## High-performance flexible and weavable asymmetric fiber-shaped solid-state supercapacitor enhanced by surface modifications of carbon fibers with carbon nanotubes

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## **Supplementary Information**



Figure S1 SEM image of a typical bundle of CF@CNC with average diameter of about 0.92 mm.



Figure S2 Water contact angles for (a) pristine CF and (b) air plasma treated CF.

 Table S1
 Measured diameters of carbon fiber and carbon fiber based hybrid fibers

	CF	CF@C	CF@NC	CF@PNC	CF@CNC
Diameter (µm)	6.98	6.92	7.58	7.74	7.04



Figure S3 EDS element (C, Co and Ni)-mapping images of CF@CNC from the rectangular area shown in the above SEM image.

Table S2 Elemental ratio of CF@CNC obtained from EDS element mapping

Element	Туре	wt%
С	K	75.65
Ο	Κ	15.74
Co	Κ	5
Ni	Κ	3.61



Figure S4 (a-d) SEM images revealing surface morphologies of the CF@CNC electrodeposited for 10, 25, 40 and 60 minutes, respectively.



Figure S5 SEM images of (a) the CF@PNC and (b) the CF@CNC fibers after 10 CV cycles.



Figure S6 CV curves obtained from CF@CNC and CF@AC electrodes at 10 mV s<sup>-1</sup> in 2 M KOH solution.

As for asymmetric supercapacitor, mass loading ratio of both electrodes are determined by the charge balance:  $q_+=q_-$ , where  $q_+$  means the charges stored in the positive electrode and  $q_-$  means the charges stored in the negative electrode. The charge is calculated from CV curves according to the following equation:  $q=C \times \Delta E \times m$ , in which C represents the specific capacitance,  $\Delta E$  means the voltage range operated on positive (-0.4–0.6 V) and negative (-1.0–0.0 V) electrodes and m refers to the mass of the active material in both electrodes. The mass ratio is thus calculated by:

$$\frac{m_{-}}{m_{+}} = \frac{C_{+} \times \Delta E_{+}}{C_{-} \times \Delta E_{-}}$$

As a result, the mass ratio of the two electrodes is  $m_{-}/m_{+}=16.0$ .



Figure S7 Calculated capacitances of the a-FSSCs operated at increased voltage windows.



Figure S8 Voltage drop with respect to charge-discharge current densities and the corresponding fitting line.



Figure S9 Cyclic galvanostatic charge-discharge test of the a-FSSC under a current density of 4.0 mA cm<sup>-2</sup> at 1.6 V operating potential.



Figure S10 The Ragone plot camparing the volumetric energy density and power density of the a-FSSC with previous works <sup>1-10</sup>.



Figure S11 Stress-strain curves of the pristine CF bundle.



Figure S12 Linear sweep voltammetry of a blue LED with working potential above 2.5 V.



Figure S13 A digital photograph of the a-FSSCs fabricated with different length and diameters.

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