Electronic Supplementary Information (ESI)

Experimental Section

The natural cotton was stretched into the CM with a uniform thickness (ca. 0.5mm) and a specifically cut size as presented in Figure 1b, which was carbonized at 1000 °C for about 2h with flow of 300 sccm Ar gas to get the CCM.

For the fabrication of *all*C-EC, two pieces of CCMs (typically, ca. $1.6 \times 1 \text{ cm}^2$) were used as the electrodes, and a thin CM (ca. 0.4mm in thickness) acted as the separator. Due to the morphologic compatibility of CM and CCM, the sandwich configuration of a CM within two CCMs has sufficient integrity and flexibility for arbitrary deformation. The CCMs will act as both the active material and current collector in the device. After the electrolyte of 1M Na₂SO₄ aqueous solution was spontaneously sucked into the device body, two pieces of Au sheets are in contact with CCMs to connect with electrochemical workstation. The supercapacitor is simply packed with plastic foil to form a proof-of-concept prototype of a device unit for electrochemical measurements.

All electrochemical characterization was carried out using a CHI 660D electrochemical workstation (CH Instruments Inc.) under ambient environment. Cyclic voltammetry (CV) was performed at scan rates of 100 mV s⁻¹ to 50 V s⁻¹. Galvanostatic charge/discharge curves were obtained at various current densities to evaluate the specific capacitance. Electrochemical impedance spectra (EIS) were measured in the frequency range from 100 000 to 1 Hz.

The area-specific capacitance, C_A (μ F cm⁻²), was calculated by the use of

following equation: $C_A = 2I_D/(dV/dt)$, where I_D is the current density ($\mu A \text{ cm}^{-2}$), dV/dt is the slope of the discharge curve (V s⁻¹).^{s1} The mass specific capacitances (C_m) were calculated by using the equation: $C_m = 2(I\Delta t/m\Delta V)$, where *I* is the constant discharge current, Δt is the discharging time, *m* is the mass of one electrode, and ΔV is the voltage drop upon discharging (excluding the IR drop).^{s2}

The morphology of CMs and CCMs was examined by scanning electron microscope (SEM, JSM-7500F) and transmission electron microscope (TEM, JEM-2010). Raman microscope was carried out with a 514 nm wavelength laser. X-ray diffraction (XRD) patterns were obtained by using a Netherlands 1,710 diffractometer with a Cu K α irradiation source ($\lambda = 1.54$ Å), and a self-calibration process was performed with a SiO₂ internal standard sample prior to target measurement. Nitrogen adsorption experiments were conducted at 77K using a TriStarII3020 (Micromeritics Instrument Corporation) apparatus. Before adsorption measurements, the samples were degassed in vacuum at 473K for 3h. The specific surface areas (SSAs) of the freeze-dried samples were calculated by Brunauer Emmett Teller (BET) analyses of their adsorption isotherms.



Fig. S1. Photograph of CCMs cut into different size.



Fig. S2. (a, b) SEM images of the initial cotton with different magnification. (c, d) SEM images of individual cellulose fiber of cotton at different magnification.



Fig. S3. (a, b) SEM images of carbonized cotton. (c, d) SEM image of individual carbonized cellulose fiber of cotton.



Fig. S4. (a) High-resolution TEM image of the carbonized cotton fiber and (b) the corresponding selected area electron diffraction pattern.



Fig. S5. (a) SEM image and (b) the corresponding EDS spectrum of CCMs.



Fig. S6. CV of the *all*C-ECs obtained at scan rate of 50 V s^{-1} .



Fig. S7. CVs of the *all*C-ECs in comparison with the capacitor using gold foil as electrodes, in which gold foils replace the CCMs in Figure 2a.



Fig. S8. Specific capacitance versus discharge current density in the range of 0.216A g^{-1} to 2.16A g^{-1} .



Fig. S9. Electrochemical characterization of the *all*C-ECs for a folding cycle. (a, b) CVs of *all*C-ECs at a scan rate of 30 V s⁻¹ and 50 V s⁻¹, respectively. (c) The plane plot of the impedance of *all*C-ECs. (d) Impedance phase angle versus frequency for an *all*C-ECs under the different folded states.



Fig. S10. CVs of *all*C-ECs at the flat and fully folded states after 1000 folding cycles in comparison with the initial one at the scan rate of 30 V s⁻¹ and 50 V s⁻¹, respectively.

References

s1. Chen, J.; Sheng, K. X.; Luo, P.H.; Li,C.; Shi, G. Q. Adv. Mater. 2012, 24, 4569–4573.

s2. Zhang, L.; Shi, G. Q. J. Phys. Chem. C 2011, 115, 17206–17212.