



Self-propagating high-temperature synthesis of activated carbon/graphene composites for high-performance lithium-ion capacitors

Hu Tao^{1,2,4}, Zhang Xiong^{1,2,3,*}, Li Chen^{1,3}, An Yabin^{1,2,3,*}, Sun Xianzhong^{1,2,3},
Wang Kai^{1,2,3}, Ma Yanwei^{1,2,3,*}

(1. State Key Laboratory of High Density Electromagnetic Power and Systems, Institute of Electrical Engineering,
Chinese Academy of Sciences, Beijing 100190, China;

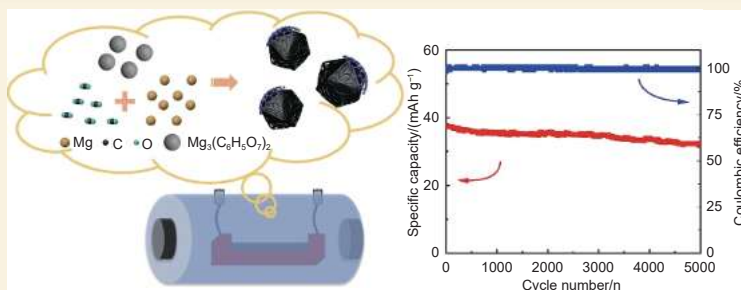
2. University of Chinese Academy of Sciences, Beijing 100049, China;

3. Shandong Key Laboratory of Advanced Electromagnetic Conversion Technology, Institute of Electrical Engineering and
Advanced Electromagnetic Drive Technology, Qilu Zhongke, Jinan 250013, China;

4. Guangdong Provincial Key Laboratory of Electric Power Equipment Reliability, Electric Power Research Institute of
Guangdong Power Grid Co., Ltd., Guangzhou, Guangdong 510080, China)

Abstract: Lithium-ion capacitors (LICs) are hybrid energy storage devices designed to bridge the gap between the high energy density of lithium-ion batteries and the high power density of supercapacitors. However, their performance is often limited by the low specific capacitance and conductivity of conventional activated carbon cathodes. We report the preparation of an activated carbon/graphene composite by a self-propagating high-temperature synthesis method in a CO₂ atmosphere, using magnesium powder and magnesium citrate as the precursors. The resulting composite is designated as MCG, where M denotes magnesiothermic reduction and CG denotes the activated carbon/graphene composite. Small two-dimensional graphene sheets are attached to the activated carbon surface, suppressing graphene restacking and forming a continuous conductive network. The best MCG produced has a high specific surface area (1173.5 m² g⁻¹) and a rich mesoporous structure. As a cathode material, it has a specific capacity of 77.2 mAh g⁻¹ at a current density of 0.05 A g⁻¹ and exceptional cycling stability, with 99.9% capacity retention after 10 000 cycles. A full LIC assembled with an MCG cathode and soft carbon anode achieved a high energy density of 127.3 Wh kg⁻¹ and retained 84.2% of its capacity after 5000 cycles, highlighting its potential for advanced energy storage applications.

Key words: Activated carbon; Graphene; Self-propagating reaction; Lithium-ion capacitor



1 Introduction

In recent decades, the escalating global demand for energy and the push for sustainable development have intensified research into energy storage technologies^[1–6]. Lithium-ion batteries (LIBs) and supercapacitors (SCs) are currently the dominant energy storage devices for portable electronics, electric vehicles, and renewable energy grids. LIBs are renowned for their high energy density (150–300 Wh kg⁻¹) but they suffer from low power density (<1000 W kg⁻¹) and limited cycle life due to volume expansion and struc-

tural degradation during intercalation^[7–12]. Conversely, SCs offer high power density (>5000 W kg⁻¹) and long cycle life (>10⁵ cycles) via surface adsorption mechanisms but are limited by low energy density (<10 Wh kg⁻¹)^[13–18]. To address this challenge, lithium-ion capacitors (LICs) have emerged as a hybrid

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energy storage system that combines the Faradaic intercalation mechanism of LIB anodes with the non-Faradaic adsorption mechanism of SC cathodes^[19–25]. However, the overall energy density and rate capability of LICs are often limited by the cathode material, typically activated carbon (AC), which suffers from relatively low specific capacitance and poor electronic conductivity^[26–33]. Therefore, the development of cathode materials with high specific capacitance, excellent conductivity, and robust electrochemical stability is of critical importance.

Graphene, owing to its unique two-dimensional (2D) structure, high theoretical surface area ($2630 \text{ m}^2 \text{ g}^{-1}$), and outstanding electronic conductivity, has attracted considerable attention as a promising candidate for enhancing the performance of LIC cathodes^[34–38]. For example, recent studies have demonstrated that incorporating graphene into AC can effectively overcome its intrinsic limitations. Hu et al.^[39] showed that adding graphene into commercial AC can establish a conductive network, reduces interfacial resistance, optimizes current distribution, and enhances the electrochemical performance of the cathode. An et al.^[40] reported that incorporating graphene into commercial AC by a self-propagating high-temperature synthesis strategy significantly boosted the electrical conductivity from 389 S m^{-1} (pristine AC) to 2941 S m^{-1} , thereby facilitating faster electron transport. Furthermore, these findings underscore graphene's critical importance in upgrading traditional porous carbon into high-performance cathodes for advanced LICs. However, the use of commercial AC presents inherent limitations, as its specific surface area and pore structure are difficult to precisely regulate. Consequently, the fabrication of activated carbon/graphene composites with well-controlled structures remains a significant challenge for the development of advanced LIC cathodes.

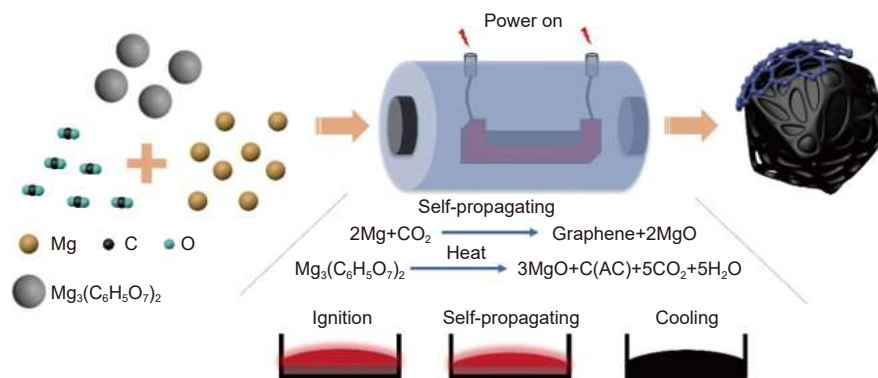
Herein, we proposed an one-pot self-propagating high-temperature synthesis (SHS) strategy for the in-situ formation of activated carbon/graphene composites. Using magnesium powder and magnesium citrate as precursors in a CO_2 atmosphere, Mg reacts with CO_2 to generate graphene via a magnesiothermic

reduction process^[41–43]. Meanwhile, the intense heat released from the self-propagating reaction rapidly carbonizes magnesium citrate into activated carbon. This process enables the in-situ formation of activated carbon/graphene composites, in which graphene sheets effectively suppress agglomeration and enhance electrical conductivity, thereby leading to improved electrochemical performance. Specifically, the optimized activated carbon/graphene (MCG) composite possesses a high specific surface area of $1173.5 \text{ m}^2 \text{ g}^{-1}$ with a rich mesoporous structure. As a cathode material, it delivers a specific capacity of 77.2 mAh g^{-1} at a current density of 0.05 A g^{-1} and exhibits remarkable cycling stability with 99.9% retention after 10 000 cycles. Furthermore, a full lithium-ion capacitor assembled with the MCG cathode and a soft carbon anode achieves a high energy density of 127.3 Wh kg^{-1} , maintaining 84.2% of its capacity after 5000 cycles.

2 Experiment

2.1 Preparation of MCG composites

The MCG composites were synthesized using magnesium powder (Mg) and magnesium citrate ($\text{Mg}_3(\text{C}_6\text{H}_5\text{O}_7)_2$) by the SHS method (Scheme 1). Mg and $\text{Mg}_3(\text{C}_6\text{H}_5\text{O}_7)_2$ powders were weighed according to specific mass ratios. The powders were premixed in a beaker by shaking and then thoroughly homogenized using a high-speed pulverizer. The mixture was placed in a graphite boat with a buried nickel-chromium wire and then transferred into a sealed 20 L reactor. Before ignition, the reactor was purged with CO_2 several times to remove residual air and then filled with CO_2 to 4 MPa. The reaction was conducted under a static CO_2 atmosphere without continuous gas flow. The SHS reaction was triggered by electrically heating the wire and was completed within a few seconds. Under the optimized precursor ratios from 1 : 1 to 1 : 5, the SHS process was reproducible, showing similar ignition behavior and product formation in repeated experiments. However, when the $\text{Mg}/\text{Mg}_3(\text{C}_6\text{H}_5\text{O}_7)_2$ mass ratio exceeded 1 : 6, the reaction could not be self-sustained because of insufficient heat generation. And then the product was



Scheme 1 Sample preparation diagram

washed with dilute sulfuric acid (2 mol L^{-1}) to remove MgO and other impurities, vacuum-filtered until neutral, and freeze-dried.

Four samples were prepared with Mg to $\text{Mg}_3(\text{C}_6\text{H}_5\text{O}_7)_2$ mass ratios of 1 : 1, 1 : 2, 1 : 3 and 1 : 5 (total mass 70 g), designated as MCG-1, MCG-2, MCG-3 and MCG-5, respectively. Ratios above 1 : 6 failed to sustain the self-propagating reaction because of insufficient heat generation.

2.2 Morphology and structure characterization

X-ray diffraction (XRD) patterns of MCG were recorded using a Bruker D8 Advance diffractometer at $10^\circ \text{ min}^{-1}$. The crystalline structures of MCG were investigated by Raman spectroscopy (LabRAM HR-800). Nitrogen adsorption-desorption measurements were carried out to investigate the pore structures using a Micromeritics ASAP 2020 HD analyzer. Based on the results, the SSA and pore size distribution were calculated according to Brunauer-Emmett-Teller (BET) model and Barrett-Joyner-Halenda (BJH) model as well as density functional theory (DFT), respectively. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were performed to investigate the morphology of MCG samples using a Hitachi S4800 and JEOL JSM-2010, respectively. The surface chemical states were characterized by X-ray photoelectron spectroscopy (XPS, ESCALAB 250XI).

2.3 Assembly of half-cells and LIC full-cells

The electrochemical performance of MCG and soft carbon electrodes was assessed using 2032-type coin cells. Specifically, the active materials (MCG or soft carbon) were thoroughly mixed with

polyvinylidene fluoride (PVDF) and Super C45 carbon black at a mass ratio of 7 : 2 : 1. A moderate amount of N-methylpyrrolidone (NMP) was added to the mixed powder and the mixture was ground to form a homogeneous slurry. This slurry was then coated onto copper foil (for soft carbon) or aluminum foil (for MCG), dried in a vacuum oven at 100°C for 10 h, and punched into discs with a diameter of 11 mm to produce both the anode and cathode electrodes. The mass loading of soft carbon anode was maintained at $1 \text{ mg} \cdot \text{cm}^{-2}$, while that for MCG cathode varied from 2 to $3 \text{ mg} \cdot \text{cm}^{-2}$. For half-cell tests, the soft carbon electrodes were used as working electrodes to evaluate their anode performance with Li foil as the counter electrode in the voltage range of 0.01–3.0 V. Separately, the MCG electrodes were used as working electrodes to evaluate their cathode performance with Li foil as the counter electrode in the potential range of 2.0–4.2 V. Prior to assembling the LIC full cells, the soft carbon electrodes were prelithiated by cycling 5 times between 0.01 and 3.0 V at a current density of 0.1 A g^{-1} . After the initial 5 activation cycles, the soft carbon electrodes were finally discharged to 0.01 V vs. Li/Li^+ to ensure a lithiated state. The prelithiated soft carbon electrodes were then disassembled and used as the anodes for assembling LIC full cells with MCG cathodes. The specific capacity, energy density, and power density of the LIC full cells were calculated based on the total mass of active materials in both the MCG cathode and soft carbon anode, excluding the masses of current collectors, separator, electrolyte, binder, conductive additive and cell package.

3 Results and discussion

3.1 Structural and morphological analyses

First, a systematic investigation of the structural characteristics of MCG-1, MCG-2, MCG-3 and MCG-5 was conducted. Fig. 1a presents the XRD patterns of these samples. As shown in the patterns, MCG-1 and MCG-2 exhibit distinct characteristic peaks corresponding to MgO, indicating the presence of MgO impurities within these materials^[41]. Conversely, as the proportion of magnesium citrate increases, neither MCG-3 nor MCG-5 displays significant characteristic peaks of MgO. This phenomenon can be attributed to the origin of the primary inorganic by-products in this system, which arise from 2 sources: (1) MgO generated via the magnesium-thermal reaction and (2) MgO derived from the pyrolysis/carbonization of magnesium citrate. The magnesium-thermal reaction is exothermic, whereas the cracking of magnesium citrate is endothermic and accompanied by gas evolution. The coupling of these two processes promotes the preferential nucleation of

carbon radicals on the surface or within the nascent MgO derived from magnesium citrate, thereby forming an “open-type” graphene-MgO contact interface. Simultaneously, the gases generated during cracking facilitate the construction of interconnected mesopores and inhibit the formation of dense carbon films, enhancing both the accessibility of the acid solution and the dissolution efficiency^[44–45]. Consequently, the MgO is no longer encapsulated by a dense carbon layer that hinders dissolution, resulting in a more thorough acid-washing process.

Furthermore, a diffraction peak corresponding to the (100) plane of graphene sheets is detected at approximately 42° . Notably, as the proportion of magnesium citrate in the feedstock increased, the intensity of this (100) diffraction peak gradually diminished, indicating a reduction in the degree of graphitization within the activated carbon/graphene composites^[46]. This structural evolution is accompanied by the formation of abundant sp^3 -hybridized structures within the products, which are attributed to edge defects and porous architectures in the samples^[47].

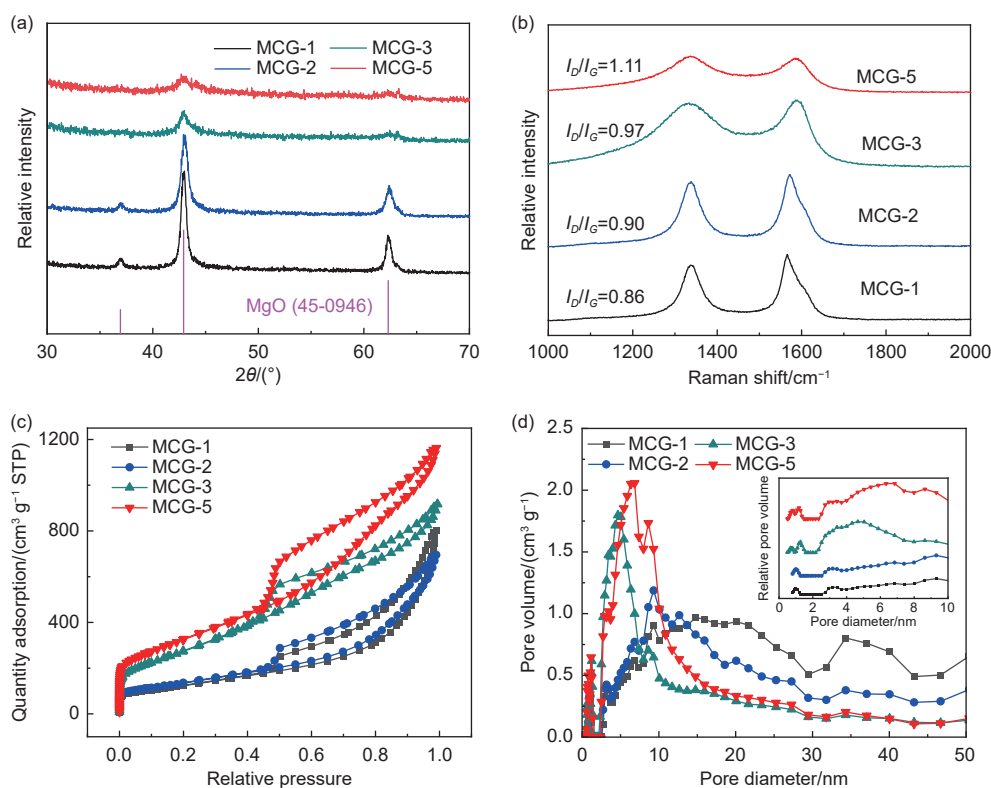


Fig. 1 (a) XRD patterns. (b) Raman diagram. (c) Low-temperature N₂ adsorption-desorption isotherm diagram, and (d) pore size distribution of MCG-1, MCG-2, MCG-3 and MCG-5

Moreover, the Raman spectroscopy results (Fig. 1b) reveal that the spectra for MCG-1, MCG-2, MCG-3 and MCG-5 all exhibit characteristic *D* and *G* bands located at approximately 1332 and 1585 cm^{-1} , respectively. The *D* band corresponds to disordered carbon atoms (specifically, sp^3 -type disordered carbon), while the *G* band represents graphitic carbon. The calculated I_D/I_G intensity ratios for MCG-1, MCG-2, MCG-3 and MCG-5 are 0.86, 0.90, 0.97 and 1.11, respectively. These values indicate that a higher proportion of magnesium citrate in the feedstock leads to an increased degree of disorder in the composites^[48], which is consistent with the XRD results. Although graphene is introduced into the composite, the decreased graphitization degree is not contradictory because XRD and Raman results represent the averaged structure of the whole MCG composite. With increasing magnesium citrate content, more defect-rich porous activated carbon is formed, which dilutes the graphitic signal and increases structural disorder. Meanwhile, the in-situ formed graphene mainly consists of small and wrinkled nanosheets with abundant edge defects, contributing to the increased I_D/I_G ratio while still providing an effective conductive network.

The specific surface area and pore size distribution of MCG-1, MCG-2, MCG-3 and MCG-5 were characterized via nitrogen adsorption-desorption isotherms, as illustrated in Fig. 1c-d. Calculated using the BET theory, the specific surface areas of MCG-1, MCG-2, MCG-3 and MCG-5 are determined to be 448.7, 488.0, 1030.9 and 1173.5 $\text{m}^2 \text{g}^{-1}$, respectively. According to the IUPAC classification, the samples exhibit typical Type IV isotherms featuring distinct H4-type hysteresis loops in the relative pressure (p/p_0) range of 0.5–1.0, which confirms the presence of

abundant mesopores (2–50 nm) within the MCG samples. Further analysis of the pore size distribution based on the DFT method reveals that the mesopore sizes in the MCG samples are concentrated between 2 and 20 nm (Table 1). Notably, as the proportion of magnesium citrate increases, the average pore diameter of the composite decreases. These abundant mesopores shorten ion diffusion pathways, mitigate concentration polarization, and enhance electrolyte wettability. When combined with a high specific surface area, these features significantly increase the number of active sites for charge storage and the interfacial reaction area. Consequently, this structural synergy enhances ion transport and rapid charge-discharge performance for the cathodes of lithium-ion capacitor.

The microstructures of MCG-1, MCG-2, MCG-3 and MCG-5 were examined by SEM. As shown in Fig. 2, all MCG samples consist of irregular carbonaceous particles, while their microstructures evolve markedly with increasing magnesium citrate content. MCG-1 (Fig. 2a-b) exhibits relatively dense agglomerates with limited pore development, although some wrinkled layered features can already be observed at higher magnification. For MCG-2 (Fig. 2c-d), obvious macropores and a more open carbon framework emerge, indicating the gradual transition from compact blocks to porous architectures. When the magnesium citrate ratio is further increased, MCG-3 (Fig. 2e-f) and especially MCG-5 (Fig. 2g-h) display much looser and thinner sheet-like structures with abundant folds and interconnected pores. The high-magnification image of MCG-5 clearly shows a crumpled and cross-linked nanosheet network, which is characteristic of graphene nanosheets anchored on a porous activated carbon matrix. Such a structural evolution suggests that the increased amount of mag-

Table 1 The pore structure parameters of MCG

Sample	$S_{\text{BET}}^{(a)}$ /($\text{m}^2 \text{g}^{-1}$)	$V_{\text{Total}}^{(b)}$ /($\text{cm}^3 \text{g}^{-1}$)	$V_{\text{micro}}^{(c)}$ /($\text{cm}^3 \text{g}^{-1}$)	$V_{\text{meso}}^{(d)}$ /($\text{cm}^3 \text{g}^{-1}$)	$D_{\text{ave}}^{(e)}$ /(nm)
MCG-1	448.7	1.0705	0.0515	0.8359	11.16
MCG-2	488.0	0.9153	0.0620	0.7578	8.82
MCG-3	1030.9	1.0592	0.1091	0.9244	6.13
MCG-5	1173.5	1.2770	0.1470	1.1107	5.51

Notes: (a): Specific surface area. (b): Total pore volume. (c): Micropore volume. (d): Mesopore volume. (e): Average pore size.

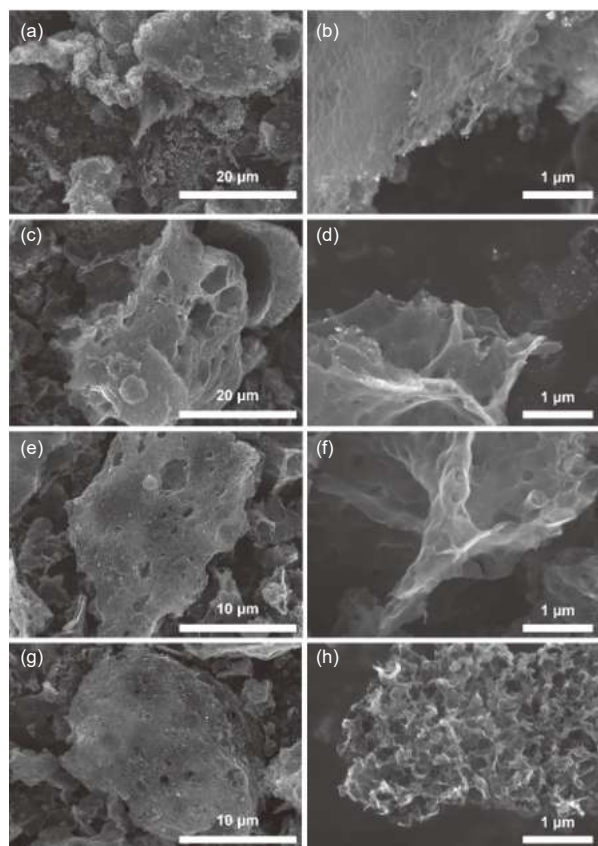


Fig. 2 SEM images: (a-b) MCG-1, (c-d) MCG-2, (e-f) MCG-3 and (g-h) MCG-5

nesium citrate promotes gas-release-induced pore generation and suppresses structural densification during the SHS process, leading to a hierarchically porous and conductive graphene/activated carbon composite. This unique morphology is expected to facilitate electrolyte penetration, expose more active sites, and accelerate electron/ion transport.

From the perspective of the growth mechanism, magnesium citrate serves not only as a carbon source during the self-propagating high-temperature reaction but also acts as an inorganic template and expansion agent. As the reaction temperature rises, the thermal decomposition of magnesium citrate releases gases such as CO_2 , which generate expansive forces within the material, thereby facilitating the formation of a porous structure. With an increased proportion of magnesium citrate, the volume of released gas increases, causing the pore formation process to experience stronger expansion effects. This results in reduced particle size and a more uniform pore distribution biased toward the nanoscale. Furthermore, the

formation of this pore structure could be attributed to the spatial confinement effect imposed by the residual magnesium oxide (MgO) resulting from the decomposition of magnesium citrate on the carbon growth process. Additionally, the incorporation of magnesium powder further promotes graphene formation; its reducing properties likely assist in the layered stacking of carbon atoms, leading to the formation of graphene crystallites.

Fig. 3 presents the TEM images of the MCG-5 material. As shown in Fig. 3a-b, the small graphene nanosheets in MCG-5 exhibit surfaces characterized by abundant wrinkled structures. The presence of these wrinkles is not only a result of the spontaneous folding and scrolling of graphene nanosheets at the nanoscale but is also likely closely associated with the dynamic reaction environment induced by gas evolution during preparation. These wrinkled structures increase the specific surface area of the material and provide additional active sites, which are conducive to enhancing the electrochemical performance of the material in energy storage devices. In the selected-area electron diffraction (SAED) pattern (Fig. 3c), clear and bright diffraction spots are observed. These spots confirm the crystalline structure of the graphene nanosheets within the MCG material, indicating a highly ordered lattice arrangement of the graphene layers and further validating the efficacy of the self-propagating high-temperature reaction. EDS elemental mapping (Fig. 3d-f) further reveals the compositional characteristics of the MCG material. Carbon and oxygen elements are uniformly distributed across the material surface, suggesting the presence of oxygen-containing functional groups. These groups likely originate from the pyrolysis products of magnesium citrate or surface oxidation reactions. The introduction of oxygen-containing functional groups not only improves the wettability of the material to the electrolyte of the material but may also significantly enhance its energy storage performance through pseudocapacitive effects.

X-ray photoelectron spectroscopy (XPS) analyses were conducted to investigate the chemical com-

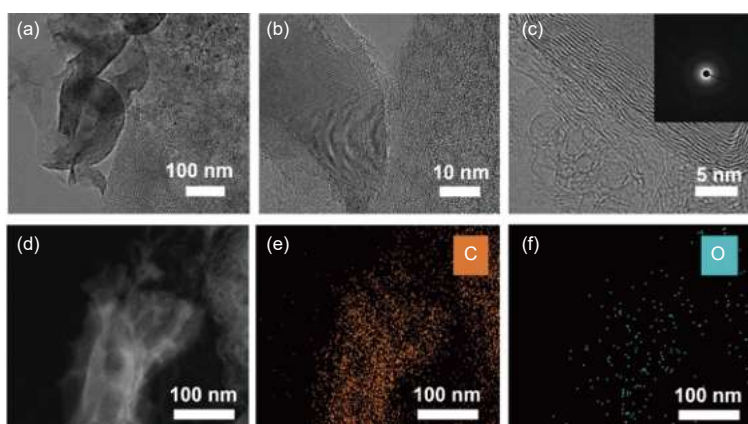


Fig. 3 TEM images of MCG-5: (a-b) TEM images, (c) High-resolution TEM image (inset shows SAED image of graphene in MCG-5), and (d-f) High-angle annular dark-field (HAADF) TEM image and element distribution maps

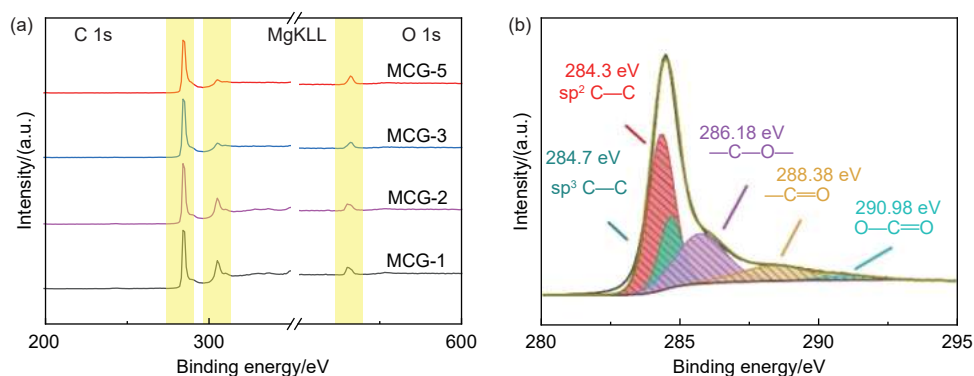


Fig. 4 XPS analysis: (a) Full spectrum of MCG. (b) C 1s spectrum of MCG-5

position and atomic bonding states of the MCG materials (Fig. 4a-b). According to the XPS survey spectra (Fig. 4a), in addition to the C and O elements, Mg Auger peaks are detected in MCG-1 and MCG-2. This indicates the presence of Mg impurities, which is consistent with the XRD results. Fig. 4b presents the high-resolution C 1s XPS spectrum of MCG-5. The peaks located at binding energies of 284.7 and 284.3 correspond to the dominant sp^3 C—C and sp^2 C—C bonds, respectively. Furthermore, peaks observed at 286.18, 288.38 and 290.98 eV are attributed to —C—O—, —C=O and O—C=O species, respectively^[49].

3.2 Electrochemical performance of the MCG cathode

The electrochemical performance of the MCG cathode was evaluated in a half-cell configuration (Fig. 5). Within the voltage window of 2.0–4.2 V (vs. Li/Li^+) and at scan rates of 5, 10 and 20 mV s^{-1} , the CV profiles exhibit typical quasi-rectangular shapes

without discernible redox peaks. This result indicates that the energy storage mechanism of the MCG-5 cathode is predominantly governed by the rapid physical adsorption–desorption of electrolyte anions at the electrode surface, which is characteristic of an electric double-layer capacitance (EDLC) process (Fig. 5a). As the scan rate increases, the response current scales up proportionally, and the curve retains a well-defined quasi-rectangular geometry without significant distortion even at the relatively high rate of 20 mV s^{-1} , demonstrating excellent electrochemical reversibility, low internal resistance, and outstanding rate capability. This superior kinetic performance is attributed to the unique material architecture of MCG-5. Its high specific surface area of 1173.5 $\text{m}^2 \text{g}^{-1}$ affords abundant active sites for ion adsorption. The mesoporous network with an average pore size of 5.51 nm effectively shortens ion diffusion pathways and alleviates concentration polarization at high scan rates. In addition, the two-dimensional graphene intro-

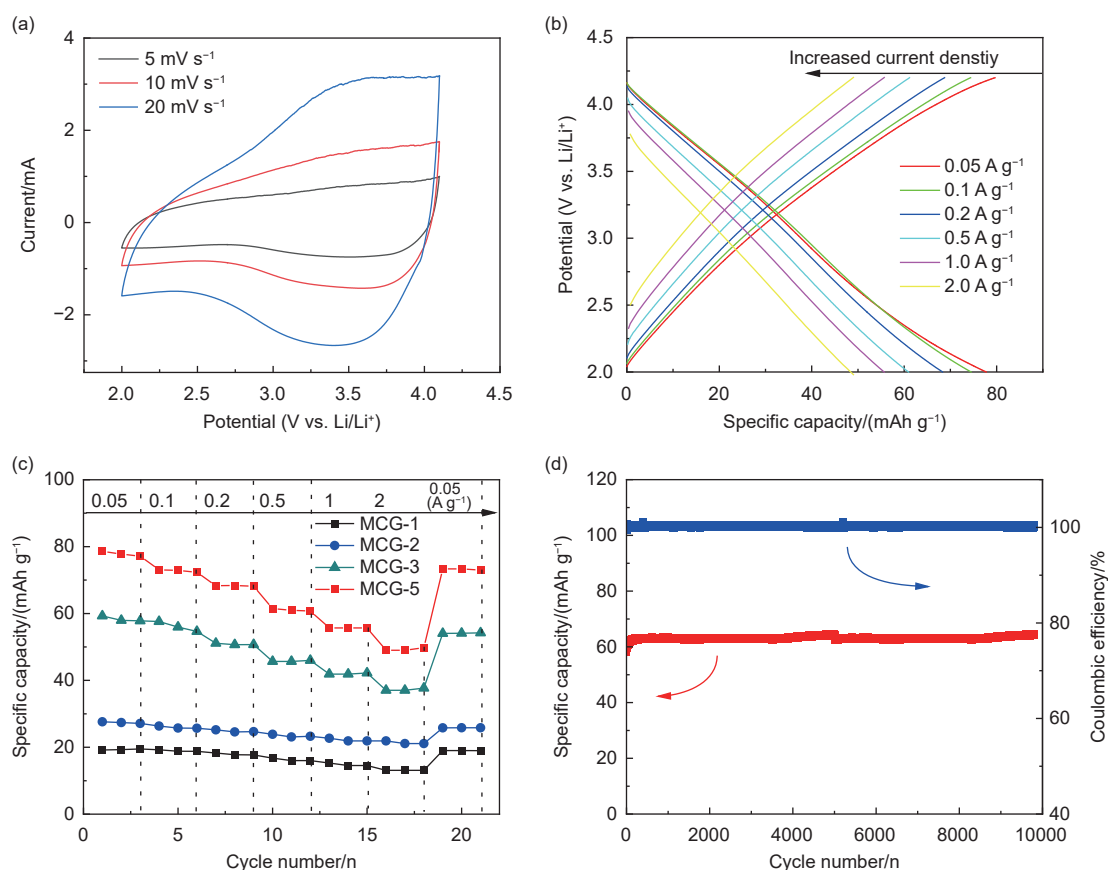


Fig. 5 MCG cathode: (a) CV curve, (b) GCD curve, (c) rate performance graphs and (d) MCG-5 cyclic performance at a current density of 1 A g^{-1}

duced via the self-propagating high-temperature synthesis method constructs a continuous and efficient electronic conduction network on the surface of the activated carbon, thereby substantially reducing interfacial resistance.

As shown in Fig. 5b, the GCD curves of MCG exhibit symmetric linear traces at different current densities. This characteristic isosceles triangle shape further corroborates the conclusions drawn from the CV curves, indicating that the energy storage process of the MCG-5 cathode is governed by a highly reversible, ideal EDLC mechanism reliant on the rapid physical adsorption and desorption of anions at the electrode surface. The initial coulombic efficiency of the MCG-5 cathode was calculated to be 98.2%, indicating the highly reversible anion adsorption/desorption behavior of the porous carbon/graphene composite. In terms of capacity, MCG-5 delivers an excellent specific discharge capacity of 77.2 mAh g^{-1} at a low current density of 0.05 A g^{-1} . As the current density increases to 2 A g^{-1} , polarization rises modestly

and leads to some capacity attenuation, yet the charge-discharge curves retain ideal symmetry and linearity.

As shown in Fig. 5c, at current densities of 0.05, 0.1, 0.2, 0.5, 1 and 2 A g^{-1} , MCG-5 delivers specific capacities of 77.2, 72.8, 68.4, 60.9, 55.7 and 49.2 mAh g^{-1} , respectively, and its specific capacity at 0.05 A g^{-1} exceeds that of MCG-2 (27.6 mAh g^{-1}) and MCG-1 (19.2 mAh g^{-1}) by 180% and 302%, respectively, clearly demonstrating that increasing the specific surface area and judiciously tailoring the pore size distribution can markedly enhance the capacity and rate performance of carbon-based electrode materials. The capacity decrease at high current densities can be mainly attributed to the shortened charge/discharge time, which prevents electrolyte ions from fully accessing the internal micropores and deeper active sites. In addition, increased ion diffusion resistance and electrode polarization under high-rate conditions further limit charge storage. Nevertheless, MCG-5 still delivers a capacity of 49.2 mAh g^{-1} at 2 A g^{-1} , be-

nefiting from its abundant mesoporous channels and graphene nanosheets conductive network, which promote rapid ion/electron transport and effectively alleviate high-rate polarization. The cycling performance of MCG-5 (Fig. 5d), indicates that after 10 000 cycles at a current density of 1 A g^{-1} , the electrode retained a specific capacity of 62 mAh g^{-1} with a capacity retention of 99.9%. Throughout the entire cycling process, the Coulombic efficiency of MCG-5 consistently remained above 99%, evidencing excellent electrochemical cycling stability.

3.3 Electrochemical performance of the soft carbon anode

As shown in Fig. 6a, the CV measurements of the soft carbon anode were conducted over a potential range of 0.01–3 V at scan rates ranging from 0.2 to 1.0 mV s^{-1} . The CV curves indicate that Li^+ intercalation occurs primarily between 0 and 0.2 V, demonstrating the electrode material's strong capability for lithium storage, while a pronounced oxidation peak appears around 0.5 V, corresponding to the deintercalation of Li^+ from the electrode. Notably, the positions of the oxidation and reduction peaks remain essentially unchanged with varying scan rates, indicating a highly stable electrochemical reaction mechanism for the soft-carbon material. In addition, the first three GCD curves (Fig. 6b) show that the initial coulombic efficiency of the soft carbon anode is 73.7%. The initial irreversible capacity loss mainly originates from solid electrolyte interphas (SEI) formation and electrolyte decomposition during the first lithiation process. At current densities of 0.05, 0.1, 0.2, 0.5, 1 and 2 A g^{-1} , the corresponding discharge specific capacities of the soft-carbon anode are recorded as 232.7, 200.8, 179.3, 151.1, 128.1 and 98.9 mAh g^{-1} (Fig. 6c). After 1000 GCD cycles at a current density of 0.1 A g^{-1} , the soft-carbon anode still retained a specific capacity of 160.1 mAh g^{-1} (Fig. 6d).

3.4 Electrochemical performance of lithium-ion capacitor

A full LIC was assembled using MCG-5 as the cathode and soft carbon as the anode. The optimal

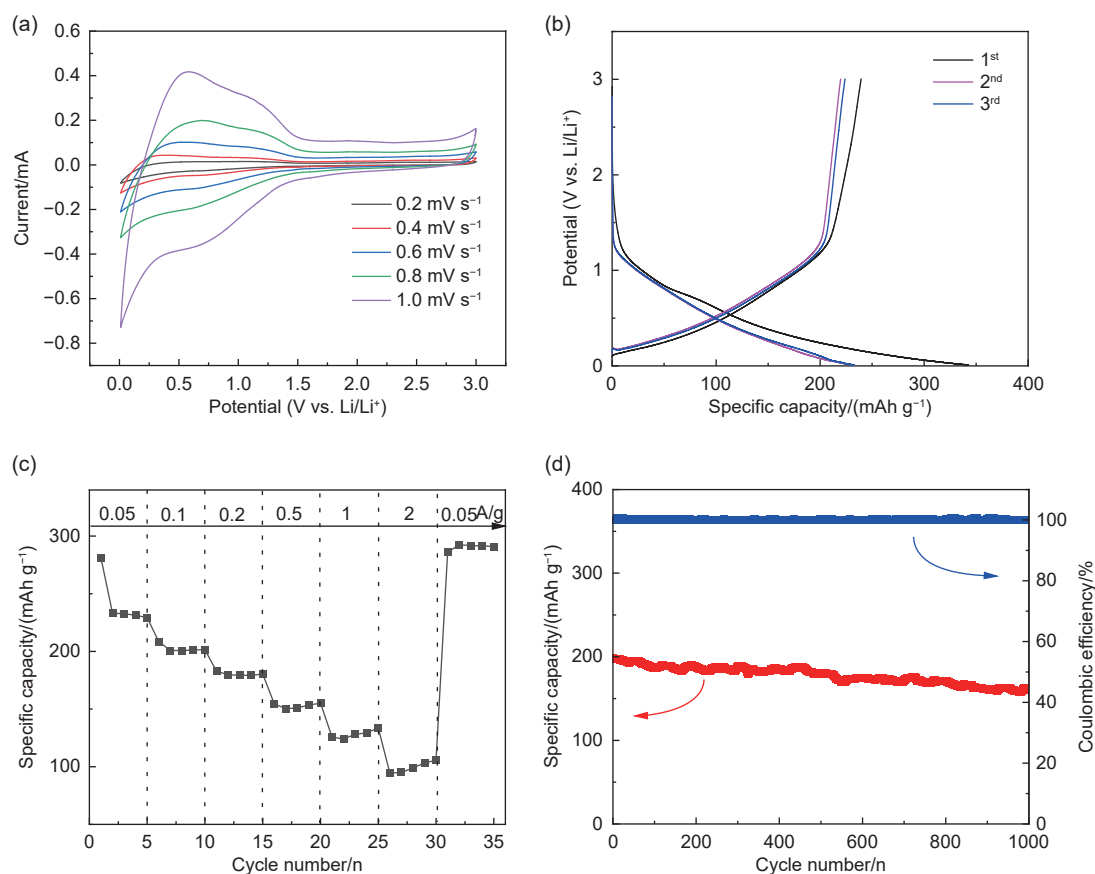


Fig. 6 Soft carbon anode: (a) CV curve, (b) GCD curve, (c) rate performance graphs, and (d) cyclic performance at a current density of 1 A g^{-1}

mass ratio of anode to cathode was determined to be 1 : 2. The device showed quasi-rectangular CV curves even at a high scan rate of 50 mV s^{-1} . Discharge specific capacities ranged from 46.3 mAh g^{-1} (0.05 A g^{-1}) to 30 mAh g^{-1} (2 A g^{-1}). The MCG-5//soft carbon LIC achieved a high energy density of 127.3 Wh kg^{-1} at a power density of 117.8 W kg^{-1} , which were calculated based on the total mass of active materials in both electrodes. Even at a high power

density of 4.4 kW kg^{-1} , it retained an energy density of 69 Wh kg^{-1} . Besides, the performance of MCG-5//soft carbon LIC is better than those reported, such as AC//soft carbon^[39], AC//MHC^[50], AC@LFP//AC^[51], AC//TiO₂@PCNF-GA^[52], AC-20//C@LTO^[53], AC//TAB-COF^[54], AC//PrGO/LTO80^[55], PAC//PDC^[56], AC//HC100^[57], AC//AlCl₃-GICs 115^[58]. The full cell retains 84.2% of its capacity after 5000 cycles at 1 A g^{-1} (Fig. 7).

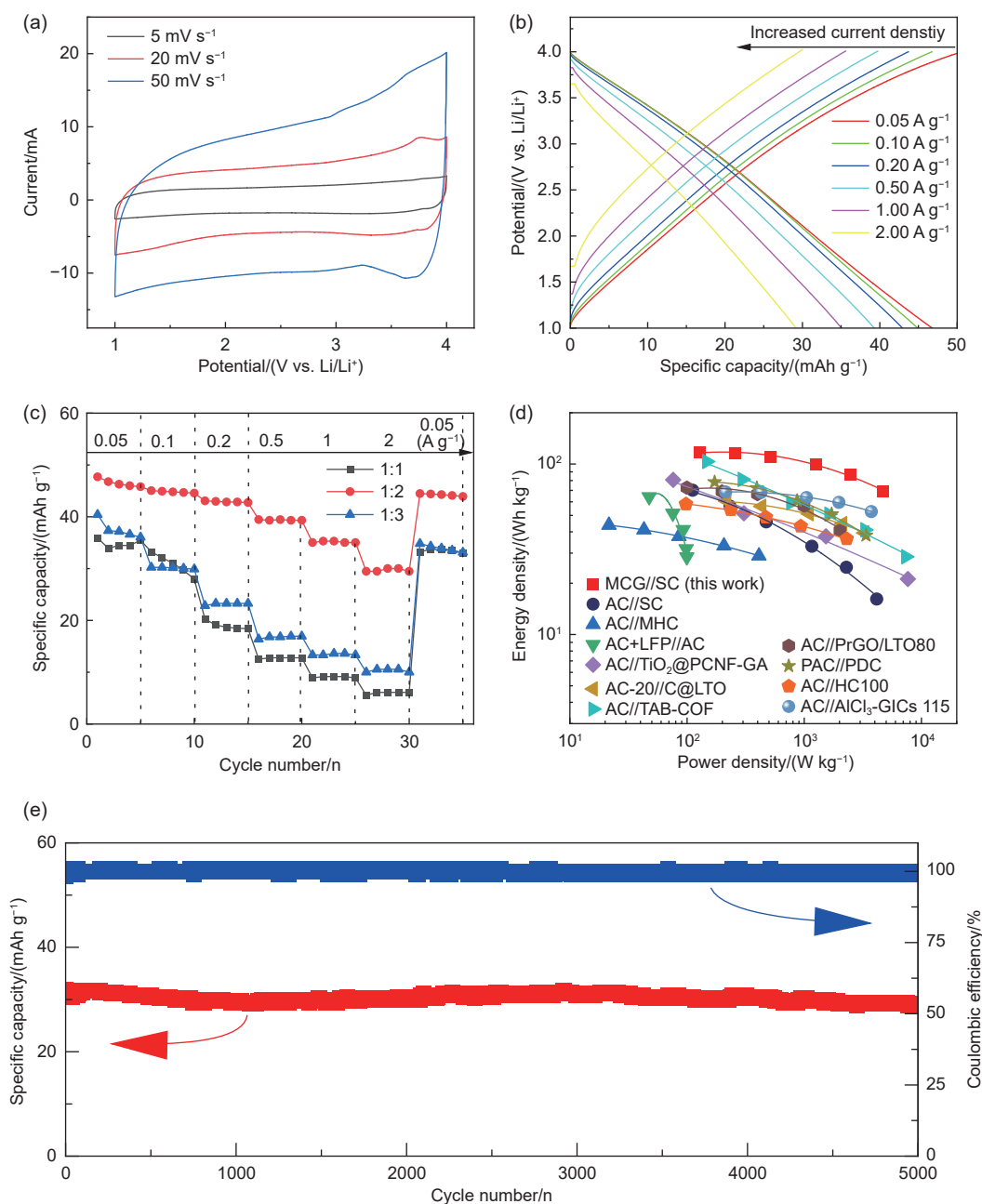


Fig. 7 (a) CV curve. (b) GCD curve. (c) Rate performance plot. (d) Ragone plot. (e) Cyclic performance plot of MCG-5//soft carbon LICs at a current density of 1 A g^{-1}

4 Conclusion

In summary, an activated carbon/graphene composite was successfully prepared by a rapid in-situ self-propagating high-temperature synthesis strategy under a CO₂ atmosphere. The optimized MCG-5 sample exhibits a high specific surface area of 1173.5 m² g⁻¹, abundant mesoporosity, and a conductive graphene nanosheet network. Benefiting from this structure, the MCG-5 cathode delivers a high specific capacity of 77.2 mAh g⁻¹ at 0.05 A g⁻¹ and retains 99.9% of its capacity after 10 000 GCD cycles at 1 A g⁻¹. When assembled with a prelithiated soft carbon anode, the MCG-5//soft carbon LIC achieves a maximum energy density of 127.3 Wh kg⁻¹ at 117.8 W kg⁻¹ and maintains 84.2% capacity retention after 5000 cycles. These results demonstrate that the SHS strategy offers a rapid and effective route for constructing graphene-integrated porous carbon cathodes with high capacity, fast charge-storage kinetics, and long-term stability, showing promising potential for practical high-performance lithium-ion capacitors.

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