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Supporting Information

Lithium-Carbon Nanotube Composite for Stable Lithium Anodes

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1. Experimental Section

1.1. Specific capacity of Li-CNT:

To evaluate the specific capacity of Li-CNT, Li||Li-CNT batteries were assembled, the Li-CNT mass in the battery is 25 mg, and the battery was galvanostatic charged at 0.1 mA cm^{-2}. When lithium was stripped from the Li-CNT electrode completely, the battery potential diverges obviously like other group reported.[S1]

1.2. Short-circuit measurement:

The battery configuration used for the short-circuit measurement is shown in Figure S4. A hollow paper ring was used as a separator. The inner diameter of the hole is 6 mm, and the thickness of the paper is 0.1 mm. During the measurement, the Li are continuously stripped from a Li electrode with far excess of Li and plated on the studied electrodes (Li-CNT or Li foil) at a constant current density until the battery fails due to Li dendrite-induced short circuit. By monitoring the voltage profile against the time, the time at which sharp drop-off in the voltage happens is identified as short circuit time (T_{sc}).[S2] T_{sc} can be used to estimate the capability of the studied electrode in retarding the formation of dendrites. The error bars in Figure 2b are standard deviations about the mean values obtained from three independent measurements performed under the same battery running conditions.

1.3. Semi-quantitative determination of the Coulombic efficiency（CE）of electrodes with pre-stored Li:

The accurate determination of CE for lithium-containing electrodes has been documented to be complicated. [S3-S5] Following the method developed by Liu Y. Y. et. al., S4 we paired the Li-CNT composite anode with a highly reversible cathode, in this case a commercial $LiFePO_4$ cathode was used for the CE estimation. If the irreversible capacity lost 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:

$$CE=(C_{cathode}-C_{total}/n)/\ C_{cathode}$$

Where C_{total} is the total capacity of the cathode and anode, $C_{cathode}$ is the cathode capacity, n is the cycle number at which the cycling curve starts to plunge.

For example, as shown in Figure 4a, the initial cell capacity of 20.7 mAh cm^{-2} (anode: 20.0 mAh cm^{-2}, cathode: 0.7 mAh cm^{-2}, the ratio of capcaity for cathode and anode is 1~30) the Li foil||LFP cell cycled at 1C starts to plunge at around 120 cycles, so the CE of Li foil can be estimated to be 75.4%. For the CE of the Li-CNT electrode, taking the 300 cycle times for *n* in the above equation, then the CE for the Li-CNT||LFP cell cycled at 1C is obtained to be 90.1%.

$$CE=\frac{0.7-\frac{20.7}{n}}{0.7}$$

For the cells with high cathode loading cycled at 0.5C as shown in Figure 4b, the initial cell capacity is 23.4 mAh cm^{-2} (anode: 20.8 mAh cm^{-2}, cathode: 2.6 mAh cm^{-2}, the ratio of capcaity for cathode and anode is 1~8). The Li foil||LFP cell starts to plunge at around 50 cycles, so the CE of Li foil can be estimated to be 81%. The Li-CNT||LFP start to decay at ~100 cycles, hence the CE for Li-CNT is estimated to be 91%.

$$CE = \frac{2.6 - \frac{23.4}{n}}{2.6}$$

2. Supplemental Results

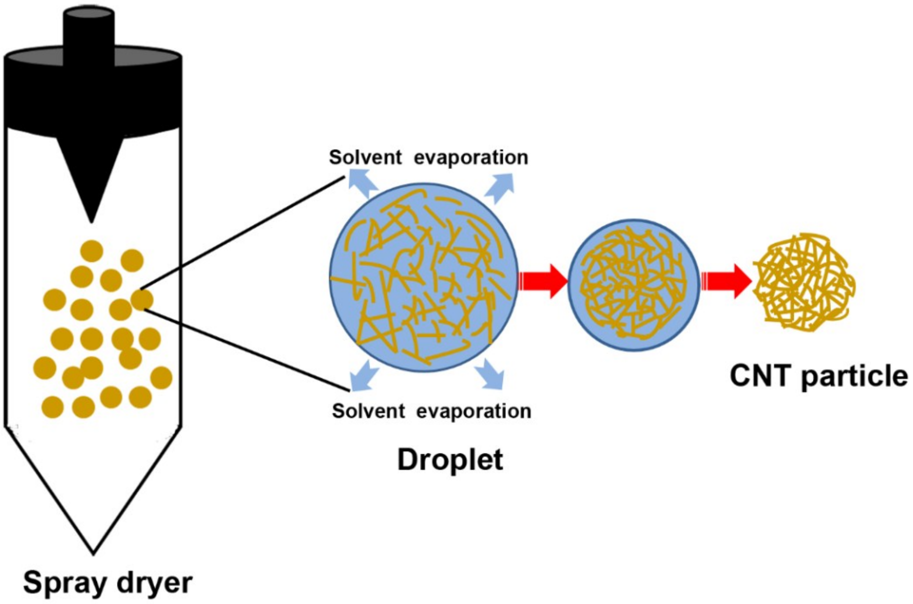

Figure S1. Schematic illustration of the spray dry preparation process of CNT particles.

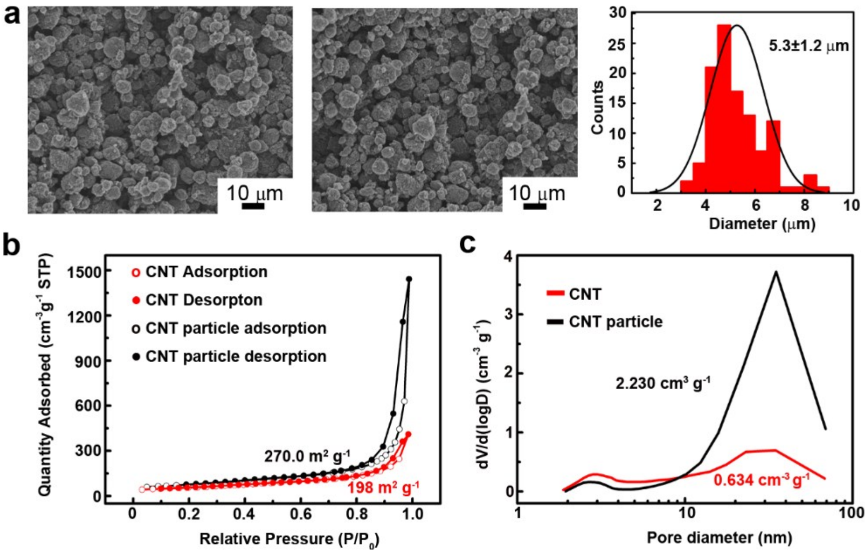

Figure S2. Characterization of the CNT particles. (a) SEM micrographs and the diameter distribution histogram of the CNT particles. (b) Argon adsorption/desorption isotherms of the pristine CNT and the CNT particles. (c) The pore-size distribution of pristine CNT and the CNT particles.

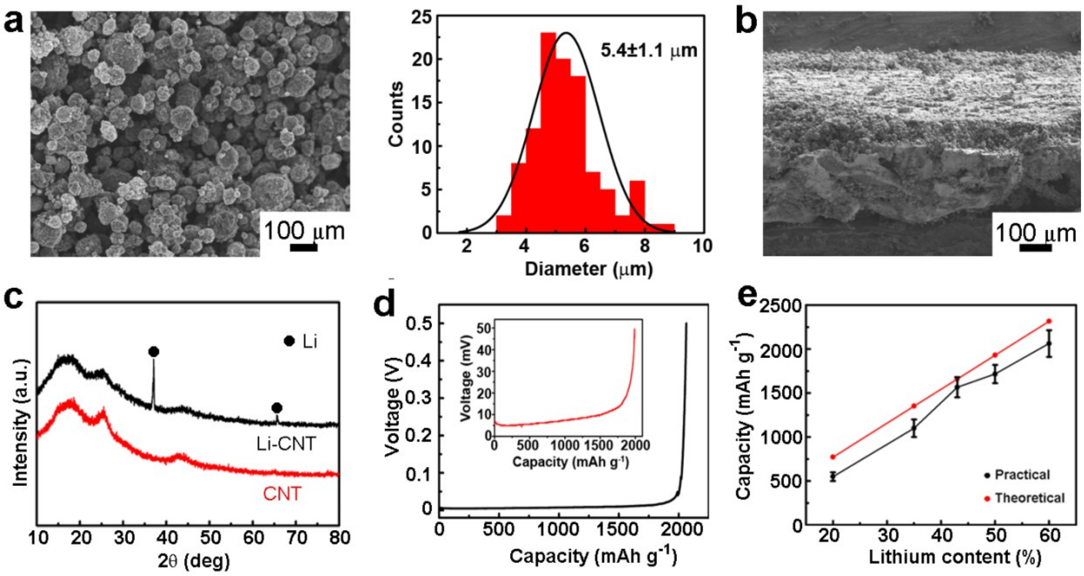

Figure S3. Characterization of the Li-CNT composite particles. (a) SEM micrographs and the diameter distribution histogram of the Li-CNT composite particles. (b) The cross-section SEM image of the Li-CNT electrode. (c) XRD patterns of the CNT particles (red line) and the Li-CNT composites particles (black line). (d) Galvanostatic discharge curve of the Li-CNT electrode. (e) The Li-CNT capacity vs. the lithium content in Li-CNT.

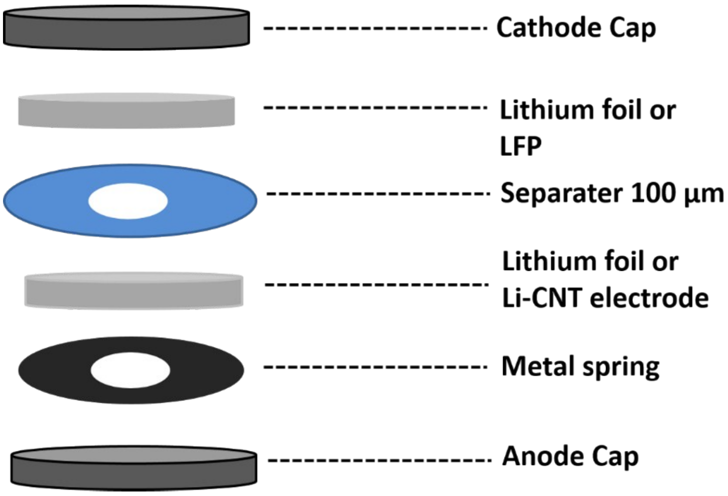

Figure S4. The configuration of the coin battery used for short-circuit time testing.

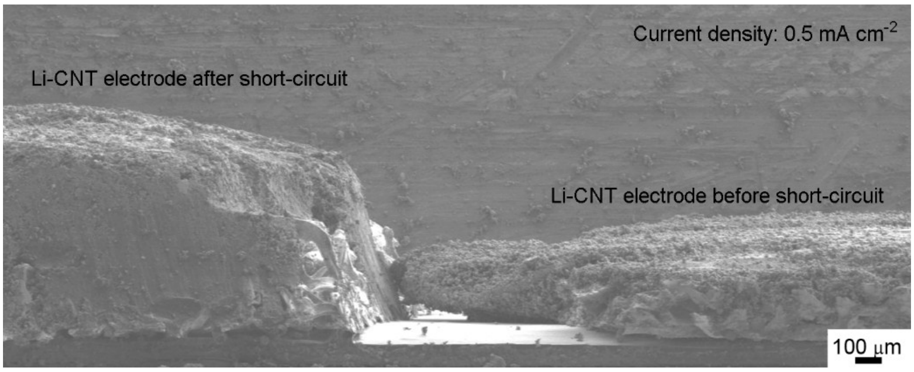

Figure S5. The cross section SEM images of the Li-CNT electrode before and after the short-circuit testing at a current density of 0.5 mA cm^{-2}.

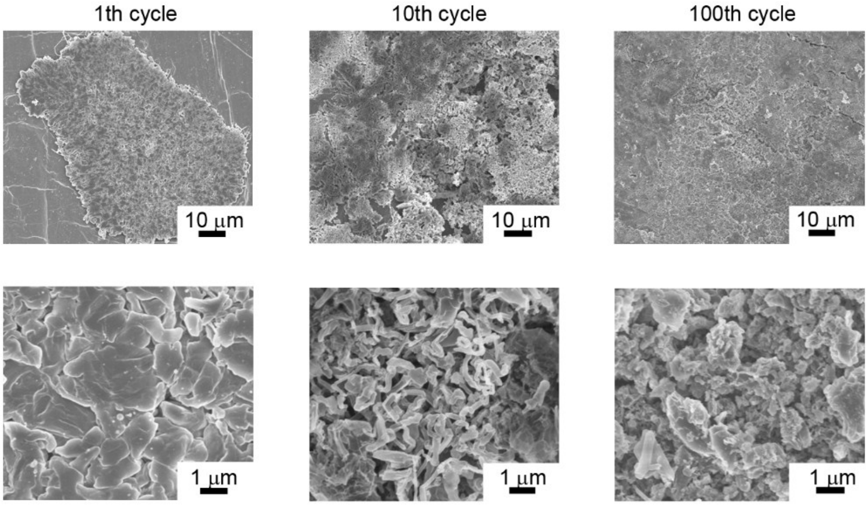

Figure S6. SEM images of the Li foil electrodes after different cycles at 0.5 mA cm^{-2}. The Li foil surface morphology evolution indicates that more and more dendrites grow on the electrode surface.

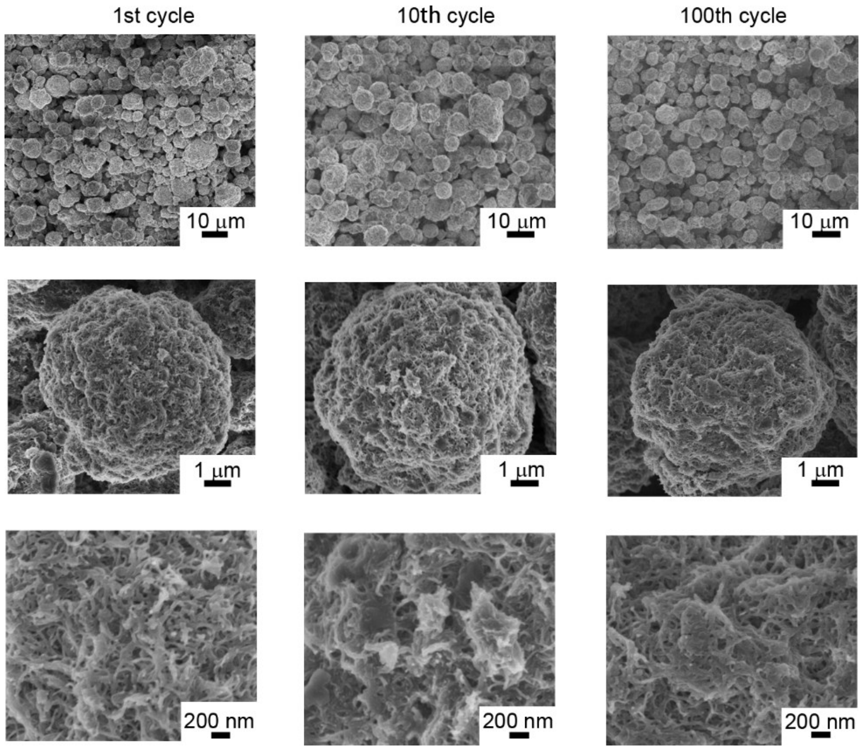

Figure S7. SEM images of the Li-CNT electrodes after different cycles at 0.5 mA cm^{-2}. The Li-CNT electrode shows almost no dendrite formation and the spherical structure remains.

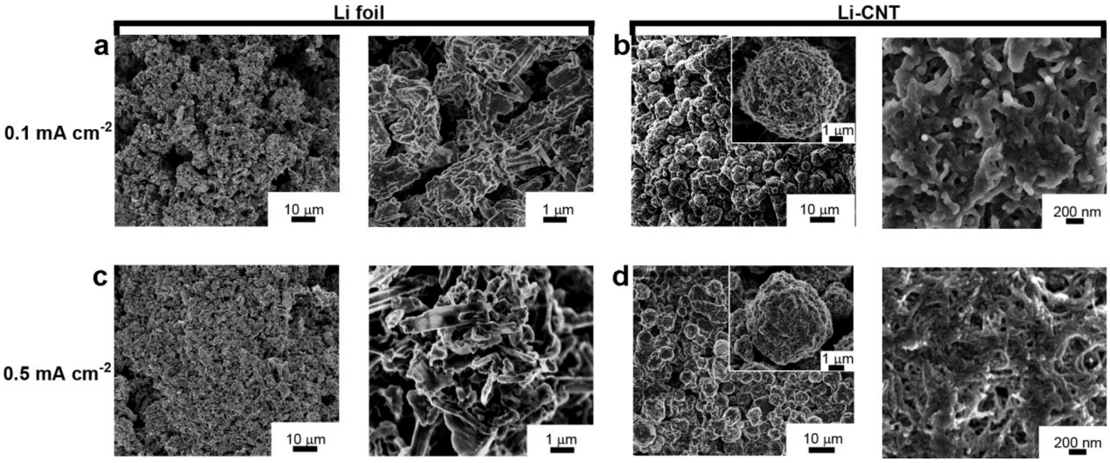

Figure S8. SEM images of (a) the Li foil electrodes and (b) the Li-CNT electrodes after 400 h cycling at 0.1 mA cm^{-2}. SEM images of (c) the Li foil electrodes and (d) the Li-CNT electrodes after 400 h cycling at 0.5 mA cm^{-2}.

References:

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