

## Supporting Information

### Activated Carbon Fiber Paper with Exceptional Capacitive Performance as Robust Electrode for Supercapacitors

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#### Experimental section

**Activation of carbon fiber paper (CFP):** The CFP was activated by a simple electrochemical oxidation approach in a conventional three-electrode cell with a mixed acid of condensed HNO<sub>3</sub> and H<sub>2</sub>SO<sub>4</sub> (v:v = 1:1) as electrolyte. Prior to oxidation, CFP (HCP010, purchased from Hesen Co., USA) with an area of 2 × 4 cm<sup>2</sup> was cleaned by alcohol, and distilled water for three times in turn. Then, the clean CFP was used as the working electrode and electrochemically oxidized with a constant voltage of 2 V for 10 min at room temperature. The counter electrode and reference electrode is a platinum wire and a saturated calomel electrode (SCE) electrode, respectively. Finally, the activated CFP was washed with deionized water and dried at 60 °C. For comparison, the CFP electrodes treated with single H<sub>2</sub>SO<sub>4</sub> and HNO<sub>3</sub> were also prepared in a similar method, which are denoted as S-OCFP and N-OCFP, respectively.

**Material Characterizations:** The morphology, phase and structure of the samples were investigated by scanning electron microscopy (JSM-6330F), X-ray diffraction (XRD, Bruker, D8 ADVANCE) and transmission electron microscope (JEM2010-HR). The composition of the samples was studied by X-ray photoelectron spectroscopy (XPS, ESCALab250), and Raman Spectroscopy (Renishaw inVia). Nitrogen adsorption/desorption isotherms of the samples at 77 K were collected on an ASAP 2020 V3.03 H instrument. Before the BET test, the CFP and OCFP were cut into small pieces and outgassed at 100 °C for 5 h under flowing nitrogen. For methylene blue

(MB)-absorption experiments, the samples were immersed in a 2 mg L<sup>-1</sup> MB aqueous solution under constant stirring in dark, and UV-vis absorption spectra of the MB solutions after treated for 12 h were collected by a UV-vis-NIR spectrophotometer (UV, Shimadzu UV-3150). Conductivities of all samples were measured by a standard four-probe method using a physical property measurement system (ST2253).

**Electrochemical measurements:** All the electrochemical tests were performed on a CHI 760D electrochemical workstation (Chenhua, Shanghai) with a Pt mesh (100 mesh, 1 cm<sup>2</sup>) with platinum black as counter electrode and a saturated calomel electrode (SCE) as reference electrode. All electrodes were prepared by directly using a piece of the CFP or the OCFP with an area of 0.5 × 2 cm<sup>2</sup>. The area immersed in electrolyte was controlled to be 0.5 × 1 cm<sup>2</sup>. The areal density of the OCFP is 13.2 mg cm<sup>-2</sup>. The electrolyte is a 1 M H<sub>2</sub>SO<sub>4</sub> aqueous solution. Electrochemical impedance spectroscopy of the samples was carried out at their open-circuit potential from a frequency of 100000 to 1 Hz, with a potential perturbation of 10 mV.

## Calculations:

### 1. Single Electrode:

Areal capacitances of all electrodes were calculated from their CVs according to the following equation:

$$C_A = \frac{Q}{\Delta V \cdot S} \quad (1)$$

where  $C_A$  (F cm<sup>-2</sup>) is the areal capacitance,  $Q$  (C) is the average charge during the charging and discharging process,  $\Delta V$  (V) is the potential window and  $S$  (cm<sup>2</sup>) is the area of OCFP electrode.

Alternatively, areal capacitances of electrodes were measured by galvanostatic charge/discharge method based on the following equation:

$$C_A = \frac{I \times \Delta t}{\Delta V \cdot S} \quad (2)$$

where  $C_A$  (F cm<sup>-2</sup>) is the areal capacitance,  $I$  (A) is the constant discharging current,

$\Delta t$ (s) is the discharging time,  $\Delta V$  (V) is the potential window, and  $S$  (cm<sup>2</sup>) is the area.

## 2. OCFP//OCFP-SCs:

The cell (device) capacitance ( $C_{cell}$ ) and areal capacitance of the OCFP//OCFP-SC devices were calculated from their CVs according to the following equation:

$$C_{cell} = \frac{Q}{\Delta V} \quad (3)$$

$$C_A = \frac{C_{cell}}{S} = \frac{Q}{S \times \Delta V} \quad (4)$$

where  $Q$  (C) is the average charge during the discharging process in the applied current,  $S$  (cm<sup>2</sup>) is the area of the whole device,  $\Delta t$  (s) is the discharging time,  $\Delta V$  (V) is the voltage window.

Alternatively, the cell (device) capacitance ( $C_{cell}$ ) and areal capacitance ( $C_s$ ) was estimated from the slope of the discharge curve using the following equations:

$$C_{cell} = \frac{I \times \Delta t}{\Delta V} \quad (5)$$

$$C_s = \frac{C_{cell}}{S} = \frac{I \times \Delta t}{S \times \Delta V} \quad (6)$$

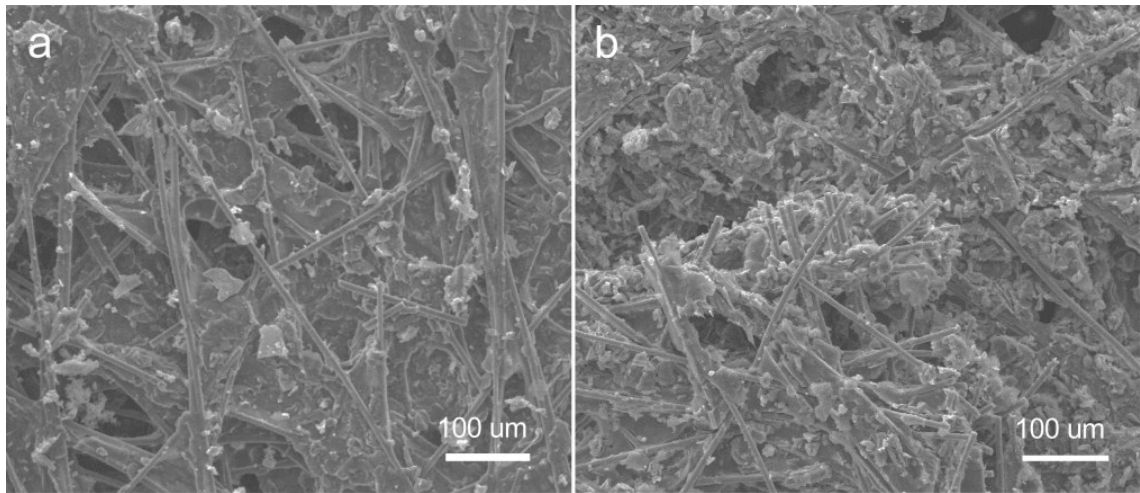
where  $I$  (A) is the applied current,  $S$  (cm<sup>2</sup>) is the area of the whole device,  $\Delta t$  (s) is the discharging time,  $\Delta V$  (V) is the voltage window.

Areal energy density and power density of the devices were obtained from the following equations:

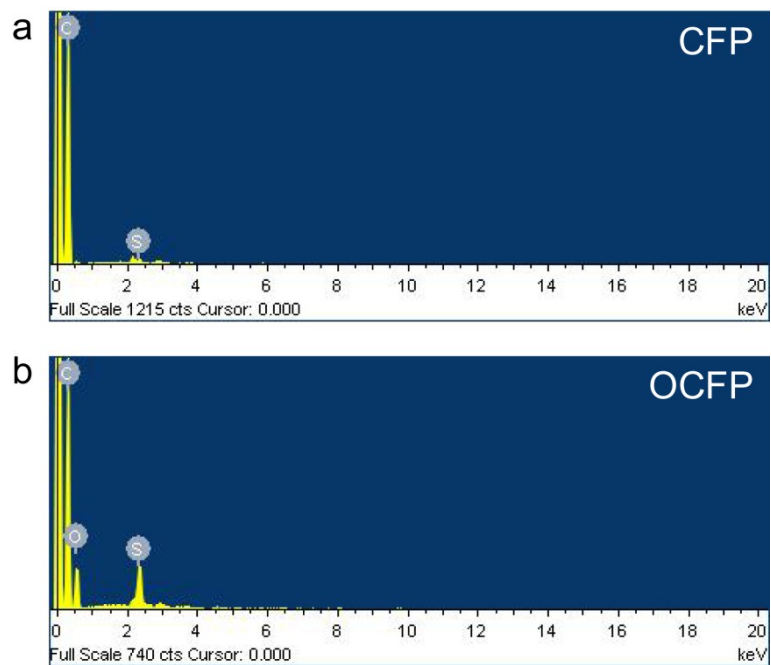
$$E = \frac{1}{2 \times 3600} C_s \Delta V^2 \quad (7)$$

$$P = \frac{3600E}{\Delta t} \quad (8)$$

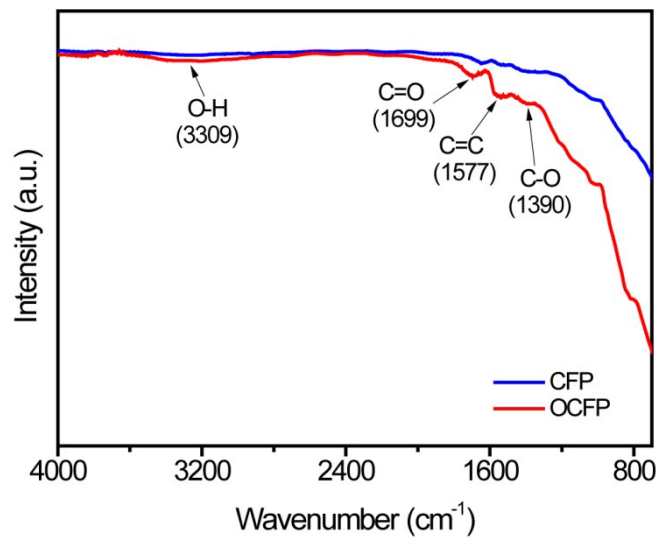
where  $E$  (Wh cm<sup>-2</sup>) is the energy density,  $C_s$  is the volumetric capacitance obtained from Equation (5) and  $\Delta V$  (V) is the voltage window.  $P$  (W cm<sup>-2</sup>) is the power density,  $\Delta t$  (s) is the discharging time.



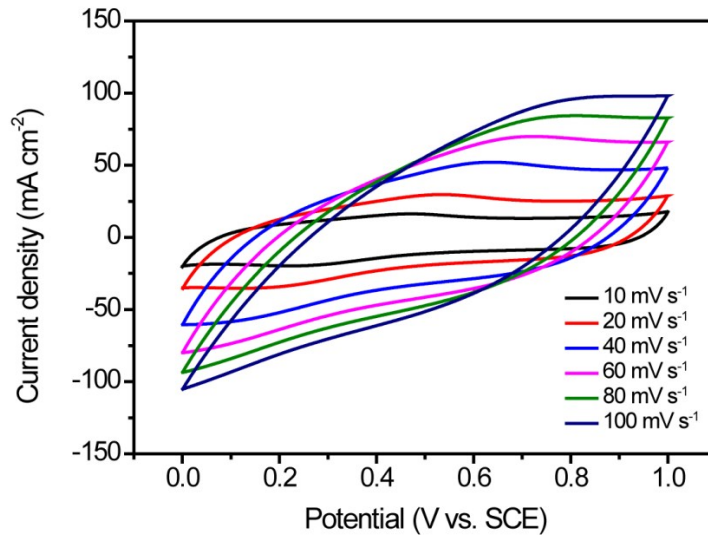
**Figure S1.** SEM images of the (a) CFP and (b) OCFP samples.



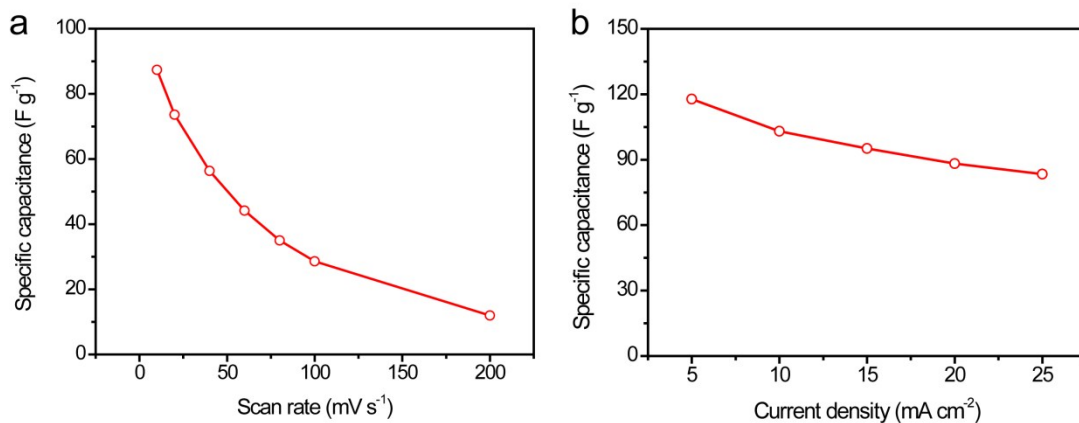
**Figure S2.** Elemental analysis of the CFP and OCFP samples.



**Figure S3.** Fourier transform infrared (FTIR) spectra of the CFP and OCFP samples.



**Figure S4.** CV curves of the OCFP electrode collected at various scan rates.



**Figure S5.** Specific capacitance of the OCFP electrode as a function of (a) scan rate and (b) current density.

