Chemical lithiation induced $Li_{4.4}Sn$ lithiophilic layer for Anode-free Lithium Metal Batteries

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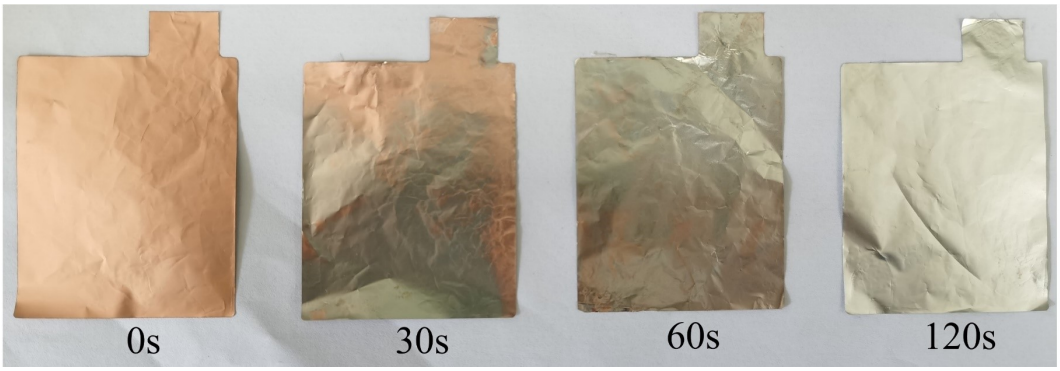

Figure S1. Optical image of Sn@Cu electrode with different processing time.

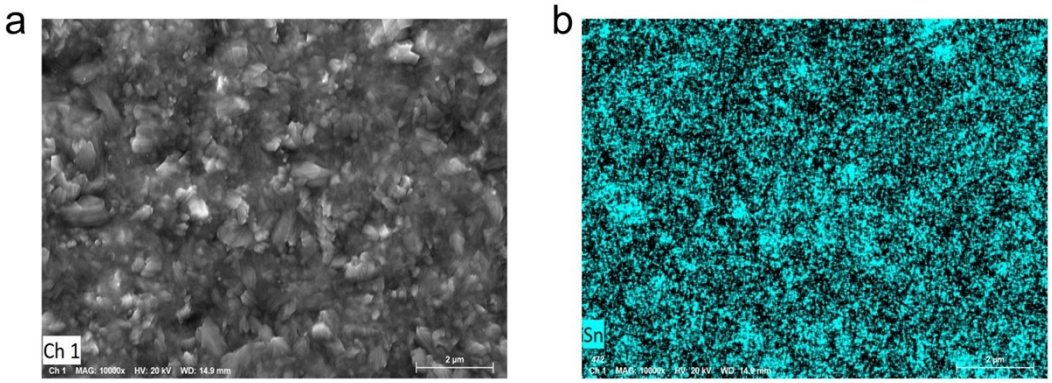

Figure S2. a) Top-view SEM image and b) elemental mapping of of Sn@Cu.

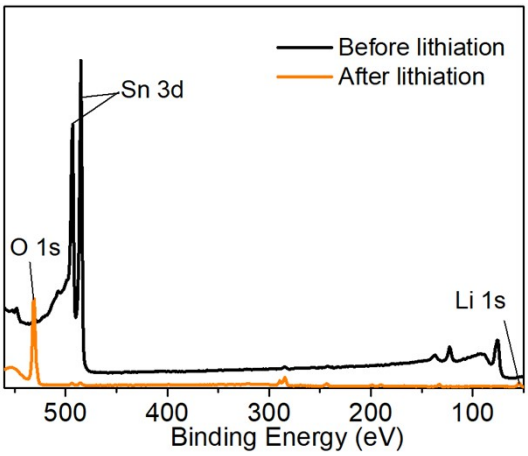

Figure S3. XPS spectra of Sn@Cu foil before and after lithiation.

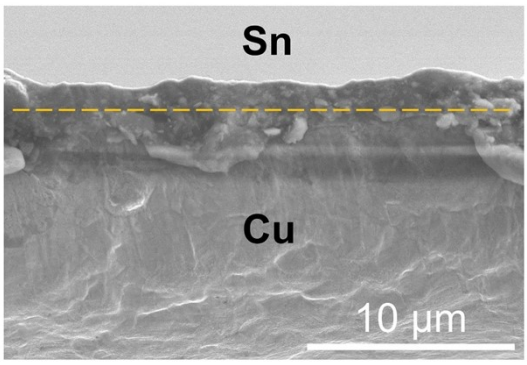

Figure S4. Cross-sectional SEM image of Sn@Cu before lithiation.

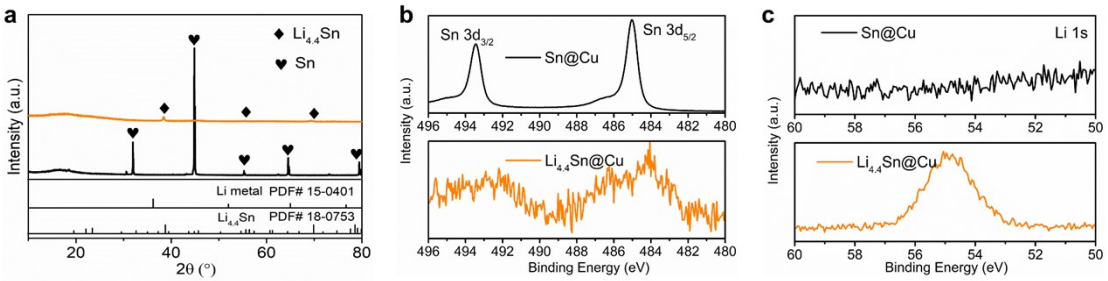

Figure S5.a) XRD patterns of $\underline{Li_{4.4}Sn@Cu}$ and Sn@Cu. b) Sn 3d XPS spectra of Sn@Cu (up) and $Li_{4.4}Sn@Cu$ (down) electrodes. c) Li 1s XPS spectra of Sn@Cu (up) and $\underline{Li_{4.4}Sn@Cu}$ (down).

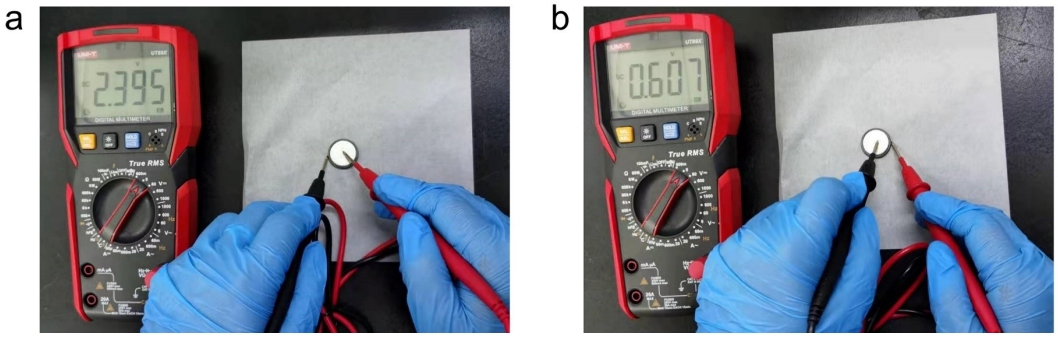

Figure S6. Measurement of open circuit voltage of a) $Li_{4.4}Sn@Cu$ / NCM811 cell and b) Cu / NCM811 cell.

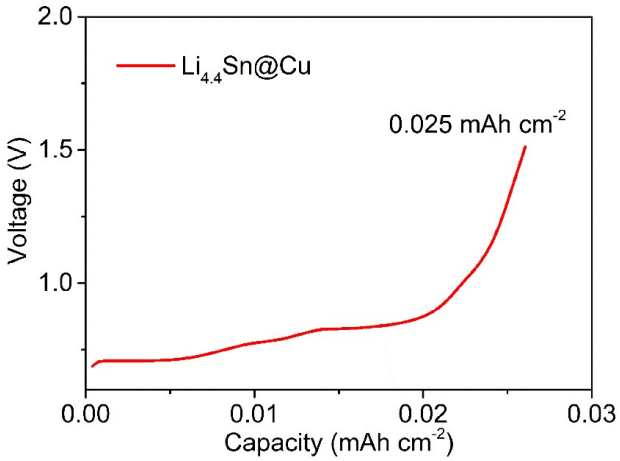

Figure S7. Measurement of lithium inventory of $Li_{4.4}Sn$@Cu electrode.

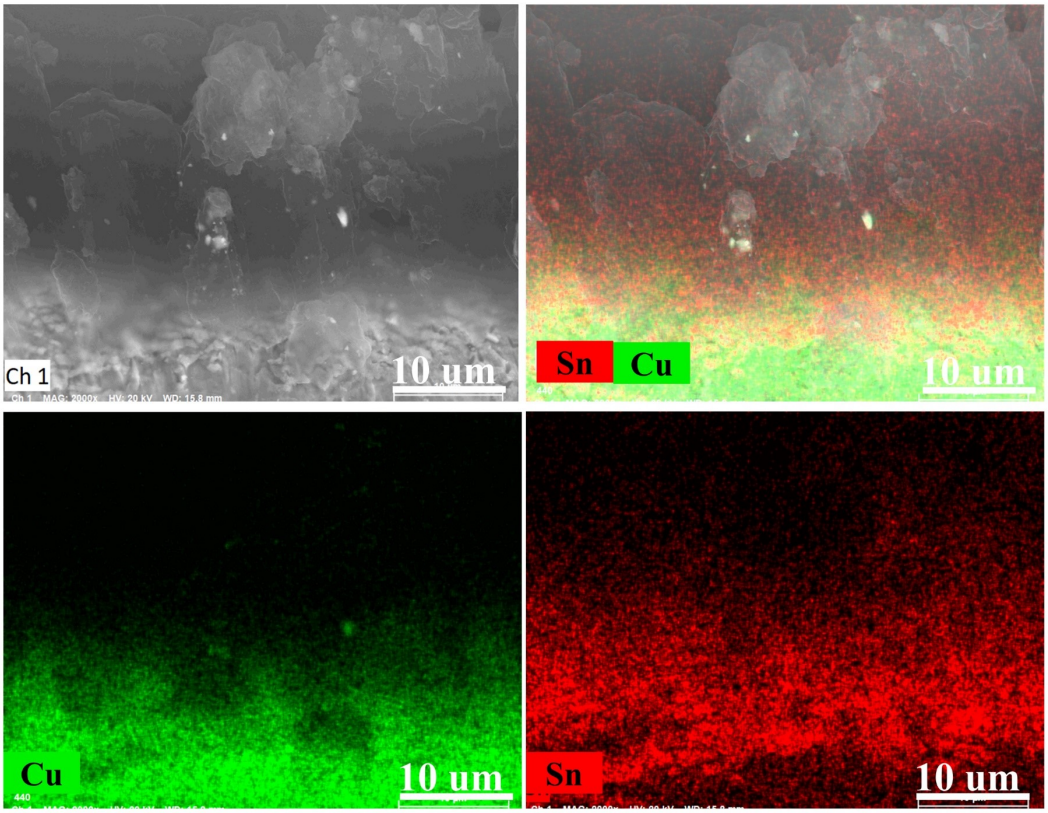

Figure S8. a) Cross-sectional SEM image of $Li_{4.4}Sn$@Cu electrode after 3 mAh cm^{-2} Li plating with a current density of 0.5 mA cm^{-2}, and b-d) the corresponding EDS mapping.

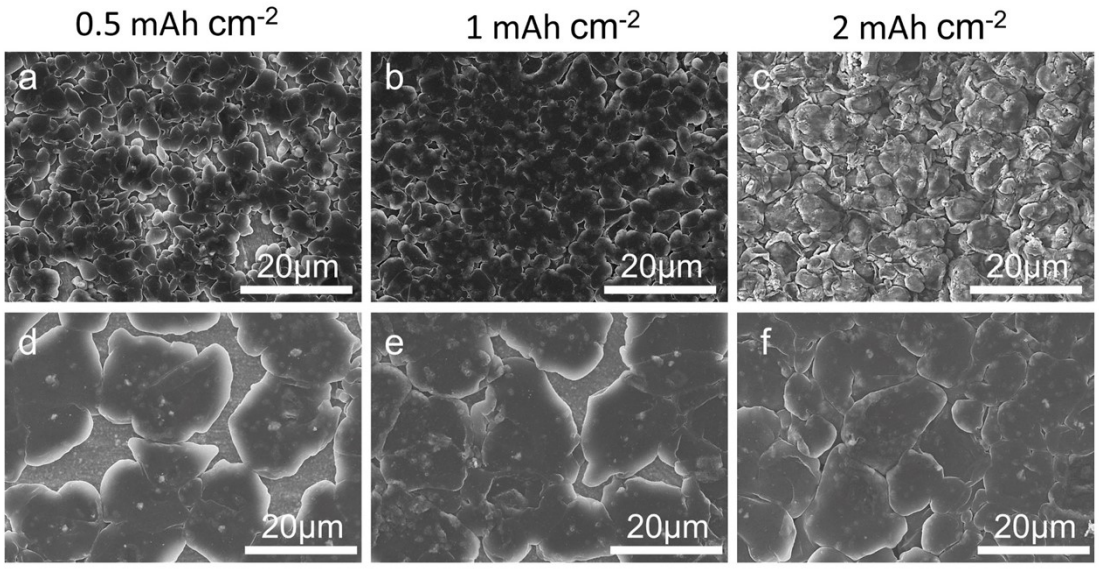

Figure S9. Surface morphology of Cu and $Li_{4.4}Sn$@Cu. a-c) Top-view SEM images of Cu after Li deposition with different areal capacities. d-f) Top-view SEM images of $Li_{4.4}Sn$@Cu after Li deposition with different areal capacities.

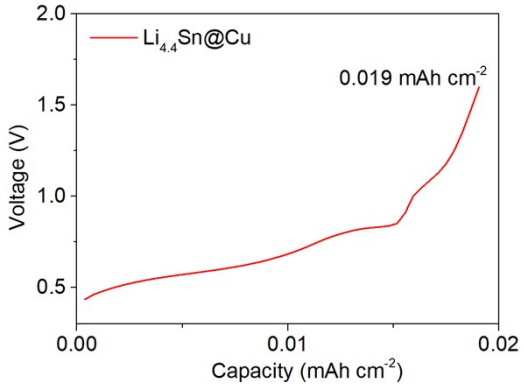

Figure S10. Lithium inventories of $Li_{4.4}Sn$@Cu electrodes after the first cycle measured by galvanostatic charging.

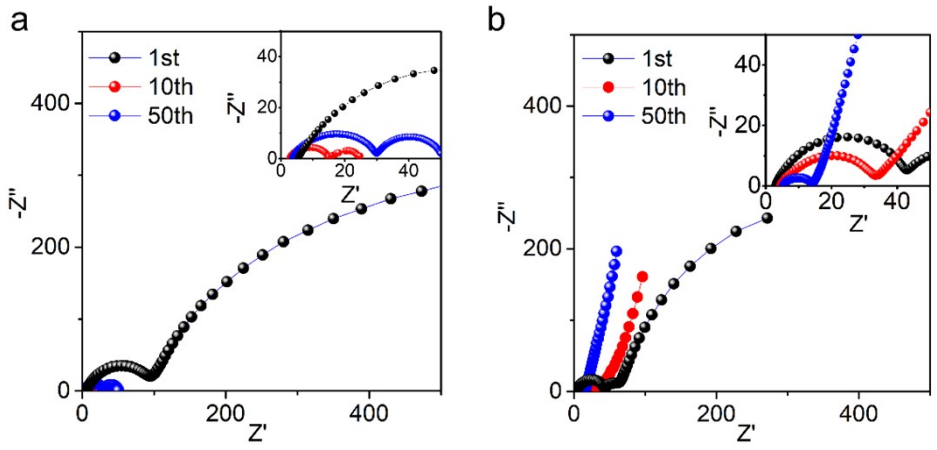

Figure S11. EIS analysis of the resistance evolution during cycling in half cells with Cu and $Li_{4.4}Sn$@Cu electrode.

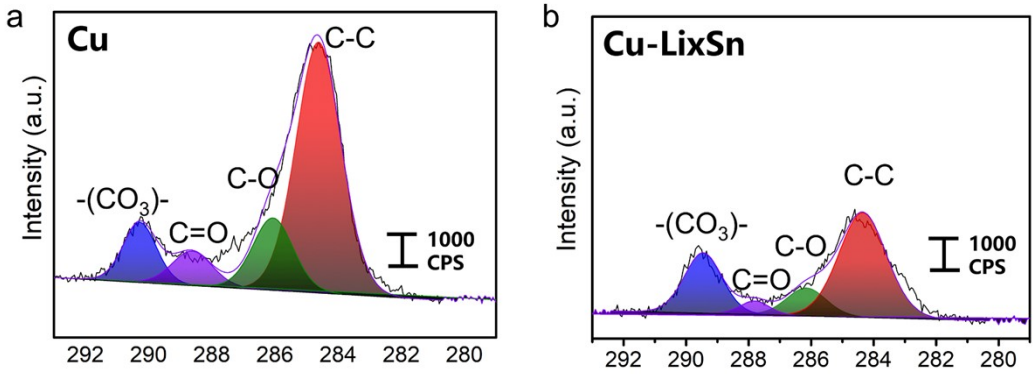

Figure S12. C 1s XPS spectra of a) Cu and b) $Li_{4.4}Sn$@Cu electrodes obtained from the cells after 10 cycles.

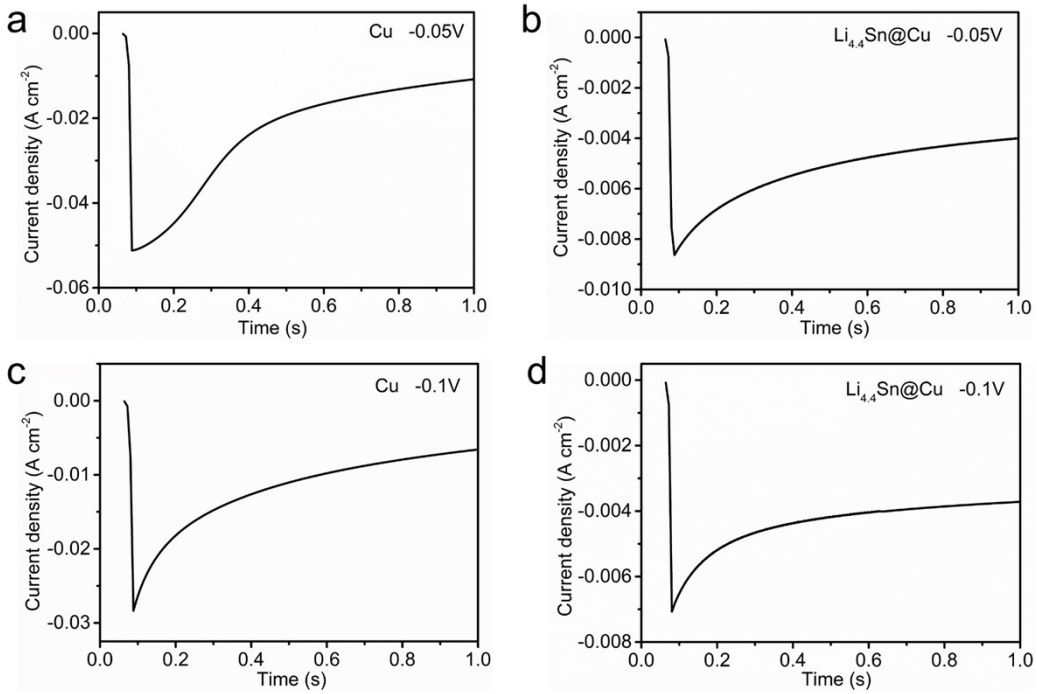

Figure S13. Current transients of Li deposition on different substrates at different potential. a, c) Cu. b, d) $Li_{4.4}Sn$@Cu.

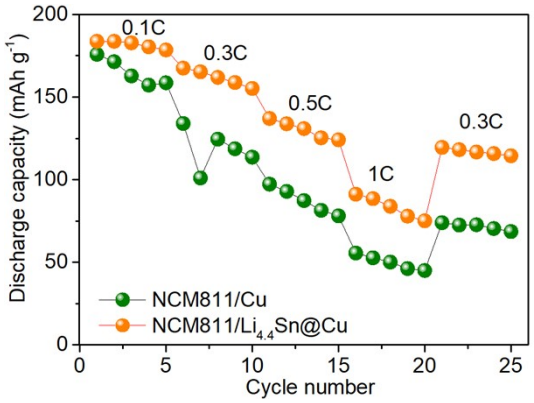

Figure S14. The rate performance of $Li_{4.4}Sn$@Cu/NCM811 and Cu/NCM81 full cells.

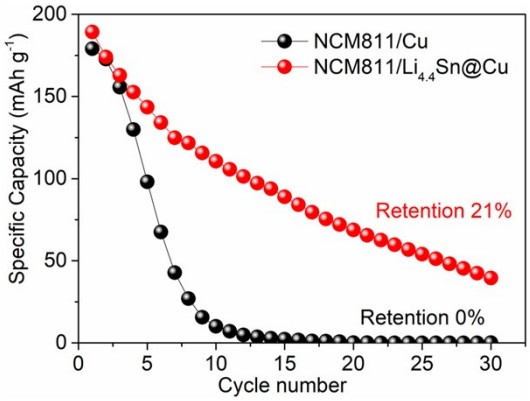

Figure S15. Cycling performance of NCM811/Cu and NCM811/$Li_{4.4}$Sn@Cu cells in carbonate-based electrolyte.

Table S1. The comparison on parameters of anode free coin cells in this work and previously reported works.

	Areal capacity (mAh cm^{-2})	Voltage (V)	Current density (mA)	Cycle number	Capacity retention (%)
This work	4	4.3	0.6	50	85.5
[18]	2.6	3.8	0.9	60	70
[22]	2	4.3	0.2	50	40
[27]	2	3.8	0.2	50	65
[29]	0.765	3.8	0.15	50	74
[35]	1.6	4.3	0.2	20	48
[36]	1.5	4	0.5	60	53
[37]	2.2	4.5	0.5	60	55.7

As the data of pouch-cell AFLMB related to current collector modification have rarely been reported, only comparisons of coin-type cell data were performed. It shows that our cell exhibits better overall performance, especially in high-capacity load cathode and cycle life.

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