ADVANCED ENERGY MATERIALS

Supporting Information

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Synthesizing Nitrogen-Doped Core–Sheath Carbon Nanotube Films for Flexible Lithium Ion Batteries

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Supporting Information

Experimental Section

Synthesis of carbon nanotube sheet. The spinnable carbon nanotube (CNT) array was synthesized by chemical vapor deposition in a tube furnace. Fe (1.2nm)/Al (3nm) on a silicon wafer was used as the catalyst. Argon (400 sccm) and hydrogen (30 sccm) were mixed as the carrying gas and ethylene served the source of carbon. The growth temperature and time were 740 ^{o}C and 10 min, respectively. An aligned CNT sheet was pulled out of the CNT array and then attached onto a silicon wafer with the two ends fixed. The CNT sheets from 1 to 20 layers had been prepared.

Characterization. The structures were characterized by scanning electron microscopy (SEM, Hitachi FE-SEM S-4800 operated at 1 KV), transmission electron microscopy (TEM, JEOL JEM-2100F operated at 200 KV) and X-ray diffraction (Bruker AXS D8). The electrochemical performances were measured by an Arbin electrochemical station (MSTAT-5V/10mA/16Ch). The mechanical measurements of the stretchable lithium ion battery (LIB) were performed at HY-0350, Shanghai Hengyi Testing Instruments Co. LTD.

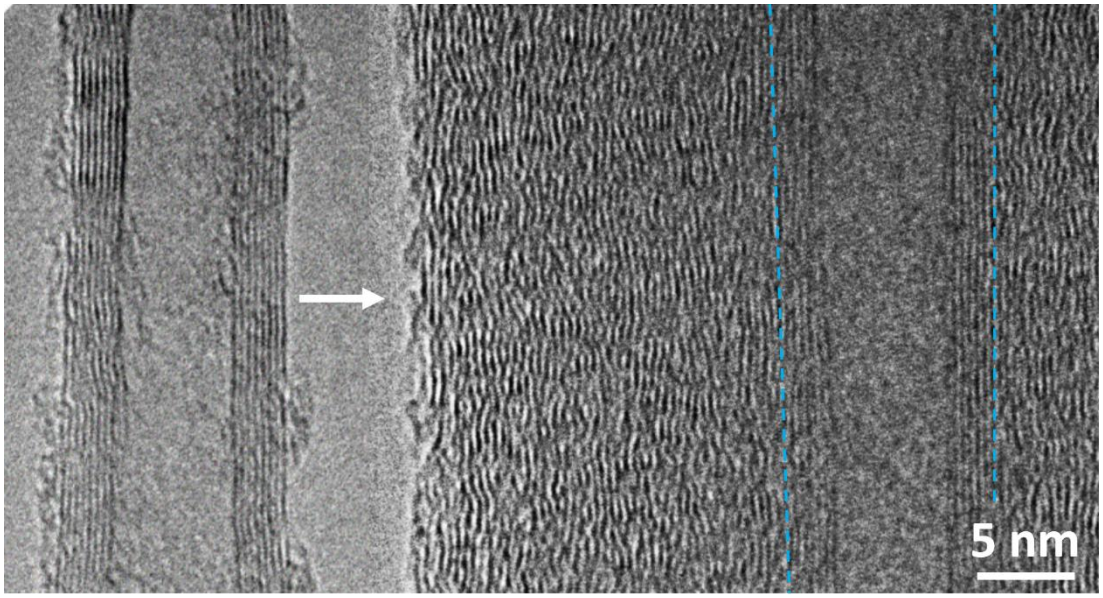

Figure S1. High-resolution TEM images of the CNT (left) and N-CNT (right). The blue lines indicate the boundary between the origin CNT wall and the new grown N-doped graphene layer.

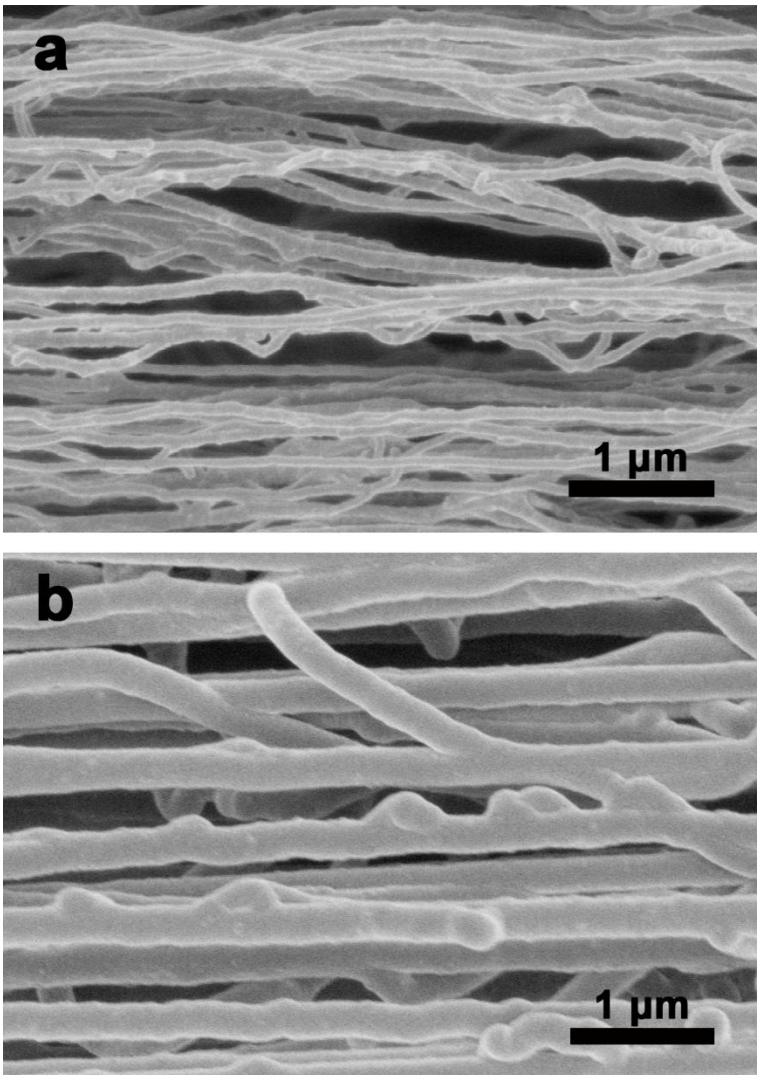

Figure S2. SEM images of aligned N-CNT films with growth times of 30 min (**a**) and 90 min (**b**).

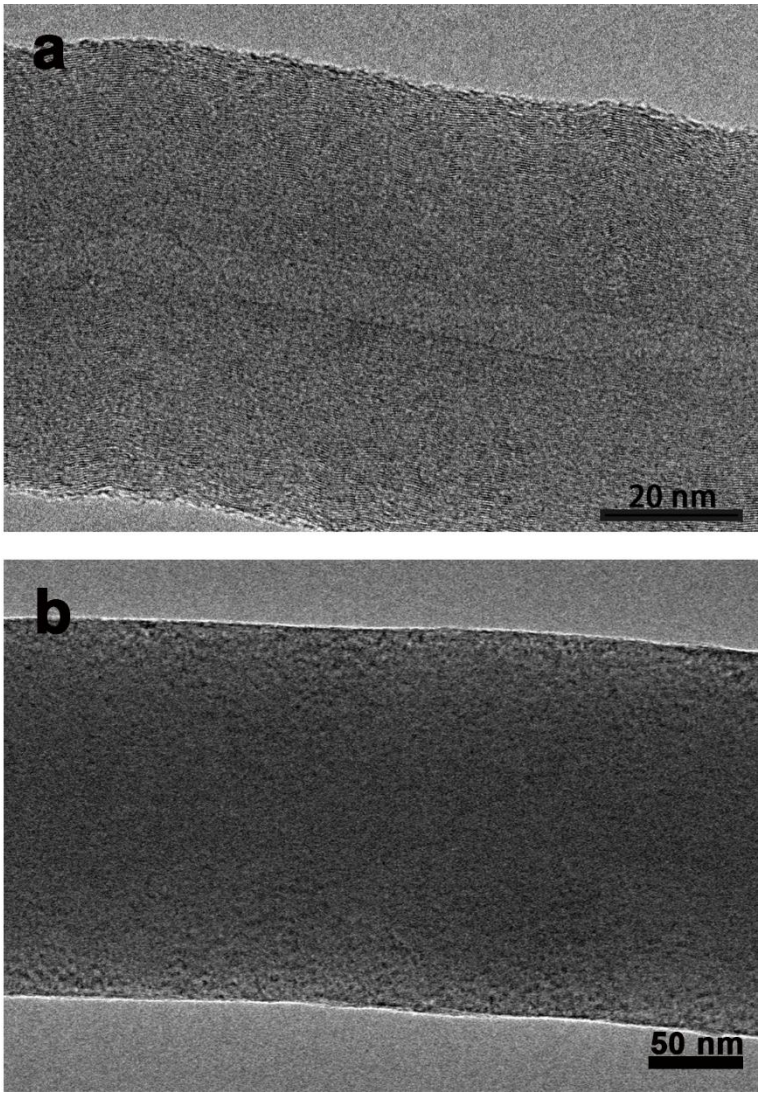

Figure S3. TEM images of N-CNTs with growth times of 30 min (**a**) and 90 min (**b**).

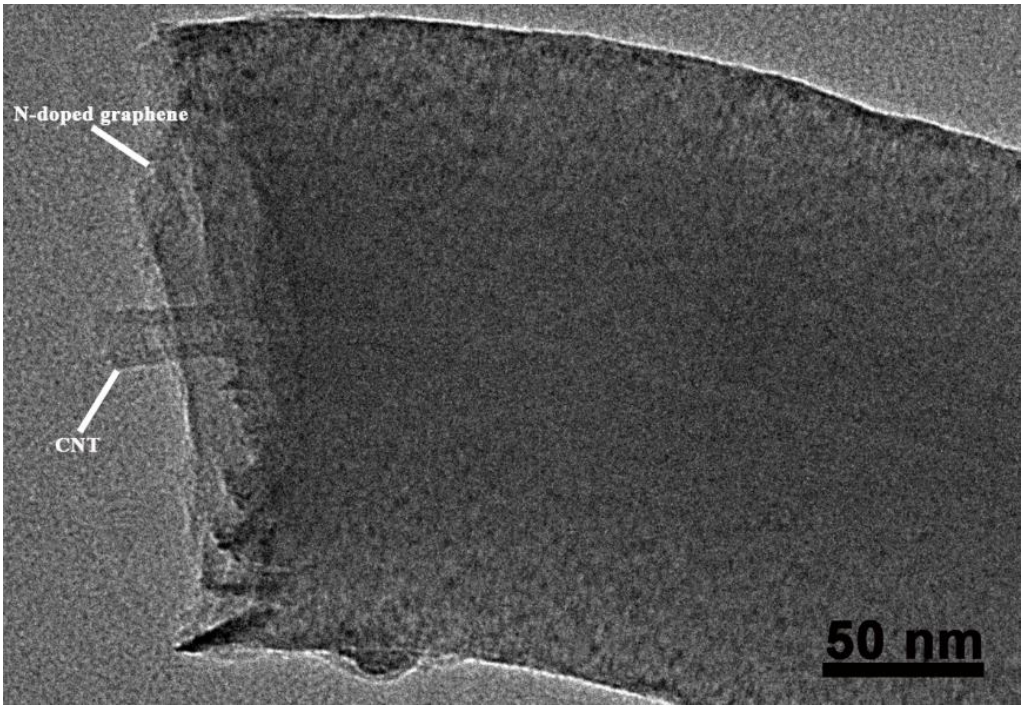

Figure S4. A core-sheath structure of the N-CNT with growth time of 60 min.

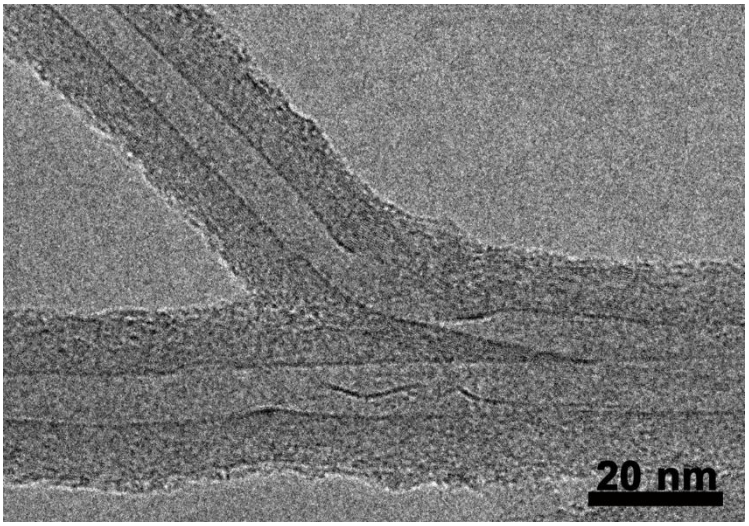

Figure S5. A Y-structure of the N-CNT with growth time of 10 min.

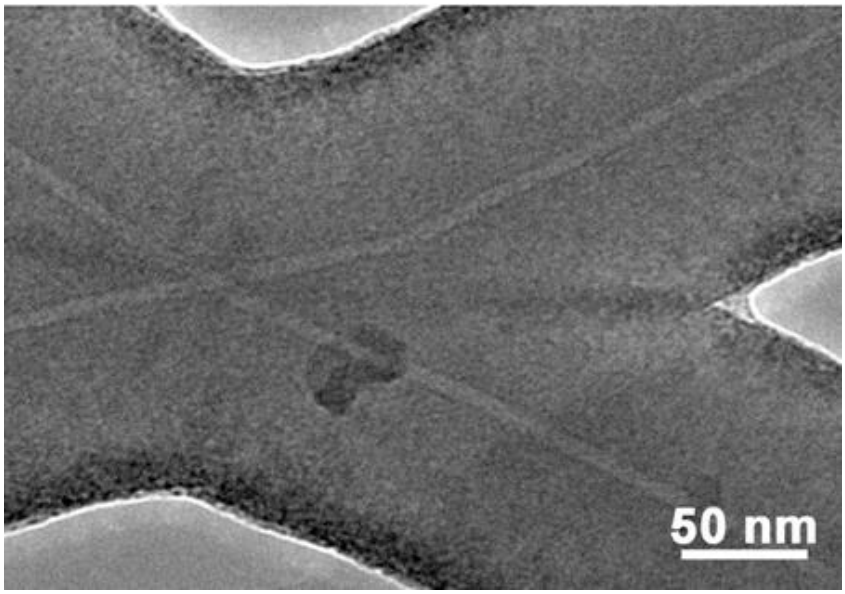

Figure S6. An X-structure of the N-CNT with growth time of 90 min.

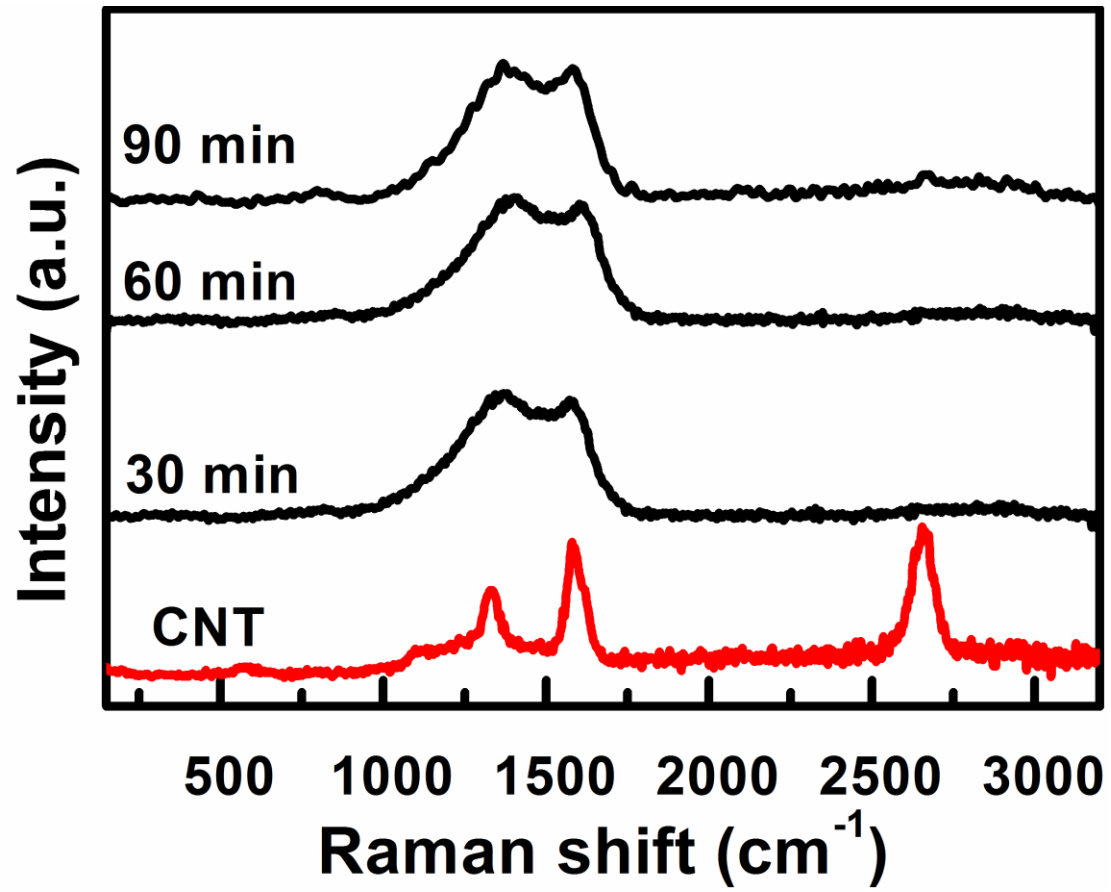

Figure S7. Raman spectra of CNTs and N-CNTs with increasing growth times.

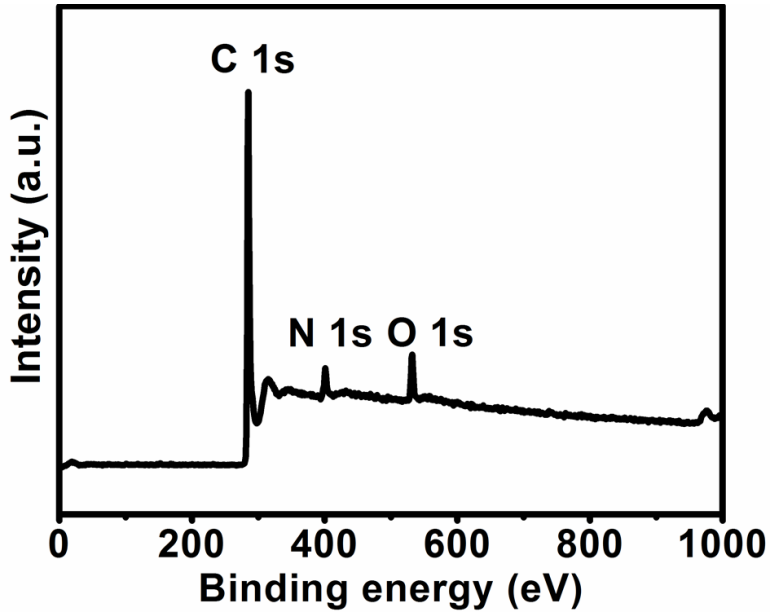

Figure S8. N 1s XPS spectrum of the N-CNT.

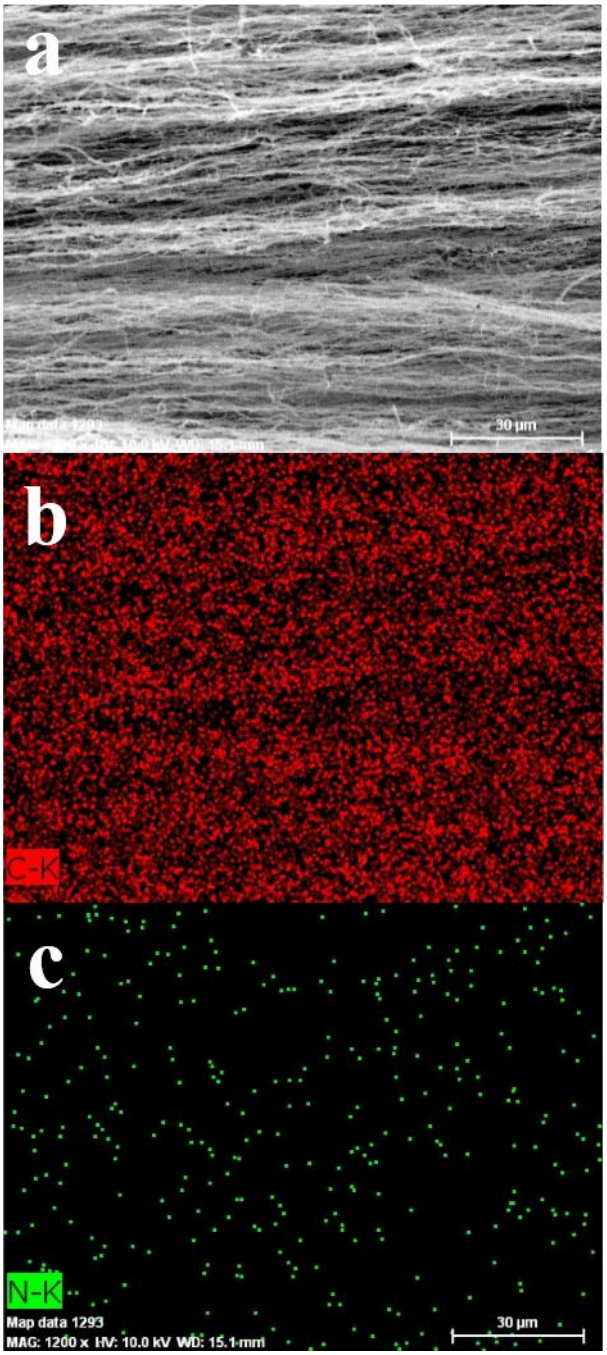

Figure S9. Energy-dispersive X-ray spectroscopy images of the aligned N-CNT sheet. (**a)** SEM image of the aligned N-CNT sheet. (**b**) The dispersion of C element. (**c**) The dispersion of N element.

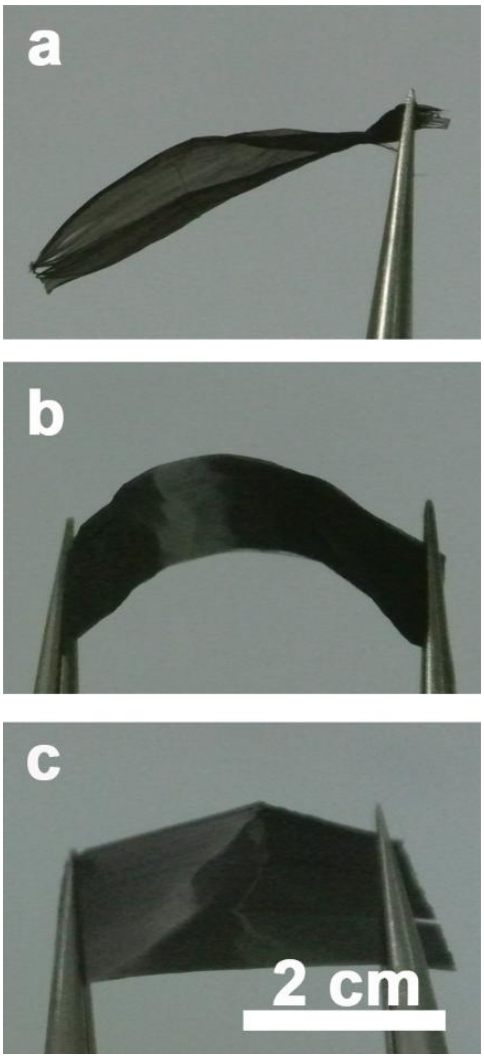

Figure S10. Photographs of CNT sheet (**a**) and N-CNT films with growth times of 60 min (**b**) and 90 min (**c**).

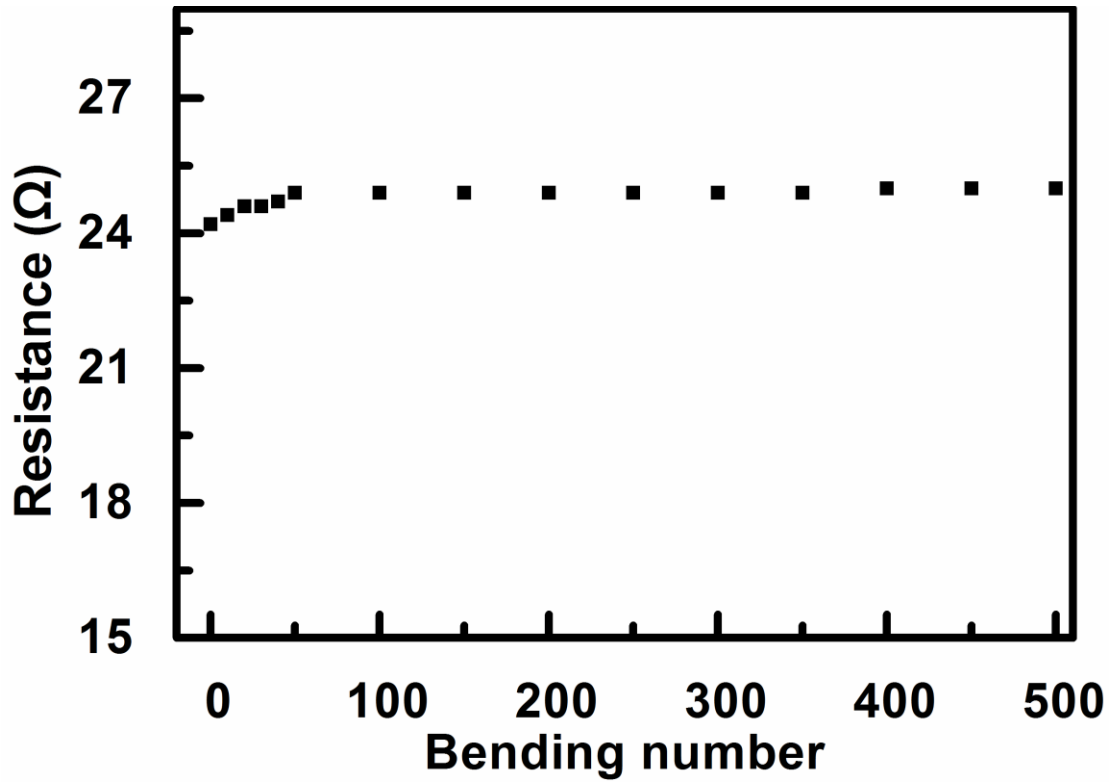

Figure S11. The electrical resistance along the aligned direction of the N-CNT film.

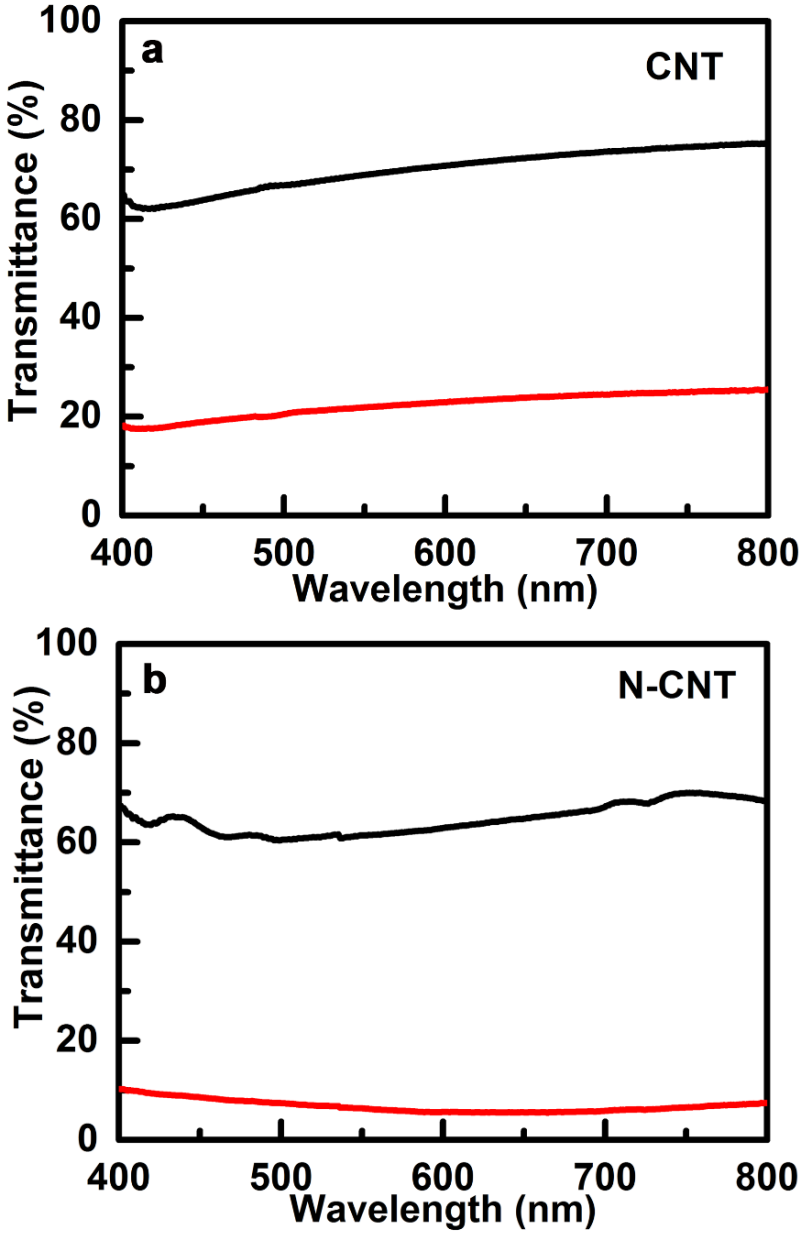

Figure S12. UV-vis transmittance spectra at being parallel (black lines) and perpendicular (red lines) to the nanotube-aligned direction. **a.** CNT sheet. **b.** N-CNT film with a growth time of 60 min.

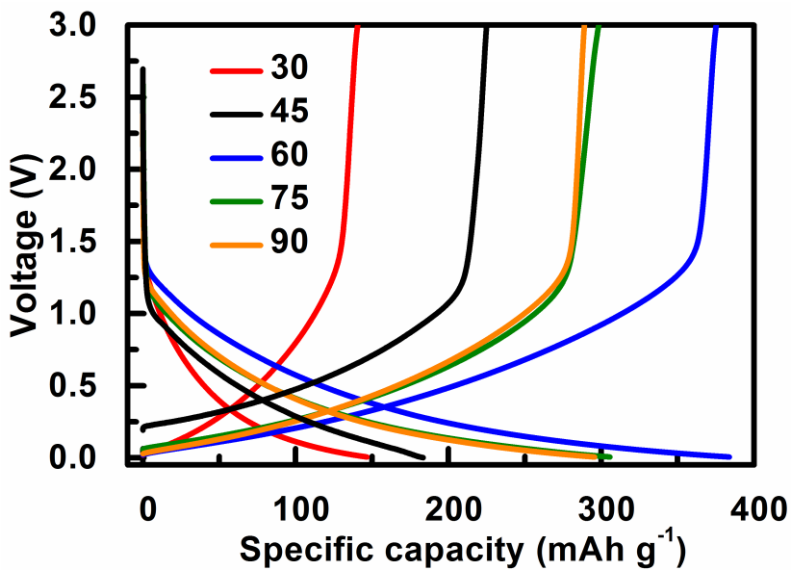

Figure S13. Galvanostatic charge and discharge curves of N-CNT films grown from 10 layers of CNT sheets with increasing growth times of 30, 45, 60 and 75 to 90 min at 0.1 $A \cdot g^{-1}$.

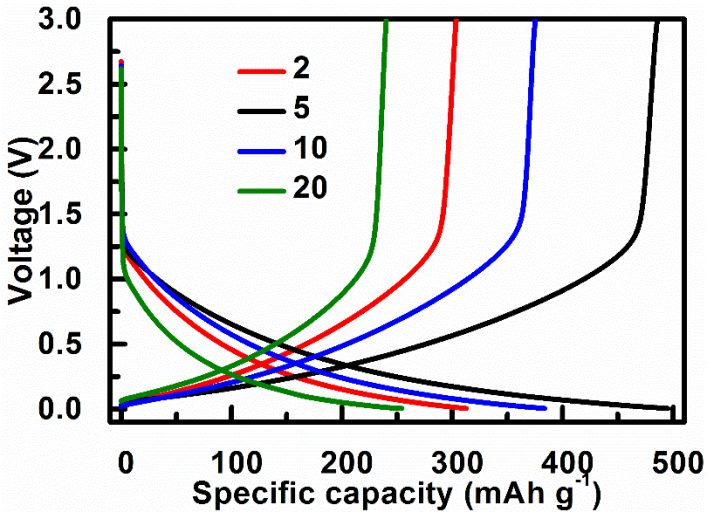

Figure S14. Galvanostatic charge and discharge curves of N-CNT films grown from CNT sheets with 2, 5, 10 and 20 layers at 0.1 $A \cdot g^{-1}$. They were grown at the same time of 60 min.

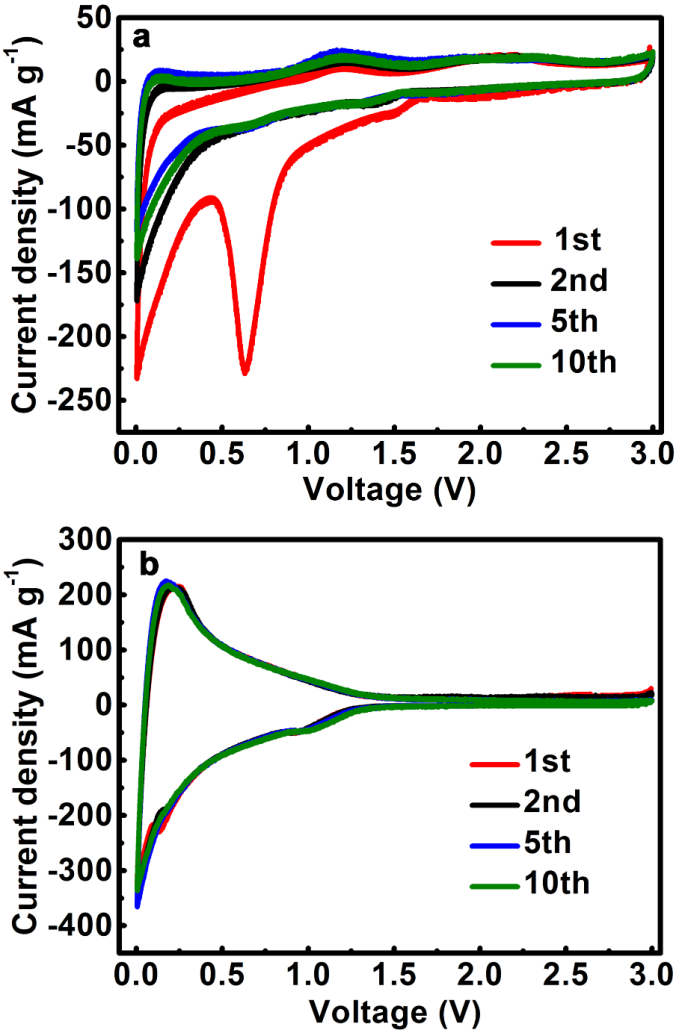

Figure S15. Cyclic voltammograms of CNT sheet (**a**) and N-CNT film (**b**) at a scan sate of 0.05 $mV \cdot s^{-1}$. The N-CNT film was synthesized from 5 layers of CNT sheets in 60 min.

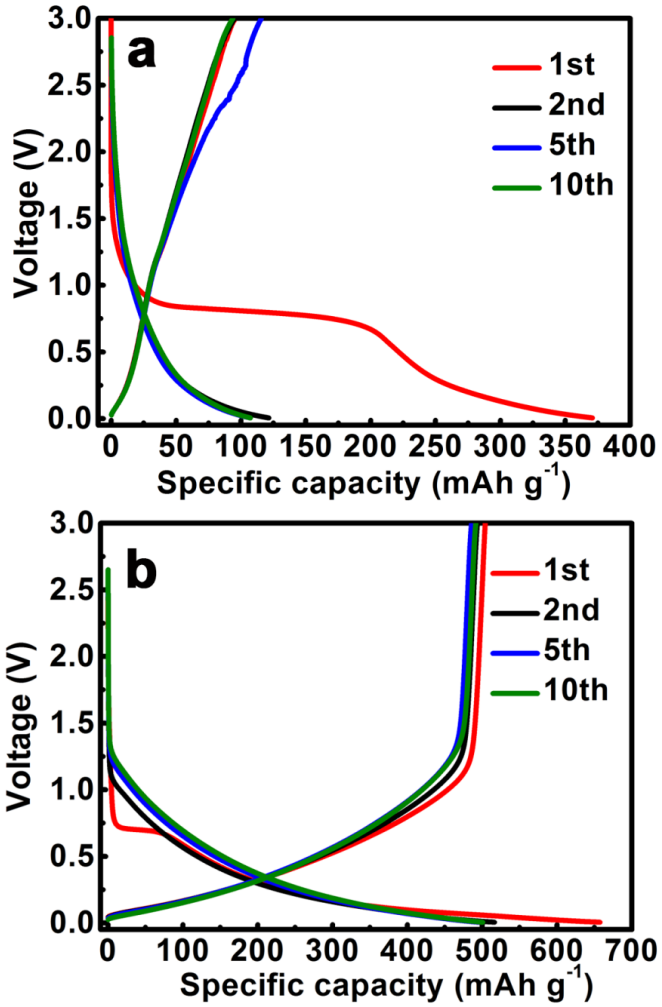

Figure S16. Galvanostatic charge/discharge curves of CNT sheet (**a**) and N-CNT film (**b**) at a current density of 0.1 $A \cdot g^{-1}$.

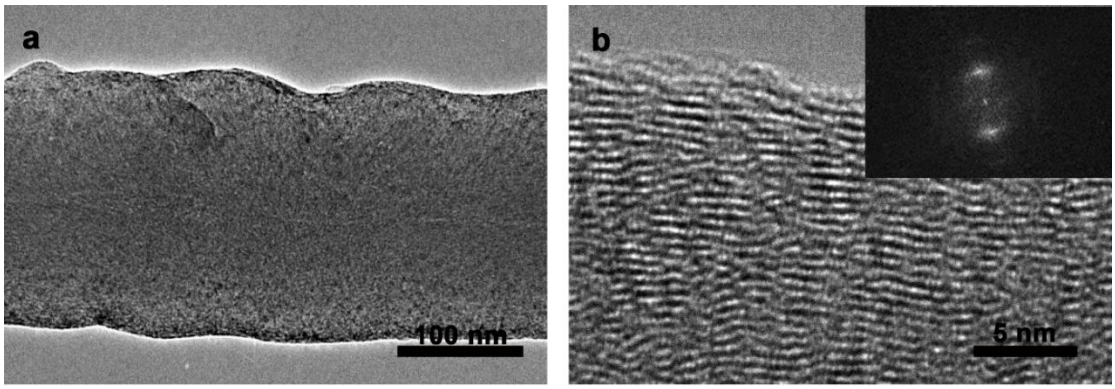

Figure S17. An N-CNT before lithiation at low (**a**) and high (**b**) magnifications.

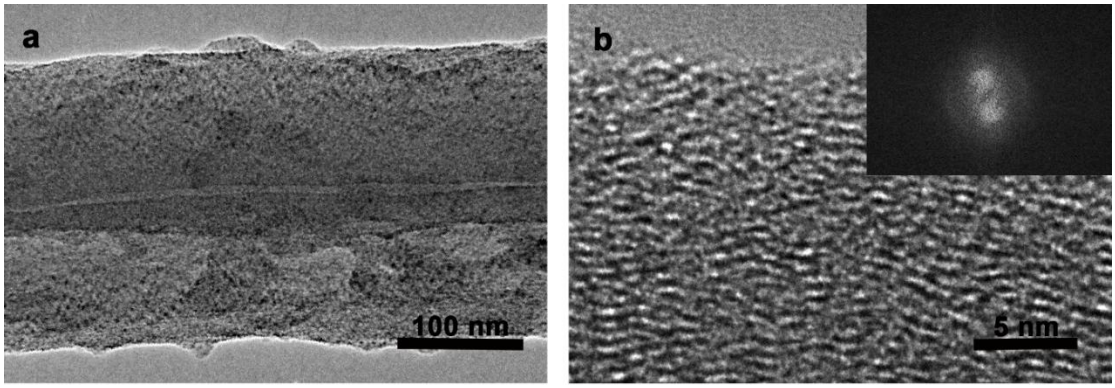

Figure S18. An N-CNT after lithiation at low (**a**) and high (**b**) magnifications.

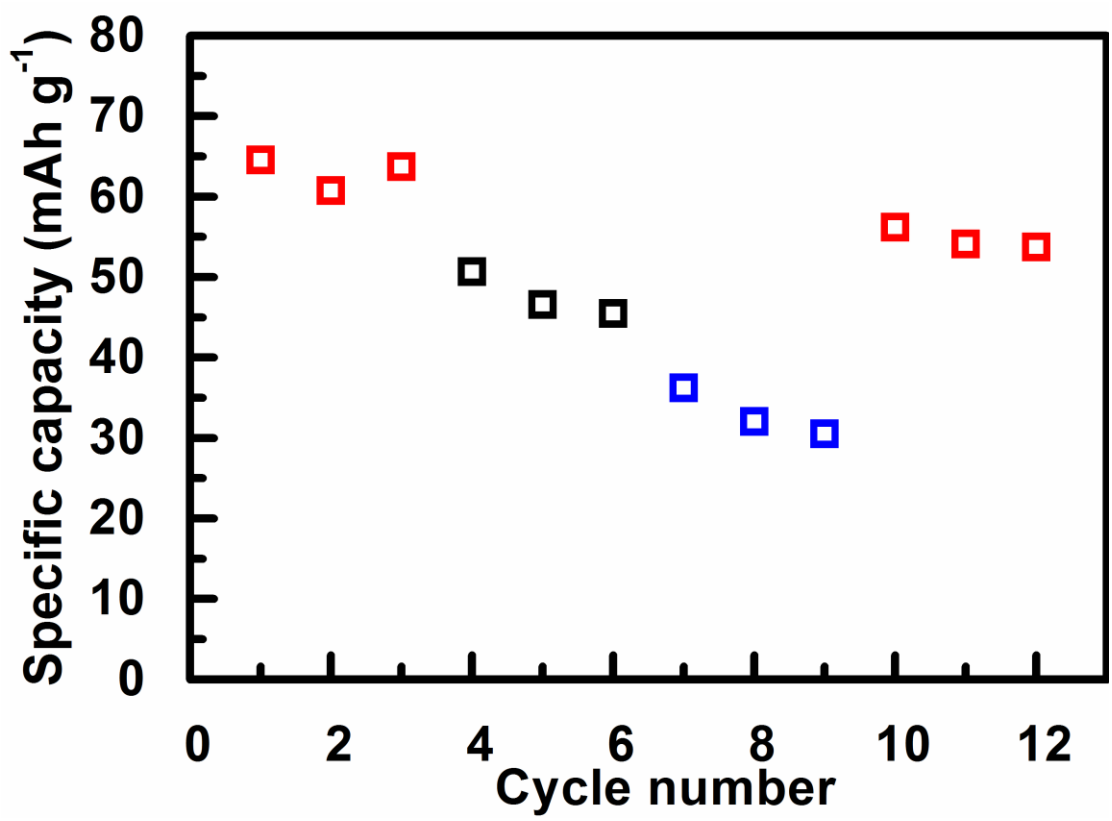

Figure S19. The rate capability of the full batttery tested with currents of 0.05 mA, 0.1 mA, 0.25 mA and the back to 0.05 mA.

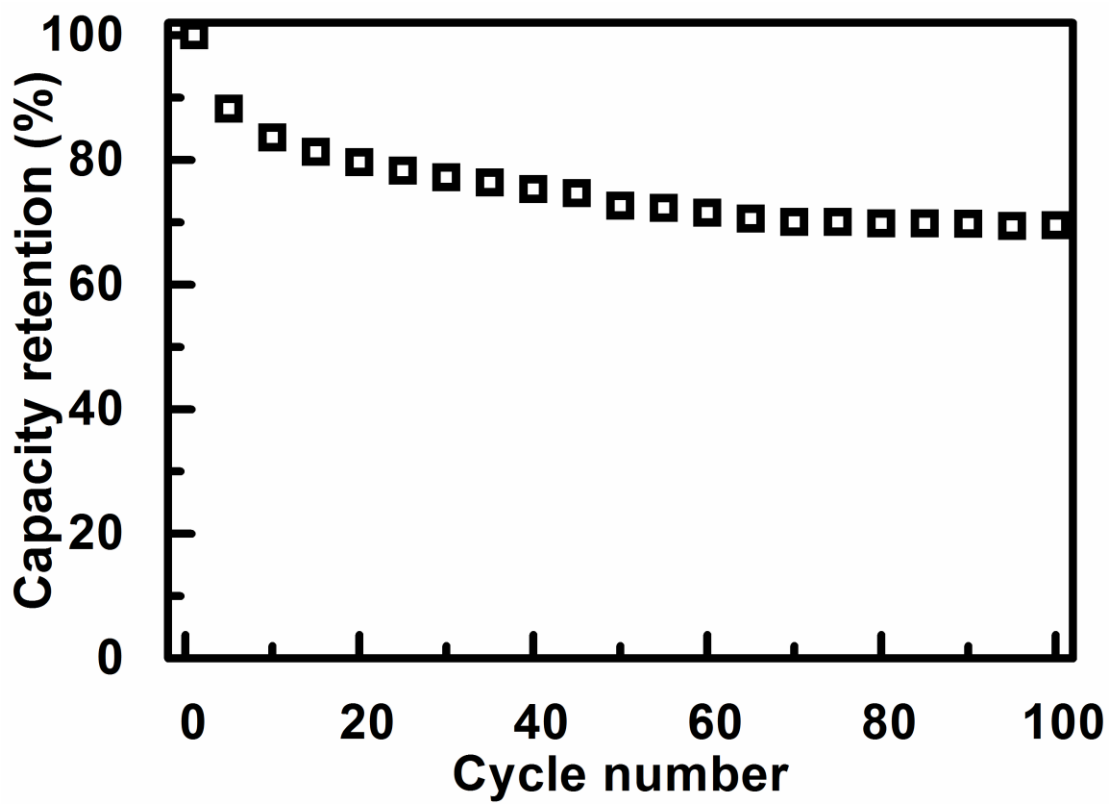

Figure S20. The capacity retention of the full battery.

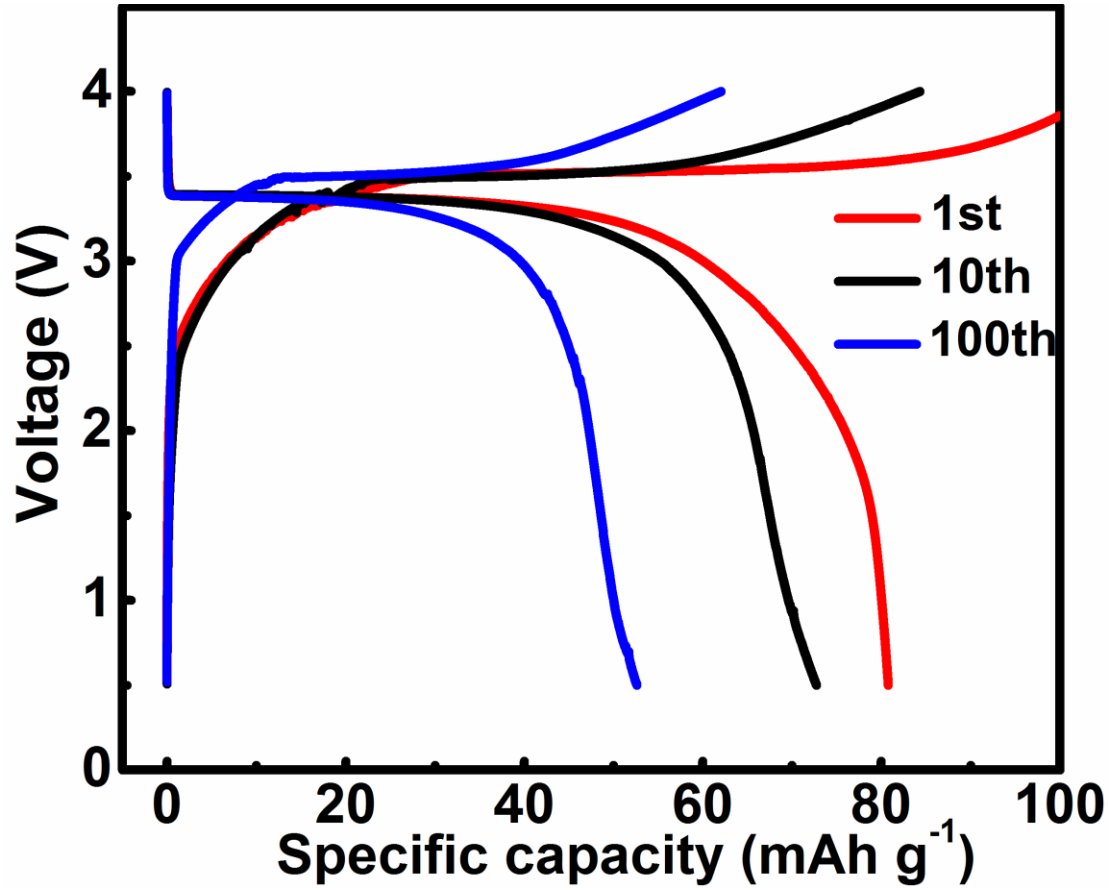

Figure S21. Charge-discharge curves of the full battery after bending for 1, 10 and 100 times.

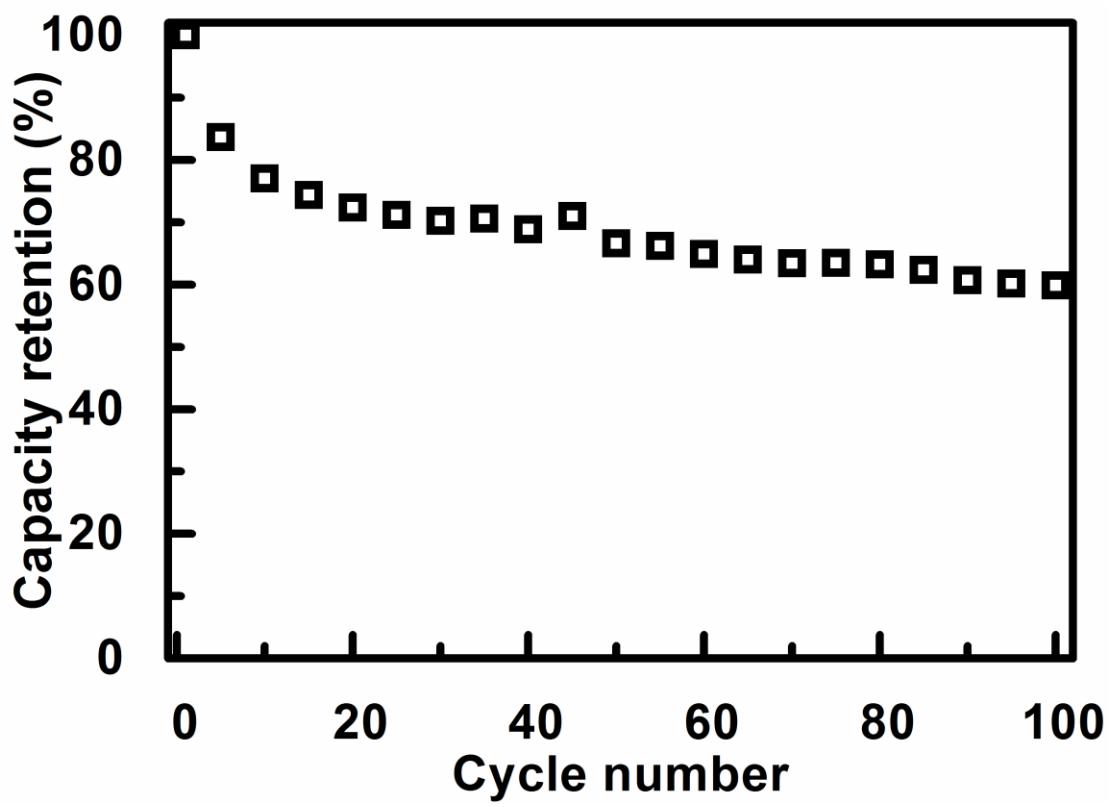

Figure S22. Dependence of the capacity retention of the full battery on bending cycle number.