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Supporting Information

Surface Engineering of Carbon Fiber Paper for Efficient Capacitive Energy Storage

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Calculations:

1. Single Electrode :

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

$$C_{a}=\frac{Q}{\Delta V \bullet S} \qquad (1)$$

where C_a (F cm^{-2}) is the areal capacitance, Q (C) is the average charge during the charging and discharging process, ΔV (V) is the potential window and S (cm^2) is the area of OCFP electrode.

Alternatively, using the galvanostatic charge/discharge method, we can determine the areal capacitances of electrodes by the following equation:

$$C_a=\frac{I \times \Delta t}{\Delta V \bullet S} \qquad (2)$$

Where C_a (F cm^{-2}) is the areal capacitance, I (A) is the constant discharging current, Δt (s) is the discharging time, ΔV (V) is the potential window, and S (cm^2) is the surface area of electrode.

2. $NiCo_2O_4$//OCFP-ASC Devices:

The cell (device) capacitance (C_{cell}) and volumetric capacitance of the as-fabricated $NiCo_2O_4$//OCFP-ASC devices were calculated from their CVs according to the following equation:

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

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

where Q(C) is the average charge during the charging and discharging process is the applied current, V represents the volume (cm^3) of the whole device (The area and thickness of our $NiCo_2O_4$//OCFP-ASC device is about 1 cm^2 and 0.08 cm. Hence, the whole volume of our $NiCo_2O_4$//OCFP-ASC device are about 0.08 cm^3), while ΔV(V) indicates the operation voltage window. It is worth mentioning that the volumetric capacitances were calculated taking into account the volume of the device stack, which includes the active material, the flexible substrate and the separator with electrolyte.

Alternatively, the cell (device) capacitance (C_{cell}) and volumetric capacitance of the electrode (C_v) was estimated from the slope of the discharge curve using the following equations:

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

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

where I is the applied current, V indicates the volume (cm^3) of the whole device (the whole volume of our $NiCo_2O_4$//OCFP-ASC device is about 0.08 cm^3), Δt represents the discharging time, ΔV (V) stand for the operation voltage window.

Volumetric energy density (E), equivalent series resistance (ESR) and power density (P) of the as-fabricated devices were obtained from the following equations:

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

$$ESR = \frac{iR_{drop}}{2 \times I} \quad (8)$$

$$P = \frac{\Delta V^2}{4 \times ESR \times V} \quad (9)$$

where E (Wh/cm^3) is the energy density, C_V represents the volumetric capacitance obtained from Equation (5) and ΔV (V) indicates the voltage operation window. ESR (Ω) is the internal resistance of the device. P (W/cm^3) is the power density. iR_{drop} is the voltage drop between first and second points from its cut-off of discharge curve. V is the volume of the device.

3. Balance the charge of electrodes in ASC device:

As for a SC, the charge balance will follow the relationship $q^+= q^-$. The charge stored by each electrode depends on the capacitance (C_s), the potential range for the charge/discharge process (ΔE) and the area of the electrode (A) following the Equation 10:

$$q = C_S \times \Delta E \times A \quad (10)$$

To obtain $q^+= q^-$ at 100 mV s^{-1}, the area balancing between $NiCo_2O_4$ and OCFP will be calculated as follow:

$$\frac{A_{NiCo_2O_4}}{A_{OCFP}} = \frac{C_{(OCFP)} \times \Delta E_{OCFP}}{C_{NiCo_2O_4} \times \Delta E_{NiCo_2O_4}} \approx \frac{1}{1}$$

Therefore, the calculated areal ration between the $NiCo_2O_4$ electrode and OCFP electrode was about 1:1.

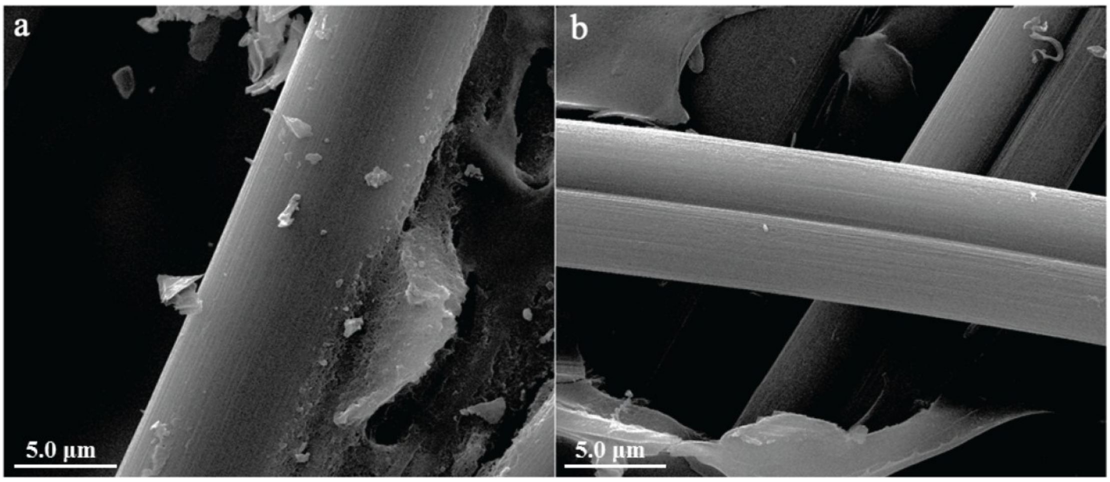

Figure S1. SEM images of the a) CFP and b) OCFP.

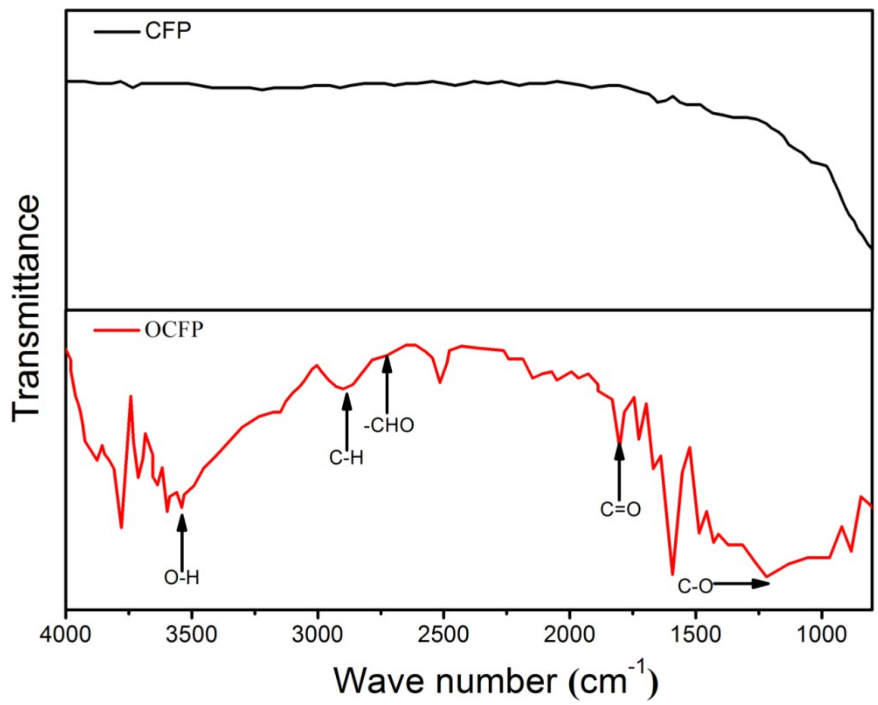

Figure S2. Fourier transform infrared (FTIR) spectra of the CFP and OCFP samples.

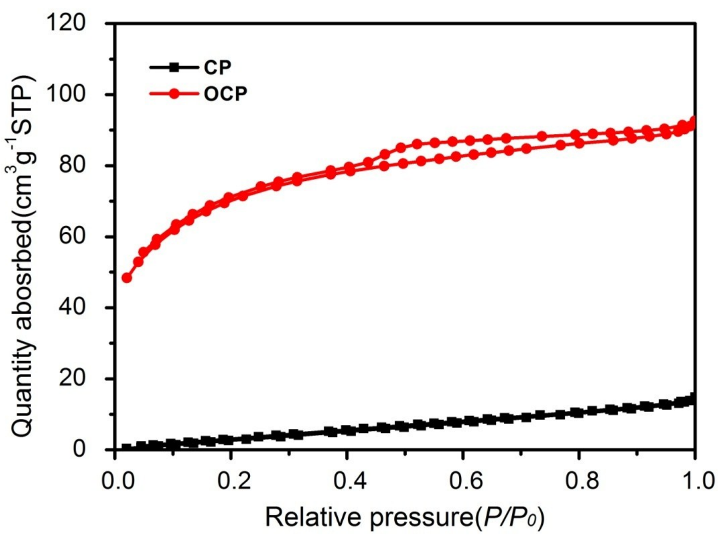

Figure S3. Nitrogen adsorption isotherms of CFP and OCFP at 77 K.

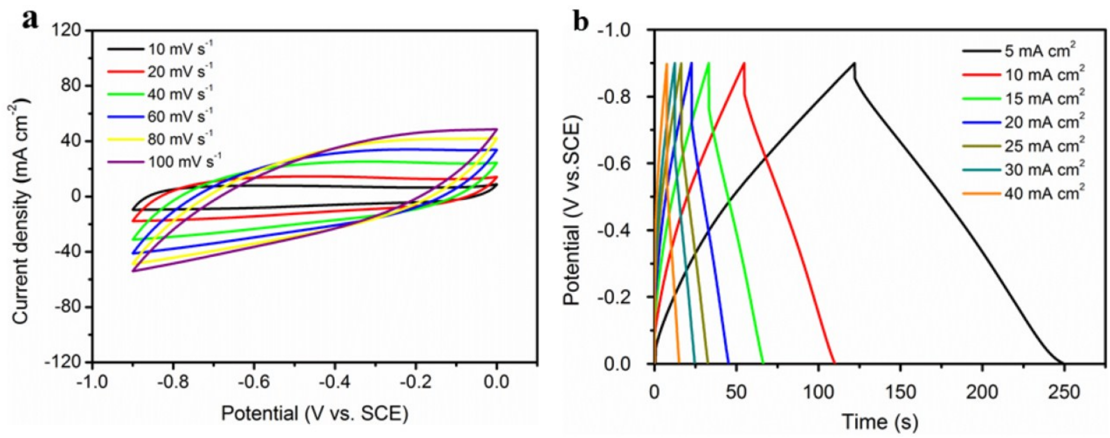

Figure S4. a) CV curves of the OCFP electrode collected at various scan rates, b) Galvanostatic charge–discharge curves of the OCFP electrode at various current densities.

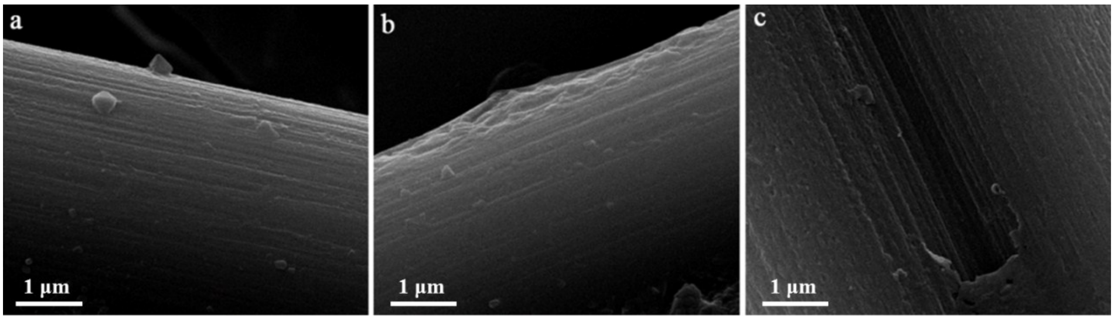

Figure S5. SEM images of 300 °C (a), 400 °C (b), and 500 °C (b) OCFP samples. As processing temperature increased, the surface of carbon fiber became rougher.

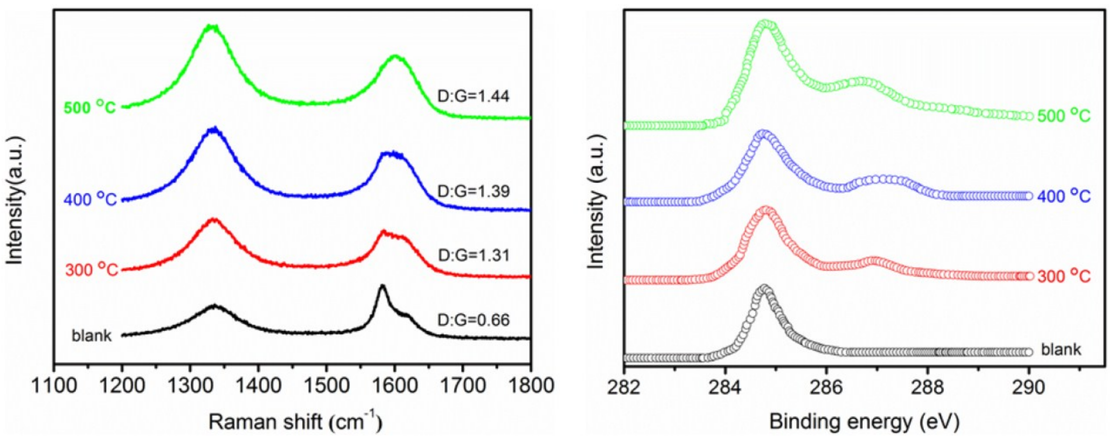

Figure S6. Raman spectra (a), and XPS spectra (b) for varies OCFP. As processing temperature increased, the broader shoulder enhanced, indicating more oxygen functional groups on the surface.

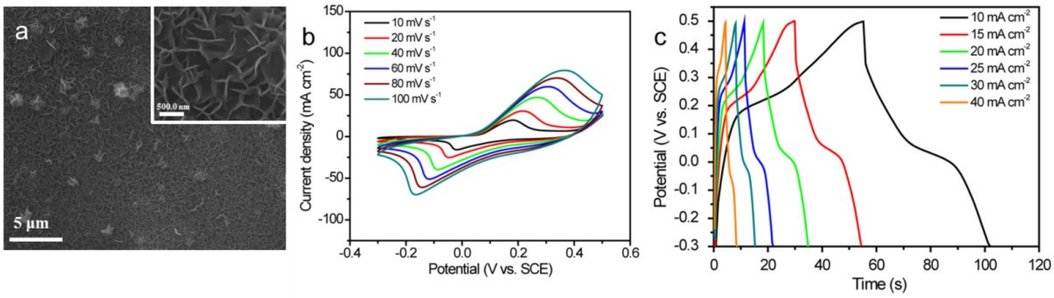

Figure S7. a) SEM images of 3D $NiCo_2O_4$ electrode, b) CV curves for the 3D $NiCo_2O_4$ electrode collected at different scan rate, c) Galvanostatic charge/discharge curve collected at different current density.

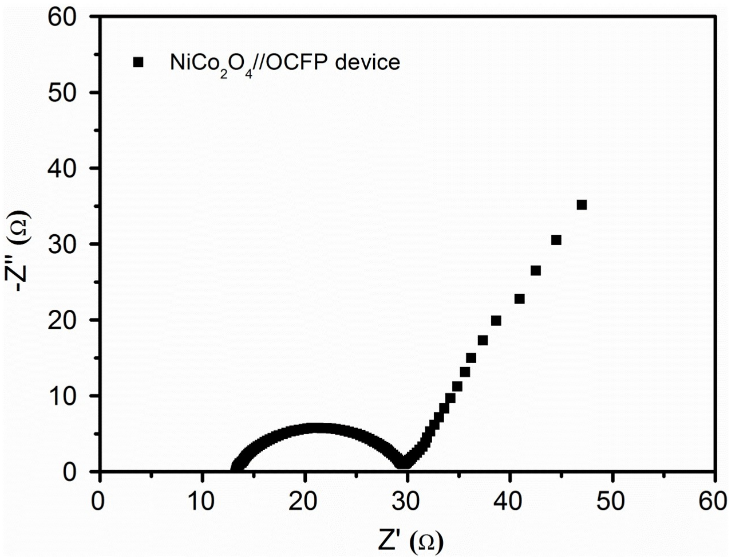

Figure S8. Nyquist plots of the as-fabricated the $NiCo_2O_4$//OCFP ASC device.