrolyte for 24h without LA.

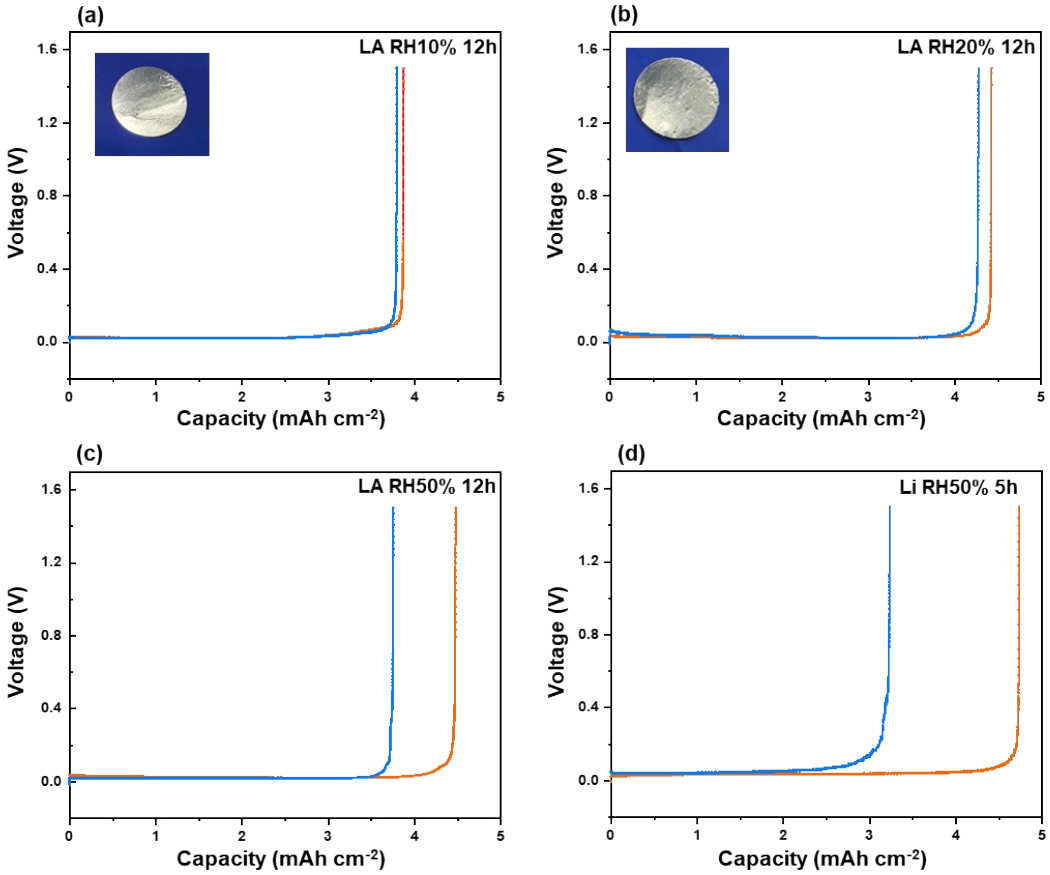

Fig. S22 Li extraction curves of LA and Li at different air stability test condition (orange line represents Li inventory before exposure, blue line represents the Li inventory after exposure): (a) LA, RH10% 12h. (b) LA, RH20% 12h. (c) LA, RH50% 12h.

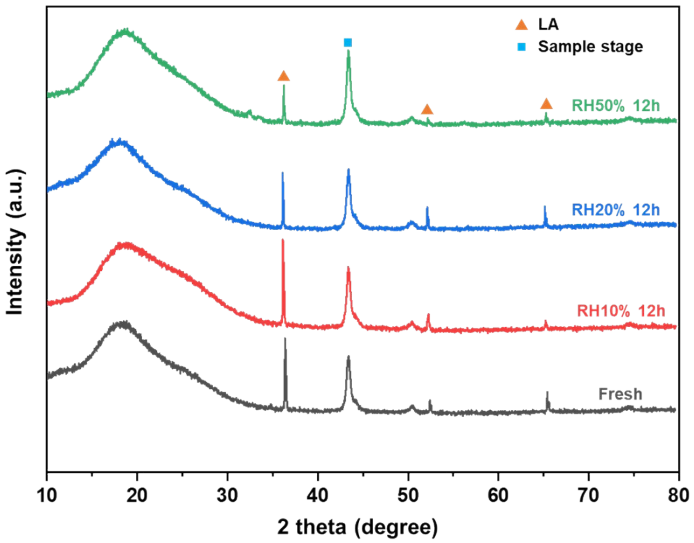

Fig. S23 XRD results of LA after different exposure condition.

Table.S1 Weighing record with a thickness of 5 μm and diameter of 12 mm for the LA12 and LA51 foil.

	1	2	3	4	5	6	7	8	9	10	11	12	Ave	per cm^2
LA12 (mg)	10	11	10	10	11	11	11	9	9	9	10	9	10.0	8.85
LA51 (mg)	4	4	4	3	4	4	3	4	4	4	4	3	3.75	3.31

Table.S2 Various parameters of LA//NCM pouch cell.

Capacity	0.82Ah
electrolyte	2.13g
cathode	5.43g
anode	0.320g
Celgard	0.355g
mass	8.235g

Table. S3 Comparison of energy density before and after prelithiation per square centimeter.

	Prelithiated	**Normal**
Capacity	2.9 mAh cm^{-2}	2.65 mAh cm^{-2}
Voltage	3.2 V	3.2 V
cathode	30.0 mg cm^{-2}	30.0 mg cm^{-2}
anode	19.5 mg cm^{-2}	19.5 mg cm^{-2}
Ultra-thin LA	3.3 mg cm^{-2}	0
GED	175.75 Wh kg^{-1}	171.31 Wh kg^{-1}

Table S4 Cost analysis of commercial Li foil and prepared LA foil

	Market price (US$ kg^{-1})	**Raw materials cost (US$ kg^{-1})**	**Processing cost (US$ kg^{-1})**
Commercial 50um Li foil	8583.7	420.6	8163.1
Commercial 5um Li foil	33488.1	420.6	33067.5
5um LA foil	/	104.8(Ag)+350.5(Li)	8163.1

Supplemental note S1

Calculation of the theoretical specific capacity of the Li_xAg_y foil:

$$C_{LA} = 3860 \times \frac{x \times M_{Li}}{x \times M_{Li} + y \times M_{Ag}} mAh\ g^{-1}$$

M_{Li} is the atomic weights of lithium and MAg is the atomic weights of sliver, and x and y are the atomic ratio of the Li and Ag, respectively.