Supporting Information

Highly Elastic Polyrotaxane Binders for Mechanically Stable Lithium Hosts in Lithium-Metal Batteries

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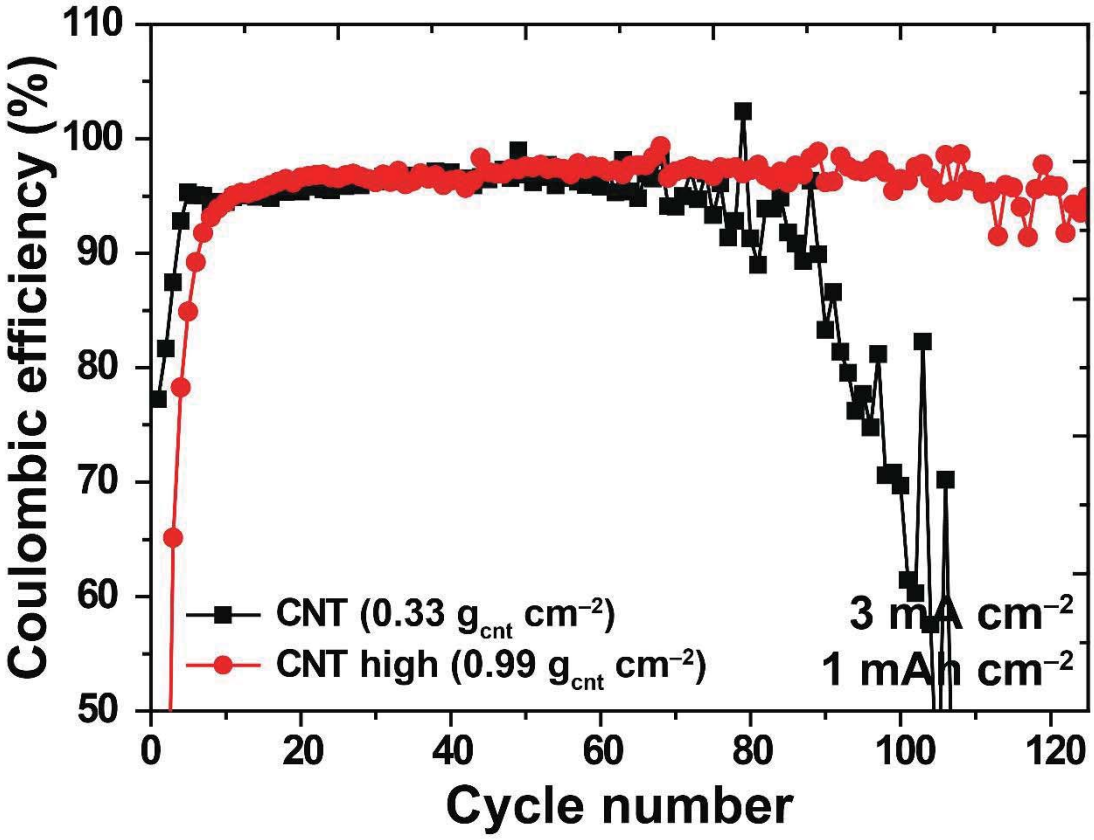

Figure S1. Cycling performance of Li-Cu asymmetric cells with Li metal anodes containing CNT network with a mass loading of 0.33 g_{cnt} cm^{-2} and 0.99 g_{cnt} cm^{-2} when tested with 1 mAh cm^{-2} at 3 mA cm^{-2}. These comparative data show the importance of CNT content in sustaining its network during repeated Li uptake-release cycles.

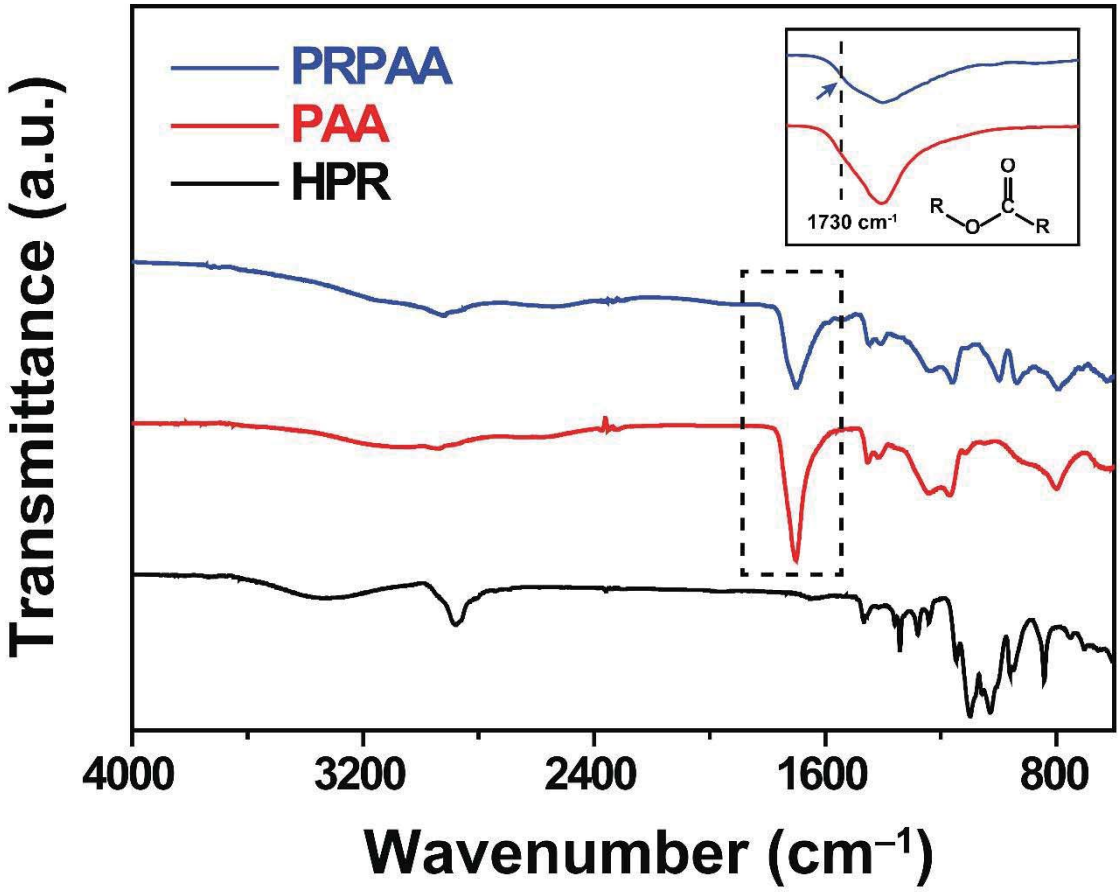

Figure S2. FT-IR spectra of hydroxylpropylated polyrotaxane (HPR), PAA and PRPAA. The peak of PRPAA at 1730 cm^{-1} indicates ester linkage between PAA and HPR.

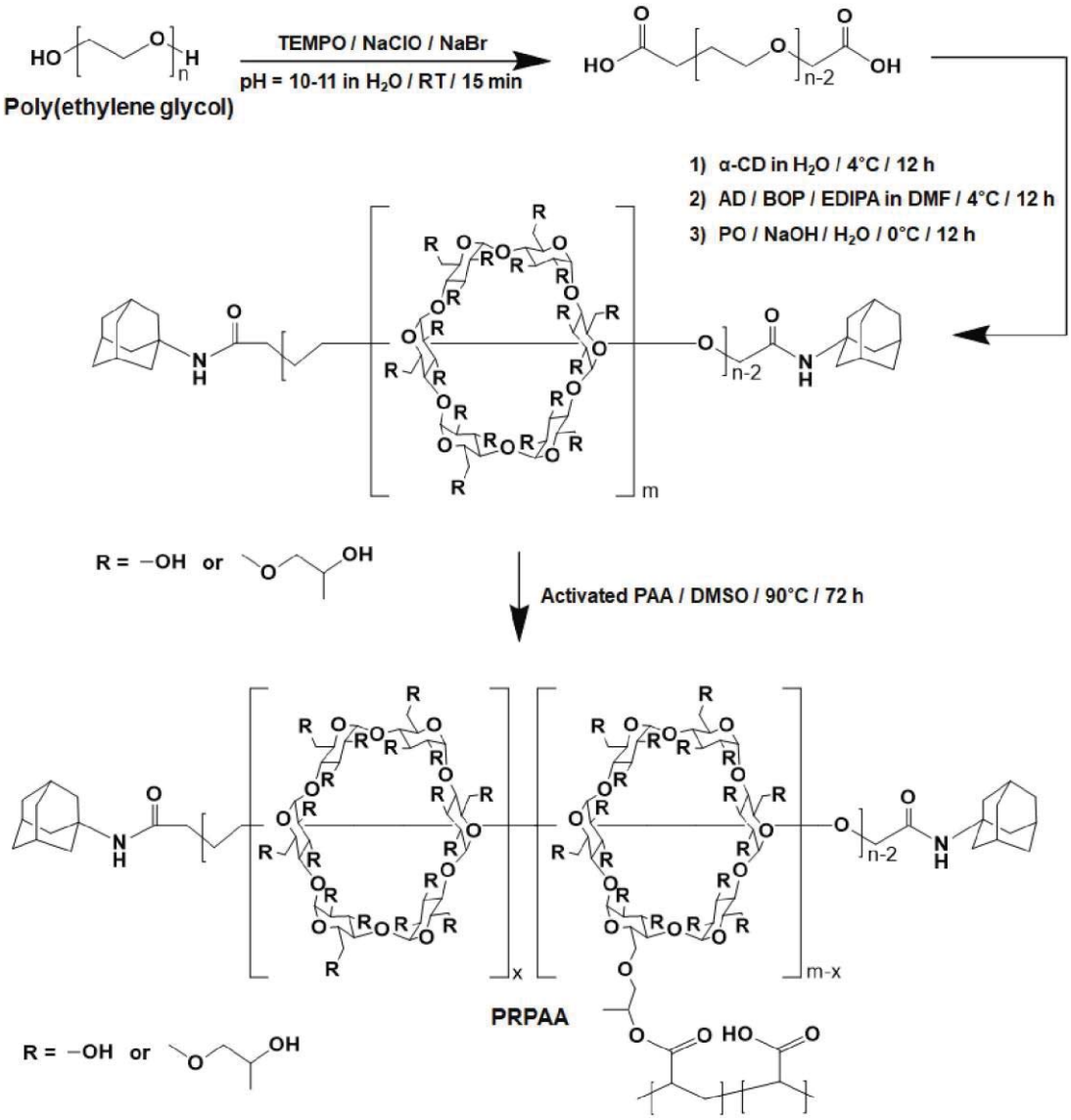

Figure S3. Synthetic scheme of PRPAA.

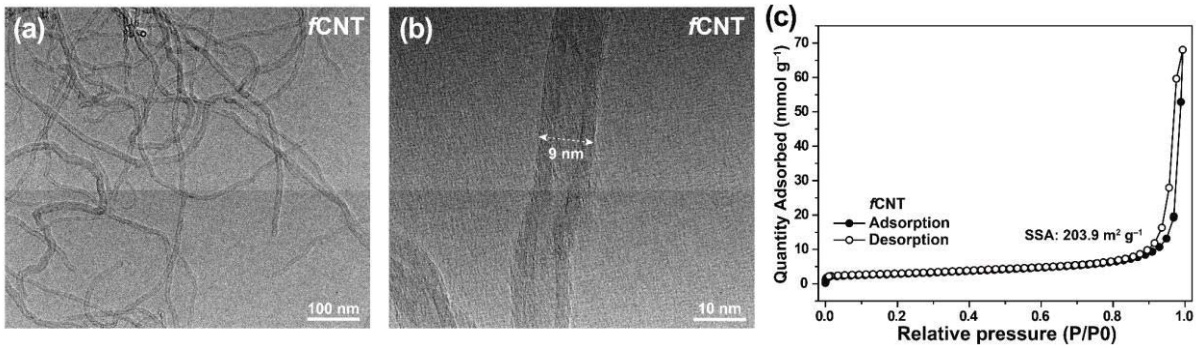

Figure S4. TEM images of *f*CNT at (a) a low magnification and (b) a high magnification. (c) BET isotherm of the same *f*CNT sample.

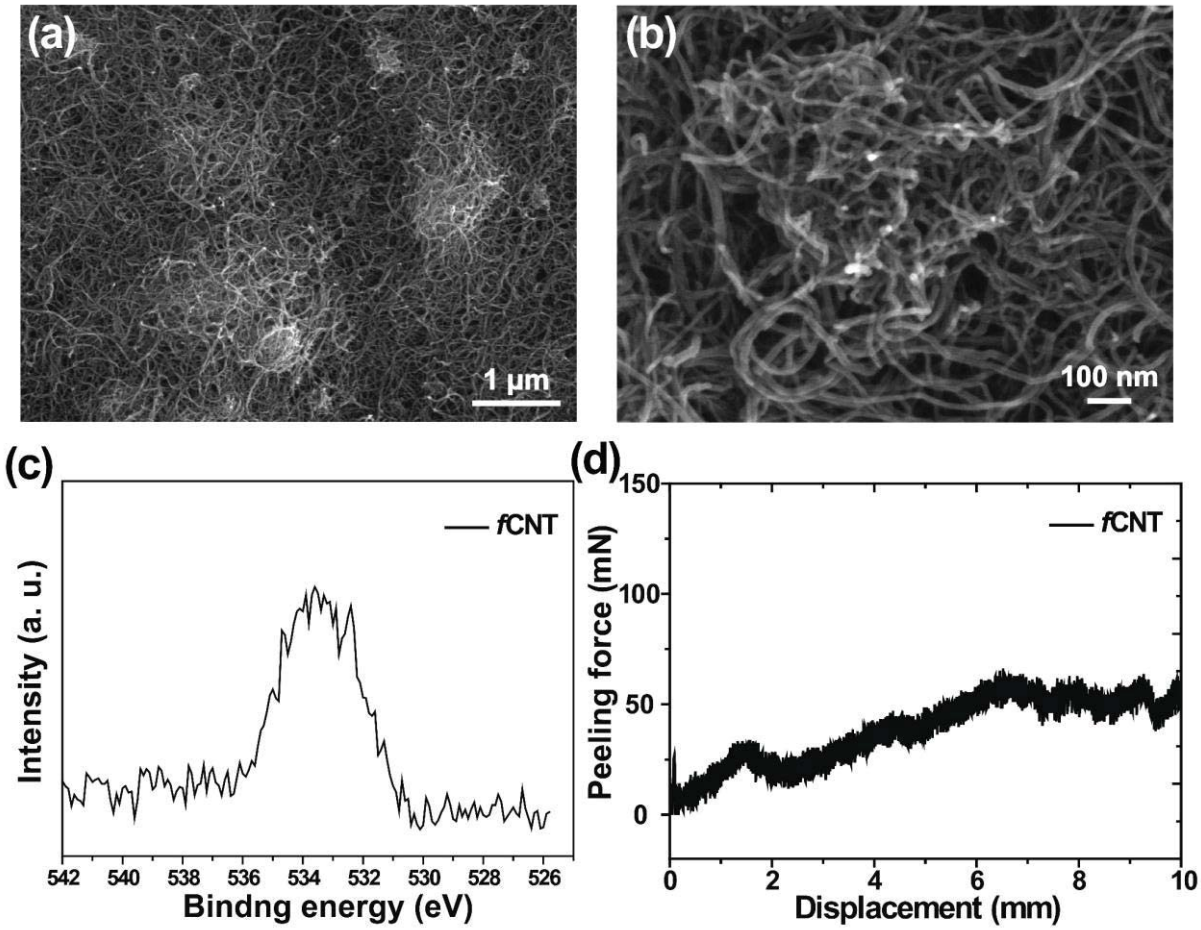

Figure S5. (a) Top-view SEM image of *f*CNT network and (b) its magnified view. (c) XPS spectrum of the *f*CNT network in O 1s branch. (d) Peeling test result of the *f*CNT network without any binder.

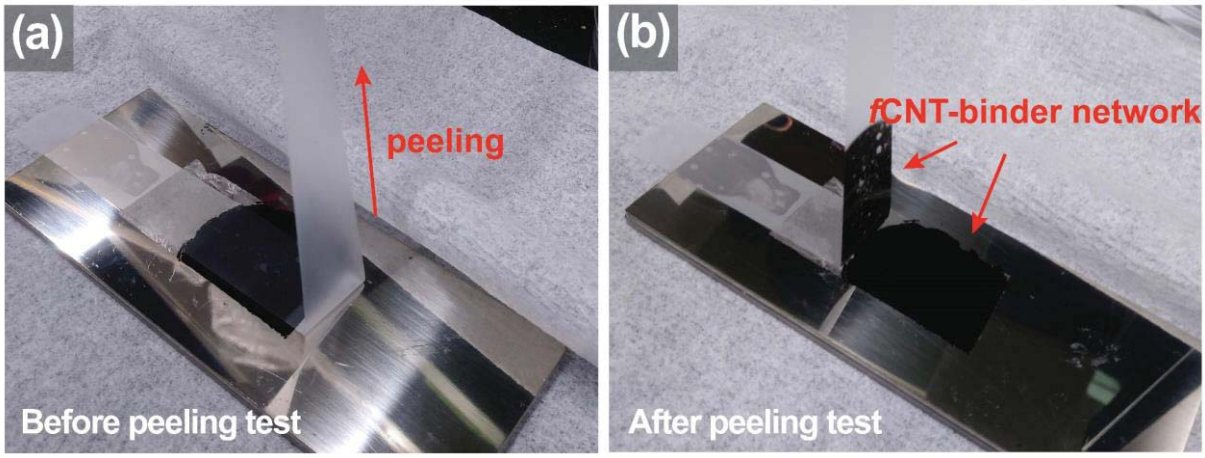

Figure S6. Photographs of *f*CNT-binder network (a) before and (b) after peeling test.

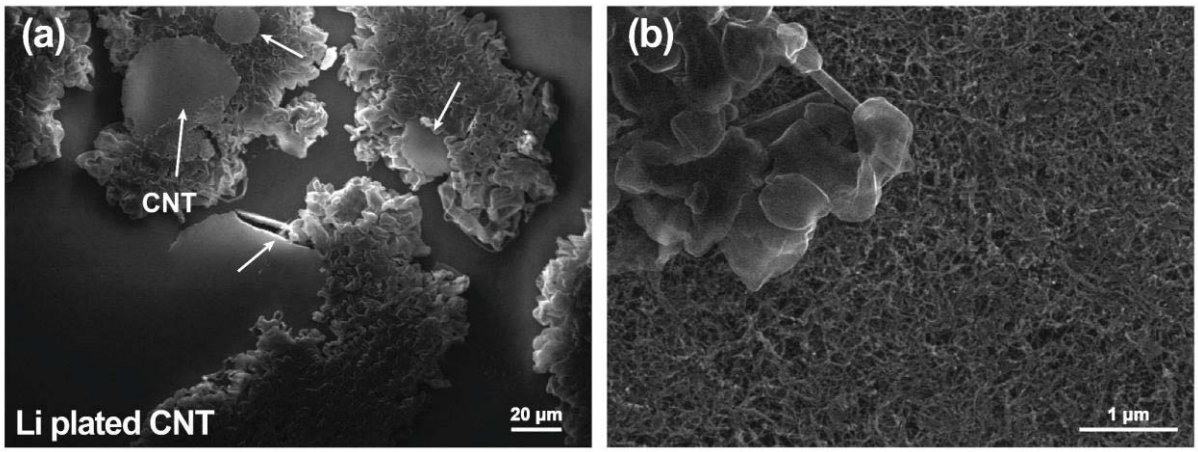

Figure S7. (a) SEM image of Li plated bare CNT network at 1 mA cm^{-2} (plating capacity = 1 mAh cm^{-2}) and (b) its magnified view.

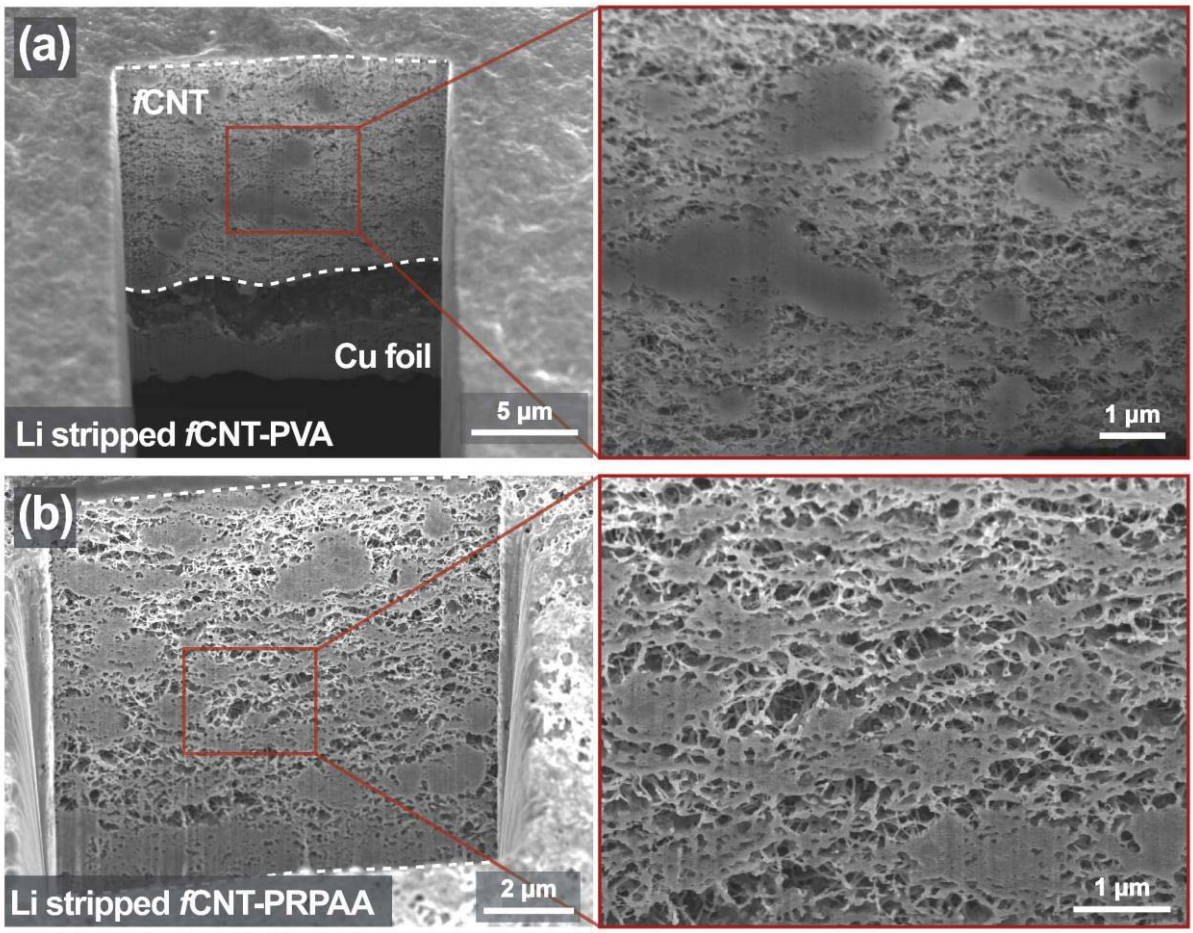

Figure S8. Cross-sectional SEM images of (a) *f*CNT-PVA network and (b) *f*CNT-PRPAA network after Li stripping. Current density = 1 mA cm^{-2}. Areal capacity = 1 mAh cm^{-2}.

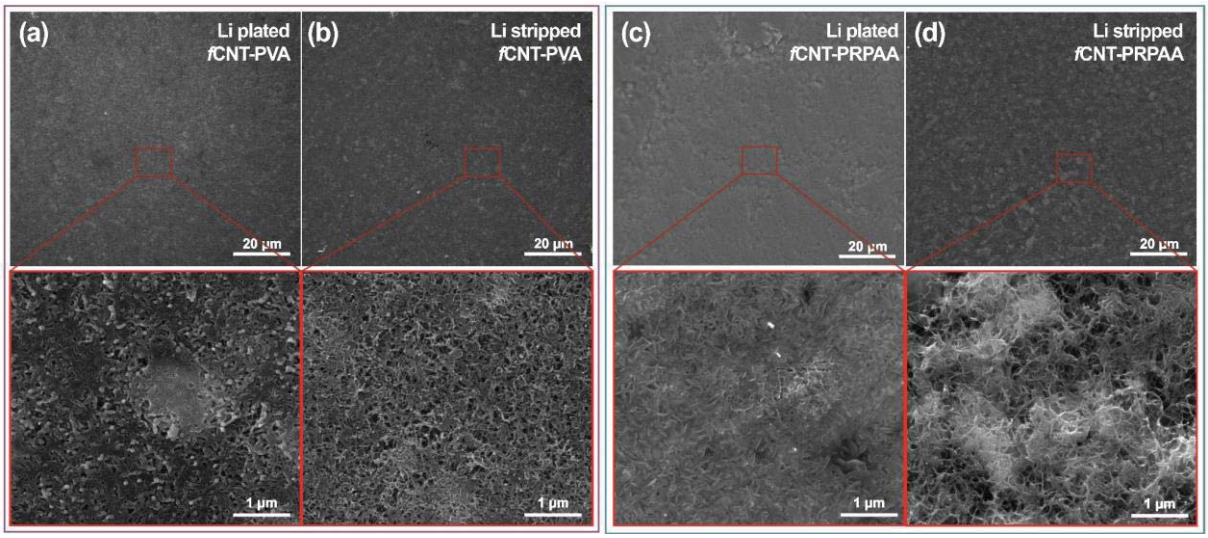

Figure S9. Top-view SEM images: (a) Li plated and (b) stripped *f*CNT-PVA network; (c) Li plated and (d) stripped *f*CNT-PRPAA network.

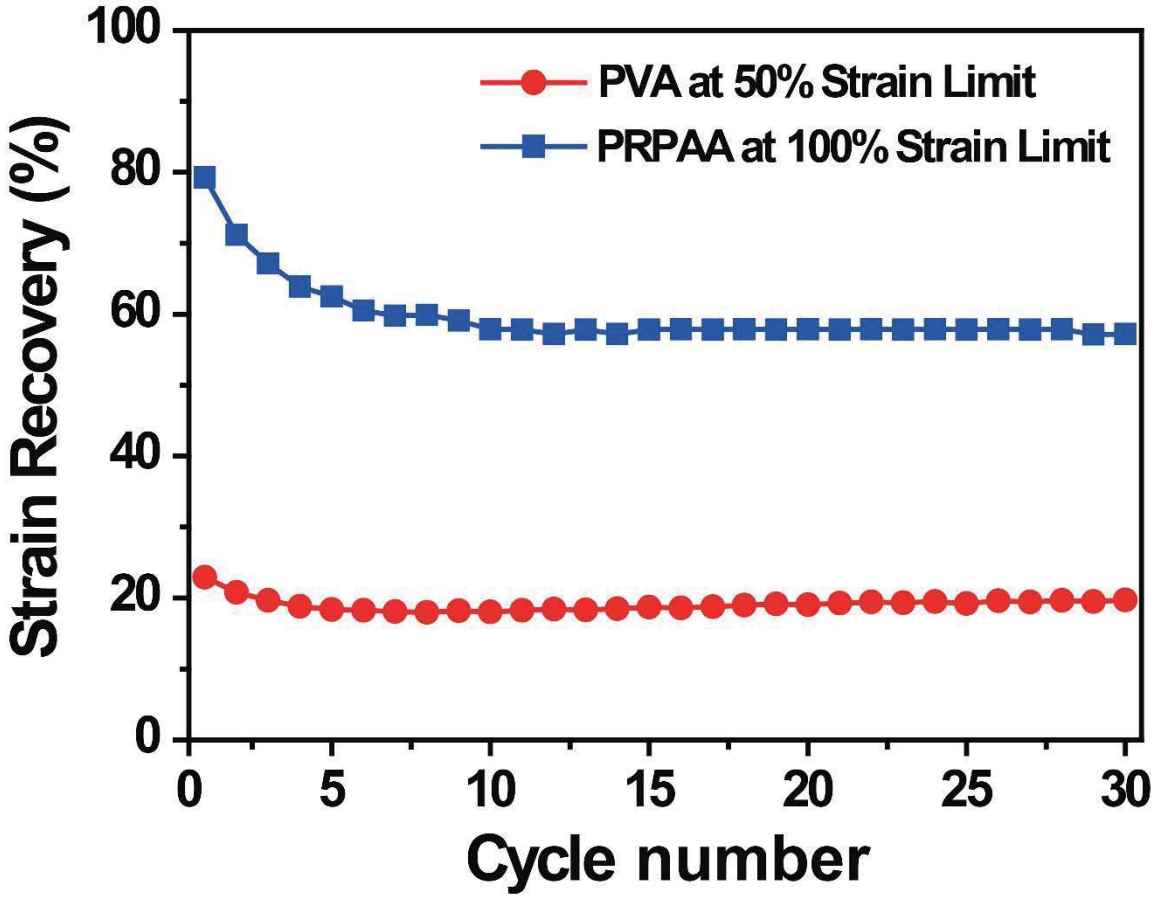

Figure S10. Strain recovery of PVA and PRPAA films over repeated stretch-recovery cycles. The recovery was estimated based on the formula: $((\boldsymbol{L}-\boldsymbol{L_0})/\boldsymbol{L_0}) \times \mathbf{100}$ (L: Length of film after cycling, L_0: Length of pristine film).

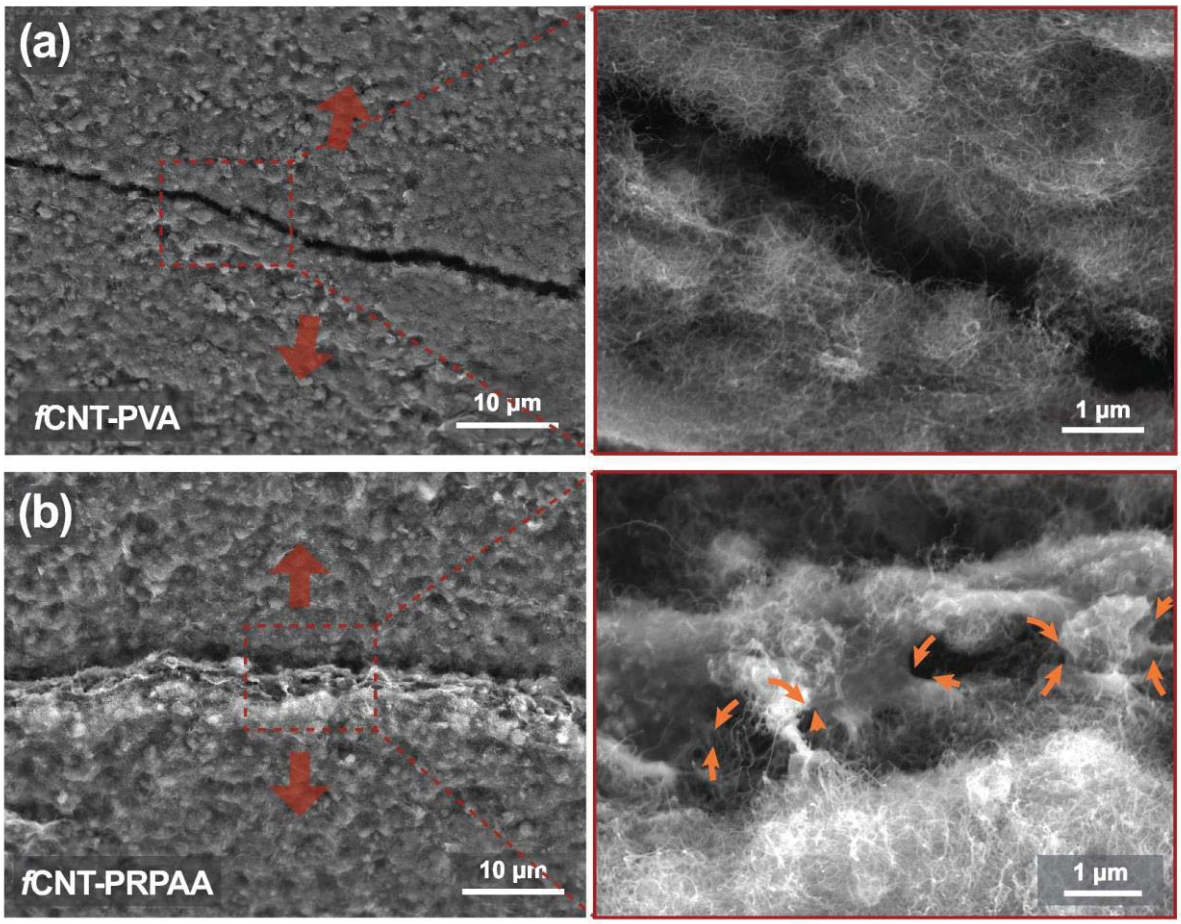

Figure S11. Top view SEM images of (a) *f*CNT-PVA and (b) *f*CNT-PRPAA networks when the CNT composite films were stretched until crack was generated.

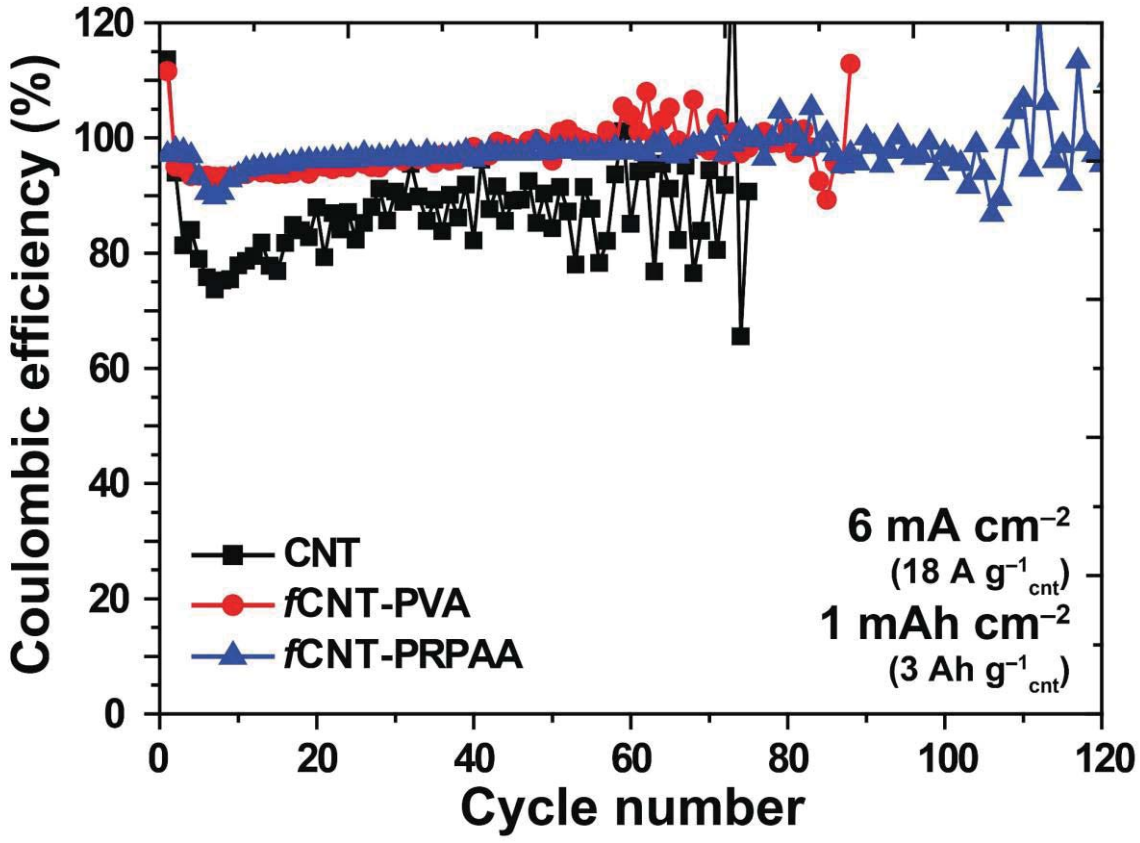

Figure S12. Coulombic efficiencies of Li-Cu asymmetric cells with Li metal anodes containing bare CNT, *f*CNT-PVA, and *f*CNT-PRPAA networks when tested at 6 mA cm^{-2} with 1 mAh cm^{-2}.

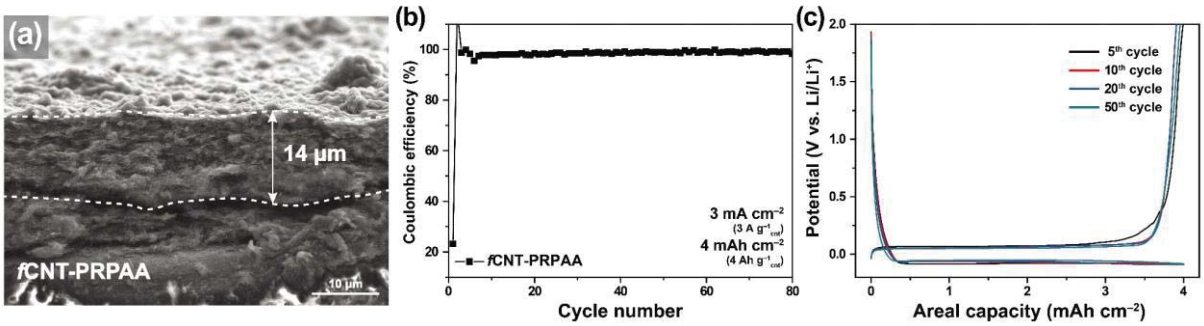

Figure S13. (a) Cross-sectional SEM image of a thick *f*CNT-PRPAA network. (b) Cycling performance under Li-Cu asymmetric cell mode and (c) voltage profiles of the thick *f*CNT-PRPAA electrode. Current density=3 mA cm^{-2}. Areal capacity=4 mAh cm^{-2}.

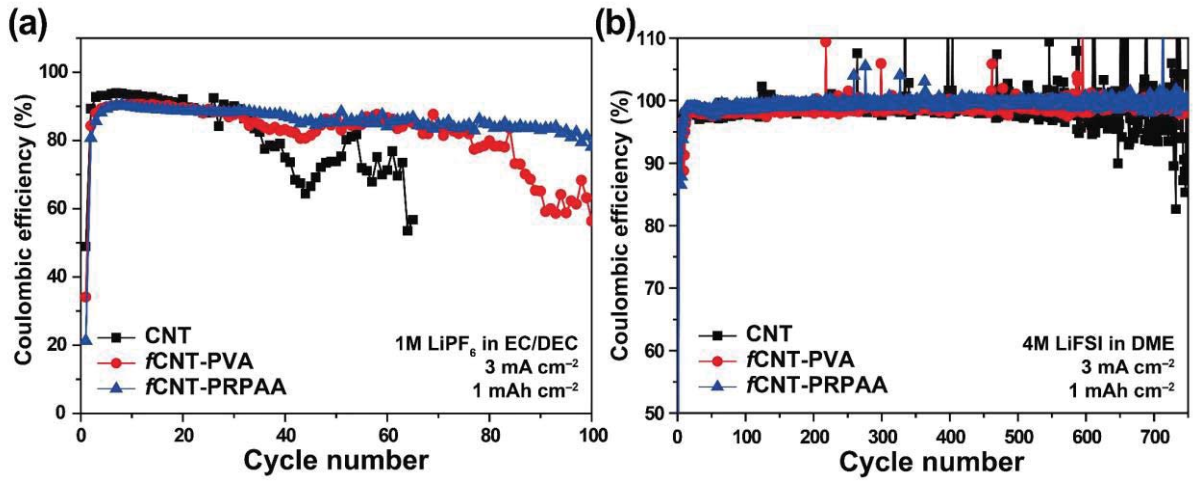

Figure S14. Cycling performance of Li-Cu asymmetric cells with Li metal anodes containing bare CNT network, *f*CNT-PVA network, and *f*CNT-PRPAA network with (a) 1M $LiPF_6$ in EC/DEC and (b) 4M LiFSI in DME when measured at a current density of 3 mA cm^{-2} with a capacity of 1 mAh cm^{-2}.

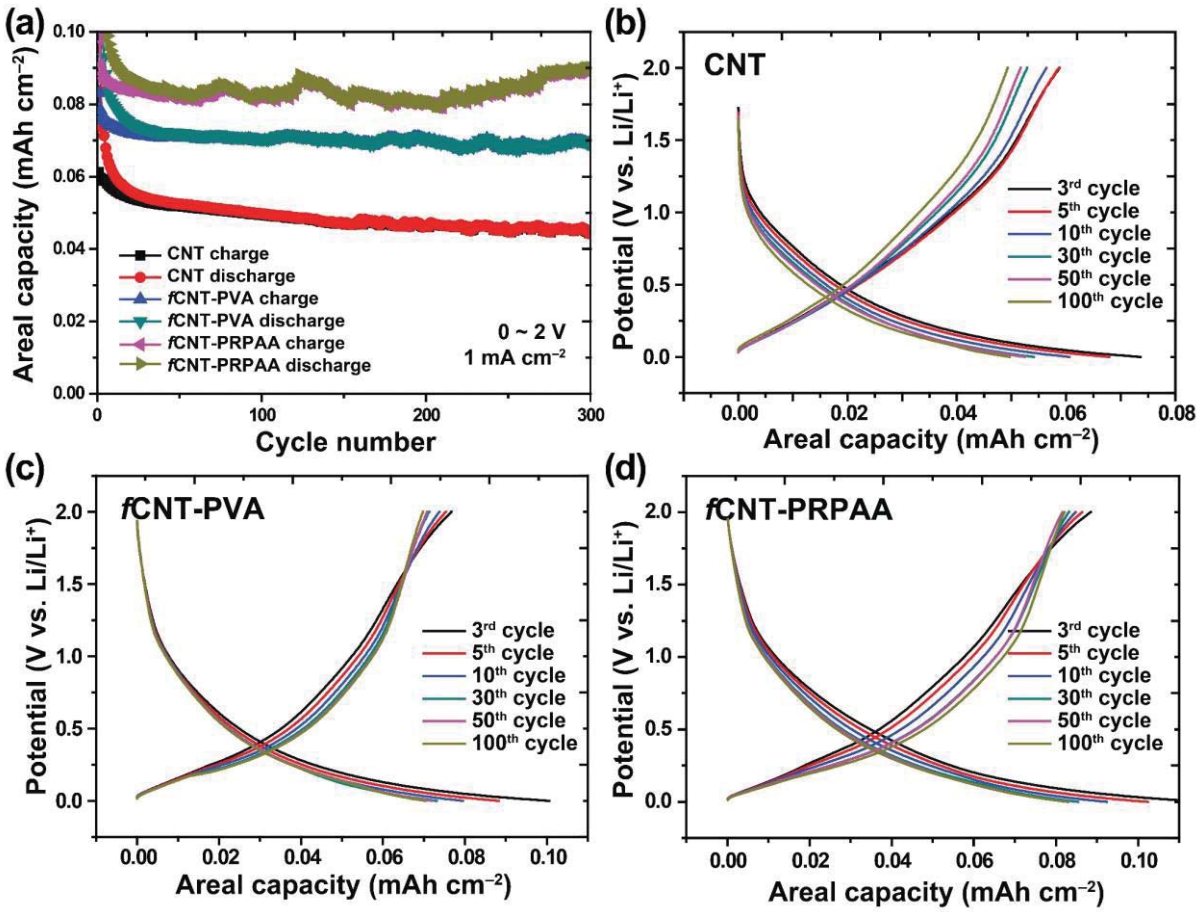

Figure S15. (a) Capacity retentions of bare CNT, *f*CNT-PVA, and *f*CNT-PRPAA networks when tested at 1 mA cm^{-2} in the potential range of 0 – 2 V, and (b-d) their corresponding voltage profiles at different cycles. The lower cut-off voltage of 0 V was to assess the capacitive ion storage exclusively.

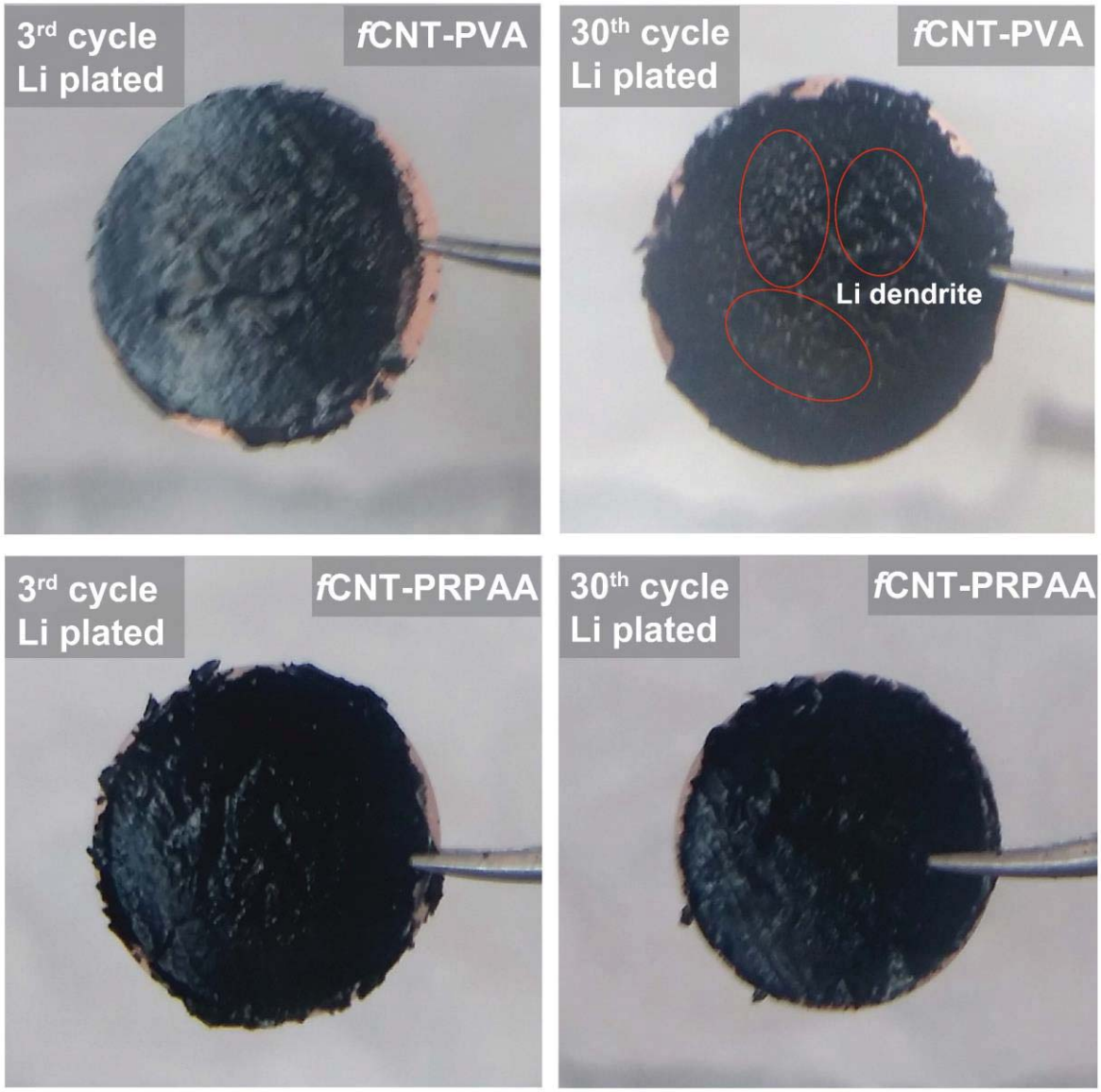

Figure S16. Photographs of Li plated *f*CNT-PVA and *f*CNT-PRPAA networks on the Cu foil after 3 and 30 cycles.

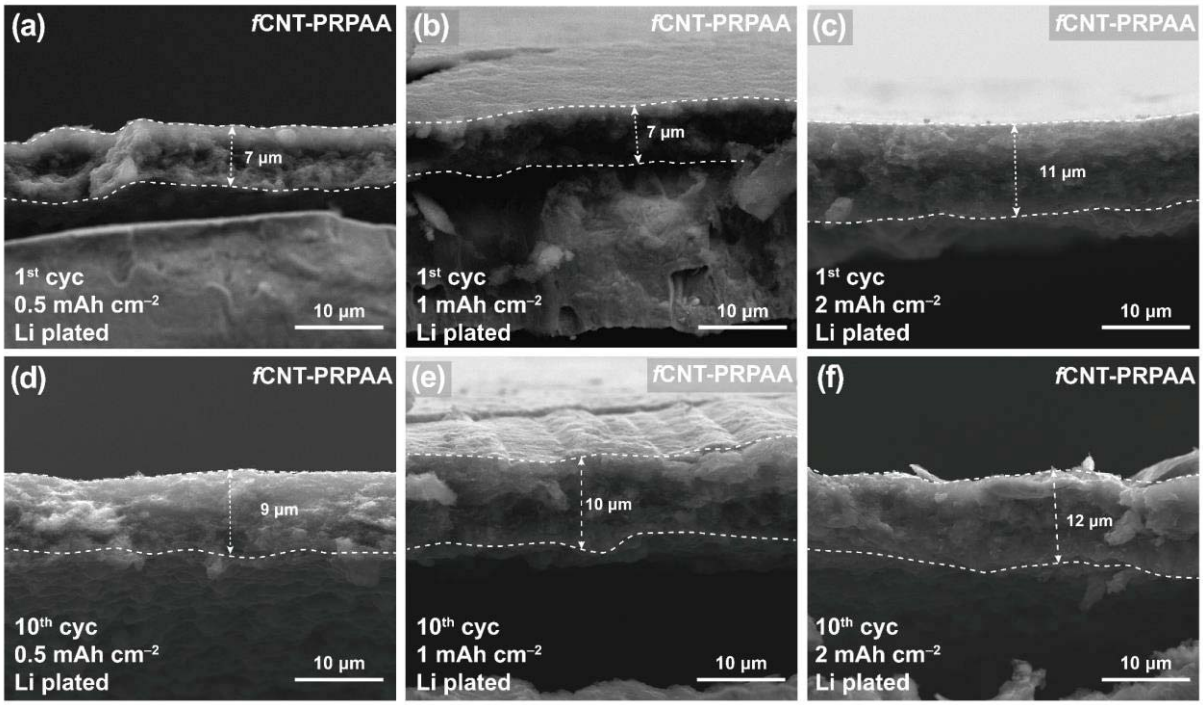

Figure S17. Cross-sectional SEM images of Li plated *f*CNT-PRPAA network after 1 cycle and 10 cycles with an areal capacity of (a, d) 0.5 mAh cm^{-2}, (b, e) 1 mAh cm^{-2}, and (c, f) 2 mAh cm^{-2}.

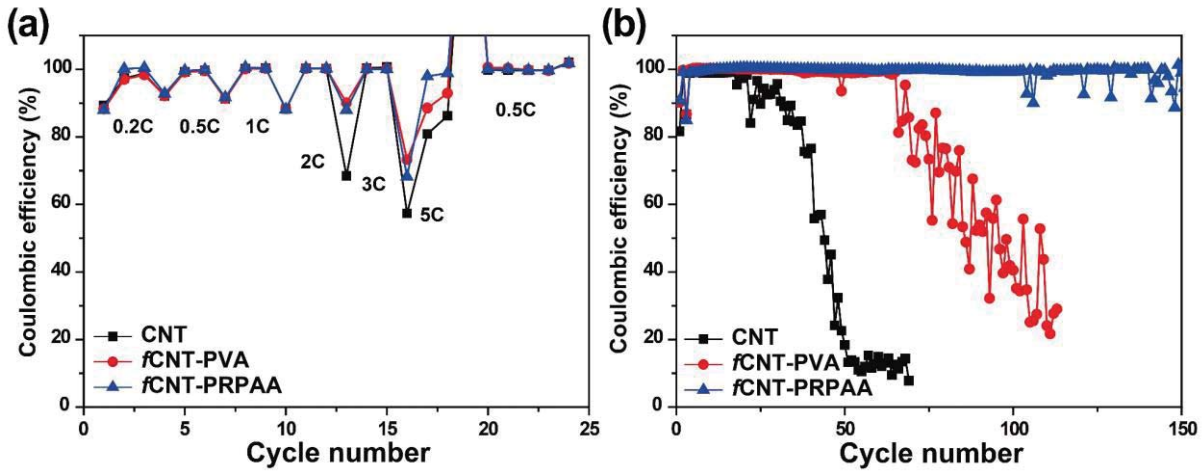

Figure S18. Coulombic efficiencies during (a) rate and (b) cycling performance tests for LFP-Li full cells in Figure 6.

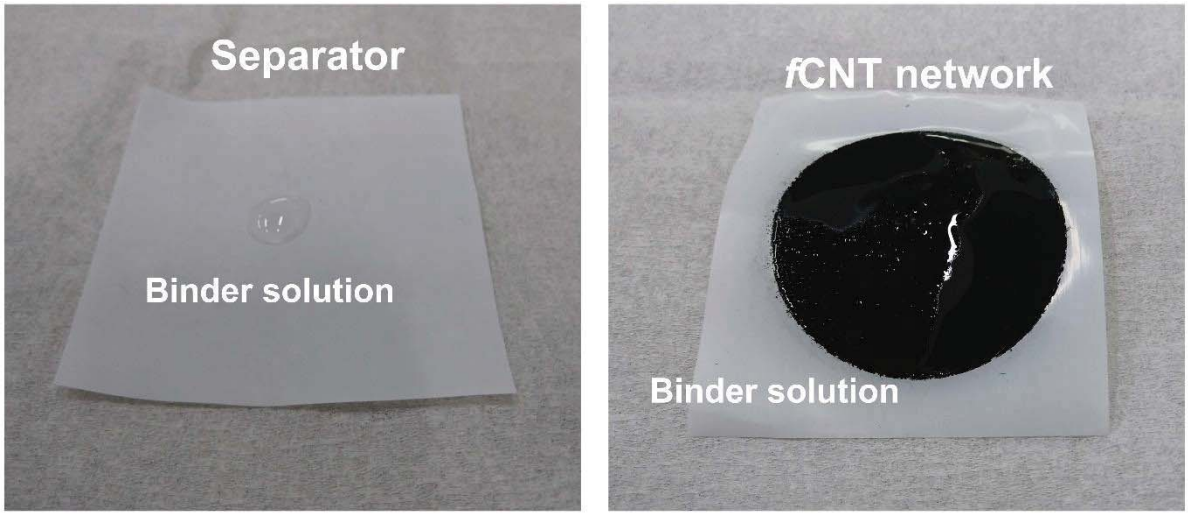

Figure S19. Photographs of wettability of binder solution with the separator and *f*CNT network.

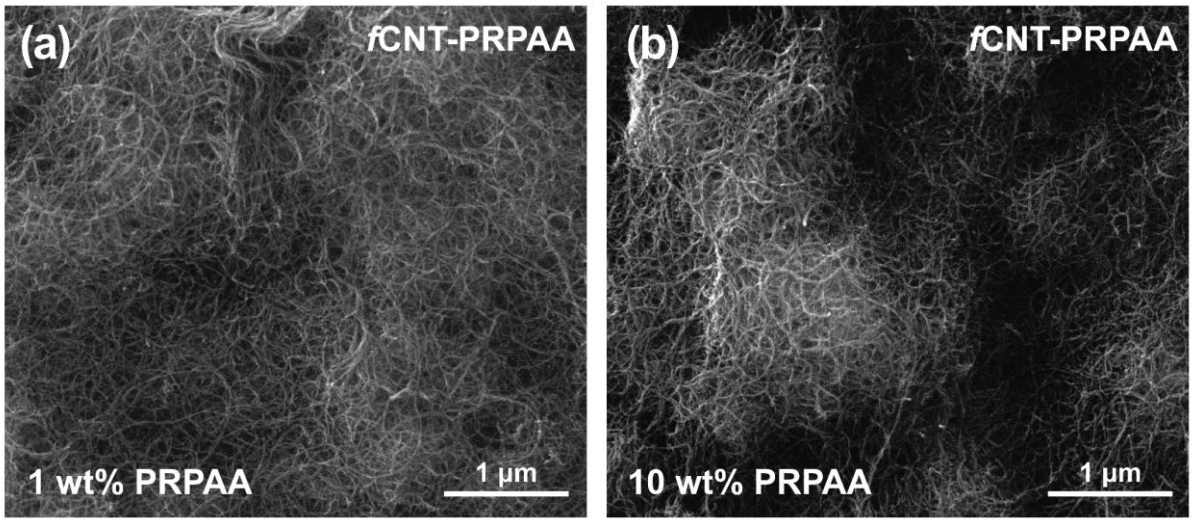

Figure S20. SEM images of *f*CNT-PRPAA networks with (a) 1 wt% of PRPAA and (b) 10 wt% of PRPAA.