Supplementary information for

Three dimensional N-doped Graphene/CNTs/AC hybrid material for high-performance supercapacitor

Zi-Ang Liu,^a Yuxi Tao,^a Xue-Zhi Song,^a Ming Bao,^a Zhenquan Tan^{*a}

Text S1 Synthesis of graphene oxide (GO)

The graphene oxide has been synthesized by a modified Hummers method. The following are the details: Graphite powder (2.000 g) was added to the round-bottom flask containing the mixture of concentrated H₂SO₄ (360.0 mL) and H₃PO₄ (40.0 mL). After vigorous stirring about 30 minutes, KMnO₄ (18.000 g) was added slowly under vigorous stirring. Then, the temperature of the mixture was increased to 50 °C, and the oxidative reaction of graphite lasted for 16 h. And then 400.0 mL of deionized water and about 20.0 mL of H₂O₂ were added at an extremely slow rate under ice-water bath to prevent the temperature of the mixture from violent abrupt increase. The mixture was stirred for 30 min to remove all impurities. Then the color of mixture turned into luminous yellow. Finally, the mixture was washed with deionized water and hydrochloric acid ($m_{\text{HCI}}:m_{\text{water}} = 1:20$) for three times, respectively.

Text S2 The computation method of specific capacitance

The specific capacitance was calculated using the following formula:

 $C_{\rm spec} = i\Delta t/m\Delta V$

Here, C_{spec} is the specific capacitance (F/g), *i* is the discharging current (mA), Δt is the discharging time (s), *m* is the mass (mg) of the active material, and ΔV is voltage window.



Fig. S1 RGO after the hydrothermal procedure.



Fig. S2 The energy- dispersive spectroscopy (EDS) mapping of the NGCA.

O-III



Fig. S3 Types of the N-containing and oxygen-containing groups in NGCA.



Fig. S4 CV curve of Ni foam without active material at 10 mV/s.



Fig. S5 The fitted EIS curves of all the samples.



Fig. S6 The equivalent circuit for the fitting of the EIS data achieved by ZView software.

Elem ental analy ses	Weight(m g)	N area	C area	H area	N(%)	C (%)	H (%)	S (%)	C/N ratio	C/H ratio
Test 1	3.0600	8923	62689	4471	7.44	72.87	1.741	0.587	9.7874	41.8674
Test 2	2.9910	8644	61610	4326	7.38	73.26	1.729	0.378	9.9304	42.3570
Test 3	3.2660	9371	66798	4982	7.31	72.81	1.795	0.345	9.9604	40.5547

Table S1 Elemental analyzer (EA) of the NGCA.

Table S2 The specific capacitance (C_m) at a current density of 1 A/g and the resistance of the contrast samples

Samples	NGCA	Graphene/CNTs/AC	NG/CNTs	NG/AC	RGO
$C_{\rm m}~({\rm F/g})$	466	139	78.9	66.1	47.1
$R_{\rm s}(\Omega)$	0.39	0.65	0.67	0.74	0.81
$R_{ m ct}\left(\Omega ight)$	0.26	0.37	0.35	0.69	0.50