Supporting information

The creation of hollow walls in carbon nanotubes for high-performance lithium ion batteries

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Experimental Section

Synthesis of spinnable CNT array. Aligned carbon nanotube (CNT) array was synthesized through chemical vapor deposition. The catalyst was synthesized *via* electron beam evaporation. Aluminum oxide and iron were deposited on silicon wafer with thicknesses of 2 and 1.2 nm, respectively. The catalyst was then transferred into a tube furnace, and Ar (400 sccm), C_2H_4 (90 sccm) and H_2 (30 sccm) were introduced to this system with the growth temperature kept at 740 ºC for 10 min.

Characterization. The structures were characterized by scanning electron microscopy (SEM, Hitachi FE-SEM S-4800 operated at 1 KV), and transmission electron microscopy (TEM, JEOL JEM-2100F operated at 200 KV). The electrochemical performances were measured by an Arbin electrochemical station (MSTAT-5V/10mA/16Ch). The batteries were assembled using hollow-structured CNTs directly as working electrode and lithium foil as counter electrode with the commonly used LB303 electrolyte. They were charged and discharged at 0.005-3.0 V at current densities of 0.1, 0.2, 0.5, 1, 2 and back to 0.1 A g^{-1}. The cyclic voltammograms were obtained at a scan rate of 0.5 mV s^{-1} at 0.01-3 V.

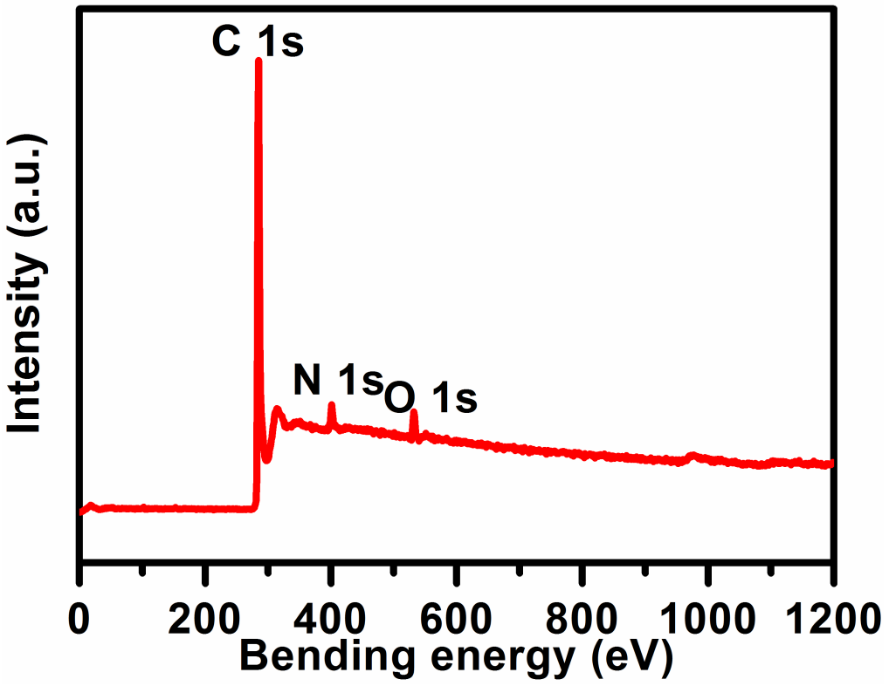

Figure S1. X-ray photoelectron spectra of the HNCNTs.

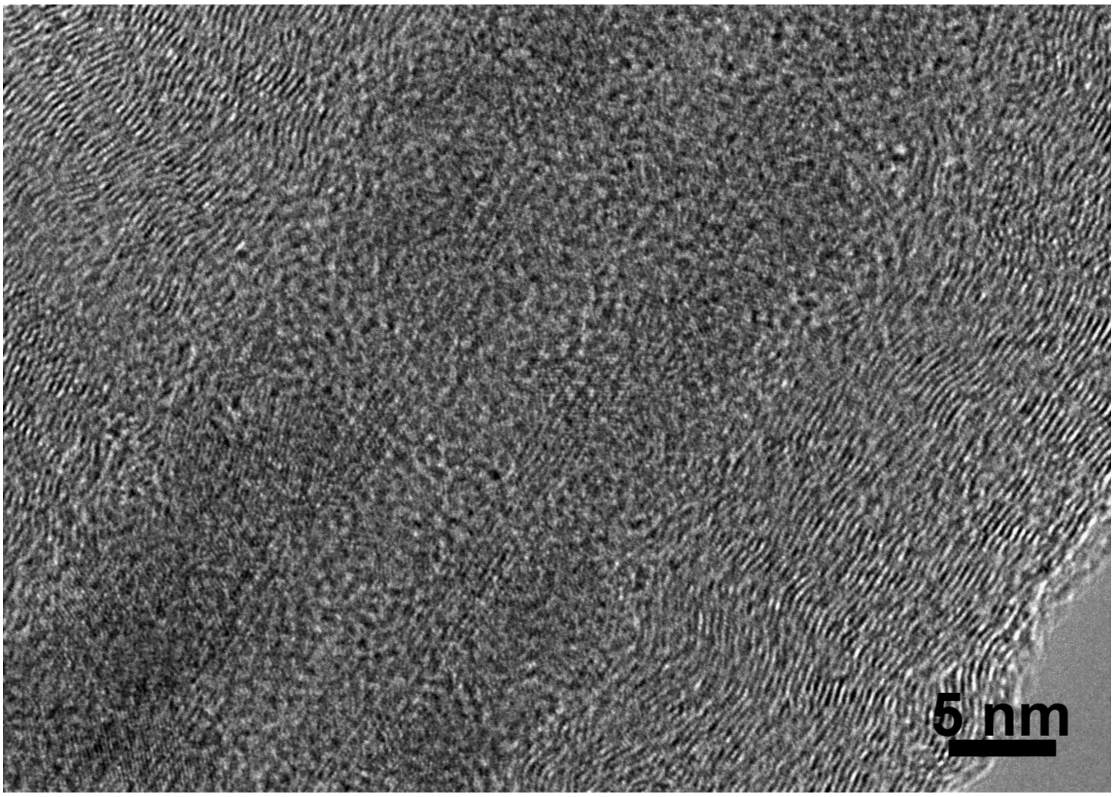

Figure S2. High-resolution TEM image of nitrogen-doped graphene/Al_2O_3/CNT.

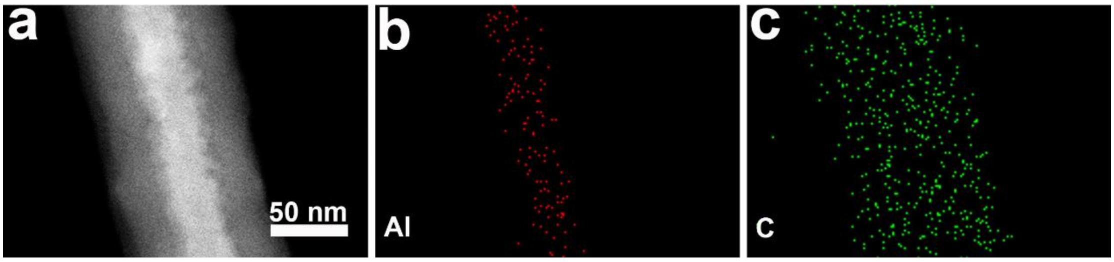

Figure S3. Energy-dispersive X-ray spectroscopy images of the nitrogen-doped graphene/Al_2O_3/CNT sheet. a) TEM image of the nitrogen-doped graphene/Al_2O_3/CNT. b) The dispersion of Al element. c) The dispersion of C element.

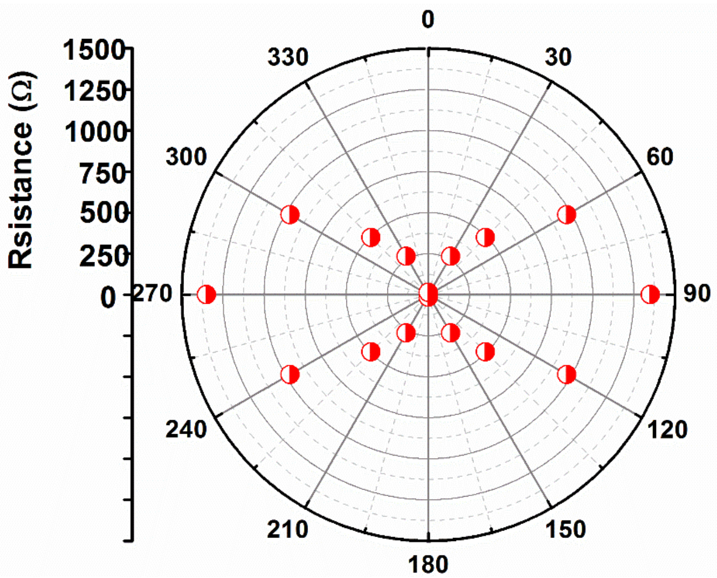

Figure S4. Electrical resistances on different angles in nitrogen-doped graphene/Al_2O_3/CNT.

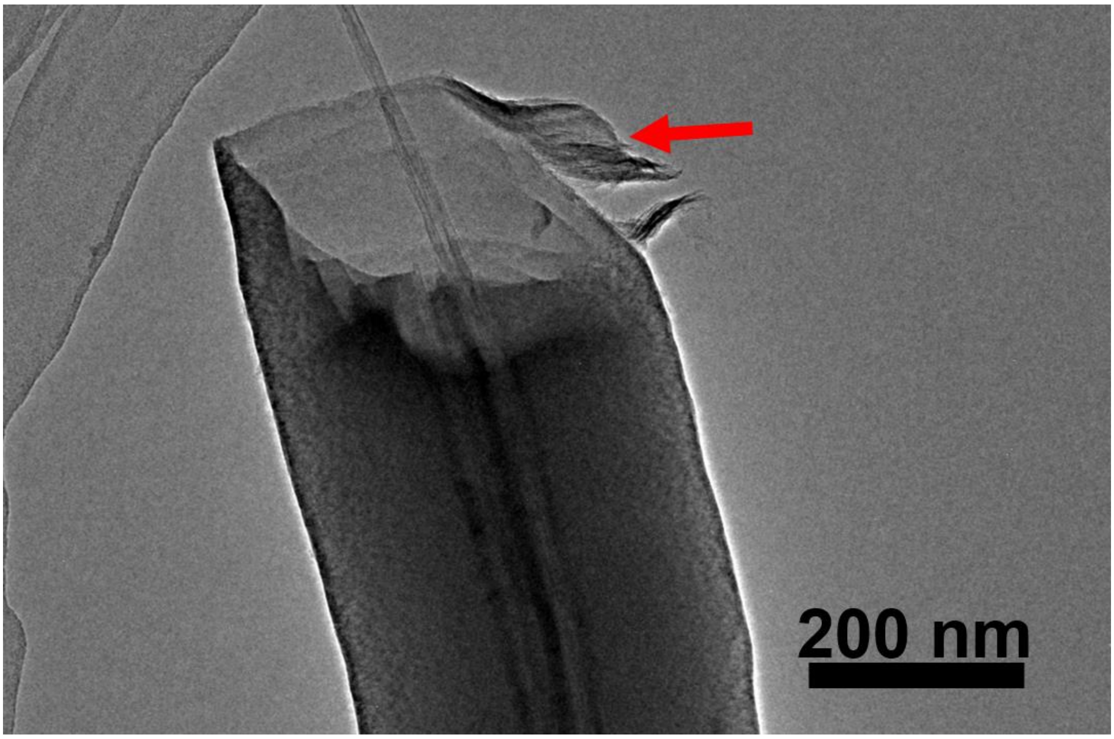

Figure S5. TEM image of the HNCNT shell (indicated by the red arrow).

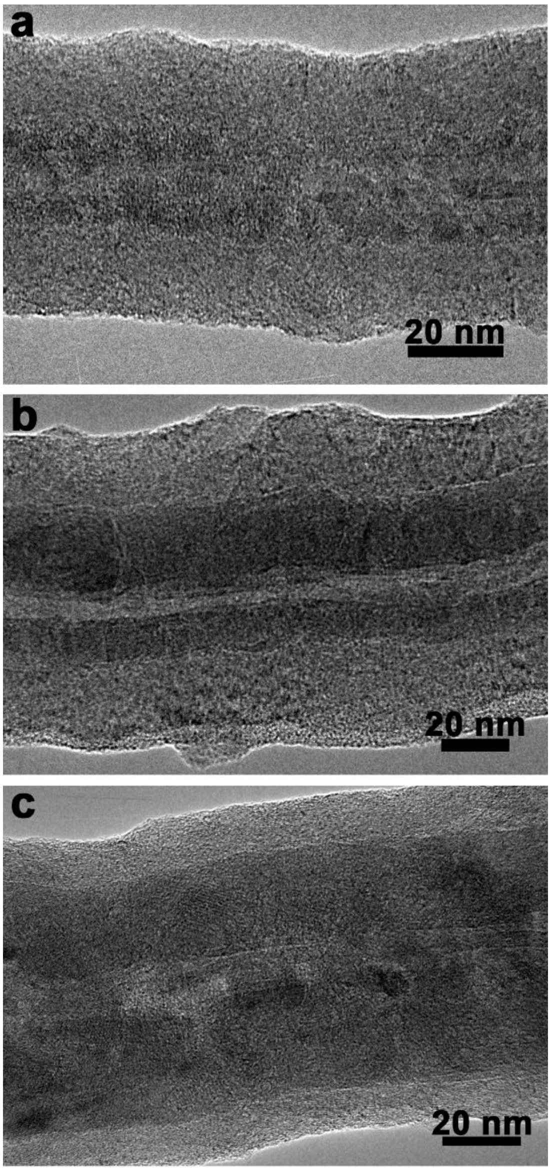

Figure S6. TEM images of nitrogen-doped graphene/Al_2O_3/CNTs with different Al_2O_3 thicknesses of 10, 20 and 30 nm. The re-growth time was 5 min.

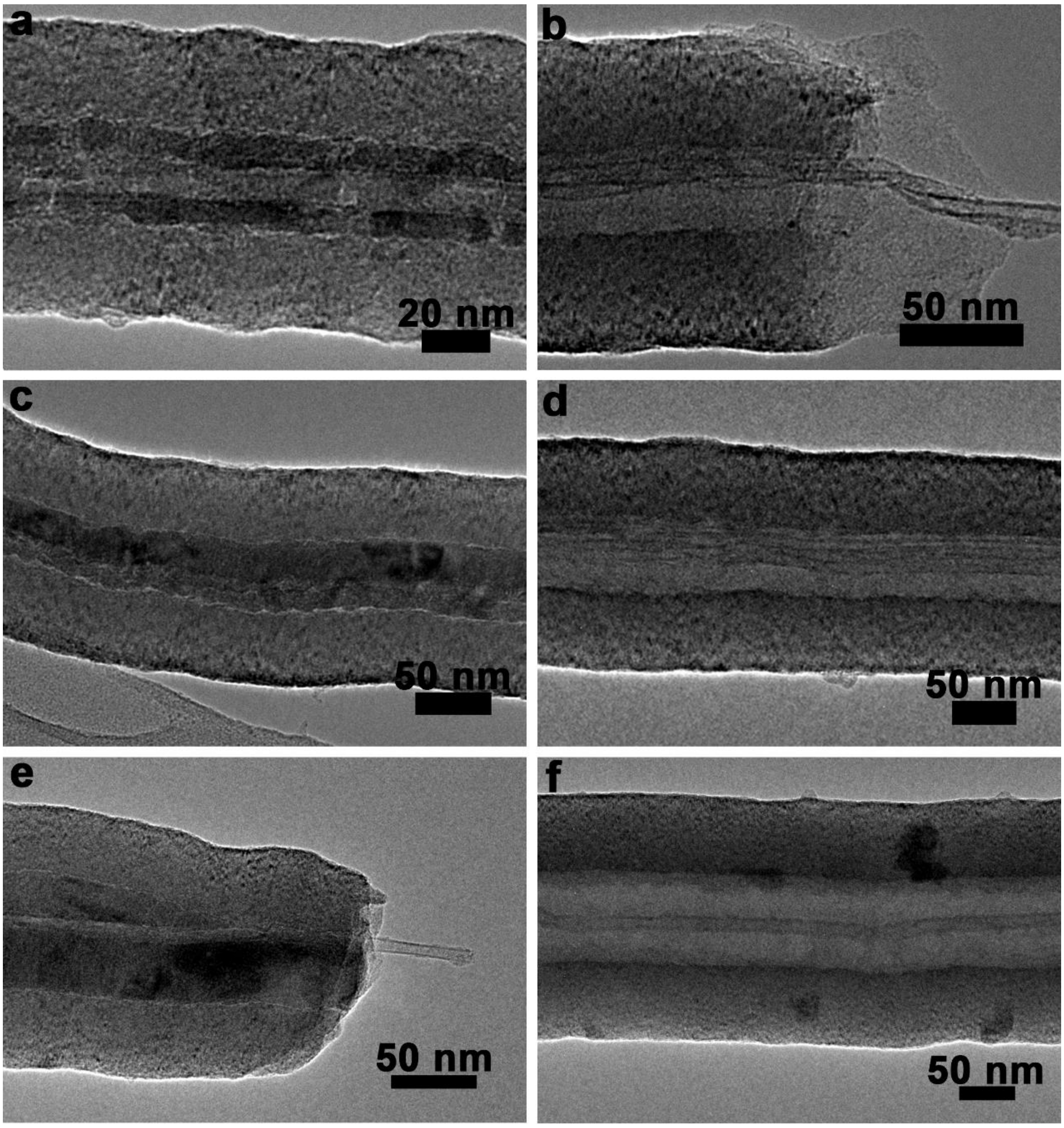

Figure S7. TEM images of HNCNTs before and after HF etching with different thicknesses of Al_2O_3 layer. a, b) 10 nm, c, d) 20 nm, e, f) 30 nm. The re-growth time was 10 min.

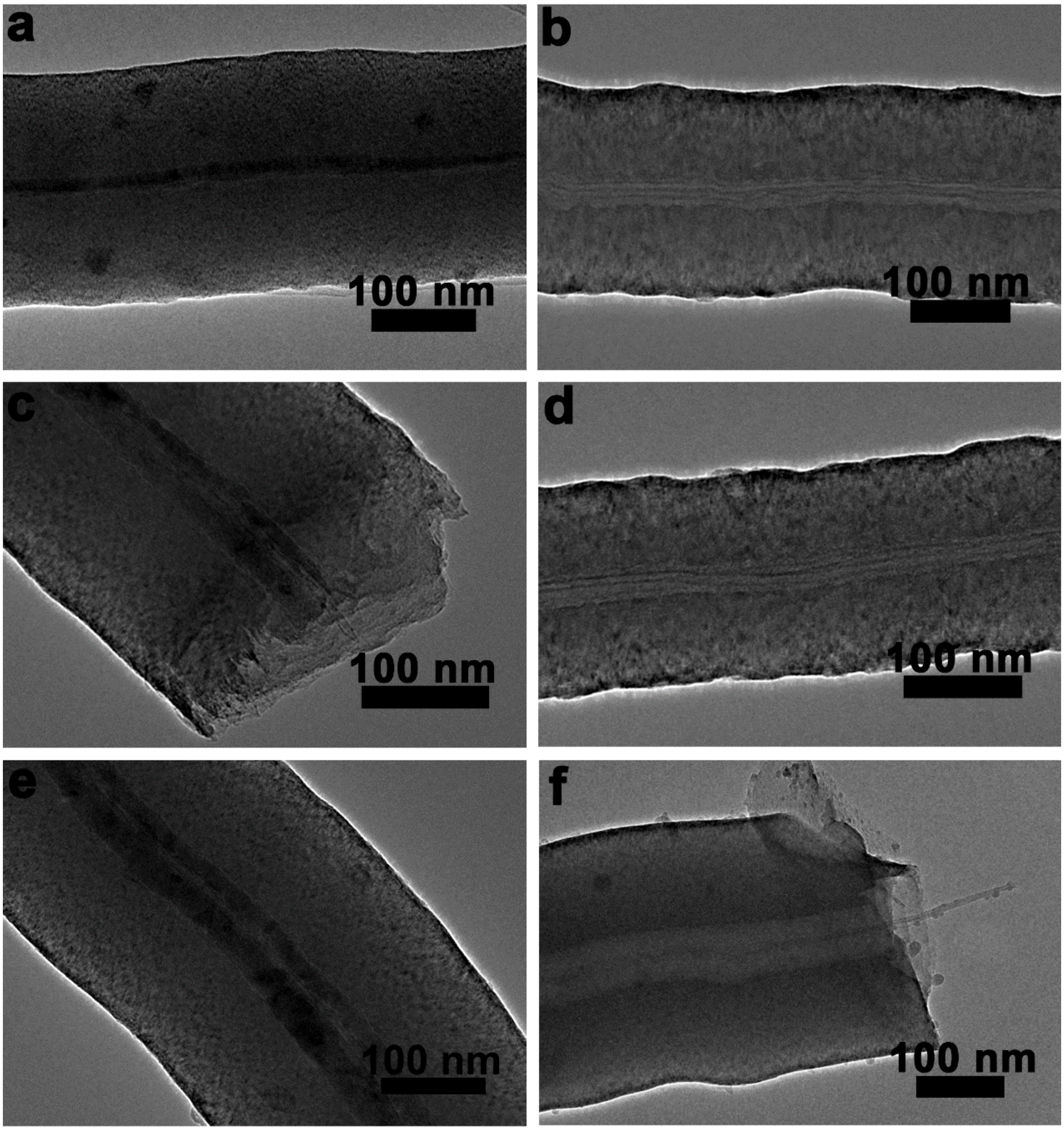

Figure S8. TEM images of HNCNTs before and after HF etching with different thicknesses of Al_2O_3 layer. a, b) 10 nm, c, d) 20 nm, e, f) 30 nm. The re-growth time was 30 min.

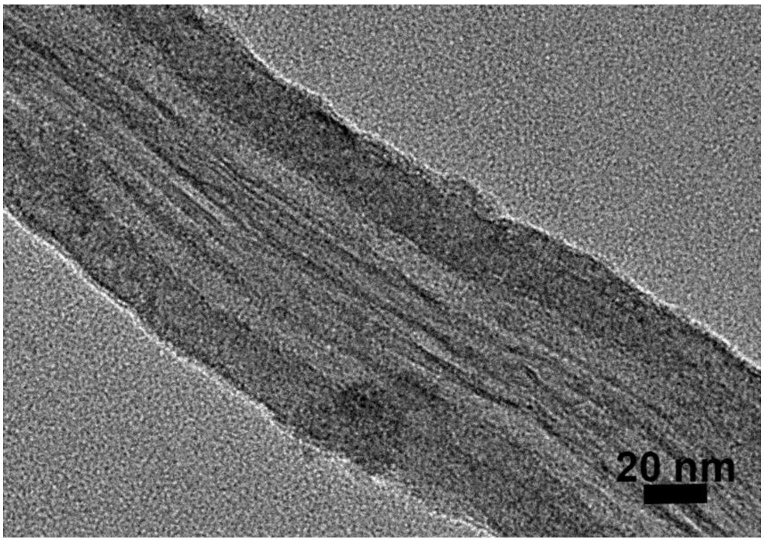

Figure S9. TEM image of an HNCNT with three CNTs bundled together.

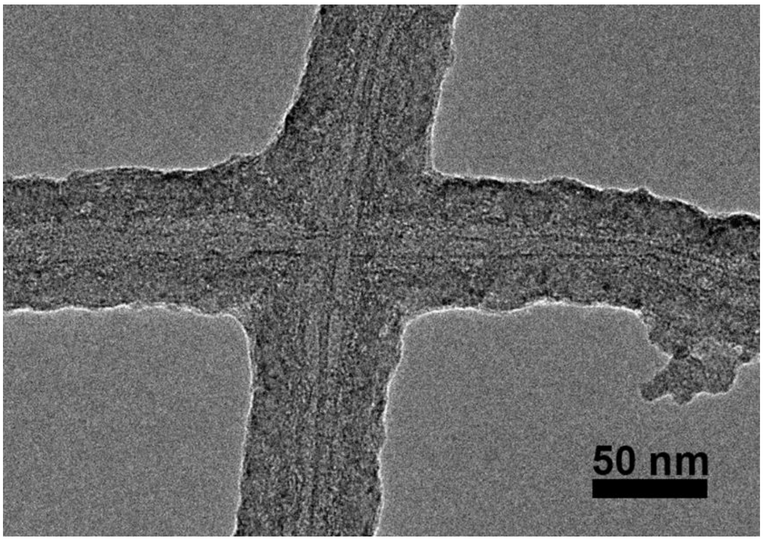

Figure S10 TEM image of two HNCNTs with a cross structure.

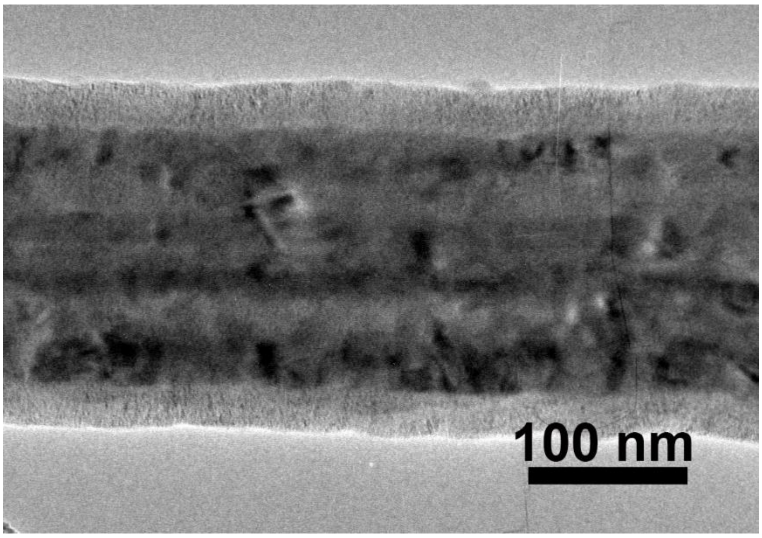

Figure S11. TEM image of an HNCNT with Matryoshka structure.

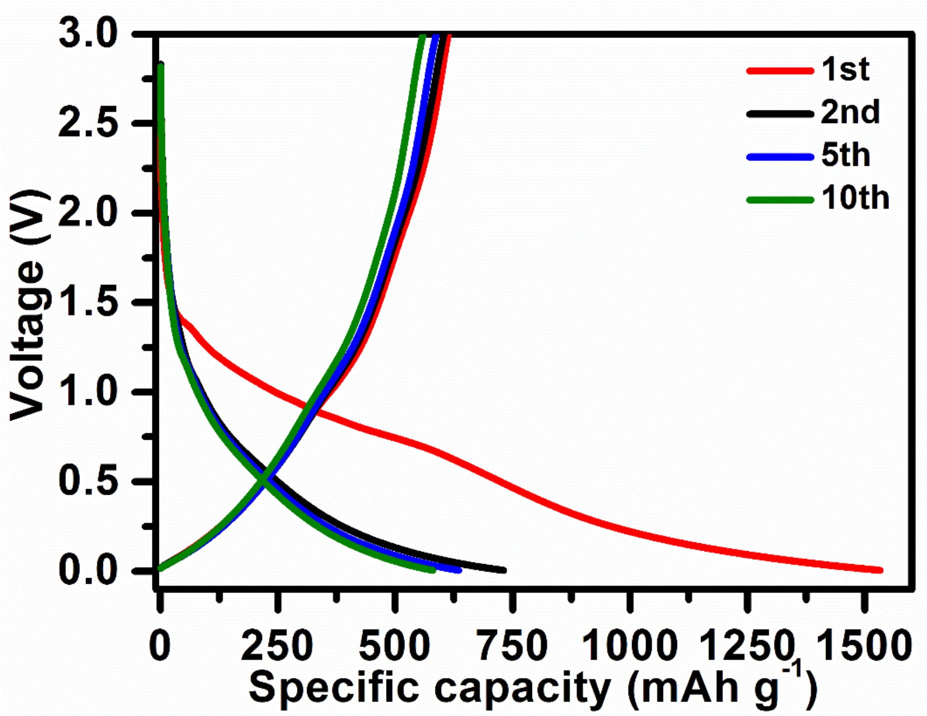

Figure S12. Charge-discharge curves of HNCNTs at different cycle numbers. The thicknesses of Al_2O_3 and nitrogen-doped graphene layer were 30 and 20 nm, respectively.

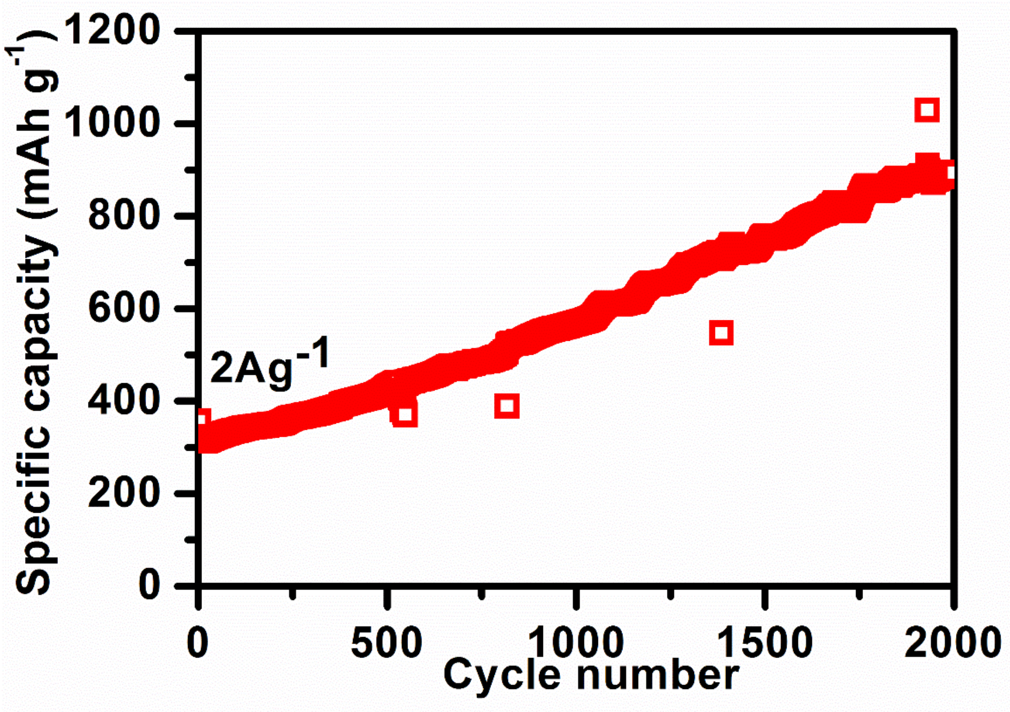

Figure S13. Specific capacitances based on HNCNTs with a nitrogen-doped graphene layer of 20 nm during 2,000 charge-discharge cycles.

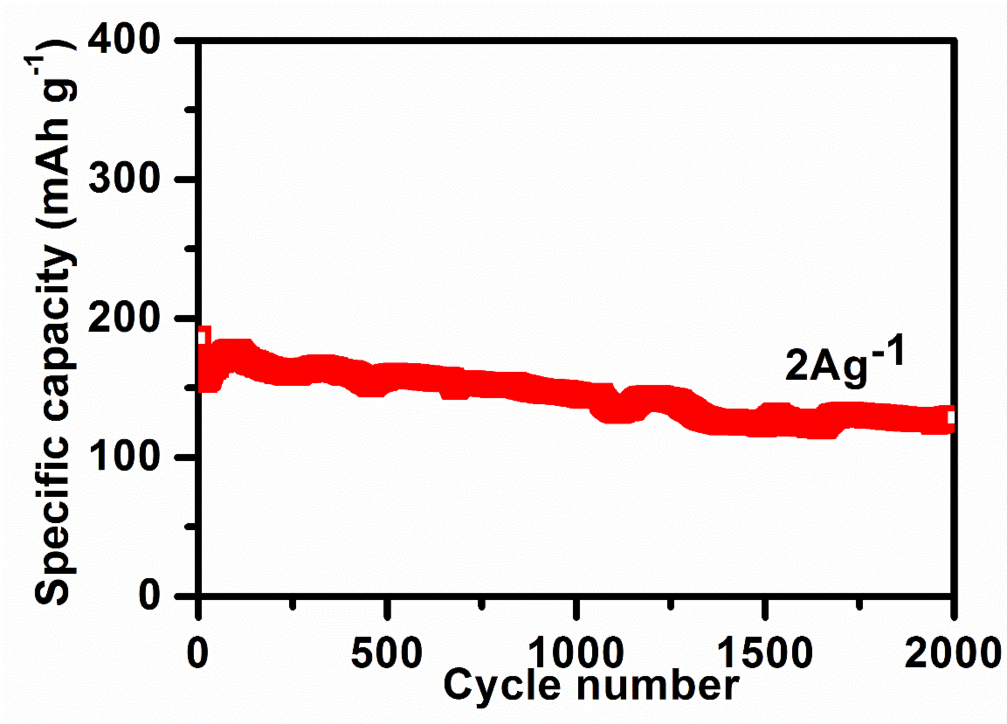

Figure S14. Specific capacitances based on HNCNTs with a nitrogen-doped graphene layer of 100 nm during 2,000 charge-discharge cycles.

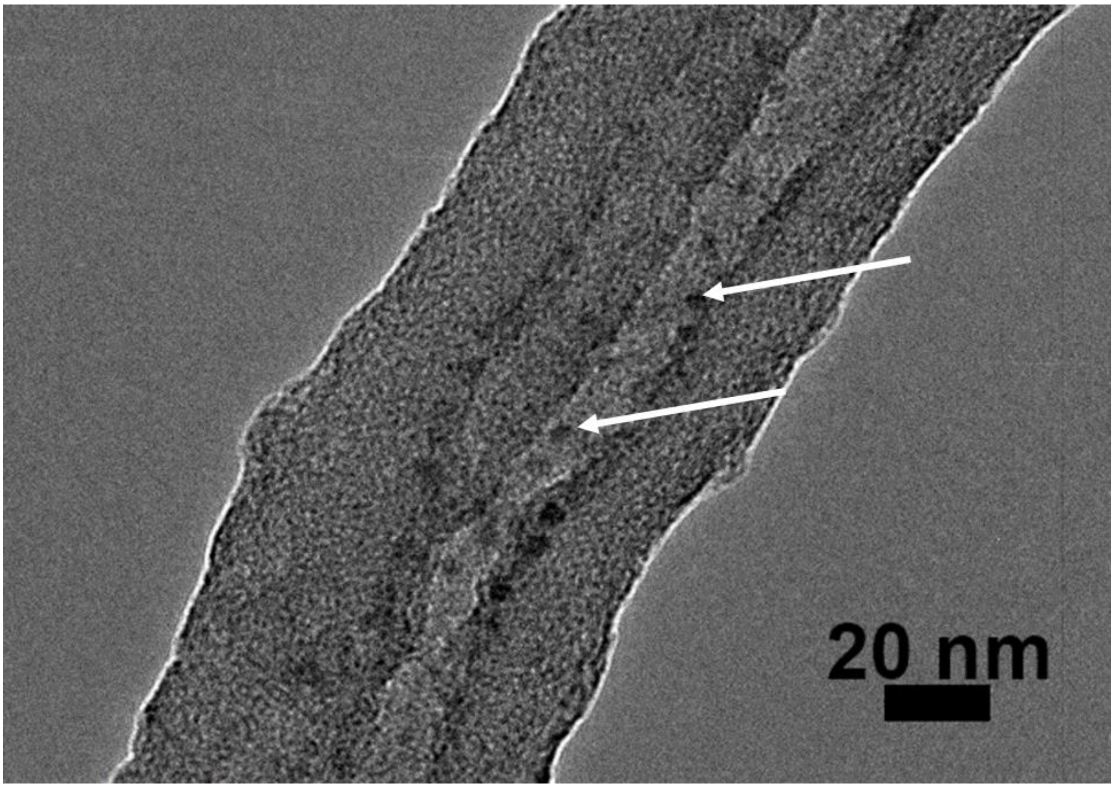

Figure S15. TEM image of an HNCNT after discharging.

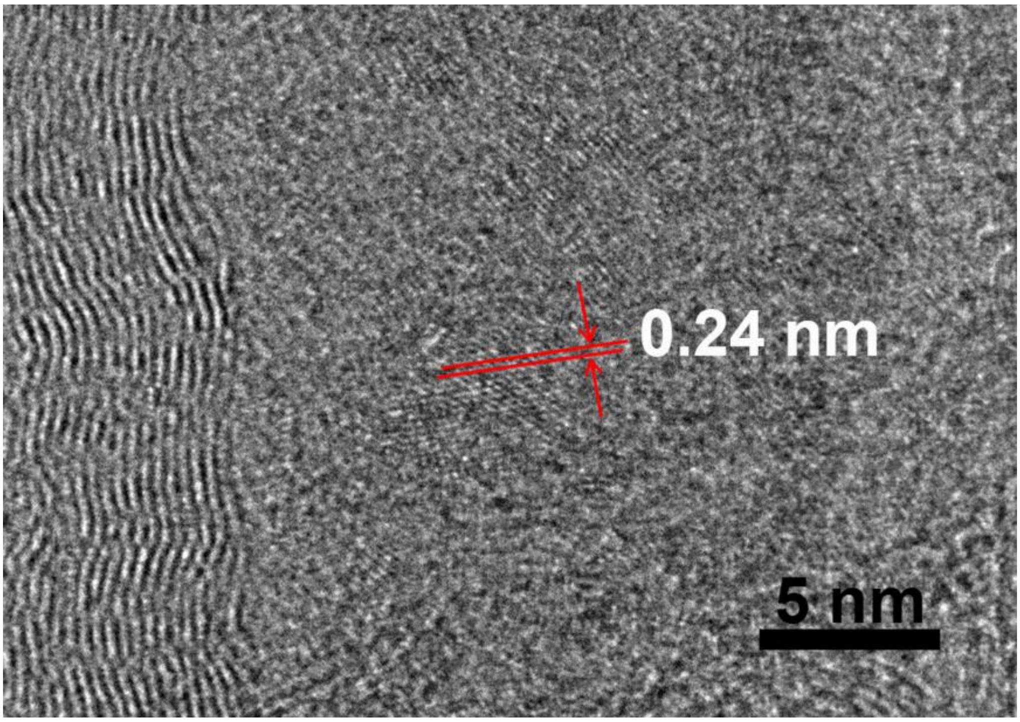

Figure S16. High-resolution TEM image of an HNCNT after discharging.

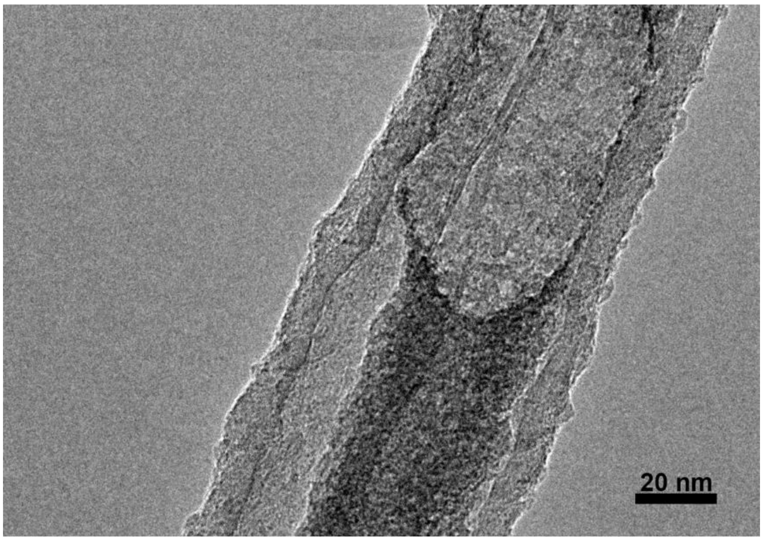

Figure S17. TEM image of Si-incorporated HNCNTs after etching by HF for 30 min. The thicknesses of Si and nitrogen-doped graphene layers were 30 and 10 nm, respectively.

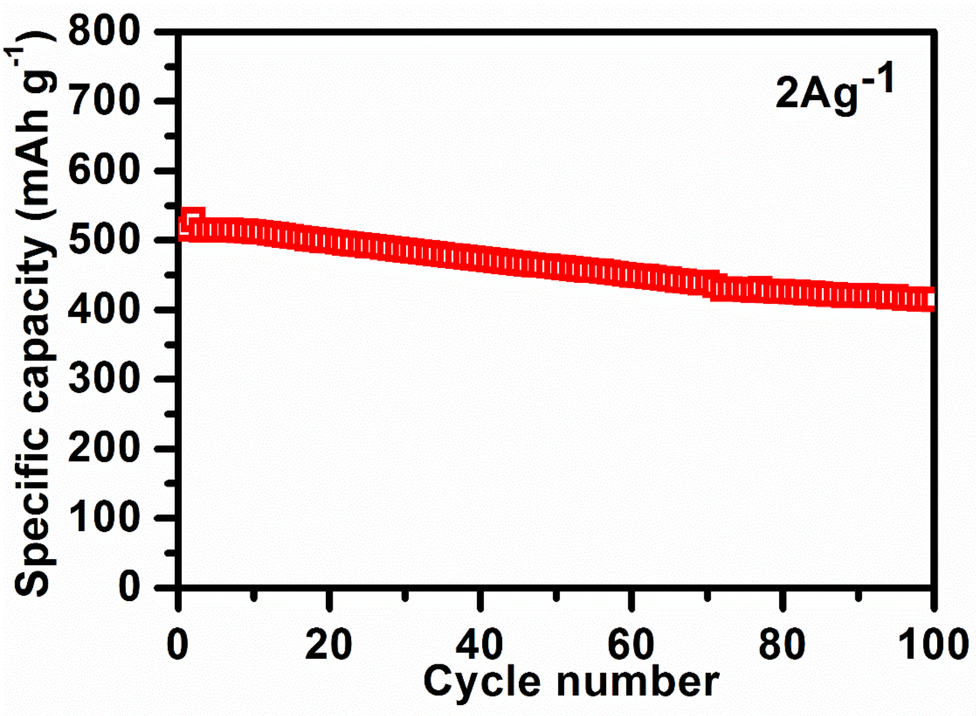

Figure S18. Specific capacitances of Si-incorporated HNCNTs during 2,000 charge-discharge cycles. The thicknesses of Si and nitrogen-doped graphene layers were 50 and 20 nm, respectively. Current density, 2 A g^{-1}.

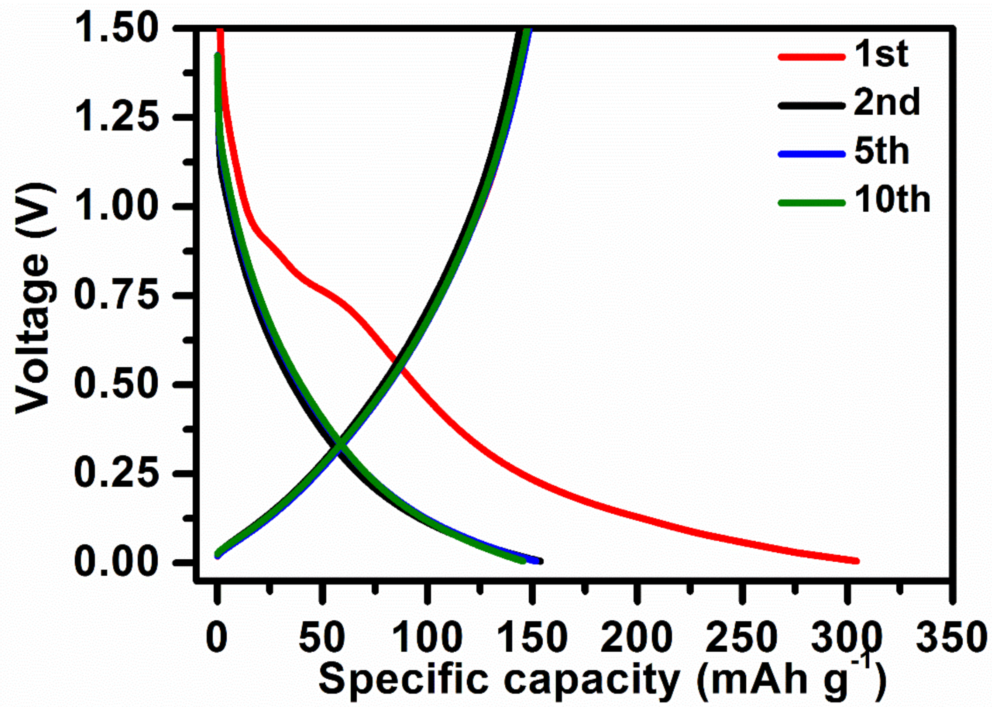

Figure S19. Charge-discharge curves of nitrogen-doped graphene/Si/CNT. The thicknesses of Si and nitrogen-doped graphene layers were 30 and 20 nm, respectively.

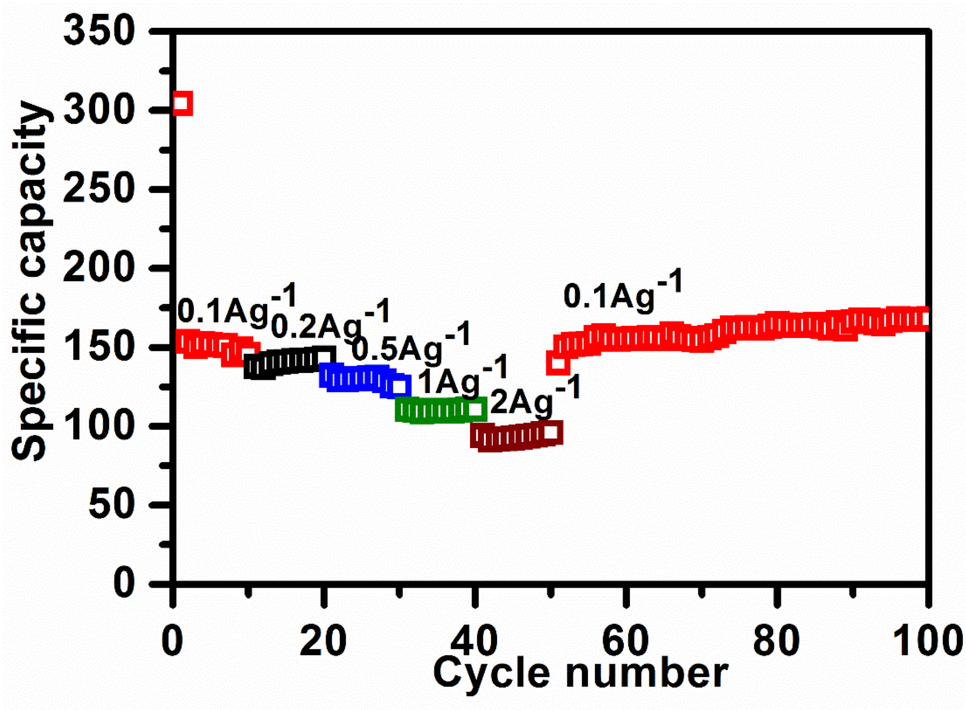

Figure S20. Rate performance of nitrogen-doped graphene/Si/CNT. The thickness of Si and nitrogen-doped graphene layers were 30 and 20 nm, respectively.

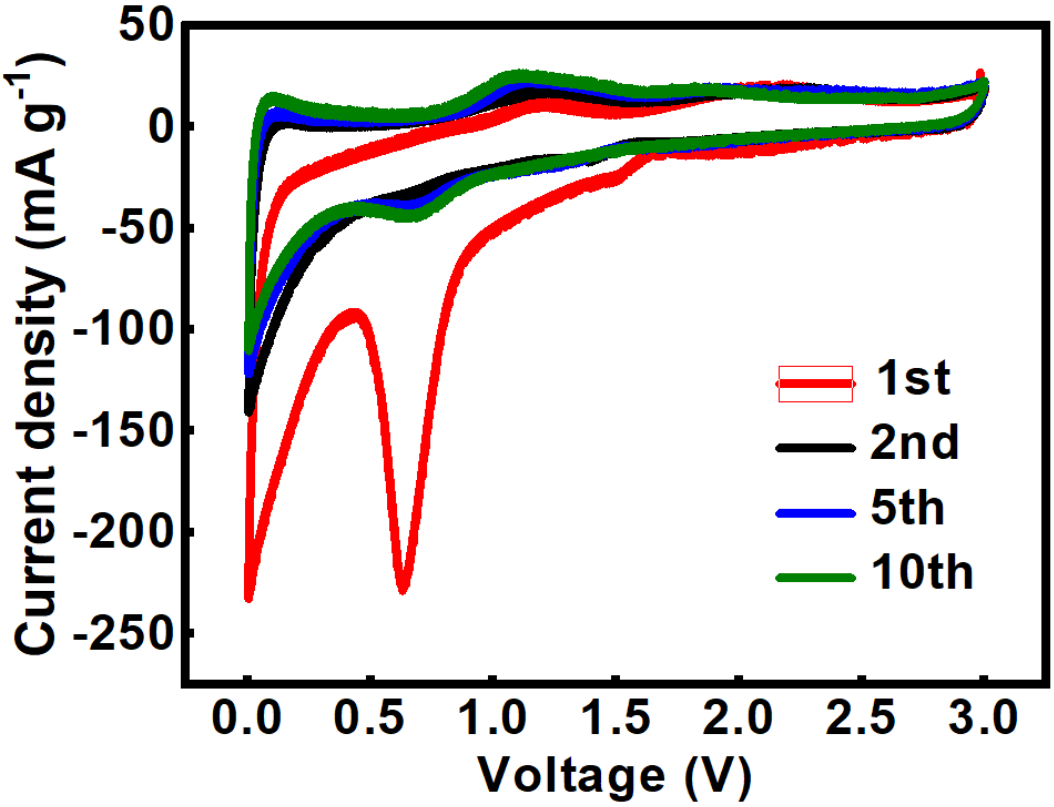

Figure S21. CV curves of CNTs at lower rate 0.2 mV s^{-1}. The coulombic efficiency was calculated by the ratio of areas of different cycles. 49% for the 1st cycle, 91% for the 2nd cycle, 95% for the 5th cycle and 94% for the 10th cycle.