# **Supporting Information**

## A general method for boosting the supercapacitor performance of

## graphitic carbon nitrides/graphene hybrids

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# Supplementary Tables and Figures

Sample name	Description			
GO	Graphene oxide			
RGO	Reduced graphene oxide (reduced by hydrothermal reduction reaction)			
CN	g-C <sub>3</sub> N <sub>4</sub>			
OCN3	Oxidized g-C <sub>3</sub> N <sub>4</sub> (oxidized by HNO <sub>3</sub> for 3 hours)			
OCN24	Oxidized $g-C_3N_4$ (oxidized by HNO <sub>3</sub> for 24 hours)			
OCN72	Oxidized $g-C_3N_4$ (oxidized by HNO <sub>3</sub> for 72 hours)			
FOCN	Fully oxidized $g-C_3N_4$ (oxidized by improved Hummer's method)			
GOCN-1	Graphene/g-C <sub>3</sub> N <sub>4</sub> composite (manufactured by hydrothermal reduction reaction with a GO/CN mass ratio of 1/1)			
GOCN-2	Graphene/g-C <sub>3</sub> N <sub>4</sub> composite (manufactured by hydrothermal reduction reaction with a GO/CN mass ratio of 2/1)			
GOCN-3	Graphene/g-C <sub>3</sub> N <sub>4</sub> composite (manufactured by hydrothermal reduction reaction with a GO/CN mass ratio of 3/1)			
GOCN-4	Graphene/g-C <sub>3</sub> N <sub>4</sub> composite (manufactured by hydrothermal reduction reaction with a GO/CN mass ratio of 4/1)			
GOOCN3	Graphene/oxidized $g-C_3N_4$ composite (manufactured by hydrothermal reduction reaction with a GO/OCN3 mass ratio of 3/1)			
GOOCN24	Graphene/oxidized g-C <sub>3</sub> N <sub>4</sub> composite (manufactured by hydrothermal reduction reaction with a GO/OCN24 mass ratio of 3/1)			
GOOCN72	Graphene/oxidized $g-C_3N_4$ composite (manufactured by hydrothermal reduction reaction with a GO/OCN72 mass ratio of 3/1)			
GOFOCN	Graphene/oxidized $g-C_3N_4$ composite (manufactured by hydrothermal reduction reaction with a GO/FOCN mass ratio of 3/1)			
PVDF	Polyvinylidene difluoride			
NMP	N-methyl-2-pyrrolidone			

 Table S1. Abbreviations of the materials and chemicals.

### **Supplementary Experimental Section**

#### Chemicals

98-99% purity natural graphite powder was purchased from Hopkin and Williams Ltd. 35 wt% hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), 37 wt% hydrochloric acid (HCl), 86 wt% potassium hydroxide (KOH), and 99 wt% melamine were purchased from Sigma-Aldrich Co. Ltd. 99.5 wt% potassium permanganate (KMnO<sub>4</sub>) was bought from BDH Chemicals Ltd. 98 wt% sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), 37 wt% nitric acid (HNO<sub>3</sub>), ethanol, and 0.22M filter paper were obtained from Fisher Scientific Ltd. Nickel foam and carbon paper were purchased from Suzhou JSD Ltd and Beijing Jinglongte Carbon Technology Co. Ltd, respectively.

#### Preparation of graphene oxide

3 g graphite powder and 69 mL concentrated sulfuric acid were mixed. Then, with the purpose of mixing the solution uniformly and controlling the system temperature at lower than 15 °C, the suspension was transferred to an ice bath and stirred for about 10 min. 3 g KMnO<sub>4</sub> was then slowly and continuously added into the system within 1 h with magnetic stirring. The suspension was moved to a 35 °C oil bath and kept for 2 h without stirring. After that, 138 mL deionized water was subsequently added into the system slowly. During the water adding process, the oil bath temperature was raised from 35 °C to 98 °C. The suspension was finally kept at 98 °C for another 15 min with stirring. In the meantime, 420 mL deionized water and 7.5 mL H<sub>2</sub>O<sub>2</sub> were mixed and preheated in an oven. Then, the preheated solution was slowly added into the suspension. While the suspension cooled to room temperature, it was purified using 750 mL deionized water and 75 mL HCl. Finally, the product was leached and dried below 60 °C for further use.

#### Preparation of g-C<sub>3</sub>N<sub>4</sub>

Briefly, 15 g melamine was annealed at 550 °C in a furnace for 3 h (temperature ramp of 0.5 °C min<sup>-1</sup>). After cooling to room temperature naturally,  $g-C_3N_4$  was obtained.

#### Fabrication of supercapacitor electrodes

Nickel foam was used as the current collector of supercapacitor electrode when tested in alkaline electrolyte in a three-electrode system. Samples ( $\sim 2 \text{ mg}$ ) were compressed between two nickel foam plates (1 cm×1 cm) without using any binders or additives and then soaked in 2 M KOH aqueous solution for future testing. Carbon paper was the current collector choice when measurements were conducted in an acid electrolyte in a three-electrode configuration. Firstly, samples were mixed with PVDF and grinded to form a homogeneous mixture (90% composite materials, 10% PVDF). Proper amount of NMP was added during the grinding process to make slurry with appropriate viscosity. Then, the homogeneous mixture ( $\sim 2 \text{ mg}$ ) was manually casted onto carbon paper (1 cm×1 cm). Finally, the electrode was dried in a vacuum oven at 110 °C for 6 h to remove NMP and soaked in 1 M H<sub>2</sub>SO<sub>4</sub> aqueous solution.

#### Fabrication of symmetrical supercapacitors

Positive (or negative) electrode of the symmetrical supercapacitor was manufactured by pressing 2 mg of the GOOCN24 sample between two round nickel foam plates (diameter =16 mm). A filter paper (diameter=20 mm) was used as the separator between electrodes. KOH aqueous solution was used as the electrolyte in the two-electrode system.

#### Fabrication of asymmetrical supercapacitor

NiCo<sub>2</sub>(OH)<sub>x</sub>/carbon nanotube (CNT) foam was prepared according to previous paper by replacing Fe<sup>2+</sup> to Co<sup>2+</sup> <sup>[S1]</sup>. The as-synthesized NiCo<sub>2</sub>(OH)<sub>x</sub>/CNT foam was chosen as the positive electrodes. The mass ratio between positive electrode and negative electrode can be balanced according to the charge balance equation. The mass ratio was calculated as *ca.* 4:1 (positive to negative part). 2 M KOH was chosen as the electrolyte and the devices were fabricated into coin cells.

#### **Electrochemical measurements**

GCD, CV, and EIS were conducted to measure the electrochemical performance of the samples on Gamry Interface 1000 stations. The potential range for GCD and CV tests was chosen between -0.8 V and 0 V versus Ag/AgCl in alkaline electrolyte (2 M KOH aqueous solution) and between 0 and 1 V versus Ag/AgCl in acid electrolyte (1 M  $H_2SO_4$  aqueous solution). Moreover, EIS tests have been performed in a frequency range between 0.01 and 10<sup>5</sup> Hz at open circuit voltage.



Fig.S1 The synthetic routes for composite materials.



**Fig. S2** Solubility of CN, OCN3, OCN24, and OCN72 in water (from left to right) after ultrasonication for 0.5 h and then stood still for another 0.5h.

As shown in Fig. S2, the solubility of the polymeric materials in water improves with an increase in the extent of oxidation. Large  $g-C_3N_4$  chunks can be observed in the CN aqueous solution which reveals the poor solubility of CN. In OCN3 aqueous solution, there are only a tiny amount of OCN3 are deposited at the bottom of the solution, indicating its improved solubility compared to CN. Furthermore, no deposit can be found in OCN24 and OCN72 aqueous solutions, which implies the gradually improved solubility of the polymeric materials due to increased oxidation treatment time.



**Fig. S3** (a-c) SEM images of (a) GOOCN3 (b) GOOCN72 and (c) GOFOCN at high magnification; (d-f) TEM images of (d) GOCN-3 (e) GOOCN24 and (f) GOFOCN at high magnification.

Fig. S3 shows the SEM and TEM images of GOCN-3, GOOCN3, GOOCN72, and GOFOCN. From these figures we can conclude that the reduced graphene oxide planes combined with the  $g-C_3N_4$  (or oxidized  $g-C_3N_4$ ) segments and formed layered 3D networks during the hydrothermal reaction. In these composites, the reduced graphene oxide acts as the skeleton on which there are  $g-C_3N_4$  (or oxidized  $g-C_3N_4$ ) deposits.

Sample	CN	OCN3	OCN24	OCN72	FOCN
Carbon content (at.%)	50.24	50.48	47.89	45.42	42.01
Nitrogen content (at.%)	42.63	36.35	36.95	36.98	1.96
Oxygen content (at.%)	7.13	13.17	15.15	17.60	56.02

 Table. S2 Carbon, nitrogen and oxygen contents of CN, OCN and FOCN samples.

Sample	GOCN-3	GOOCN3	GOOCN24	GOOCN72	GOFOCN
Carbon content (at.%)	64.09	79.22	79.66	81.82	85.67
Nitrogen content (at.%)	24.59	4.76	2.50	1.82	<0.01
Oxygen content (at.%)	11.32	16.02	17.84	16.36	14.32

Table. S3 Carbon, nitrogen and oxygen contents of GOCN, GOOCN and GOFOCN samples.



**Fig. S4**(a) FTIR spectra of OCN3, OCN72, and FOCN; (b-d) XPS (b) survey spectra of OCN3, OCN72, and FOCN; (c) C1s and (d) N1s spectra of OCN3, OCN72, and FOCN, respectively.

Fig. S4(a) shows the FTIR spectra of OCN3, OCN72, and FOCN samples. The FTIR spectra of these samples display peaks centred at ~800 cm<sup>-1</sup> which is caused by the vibration of triazine rings in g-C<sub>3</sub>N<sub>4</sub><sup>[S2]</sup>. The peaks located between 1250 to 1600 cm<sup>-1</sup> are attributed to the vibration of aromatic C-N bonds in carbon-nitrogen heterocycles [S3]. It is noteworthy that the number of characterized C-N bond peaks in FTIR spectrum of FOCN sample is much less than that in the FTIR spectra of OCN3 and OCN72 samples, revealing the relatively low nitrogen content of FOCN sample due to the strong oxidation treatment. In addition, the broad peaks centred at  $\sim$ 3100 cm<sup>-1</sup> correspond to the vibration of –OH and –NH groups. The tiny peak at 1740 cm<sup>-1</sup> is attributed to the vibration of -COOH groups. The intense peaks centred at ~2700 cm<sup>-1</sup> and ~1050 cm<sup>-1</sup> are caused by the vibration of –OH bonds in carboxyl group and C-O bonds in the hydroxyl group, respectively. The XPS survey spectra of GOOCN3, GOOCN72, and GOFOCN are shown in Fig. S4(b). There are C 1s, O 1s, C KLL, and O KLL peaks in XPS survey spectra of all the samples. However, no obvious N 1s and N KLL peaks are shown in XPS survey spectrum of GOFOCN sample, indicating the low nitrogen content of GOFOCN sample. Furthermore, according to the XPS C 1s spectra displayed in Fig. S4(c), there are C-O, C=O, C-O-C, and C-O=C coordination in GOOCN3, GOOCN72, and GOFOCN samples. Nevertheless, similar to GOOCN3 and GOOCN72, no obvious C-N=C peak is displayed in the XPS C 1s spectrum of the GOFOCN sample, which further confirms GOFOCN's low nitrogen content. Fig. S4(d) confirms that all the nitrogen atoms in GOOCN3, GOOCN72, and GOFOCN samples are pyridinic nitrogen and pyrrolic nitrogen, revealing that the oxidation

treatment has successfully converted graphitic nitrogen to edge nitrogen in the polymeric materials.



**Fig. S5** Kinetic analysis results of (a) GOCN-3 (b) GOOCN3 (c) GOOCN72 (d) GOFOCN from CV curves.

Fig. S5 shows the kinetic analysis results of GOCN-3, GOOCN3, GOOCN72, and GOFOCN electrodes. The kinetic analysis was performed using the Dunn method recorded in previous papers <sup>[S4, S5]</sup>. It is obvious that the surface capacitive effect dominates the capacitance.



**Fig. S6** Rate performances in alkaline electrolyte of (a) GOCN-1, GOCN-2, GOCN-3, and GOCN-4 samples; (b) GOCN-3, GOOCN3, GOOCN24, GOOCN72, and GOFOCN samples.

The rate performances of GOCN-1, GOCN-2, GOCN-3, and GOCN-4 electrodes are illustrated in Fig. S6 (a). It was found that GOCN-3 electrode possesses the highest specific capacitance as well as the best rate performance among all the GOCN samples, indicating that GOCN composites reach their optimized performances when the mass ratio of graphene oxide/g-C<sub>3</sub>N<sub>4</sub> is 3/1. From Fig S6 (b), the pre-oxidation treatment of  $g-C_3N_4$  significantly increases specific capacitance of the composite materials. Moreover, it can be seen that the effect of the oxidation treatment reaches its maximum when the treatment time is 24 h. It is worth mentioning that although the specific capacitance of GOFOCN is very high it is only slightly lower than the specific capacitance of GOOCN24 at low current densities (224.5 F g<sup>-1</sup> at 1 A g<sup>-1</sup>), its rate performance is relatively poor (whereby the specific capacitance value deteriorates to 44.1 F g<sup>-1</sup> at a high current density of 50 A g<sup>-1</sup>).



Fig. S7 Equivalent circuit employed for fitting Nyquist plots

The Nyquist plot is fitted to an equivalent circuit shown in Fig. S7 using the following equation <sup>[S6]</sup>:

$$Z = Rs + \frac{1}{j\omega CPE1 + \frac{1}{Rct + Zw}} + \frac{1}{j\omega CPE2 + \frac{1}{Rleak}}$$

 $R_{ct}$  and  $R_{leak}$  are the charge transfer resistance and low frequency leakage resistance, respectively.  $R_s$  is the cell internal resistance that includes the intrinsic electronic resistance of the electrode material, the ohmic resistance of the electrolyte and the interfacial resistance between the electrode and the current collector.  $C_{PE1}$  and  $C_{PE2}$  are the double layer capacitance and low frequency mass capacitance, respectively.  $Z_W$  governs the Warburg element. ESR stands for the sum of  $R_s$  and  $R_{ct}$ .

Sample	R <sub>s</sub> (ohm)	R <sub>ct</sub> (ohm)	ESR (ohm)	
GOCN-3 (alkaline electrolyte)	1.04	1.05	2.09	
GOOCN24 (alkaline electrolyte)	0.95	0.06	1.01	
GOOCN24 (acid electrolyte)	0.78	0.04	0.82	
GOOCN24 supercapacitor	2.61	6 75	0.26	
(2 M KOH electrolyte)	2.01	0.75	9.30	

**Table. S5** R<sub>s</sub>, R<sub>ct</sub>, and ESR values of GOCN-3 and GOOCN24 electrodes or supercapcitors in alkaline and acid electrolyte.



**Fig. S8** Electrochemical performance of a combined device that consists of two symmetrical supercapacitors (connected in series) using GOOCN24 and 2 M KOH as the electrode materials and electrolytes, respectively (a) CV plots measured at a scan rate of 20 mV s<sup>-1</sup>; (b) GCD tests conducted at a current density range between 0.5 and 10 mA cm<sup>-2</sup>; (c) Rate performance calculated from GCD test; (d) Ragone plot obtained from GCD test.

Fig. S8 illustrates the electrochemical performance of a combined device that consists of two GOOCN24 symmetrical supercapacitors (connected in series). Although the CV curves of the combined device displayed in Fig. S8(a) show less ideal rectangular shapes when compared to the CV plots shown in Fig. 4(a) which belong to single GOOCN24 supercapacitor, the electrical double layer behaviour of the combined device is still good. This implies GOOCN24 has promising potentially practical application. The specific capacitance of the combined device can be calculated using GCD test result shown in Fig. S8(b). The combined device possesses a specific capacitance of 40.3 F g<sup>-1</sup> at 0.5 mA cm<sup>-2</sup> current density. It still maintains a specific capacitance of 25.9 F g<sup>-1</sup> when the current density increases to 5 mA cm<sup>-2</sup>. Fig. S8(c) shows that the rate performance of the combined device is good. A Ragone plot depicted in Fig. S8(d) was obtained from the GCD test. The gravimetric energy density and gravimetric power density of the GOOCN24 electrode in combined device can reach 13.90 Wh kg<sup>-1</sup> and 430.57 W kg<sup>-1</sup>, respectively at 0.5 mA cm<sup>-2</sup> current density. Hence, from the electrochemical tests results of the combined device, we can conclude GOOCN24 has marked potential as supercapacitor electrode in industry.



**Fig. S9** Ragone plot of gravimetric energy density *vs.* gravimetric power density of the GOOCN24 material on supercapacitor electrodes.



**Fig. S10** Electrochemical performance of a GOOCN24 symmetrical supercapacitor using 6 M KOH as the electrolyte (a) CV plots measured at scan rates ranging from 1 to 50 mV s<sup>-1</sup>; (b) Kinetic analysis result from CV curves; (c) Nyquist plot; (d) Cycling stability test conducted at a high scan rate of 100 mV s<sup>-1</sup>.

Fig. S10(a) displays the CV plots of a GOOCN24 symmetrical supercapacitor in 6 M KOH electrolyte. The CV curves possess rectangular shapes at both low and high scan rates which imply the ideal capacitive performance as well as good rate performance of GOOCN24 supercapacitor. The kinetic analysis shown in Fig. S10(b) demonstrates that most of the capacitance of the supercapacitor is governed by surface capacitive effects. The Nyquist plot shown in Fig. S10(c) indicates low internal resistance and charge transfer resistance values in the device. Cycling stability test was also performed to explore long term performance of the supercapacitor. The results shown in Fig. S10(d) proves that the device has tremendous cycling stability of 94.4% capacitance retention after 24000 CV cycles.



**Fig. S11** (a) CV curves of the asymmetrical supercapacitor at 50 mV/s in different voltage ranges; (b) GCD tests at various current densities.

Material	Electrolyte	Current density or scan rate	Specific capacitance	Stability
This work	1 M H <sub>2</sub> SO <sub>4</sub>	1 A g <sup>-1</sup>	265.6 F g <sup>-1</sup>	94 % capacitance retention after 5000 CV cycles
Laser reduced graphene [S7]	0.5 M Na <sub>2</sub> SO <sub>4</sub>	1.04 A g <sup>-1</sup>	141 F g <sup>-1</sup>	47 % capacitance retention after 2000 GCD cycles
Polyaniline/activated carbon composite [S8]	1 M H <sub>2</sub> SO <sub>4</sub>	0.2 A g <sup>-1</sup>	240 F g <sup>-1</sup>	83 % capacitance retention after 500 GCD cycles
Activated carbon [S9]	6 M KOH	1 A g-1	80 F g <sup>-1</sup>	97 % capacitance retention after 1000 GCD cycles
N-doped laminated graphene [S10]	6 M KOH	0.25 A g <sup>-1</sup>	245 F g <sup>-1</sup>	94.8 % capacitance retention after 2000 GCD cycles
Chemical vapour deposition graphene <sup>[S11]</sup>	1 M Na <sub>2</sub> SO <sub>4</sub>	0.5 A g <sup>-1</sup>	345.5 F g <sup>-1</sup>	84 % capacitance retention after 2000 GCD cycles
Flexible graphene film [S12]	1 M KOH	0.2 A g <sup>-1</sup>	211 F g <sup>-1</sup>	96 % capacitance retention after 5000 GCD cycles
3D N-doped mesoporous graphene <sup>[S13]</sup>	1 M H <sub>2</sub> SO <sub>4</sub>	100 mV s <sup>-1</sup>	279 F g <sup>-1</sup>	90.6 % capacitance retention after 5000 GCD cycles
Wire-like all-carbon <sup>[S14]</sup>	1 M H <sub>2</sub> SO <sub>4</sub>	0.2 A g <sup>-1</sup>	110 F g <sup>-1</sup>	98.5 % capacitance retention after 5000 GCD cycles
Ag NWs/3D-graphene foam/ordered mesoporous carbon <sup>[S15]</sup>	6 M KOH	1 A g <sup>-1</sup>	213 F g <sup>-1</sup>	91 % capacitance retention after 10000 CV cycles
Graphene quantum dots- carbon fibre grafted structure <sup>[S16]</sup>	1 M H <sub>2</sub> SO <sub>4</sub>	1 A g <sup>-1</sup>	213 F g <sup>-1</sup>	97 % capacitance retention after 5000 GCD cycles
Graphene hydrogels <sup>[S17]</sup>	6 M KOH	0.5 A g <sup>-1</sup>	190 F g <sup>-1</sup>	93 % capacitance retention after 2000 GCD cycles

KOH activated C <sub>70</sub> microstructure [S18]	1 M H <sub>2</sub> SO <sub>4</sub>	5 A g <sup>-1</sup>	191.6 F g <sup>-1</sup>	92.5 % capacitance retention after 5000 GCD cycles
B, N co-doped porous carbon <sup>[S19]</sup>	1 M H <sub>2</sub> SO <sub>4</sub>	3 A g <sup>-1</sup>	176 F g <sup>-1</sup>	99.4 % capacitance retention after 4000 GCD cycles
Porous N-doped carbon [S20]	1 M H <sub>2</sub> SO <sub>4</sub>	20 A g <sup>-1</sup>	134 F g <sup>-1</sup>	98.7 % capacitance retention after 11000 GCD cycles

Table. S6 Performance comparison of different carbon-based materials.

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