

## Supplementary Information

### Cellulose Nanofibers Derived Carbon Aerogel with 3D Multiscale Pore Architecture for High-Performance Supercapacitors

Lumin Chen<sup>a</sup>, Houyong Yu <sup>a\*</sup>, Ziheng Li<sup>a</sup>, Xiang Chen<sup>a</sup>, Wenlong Zhou<sup>a</sup>

<sup>a</sup>*National Engineering Lab for Textile Fiber Materials & Processing Technology,  
College of Textile Science and Engineering, Zhejiang Sci-Tech University, Hangzhou  
310018, China*

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*\*Corresponding authors. Tel: +86-571-86843618; Fax: +86-571-86843619.  
E-mail addresses: phdyu@zstu.edu.cn*

## Electrochemical measurements

The electrochemical performance of electrodes was performed using a three-electrode system in an electrochemical workstation (CHI660E, CH Instruments Ins., USA). A saturated calomel electrode (SCE), platinum wire, and 1 M H<sub>2</sub>SO<sub>4</sub> were used as the reference electrode, the counter electrode, and the electrolyte, respectively. The Cyclic voltammetry (CV) tests were carried out in a scan rate range of 10-600 mV s<sup>-1</sup> with a potential window of 1.6 V. Galvanostatic charge-discharge test was performed between -0.4 and 1.2 V at current densities of 1-30 A g<sup>-1</sup>. The electrochemical spectroscopy (EIS) measurements were conducted in a frequency range of 0.01-100 kHz with an AC amplitude of 5 mV at open circuit potential. The electrochemical tests of symmetric supercapacitors were performed in a two-electrode system.

For three-electrode system, the specific capacitances ( $C_s$ ) can be calculated from CV via the following equation (1):

$$C_s = \frac{Q}{mS\Delta V} \quad (1)$$

Where Q is the integrated area of CV curve, m is active material mass, S is the scan rate of CV curve,  $\Delta V$  is the potential window.

or from GCD curve by the equations (2):

$$C_s = \frac{It}{m\Delta V} \quad (2)$$

Where I is the discharge current, t is the discharge time, m is active material mass,  $\Delta V$  is the voltage after IR drop, respectively.

For two-electrode system, the areal specific capacitances ( $C_A$  in mF cm<sup>-2</sup>) and

gravimetric specific capacitances ( $C_g$  in  $F g^{-1}$ ) can also be calculated by the following equations:

$$C_A = \frac{It}{S\Delta V} \quad (3)$$

$$C_g = \frac{It}{m\Delta V} \quad (4)$$

Where  $I$  is the discharge current,  $t$  is the discharge time,  $S$  is the area of the hydrogel electrode,  $m$  is active material mass of two electrodes,  $\Delta V$  is the voltage after IR drop.

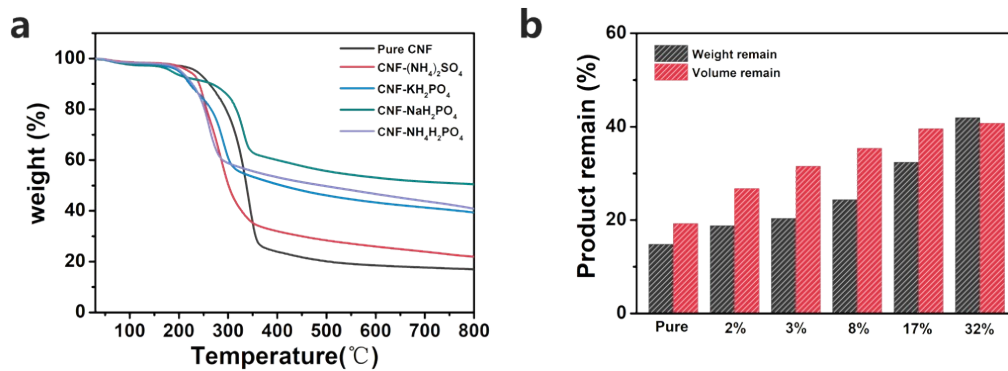
The areal energy density  $E_A$  ( $mWh cm^{-2}$ ) and power density  $P_A$  ( $mW cm^{-2}$ ) were obtained from the following equations:

$$E_A = \frac{1}{2} \times C_A \times \frac{\Delta V^2}{3.6} \quad (5)$$

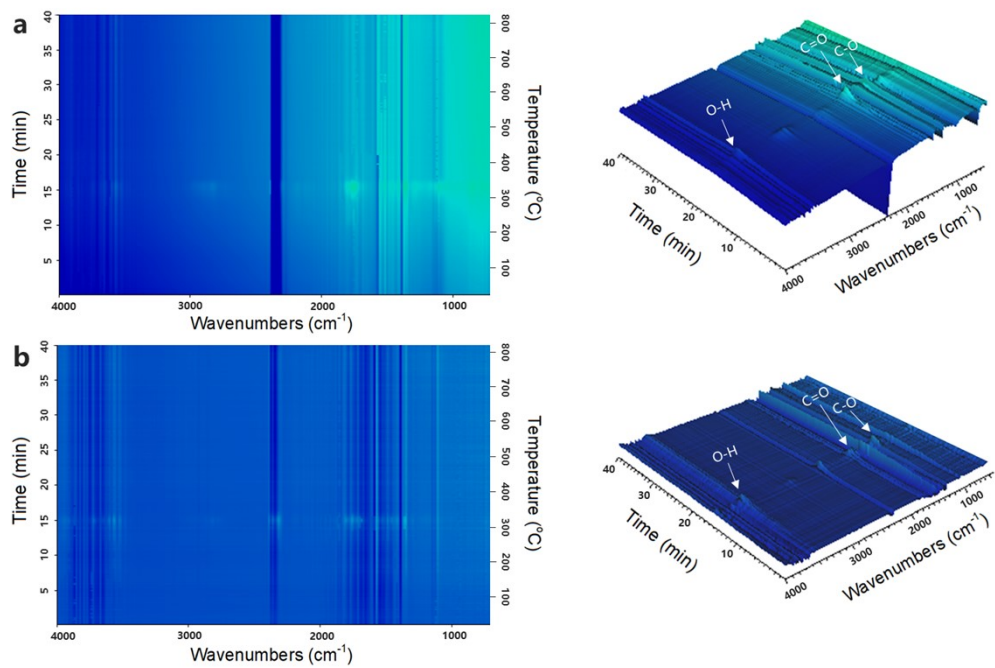
$$P_A = \frac{3600 \times E_A}{t}$$

(6)

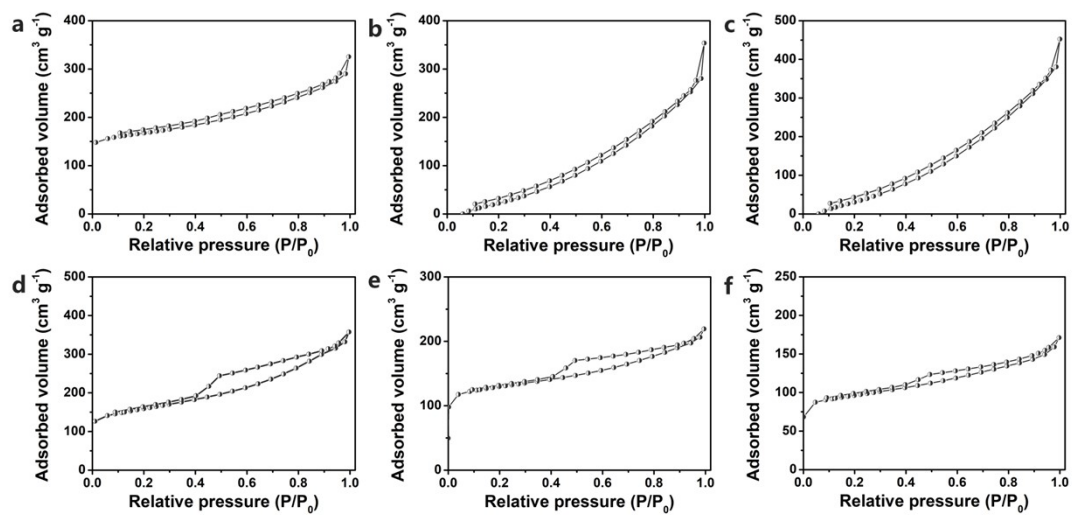
Where  $C_A$ ,  $\Delta V$  and  $t$  are the areal specific capacitances of supercapacitor, the voltage after IR drop and discharge time, respectively.



**Figure S1.** (a) TG curves of pure CNF and CNF soaked with (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, KH<sub>2</sub>PO<sub>4</sub>, NaH<sub>2</sub>PO<sub>4</sub>, and NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub>. (b) Weight and volume remain of CNFAs prepared by carbonization from pure CNF and CNF soaked with different amounts of NaH<sub>2</sub>PO<sub>4</sub>.



**Figure S2.** 3D FTIR spectra of (a) pure CNF and (b) CNF-32 wt% NaH<sub>2</sub>PO<sub>4</sub> composite.



**Figure S3.** Nitrogen adsorption-desorption isotherm of (a) pure CNFAs, (b) CNFAs-2%, (c) CNFAs-3%, (d) CNFAs-8%, (e) CNFAs-17%, and (f) CNFAs-32%.

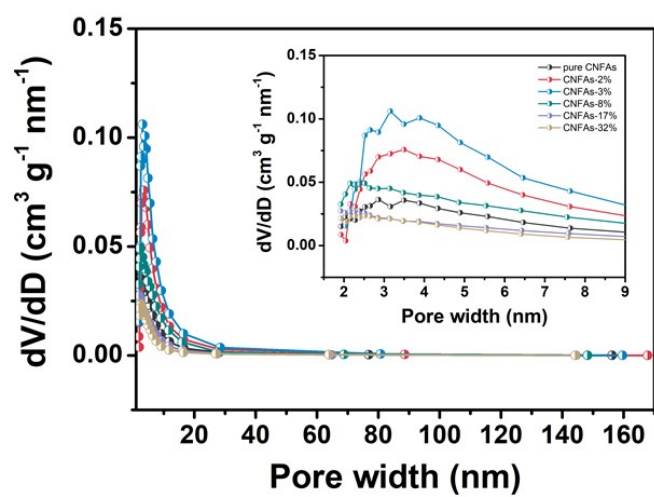
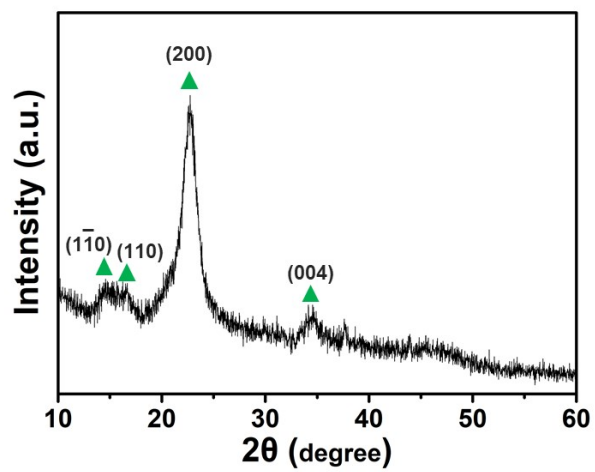
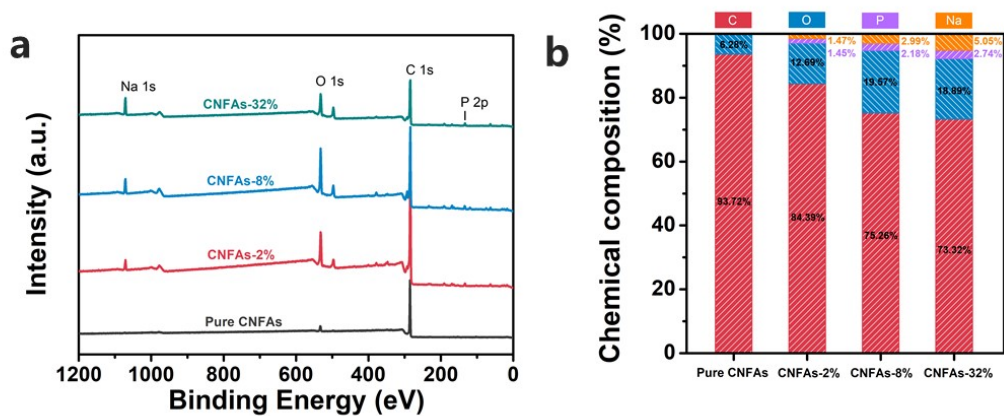


Figure S4. The pore-size distribution of CNFAs.

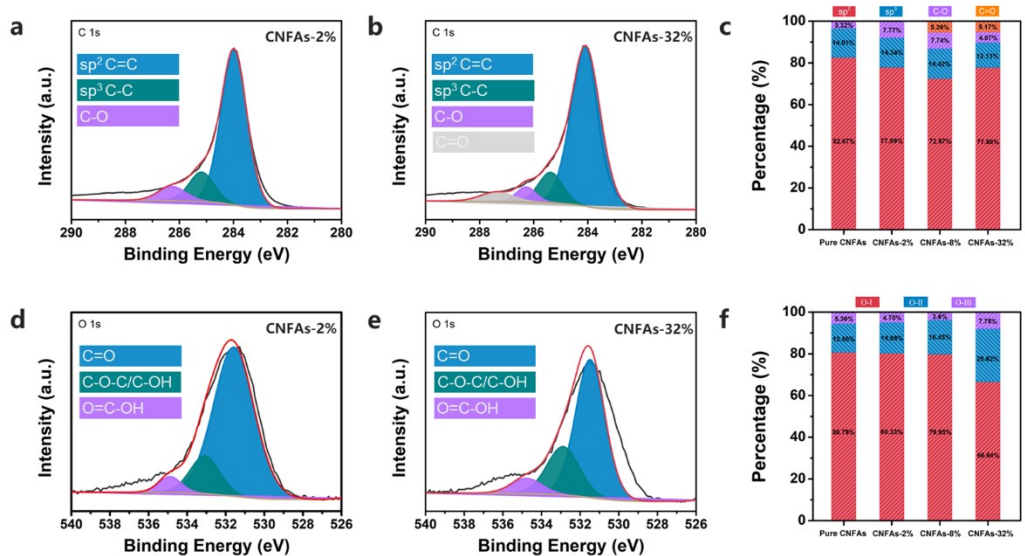


**Figure S5.** The XRD pattern of CNF.

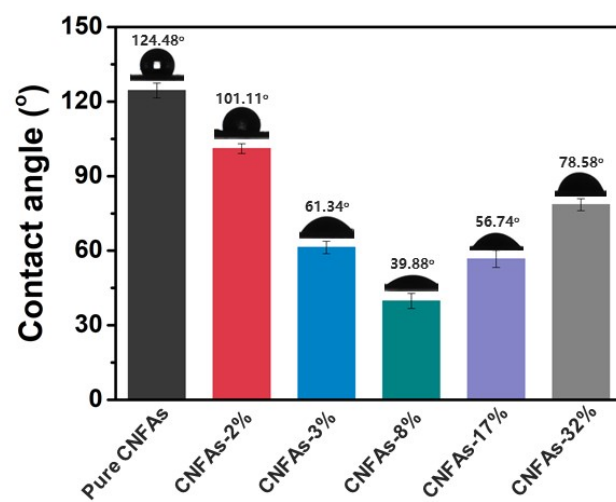




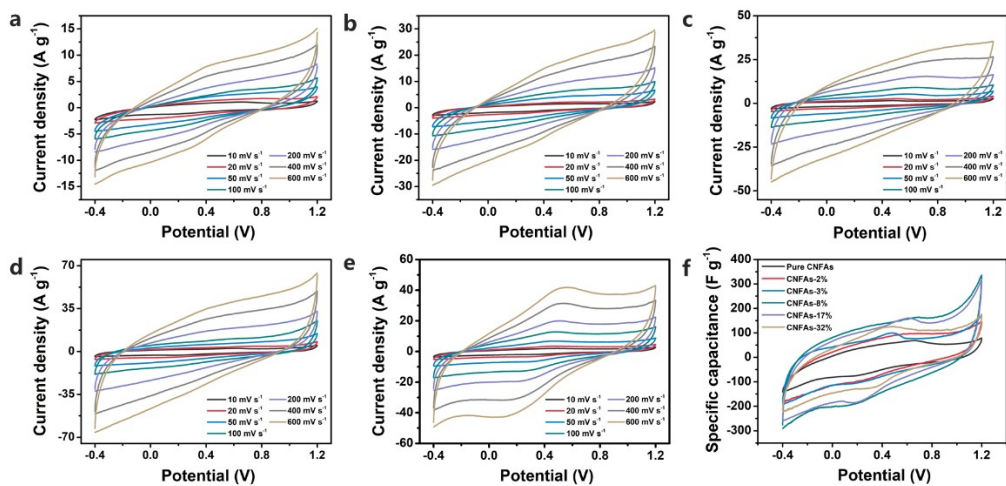
**Figure S6.** (a) XPS survey spectra of pure CNFAs, CNFAs-2%, CNFAs-8%, and CNFAs-32%. (b) The chemical composition content of pure CNFAs, CNFAs-2%, CNFAs-8%, and CNFAs-32% derived from XPS results.



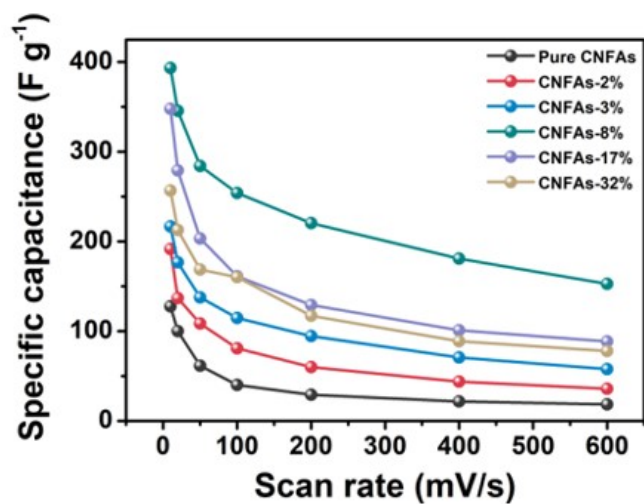
**Figure S7.** C 1s XPS spectra of (a) CNFAs-2% and (b) CNFAs-32%. (c) The functional group content of pure CNFAs, CNFAs-2%, CNFAs-8%, and CNFAs-32% derived from C 1s XPS spectra results. O 1s XPS spectra of (d) CNFAs-2% and (e) CNFAs-32%. (f) The functional group content of pure CNFAs, CNFAs-2%, CNFAs-8%, and CNFAs-32% derived from O 1s XPS spectra results.



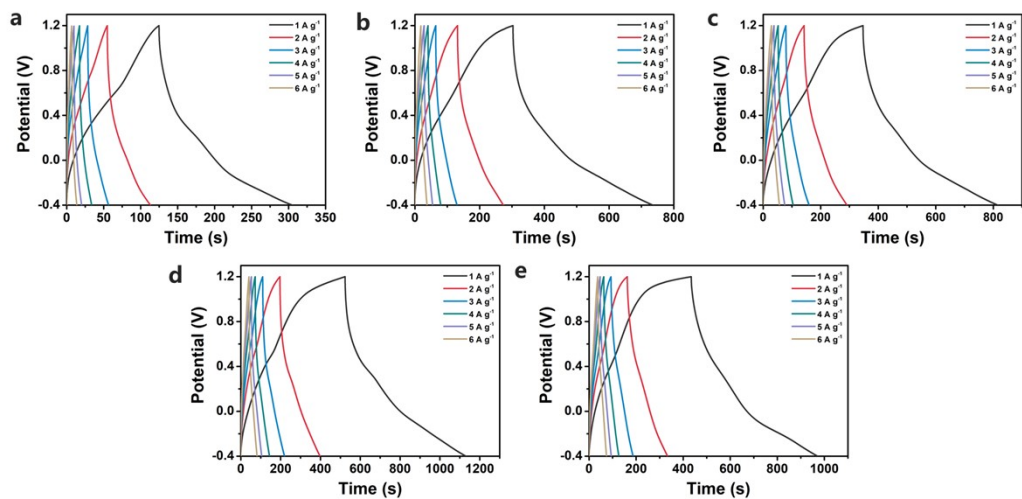
**Figure S8.** Water contact angle values of CNFAs.



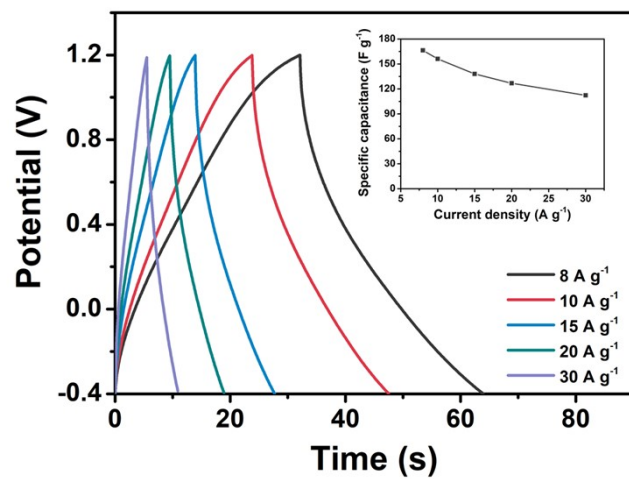
**Figure S9.** CV curves of (a) pure CNFAs, (b) CNFAs-2%, (c) CNFAs-3%, (d) CNFAs-8%, (e) CNFAs-17%, and (f) CNFAs-32% at different scan rates. (f) CV curves of the six samples at a scan rate of  $10 \text{ mV s}^{-1}$ .



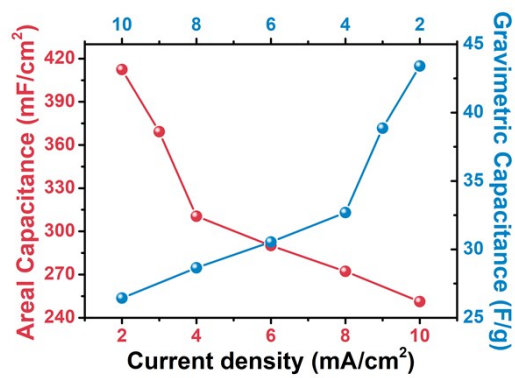
**Figure S10.** Specific capacitance of pure CNFAs, CNFAs-2%, CNFAs-3%, CNFAs-8%, CNFAs-17%, and CNFAs-32% at different scan rates.



**Figure S11.** GCD curves of (a) pure CNFAs, (b) CNFAs-2%, (c) CNFAs-3%, (d) CNFAs-17%, and (e) CNFAs-32% at different current densities.

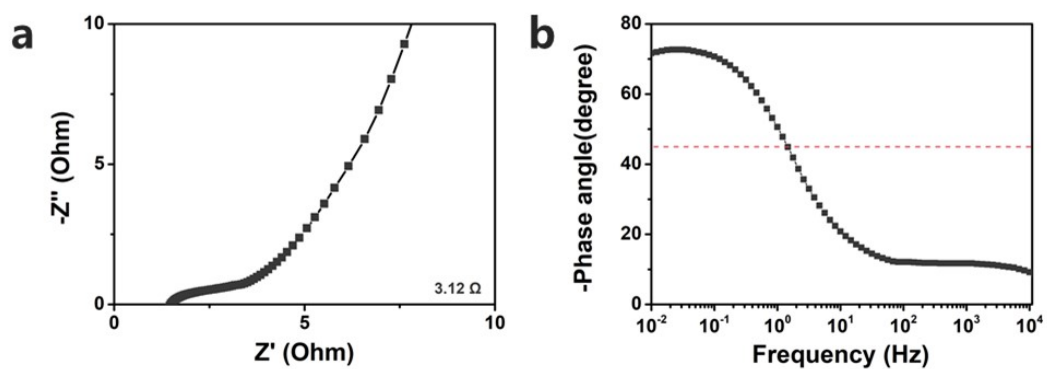


**Figure S12.** GCD curves of CNFAs-8% electrode at different current densities, the inset shows the corresponding specific capacitance.



**Figure S13.** Variation of areal capacitances and gravimetric capacitances at different current densities.





**Figure S14.** (a) Nyquist plot and (b) bode plot of CNFAs-8% // CNFAs-8% symmetric supercapacitor.



**Figure S15.** Photographs of electronic watch powered using CNFAs-8%/CNFAs-8% symmetric supercapacitor.