Supporting Information

Self-assembled Monolayer Enables Slurry-coating of Li Anode

Tuo Kang,[†] Yalong Wang,[‡] Feng Guo,[‖] Chenghao Liu,[‡] Jianghui Zhao,[‖] Jin Yang,[‖] Hongzhen Lin,[‖] Yejun Qiu,[†*] Yanbin Shen,[‖*] Wei Lu,[‖] and Liwei Chen[‖*]

*To whom Correspondence should be addressed:

lwchen2008@sinano.ac.cn (L.W.C.);

yejunqiu@hit.edu.cn (Y.J.Q)

ybshen2017@sinano.ac.cn (Y.B.S)

†Shenzhen Engineering Lab of Flexible Transparent Conductive Films, Department of Materials Science and Engineering, Harbin Institute of Technology, Shenzhen 518055, China.

‡China Energy Lithium Co., No. 100, the 9th Avenue of Xinye, West TEDA, Tianjin, China

‖*i*-Lab, CAS Center for Excellence in Nanoscience, Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences, Suzhou 215123, China.

Calculation of Coulombic efficiency (CE): The method used to measure CE of the lithium metal anode follows our previous papers.[1,2] The Li-CNT composite anode was paired with a highly reversible cathode, in this case a commercial $LiFePO_4$ cathode, for the estimation of CE. If the irreversible capacity loss of the LFP electrode during the cycling is ignored, and assume that all the capacity from the lithium anode have been consumed at the plunge point in the cycling curve, then the CE of the anode can be estimated from the initial capacity and the cycle number, as shown in the following equation:

CE=(Ccathode-Ctotal/n)/ Ccathode

Where Ctotal is the total capacity of the cathode and anode, Ccathode is the cathode capacity, n is the cycle number at which the cycling curve starts to plunge.

For example, as shown in Figure 4e, the initial cell capacity of 3.75 mAh/cm^2 (anode: 2.5 mAh/cm^2, cathode: 1.25 mAh/cm^2), and the capacity retention curve of the OPA-Li-CNT||LFP cell cycled at 1C starts to plunge at around 250 cycles, so the CE of OPA-Li-CNT can be estimated to be 98.8%.

$$CE = \frac{1.25 - \frac{3.75}{250}}{1.25} = 98.8\%$$

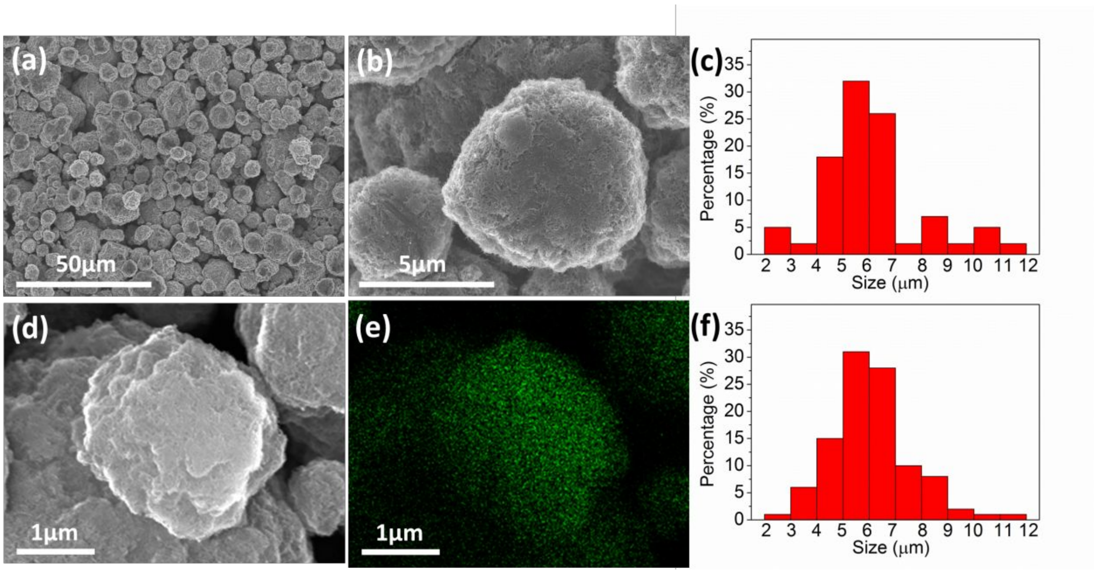

Figure S1. (a, b) Scanning electron micrograph (SEM) and (c) histogram of diameter distribution of the Li-CNT particles; (d) SEM, (e) phosphorus elemental mapping, and (f) histogram of diameter distribution of the OPA-Li-CNT.

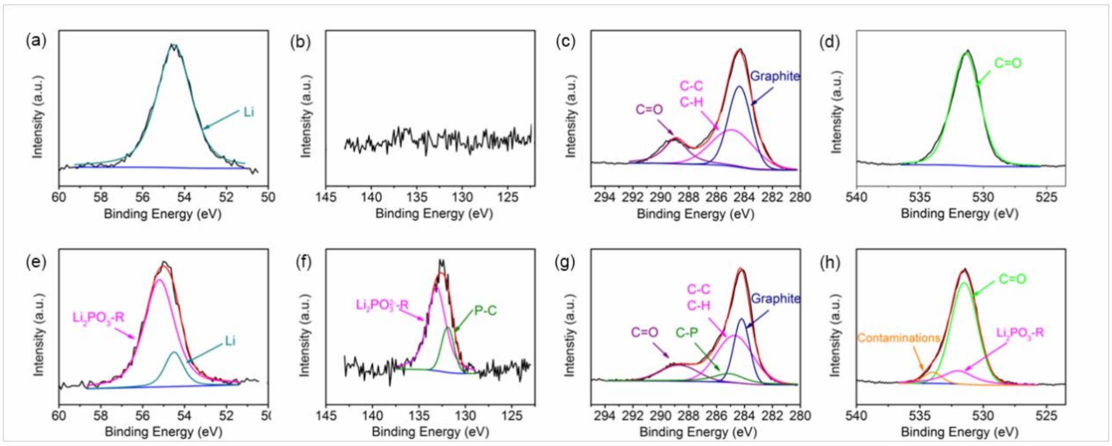

Figure S2. XPS spectra of Li-CNT: (a) Li 1s spectrum, (b) P 2p spectrum, (c) C 1s spectrum, and (d) O 1s spectrum; and XPS spectra of OPA-Li-CNT: (e) Li 1s spectrum, (f) P 2p spectrum, (g) C 1s spectrum, and (h) O 1s spectrum.

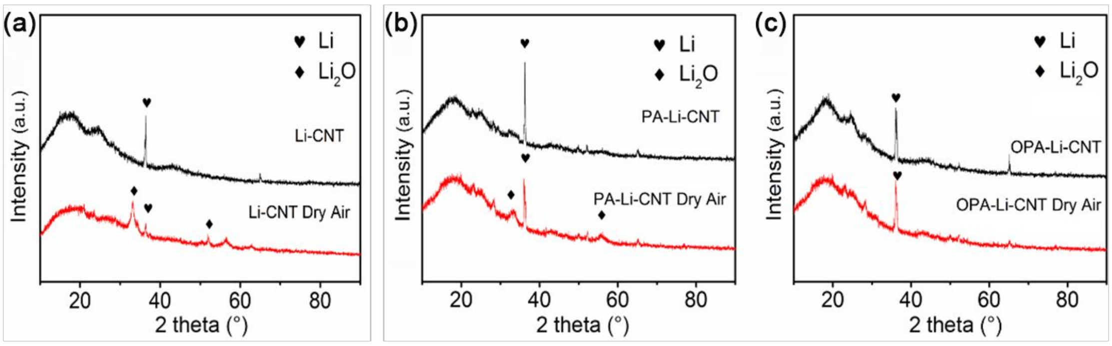

Figure S3. XRD of (a) Li-CNT, (b) PA-Li-CNT, and (c) OPA-Li-CNT before and after 1 hr exposure to dry air.

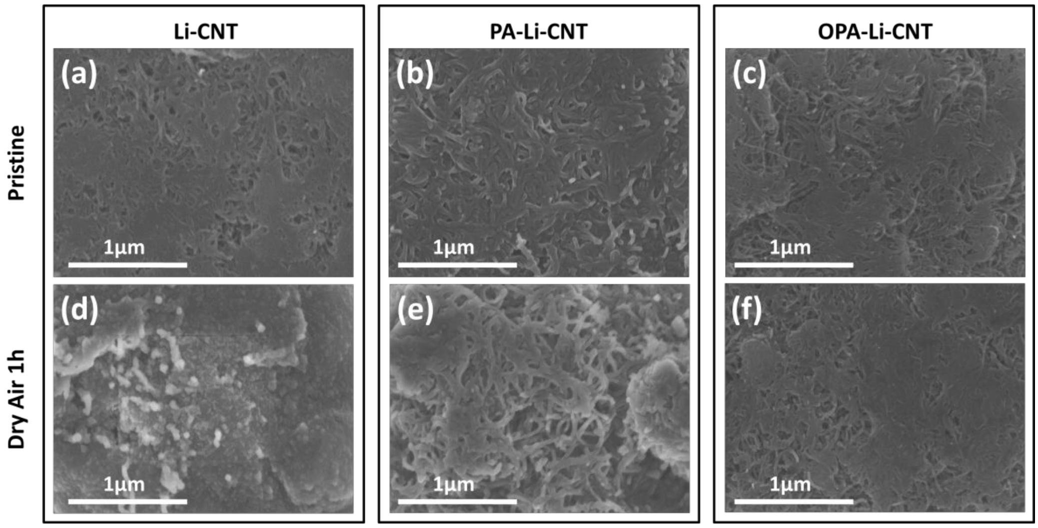

Figure S4. SEM micrographs of pristine (a) Li-CNT, (b) PA-Li-CNT, and (c) OPA-Li-CNT and after 1 hr exposure to dry air (d) Li-CNT, (e) PA-Li-CNT, and (f) OPA-Li-CNT

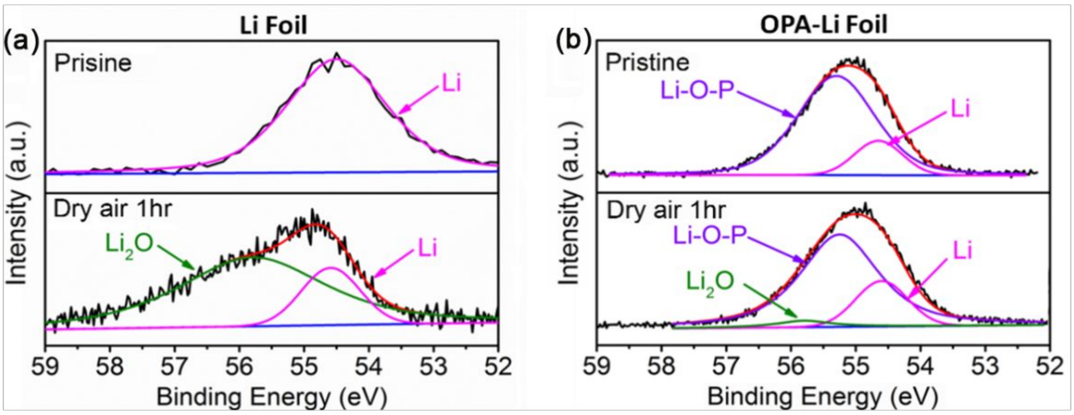

Figure S5. XPS spectra of pristine and dry air (dew point: -40 ℃) exposed samples: (a) Li Foil, (b) OPA-Li Foil

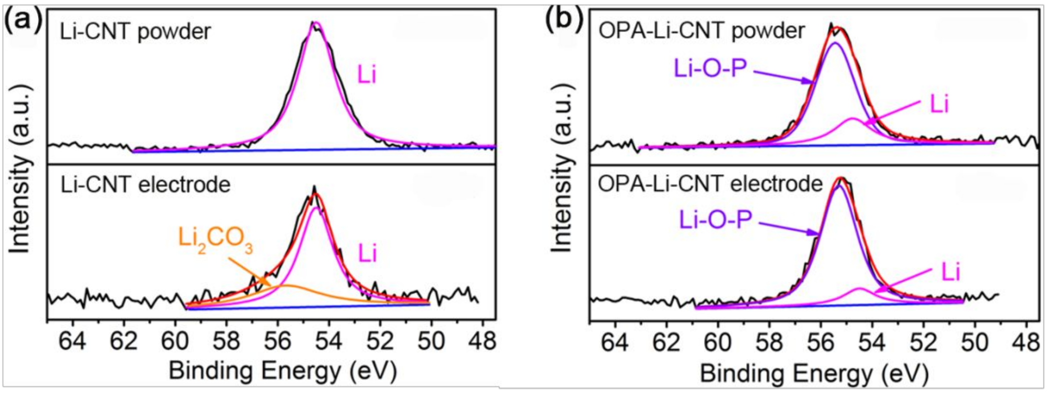

Figure S6. XPS spectra of Li-CNT and OPA-Li-CNT materials before and after slurry coating: (a) Li-CNT, (b) OPA-Li-CNT

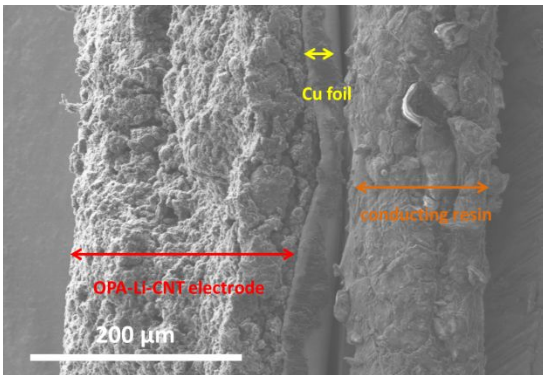

Figure S7. SEM on the cross section of the OPA-Li-CNT electrode

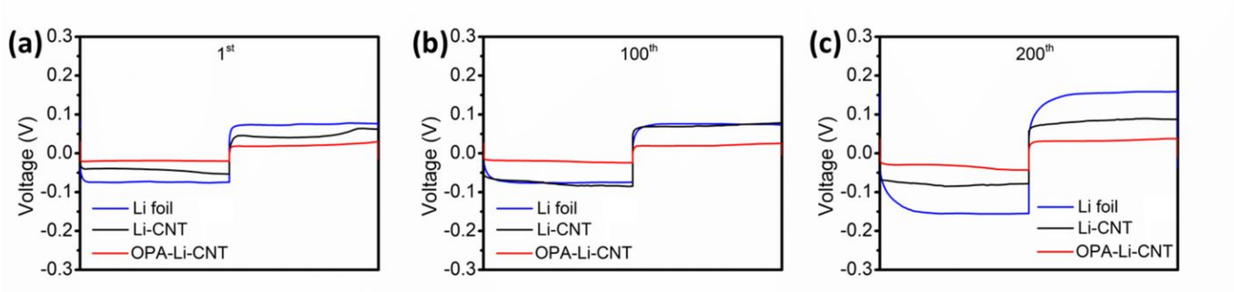

Figure S8. Overpotential of different anodes at different Li stripping/plating cycles. (a) the 1st cycle, (b) the 100th cycle, (c) the 200th cycle.

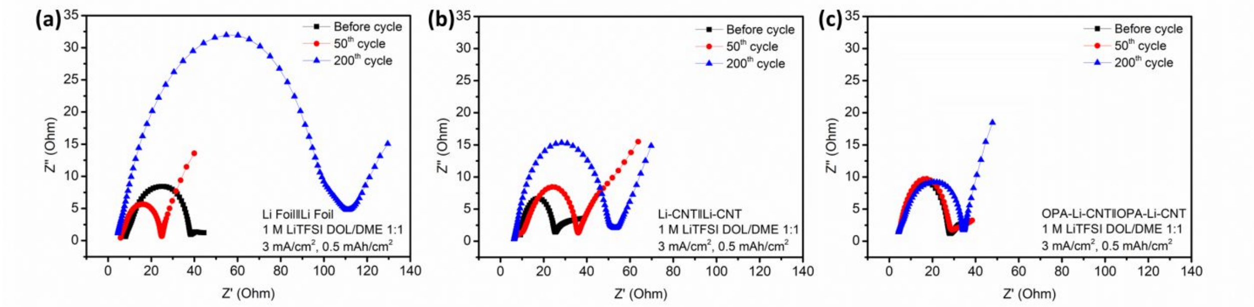

Figure S9. Nyquist plots of (a) Li foil||Li foil, (b) Li-CNT||Li-CNT and (c) OPA-Li-CNT||OPA-Li-CNT cells measured at different stripping/platting cycles.

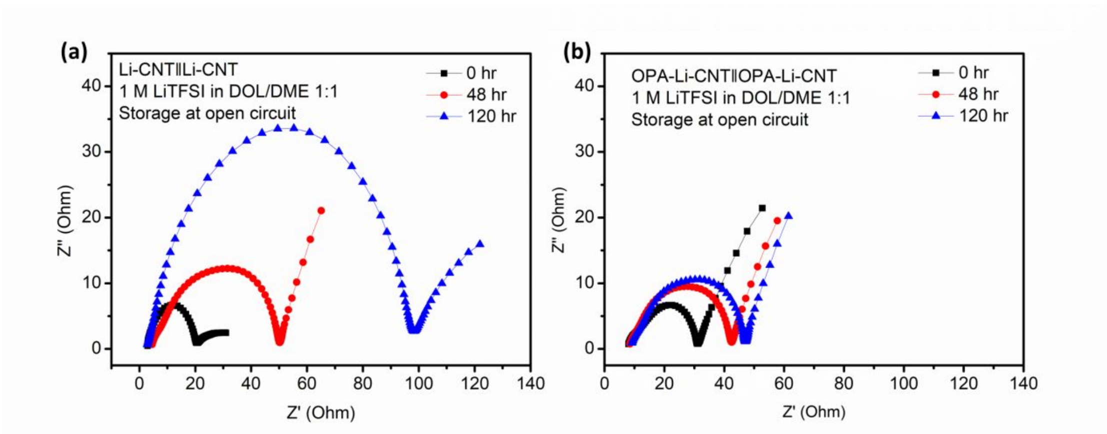

Figure S10. Nyquist plots of (a) Li-CNT||Li-CNT and (b) OPA-Li-CNT||OPA-Li-CNT cells measured at different storage time

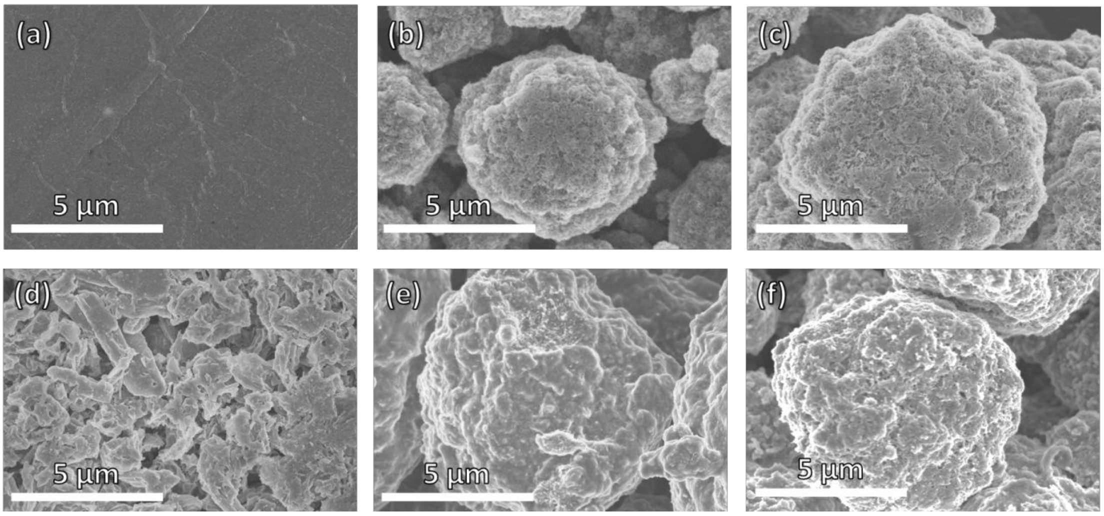

Figure S11. SEM images of Li foil (a), Li-CNT (b), OPA-Li-CNT (c), before cycle, and Li foil (d), Li-CNT (e), OPA-Li-CNT (f) after 200 cycles; at current density of 3 mA/cm^2, capacity density is 0.5 mAh/cm^2.

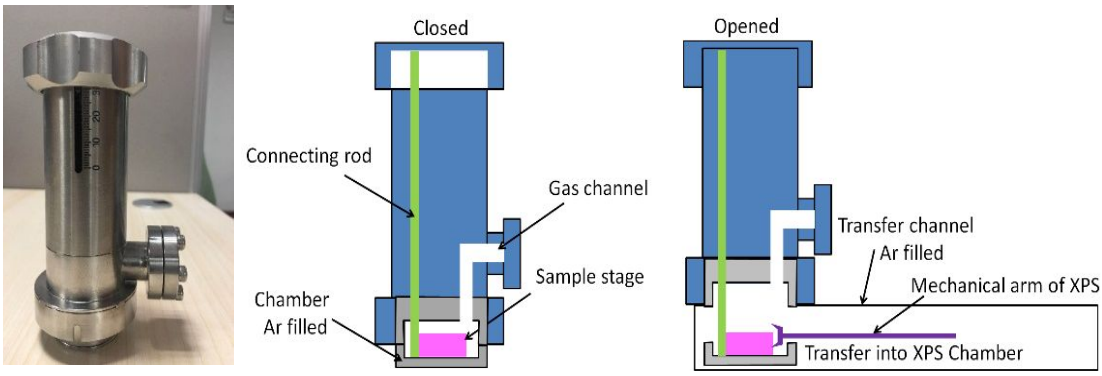

Figure S12. Photo and schematic illustration of the XPS sample transfer chamber

References

(1) Guo, F.; Wang, Y.; Kang, T.; Liu, C.; Shen, Y.; Lu, W.; Wu, X.; Chen, L. A Li-dual carbon composite as stable anode material for Li batteries. *Energy Storage Materials*, **2018**, *15*, 116-123.

(2) Wang, Y.; Shen, Y.; Du, Z.; Zhang, X.; Wang, K.; Zhang, H.; Kang, T.; Guo, F.; Liu, C.; Wu, X.; Lu, W.; Chen, L. A lithium-carbon nanotube composite for stable lithium anodes. *J. Mater. Chem. A*, **2017**, *5*, 23434-23439.

Safety statement

Li-CNT particles spontaneously react with dry or moist air, and thus must be handled with care in Ar atmosphere. OPA-Li-CNT can be processed in dry and moist air. Waste Li-CNT and OPA-Li-CNT materials need to be slowly oxidized under controlled conditions before disposed as chemical wastes.