

Supporting Information

A three-dimensionally stretchable high performance supercapacitor

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Experimental section

Characterization. The structures were characterized by scanning electron microscopy (SEM) (Hitachi FE-SEM S-4800) and transmission electron microscope (TEM) (JEOL JEM-2100F). 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 (Sartorius SE2). The oxygen plasma treatment was performed in an oxygen microwave (Plasma System 690, PVA Tepla). Galvanostatic charge-discharge curves and cyclic voltammograms were conducted at an electrochemical workstation (CHI 660D). Cyclic galvanostatic charge-discharge measurements were characterized from an ARBIN electrochemical station (MSTAT-5 V/10 mA/16 Ch).

Calculation of electrochemical parameters of supercapacitors. The areal capacitance (C_A), gravimetric capacitance (C_M) and volumetric capacitance (C_V) are calculated by the following equations: $C_A = 2 \times I \times \Delta t / (S \times U)$, $C_V = 2 \times I \times \Delta t / (V \times U)$ and $C_M = 2 \times I \times \Delta t / (m \times U)$. Here I , Δt and 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. In this work, m was measured by using a microbalance, typically 0.619 mg for a CNT film with an area of 0.36 cm². The volume 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, the specific capacitance of supercapacitor $C_S = 0.25 \times C_E$ where C_E is the capacitance of a single electrode. The volumetric energy density (E_V) and volumetric power density (P_V) of the supercapacitor can be expressed as: $E_V = 0.5 \times C_S \times U^2 = 0.125 \times C_V \times U^2$ and $P_V = E_V \times 3600 / \Delta t$.

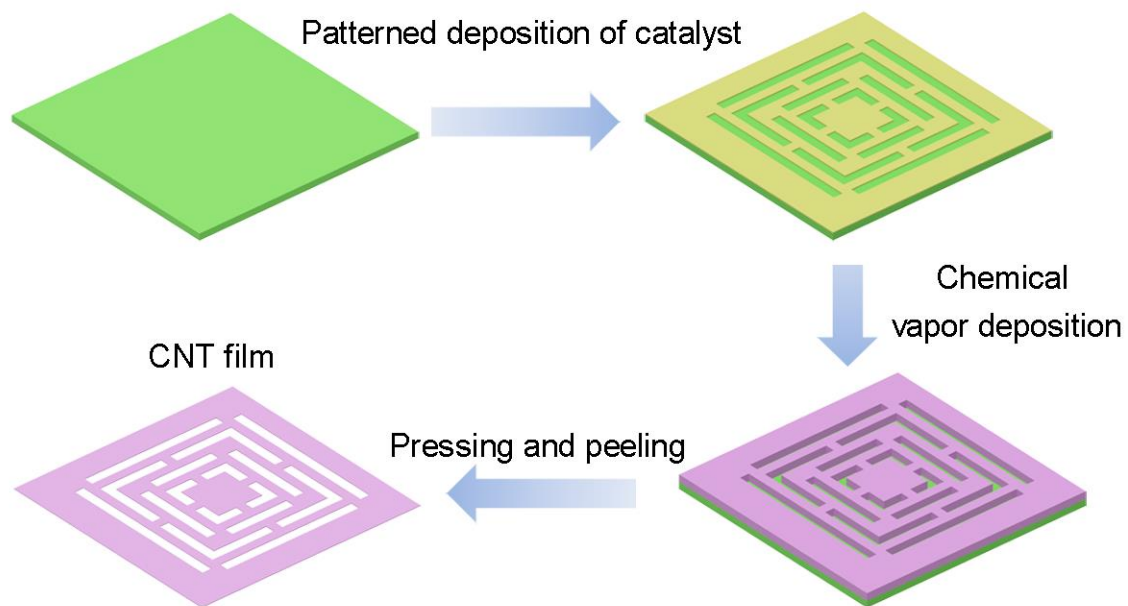


Figure S1. Schematic illustration to the preparation of a pyramid-shaped CNT film. The preparation included the selected deposition of catalyst and growth of aligned CNT array, followed by pressing and peeling to obtain the desired CNT film.

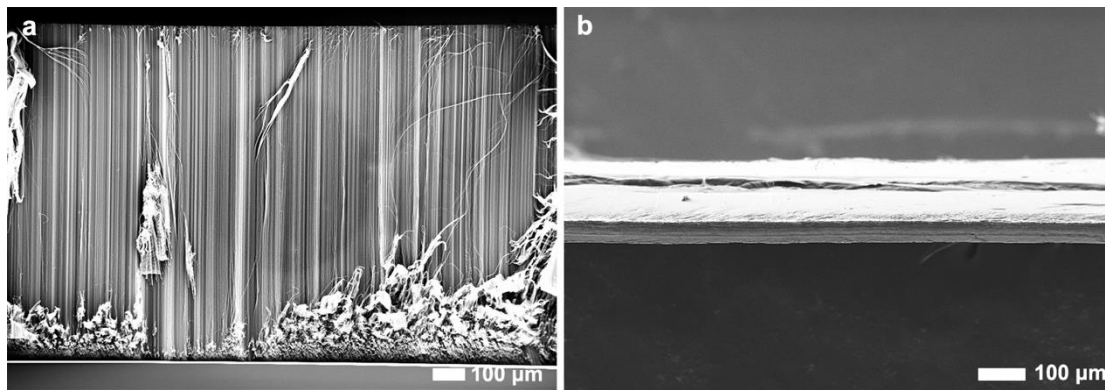


Figure S2. SEM images of the CNT array before (a) and after pressing (b).

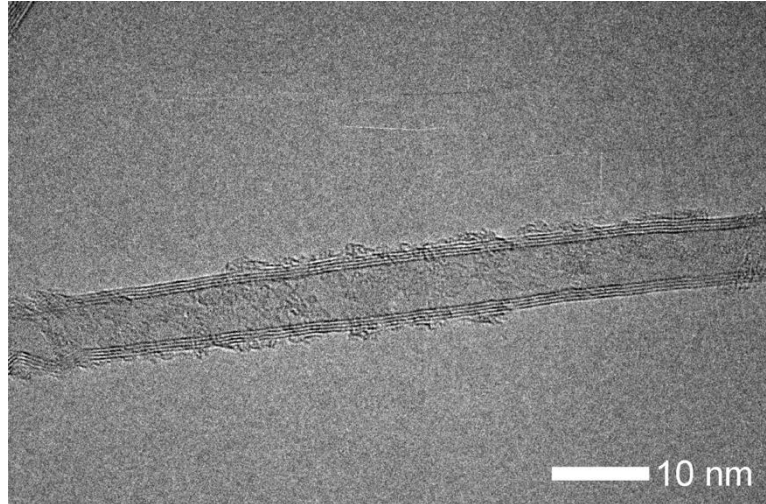


Figure S3. TEM image of a CNT.

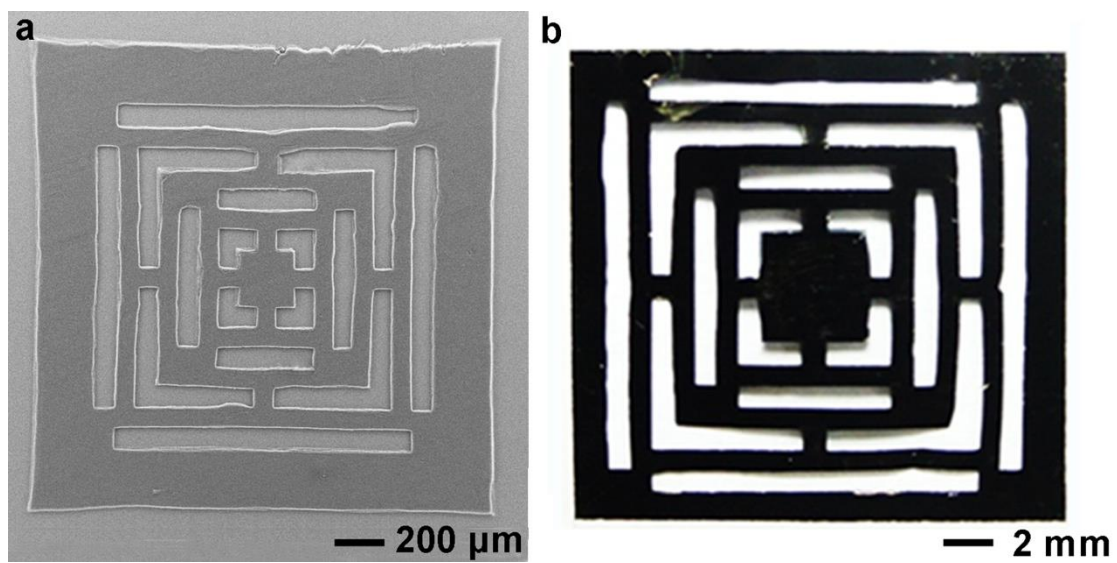


Figure S4. (a) SEM image of a pyramid-shaped CNT film with widths of millimeters. (b) Photograph of a pyramid-shaped CNT film with widths of centimeters.

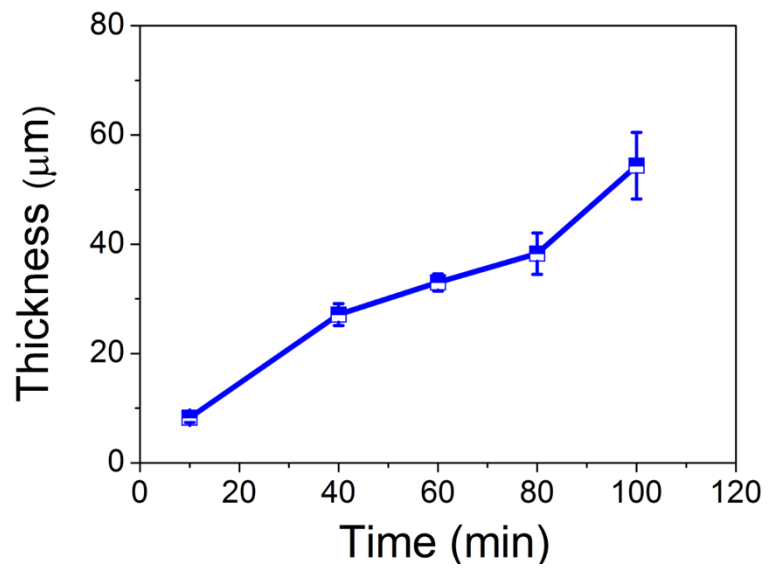


Figure S5. Dependence of the CNT film thickness (after pressing) on the growth time.

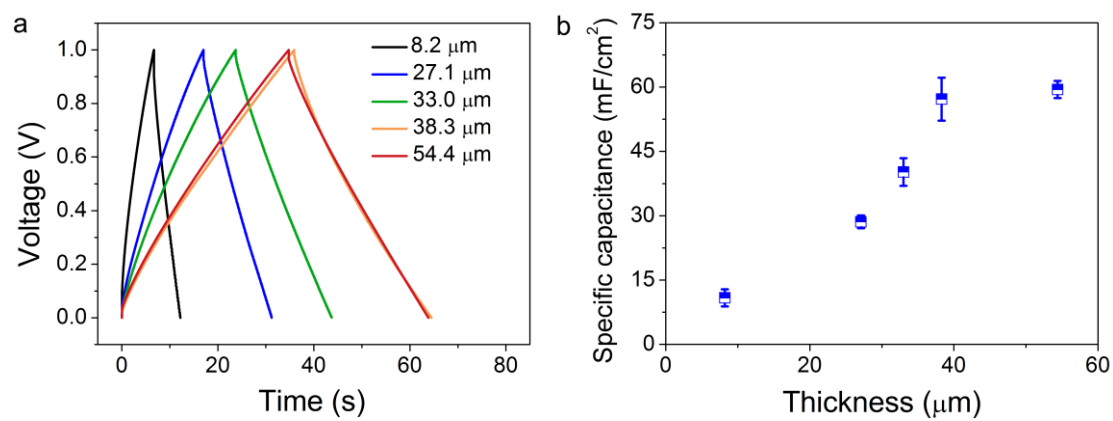


Figure S6. (a) Galvanostatic charge-discharge profiles with increasing thicknesses of the CNT film electrodes at the current density of $1 \text{ mA}/\text{cm}^2$. (b) Dependence of the specific capacitance on the CNT thickness.

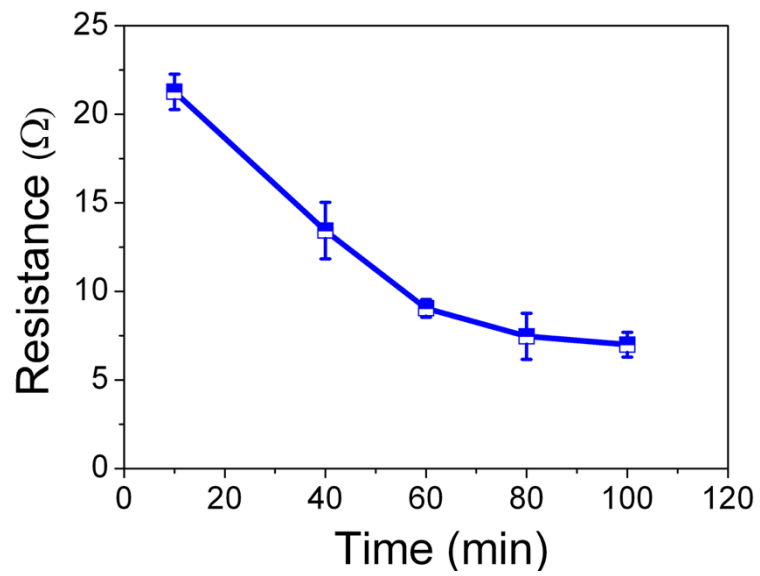


Figure S7. Dependence of the resistance of CNT film on the growth time. The CNT films shared a width of 2 mm and length of 5 mm.

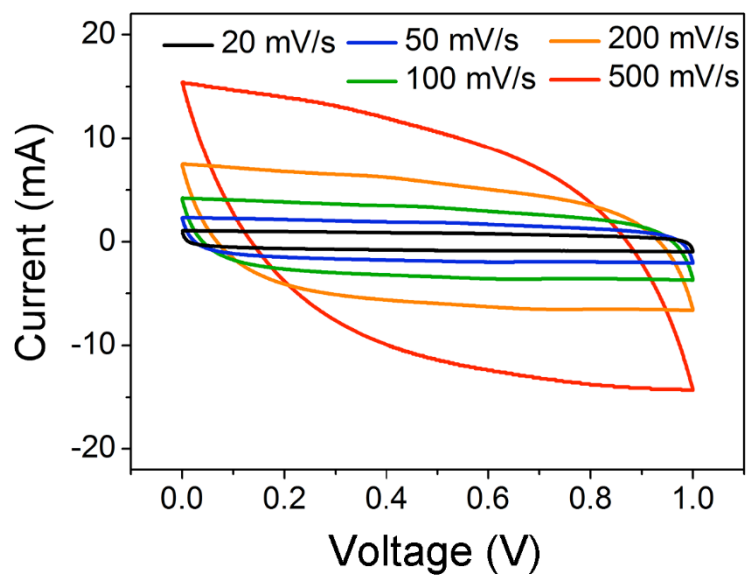


Figure S8. Cyclic voltammograms at increasing scan rates from 20 to 500 mV/s.

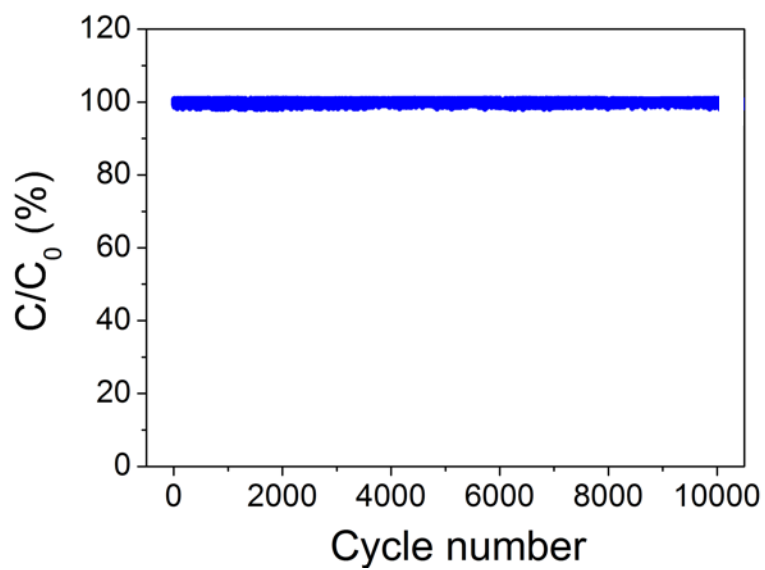


Figure S9. Dependence of the specific capacitance on the cycle number at a current density of 1 A/cm^2 . Here C_0 and C correspond to specific capacitances before and after cycling, respectively.

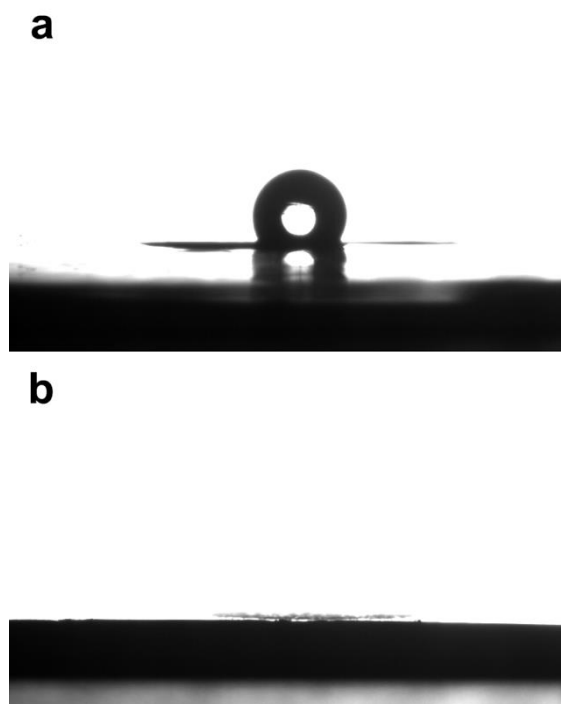


Figure S10. The surface properties of a CNT film before and after oxygen plasma treatment. **(a)** Hydrophobic. **(b)** Hydrophilic.

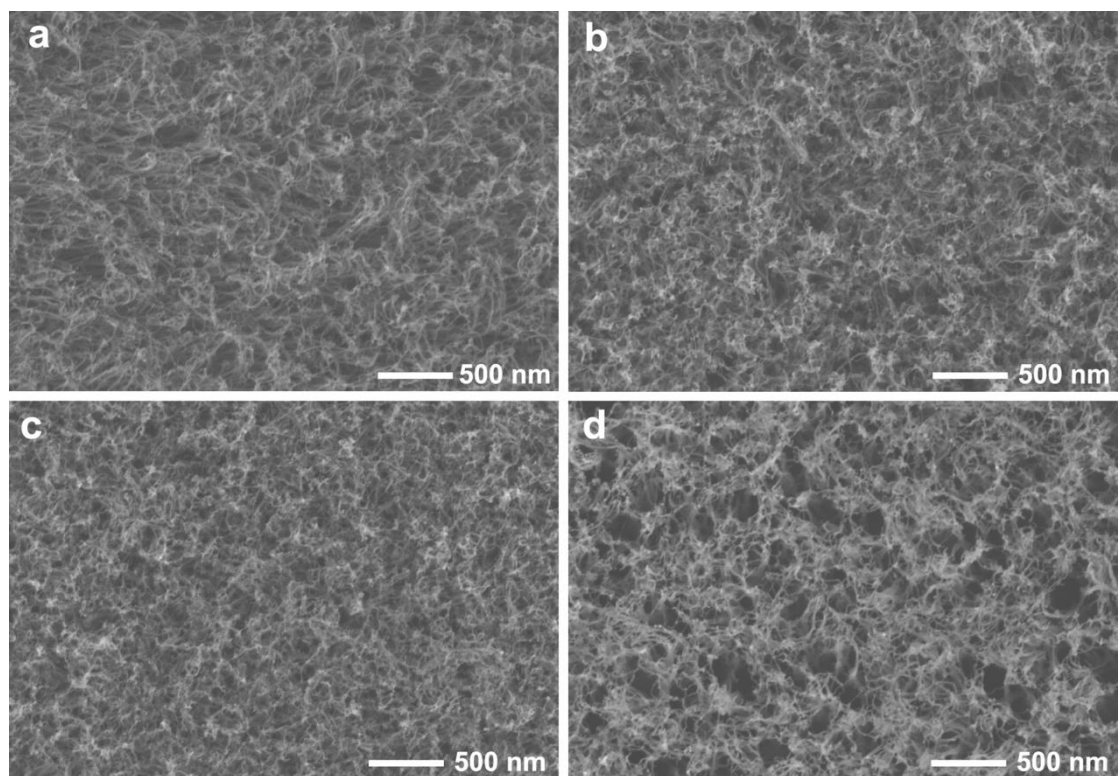


Figure S11. Surface morphologies of CNT films after oxygen plasma treatment with increasing oxygen contents. **(a)** 5.29 wt%. **(b)** 6.67 wt%. **(c)** 8.26 wt%. **(d)** 9.27 wt%.

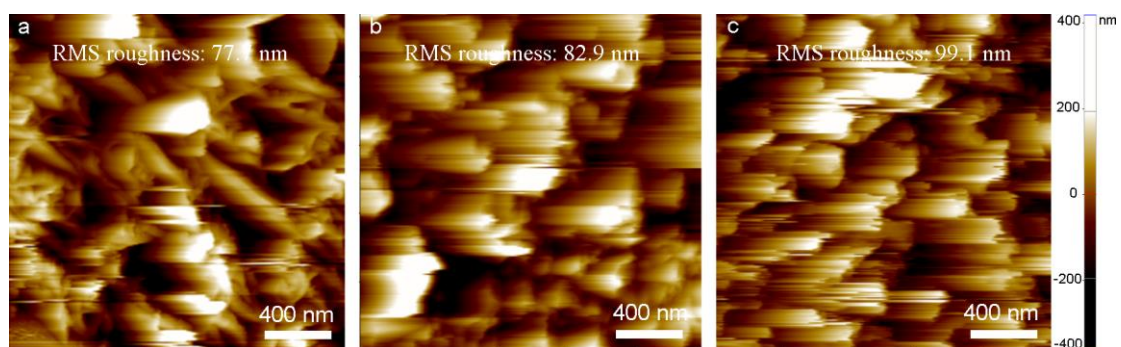


Figure S12. AFM images of the CNT films after oxygen plasma treatment with increasing oxygen contents with the primary film as a comparison. **(a)** 3.06 wt%, **(b)** 5.29 wt% and **(c)** 9.27 wt%.

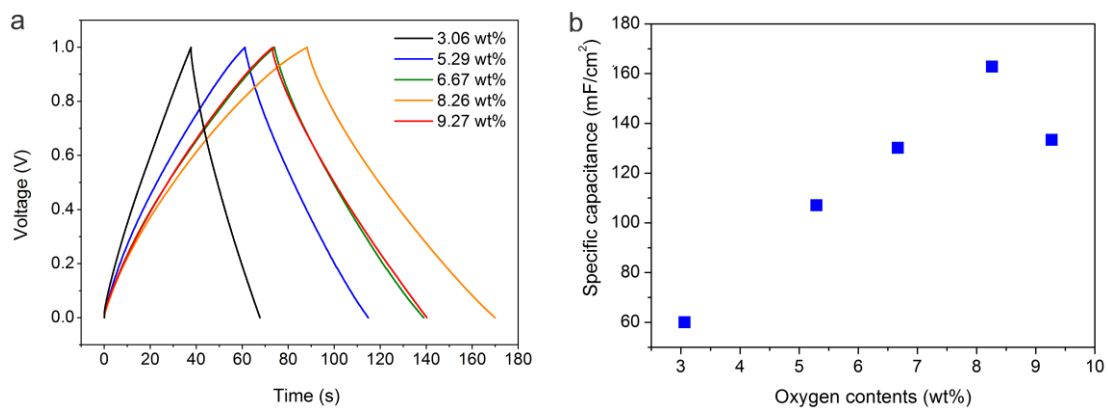


Figure S13. (a) Galvanostatic charge-discharge profiles at increasing oxygen contents. **(b)** Dependence of the specific capacitance on the oxygen content.

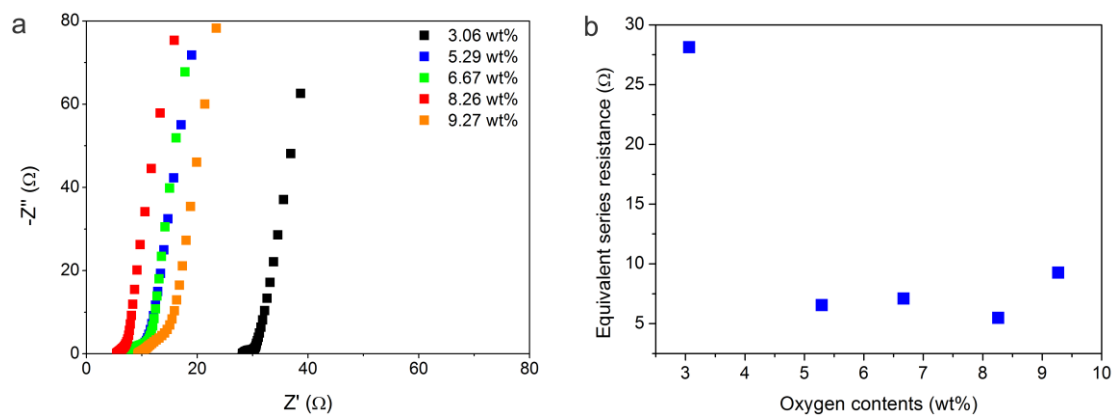


Figure S14. (a) Nyquist plots of the supercapacitors fabricated from CNT films with increasing oxygen contents. **(b)** The equivalent series resistance of the supercapacitors fabricated from CNT films with increasing oxygen contents.

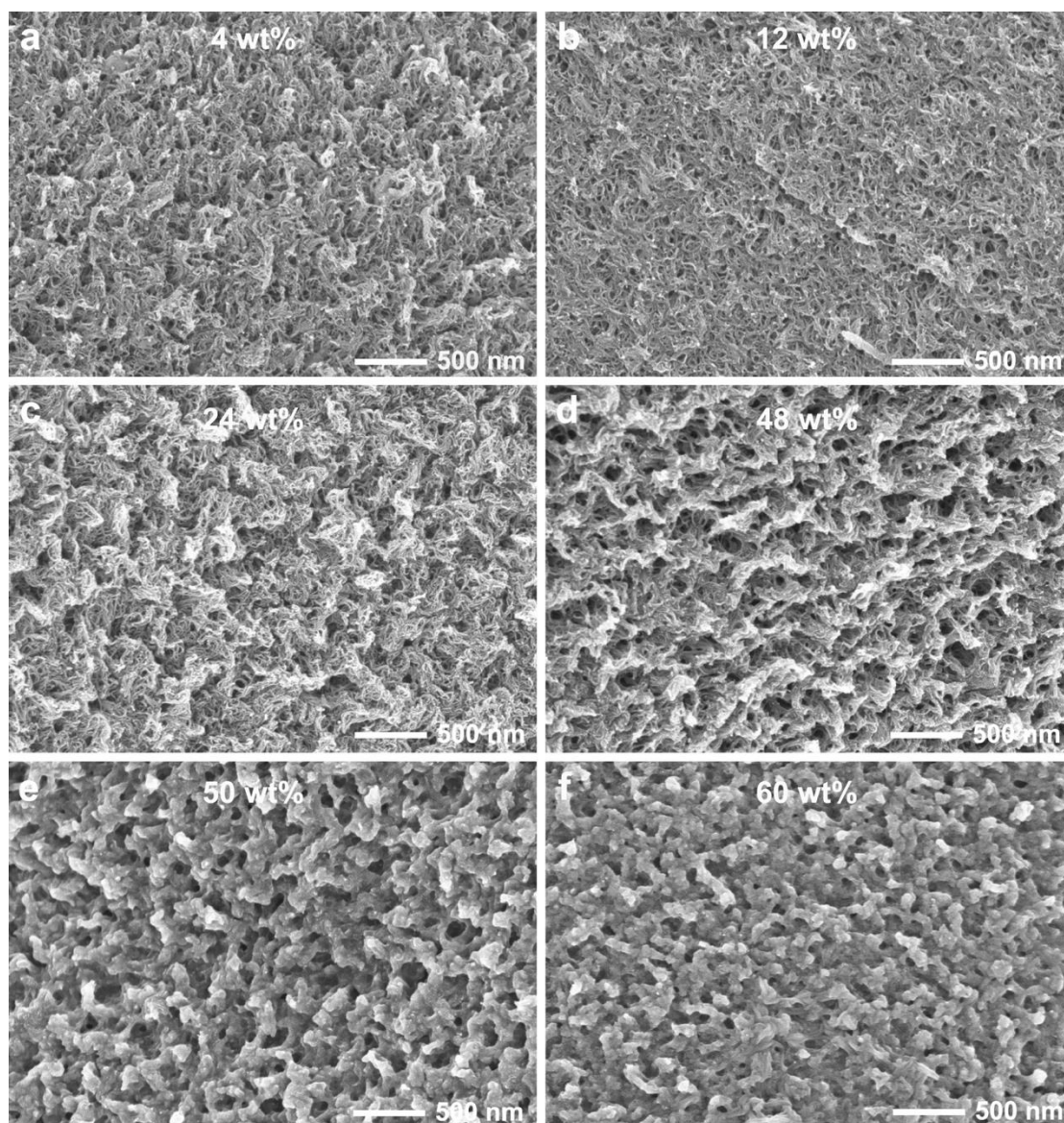


Figure S15. SEM images of the CNT/PANI composite films with increasing PANI weight contents from 4 wt% (a), 12 wt% (b), 24 wt% (c), 48 wt% (d), and 50 wt% (e) to 60 wt% (f).

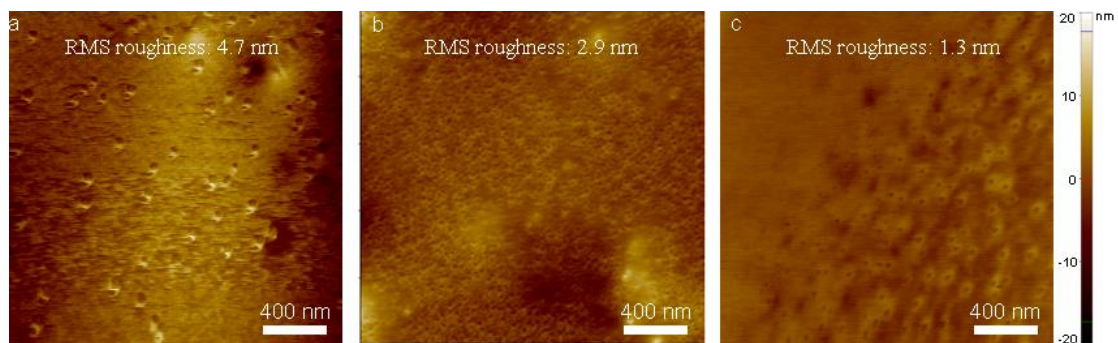


Figure S16. AFM images of the CNT/PANI composite films with increasing PANI weight contents. **(a)** 4 wt%, **(b)** 48 wt% and **(c)** 60 wt%.

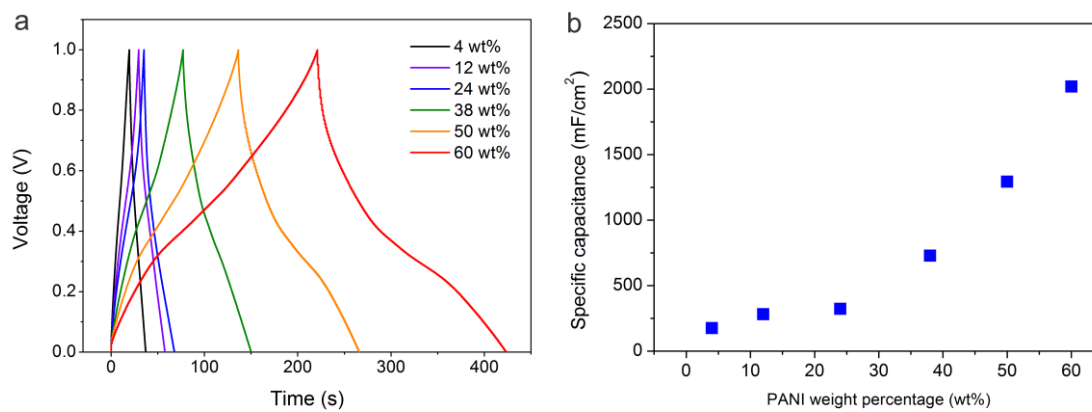


Figure S17. (a) Galvanostatic charge-discharge profiles at increasing PANI weight percentages. (b) Dependence of the special capacitance on the PANI weight percentage.

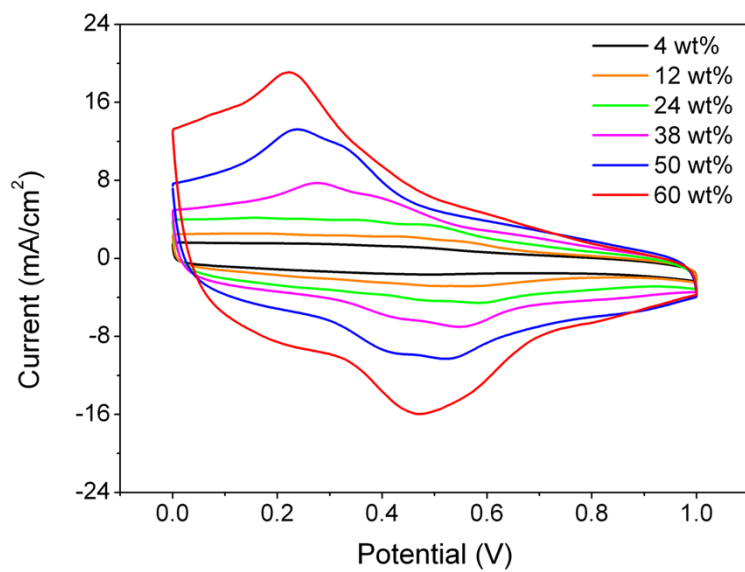


Figure S18. Cyclic voltammograms at increasing PANI weight percentages.

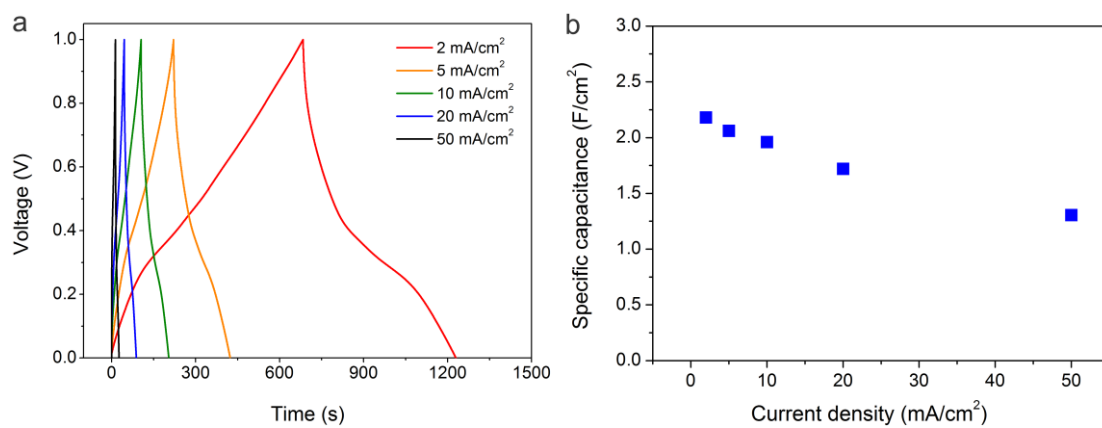


Figure S19. (a) Galvanostatic charge-discharge profiles of the supercapacitor from the CNT/PANI composite film electrode (60 wt% PANI) at increasing current densities. (b) Dependence of the specific capacitance on the current density.

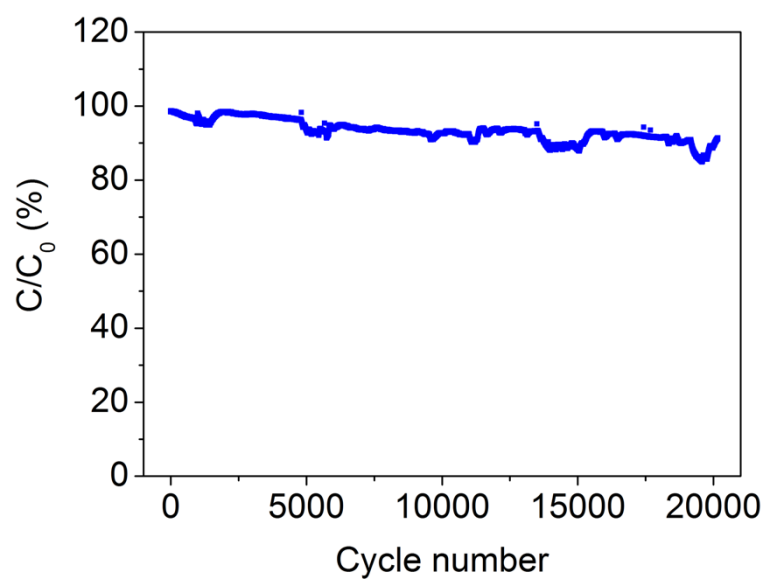


Figure S20. Dependence of the specific capacitance on the cycle number at a current of 1 mA. Here C_0 and C correspond to specific capacitances before and after cycling, respectively.

Table S1. Oxygen contents of CNT films as verified by energy-dispersive X-ray spectra after oxygen plasma treatment with increasing times.

Treatment time	C/wt%	O/wt%
0	96.94	3.06
1	94.71	5.29
2	93.33	6.67
5	91.74	8.26
10	90.73	9.27