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Supporting Information

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A Highly Stretchable, Fiber-Shaped Supercapacitor**

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Supporting Information

Video S1: The highly stretchable property of the fiber-shaped supercapacitor.

Experimental section

The synthesis of spinnable CNT arrays that were used to prepare aligned CNT sheets was previously reported[S1-S3]. To wind the aligned CNT sheet, (1) an elastic rubber fiber was firstly stabilized at two motors by the two ends, (2) a spinnable CNT array was then fixed onto a precisely motorized translation stage, and (3) a continuous, aligned CNT sheet with thickness of ~18 nm was drawn out of the array and attached on the rubber fiber with an angle of 60° with the two motors and the translation stage being operated simultaneously (Figure S17). The helical angle of the wound CNT sheet was maintained at 60° across the fiber when the precession velocity of CNT sheet equals to the moving velocity of translation stage.

The OMC particles with diameters of 1-5 μm (pore diameter of 3.8-4.0 nm and specific surface area of 900 m^2 g^{-1}) were purchased from the Nanjing XFNANO Materials Tech Co. Ltd. They were firstly dispersed in ethanol with a concentration of 1 mg/mL, followed by coating onto the CNT sheet. As the voids in the CNT sheet were ranged from tens to hundreds of nanometers, these OMC nanoparticles should be mainly coated at the top of the CNT sheet. Note that the particle solution could be quickly absorbed by and then uniformly distributed among the aligned CNTs to form uniform composite materials. The content of the OMC at a weight percentage can be controlled by varying the dispersion volume. The PVA-H_3PO_4 gel electrolyte was prepared by dissolving PVA powder (1 g) in deionized water (9 mL) and H_3PO_4 (1 mL)[S4].

The structures were characterized by TEM (JEOL JEM-2100F operated at 200 KV) and SEM (Hitachi FE-SEM S-4800 operated at 1 KV). The cyclic voltammetry and galvanostatic charge-discharge curves were recorded by a CHI 660a electrochemical workstation. The specific capacitance (C) was calculated from the galvanostatic discharge curve according to the equation of $C=2(I*\Delta t)/(m*\Delta V)$, where I, Δt, m, and ΔV are discharge current, discharge time, electrode weight, and voltage variation in the time range, respectively[S3]. A Shimadzu Table-Top Universal Testing Instrument was used for the stretching measurement.

Calculations of the thicknesses of carbon nanotube (CNT) sheet electrodes. During the fabrication of CNT sheet electrodes, the angle between rubber fiber and CNT sheet was 60°, which had been maintained when the precession velocity of the MWCNT sheet was equal to the moving velocity (v) of the translation stage. At a specific time during the fabrication (i.e., t), the total area of the used CNT sheet (S_1) can be calculated from $S_1=v\times t\times a/\cos 60°$, and the surface area of the rubber fiber (S_2) can be calculated from $S_2=2\pi r\times v\times t$. Here a and r correspond to the width of the MWCNT sheet (with a width of 1 cm) and radius of the electrode, respectively. The layer numbers of CNT sheets (N) were calculated by $N=S_1/S_2$. For a continuous fabrication from one to the other end of the rubber fiber, the layer number of CNT sheet was calculated to be 6.3 and the thickness was ~110 nm (here a single layer of CNT sheet was ~18 nm[S5]).

Calculations of the mass of CNT sheet electrodes. The mass of CNT sheet electrodes can be obtained by multiplying the area and area density of CNT sheet in the electrode. Here the area density of CNT sheet was 1.41 μg cm^{-2}[S6]. For instance, for the CNT sheet with thickness of ~110 nm and length of 4 cm, the area is $2\pi r\times 4$, and the layer number is 6.3, so the mass is 11.2 μg.

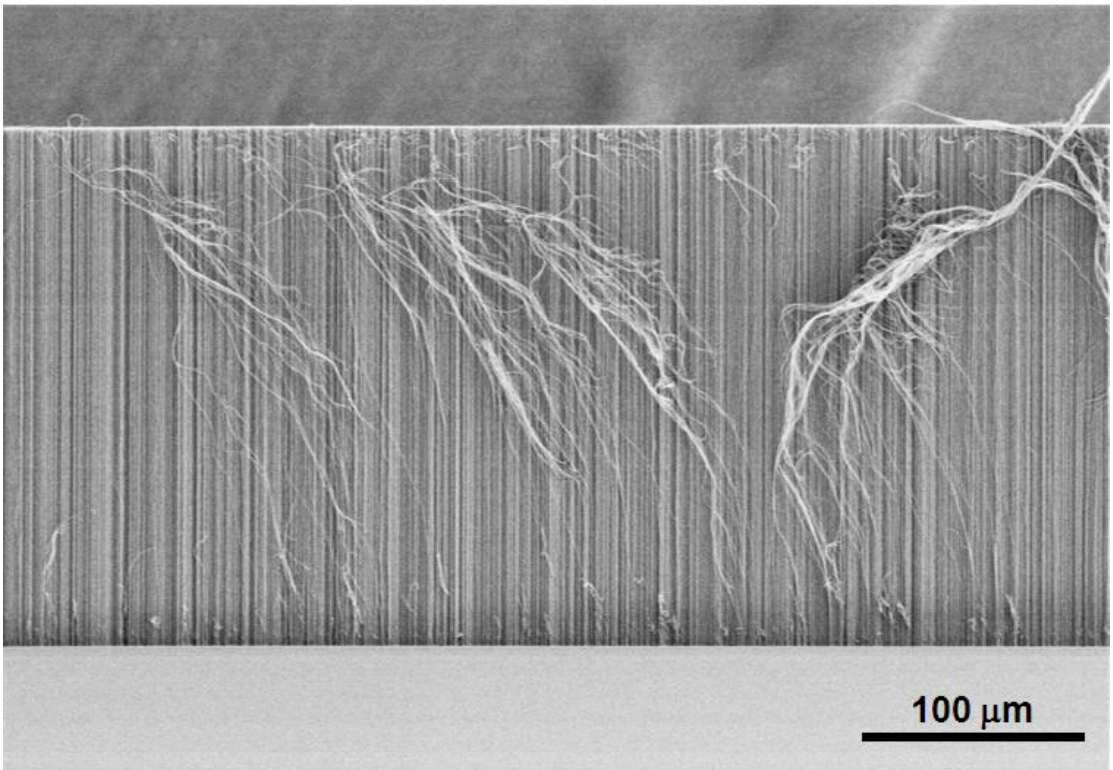

Figure S1. Scanning electron microscopy (SEM) image of a spinnable CNT array.

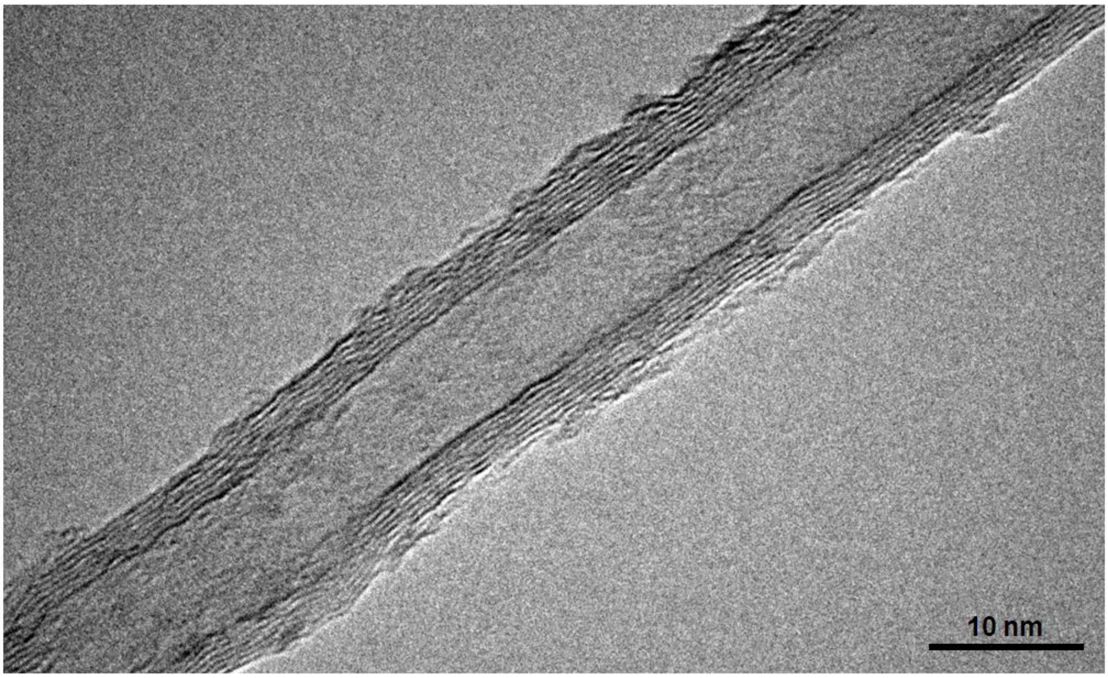

Figure S2. Transmission electron microscopy image of a CNT.

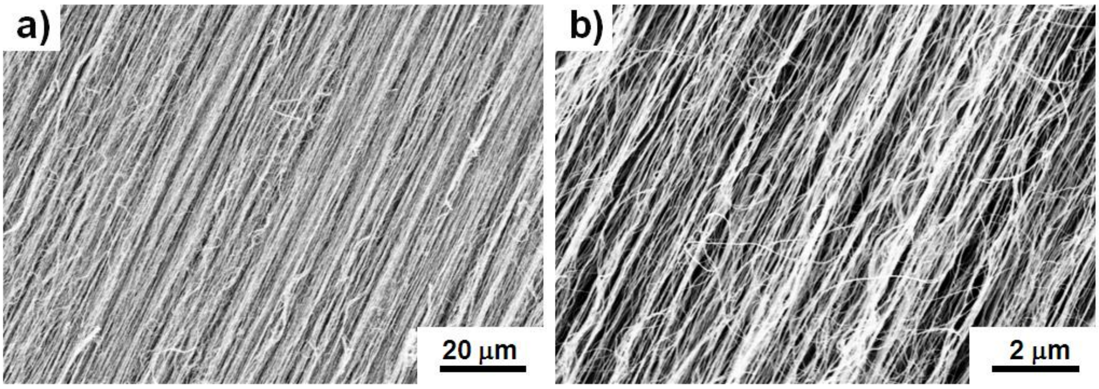

Figure S3. SEM images of the CNT sheet electrode at different maganificaitons.

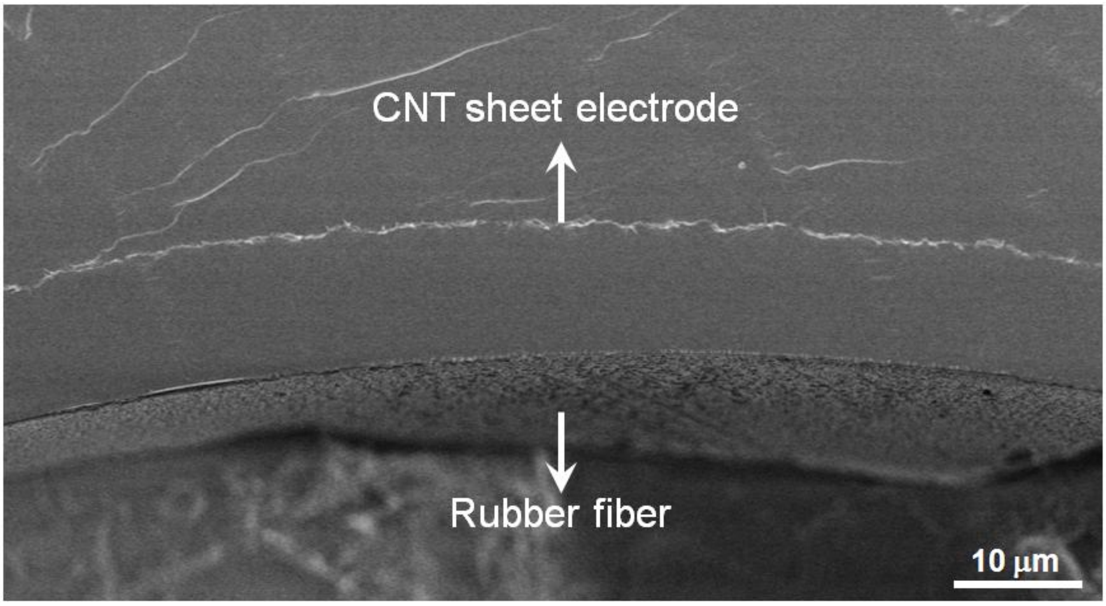

Figure S4. Cross-sentional SEM image of the fiber-shaped supercapacitor.

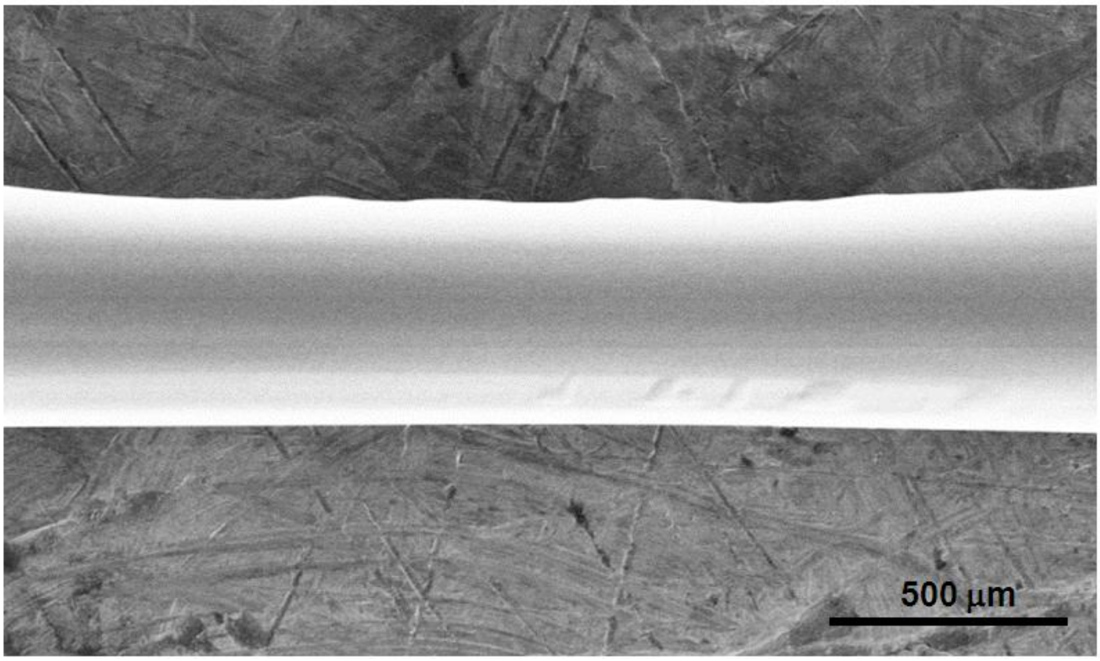

Figure S5. SEM image of the fiber-shaped supercapacitors after stretch by 100%.

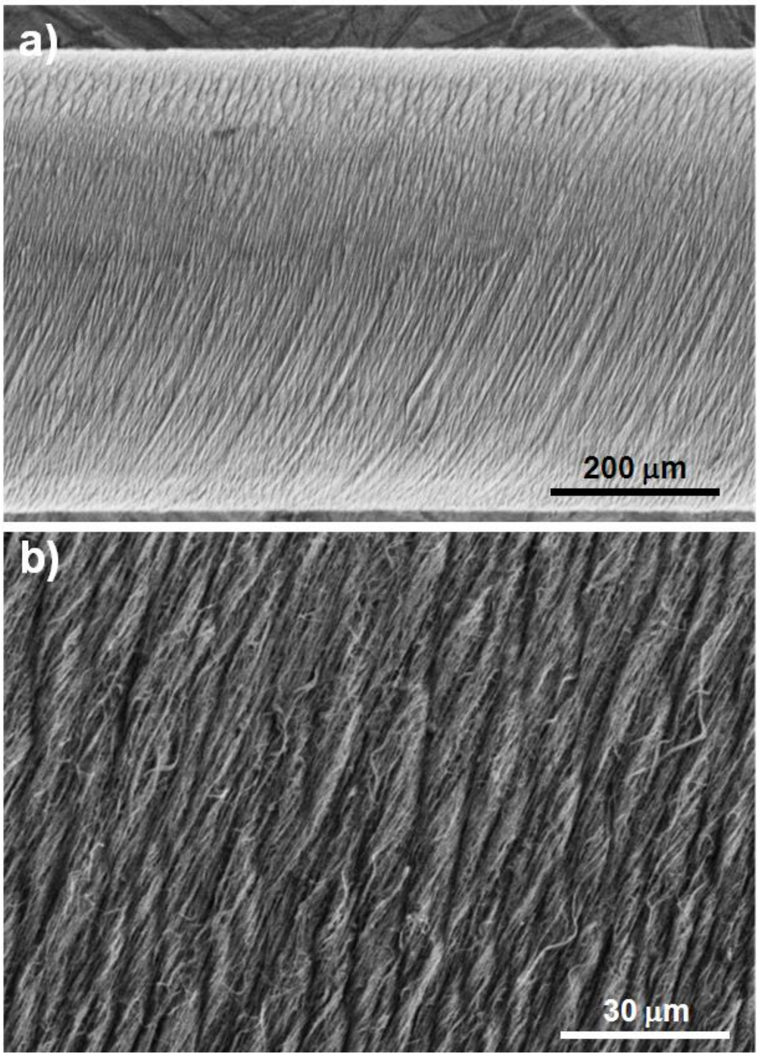

Figure S6. SEM images of the inner CNT sheet electrode after stretch by 100% for 100 cycles at different maganificaitons.

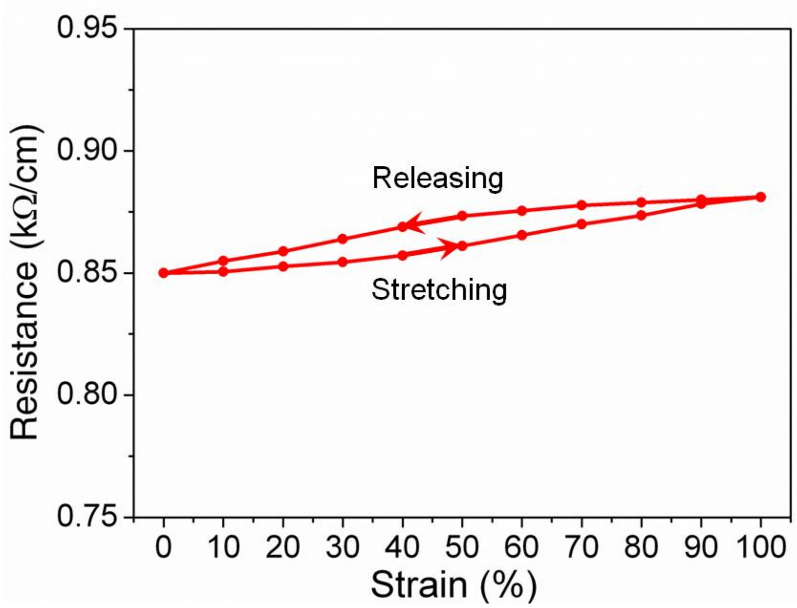

Figure S7. Resistance change of the inner CNT sheet electrode (responding to Figure 1c, thickness of 330 nm) during the stretching and releasing process with a strain of 100%.

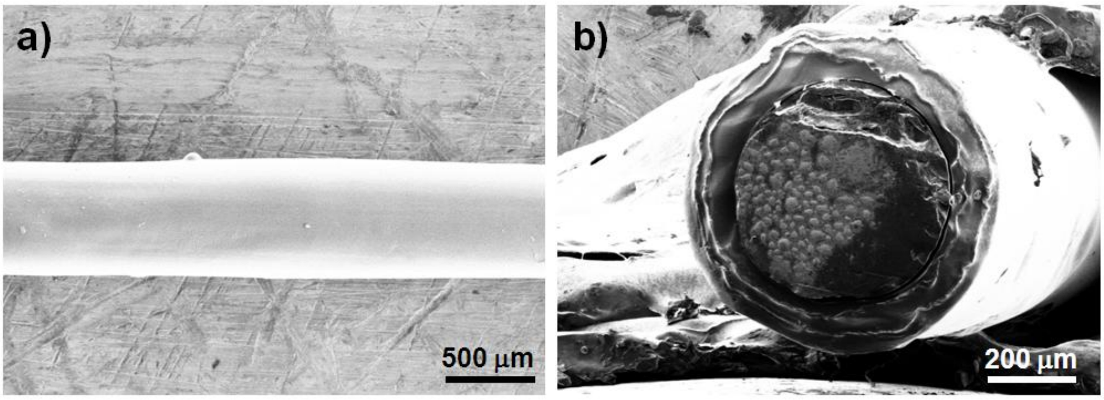

Figure S8. A top (**a**) and cross-sectional (**b**) view of the fiber-shaped supercapacitor after 1000 charge-discharge electrochemical cycles.

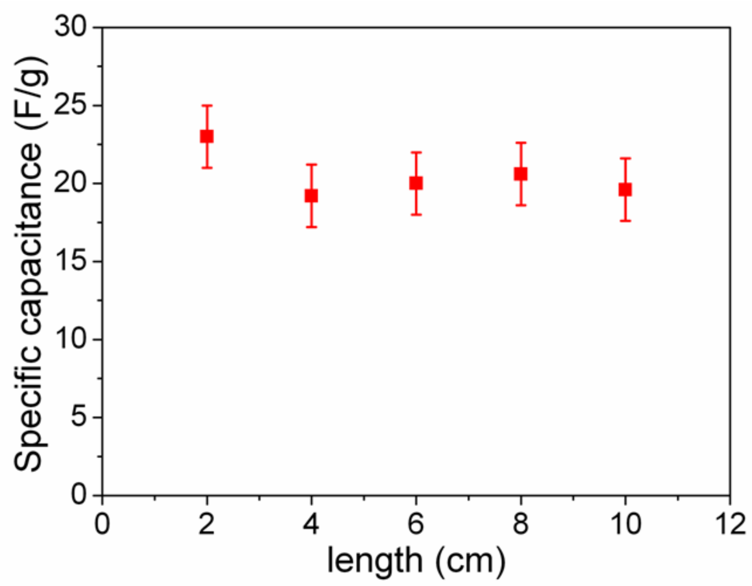

Figure S9. Dependence of specific capacitance on supercapacitor length.

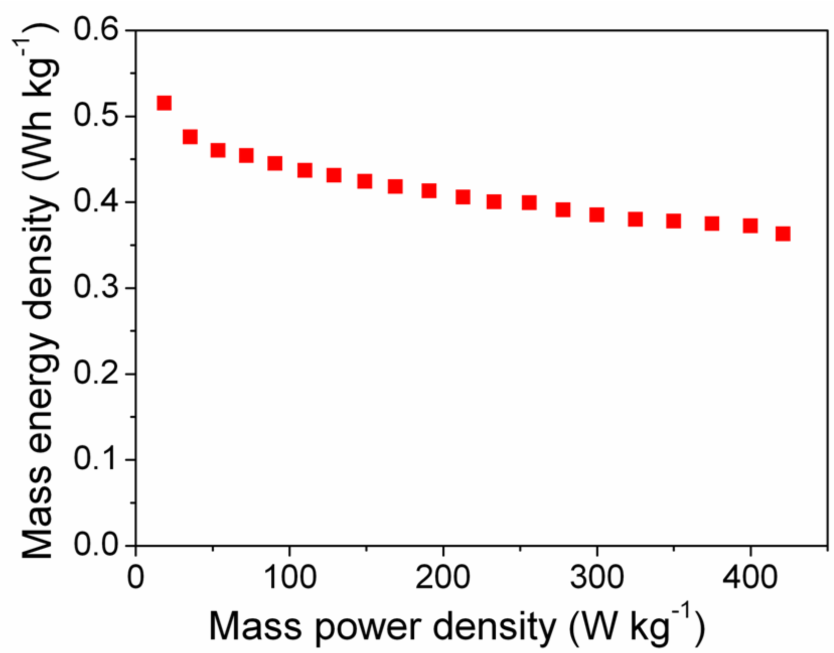

Figure S10. Ragone plots of mass energy and power densities for the fiber-shaped supercapacitor with the increasing charge-discharge current density from 0.05 to 1 A/g. Here the mass of the electrolyte has not been considered.

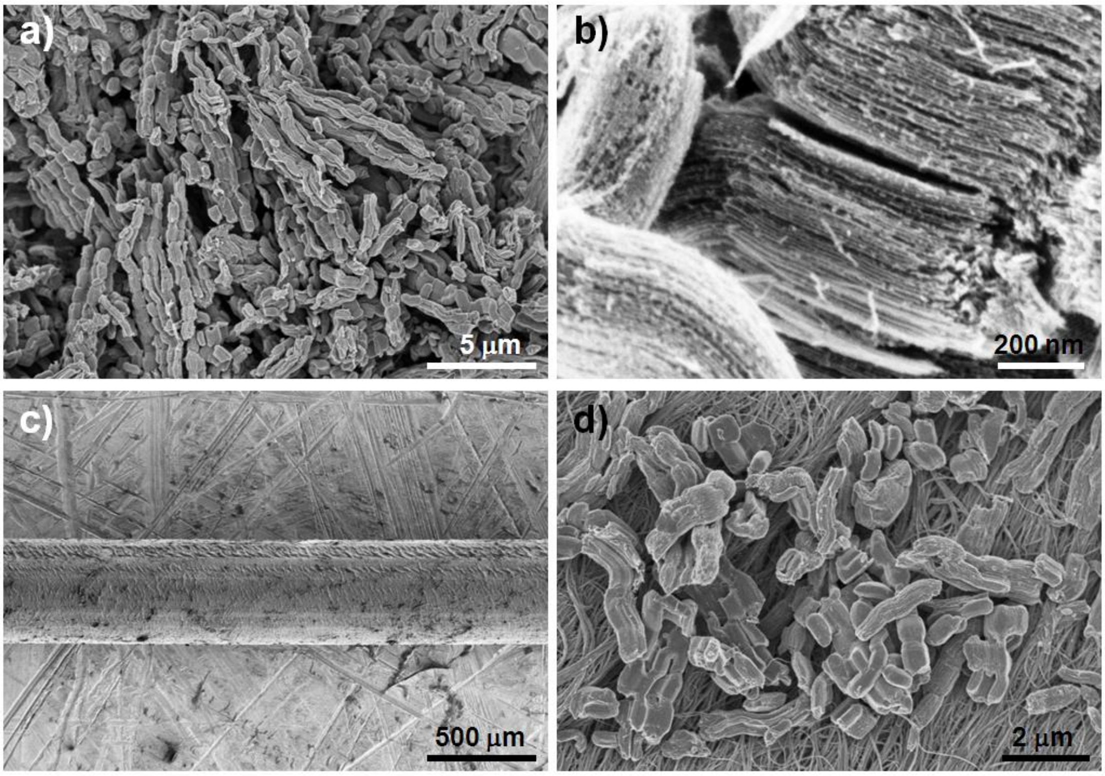

Figure S11. **a** and **b**, SEM images of the ordered mesoporous carbon (OMC) at different maganificaitons. **c** and **d**, SEM images of the inner CNT/OMC composite with the OMC weight percentage of 50%.

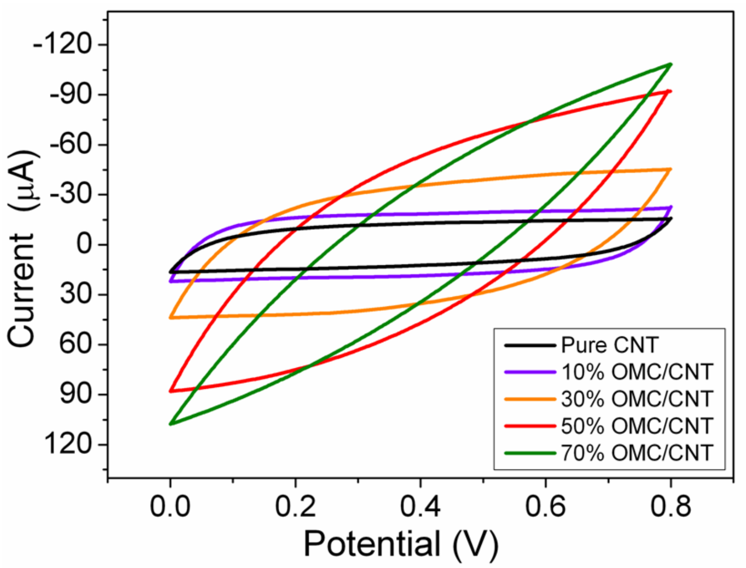

Figure S12. CV curves of fiber-shaped supercapacitors with bare CNT sheet and CNT/OMC composite films with different weight percentages (10%, 30%, 50%, and 70%) of OMC as electrodes at a potential range of 0 to 0.8 V with a scan rate of 50 mV s^{-1} in H_3PO_4-PVA gel electrolyte.

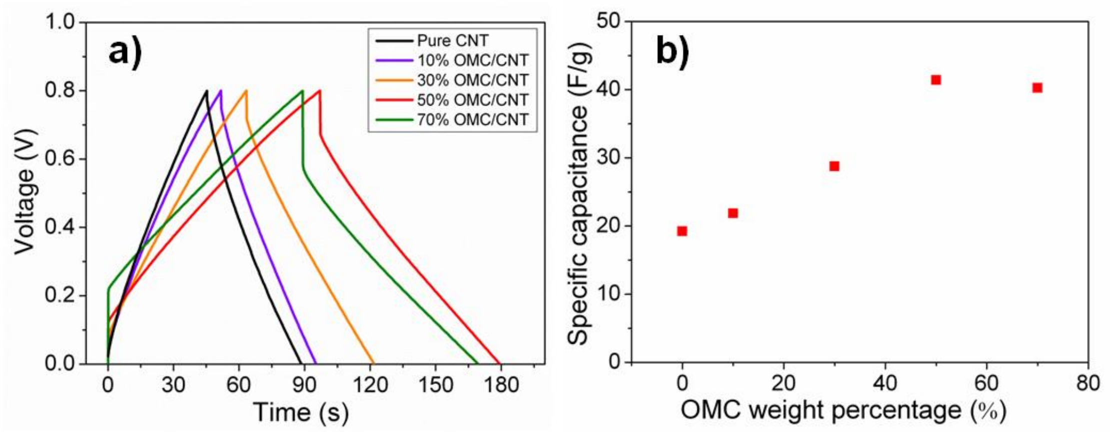

Figure S13. **a,** Galvanostatic charge-discharge curves of fiber-shaped supercapacitors with bare CNT sheet and composite films with different OMC weight percentages (10%, 30%, 50%, and 70%) as electrodes at a current density of 0.1 A/g. **b**, Dependence of specific capacitance on OMC weight percentages. The specific capacitance was calculated from the charge-discharge curves at the current density of 0.1 A/g in H_3PO_4-PVA gel electrolyte.

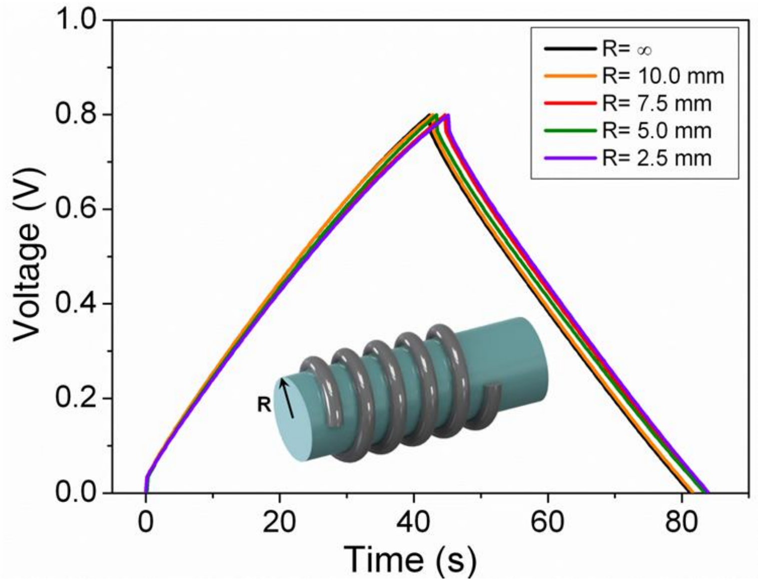

Figure S14. Galvanostatic charge-discharge curves of the fiber-shaped supercapacitor without bending (i.e., the one labeled with ∞) and being bent with different radii of curvatures that are shown in the inserted scheme at a current density of 0.1 A/g.

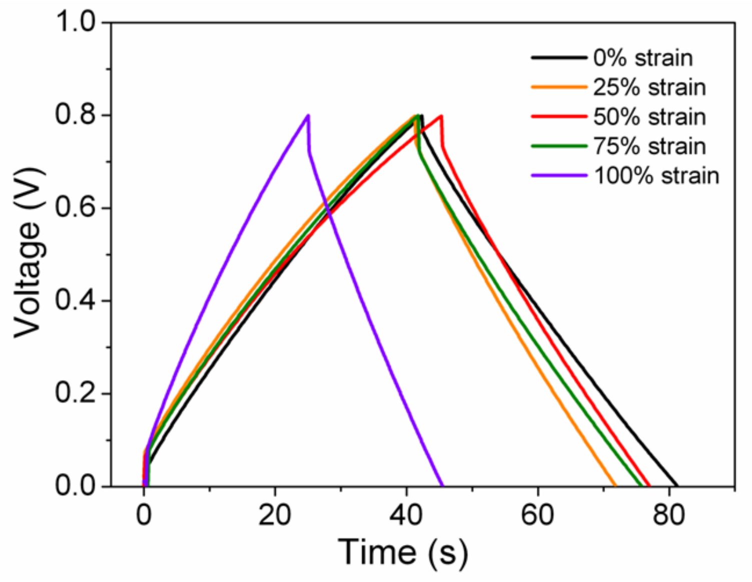

Figure S15. Galvanostatic charge-discharge curves of fiber-shaped supercapacitors with increasing strains from 0 to 100% at a current density of 0.1 A/g.

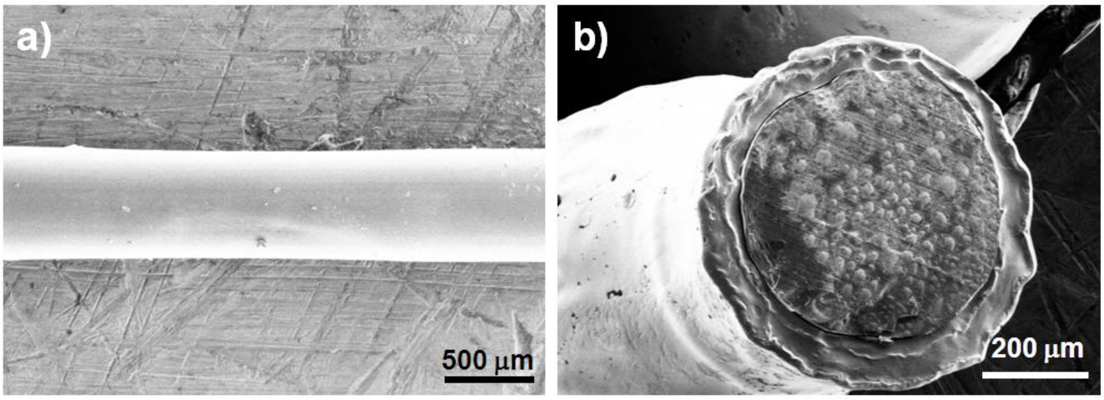

Figure S16. Top (**a**) and cross-sectional (**b**) views of the fiber-shaped supercapacitor after being stretched by 100% for 100 cycles.

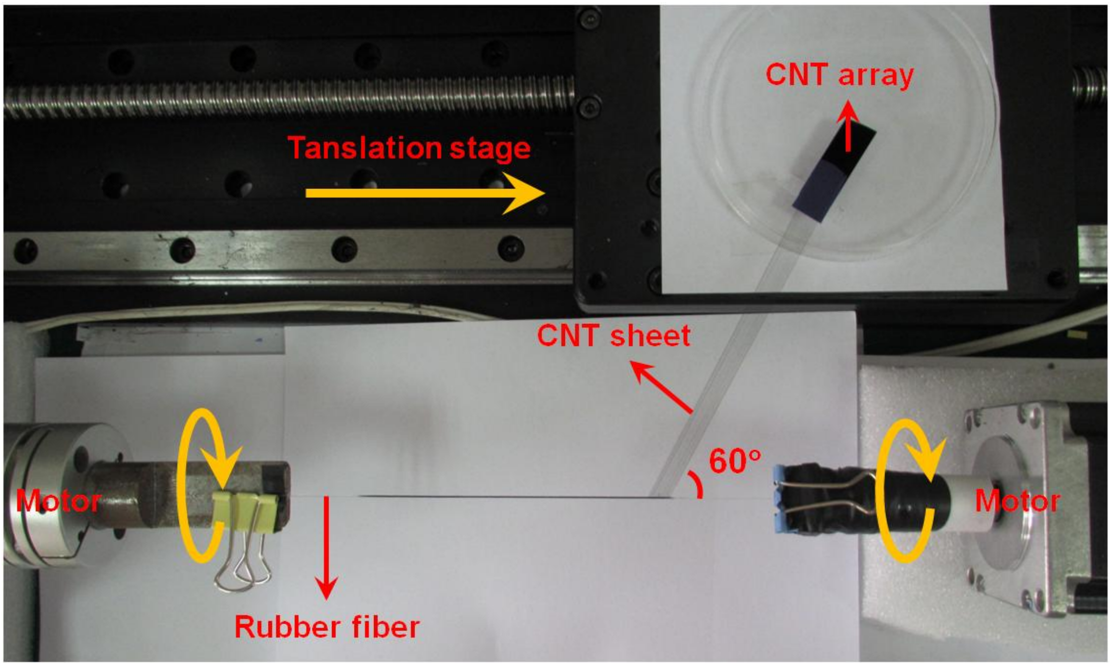

Figure S17. Photograph of the experimental setup to wrap the CNT sheet electrode.

Refencences for the Supporting Information

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[S2]. X. Sun, T. Chen, Z. Yang, H. Peng, *Acc. Chem. Res.* **2012**, *46* , 539-549.

[S3]. Z. Yang, T. Chen, R. He, G. Guan, H. Li, L. Qiu, H. Peng, *Adv. Mater.* **2011**, *23*, 5436-5439.

[S4]. H. Lin, L. Li, J. Ren, Z. Cai, L. Qiu, Z. Yang, H. Peng, *Sci. Rep.* **2013**, *3*.

[S5]. Z. Yang, T. Chen, R. He, G. Guan, H. Li, L. Qiu, H. Peng, *Adv. Mater.* **2011**, *23*, 5436.

[S6]. L. Qiu, X. Sun, Z. Yang, W. Guo, H. Peng, *Acta Chim. Sinica* **2012**, *70*, 1523.