Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A

Supporting Information

1. Supporting video

Video S1. Preparation of the aligned MWCNT sheet from a spinnable array. It mainly includes the following three steps: (1) a spinnable MWCNT array on the silicon substrate was firstly stabilized to the left sample hold; (2) an aligned MWCNT ribbon was first pulled off from the array and then attached on the right collecting roller; (3) the roller was rotated to continuously produce the MWCNT sheet. The rotary speed of the roller had been accurately controlled by an external motor.

2. Experimental Section

(1) Synthesis of spinnable MWCNT arrays

Chemical vapor deposition was used to synthesize spinnable MWCNT arrays in a quartz tube furnace. Fe (1.2 nm)/Al_2O_3 (5 nm) was used as the catalyst on a silicon wafer, ethylene served as carbon source with a flowing rate of 90 sccm, and a gas mixture of Ar (480 sccm) and H_2 (30 sccm) was used as the carrier gas. The growth was conducted at 750°C, and the thickness of spinnable CNT array was ~200 μm.

(2) Preparation of the quasi-solid-state electrolyte

The synthesis of ionic liquid crystal, 1-ethyl-3-methylimidazolium iodide (EMII), and ion liquid, 1-propyl-3-methylimidazolium iodide (PMII), was made according to Figure S1 (*J. Phys. Chem. B* 2011, 115, 14688).

Synthesis of 1-ethyl-3-methylimidazolium iodide (EMII). In a three neck flask which had been purged with dry nitrogen, 0.1 mol (7.97 mL) 1-methylimidazole was dissolved in 15 mL of acetonitrile, and 0.1 mol (8.08 mL) ethyl iodide was added dropwise under vigorous stirring in 1 h. The mixture was stirred for another 24 h at room temperature, followed by the addition of ethyl acetate under vigorous stirring. White crystal was precipitated immediately. The resulting product was then collected and washed with ethyl acetate and diethyl ether, each for three times, with a yield of ~70%. The product had been characterized by H^1-NMR as below.
H^1-NMR of EMII: (400 MHz, acetone-D6, δ) 1.56(t, 3H), 4.10(s, 3H), 4.46(q, 2H), 7.85 (br d, 2H), 9.68(s, 1H).

Synthesis of 1-propyl-3-methylimidazolium iodide (PMII). It was similar to EMII. In a three neck flask which had been purged with dry nitrogen, 0.1 mol (7.97 mL) 1-methylimidazole was dissolved in 15 mL isopropanol, and 0.1 mol (9.71 mL) propyl iodide was added dropwise under vigorous stirring in 1 h. The mixture was stirred for another 24 h at 80 ºC and cooled down to room temperature, followed by washing with ethyl acetate and diethyl ether, each for 3 times. The solvent was vaporized and the product was dried under vacuum for 3 days with a yield of ~70%. The resulting product had been also characterized by H^1-NMR as below.
H^1-NMR of PMII: (400 MHz, acetone-D6, δ) 0.94(t, 3H), 1.97(sx, 2H), 4.10(s, 3H),

4.40(q, 2H), 7.90 (br d, 2H), 9.69(s, 1H).

Preparation of quasi-solid-state electrolyte. 1 g of EMII, 0.35 g of PMII and 0.03 g of iodine were solved in 1 mL of acetonitrile. The mixture was stirred for 8 h and then heated to 80 ºC over night, followed by drying in vacuum to remove the acetonitrile. The quasi-solid-state electrolyte appeared dark.

(3) Characterization

The structures were characterized by scanning electron microscopy (Hitachi FE-SEM S-4800 operated at 1 kV). H^1-NMR spectra were obtained on a Varian Mercury plus 400 MHz NMR spectrometer. Thermal analysis was performed on a TA DSC Q2000 differential scanning calorimeter at a heating rate of 10 ºC min^{-1} from -50 to 150 ºC in N_2 atmosphere. The J-V curves were produced by a Keithley 2400 Source Meter under illumination (100 mW/cm^2) of simulated AM1.5 solar light coming from a solar simulator (Oriel-Sol3A 94023A equipped with a 450 W Xe lamp and an AM1.5 filter). The light intensity was calibrated using a reference Si solar cell (Oriel-91150). The transmittance characterizations of MWCNT sheet was made by UV-vis spectrophotometer (SHIMADZU UV-2550). The temperature of the DSC was controlled by a hot plate (MODELKW-4AH).

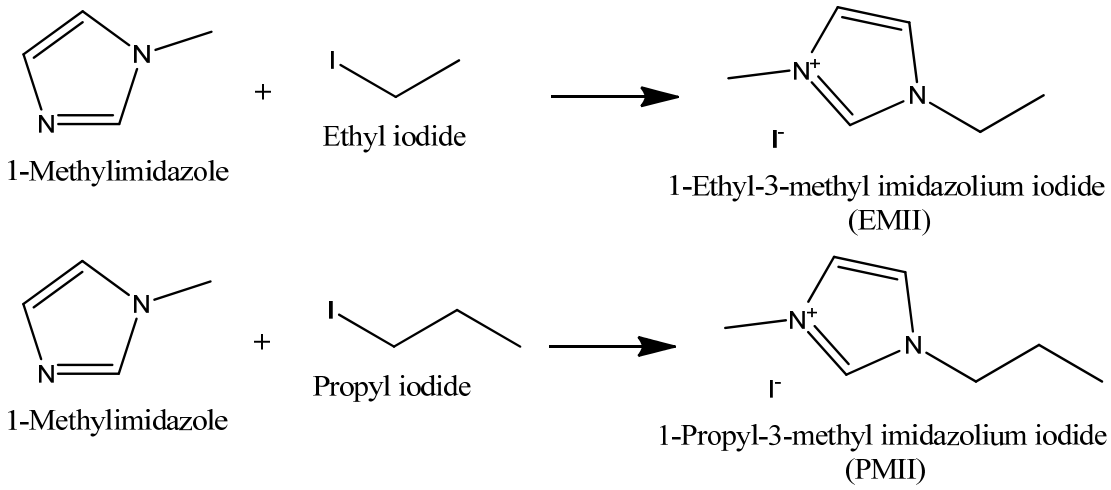

Figure S1. Schematic illustration to the synthesis of the ionic liquid crystal of 1-ethyl-3-methylimidazolium iodide (EMII) and ion liquid of 1-propyl-3-methylimidazolium iodide (PMII).

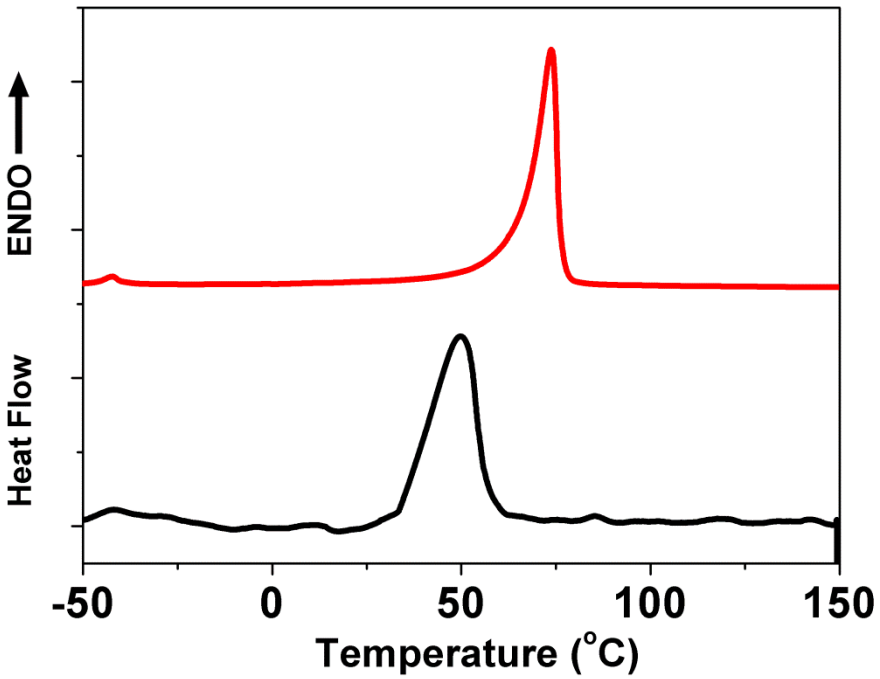

Figure S2. Thermal analysis of EMII (red line) and quasi-solid-state electrolyte (black line).

Figure S3. Photograph of the electrolyte in an inverted bottle. The arrow shows the quasi-solid-state electrolyte.

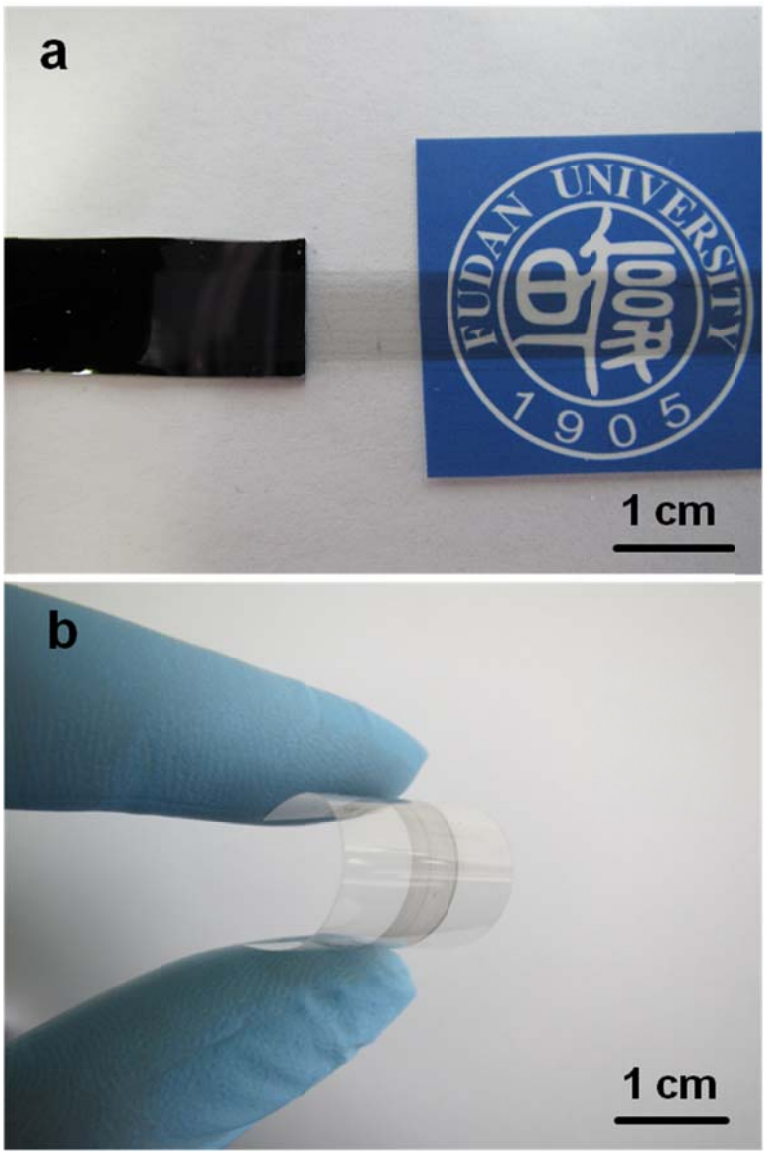

Figure S4. Photographs of MWCNT sheets. **a.** A single layer of MWCNT sheet with thickness of 20 nm being pulled out of a spinnable MWCNT array. The logo of Fudan University below the sheet can be clearly observed. **b.** A MWCNT sheet being attached on a flexible and transparent polyethylene terephthalate film.

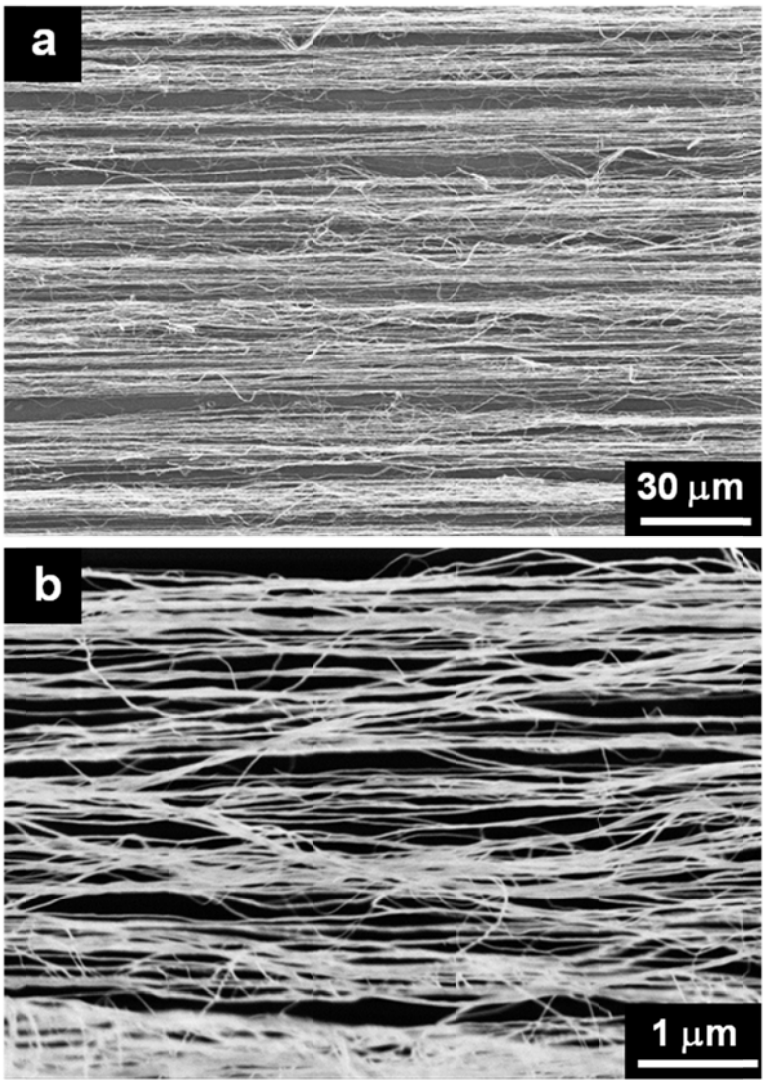

Figure S5. Scanning electron microscopy images of a highly aligned MWCNT sheet at (a) low and (b) high magnifications.

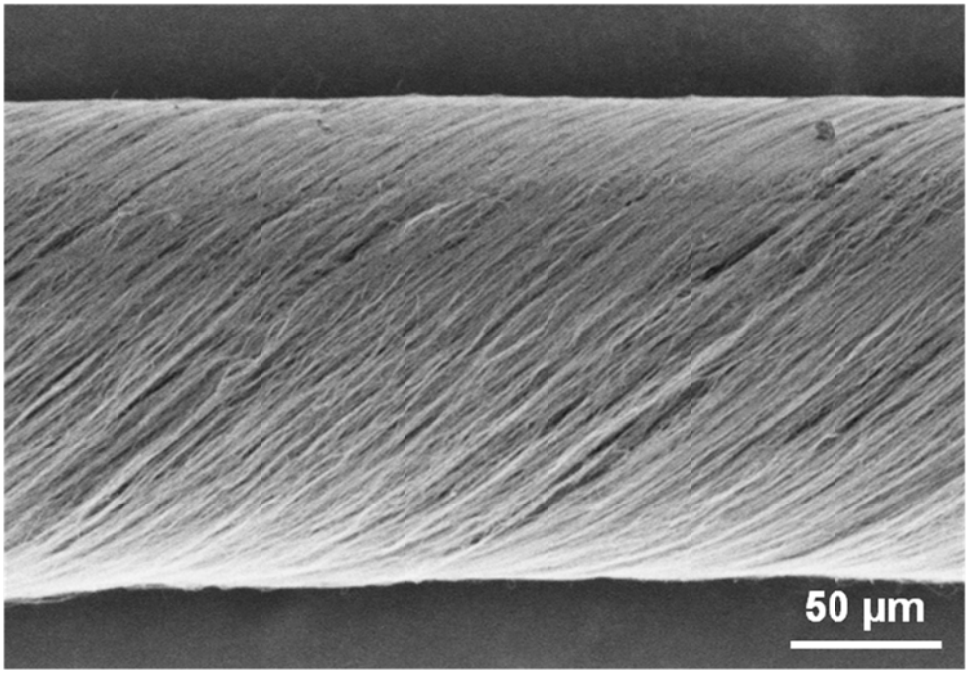

Figure S6. Scanning electron microscopy image of a MWCNT sheet with thickness of 60 nm being wrapped on a titania nanotube-modified Ti wire.

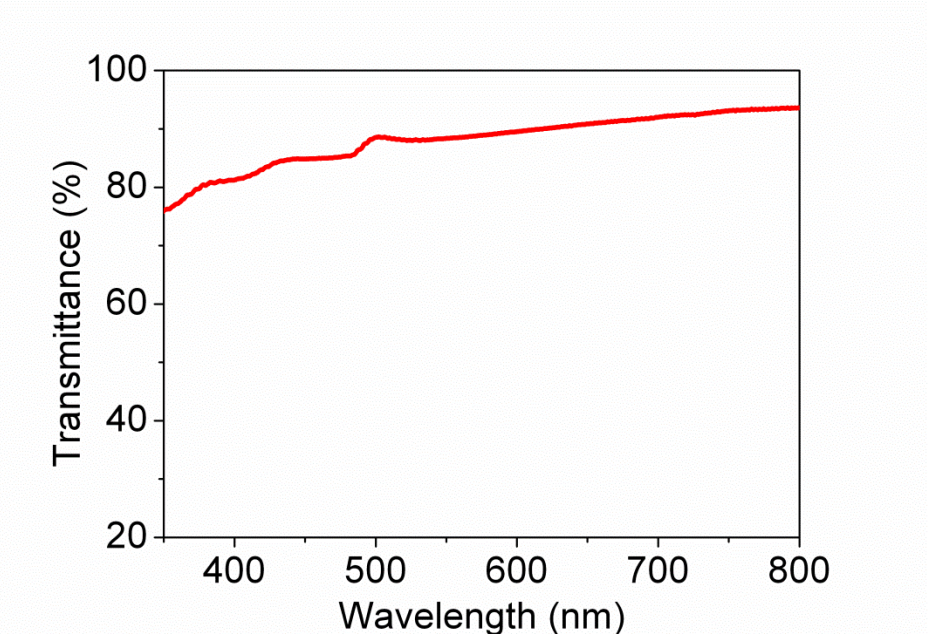

Figure S7. UV-vis spectra of an aligned MWCNT sheet with a thickness of 20 nm.

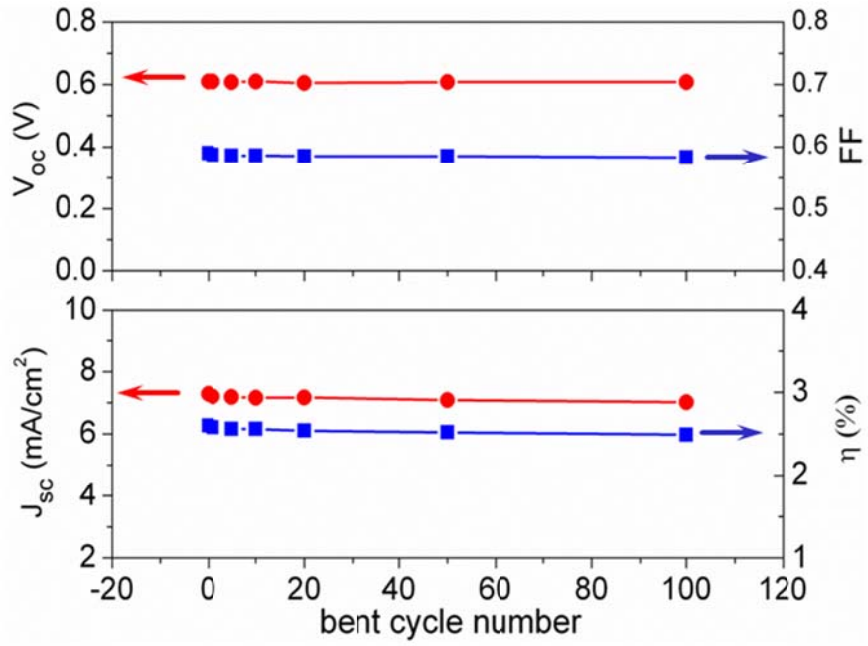

Figure S8. Photovoltaic parameters before and after bending for different cycles.

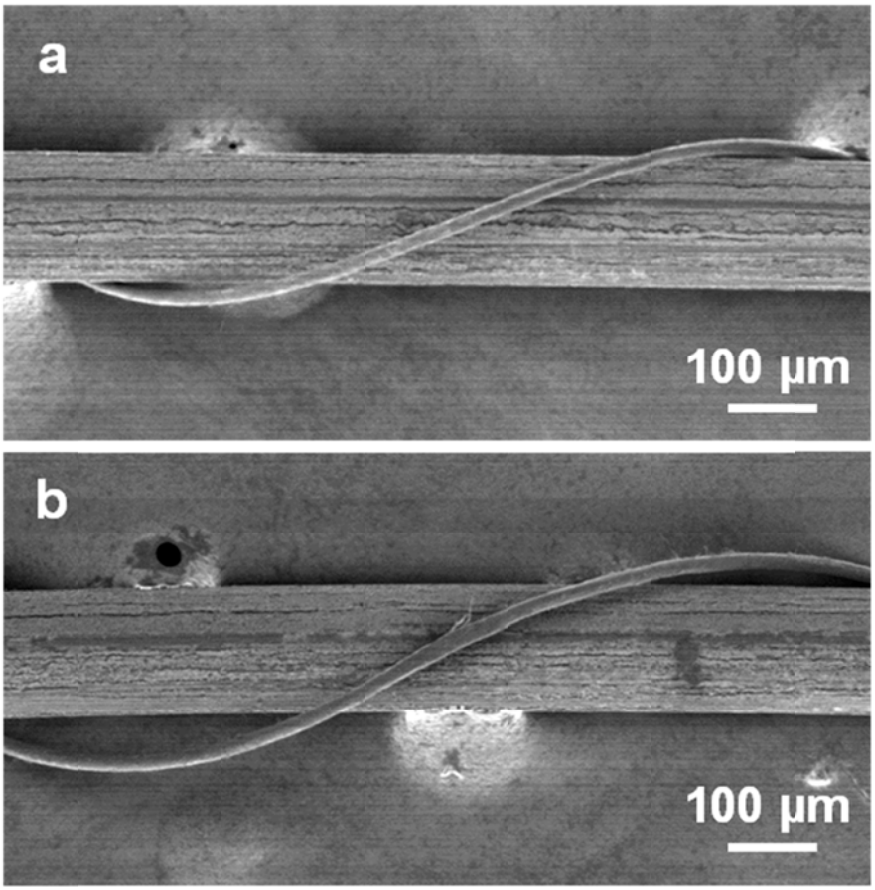

Figure S9. a and **b.** Scanning electron microscopy images of a twisted wire-shaped DSC before and after bending for 10 cycles, respectively.

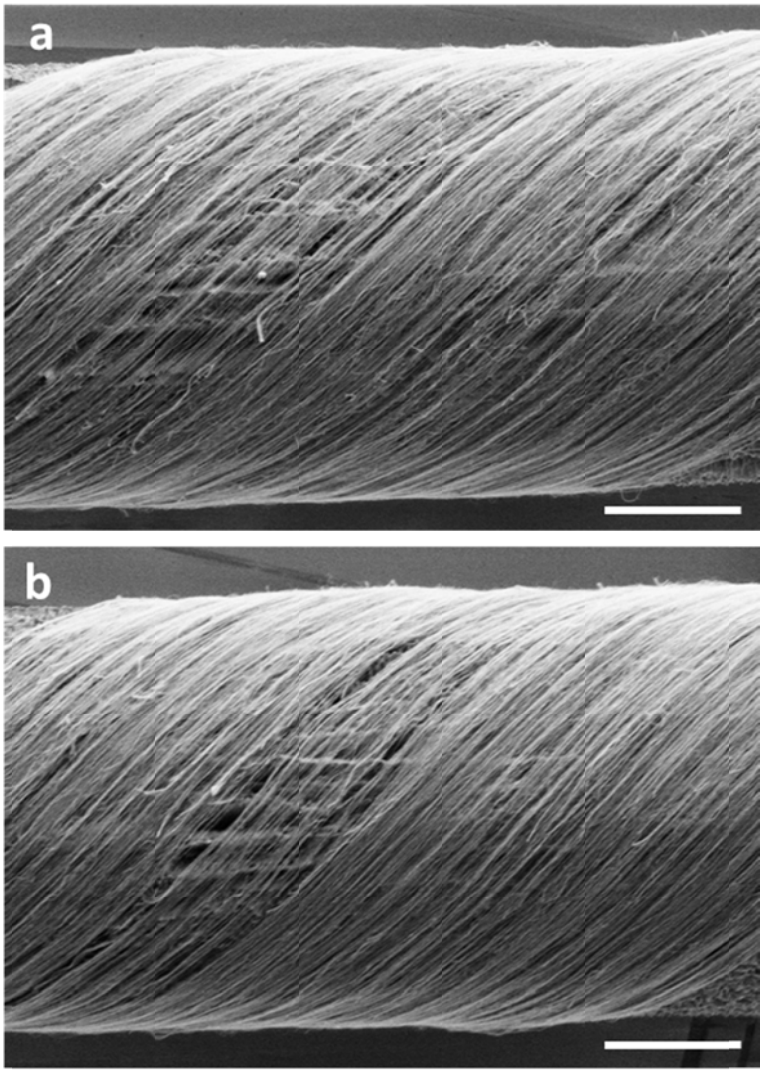

Figure S10. a and **b.** Scanning electron microscopy images of a coaxial fiber-shaped DSC before and after bending for 10 cycles, respectively. The scale bars are the same of 50 μm.

Table S1. Photovoltaic parameters of quasi-solid-state coaxial photovoltaic fibers based on different lengths of titania nanotubes as working electrodes.

Length (μm)	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	η (%)
10	0.611	4.54	0.590	1.64
22	0.618	5.93	0.588	2.15
32	0.610	7.29	0.585	2.60
40	0.611	6.49	0.590	2.34

Table S2. Photovoltaic parameters of quasi-solid-state coaxial photovoltaic fibers based on different thicknesses of CNT sheets as counter electrodes.

Thickness (nm)	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	η (%)
20	0.610	7.29	0.585	2.60
40	0.603	6.37	0.570	2.19
60	0.613	5.56	0.516	1.76
80	0.604	3.99	0.489	1.18

Table S3. Photovoltaic parameters of quasi-solid-state coaxial photovoltaic fibers and liquid-state twisted photovoltaic wires. The two solar cells showed the same length of ~1.5 cm.

Group	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	η (%)
Liquid-state twisted	0.723	11.46	0.386	3.20
Quasi-solid-state coaxial	0.610	7.29	0.585	2.60