

## Supplementary Information for

# Self-weaving CNT -LiNbO<sub>3</sub> nanoplate-polypyrrole hybrid as a flexible anode for Li-ion batteries

## Experimental section

### 1. Synthesis

In a typical synthesis, CNTs were well dispersed into the dimethylformamide/water solution with the dispersant rhodanineacetic acid-pyrene (RAAP) added. As the strong  $\pi$ - $\pi$  stacking force between the RAAP and CNTs, the RAAP molecules were well decorated on the CNTs. Due to their strong hydrophilicity, the CNTs can be easily separated from each other and the surface of them became negative charged. Then (NH<sub>4</sub>)<sub>3</sub>NbO (C<sub>2</sub>O<sub>4</sub>)<sub>3</sub> and CH<sub>3</sub>COOLi solution were added into such dispersion drop by drop. The carboxyl groups of RAAP on the surface of CNTs supply nucleation sites for the formation of LiNbO<sub>3</sub> nanoplates. Simultaneously, CNTs were self weaved with each other to appear as a flexible network and knitted the LiNbO<sub>3</sub> into the networks.

The acquired mixture was dried and then annealed at 500 °C under purified N<sub>2</sub> gas flow for 10 hours to obtain CNT-LiNbO<sub>3</sub> nanoplate hybrids. As-synthesized hybrids were decorated with polypyrrole (PPy) using a surface polymerization method (*J. Mater. Chem.*, 2012, 22, 11159.), and later dispersed into distilled water to form a suspension which was poured into the funnel and filtered through a porous poly-vinylidene fluoride (PVDF) membrane. The resultant mat with its PVDF membrane was then washed twice using distilled water, followed by ethanol to remove any remaining solvent. The mat with the PVDF membrane still attached was dried in an oven for 2 hours and then peeled off the membrane. The acquired composites can maintain the flexibility and be cut into free-standing electrodes with required shapes.

## 2. Characterization

X-ray diffraction measurements were conducted on an X-ray powder diffractometer using Cu-K $\alpha$  radiation (D/Max-RA). Field-emission scanning electron microscopy (FE-SEM) experiments were conducted on a Hitachi S-4800 instrument. Transmission electron microscopy (TEM) experiments were carried out on a Titan 80-300 S/TEM operated at 150.0 kV.

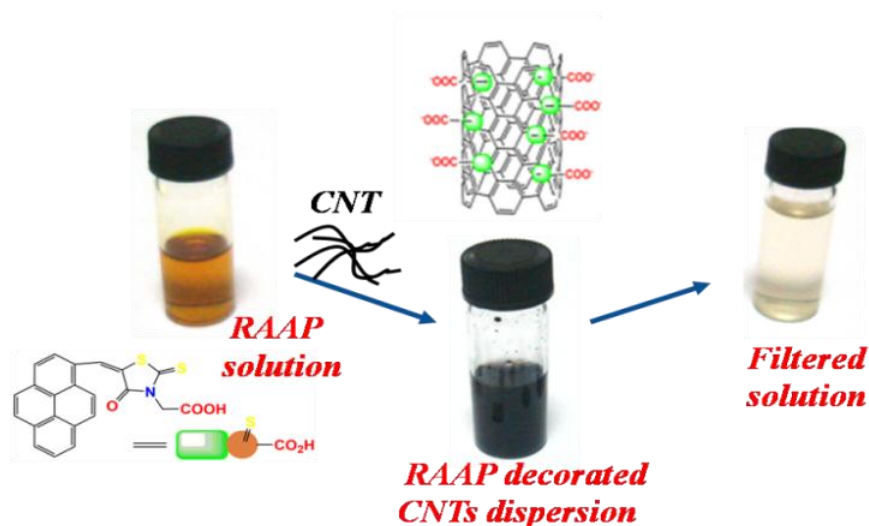
To examine the flexibility and durability of the LNO/CNT membrane, a cyclic bend test was performed (*Energy Environ. Sci.*, 2012, 5, 8901.). The electric conductivity was measured by the 4-probe method with Keithley 2400 Digital Source Meter.

## 3. Electrochemical measurement

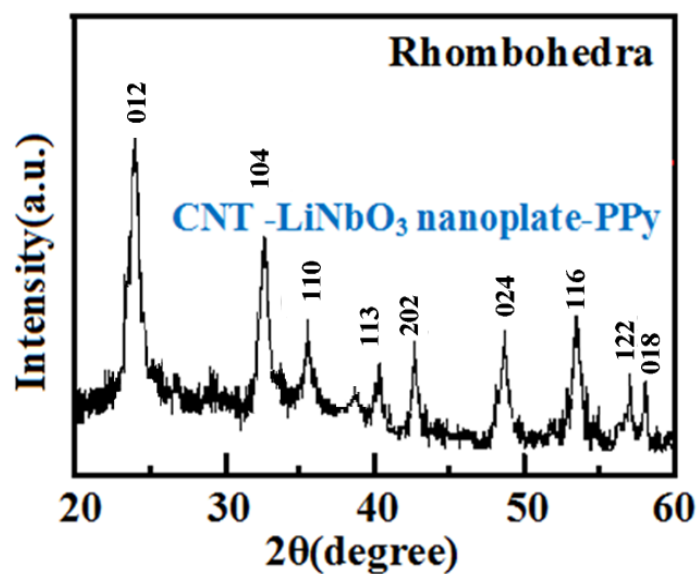
To test the electrochemical performance of the flexible lithium-ion anodes, flexible CNT-LiNbO<sub>3</sub> nanoplate-PPy hybrid (LNO-CNT) electrodes were cut into a desired size. By comparison, to make composite LiNbO<sub>3</sub> electrodes (LNO/C and LNO), 80% of as synthesized electrode materials, 10% carbon black, and 10% poly(vinylidene fluoride) (PVDF) dispersed in N-methylpyrrolidinone (NMP) were mixed to form slurries. The homogenous slurries were coated on a Cu substrate and dried at 60 °C for 1 h under vacuum. As formed electrodes were then pressed at a pressure of 2 MPa and further dried under vacuum at 100 °C for another 10 h. The mass loading was 3 ~ 4 mg cm<sup>-2</sup> on the current collect.

The flexible LNO-CNT and composite LiNbO<sub>3</sub> electrodes (LNO/C and LNO) were then assembled into 2032-type coin cells, where Lithium metal was used as the negative electrode and a porous polypropylene separator (Celgard 3501)) was saturated with 1 M LiPF<sub>6</sub> in 1:1 ethylene carbonate (EC)/dimethyl carbonate (DMC) as the electrolyte. All the cells were assembled in an argon-filled glove box. The constant-current charge and discharge measurements were conducted on a LAND CT2001-A at different current densities in a voltage range between 1.0 and 3.0 V (vs. Li/Li<sup>+</sup>). All of the electrochemical performance measurements were obtained at a constant temperature of 25 °C.

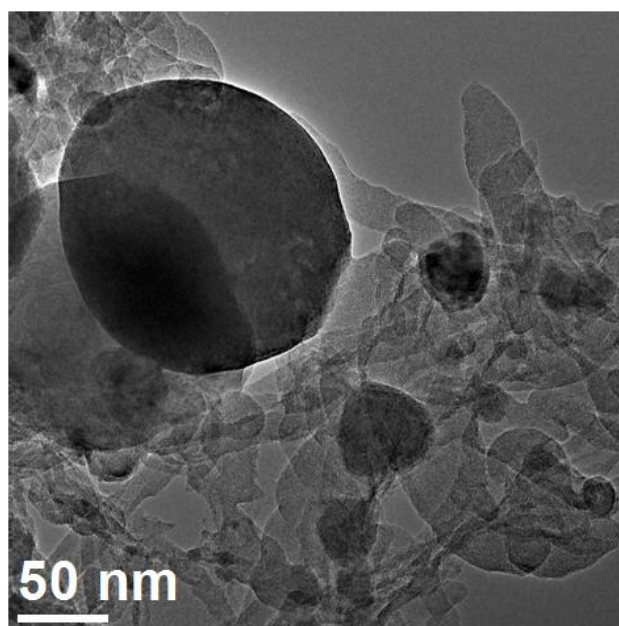
The pure CNT-PPy composite was synthesized and characterized as the controlled group under the same testing condition (Fig. S7). It shows a poor reversible capacity as  $16 \text{ mAh g}^{-1}$  in the voltage range of 3-1 V at the current density of  $0.4 \text{ A g}^{-1}$ . As the mass of CNTs and PPy in the flexible CNT-LiNbO<sub>3</sub>-PPy composite electrode is 26.2 %wt, the capacity contributions of the CNT-PPy in our samples is less than  $5 \text{ mAh g}^{-1}$ .



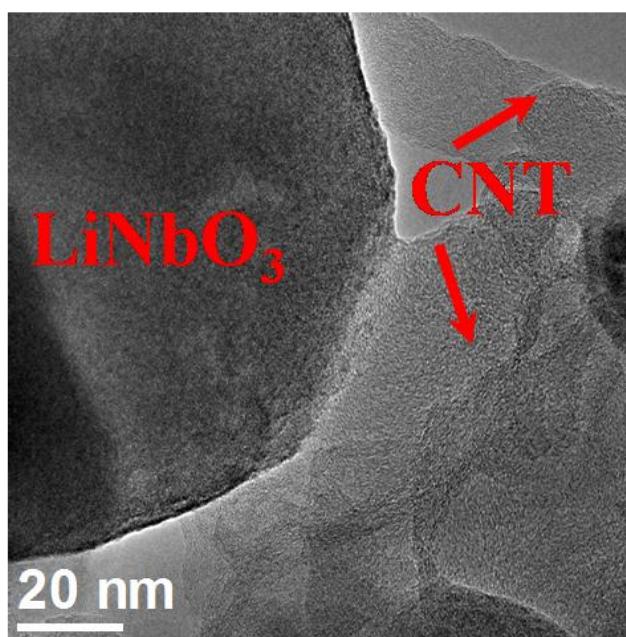
**Fig. S1** Flow diagram shows the process as RAAP decorated on the CNTs



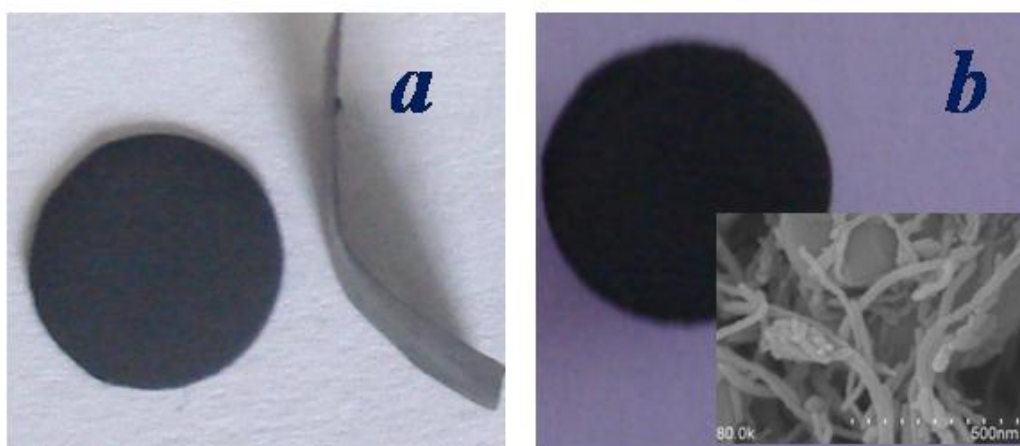
**Fig. S2** XRD pattern of the CNT-LiNbO<sub>3</sub> nanoplate-PPy hybrids



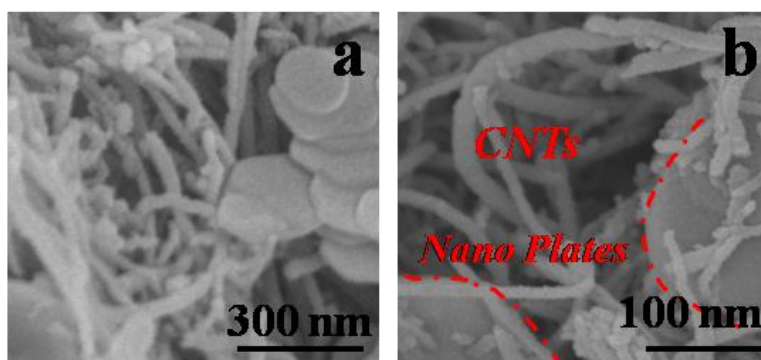
**Fig. S3** TEM image of LiNbO<sub>3</sub> –CNT-Matrix



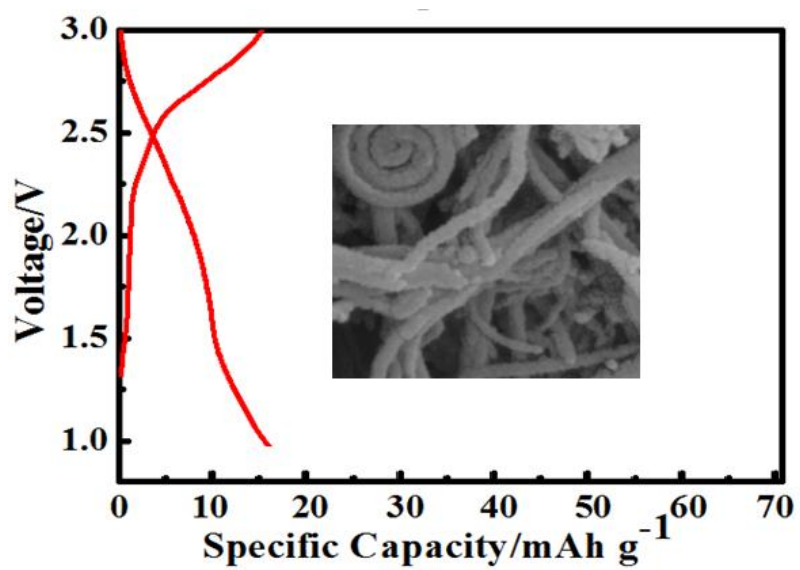
**Fig. S4** TEM image shows the nice contact between LiNbO<sub>3</sub> and CNTs



**Fig. S5** (a) Photograph of the flexible CNT -LiNbO<sub>3</sub> nanoplates/polypyrrole anodes;  
(b) A shape circular flexible electrode with the insert SEM image shows the morphology of polypyrrole decorated hybrids.



**Fig. S6** (a)-(b) SEM images shows the morphology of the CNT-LiNbO<sub>3</sub>-polypyrrole hybrids after 40 cycles at different charge-discharge rates.



**Fig. S7 Charge–discharge profiles of the pure CNT-PPy composite at 0.4 A g<sup>-1</sup>.**