

## Supporting Information

### Preparation of aligned carbon nanotube sheet

Aligned carbon nanotube (CNT) sheets were dry-drawn from a CNT array that was synthesized by chemical vapor deposition at 740 °C. Fe (1.2 nm)/Al<sub>2</sub>O<sub>3</sub> (3 nm) on a silicon substrate served as the catalyst. The ethylene (flowing rate of 90 sccm) was used as the carbon source. A mixture of Ar (flowing rate of 400 sccm) and H<sub>2</sub> (flowing rate of 30 sccm) was used as the carrier gas.

### Preparation of fiber-shaped supercapacitors based on CNT fibers

Fiber-shaped supercapacitors were fabricated according to the previously reported methods.<sup>1</sup> In brief, CNT fibers were prepared by twisting the stacked CNT sheets with a rotating motor. To ensure the supercapacitors in strip and fiber configuration have the same amount of active materials, 40 layers of CNT sheets were twisted at a rotating speed of 1000 rpm to prepare the fibrous electrode. Two fibrous electrodes were coated with gel electrolyte and twisted into a fiber-shaped supercapacitor.

### Calculation of electrochemical parameters of supercapacitors

The volumetric capacitance ( $C_V$ ), gravimetric capacitance ( $C_M$ ) and areal capacitance ( $C_A$ ) of the strip-shaped supercapacitor are calculated based on single electrode by the following equations:

$$C_V = 2 \times \frac{I \times \Delta t}{V \times \Delta U}$$

$$C_M = 2 \times \frac{I \times \Delta t}{m \times \Delta U}$$

$$C_A = 2 \times \frac{I \times \Delta t}{S \times \Delta U}$$

where  $I$ ,  $\Delta t$ ,  $V$ ,  $m$ ,  $S$ , and  $\Delta U$  are discharge current, discharge time, volume, mass and surface area of active material in the electrode and voltage window, respectively.

The volume of active material—CNT sheet or CNT/PANI composite film—is calculated by assuming it as a cuboid. The volume can be obtained by

$$V = L \times W \times H$$

where  $L$ ,  $W$ , and  $H$  represent the length, width and thickness of the electrode,

respectively.

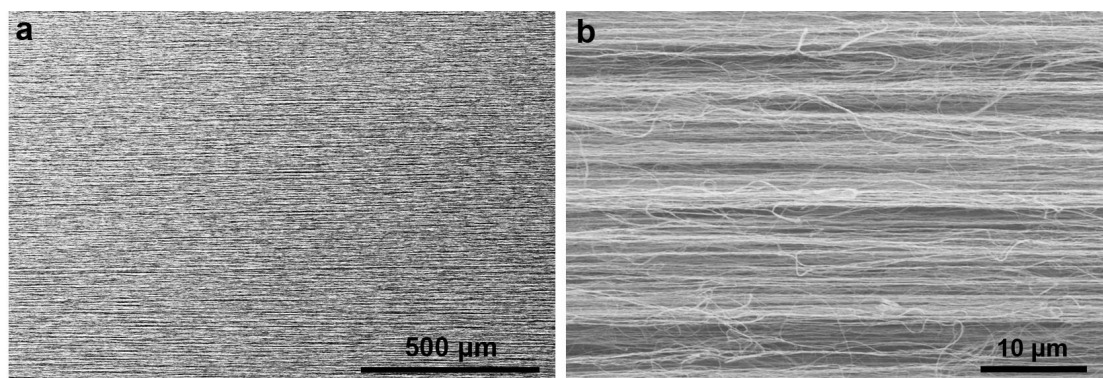
In a symmetrical supercapacitor, we have:

$$\frac{1}{C} = \frac{1}{C_1} + \frac{1}{C_2} = \frac{2}{C_E}$$

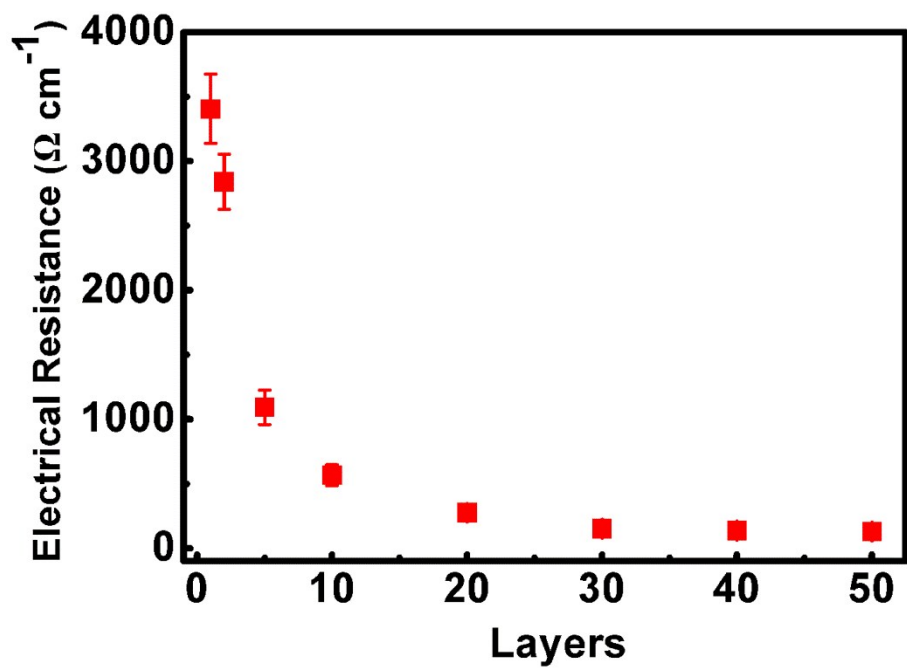
where  $C$  is the capacitance of the supercapacitor and  $C_E$  is the capacitance of a single electrode. Therefore, the specific capacitances  $C_S = 0.25 \times C_V$ . According to  $E = 0.5 \times C_S \times \Delta U^2$ , the volumetric energy density ( $E_V$ ) and volumetric power density ( $P_V$ ) can be expressed as:

$$E_V = 0.125 \times C_V \times \Delta U^2$$

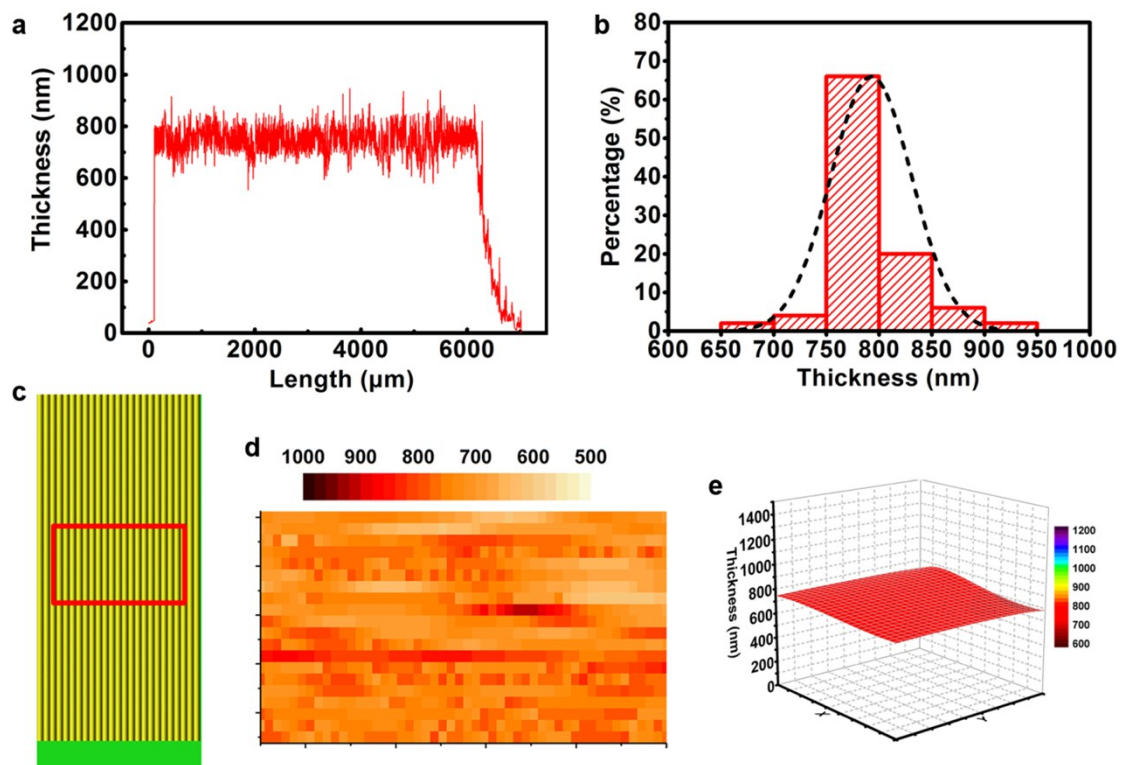
$$P_V = E_V \times 3600/\Delta t$$



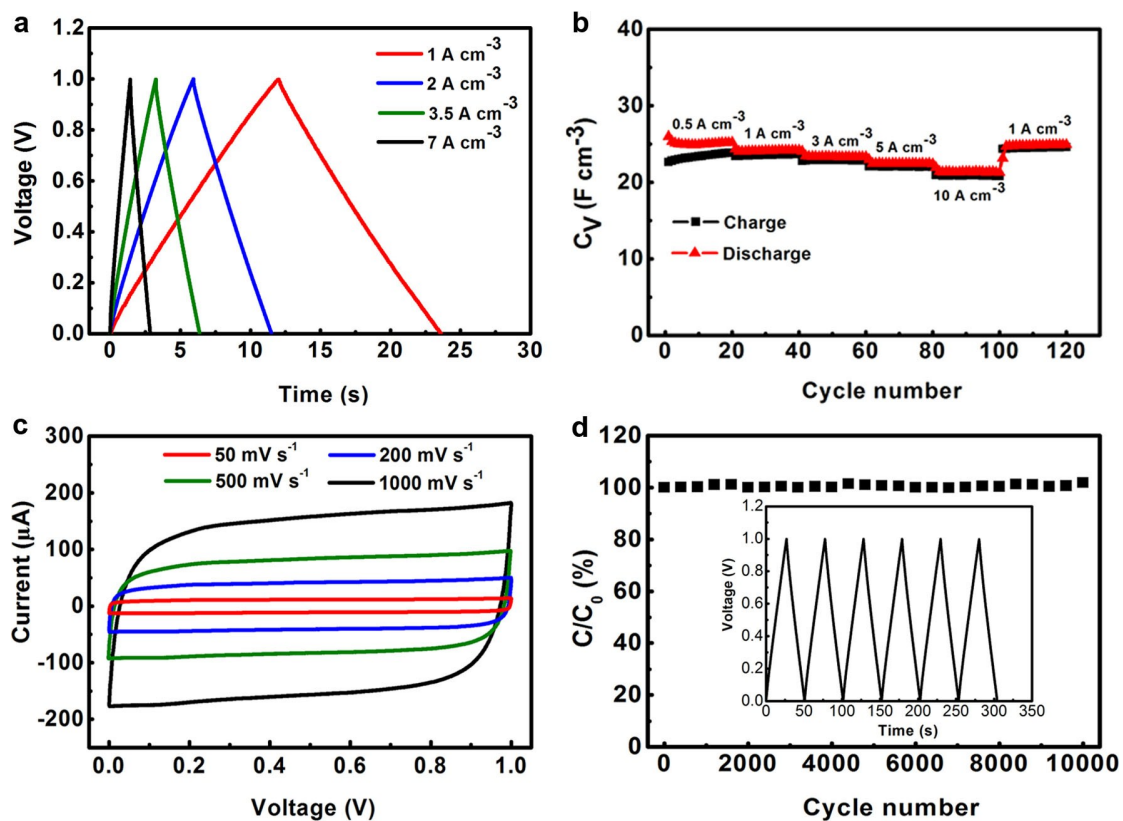
**Fig. S1** Low (a) and high (b) magnification SEM images of the CNT strip.



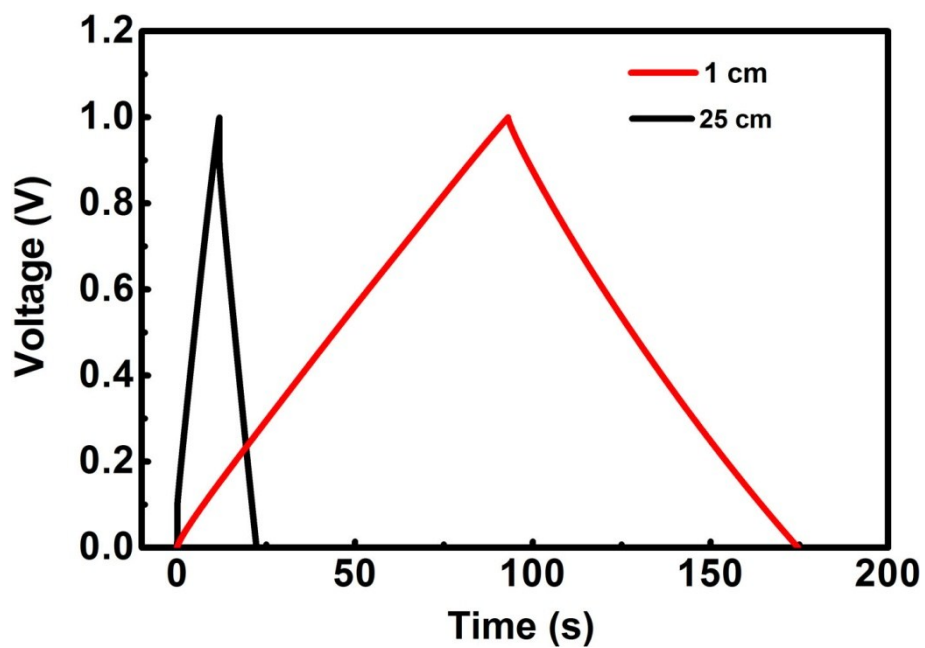
**Fig. S2** Dependence of electrical resistance on the layer number of CNT strips. The tested strip electrodes shared the length of 10 cm and width of 2 mm.



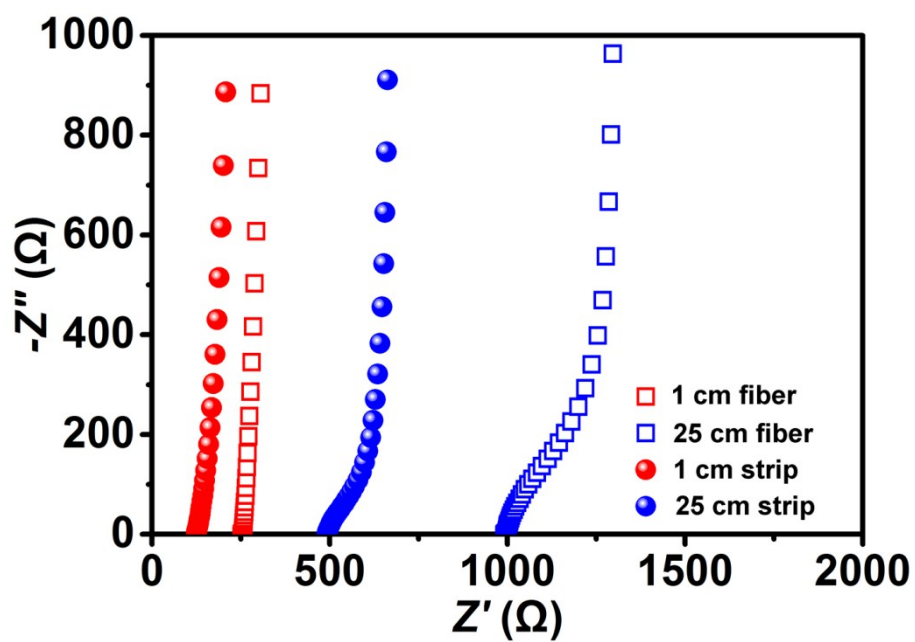
**Fig. S3** Thickness of 40 layers of CNT strips. (a) Cross scanning profile. (b) Histogram of the thicknesses. (c and d) Schematic illustration and thickness mapping of the selected area, respectively. (e) Surface fitting of the selected area at c.



**Fig. S4** Electrochemical performance of strip-shaped supercapacitors with a length of 1 cm. (a) Galvanostatic charge-discharge curves with increasing current densities. (b) Rate performances. (c) Cyclic voltammograms. (d) Cyclic performances at 0.5 A cm<sup>-3</sup>. The inserted graph displays the galvanostatic charge-discharge curves in the first 6 cycles.

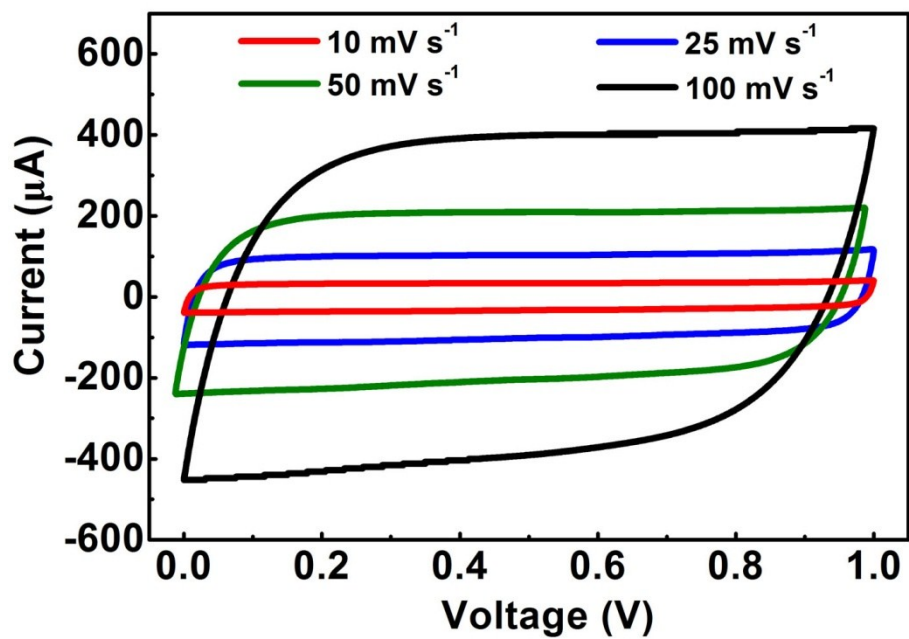


**Fig. S5** Galvanostatic charge-discharge curves of 1-cm and 25-cm fiber-shaped supercapacitors at  $0.1 \text{ A cm}^{-3}$ .

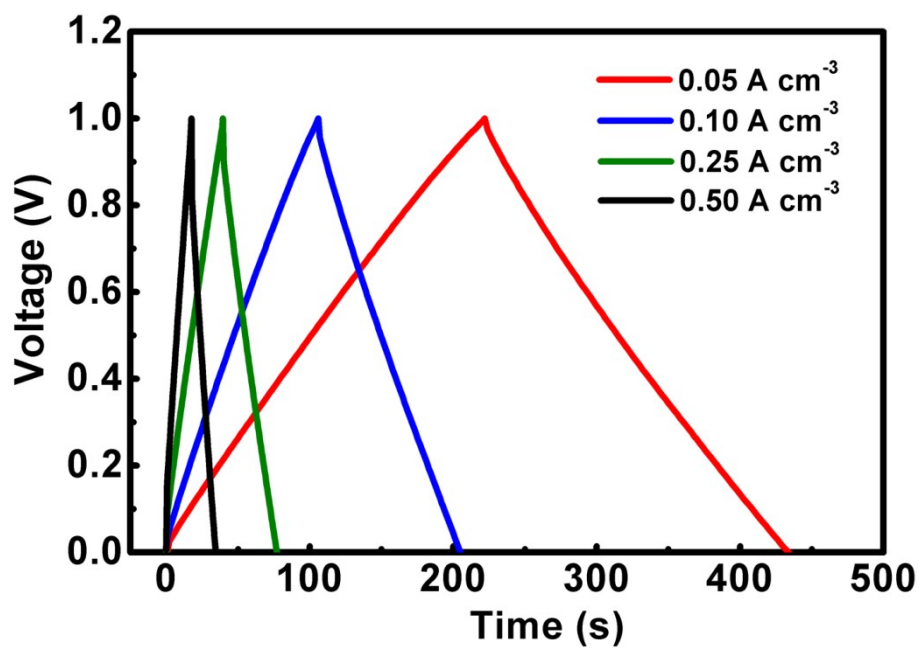


**Fig. S6** Comparison on the electrochemical impedance spectra of strip-shaped (solid sphere) and fiber-shaped supercapacitors (empty square) at the same two lengths, i.e., 1 and 25 cm. The compared supercapacitors with different configurations had the same amount of CNTs.

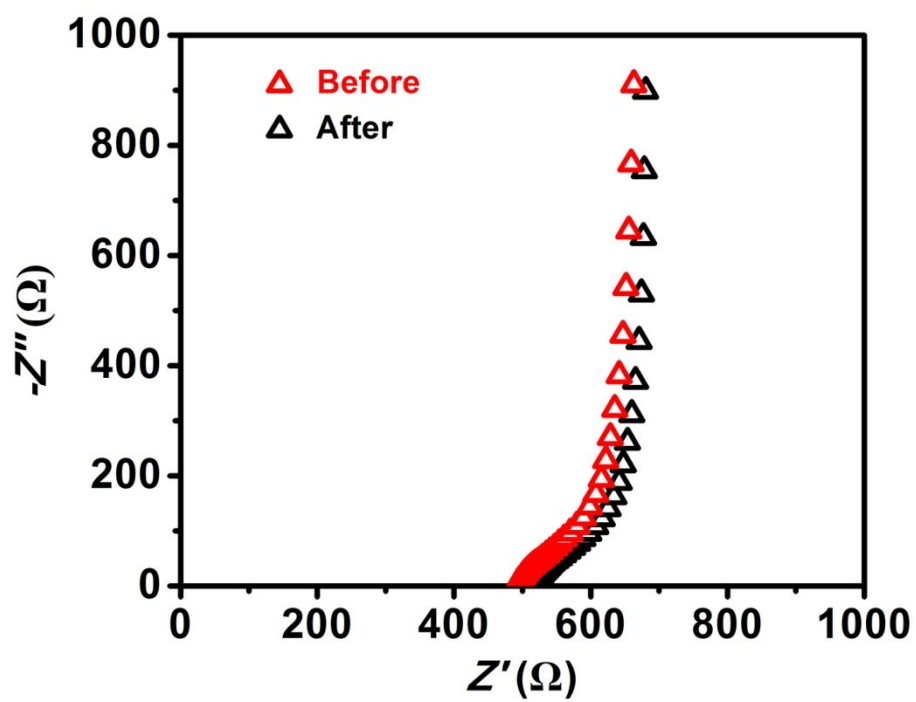




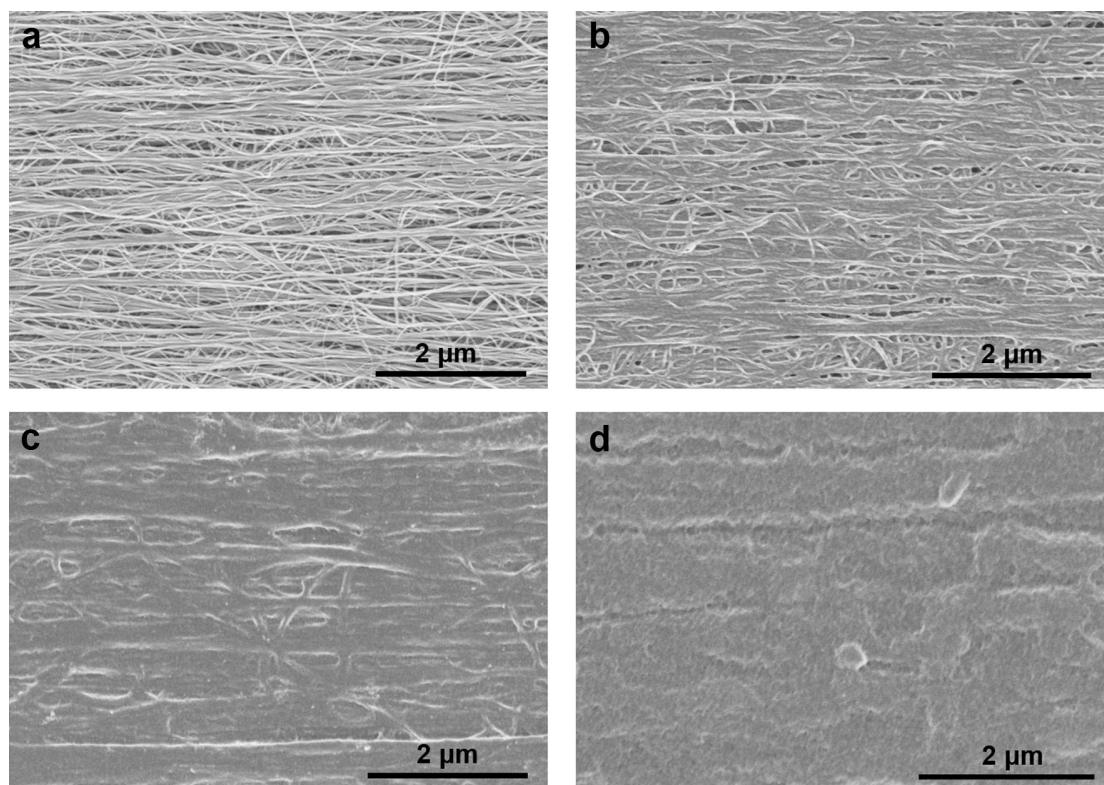
**Fig. S7** Cyclic voltammogram curves of a 25-cm-long supercapacitor at increasing scanning rates.



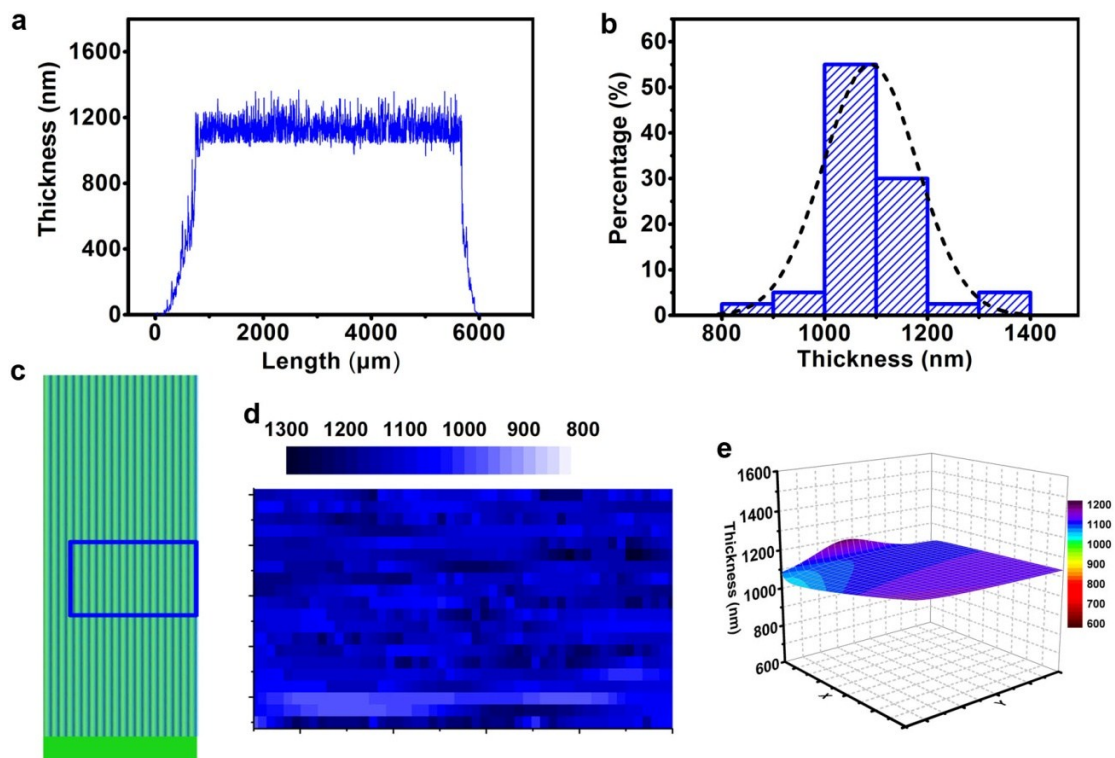
**Fig. S8** Galvanostatic charge-discharge curves of a 25-cm-long supercapacitors at increasing current densities.



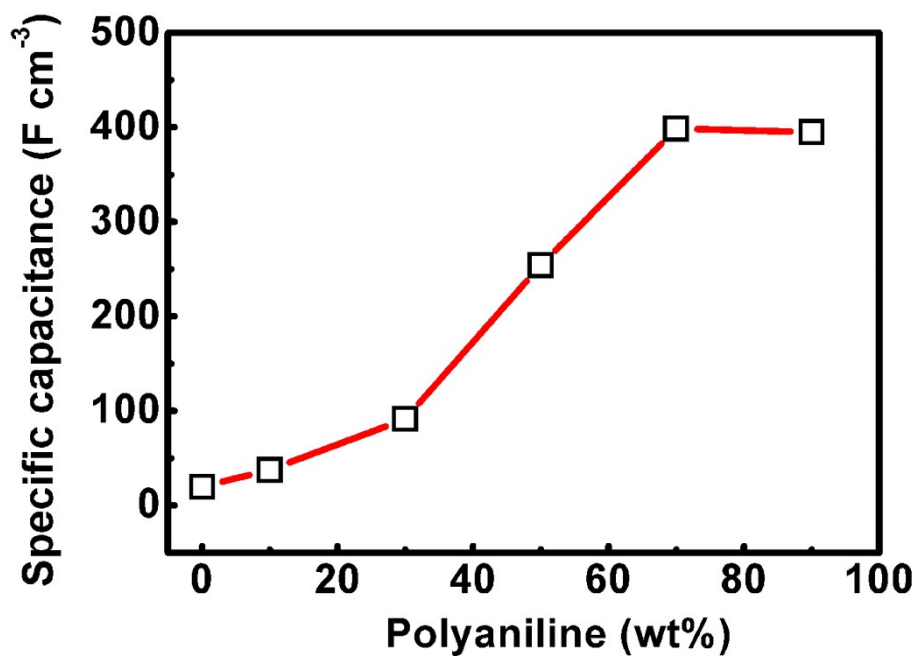
**Fig. S9** Electrochemical impedance spectra of a strip-shaped supercapacitor (length of 25 cm) before (red) and after (black) bending for 1000 cycles.



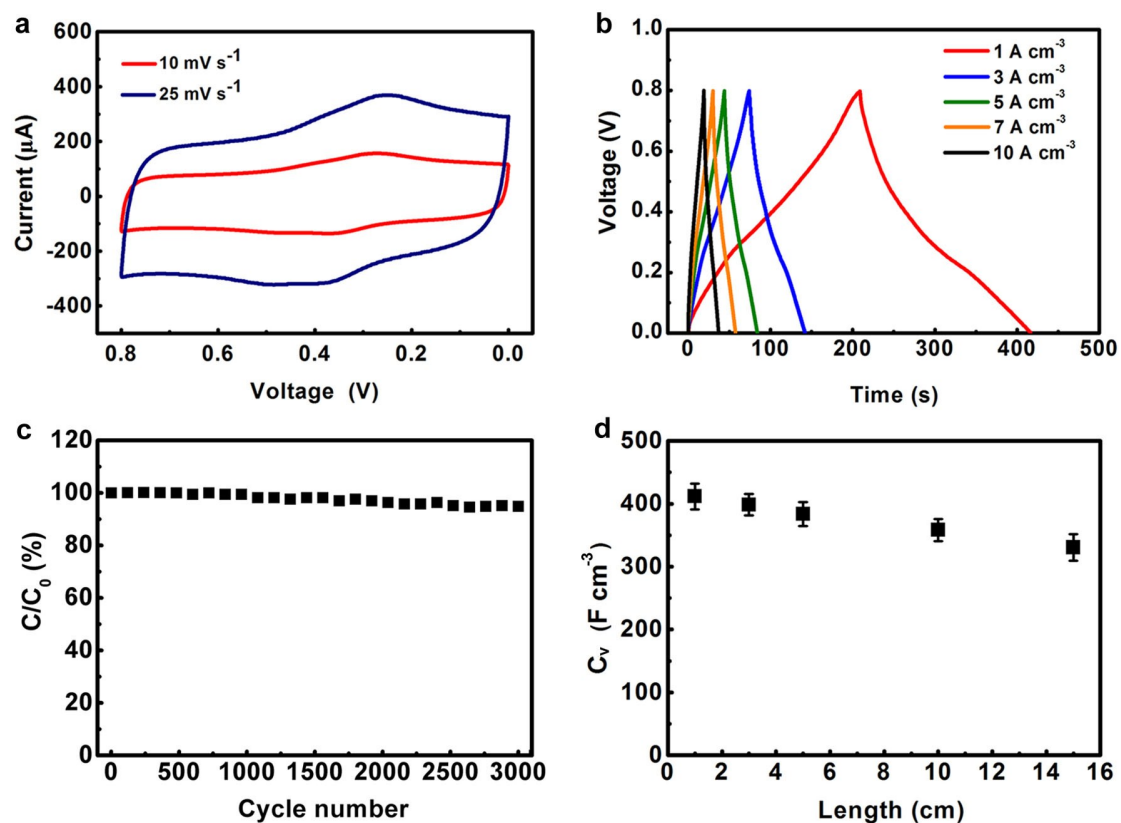
**Fig. S10** SEM images of aligned CNT/PANI composites with increasing PANI weight percentages. (a) Bare CNT strip. (b) 30%. (c) 50%. (d) 70%. The content of PANI was controlled by the time for electrochemical polymerization.



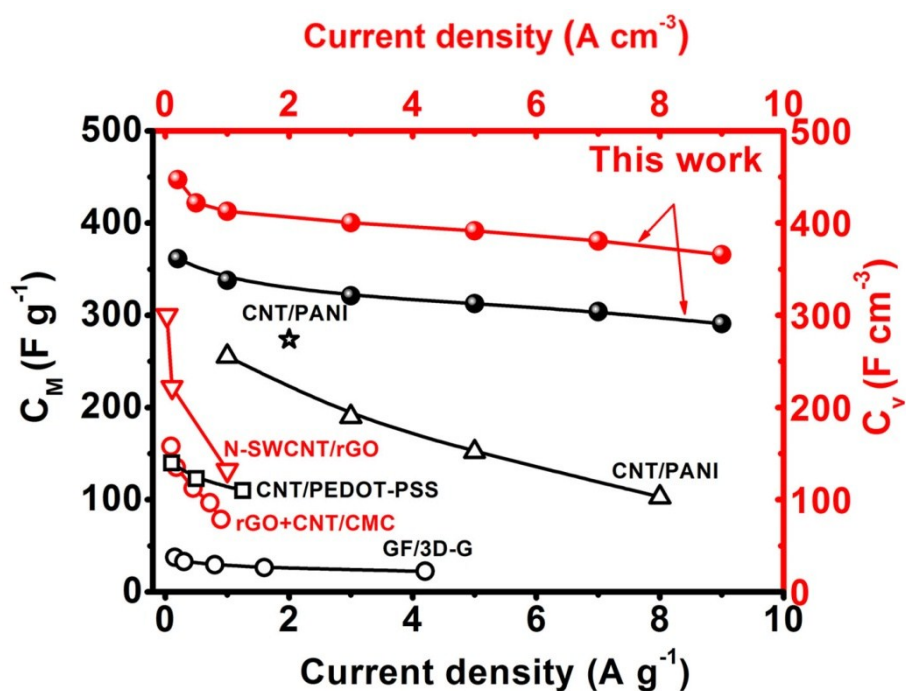
**Fig. S11** Thickness of aligned CNT/PANI composite strip. (a) Cross scanning profile. (b) Histogram of the thicknesses. (c and d) Schematic illustration and thickness mapping of the selected area, respectively. (e) Surface fitting of the selected area at c. PANI was electrochemically deposited on the stacked CNT strip (40 layers).



**Fig. S12** Dependence of specific capacitance on the weight percentage of PANI in the strip-shaped supercapacitors.

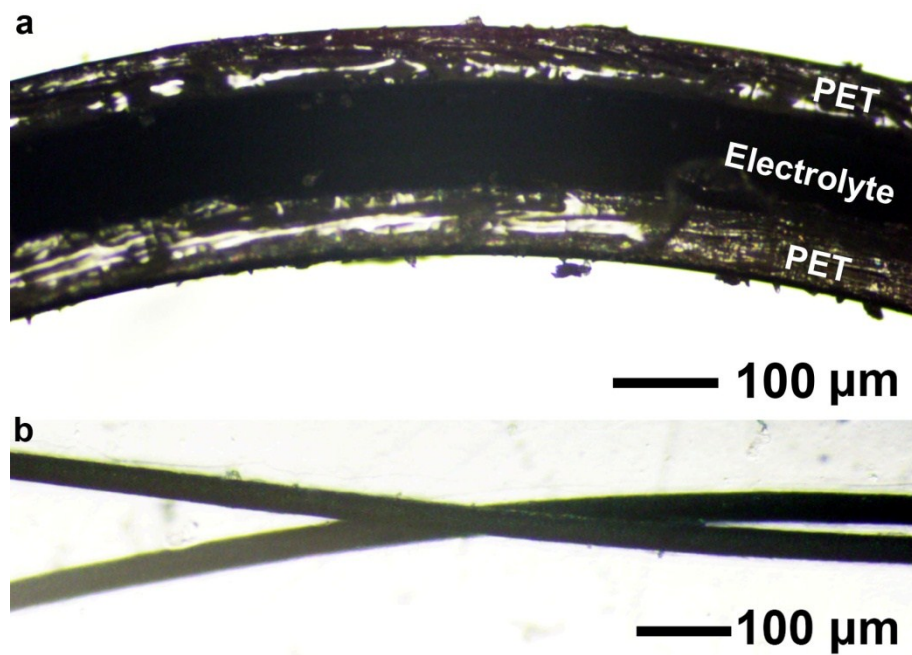


**Fig. S13** Electrochemical performance of strip-shaped supercapacitors based on aligned CNT/PANI composite. (a) Cyclic voltammograms. (b) Galvanostatic charge-discharge curves. (c) Cyclic performances at  $1 \text{ A cm}^{-3}$ . The supercapacitors at a-c shared the length of 1 cm. (d) Dependence of specific capacitance on the length of supercapacitor. The measurements were made at a current density of  $1 \text{ A cm}^{-3}$ . Each point was collected from five parallel samples.

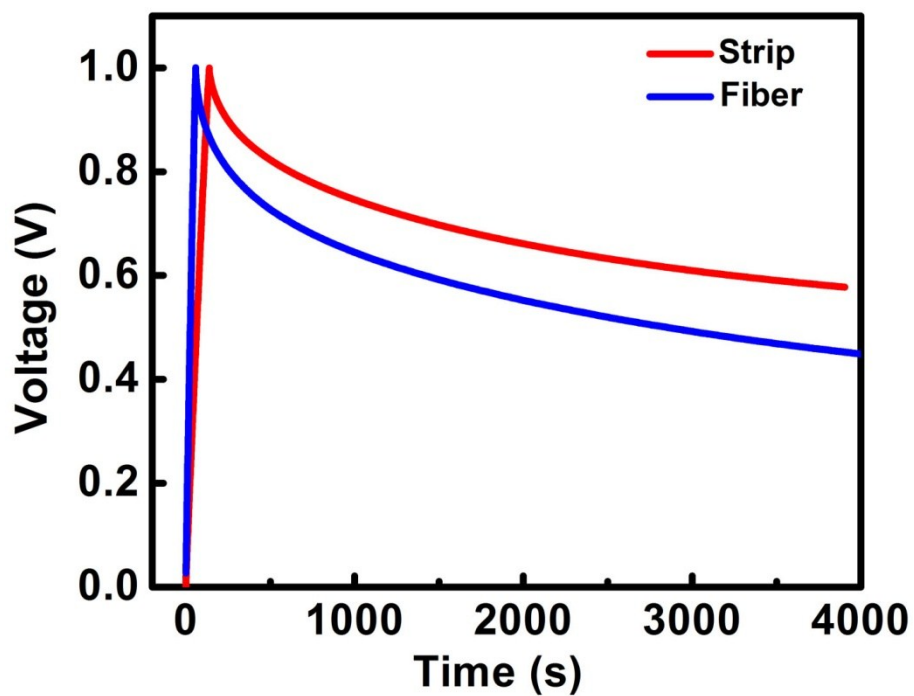


**Fig. S14** Comparison on specific gravimetric capacitances ( $C_M$ ) (black) and volumetric capacitances ( $C_V$ ) (red) of the strip-shaped supercapacitor with the reported fiber-shaped supercapacitors based on GF/3D-G,<sup>S2</sup> PEDOT-PSS/CNT,<sup>S3</sup> rGO+CNT/CMC,<sup>S4</sup> SWCNT/N-doped rGO,<sup>S5</sup> and CNT/PANI<sup>S6,7</sup> at different current densities. GF, 3D-G, PEDOT-PSS, rGO and CMC represent graphene fiber, three-dimensional graphene, poly(3,4-ethylenedioxythiophene)/poly(styrene sulfonate), reduced graphene oxide, and carboxymethyl cellulose, respectively. Here the strip-shaped supercapacitor was made from the CNT/PANI (70 wt%) composite.





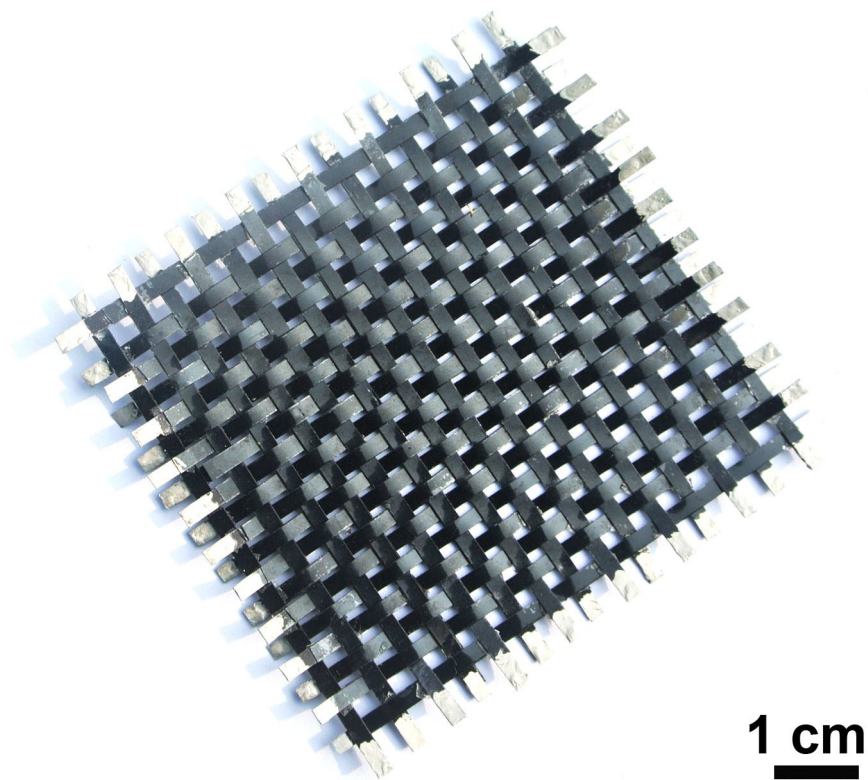
**Fig. S15** Optical micrographs of strip- and fiber-shaped supercapacitors under bending deformation. (a) Strip-shaped supercapacitor. (b) Fiber-shaped supercapacitor twisted from two fibrous electrodes. Both strip- and fibre-shaped supercapacitors were bent by 180° with a bending radii of 1 cm.



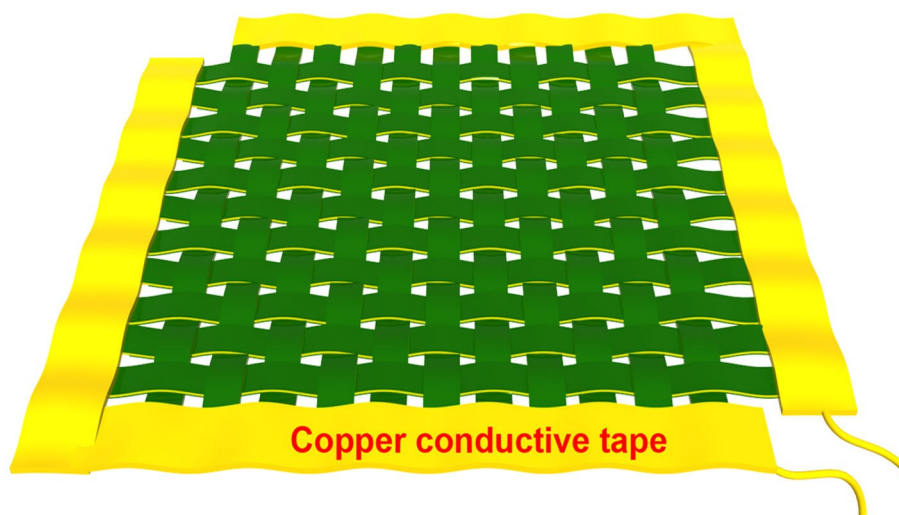
**Fig. S16** Self-discharge tests of strip- and fiber-shaped supercapacitors. The strip- and fiber-shaped supercapacitors were 25 cm in length and had same weight of CNTs. They were charged at  $5 \times 10^{-5}$  A and then rested for 1 h.



**Fig. S17** Photograph of a bent energy storage fabric woven from strip-shaped supercapacitors with a length of 25 cm.



**Fig. S18** Photograph of a fabric woven from 8-cm strip-shaped supercapacitors.



**Fig. S19** Schematic illustration of the energy storage fabric for galvanostatic charge-discharge tests. The supercapacitor units in the fabric were connected in parallel.

### References for the Supporting Information

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