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# Supplemental Information

# Long-Life, High-Efficiency Lithium/Sulfur Batteries from Sulfurized Carbon Nanotube Cathodes

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**Detailed fabrication procedures for SCNT cathode materials**

First, Multi-walled carbon nanotubes (CNTs, 10~50 nm in diameter and 5~9 μm in length, Sigma-Aldrich, St. Louis, MO) were soaked in nitric acid (70 wt.%) and sulfuric acid (98 wt.%) (v 1:3) in an ultrasonic container for 1 h, kept in an oven of 70 $^0$C for 2 h, and then rinsed with distilled water to obtain the treated CNTs.

Second, elemental sulfur was dissolved into carbon disulfide solution ($CS_2$, Sigma-Aldrich) for 1 h to reach saturation. Meanwhile, the treated CNTs in the first step were immersed in hydrogen peroxide ($H_2O_2$, Sigma-Aldrich) solution for 1 h to obtain the functionalized CNTs.

Third, the prepared sulfur in $CS_2$ suspension were added dropwise to the CNTs in $H_2O_2$ solution (with pH=7, adjusted by LiOH) at a slow rate of 2 ml per minute. Next, the mixture was stirred under magnetic at 50 °C in a sealed glass box for 2 h. Then, the solution was evaporated at 50 °C in a fume hood for 1.5 h to obtain the pristine SCNTs.

Last, the samples were treated in a vacuum oven at 159 °C for 2 h followed at 300 °C for 5 h, and designated as SCNT-300. Two types of control samples were prepared: (i) SCNT-159, obtained by heating the pristine SCNTs in a vacuum oven at 159 °C for 10 h; (ii) S/CNT-159, fabricated by mixing sulfur and functionalized CNTs together and heating the mixture in a vacuum oven at 159 °C for 10 h.

## Figures and Tables

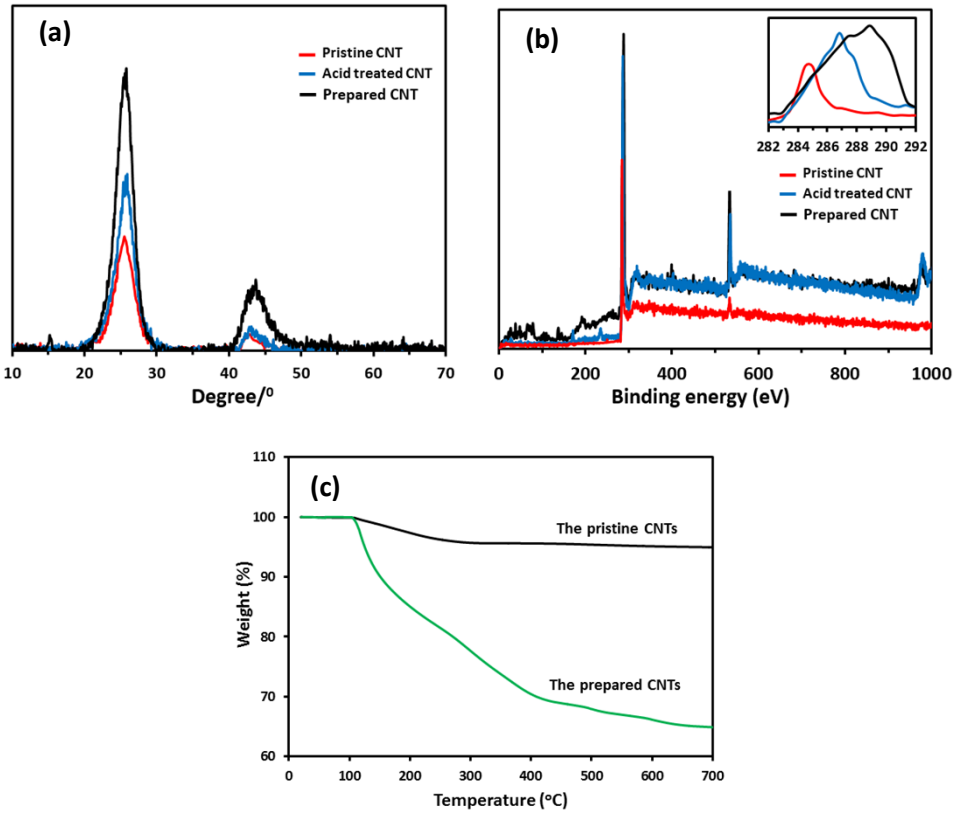


**Figure S1.** Characterization of the pristine and prepared CNTs. (**a**) XRD pattern and (**b**) XPS analysis of three kinds of CNT: pristine CNTs, CNTs treated with two mixed concentrated acids (i.e., nitric acid and sulfuric acid), and CNTs treated with the two mixed concentrated acids followed with $H_2O_2$ treatment. The intensities of both carbon peaks and oxygen peaks in Figure S1b increased with the mixed acids and $H_2O_2$ treatments. Compared to the non-treated CNTs, the C1s peak exhibited both a shift and an asymmetric broadening to higher binding energies (Figure S1b inset). The tip-shift of this peak, due to the polar character of the carbon oxygen bonds, was an evidence of the incorporation of oxygen into the nanotube structure. A contribution at about 288.4 eV described surface oxygen groups with multiple carbon oxygen bonds. (**c**) TGA profiles of the pristine and prepared CNTs.

The oxygen and corresponding -COOH contents in the CNTs were calculated in **Tables S1** and **S2**, respectively. The oxygen content in the CNTs significantly increased from 1.1 wt.% of pristine CNT to 10.4 wt.% and 19.7 wt.% respectively, after being treated with the mixed concentrated nitric and sulfuric acids or treated with the two mixed concentrated acids followed with $H_2O_2$ treatment.

| Sample | Peak | Center | Wt.% | Stoichiometric amount |
|---|---|---|---|---|
| Pristine CNT | C1s | 284.5 | 98.5 | 1.00 |
| | O1s | 531.4 | 1.5 | 0.01 |
| CNT treated with mixed acids | C1s | 286.4 | 89.6 | 1.00 |
| | O1s | 530.9 | 10.4 | 0.09 |
| CNT treated with mixed acids and $H_2O_2$ | C1s | 288.4 | 80.3 | 1.00 |
| | O1s | 530.4 | 19.7 | 0.18 |

**Table S1.** Surface element contents of CNTs with different treatment methods.

| Sample | O stoichiometric amount (%) | -COOH stoichiometric amount (%) | -COOH weight (wt.%) |
|---|---|---|---|
| Pristine CNT | 1.1 | 0.55 | 4.2 |
| CNT treated with mixed acids | 8.7 | 4.35 | 12.2 |
| CNT treated with mixed acids and $H_2O_2$ | 18.4 | 9.2 | 25.9 |

**Table S2.** Relationship between O content and –COOH content on the surface of CNTs.

| Sample | Weight loss (wt.%) | CNT content (wt.%) | -COOH content (wt.%) | Sulfur content (wt.%) |
|---|---|---|---|---|
| FCNT | 36.2 | 63.8 | 36.2 | 0 |
| SCNT-159 | 90.3 | 9.7 | 5.5 | 84.8 |
| SCNT-300 | 79.6 | 20.4 | 11.6 | 68 |

**Table S3.** Sulfur contents in SCNT-159 and SCNT-300.

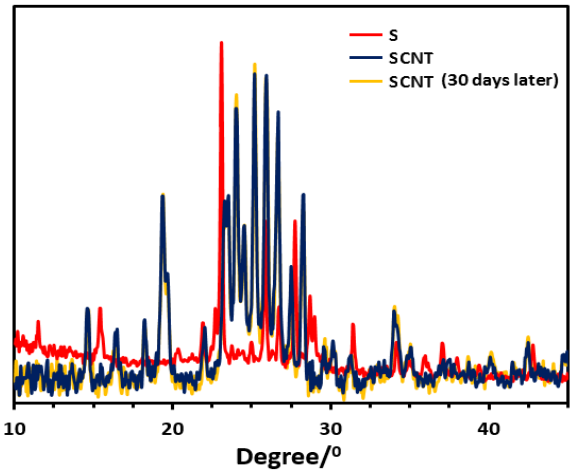


**Figure S2.** XRD pattern of SCNT-300. The majority of sulfur in SCNT-300 adopted the uncommon monoclinic phase rather than the typical orthorhombic phase (sulfur), and the monoclinic sulfur was very stable even after stored for 30 days at room temperature.

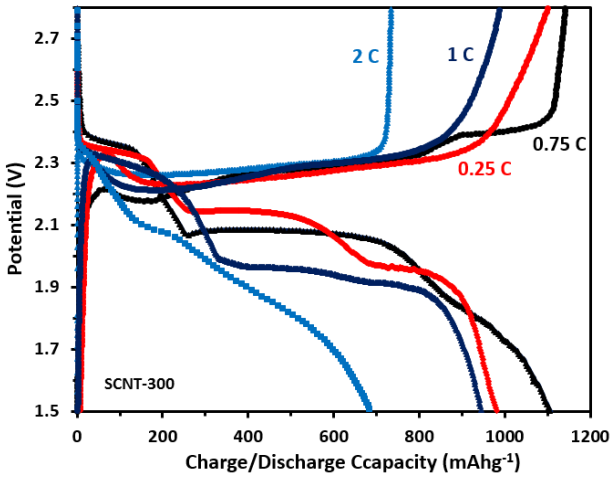


**Figure S3**. Voltage profiles of SCNT-300 cathodes at different current rates.

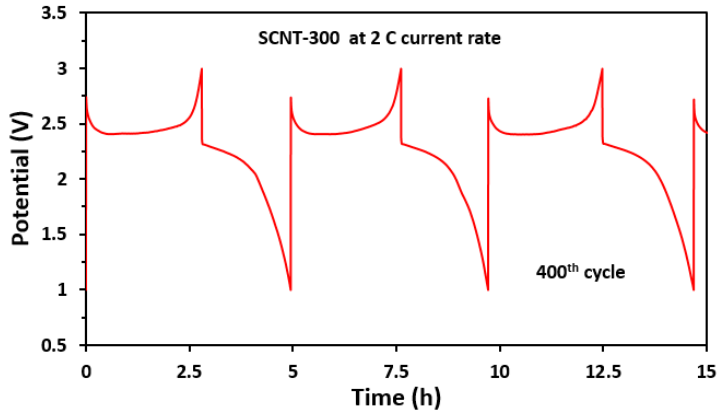


**Figure S4**. Voltage profiles (400$^{th}$ cycle) of SCNT-300 cathodes at 2 C.

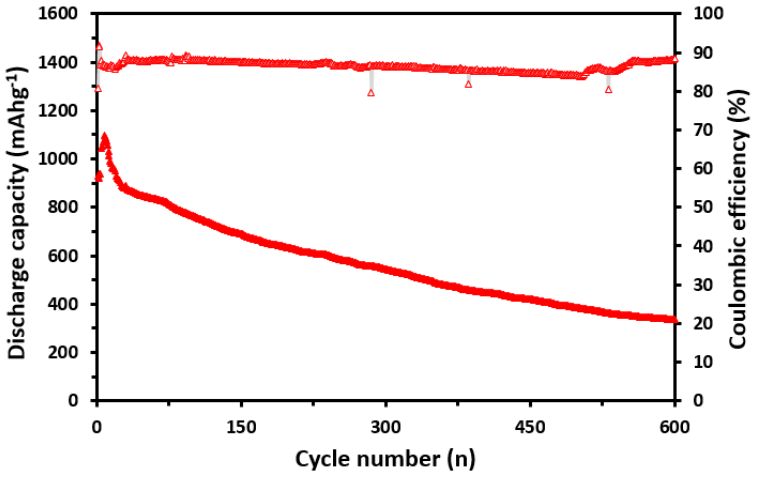


**Figure S5.** Cycling properties of Li-S cells comprised of SCNT-159 cathodes, which contained 10 wt.% of PVDF and 90 wt.% SCNT-159 material; Capacities were normalized by the mass of sulfur. The current rate for discharge/charge was 0.75 C. Sulfur load was 3.07 mg cm$^{-2}$ in the cathode.

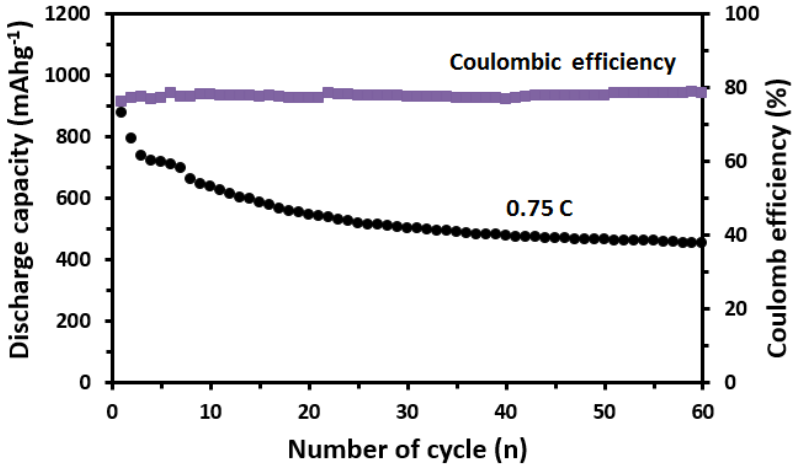


**Figure S6.** Cycling properties of Li-S cells comprised of S/CNT-159 (mixing sulfur and functionalized CNTs) cathodes, which contained 10 wt.% of PVDF and 90 wt.% S/CNT-159 material; Capacities were normalized by the mass of sulfur. The current rate for discharge/charge is 0.75 C. Sulfur load was 3.01 mg $cm^{-2}$ in the cathode.