Supporting Information

Hydroxylated CNTs Enhanced Sulfur Cathode for Improved Electrochemical Performance of Lithium-Sulfur Batteries

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Experimental

1. Preparation of Sulfur-MWNT composite

First, 3.2 g of $Na_2S_2O_3$ was dissolved in 1 L of Deionized water with the bath sonicator for 15 min. 0.1 g of OH-MWNT (20-30 nm of diameter, Cheaptubes) was introduced into the as-prepared precursor solution, followed by tip sonication (350 W, 10 min) to debundle the OH-MWNT and make the homogeneous solution. 1 ml of hydrochloric acid (36.5-38 %, DAEJUNG CHEMICALS) was dropped very slowly into the as-prepared solution with vigorously stirring. The reaction was maintained for 1 hour, and then the color changed from the black to greenish. After filtering the precipitates, they were washed several times with DI water. The Sulfur-MWNT powder was dried in a vacuum oven at 40^{o}C for 48 hours to remove the humidity contained on the surface of Sulfur-MWNT.

2. Fabrication and evaluation of the Lithium-sulfur battery

The electrochemical properties of S/MWNT, S/MWNT-OH25, and S/MWNT-OH50 were evaluated using a coin-type half cell (2032R type). Lithium metal was used as a counter electrode. The preparation of S/MWNT cathode was prepared by mixing 80 % of active material, 10 % of Super P and 10 % of PVdF, and then casted on the Al foil, followed by drying in a vacuum oven at 50 ^{o}C for 24 hours. In cases of preparing each S/MWNT-OH25 and S/MWNT-OH50 electrodes, Sulfur-MWNT and OH-MWNT were mixed in the proportion of 60 % to 20 % and 40 % to 40 %, respectively. The procedure of mixing Sulfur-MWNT and OH-MWNT was done by planetary centrifugal mixer (2000 rpm, 20 min). The specific capacity of sulfur cathode is normalized to the sulfur mass. In the electrodes, the sulfur loading for S/MWNT, S/MWNT-OH25, and S/MWNT-OH50 are assigned to 1.50 mg/cm2, 1.19 mg/cm2, and 0.72 mg/cm2, respectively. The electrolyte used in this work was 1 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) in a solvent mixture of 1,3-dioxolane and 1,2-dimethoxyethane (volume ratio 1:1) dissolved with 0.2 wt% of $LiNO_3$. The galvanostatic discharge-charge evaluations of the cell were done between 1.5 V and 3 V vs. Li/Li^+ using the battery cycle tester TOSCAT 3100 (Toyo Systems, Japan).

3. Characterizations

The morphologies of OH-MWNT and Sulfur-MWNT were observed using a JSM 4700F field emission scanning electron microscope (JEOL) and a JEM 2100F transmission electron microscope (JEOL). The Sulfur-

MWNT was estimated by thermogravimetric analysis (TGA) with a Q500 thermogravimetric analyser (TA Instruments) in the temperature range of 80 to 800 ^{o}C at a ramping rate of 10 ^{o}C /min in air. The relative quantity of polysulfide was measured by UV-Visible spectroscopy (Cary 5000 UV-Vis-NIR). The sample of UV-visible measurement was prepared by disassembling the cell in the glove box, followed by collecting the remaining electrolyte. The collected electrolyte was carefully put into the cell and then sealed with the cap to prevent the direct contact with air. The crystallographic patterns of the OH-MWNT and Sulfur-MWNT were determined using X-ray diffraction patterns obtained by a Bruker Miller diffractometer using Cu-K α radiation. The electrochemical impedance spectroscopy (EIS) was evaluated using an Autolab PGSTAT 302N potentiostat/galvanostat apparatus (Metrohm AG) in the frequency range of 250 kHz to 10 mHz at an excitation amplitude of 10 mV. XPS analysis was conducted with a Sigma Probe (Thermo VG Scientific) with Al-Kα X-ray radiation.

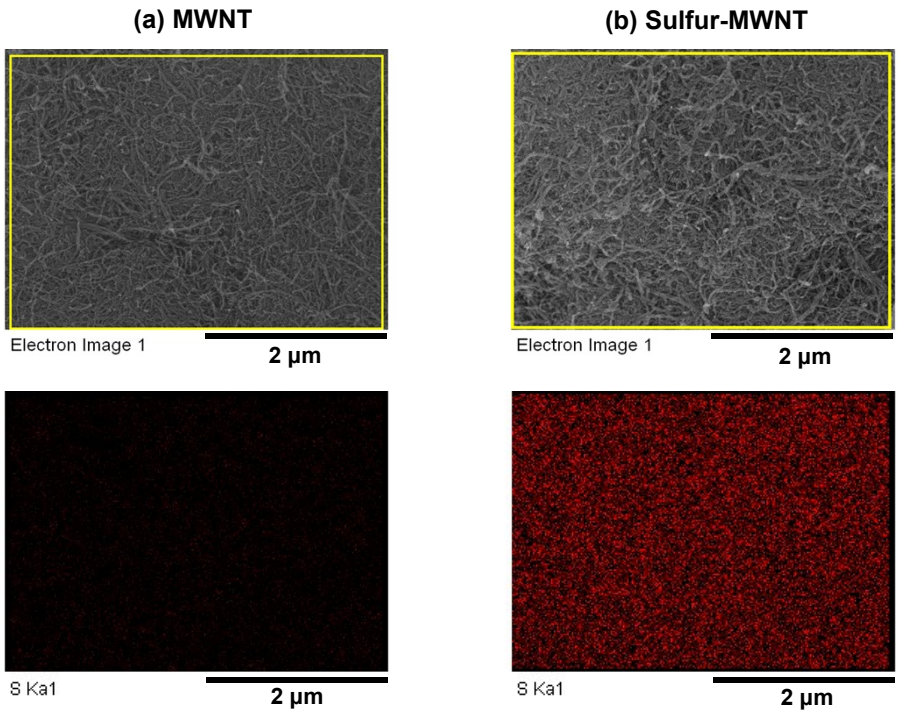

Figure S1. SEM EDS analysis of MWNT and Sulfur-MWNT.

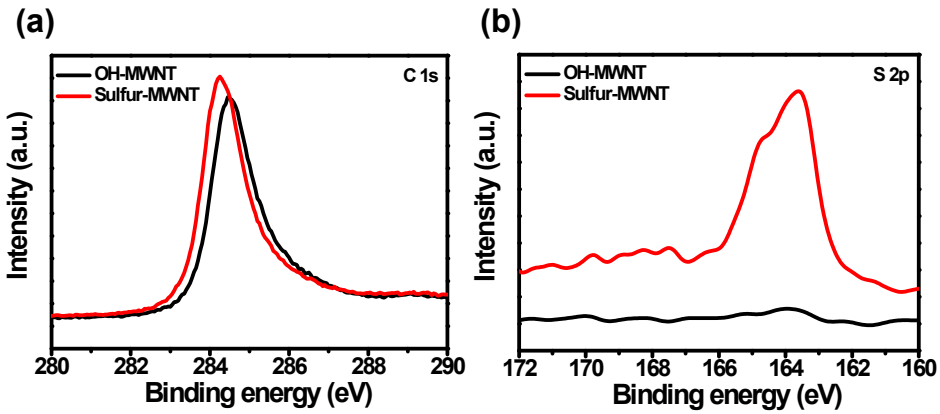

Figure S2. (a) C 1s XPS spectra of OH-MWNT and Sulfur-MWNT, (b) S 2p XPS spectra of OH-MWNT

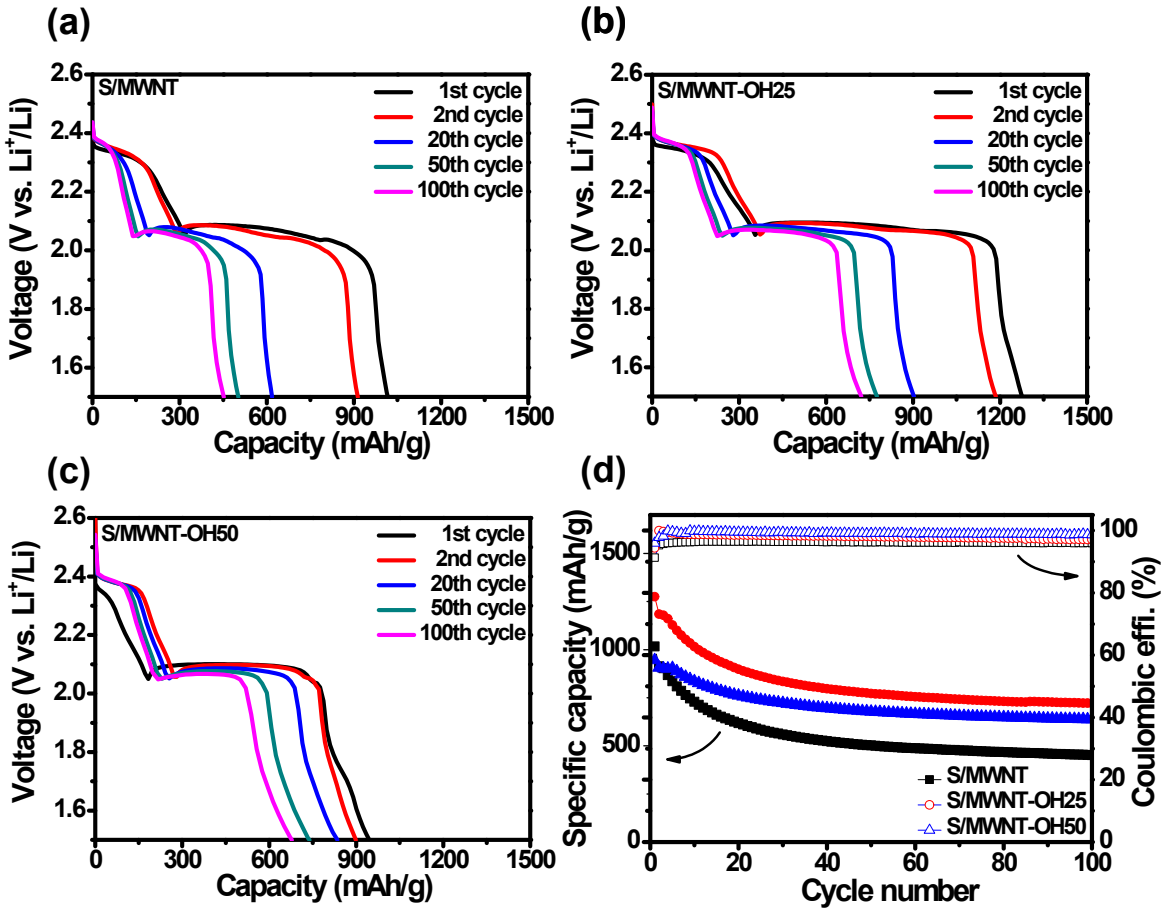

Figure S3. Discharge profiles of (a) S/MWNT, (b) S/MWNT-OH25, (c) S/MWNT-OH50. (d) Cycle performance of each electrodes at 0.2C

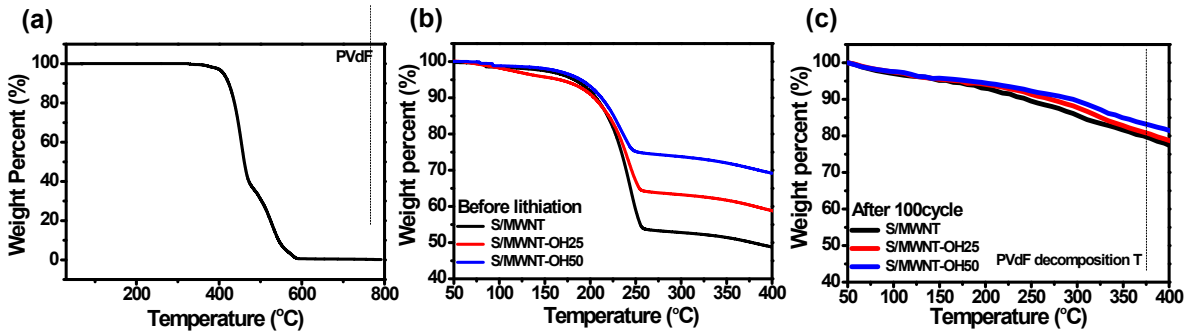

Figure S4. TGA curves of (a) PVdF and (b) before, (c) after 100 cycles of each S/MWNT, S/MWNT-OH25, and S/MWNT-OH50 electrodes

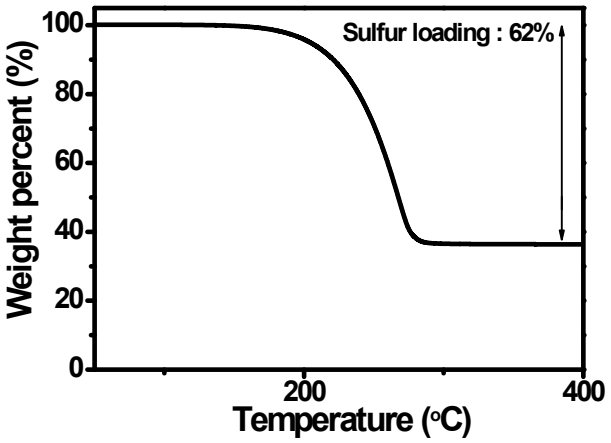

Figure S5. TGA curves of Sulfur-MWNT composite

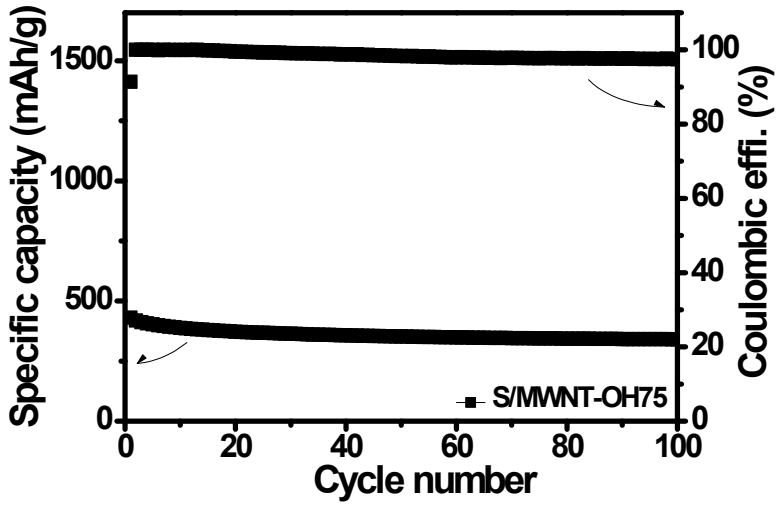

Figure S6. Cycle performance and Coulombic effiiciency of S/MWNT-OH75 electrode during 100 cycles