



Melamine-Fe complex induced GO assembly to form nitrogen-doped carbon for high-gravimetric/volumetric supercapacitors

Tian Ting^{1,†}, Xie Xingyan^{1,†}, Du Cheng^{1,*}, Zhang Yan¹, Wan Liu¹, Xie Mingjiang^{1,*},
Chen Jian¹, Wang Lei^{2,*}

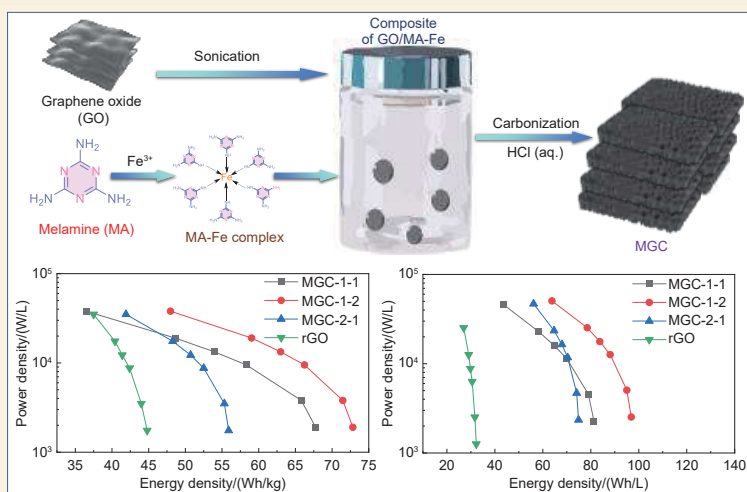
(1. Hubei Key Lab for Processing and Application of Catalytic Materials, Hubei Provincial Engineering Research Center of High Purity Raw Material

Processing Technology of Electronic Materials, Huanggang Normal University, Huanggang 438000, China;

2. Key Laboratory of Optic-electric Sensing and Analytical Chemistry of Life Science, College of Chemistry and Molecular Engineering,
Qingdao University of Science and Technology, Qingdao 266000, China)

Abstract: As electrodes for supercapacitors, carbon-based materials are facing challenging task of simultaneously achieving high gravimetric and volumetric energy storage capabilities because of the trade-off relationship between the two. In this study, we used a coordination compound of melamine and iron to induce the assembly of graphene oxide (GO) and fabricate a porous carbon material through coordination and hydrogen bonding interactions between the complex and GO. The resulting melamine-graphene-carbon product (MGC) is highly compact with a packing density of 1.20-1.34 g/cm³ (vs. reduced GO of 0.72 g/cm³), a high electrical conductivity (25 S/cm), and is superhydrophilic. As an electrode for supercapacitors, a MGC-based symmetric device has a superior energy storage ability with high gravimetric and volumetric energy densities of 72.8 Wh/kg@1900 W/kg and 96.9 Wh/L@2527 W/L, respectively. These values are 1.6 times higher than those of a reduced GO-based device and 1.79 times higher than those of devices based on an industrial activated carbon (YP-80F, Kuraray, with a maximum energy density of 40.6 W/kg@1500 W/kg). The exceptional energy storage performance of the developed material indicates that the synthesis route presented in this study is feasible and provides new insights into creating highly compact carbon materials for a range of applications.

Key words: Melamine-Fe complex; GO; Superhydrophilicity; High conductivity; Supercapacitor



1 Introduction

Developing new energy storage devices to meet the growing demand for electric power storage in portable devices is an urgent imperative. Among these devices, supercapacitors (SCs) possess unique characteristics such as high power density and long cycling life^[1-3], which make them suitable for applications that require rapid charge/discharge, such as a backup power for the initial acceleration and climbing of electric vehicles. The high power density of SCs can be used to compensate for the power limitations of sec-

ondary batteries. However, the drawback of SCs lies in their low energy density compared to secondary batteries, typically ranging from 10 to 40 Wh/kg. A current focus in this field is the development of new electrode materials or electrolytes to increase the energy density of SCs. Given that the energy density of

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a SC is determined by the formula $E = \frac{1}{2} CV^2$ (where C represents capacity and V is working voltage), exploring electrode materials with high capacity (C) is normally used to enhance energy density^[4-6].

Carbon materials^[7-10] with various morphologies and structures are commonly utilized as electrodes for SCs due to their large surface area, good conductivity, and excellent structural stability. Carbon-based materials, particularly activated carbons (ACs)^[11-12], have been successfully utilized as supercapacitor electrode materials as they are able to achieve high power density and long cycling life. However, the energy storage mechanism in carbon-based SCs primarily relies on the reversible adsorption and desorption of electrolytes onto the surface of carbon materials to form electric double layers (EDLs)^[13-14], with the capacitance determined mainly by their active surface areas. Increasing the surface area can lead to a disruption in the ordered graphitic microstructure, resulting in a decrease in inherent electric conductivity. Consequently, there exists a trade-off between surface area and electric conductivity in carbon materials. It is challenging to simultaneously achieve a carbon material with both a large surface area and high electric conductivity to ensure a SC with high energy and power density. Moreover, increasing the surface area is typically achieved by creating high microporosity, leading to a decrease in the packing density (ρ) of the resulting carbon material. Packing density plays a crucial role in the volumetric energy density of SCs, which is key for practical applications in electric vehicles. The relationship between volumetric and gravimetric energy

density can be expressed as E_v (volumetric energy density) = $\rho \cdot E_g$ (gravimetric energy density)^[15-17]. Therefore, it is essential to develop carbon materials with high microporosity and packing density in order to achieve high energy density. Microporosity is crucial for maximizing gravimetric energy density^[18-20], while large packing density is necessary for enhancing volumetric energy density. However, achieving both high microporosity and packing density in a carbon material while maintaining good conductivity is challenging. Recent research has shown that incorporating reduced graphene oxide (rGO) into carbon structure can significantly improve their electric conductivity^[4]. Therefore, one approach to address this challenge is to introduce GO during the fabrication of microporous carbon materials in order to achieve the desired carbon material with high microporosity, packing density and electrical conductivity.

In this study, as depicted in Fig. 1, a carbon material with high packing density (1.20-1.34 g/cm³), high conductivity (up to 25 S/cm), and abundant N/O functionalities was developed through the coordination of melamine and iron (MA-Fe) induced assembly of GO via coordination and hydrogen bonding interactions. In carbonization, the coordinated structure of MA-Fe formed microporosity, the introduced GO was transformed in situ to rGO to enhance the electric conductivity and N in MA and O in GO were partially retained as N/O functional groups to provide additional capacitance through reversible faradaic reactions. Melamine has been widely employed as a nitrogen source for fabricating nitrogen-doped graphene.

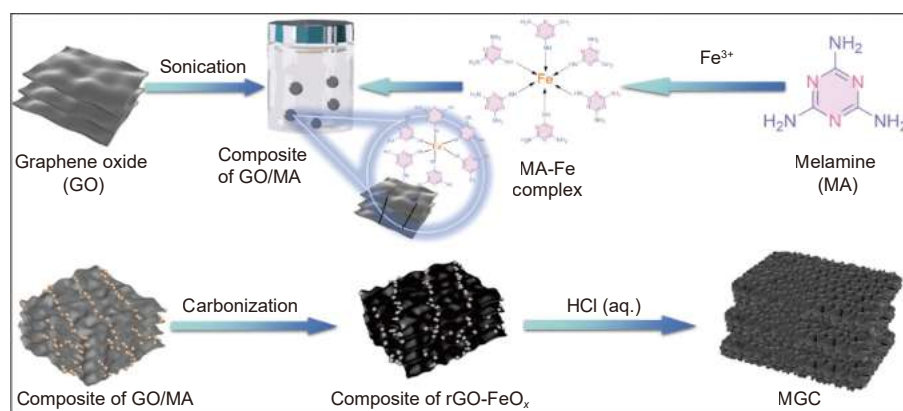


Fig. 1 Schematic of the fabrication of MGCs by the complex induced assembly of GO and MA-Fe

However, the fabrication methods of nitrogen-doped graphene are mainly thermal annealing or the CVD method^[21–22]. Melamine is not dissolved in deionized water, which is unfavorable for its assembly with GO. In this report, the dissolution of melamine in deionized water was achieved by adding coordination of Fe^{3+} , which enabled the formation of a melamine-Fe complex that dissolved in water to allow subsequent assembly with GO. Moreover, the introduced Fe could be uniformly distributed in the assembled GO-melamine composite, so that catalytic graphitization of the carbon skeleton by Fe could be effectively achieved, thereby increasing the conductivity of the final carbon. The products obtained through this method can also be used in various fields, such as catalyst supports for anchoring metal ions and as adsorbents for contaminant removal.

2 Experiment

2.1 Synthesis

The experimental procedure involved the dissolution of 1.0 g of iron chloride in 20 mL of deionized water to obtain a solution of Fe^{3+} . Subsequently, x grams of melamine (where $x=0.5$ or 1.0) were added to the Fe^{3+} solution with vigorous stirring to achieve a homogeneous solution of MA-Fe. In a separate step, y grams of GO (where $y=0.5$ or 1.0) were dissolved in 500 mL of distilled water under ultrasonic stirring to obtain the GO solution. The MA-Fe solution was then added dropwise to the GO solution with stirring for 3 h. The resulting mixture of MA-Fe and GO was evaporated at 60 °C for 12 h to form a composite mixture. The composite mixture was subsequently annealed at 700 °C for 2 h under nitrogen atmosphere to yield carbonized products. To remove any remaining Fe residue, acid etching was conducted and the mixture was filtered and rinsed for multiple times with ionized water. The final products obtained were labelled as MGC- x - y , where x and y respectively indicate the mass of MA and GO used in the preparation process.

2.2 Characterization

The samples underwent analysis for morphology,

microstructure, and element distribution using a Scanning Electron Microscope (SEM, Zeiss Sigma 300) and Transmission Electron Microscopy (TEM, Thermo Fisher Talos F200X). X-ray Diffraction (XRD, LabX XRD-6100, SHIMADZU) and Raman Spectroscopy (Renishaw Invia Raman microscope) were utilized to investigate crystallinity. Textural properties were measured with a Tristar 3020 instrument. Surface chemistry was examined using Fourier Transform Infrared Spectroscopy (FT-IR, Nicolet 6700, Thermo fish) and X-ray Photoelectron Spectroscopy (XPS, Thermo Scientific ESCALAB). Water contact angles were determined with a Dataphysics Oca20 instrument. The conductivity was evaluated through the four-probe method on the ST2742B automated powder resistivity tester. Thermogravimetric Analysis (TGA) of the mixture was carried out on a TA SDT Q600. The density of electrode material is determined by weighing a sample of a certain mass (m) and pressing it into a uniform circular shape at 10 MPa. The volume of the circular shape is calculated by measuring its diameter and thickness, and the density was then calculated by formula of $\rho=m/V$ (g/cm^3).

2.3 Electrochemical test

The working electrode was prepared by mixing the obtained sample powder with carbon black and polytetrafluoroethylene (PTFE) at a mass ratio of 80 : 10 : 10 in absolute ethanol to a sticky slurry, followed by coating the resulted mixture into coin-type nickel foam with the size of 1.0 cm^2 before being pressed together at 10.0 MPa. The loading amount of active material is about 1.0 mg/cm^2 . The electrochemical properties of the symmetric supercapacitor were investigated with a two-electrode cell configuration using the fabricated electrode as positive as well as negative electrode (with the loading of active material in equal amount) in 1.0 mol L^{-1} TEABF₄/AN electrolyte solution (1.5 mL). Cyclic voltammetry and galvanostatic charge-discharge were conducted on a CHI 660D electrochemical workstation (Shanghai CH Instrument Company, China). The specific capacitance of the symmetric supercapacitor was calculated from the discharge curve according to Eq. (1)

$$C_s = 2 \times \frac{I\Delta t}{m\Delta V} (\text{F/g}), \quad (1)$$

where I is the current density (A), Δt is the discharge time (s), m is the mass of the single electrodes, ΔV is the potential window of discharging (V). The energy densities and power densities were calculated according to Eq. (2-3)

$$E = \frac{1}{8} \times \frac{1}{3.6} \times C_s V^2 (\text{Wh/kg}) \quad (2)$$

$$P = \frac{E}{t} (\text{W/kg}) \quad (3)$$

where C_s (F/g) is the capacity of the asymmetric supercapacitor, V (V) is the working voltage of the cell, t is the discharge time.

3 Results and discussion

The thermal degradation behavior of the composite of MA-Fe and GO was examined by thermogravimetric analysis (TGA). The TGA results presented in Fig. S1 reveal that pure GO exhibits a notable endothermic peak at approximately 219 °C, corresponding to deoxygenation of GO to rGO. Interestingly, this peak is observed to shift to temperatures exceeding 280 °C in the presence of MA-Fe, indicating that the decomposition of GO is delayed possibly due to the stabilizing effect of MA-Fe for GO. This effect is attributed to the coordination of iron ions with GO and the formation of hydrogen bonds between GO and the MA-Fe complex. Furthermore, an additional endothermic peak at around 600 °C is observed for the three composites of MA-Fe and GO, which can be linked to carbonization of the composites. This observation suggests that the selected annealing temperature of 700 °C is high enough to promote carbonization of the composites, resulting in the formation of carbonaceous materials.

The evolution of crystallinity was monitored using X-ray diffraction (XRD). Fig. S2a displays the XRD patterns of composites of MA-Fe and GO, revealing a broad diffraction peak at 16°, attributable to the diffraction of restacked GO, as well as sharp diffraction peaks corresponding to the MA-Fe complex. Fig. S2b illustrates the XRD patterns of the annealed composites, clearly showing the disappearance of dif-

fraction peaks associated with the MA-Fe complex due to carbonization, leading to the breakdown of the complex structure. After the removal of Fe species through acid etching, the resulting carbon products (MGCs) were obtained. To investigate the morphology evolution of the 3 MGCs, scanning electron microscope (SEM) analysis was conducted. Fig. 2 presents the SEM of the three MGCs at different magnifications. It is evident that all three MGCs exhibit a nanosheet morphology, with the MGC-1-2 product derived from a low ratio of MA-Fe to GO displaying a stacked structure as depicted in Fig. 2e and 2f. This observation can be attributed to the high proportion of GO used during the synthesis process, leading to the tendency of GO to restack in solution and the stacked morphology after carbonization. The microstructures of the resulting MGCs were evaluated using transmission electron microscopy (TEM). Taking MGC-12 as an example, Figs. 3a-c show TEM micrographs captured at different magnifications. These images all reveal a two-dimensional (2D) microstructure, which is in agreement with the findings from SEM analysis. The high-resolution TEM image in Fig. 3d displays disordered fringes of domains, indicative of the graphitic structure of graphite nanocrystallites. Fig. 3e depicts a scanning transmission electron microscopy (STEM) image of the synthesized MGC along with corresponding element maps of carbon, nitrogen and oxygen. The element maps consistently exhibit a similar pattern to that of the parent STEM image, indicating a uniform distribution of elements in MGC.

The crystallinity of the resulting MGC was analyzed using XRD and Raman spectroscopy. Fig. 4a presents the XRD patterns of MGCs, showing two broad diffraction peaks at approximately 16° and 26°, which can be attributed to the diffraction of stacked rGO and nanographite crystallites, respectively. Raman spectroscopy was employed to further verify the crystallinity of the MGCs. Fig. 4b displays the Raman spectra of the 3 MGCs, demonstrating two broad Raman peaks at around 1350 and 1560 cm⁻¹, corresponding to the *D*-band and *G*-band vibrations of sp³ and sp² carbon bonds, respectively^[23,24]. The graphitiz-

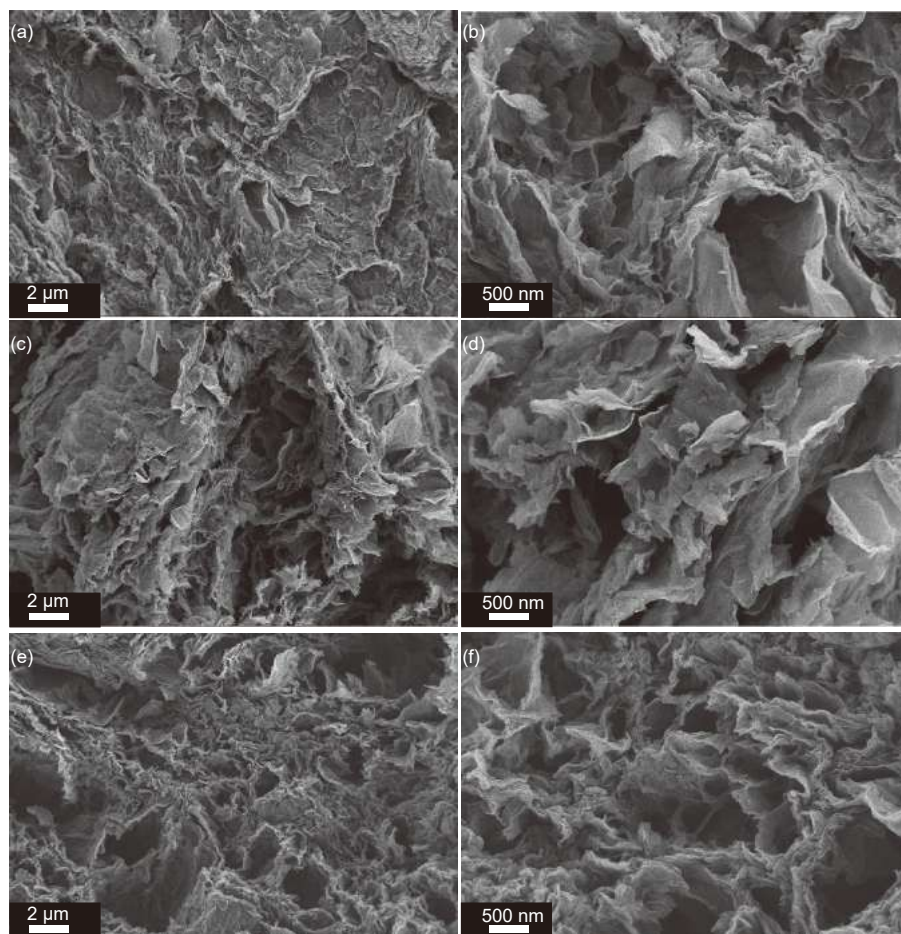


Fig. 2 The morphology evolution analysis of the resultant MGCs: (a, b) MGC-2-1. (c, d) MGC-1-1. (e, f) MGC-1-2

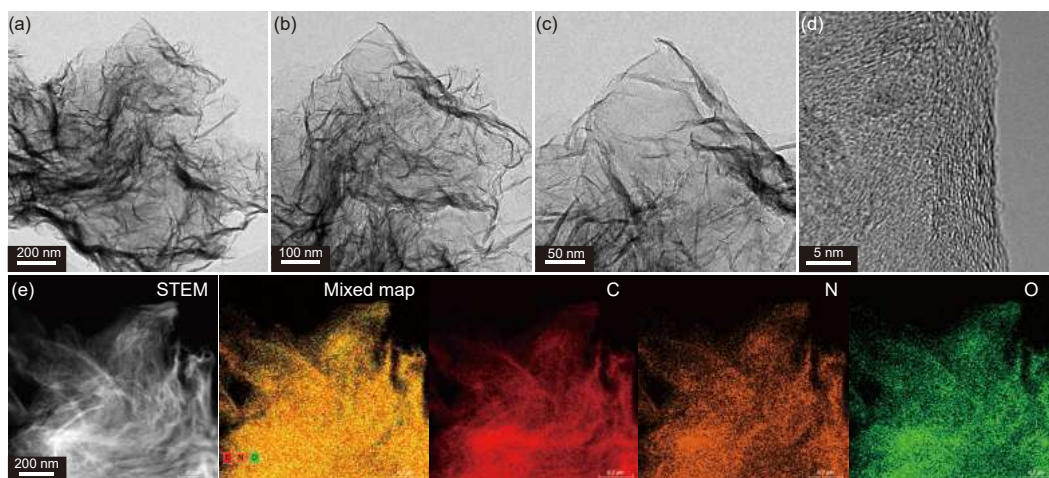


Fig. 3 Microstructure analysis of the resultant MGC-1-2: (a-c) TEM micrographs viewed at different magnifications. (d) high-resolution TEM image. (e) STEM and the corresponding elemental mapping of C, N and O

ation degrees of the three MGCs were assessed by calculating the intensity ratio of the D -band to the G -band, denoted as I_D/I_G , where a lower I_D/I_G value indicates a higher graphitic degree. As shown in Fig. 4b, the calculated I_D/I_G values were 1.166 for MGC-1-1, 1.058 for MGC-1-2 and 0.984 for MGC-2-1. These

results suggest that MGC-2-1 exhibits a slightly higher degree of graphitization compared to the other MGCs. The enhanced graphitization degree of MGC-2-1 can be ascribed to the efficient catalytic graphitization of carbon substrates induced by the incorporated Fe species. During the synthesis of MGC-2-1, an

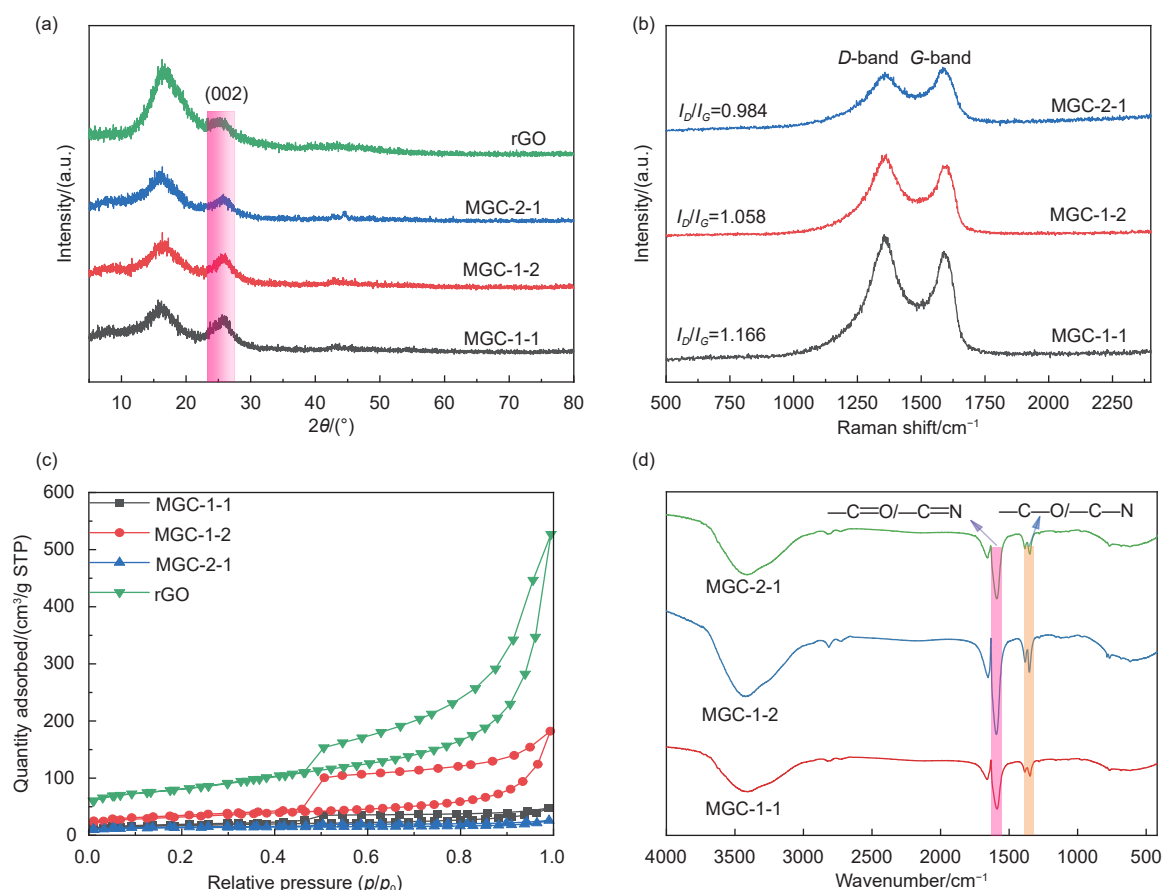


Fig. 4 The analysis results on crystallinity and surface chemistry. (a) XRD patterns. (b) Raman spectra. (c) nitrogen sorption isotherms and (d) FT-IR spectra

increased melamine dosage facilitates sufficient coordination with Fe ions, enabling the uniform dispersion of Fe active sites throughout the carbon composite. Well-distributed and high content Fe species of MGC-2-1 thereby promote high-efficiency catalytic graphitization and ultimately yield a high graphitization degree. In contrast, insufficient melamine addition triggers severe self-aggregation of Fe nanoparticles, which gives rise to heterogeneous metal dispersion and consequently reduces the graphitization degree of carbon materials. The nitrogen sorption isotherms test results presented in Fig. 4c indicate that both the resultant MGCs and rGO exhibit an IV-shaped pattern with a distinct hysteresis loop at a relative pressure range of 0.4-1.0, indicating the pres-

ence of a mesoporous structure. The pore size distribution plots shown in Fig. S3 reveal a mean pore size of 2.6 nm, further confirming the presence of a mesoporous structure. The parameters of the resultant MGCs are provided in Table 1, which shows that the surface area decreases to some extent after GO was assembled with the MA-Fe complex. This decrease could be attributed to the destruction of melamine, which can generate corrosive gases leading to the collapse of the microstructure and subsequent reduction in surface area. The packing density of the resultant products was evaluated by compressing the samples into regular discs and calculating the density using the formula $\rho = m/V$. Compared to rGO with a density of 0.72 g/cm^3 , the resultant MGCs demonstrate in-

Table 1 The texture properties of the MGCs and rGO

Sample	$S_{\text{Total}}/(\text{m}^2/\text{g})$	$V_{\text{Total}}/(\text{cm}^3/\text{g})$	$S_{\text{Micro}}/(\text{m}^2/\text{g})$	$S_{\text{Exter}}/(\text{m}^2/\text{g})$	$S_{\text{Exter}}/S_{\text{Total}}/\%$	Packing density/ $(\rho, \text{g}/\text{cm}^3)$	Conductivity $(\sigma, \text{S}/\text{cm})$
rGO	278	0.84	77	201	72	0.72	4.91
MGC-1-1	56	0.07	18	38	68	1.20	25.0
MGC-1-2	109	0.29	24	85	78	1.33	7.1
MGC-2-1	42	0.05	22	20	48	1.34	13.7

creased packing densities of 1.20 g/cm³ for MGC-1-1, 1.33 g/cm³ for MGC-1-2, and 1.34 g/cm³ for MGC-2-1. The inherent conductivity (σ) of the resulting MGCs was evaluated using a four-probe method. The conductivity values for the three MGCs are 25 S/cm for MGC-1-1, 7.11 S/cm for MGC-1-2 and 13.7 S/cm for MGC-2-1. In contrast, rGO has a conductivity of 4.91 S/cm. It is evident that the introduction of rGO component has significantly enhanced the conductivity of the MGC materials. These findings support our research hypothesis, which indicated that incorporating GO during the synthesis process and subsequently converting it to rGO in carbonization at high temperatures would lead to improved conductivity in the final material.

Fig. 4d illustrates the Fourier-transform infrared (FT-IR) spectra of the MGCs, which reveal two prominent absorption peaks at 1580 and 1350 cm⁻¹, which correspond to the vibrations of $-\text{C}=\text{O}/-\text{C}=\text{N}$ and $-\text{C}-\text{O}/-\text{C}-\text{N}$ functional groups, respectively. Further analysis of the surface species of residual N and O was conducted using X-ray photoelectron spectroscopy (XPS). Fig. S4a depicts the overall survey XPS

spectra of the 3 MGCs, indicating the presence of 3 elemental constituents: C, N and O. The survey XPS shows only a faint peak around 710 eV, suggesting that Fe was effectively removed by acid etching, which is consistent with the XRD results. Fig. S4b-d present the high-resolution C 1s XPS spectra of the 3 MGCs, which were resolved into 3 distinct species located at 284, 285 and 288 eV, corresponding to $-\text{C}=\text{C}-$, $-\text{C}-\text{C}-$ / $-\text{C}-\text{O}/-\text{C}-\text{N}$ and $-\text{C}=\text{O}/\text{C}=\text{N}$ groups, respectively. Figs. 5a-c exhibit the high-resolution N 1s XPS spectra of the 3 MGCs, revealing three distinct peaks at 398 eV (pyridinic N, N-6), 400 eV (pyrrolic N, N-5), and 401 eV (graphitic N, N-Q)^[25-26]. The residual O species in the 3 MGCs, as shown in Figs. 5d-5f, were fitted into 2 species located at 531 eV ($-\text{C}=\text{O}$, O-II) and 533 eV ($-\text{C}-\text{O}$, O-I)^[27].

The specific composition of the three MGCs is tabulated in Table 2. It is evident that MGC-1-1 and MGC-1-2 exhibit relatively stable levels of N-containing and oxygenic species, whereas MGC-2-1 shows a pronounced decrease in N species. The possible reason is the catalytic graphitization effect of Fe at the

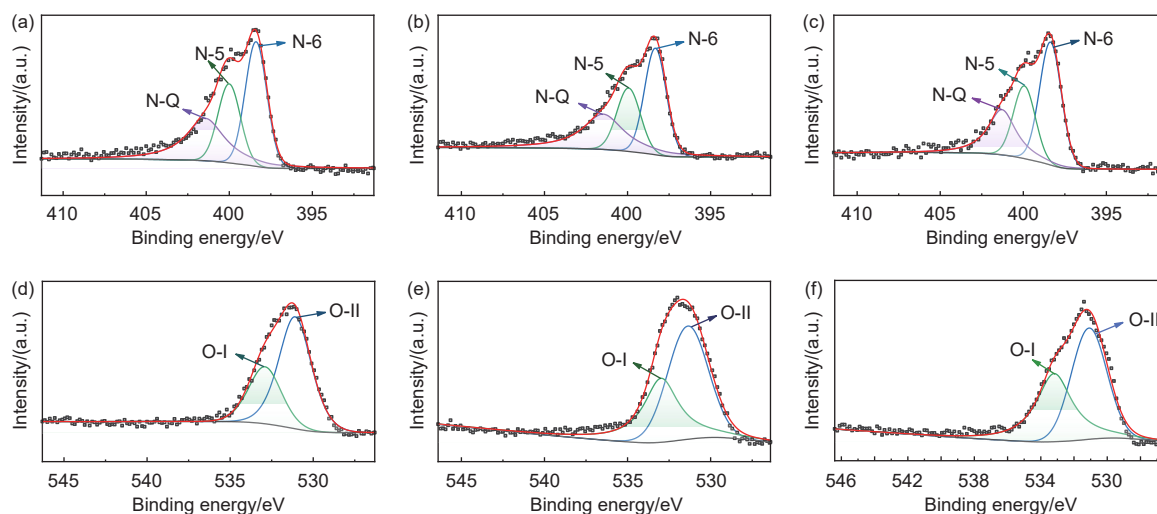


Fig. 5 The high-resolution N 1s XPS of (a) MGC-1-1, (b) MGC-1-2, (c) MGC-2-1, and O 1s XPS of (d) MGC-1-1, (e) MGC-1-2, and (f) MGC-2-1

Table 2 The surface elements and chemical species of the MGCs

Samples	C/(%, atom fraction)	N/(%, atom fraction)	O/(%, atom fraction)	N species/(%, atom fraction)			O species/(%, atom fraction)	
				N-6	N-5	N-Q	O-I	O-II
MGC-1-1	82.37	10.33	7.31	4.17	2.76	3.40	2.45	4.86
MGC-1-2	81.99	10.27	7.74	4.10	2.65	3.52	3.11	4.63
MGC-2-1	82.19	9.25	8.56	4.24	2.50	2.51	3.76	4.80

MA-to-GO ratio of 2 : 1. This ratio may facilitate the coordination of Fe with MA toward a nearly saturated coordination state, thereby promoting the homogeneous distribution of Fe throughout the melamine framework and enhancing catalytic graphitization. Due to catalytic graphitization, nitrogen and oxygen tends to escape rather than doped into the formed carbon skeleton. In addition, Raman spectra analysis indicates that the resultant MGC-2-1 has the highest graphitization degree, which indirectly indicates that, as compared with other ratios, the catalytic graphitization effect of iron leads to a decrease in nitrogen content and an increase in graphitization degree when the ratio of MA to GO is 2 : 1.

The surface wettability was investigated by measuring the water contact angle (WCA). Fig. 6 displays the process of water droplets when they contact with the materials, which was captured through photography. The images obtained demonstrate that the final WCA values of 3 modified graphene composites (MGCs) are all 0°, suggesting superhydrophilicity, attributed to the abundant N and O functionalities that facilitate strong interactions between the surface functionalities and water molecules. The inclusion of N and O functionalities not only enhances hydrophilicity, but also increases surface polarity, thereby facilitating the adsorption of ions. Such properties are advantageous for various applications such as adsorp-

tion and supercapacitors.

The energy storage performance of the MGCs was systematically investigated. Symmetric SCs were assembled using organic electrolytes consisting of 1.0 mol L⁻¹ TEABF₄/AN. The organic electrolyte was chosen due to its ability to provide a wide operating voltage range, allowing for high energy density of SCs. The cyclic voltammetry (CV) plots of the assembled SCs based on the MGC-1-1 (Fig. S5a), MGC-1-2 (Fig. 7a), MGC-2-1 (Fig. S6a), and rGO (Fig. S7a) all exhibit rectangular-shaped curves, indicating typical electric double-layer capacitance (EDLC) behavior.

The galvanostatic charge-discharge (GCD) curves of the assembled SCs for MGC-1-1 (Fig. S5b), MGC-1-2 (Fig. 7b), MGC-2-1 (Fig. S6b), and rGO (Fig. S7b) all display triangular-shaped patterns, further confirming the EDLC behavior. The gravimetric capacities of the 4 devices were calculated using formula (1) provided in the supporting information, and correspondingly, the volumetric capacities were also determined using the formula $C_v = C_g \cdot \rho$, where C_g represents the gravimetric capacities, C_v denotes the volumetric capacities, and ρ represents the packing density. Fig. 7c illustrates the gravimetric capacities of the four devices, with the MGC-1-2 based device exhibiting the highest capacity of 145 F/g@1.0 A/g, surpassing the capacities of the other MGCs (135 F/g@1.0 A/g for MGC-1-1, 131 F/g@1.0 A/g for MGC-2-1) and rGO (105 F/g@1.0 A/g). Fig. 7d displays the volumetric capacities of the 4 devices, with the maximum capacities being 193 F/cm³@1.0 A/g for MGC-1-2, 176 F/cm³@1.0 A/g for MGC-2-1, 162 F/cm³@1.0 A/g for MGC-1-1, and 76 F/cm³@1.0 A/g for the rGO. The enhanced energy storage performance of the MGCs compared to rGO can be attributed to the introduction of N and O functionalities, which not only increase surface polarity facilitating the formation of EDLs, but also provide additional faradaic capacities. To quantitatively assess the specific contributions of EDLC and faradaic capacitance, trasatti analysis (TA) was conducted. In TA, the capacitance was plotted against the square root of

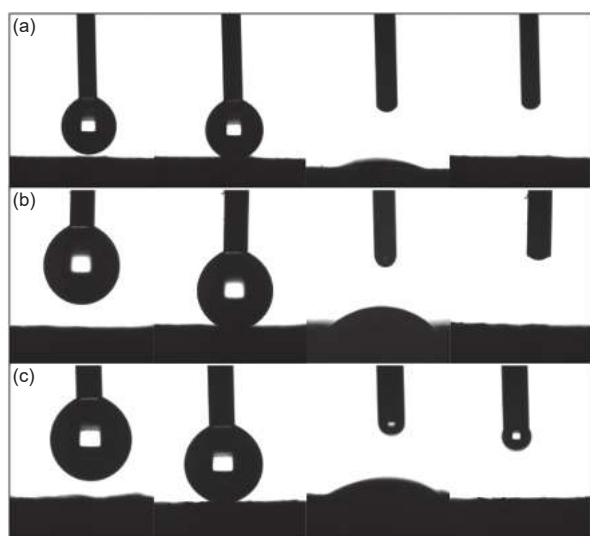


Fig. 6 Water contact angle (WCA) test results of the 3 MGCs recorded at different times: (a) MGC-1-1, (b) MGC-1-2 and (c) MGC-2-1

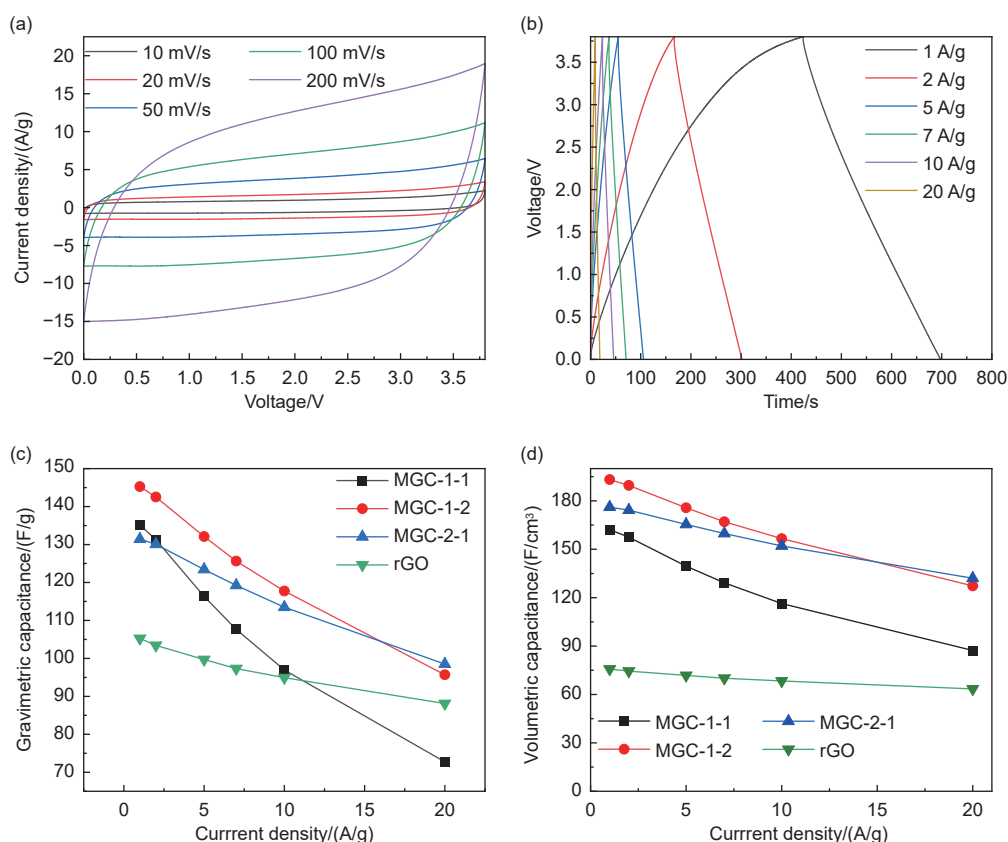


Fig. 7 The electrochemical performance of the samples. (a) CV plots of MGC-1-2 based SC. (b) GCD curves of the MGC-1-2 based SC. (c) Gravimetric capacitances and (d) volumetric capacitances of the 4 devices

charge/discharge time (Fig. S8). The intersection of the dotted line with the vertical axis represents the rate-independent capacitance, indicating that the EDLC contributions of the 4 devices are 81 F/g for MGC-1-1, 109 F/g for MGC-1-2, 104 F/g for MGC-2-1 and 92 F/g for rGO. This suggests that the faradaic capacitances contributed from nitrogen and/or oxygen species are approximately 54 F/g for MGC-1-1, 36 F/g for MGC-1-2, 27 F/g for MGC-2-1, and 13 F/g for rGO. It is evident that the faradaic capacitance contribution is directly proportional to the N content. Although the conductivity of MGC-1-1 is much higher than that of MGC-1-2, its capacitance performance is still inferior to that of MGC-1-2. This difference can be attributed to the larger surface area of MGC-1-2, which may provide more effective surface area for the formation of EDLCs. This hypothesis can be verified by the TA analysis. Therefore, the superior SC performance of MGC-1-2 can be attributed to its increased electrochemically-active surface area, which provides high EDLC, as well as the high content of

N/O species, offering additional faradaic capacitance.

The energy storage capacity of the MGCs was evaluated by plotting the energy density to power density (Ragone plots). Fig. S9a illustrates the gravimetric Ragone plots, indicating that the MGC-1-2-based device outperforms other MGCs at various power densities. The maximum energy density of 72.8 Wh/kg@1900 W/kg achieved by MGC-1-2 surpasses those of MGC-1-1 (67.8 Wh/kg@1900 W/kg), MGC-2-1 (55.9 Wh/kg@1750 W/kg), and rGO-based devices (44.8 Wh/kg@1750 W/kg). The enhanced energy storage capabilities of the MGC-based devices highlight the effectiveness of the current fabrication approach for carbon materials in energy storage applications. Additionally, the stacked structure of the MGCs enables an increased packing density, leading to superior volumetric performance.

Fig. S9b presents the volumetric performance of the MGC-based SCs in comparison to the rGO-based counterpart, demonstrating significantly higher volumetric energy densities for MGC-1-2 (96.9 Wh/L@

2527 W/L), MGC-1-1 (81.3 Wh/L@2280 W/L), and MGC-2-1 (74.9 Wh/L@2345 W/L) compared to rGO (32 Wh/L@1260 W/L). Even at a superhigh power density of 50 540 W/L, MGC-1-2 maintains a high energy density of 63.8 Wh/L, which is twice that of the rGO-based device. Overall, the Ragone plot analysis indicates that the synthesized MGCs exhibit superior energy storage capabilities compared to the original rGO, suggesting that the assembly of GO induced by melamine-Fe complex enhances carbon packing density, superhydrophilicity from N/O functionalities, and excellent conductivity from the rGO component. The high energy storage capability provided by the densely packed structure and abundant N/O species of the developed MGC SCs is evident in both its gravimetric and volumetric performance. A comparison was conducted among various carbons reported and the best MGC (MGC-1-2) to assess the energy storage level of the MGC-1-2, the results of which are presented in Fig. 8^[11,28–31]. It is clear from the comparison that the MGC-1-2 demonstrates significantly superior energy storage compared to recently reported carbon-based symmetric SCs. Specifically, the MGC-1-2 SC exhibits a higher energy storage level than an industrial activated carbon (YP-80F, Kuraray), with the energy density of MGC-1-2 being 1.79 times higher than that of YP-80F-based SCs (maximum energy density of 40.6 W/kg@1500 W/kg). The cycling performance of the MGC-1-2 based SC was evaluated by subjecting it to continual charge/discharge cycles at a high current density of 7.0 A/g. As shown in Fig. S10, after 30 000 cycles of charge/discharge, the MGC-1-

2-based device exhibits excellent cycling stability with nearly 100% capacitance retention and coulombic efficiency. The comprehensive analysis of the energy storage performance indicates that the MGC-1-2 holds great potential as a reliable candidate for practical energy storage applications.

4 Conclusion

We have successfully developed highly compact and porous carbons (MGCs) through a novel assembly strategy utilizing coordination and hydrogen bonding interactions between a melamine–iron complex and GO. The MGCs exhibit an exceptionally high packing density (1.20–1.34 g/cm³), superior electrical conductivity (up to 25 S/cm), and excellent superhydrophilicity. When used as an electrode in a SC, the best MGC-1-2 based symmetric device delivers outstanding gravimetric and volumetric energy densities of 72.8 Wh/kg and 96.9 Wh/L, respectively, significantly outperforming both rGO and commercial activated carbon (YP-80F). These results not only demonstrate the effectiveness of our synthesis strategy in breaking the conventional trade-off between gravimetric and volumetric performance but also highlight the scalability and potential of this approach for designing advanced compact carbon materials for next-generation energy storage systems.

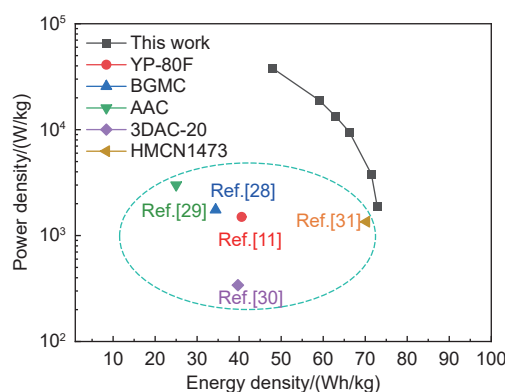


Fig. 8 Energy storage level of this work as compared to recently reported carbon-based symmetric devices

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