

Supplementary Information

Positive electrode-Li metal crosstalk behavior-induced morphology change of Li deposits

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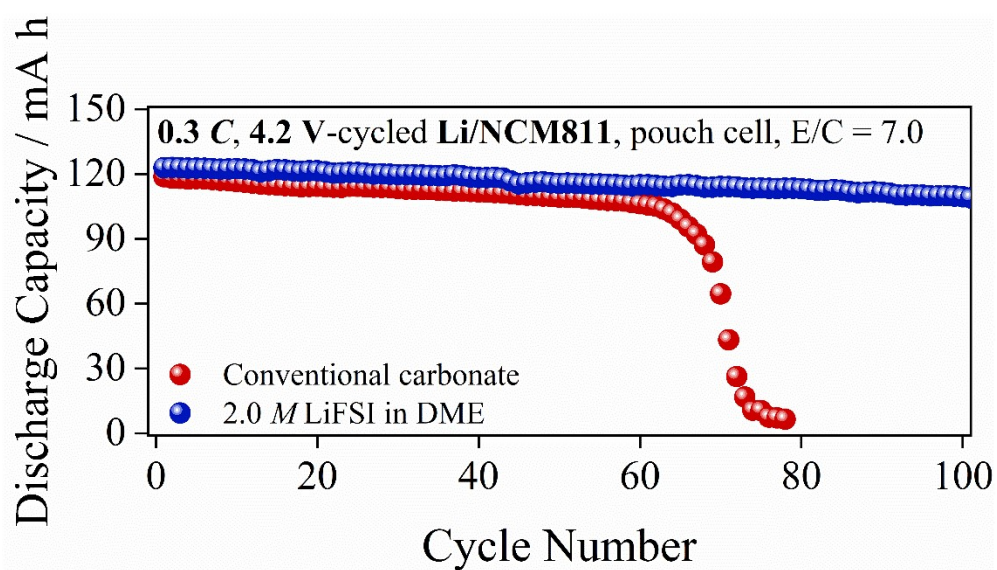


Figure S1. Li/NCM811 pouch cell evaluated with conventional carbonate electrode and 2.0 M LiFSI in DME electrolyte

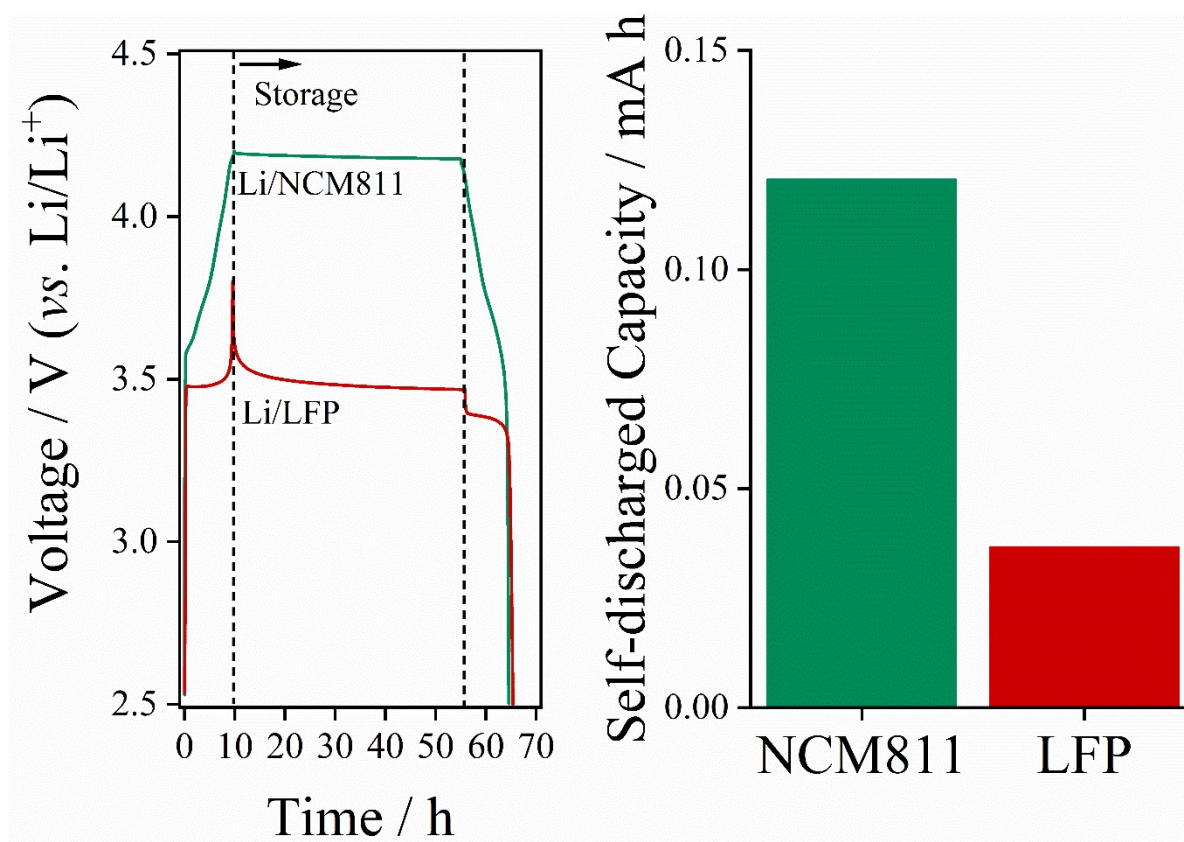


Figure S2. time vs. voltage curves during storage test of Li/NCM811 and Li/LFP cells and the calculated irreversible capacities of Li/NCM811 and Li/LFP cell, respectively. The storage was conducted with SOC 100 at 25 °C chamber

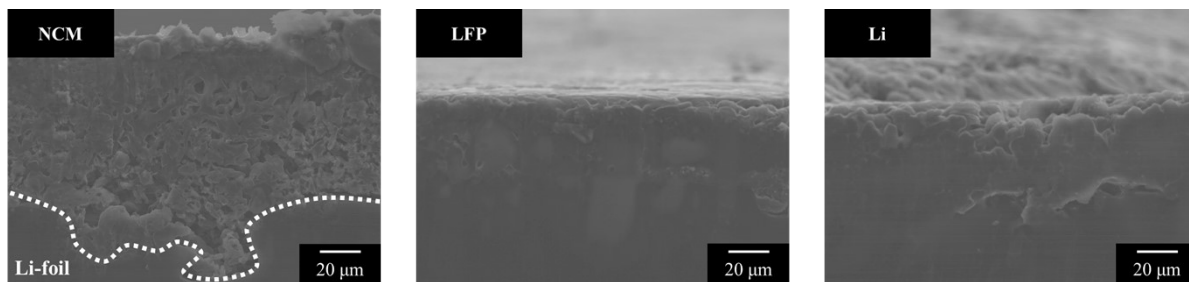


Figure S3. Cross-sectional SEM images from the 5th cycled Li electrode with Li, LFP, NCM811 counter electrodes, respectively.

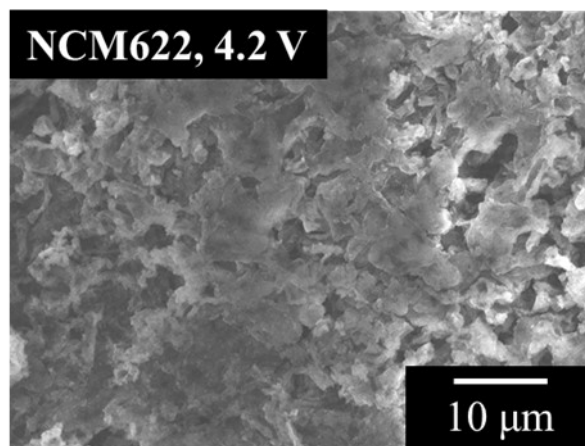


Figure S4. SEM image of 5th cycled Li electrode with NCM622 counter electrode.

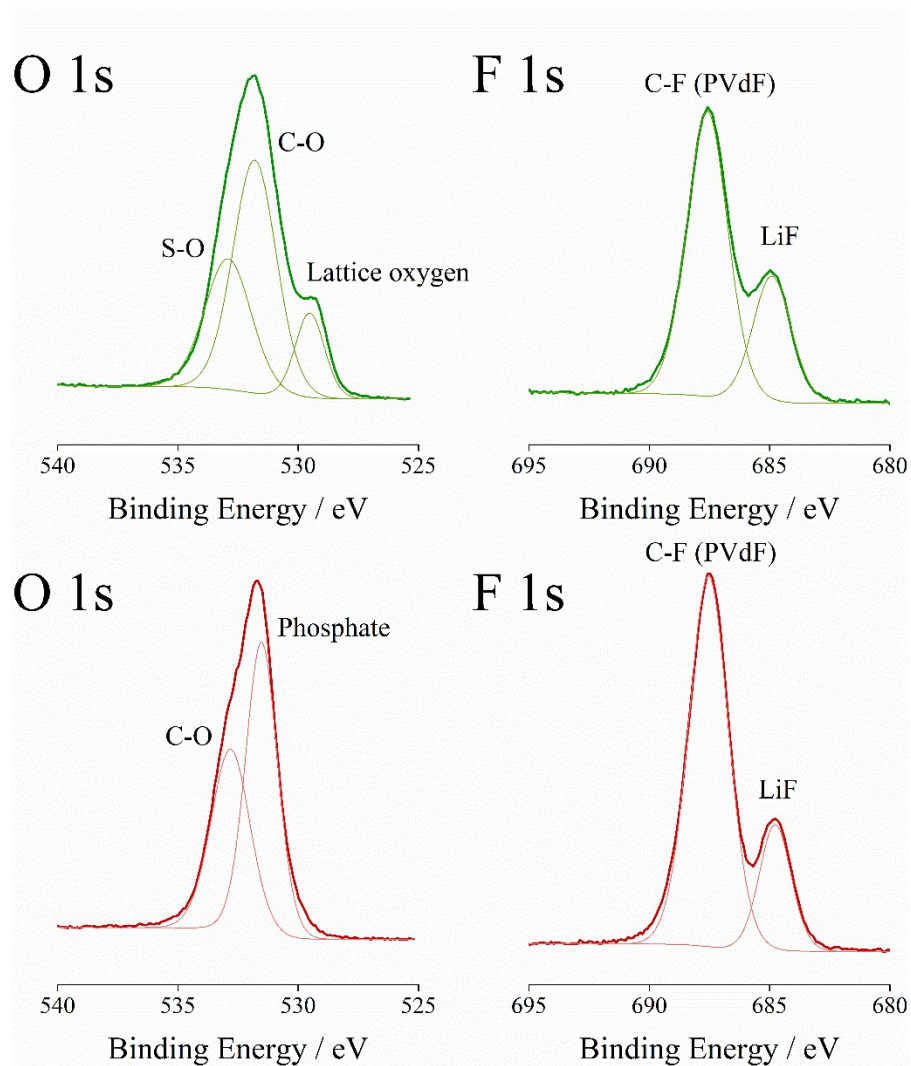


Figure S5. Post-mortem O 1s and F 1s XPS spectra obtained from the positive electrodes of (a) Li/NCM811 and (b) Li/LFP cells.

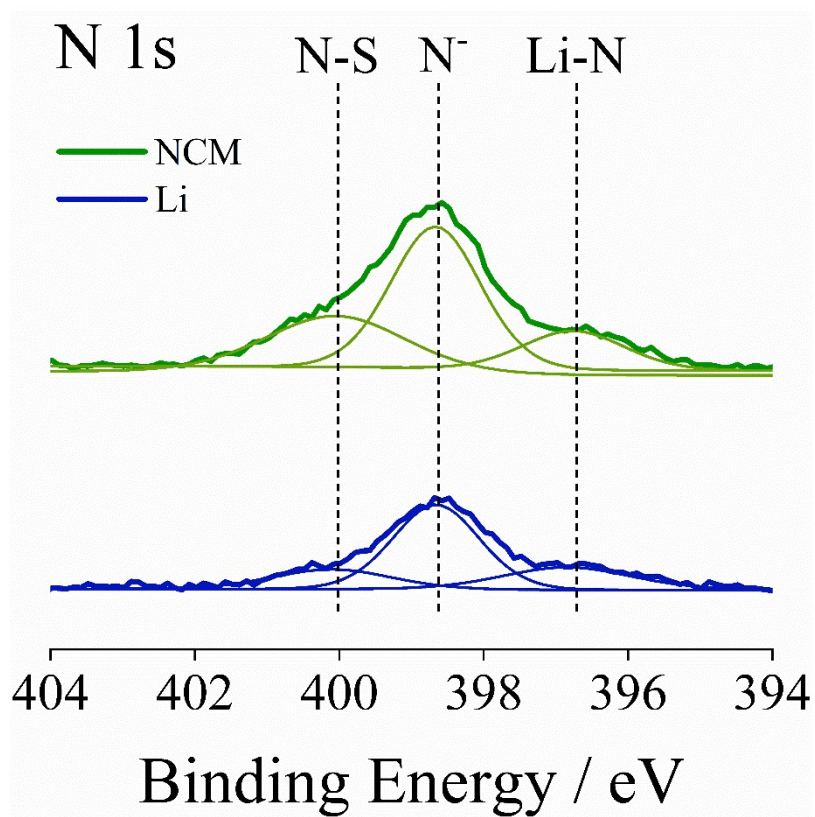


Figure S6. N 1s spectra obtained from the 5th cycled Li metals with Li/NCM811 and Li/Li cells.

XPS references are from the *Sci. Rep.*, 2016, 6, 21771, *Sci. Rep.*, 2017, 7, 15925, *ACS Energy*

Lett., 2019, 4, 411-41

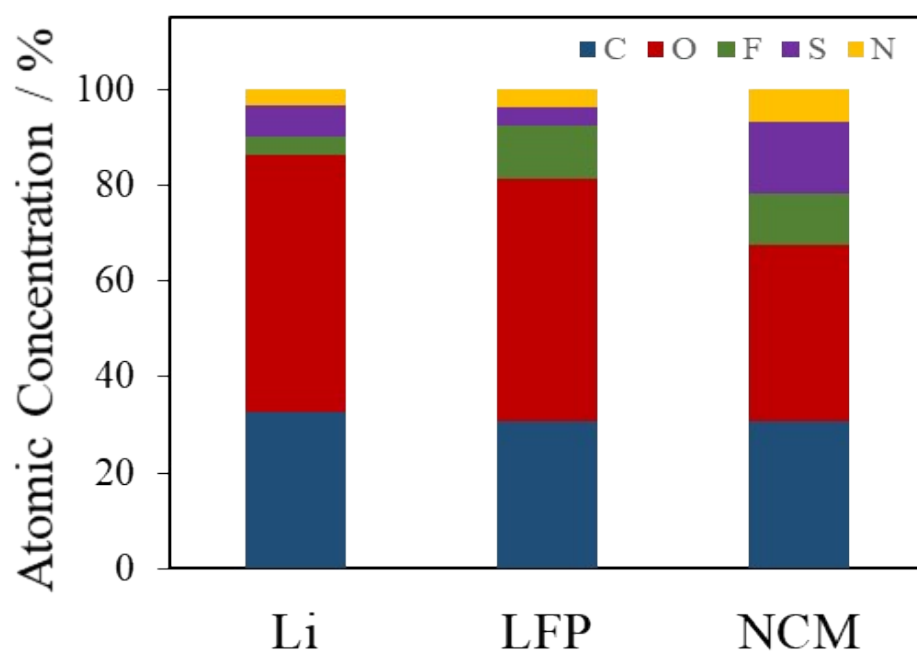


Figure S7. Atomic concentration of the SEI film on the Li metal obtained from 5th cycled Li/Li, Li/LFP and Li/NCM811 cell.

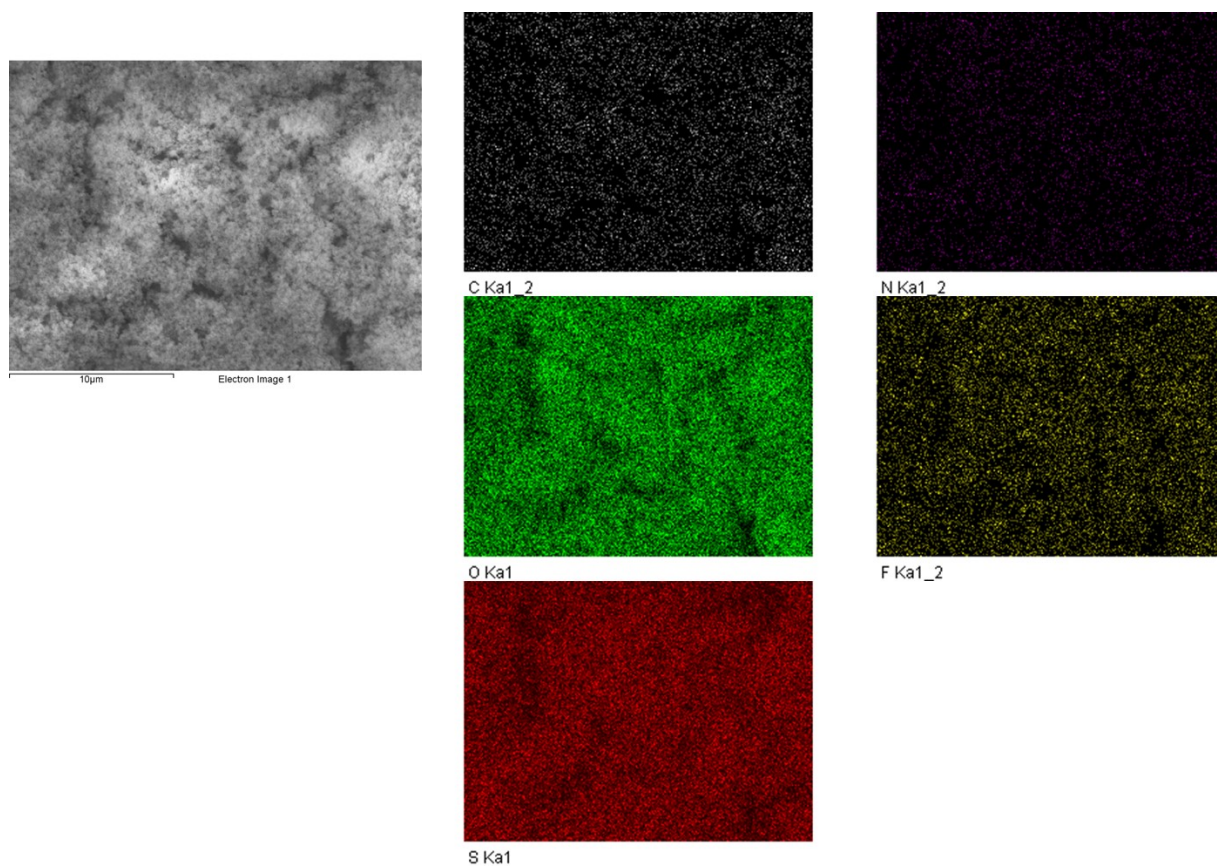


Figure S8. EDS map obtained from the 5th cycled Li metals at Li/Li symmetric cell comprising the stored electrolyte with 4.2 V charged NCM811 electrodes

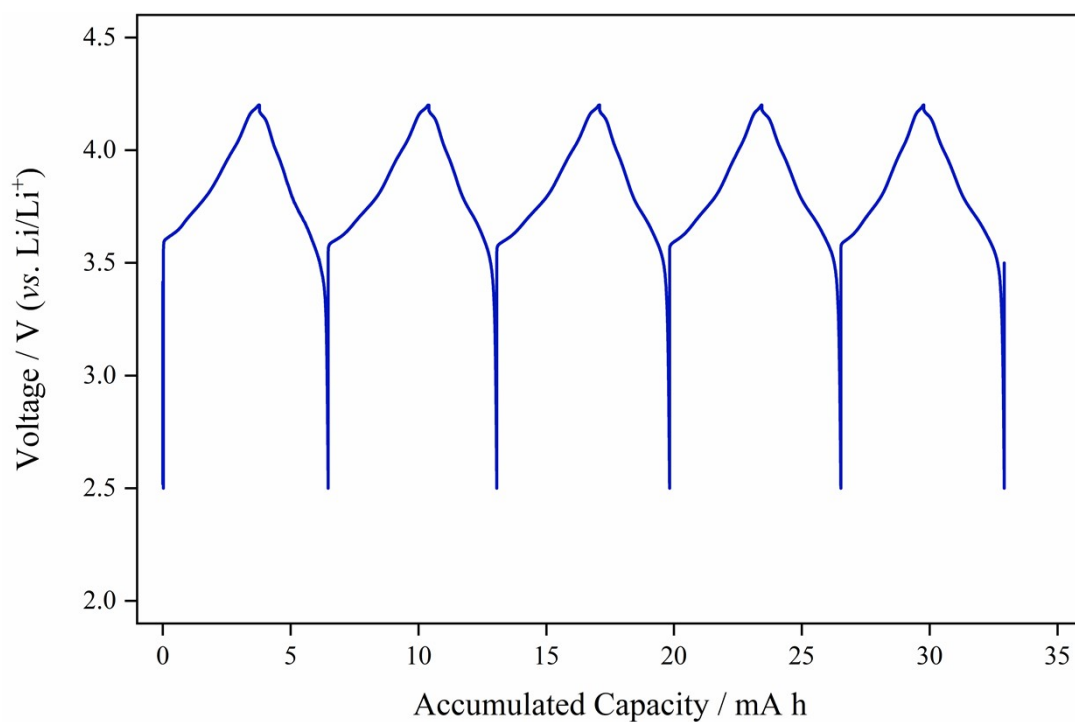


Figure S9. Capacity vs. voltage curves recorded from the reassembled fresh Li and electrolyte comprised Li/pre-cycled NCM811 cell.

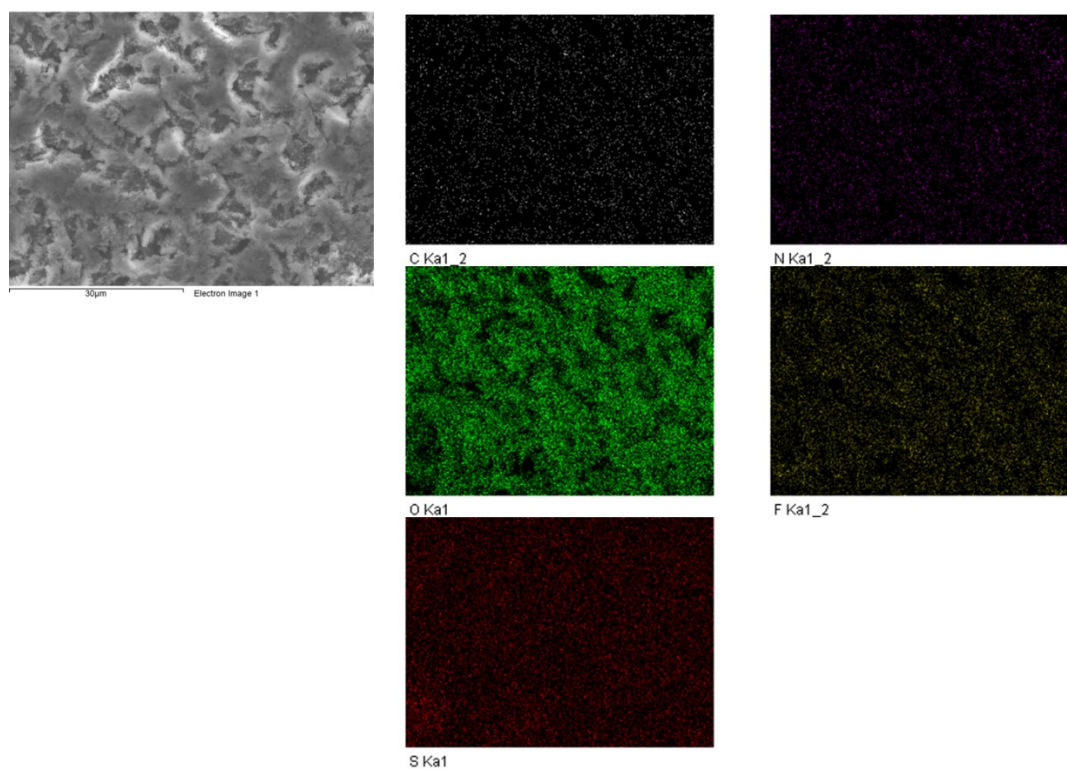


Figure S10. EDS map obtained from the 5th cycled Li metals with Li/pre-cycled NCM811 electrode comprised cell

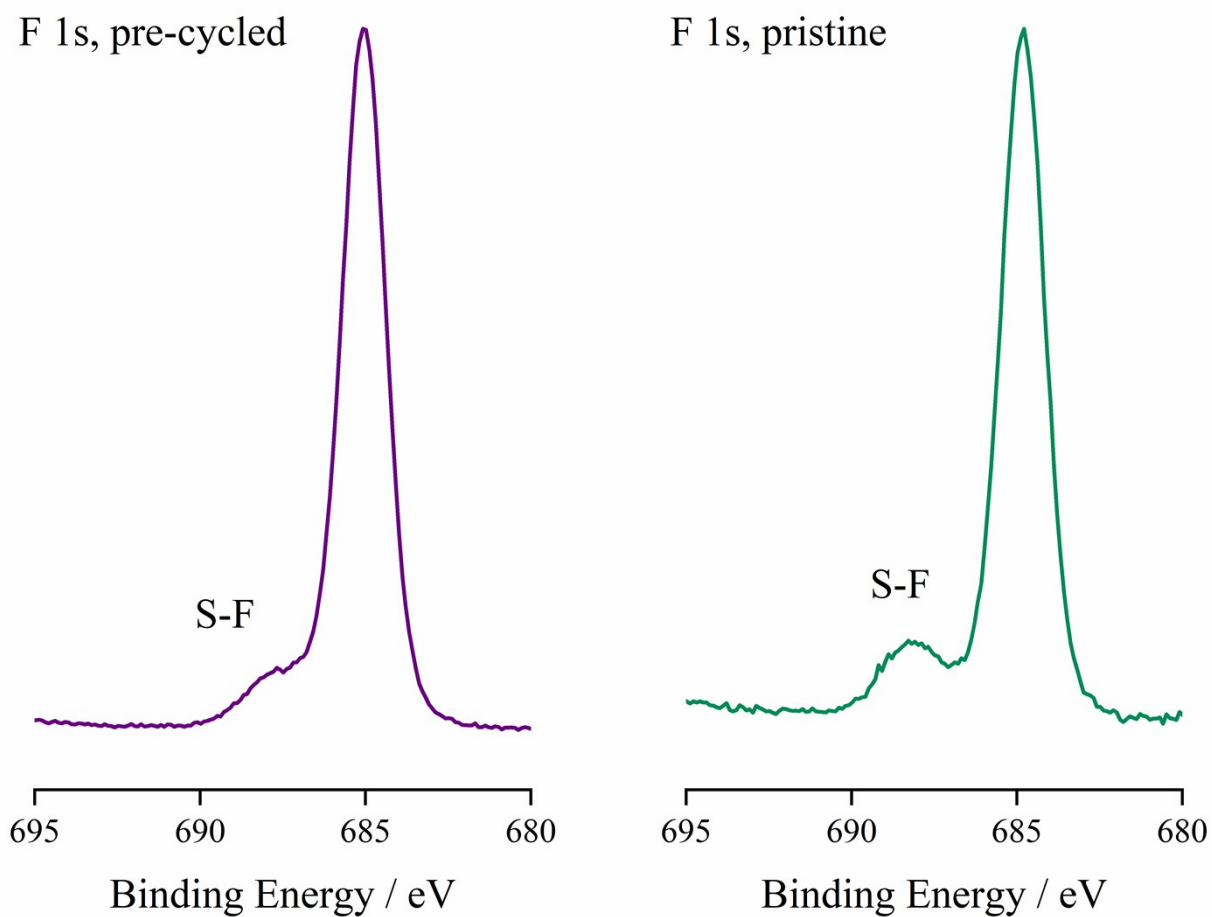


Figure S11. F 1s spectra obtained from the 5th cycled Li metals with Li/pre-cycled NCM811 and Li/NCM811 cells

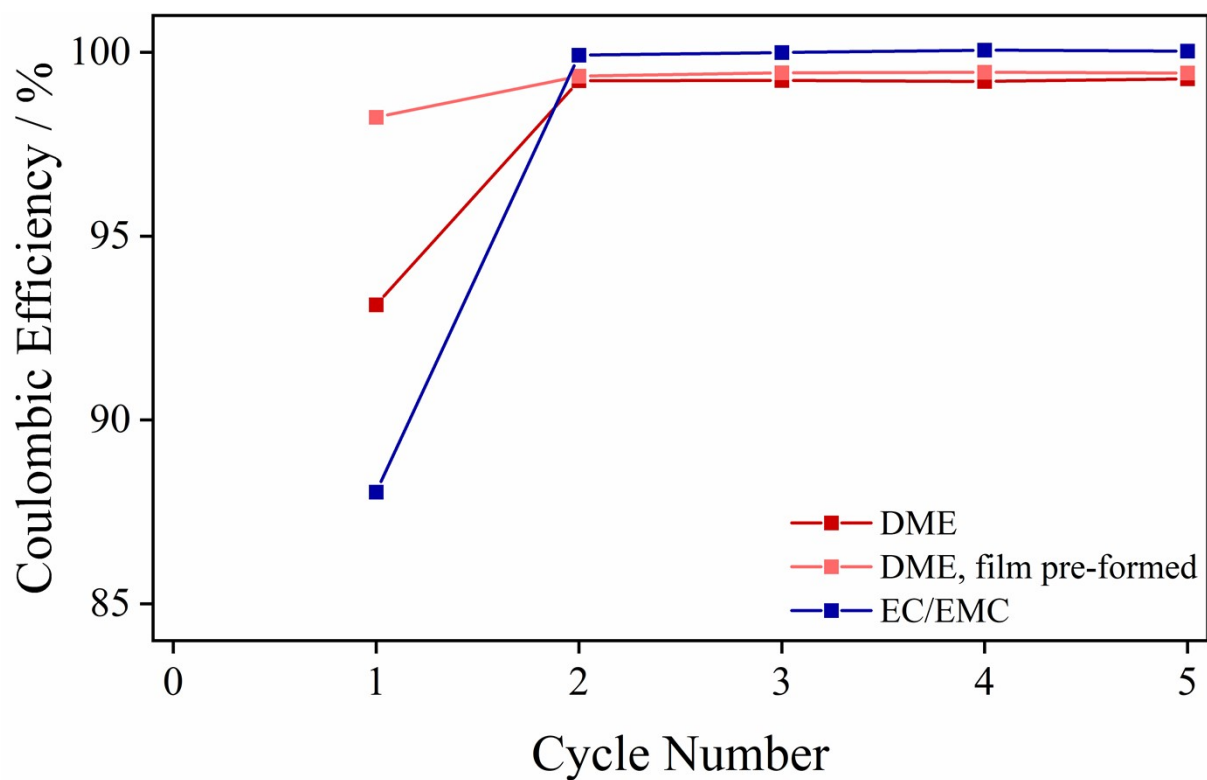


Figure S12. Coulombic efficiency obtained from 2.0 M LiFSI in DME electrolyte with pristine NCM811 and pre-film formed NCM811 electrode and 1.0 M LiPF₆ in EC/EMC (3:7 = v/v) electrolyte injected pristine NCM electrode with Li/NCM cell configuration at 0.1 C with 2.5 to 4.2 V (vs. Li/Li⁺) voltage range

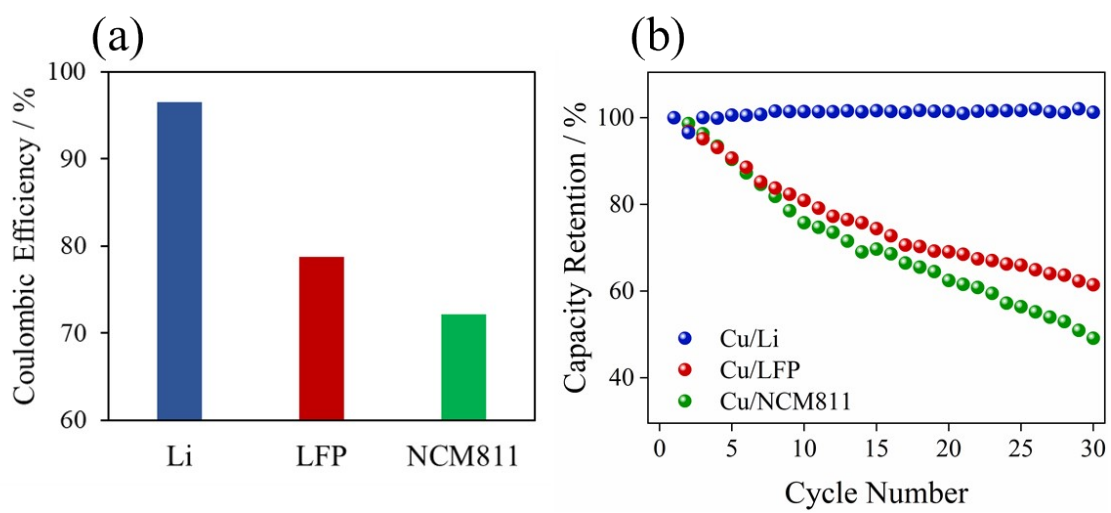


Figure S13. (a) Initial Coulombic efficiency and (b) cycleability obtained from the Cu/Li, Cu/LFP and Cu/NCM811 cells

Table S1. Atomic percent of Ni and Fe from the EDS analysis from the cycled Li from Li/NCM and Li/LFP cells.

	Li from Li/NCM cell / at. %	Li from Li/LFP / at. %
Ni	-0.01	
Fe		0.01