Supporting Information

Ultrastretchable Carbon Nanotube Composite Electrodes for Flexible Lithium-Ion Batteries

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Experimental Section

Preparation of the CNT film and the PDMS substrate:

CNT arrays with a diameter of 20–30 nm and a height of 300 μm were synthesized on silicon wafers by chemical vapor deposition. Continuous CNT films were directly drawn from the CNT arrays. Details of the experimental method can be found in previous papers[1, 2]. A PDMS substrate with a thickness of 1 mm was prepared by mixing the base and the curing agent at a weight ratio of 10:1 using Sylgard 184 (Dow Corning). The mixture was cured at 70 °C for 1 h. After cooling down to room temperature, the PDMS substrate was cut into 100 mm × 100 mm pieces.

Fabrication of the stretchable CNT composite electrodes:

First, a PDMS substrate with a length *L* was biaxially pre-strained in two perpendicular

directions to $L+\Delta L$, followed by coating of a cross-stacked 6-layer CNT film on the surface of the pre-strained PDMS substrate. $Li(Ni_{1/3}Co_{1/3}Mn_{1/3})O_2$ (NCM, Kejing Star Technology Co., Shenzhen, China) or $Li_4Ti_5O_{12}$ (LTO, Reshine, China) powder was dispersed in ethanol by ultrasonication for 45 minutes using an ultrasonic cell crusher (Scientz Co., Ningbo, China). The suspensions were dropped on the CNT film with a pipette, followed by cross-stacking a 2-layer CNT film. After the evaporation of ethanol, the dropping of the suspension and the stacking of the CNT film were repeated 10 times. Finally, another cross-stacked 6-layer CNT film was put on the top to form a sandwiched structure. After releasing the pre-strain of the PDMS substrate to its original length L, CNT composite electrodes were fabricated. The dimensions of the CNT/NCM and CNT/LTO electrodes are 12 mm×20 mm and 10 mm×20 mm, respectively, in order to make sure that the CNT/NCM electrode fully contacts with the gel electrolyte during the full battery assembly process.

Preparation of the gel electrolyte and assembly of the stretchable full batteries:

The gel electrolyte was prepared by dissolving 0.35 g poly (ethylene oxide) (PEO, Mw~60,000), 0.35 g succinonitrile, and 0.30 g lithium bis(trifluoromethane) sulfonimide (LiTFSi) in 50 mL mixed solvent of methylene chloride and acetone at a weight ratio of 40:1. The full batteries were assembled in an argon-filled glove box (M. Braun Inert Gas Systems Co. Ltd). As shown in Figure 6, a 0.6 mm thick PDMS film with a rectangular hole (10 mm×20 mm) was stacked on the anode, forming a template slot for the gel electrolyte. The gel electrolyte was dropped into the slot and then dried at room temperature for 24 h. Afterwards, the CNT/NCM cathode was stacked on the gel electrolyte. Uncured PDMS paste was used to seal the full battery. After curing of the PDMS paste, a stretchable full battery was obtained. The thickness of the whole device is 2.6 mm.

Characterization:

The structures of the CNT/NCM and CNT/LTO electrodes were characterized by X-ray diffraction (XRD) using a diffractometer (Rigaku). The resistances of the CNT/LTO and CNT/NCM electrodes at strain up to 150% were measured. The dimensions of the sample are 70 mm × 10 mm × 0.6 mm. Two Cu foils were attached at both ends of the sample by a conductive silver paste for resistance measurements. A tensile strain was applied by using an Instron 5848 microtester and the resistance of the CNT composite electrodes was monitored by a Keithley 2400 Source Meter. The durability of the CNT composites was characterized by performing 2,000 cyclic tensile tests at 0-150% strain and the resistance of the sample was measured at each stretching cycle. The morphologies of the CNT composite electrodes were examined by SEM before and after the cyclic tensile testing. Half-cells were used to measure the electrochemical properties of the CNT/NCM and CNT/LTO electrode. The CNT/NCM and CNT/LTO composites were cut into several pieces and then were stretched for different times. After cyclic tensile loading, the CNT composite electrodes were coated with porous polymer films (Celgard 2400, USA) and lithium foils to form half-cells. 1 M LiFP6 solution in ethylene carbonate and diethyl carbonate mixed at weight ratio of 1:1 was used as the electrolyte. The electrochemical properties of the electrodes were tested using a Land battery test system (Wuhan Land Electronic Co., China) with a cut-off voltage window of 2.8-4.3V for the CNT/NCM electrode and 1.0-2.5V for the CNT/LTO electrode. The voltage window of the full battery was 1.6-2.8V.

Notes and References

1. K. Jiang, J. Wang, Q. Li, L. Liu, C. Liu and S. Fan, *Advanced Materials*, 2011, **23**, 1154-1161.
2. K. Liu, Y. Sun, P. Liu, J. Wang, Q. Li, S. Fan and K. Jiang, *Nanotechnology*, 2009, **20**, 335705.

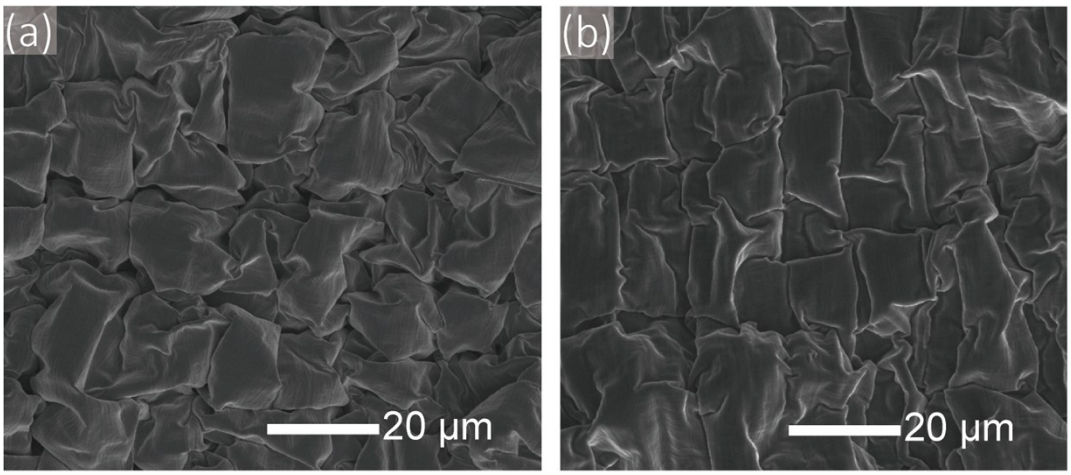

Figure S1. SEM images of the CNT/NCM electrode (a) before and (b) after 2,000 tensile cycles at 0-150% strain.

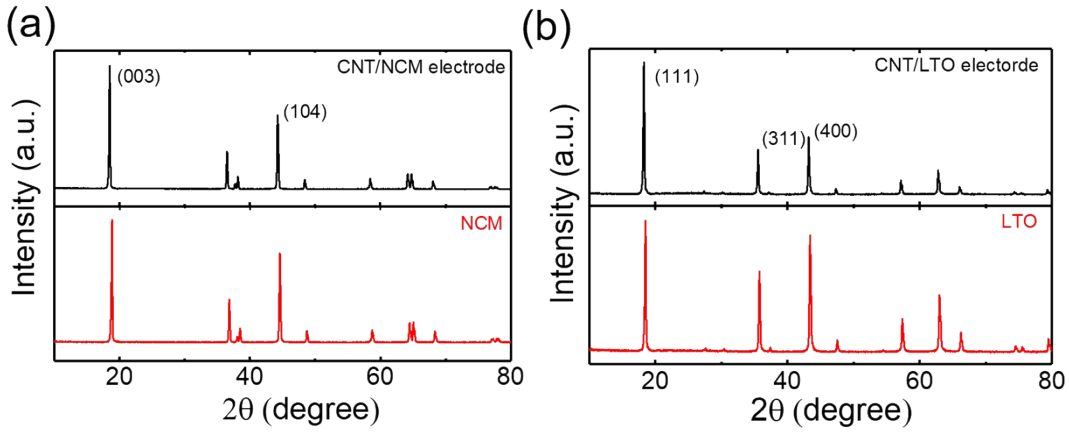

Figure S2. XRD patterns of (a) the NCM powder and CNT/NCM electrode and (b) the LTO powder and CNT/LTO electrode.

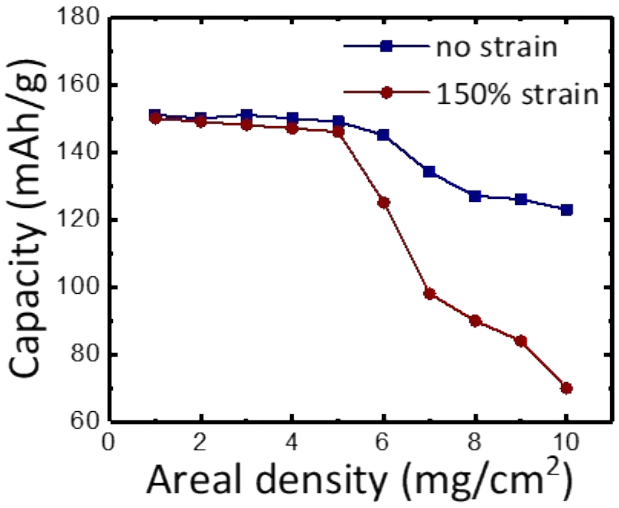

Figure S3. Specific capacities of the LTO/CNT electrode as a function of areal density of LTO at zero strain and at 150% strain.

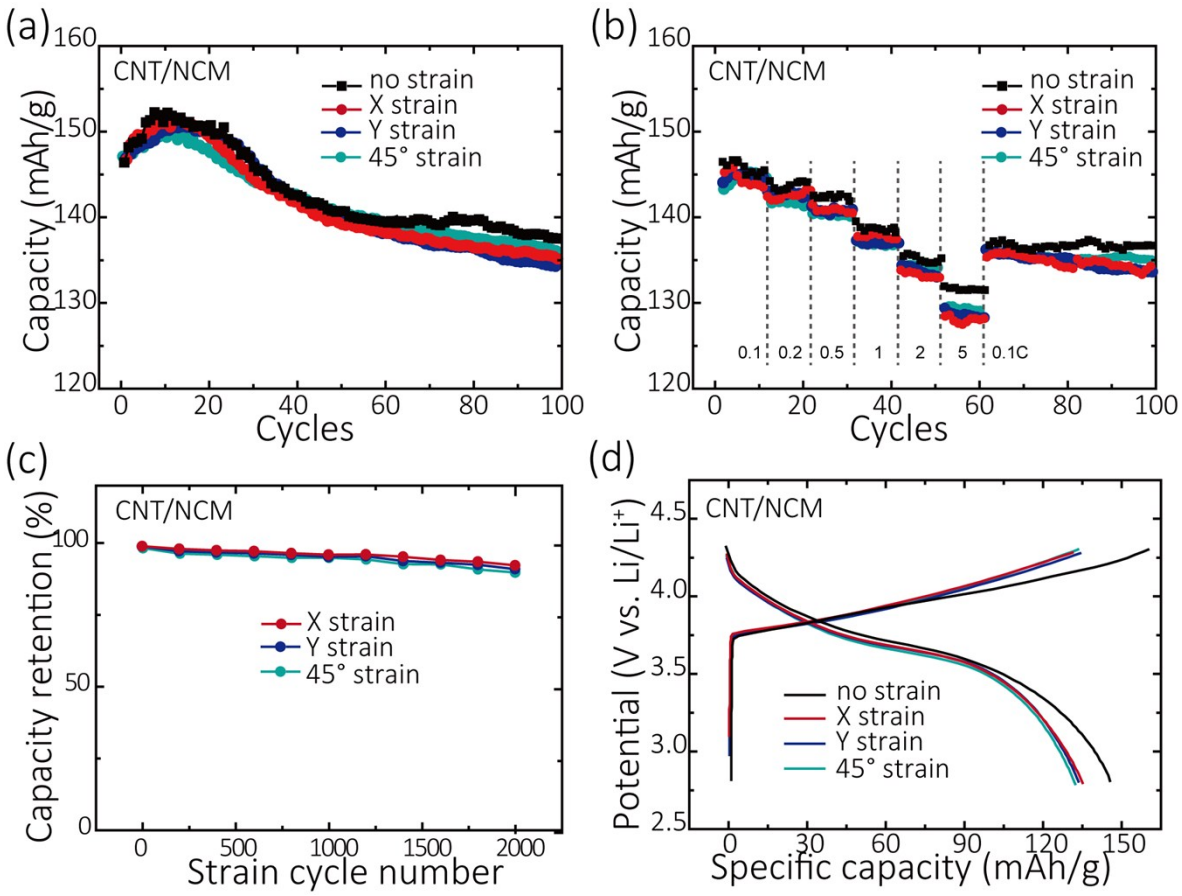

Figure S4. (a) Cycling and (b) rate performances of the CNT/NCM electrode at zero strain and at 150% strain in different axes; (c) voltage profiles and (d) capacity retention of the CNT/NCM electrode after 2,000 tensile cycles at 0-150% strain in different axes.