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Supporting Information

for Adv. Mater., DOI: 10.1002/adma.201400842

Smart, Stretchable Supercapacitors

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Supporting Information

Video S1: Chromatic transition of a supercapacitor during cyclic voltammetry characterization at a scan rate of 10 mV/s (the video is played at 15 times of the real speed).

Calculation of specific capacitance

According to the definition, the capacitance of the capacitor C is calculated by

$$C = \frac{Q}{U} \tag{1}$$

Q and U correspond to electric charge and voltage, respectively. Q is calculated by Q = It (2) I and t correspond to the current and time during the discharging process. For a

supercapacitor, the two electrodes are connected in series, so $Q_1=Q_2$ and $U_1+U_2=U$. In this work, two electrodes of the supercapacitor are the same, so $U_1 = U_2 = \frac{1}{2}U$. Therefore, it is easy to conclude that $C_1 = C_2 = 2C$. The specific capacitance of electrode (C_{s1}) can be then calculated by

$$C_{s1} = \frac{C_1}{m_1}$$
(3)

where m_1 stands for the mass of the composite electrode.

Experimental section

Spinnable CNT array was grown by chemical vapor deposition. Typically, in a quartz tube furnace, using Fe $(1.2 \text{ nm})/\text{Al}_2\text{O}_3$ (3 nm) on a silicon wafer as catalyst, ethylene as carbon source, and a mixture of Ar and H₂ gases as carrying gas, controlling the growth temperature at 740 °C for 10 to 20 min, the spinnable CNT array can be synthesized.

The gel electrolyte containing PVA and H_3PO_4 with PVA/ H_3PO_4 mass ratio of 1/0.85 was prepared by firstly immersing 1 g of PVA in 9 g of distilled water for 4 h, followed by heating to 90 °C and being maintained for 2 h. After cooling down to room temperature, 1 g of H_3PO_4 aqueous solution (85 wt%) was added to the above solution under stirring for 30 min.

The structures were characterized by SEM (Hitachi, FE-SEM S-4800 operated at 1

kV). The PDMS thickness was obtained from a surface profiler (Veeco, Dektak 150). Galvanostatic charge-discharge characterizations were carried out by an Arbin multi-channel electro-chemical testing system (Arbin, MSTAT-5 V/10 mA/16 Ch). Cyclic voltammograms were made from an electrochemical analyzer system (CHI 660D). The stretchability was characterized by an HY0350 Table-top Universal Testing Instrument. The UV-vis spectra were recorded by UV-2550 from Shimadzu. The resistances of the composite electrode under bending and stretching were traced by Keithley Model 2400 Source Meter.



Figure S1. Schematic illustration to fabrication of the supercapacitor by a side view.



Figure S2. Dependence of specific capacitance on current density at the PANI weight percentage of 90%.



Figure S3. Galvanostatic charge-discharge curves of supercapacitors based on different PANI weight percentages in the electrode at a current density of 8 A/g.



Figure S4. Dependence of the resistance of the sheet along the CNT-aligned direction and specific capacitance on CNT sheet thickness. Here the resistance is calculated from the same area of $1 \text{ cm} \times 1 \text{ cm}$ for the length and width, respectively.



Figure S5. Optical transmittances of CNT sheets with increasing thicknesses.



Figure S6. Dependence of electrical resistance on bent cycle number. R_0 and R correspond to the resistances before and after bending, respectively.



Figure S7. a. Dependence of electrical resistance on strain. R_0 and R correspond to the resistances at 0 and the other strain, respectively. **b.** Dependence of electrical resistance on stretched cycle number at a strain of 100%. R_0 and R correspond to the resistances before and after stretching, respectively.



Figure S8. Chromatic transitions of a supercapacitor at different voltages.