

# N-Doped Porous Carbon-Nanofiber-Supported Fe<sub>3</sub>C/Fe<sub>2</sub>O<sub>3</sub> Nanoparticles as Anode for High-Performance Supercapacitors

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## 1 Electrochemical measurements

All of the electrochemical measurements were carried out on a CHI660E electrochemical workstation. In a three-electrode configuration, a standard Hg/HgO was used as the reference electrode, a slice of Pt foil was acted as the counter electrode. The work electrode was prepared by coating the homogeneous slurry containing active material, carbon black and PVDF (5 wt.% NMP solution) in a ratio of 8:1:1 onto a piece of graphite paper and dried at 80 °C for 24 h. The single electrode measurements were conducted in 2 mol L<sup>-1</sup> KOH aqueous electrolyte. The electrochemical impedance spectroscopy (EIS) was performed in the frequency range from 0.01 to 100 kHz with an amplitude of 10 mV at open circuit potential. Cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) were conducted at room temperature. The specific capacitances derived from GCD ( $C_g$ , F g<sup>-1</sup>) results were calculated by means of equation (S1):

$$C_g = \frac{i\Delta t}{m\Delta V} \quad (\text{S1}),$$

where  $i$ ,  $\Delta t$ ,  $m$  and  $\Delta V$  are the current density (A), discharge time (s), mass of active material (g), and

voltage window (V), respectively.

The asymmetric supercapacitor (SC) was assembled using N-doped carbon nanofibers supporting Fe<sub>3</sub>C/Fe<sub>2</sub>O<sub>3</sub> nanoparticles hybrid architecture composite (NCFCO) anode and activated carbon (AC) cathode and the electrochemical performances were measured in a two-electrode system in 2 mol L<sup>-1</sup> KOH aqueous electrolyte. The masses of active material in the anode ( $m_-$ , g) and cathode ( $m_+$ , g) were determined according to the following equation:

$$\frac{m_+}{m_-} = \frac{C_- \Delta V_-}{C_+ \Delta V_+} \quad (S2),$$

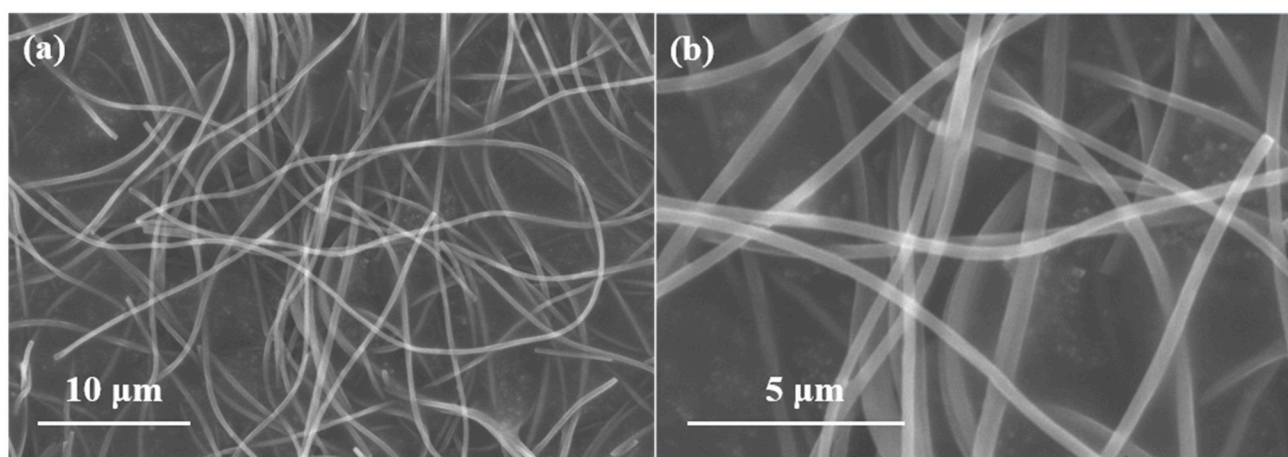
where  $C_+$  (F g<sup>-1</sup>) and  $C_-$  (F g<sup>-1</sup>) are the specific capacitances,  $\Delta V_+$  (V) and  $\Delta V_-$  (V) are the voltage windows of anode and cathode, respectively. The energy density ( $E$ , Wh kg<sup>-1</sup>) and power density ( $P$ , W kg<sup>-1</sup>) of the as-assembled device were computed in terms of equation (S3) and (S4) individually:

$$E = \frac{C \Delta V^2}{7.2} \quad (S3),$$

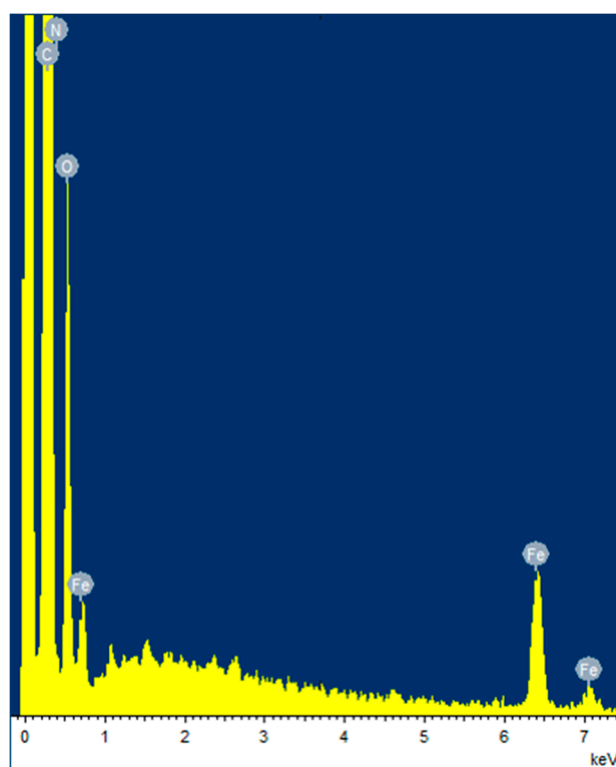
$$P = \frac{E}{\Delta t} \quad (S4).$$

## 2 Characterizations

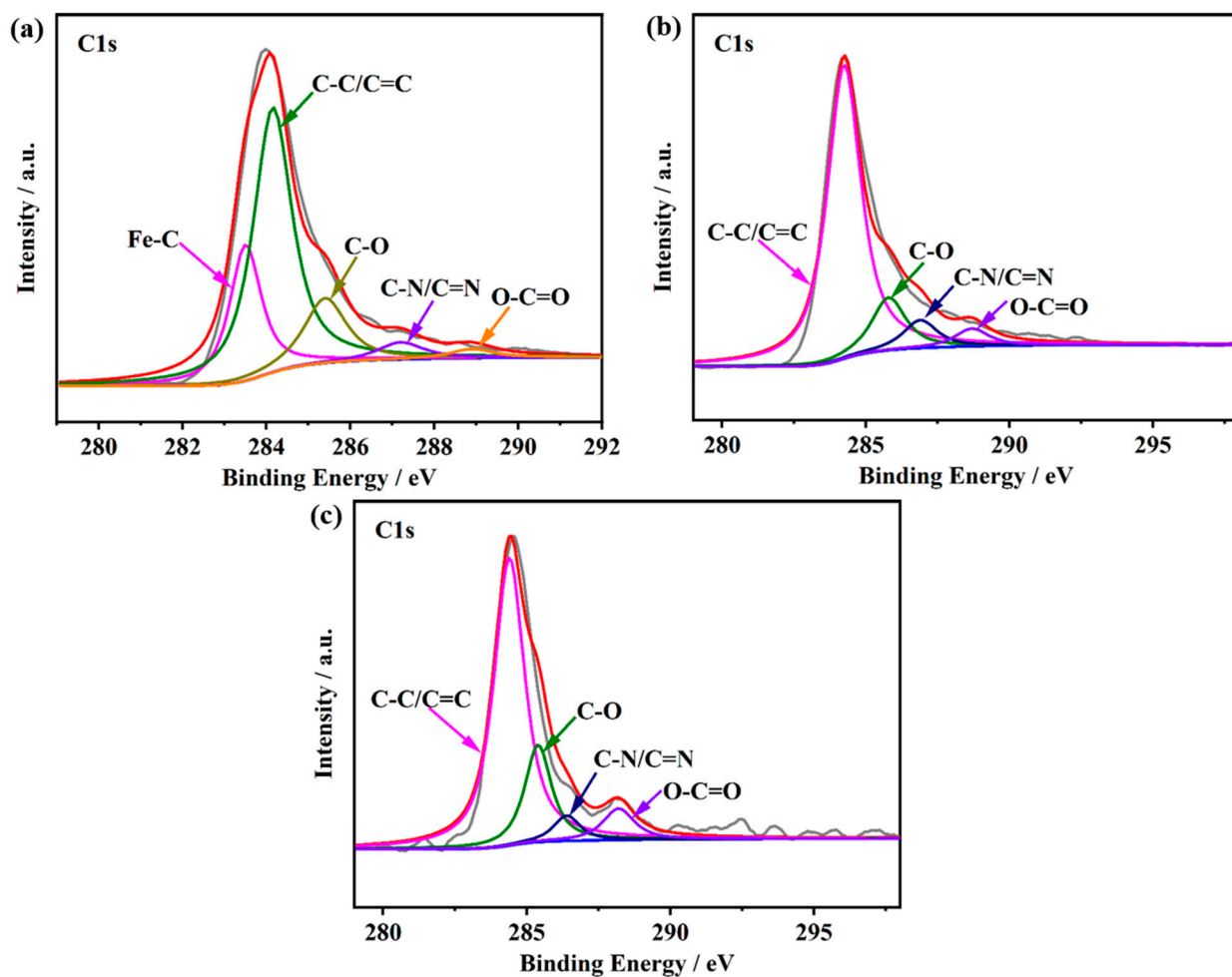
The morphology was characterized on a scan electron microscope (SEM, Hitachi S1510) equipped with energy dispersive X-ray spectroscopy (EDS, Oxford). The phase structure was examined by X-ray diffraction (XRD, Rigaku, Smartlab) and Raman spectroscopy (Horiba, Xplora Plus). The elemental composition and chemical valence on the surface of the samples were tested by X-ray photoelectron spectroscopy (XPS, Escalab 250Xi). The specific surface area (SSA) and pore size distribution were calculated on a physical analyzer (Micromeritics, ASAP 2020 Plus) applying Brunauer-Emmett-Teller (BET) method and Barrett-Joyner-Halenda (BJH) model based on N<sub>2</sub> adsorption-desorption isotherms at 77 K, respectively.



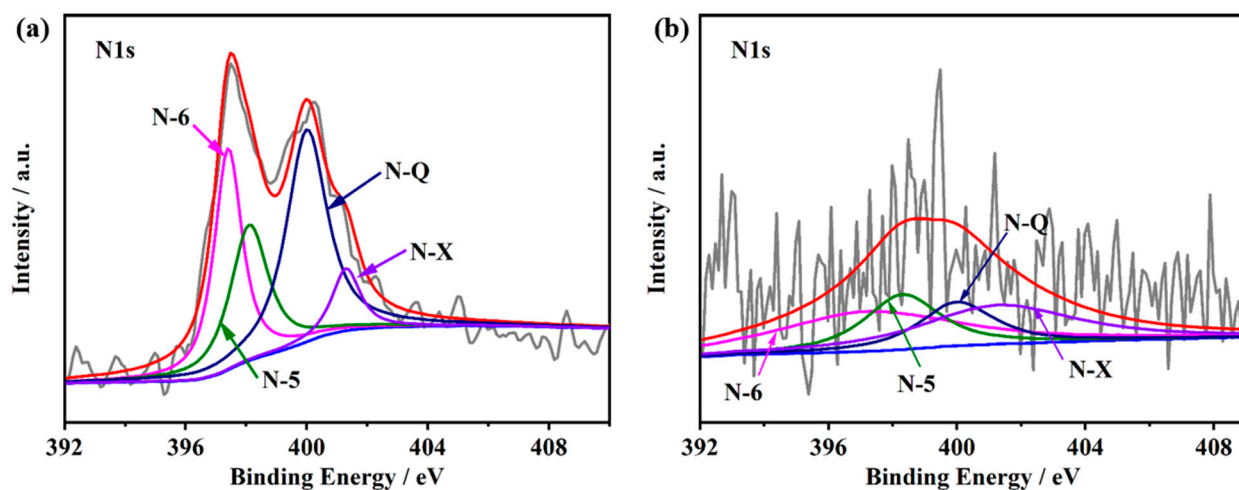
**Figure S1** Low (a) and High (b) magnification SEM images of the electrospinning nanofiber precursor before annealing.



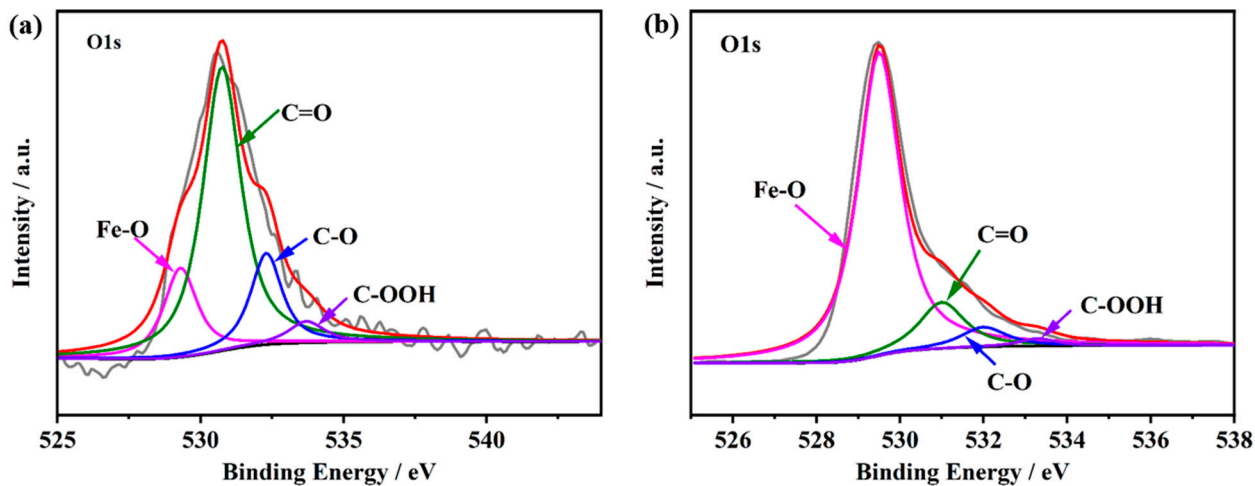
**Figure S2** EDS spectrum of NCFCO.



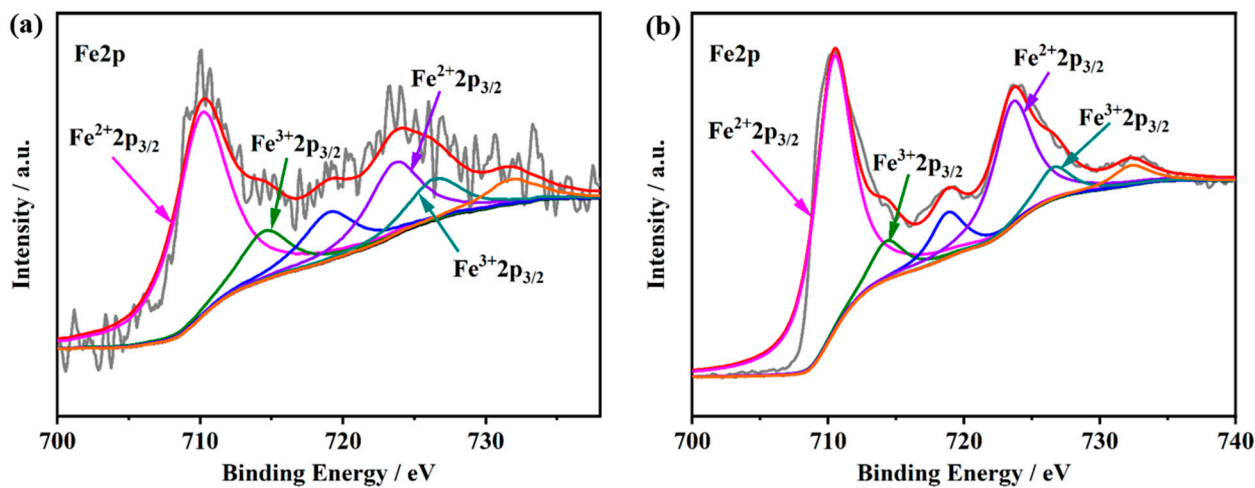
**Figure S3** High resolution  $C1s$  spectra of NCFC (a), NCFCO (b) and NCFO (c).



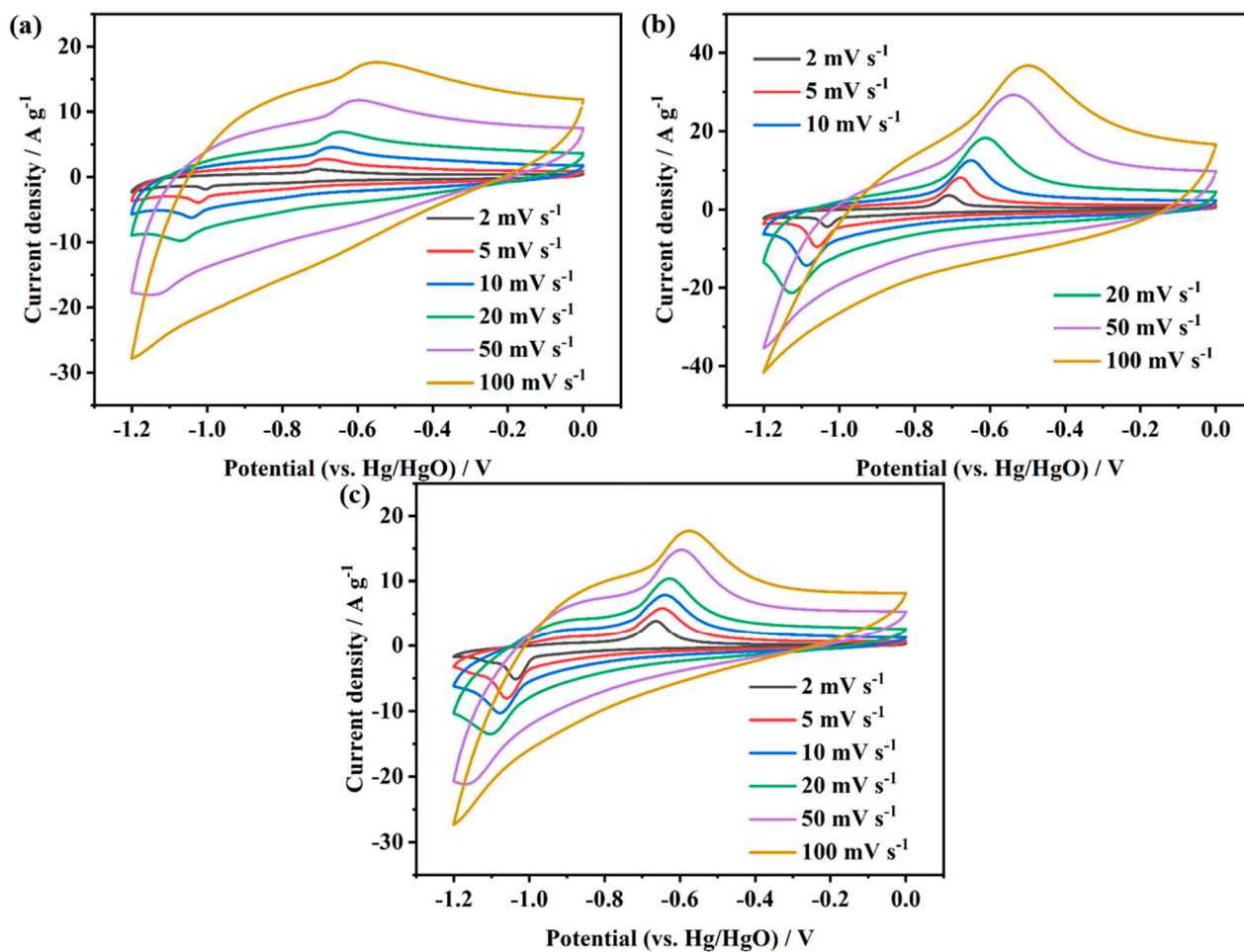
**Figure S4** High resolution  $N1s$  spectra of NCFC (a) and NCFO (b).



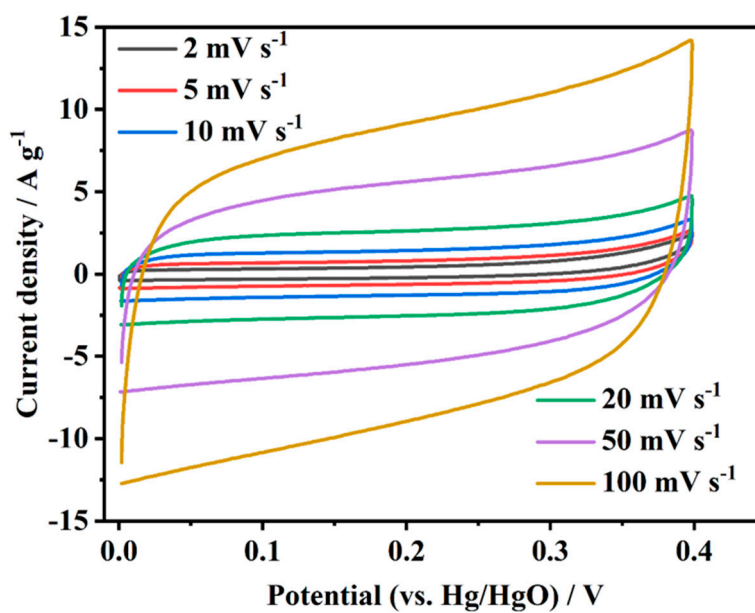
**Figure S5** High resolution O1s spectra of NCFC (a) and NCFO (b).



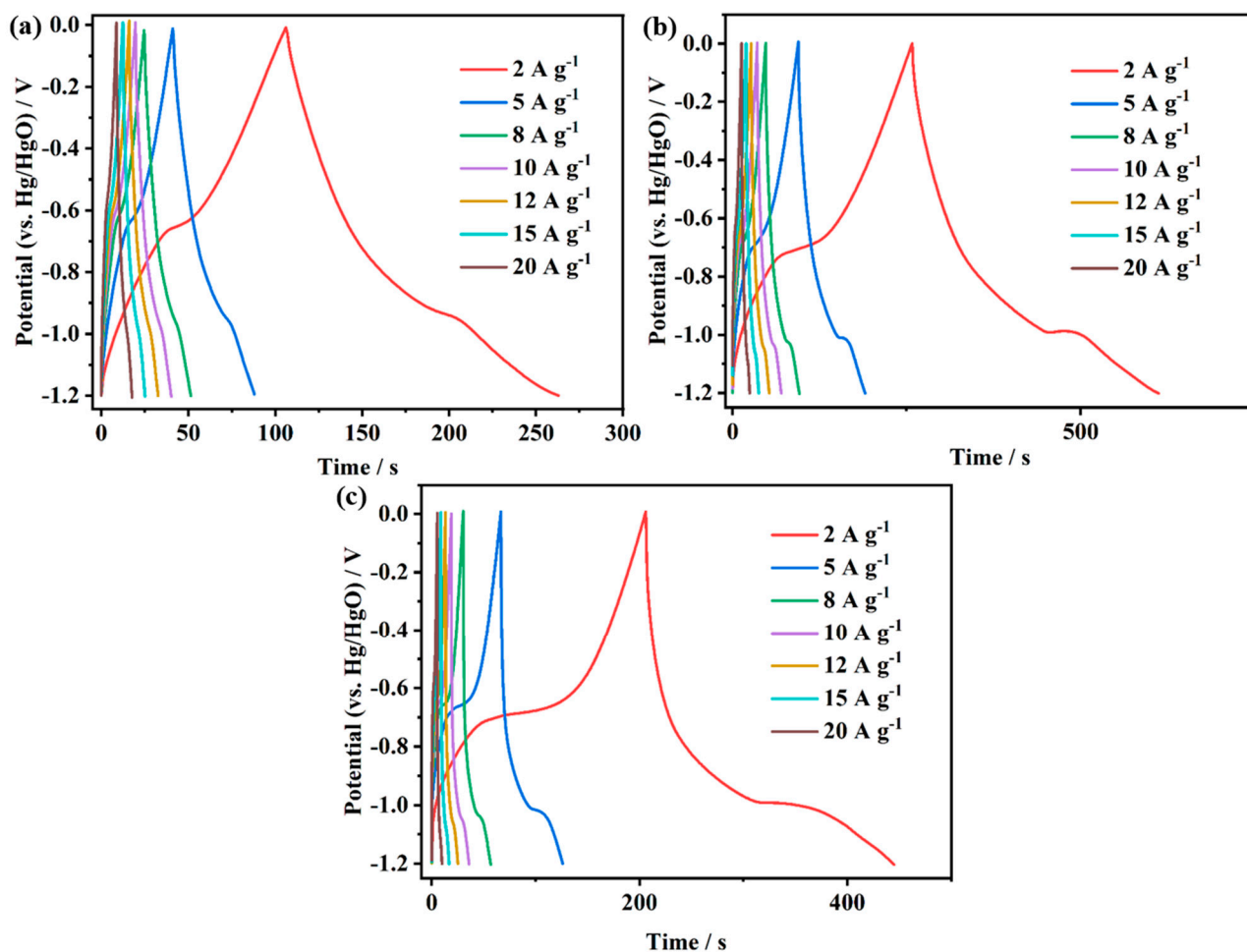
**Figure S6** High resolution Fe2p spectra of NCFC (a) and NCFO (b).



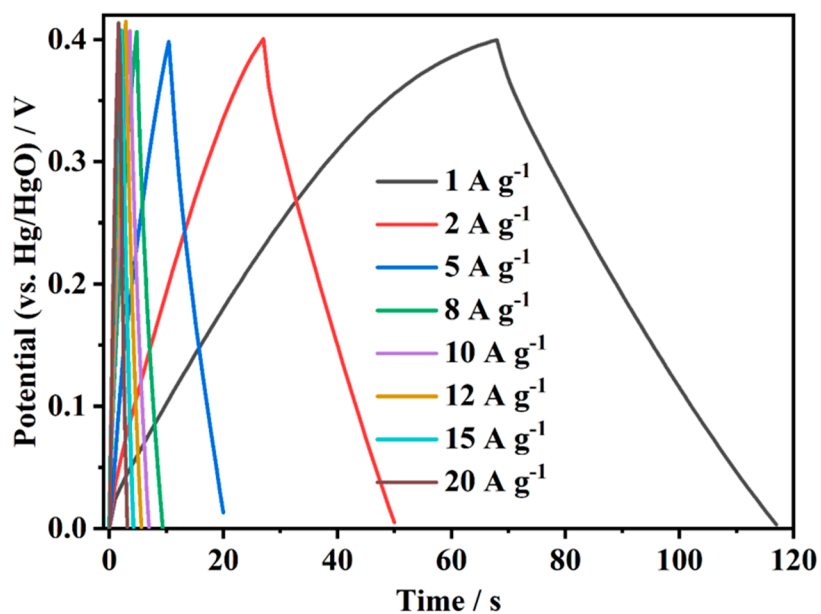
**Figure S7** CV curves of NCFC (a), NCFCO (b) and NCFO (c) at different scan rates.



**Figure S8** CV curves of AC at different scan rates.



**Figure S9** GCD curves of NCFC (a), NCFCO (b) and NCFO (c) at different current densities.



**Figure S10** GCD plots of AC at different current densities.

**Table S1.** SSA, pore volume and average pore diameter of NCFC, NCFCO and NCFO.

	NCFC	NCFCO	NCFO
SSA / m <sup>2</sup> g <sup>-1</sup>	215.9	210.7	21.6
Pore volume / cm <sup>3</sup> g <sup>-1</sup>	0.136	0.124	0.078
Average pore diameter / nm	1.85	1.97	11.8

**Table S2.** Specific capacitances of NCFCO and previously reported Fe-based oxides/carbides.

Materials	Current density / A g <sup>-1</sup>	Specific capacitance / F g <sup>-1</sup>	Reference
Fe <sub>x</sub> C <sub>y</sub> /Fe/carbon nanofibers	1	340	[1]
Fe <sub>2</sub> O <sub>3</sub> /Fe <sub>3</sub> C/N-doped carbon nanosheet	0.5	240	[2]
Fe <sub>2</sub> O <sub>3</sub> /MXene	1	486.3	[3]
Carbon coated Fe <sub>3</sub> O <sub>4</sub> /carbon cloth	1	463	[4]
Fe <sub>3</sub> O <sub>4</sub> /N-doped carbon nanosheets	0.5	522.7	[5]
Fe <sub>3</sub> O <sub>4</sub> /graphene/carbon cloth	1	406	[6]
Fe <sub>2</sub> O <sub>3</sub> /carbon	1	221.5	[7]
Fe <sub>2</sub> O <sub>3</sub> /MXene	1	182	[8]
Carbon/Fe/Fe <sub>2</sub> O <sub>3</sub>	0.1	177	[9]
Fe <sub>3</sub> C/N-doped carbon	2	325	[10]
NCFCO	2	590.1	This work

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