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Supplementary Information for

Bi-Continuous Phase Separation of Lithium Ion Battery Electrodes for Ultra-high Areal Loading

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Other supplementary materials for this manuscript include the following:

Movies S1 to S2

www.pnas.org/cgi/doi/10.1073/pnas.2007250117

Porosity of TIPS electrodes

Measuring the porosity and tortuosity of TIPS electrode is challenging. However, if we use density and neglect swelling of PVDF, the porosities of Type 1 and 3 LFP TIPS electrodes are respectively 40% and 10% larger than conventional LFP electrodes. According to the simulations, the maximum values for both radial and tangential stresses during delithiation for thinner-less porous electrodes (conventional electrode in this study) are higher compared to the thicker-more porous electrodes (TIPS electrode in this study) (44). Therefore, TIPS electrodes might experience less mechanical stresses during cycling. The density values used for this analysis are 1.78, 1.8, and 3.5 g/cc for PVDF, carbon black, and LFP.

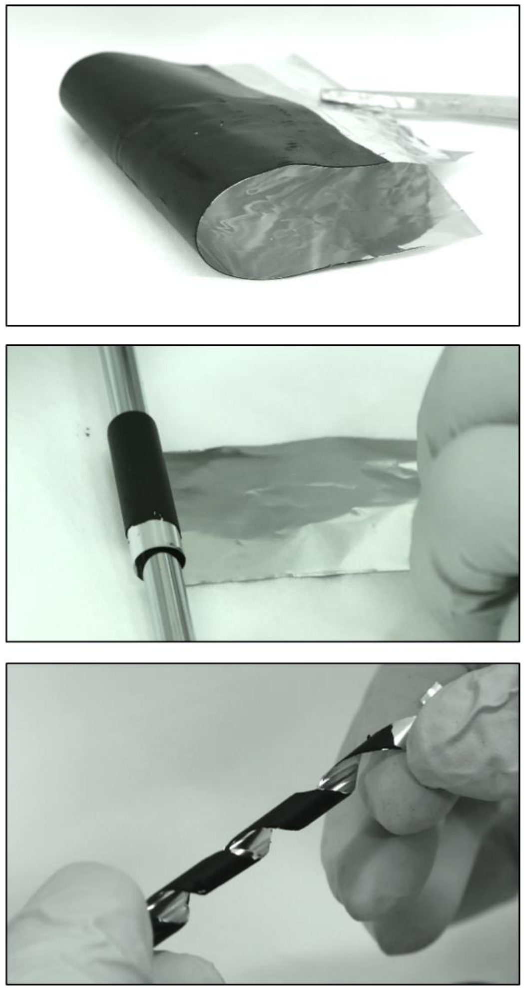

Figure S1. Roll-to-roll coated TIPS LFP electrode on Al current collector under various mechanical deformation. TIPS electrode did not show any sign of delamination confirming the manufacturability in relevant industrial processes.

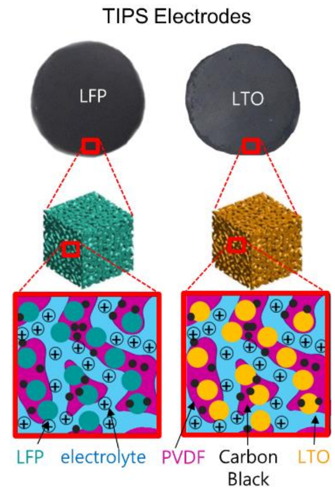

Figure S2. Thermally moulded LFP and LTO discs. The TIPS electrodes are composed of active material, carbon black, and electrolyte.

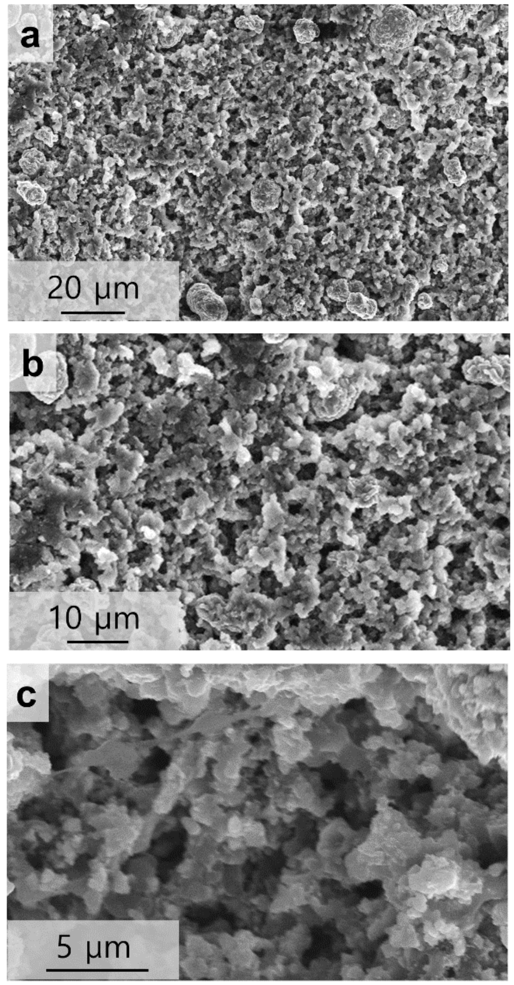

Figure S3. Cross-sectional SEM images of TIPS LFP electrode in different magnifications.

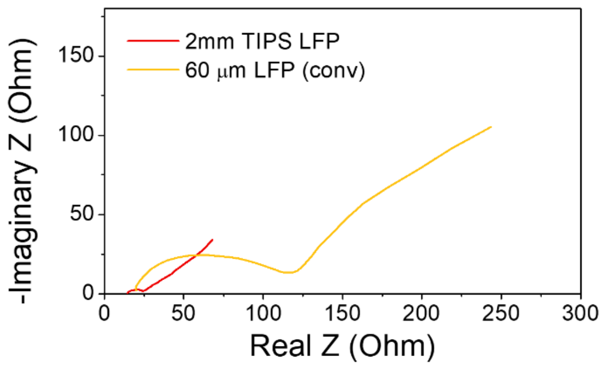

Figure S4. EIS plots of 2mm-thick TIPS LFP electrode and 60 μm-thick conventional electrode.

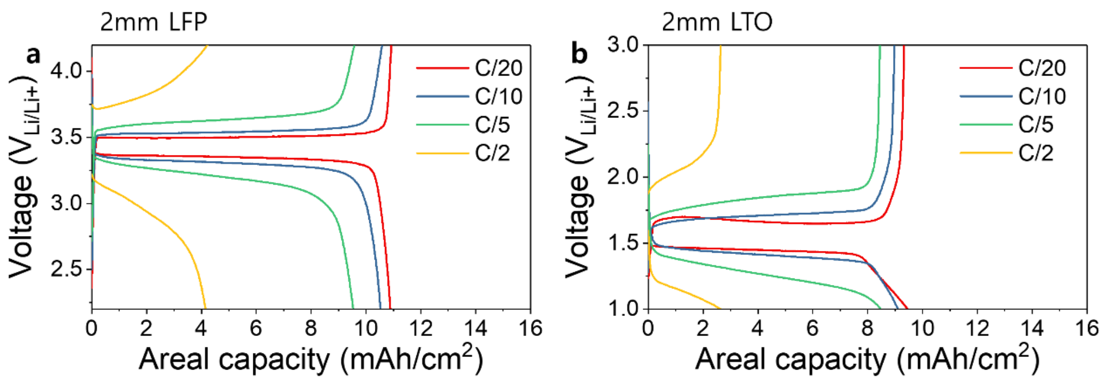

Figure S5. Voltage profiles of 2mm-thick a) LFP and b) LTO electrodes with respect to the areal capacities at different C-rate

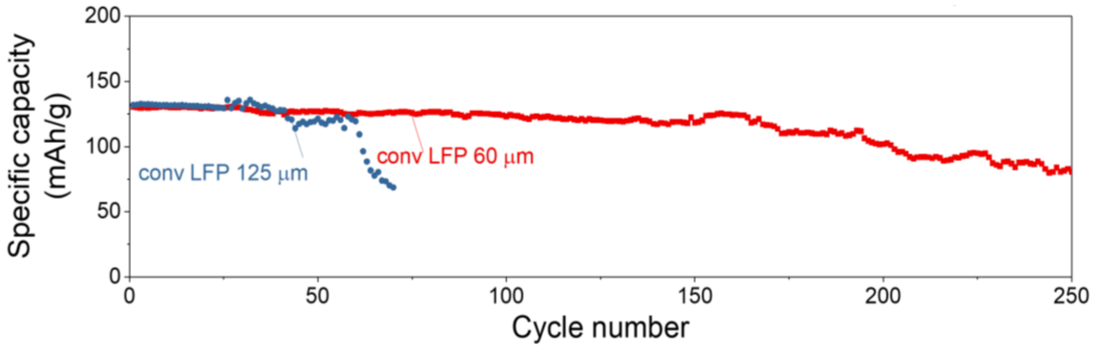

Figure S6. Cycleability of conventional 60 μm-thick and 125 μm-thick LFP electrodes at C/5

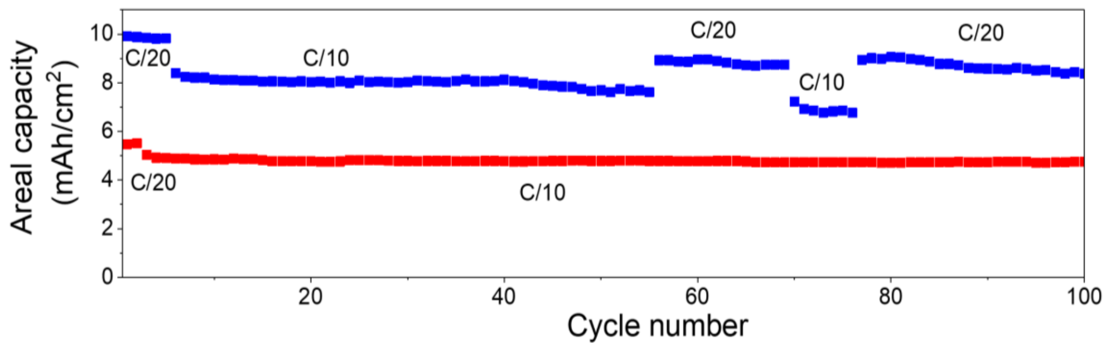

Figure S7. Capacity retention and areal capacity of 1mm LFP- 1mm LTO (red) and 2mm LFP – 2mm LTO (blue) full cells.

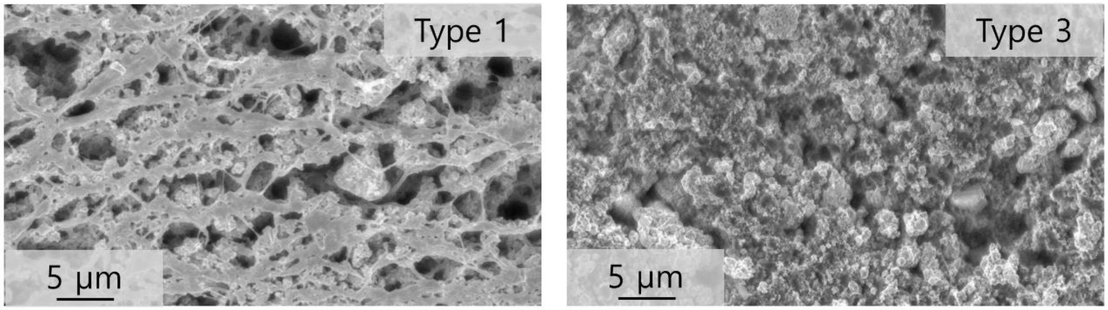

Figure S8. Morphology comparison between a) type 1 and b) type 3 LFP electrodes.

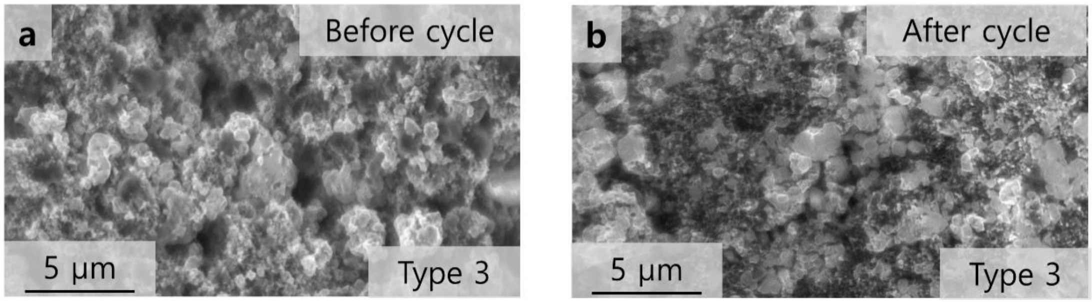

Figure S9. Morphology comparison a) before and b) after 70 charge-discharge cycling.

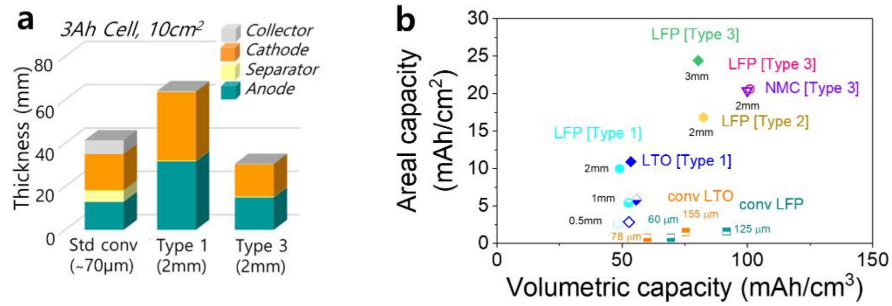

Figure S10. a) 3Ah cell configuration with different 10cm^2 electrodes in terms of thickness and b) areal capacity with respect to the volumetric capacity of TIPS electrodes compared with conventional electrodes. The volume includes 25 μm separator, 15 μm current collector, and double-side coated electrodes.

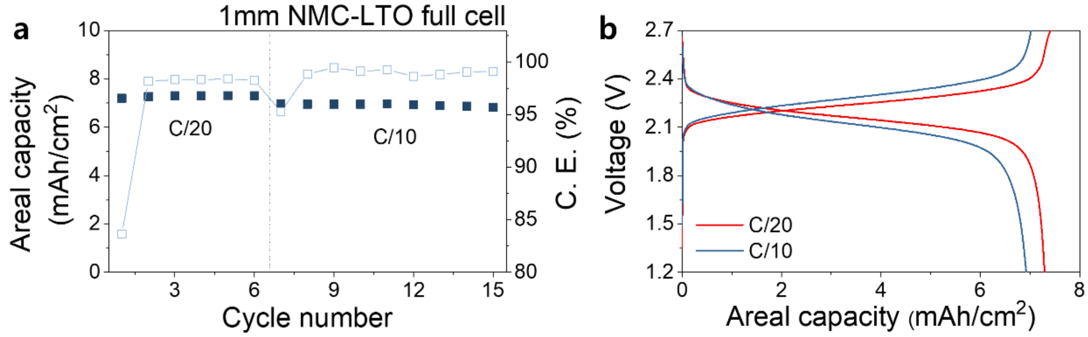

Figure S11. a) Cycleability/Coulombic efficiency and b)Charge-discharge profiles of all TIPS electrodes and electrolyte at C/20 and C/10. The cathode is NMC111 and the anode is LTO.

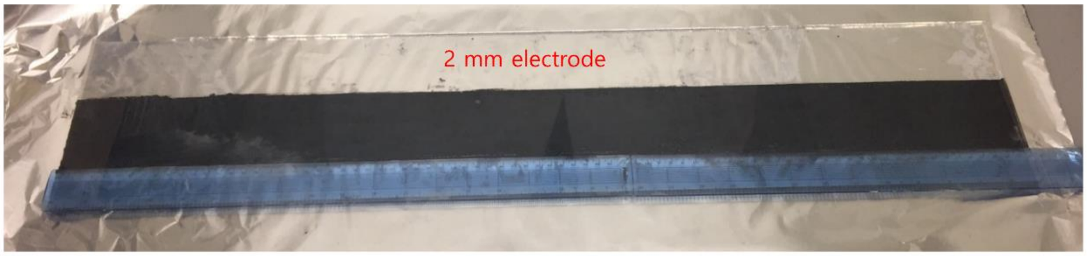

Figure S12. Large scale LFP electrode prepared by thermal moulding. The electrode size is 800 mm x 60 mm x 2 mm.

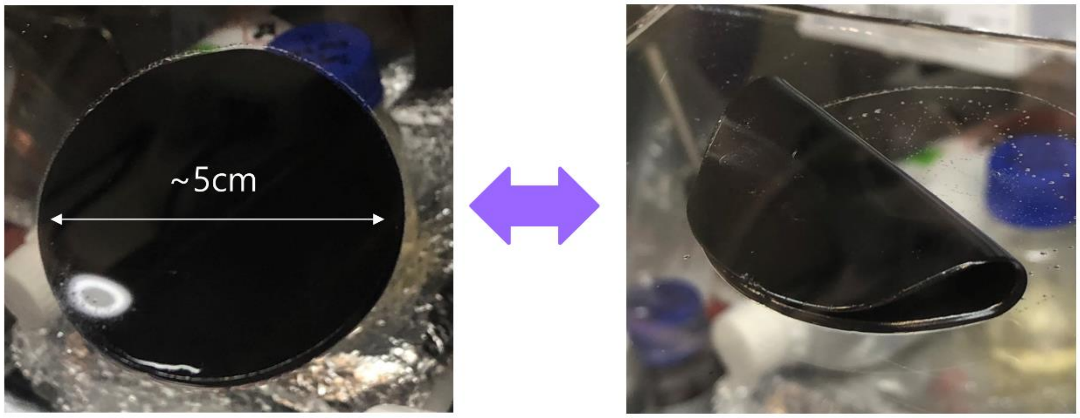

Figure S13. Mechanical integrity of TIPS electrode; the 1mm-thick TIPS NMC111 cathode is mechanically stable under repetitive bending.

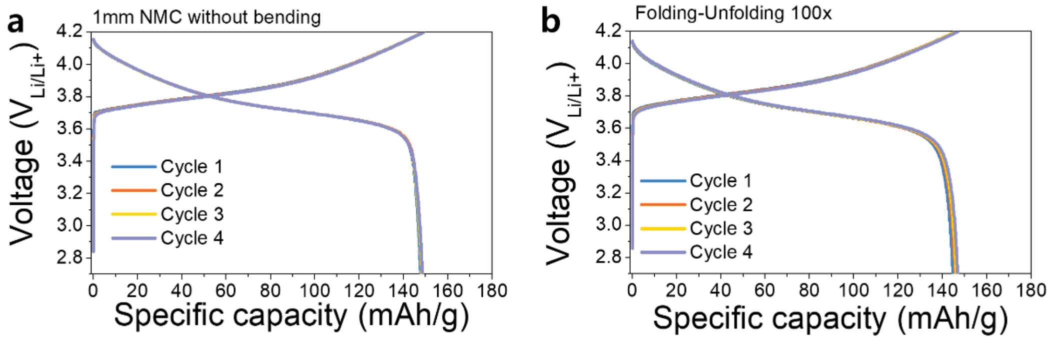

Figure S14. Voltage profiles of 1mm-thick TIPS NMC111; a) without bending and b) with hundred mechanical bending cycles. The bending radius is ~1mm.

SI References

44. Suthar B, Northrop PW, Rife D, & Subramanian VR (2015) Effect of porosity, thickness and tortuosity on capacity fade of anode. Journal of The Electrochemical Society 162(9):A1708-A1717

Movie S1 (separate file). Initial TIPS electrode supply for roll-to-roll process
Movie S2 (separate file). Roll-to-roll process to fabricate long and ultra-thick electrode.