Supporting Information

Stretchable supercapacitor based on a cellular structure

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Experimental section

Characterization. The structures were characterized by scanning electron microscopy (SEM) (Hitachi FE-SEM S-4800), transmission electron microscope (TEM) (JEOL JEM-2100F) and Raman spectrometer (XploRA, HORIBA JobinYvon, France). The photographs were taken by a digital camera (Nikon, J1). The electrical conductivities were measured by Keithley Model 2400 Source Meter. The thicknesses were obtained using a surface profiler (Veeco, Dektak 150). The weight of the carbon nanotube (CNT) film was measured by using a microbalance (Sartarious SE2). The mechanical properties of the fibers were tested by a HY0350 Table-top Universal Testing Instrument with a tensile rate of 1 mm/min. The films were fixed on a paper hole with a gauge length of 5 mm by silver paste. Galvanostatic charge-discharge curves and cyclic voltammograms were conducted at an electrochemical analyzer system (CHI 660D). Cyclic galvanostatic charge-discharge measurements were characterized from an ARBIN electrochemical workstation (MSTAT-5 V/10 mA/16 Ch).

Calculation of electrochemical parameters. The areal capacitance ($C_A$), gravimetric capacitance ($C_M$) and volumetric capacitance ($C_V$) were calculated by the following equations: $C_A = 2 \times I \times \Delta t / (S \times \Delta U)$, $C_V = 2 \times I \times \Delta t / (V \times \Delta U)$, $C_M = 2 \times I \times \Delta t / (m \times \Delta U)$, where $I$, $\Delta t$ and $\Delta U$ are discharge current, discharge time and voltage window, respectively. $S$, $m$ and $V$ are the total surface area, mass and volume of the electrode, respectively. Here, $m$ was measured by using a microbalance, typically 0.619 mg for a CNT film with an area of 0.36 cm$^2$. The weight of polyaniline (PANI) in the PANI/CNT composite film had been calculated by the total Faradic charge consumed in the electropolymerization reaction by assuming an average of 2.5 electrons per monomer.\textsuperscript{51} For instance, when the weight of aligned CNT film was 0.619 mg, the weight of PANI at the content of 5% can be calculated as 0.0295 mg. The volume ($V$) of the electrode can be obtained by $V = L \times W \times H$, where $L$, $W$, and $H$ represent the length, width and thickness of the CNT film, respectively. In a symmetrical supercapacitor, we obtain $1/C = 1/C_1 + 1/C_2 = 2/C_E$, where $C$ is the capacitance of supercapacitor and $C_E$ is the capacitance of a single electrode. Therefore, the gravimetric capacitance of supercapacitor $C_S = 0.25 \times C_M$. The gravimetric energy density ($E_M$) and gravimetric power density ($P_M$) can be expressed as: $E_M = 0.5 \times C_S \times U^2 = 0.125 \times C_S \times U^2$ and $P_M = E_M \times 3600/\Delta t$. 

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Reference
Figure S1. SEM images of the CNT film by side (a) and top (b) views.
**Figure S2.** TEM image of the CNTs.
Figure S3. Raman spectrum of the CNTs.
Figure S4. Stress-strain curve of a CNT film without the cellular structure.
Figure S5. Dependence of strain on $a_0/b_0$ with $w$ of 1 mm (inserted, the schematic of a basic unit of the stretchable CNT film).
Figure S6. Nyquist plot of the supercapacitors with the increasing thickness of the CNT film electrode.
Figure S7. Dependence of electrical resistance on thickness of the CNT film.
Figure S8. Dependence of weight of the film on thickness.
Figure S9. Dependence of specific capacitance on current density.
Figure S10. Cyclic voltammograms for the supercapacitor at increasing scan rates from 20 to 500 mV/s.
Figure S11. Dependence of specific capacitance on cycle number at a current density of 1 A/cm². Here \( C \) and \( C_0 \) correspond to specific capacitances before and after stretching for different cycles, respectively.
Figure S12. Galvanostatic charge-discharge curves for the supercapacitor with (red line) and without (black line) the cellular structure at the same area and current density of 0.5 mA/cm².
**Figure S13.** Galvanostatic charge-discharge profiles with increasing strains from 0 to 140%.
Figure S14. Cyclic voltammograms with increasing bending angles from 0 to 180°.
Figure S15. Dependence of energy and power densities on strain. Here $E_0$ and $E$ correspond to the energy densities before and after stretching, respectively; $P_0$ and $P$ correspond to the power densities before and after stretching, respectively.
Figure S16. SEM image of the PANI/CNT composite film.
**Figure S17.** TEM image of the PANI/CNT composite.
**Figure S18.** Electrochemical performances of the supercapacitors based on the CNT/PANI composites with the PANI weight percentage of 5 wt%. (a) Galvanostatic charge-discharge curve at a current density of 2 mA/cm$^2$. (b) Cyclic voltammogram at a scan rate of 100 mV/s.