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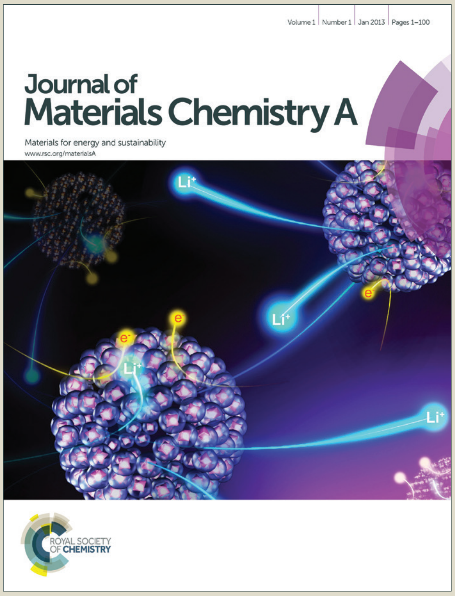

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ARTICLE TYPE

Microwave-assisted production of giant graphene sheets for high performance energy storage applications

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A simple, low-cost and energy-effective method has been developed in this work to fabricate giant graphene sheets by double microwave assisted exfoliations of expandable graphite. The graphene sheets, which are large (10-100 microns) in size and thin (1-10 nm) in thickness, manifests superior electrochemical performance for energy storage, achieving 221 F g^{-1} capacitance after 5000 cycles at 10 mV s^{-1} in a symmetric capacitor measurement and 400 mAh g^{-1} in a coin-type Li-ion battery after 300 cycles at 0.5 A g^{-1}.

Graphene is an intriguing single-atom thick, two-dimensional carbon material whose outstanding properties make it very promising in various applications, such as nanoelectronics[1], biological medicine, electrocatalyst, lithium ion battery[2-9] and supercapacitor[10-13], etc. Graphene can be prepared via various different methods, such as micromechanical cleavage[14], modified Hummers method[15], electrochemical exfoliation[16] and chemical vapor deposition[17], etc. However, Mechanical cleavage of highly ordered pyrolytic graphite or solution-based exfoliation often results in a relatively small size of graphene while epitaxial growth on metal substrates by CVD methods is constrained by its high cost and lack of controllability for forming homogenous large-area films. Hummers method is one of the most widely used approaches. But graphene oxide (GO) flakes produced by Hummers method are normally a few micrometres in size[18, 19], and the small size may limit some applications where large size of graphene flakes is needed[20]. Various strategies towards preparing of large-size GO and graphene sheets have been developed.[21, 22] However, most of these approaches are complicated, energy and cost intensive[18-23].

Recently, microwave exfoliation has proved to be an attractive method for graphene preparation.[23-25] Microwave graphene synthesis is normally realized by heating its suspensions in aqueous or organic solutions[26]. In this report, we propose a novel microwave exfoliation approach to fabricate giant graphene by using repeated (or double) microwave radiation method. The initial step started from microwave heating of commercially available expandable graphite (EG), which was followed by the oxidation and intercalation of oxidant and sulfuric acid. Finally, the second microwave irradiation process was applied, leading to effective exfoliation and reduction of graphite oxide. High-quality large-sized (up to 100 microm) thin graphene sheets (few layers only) were obtained. This method has several obvious advantages. First the average size of prepared graphene is 10 times larger than those using conventional Hummer's method[27]. Secondly, our process is time-efficient since no tedious centrifugation, ultrasound, shock and H_2O_2 treatments are needed. Thirdly, the process is cost-effective as the quantity of chemicals used is greatly reduced compared to conventional approaches. Finally our graphene products are high quality with limited structural damages and residual functional groups, showing excellent performance in supercapacitors and lithium-ion batteries. Our process is easily scalable, providing a high potential for industrial applications.

Results and discussion

Figure 1 schematically illustrates the process of microwave-assisted synthesis of giant graphene sheets (MGG). The process starts from exfoliation of commercial expanded graphite (EG) by 10 seconds of microwave irradiation in conventional microwave oven. Upon microwave heating, the volume of EG powders expands dramatically. In the second step the expanded EG is mixed with concentrated sulfuric acid and $KMnO_4$. The optimized reactant ratio of $KMnO_4$ to EG is 2:1, which is much lower than that of conventional Hummers method (6:1) [27] and modified Hummers process (3:1) in which EG is the starting material[28]. No hydrogen peroxide is added in our case because all potassium permanganate have been participated in the reaction. In the final step the oxidized and intercalated EG is heated by microwave radiation for 10 seconds, yielding a black and fluffy graphene powder.

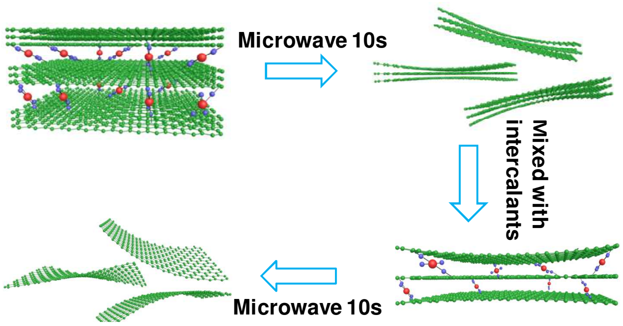

Fig. 1.Schematic of MGG fabrication process

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The graphene sheets obtained by this method are giant in lateral size and very thin in thickness (few layers only). A typical optical image of the giant-graphene-oxide (GGO) sheets is shown in Fig. 2a, which has a lateral size of 50 µm. Some of the GGO sheets are as large as 100 µm while the minimum size in all graphene sheets is about 10 µm. (see Fig. S1a and S2) Our GGO sheets are very thin, showing extremely low optical contrast with respect to the substrate surface. They are in the range between 1 and 12 nm in thickness, mostly a few nm only (see the AFM measurement results in Fig. S1b, which were obtained from 40 randomly selected graphene sheets.). To our best knowledge, there have been no reports on GO sheets with such a large size and thin layer, prepared via conventional Hummers methods[27]. Figure 2b displays a representative AFM image of GGO. A variation of the thickness across the GGO flake is clearly observed, indicating a mean GGO thickness of ~2.15 nm in a large flake with a lateral dimension of ~50 µm. The big size and thin thickness of our graphene sheets may be attributed to the effective exfoliation caused by the first microwave heating. Because large-size graphene sheets tend to restack together[29], we can easily use simple vacuum filtration to obtain a free standing graphene film (see the photo in Fig. 2c inset). The TEM and SEM images of the film in Figs. 2c and 2d exhibit a continuous, wrinkled and transparent sheet, indicating the successful synthesis of reduced graphene after the second microwave irradiation, i.e. MGG. A SEM image of MGG with relatively small magnification is shown in Fig. S3. The size of the exfoliated graphene sheets is really large though the agglomerated and wrinkled microstructure can be observed.

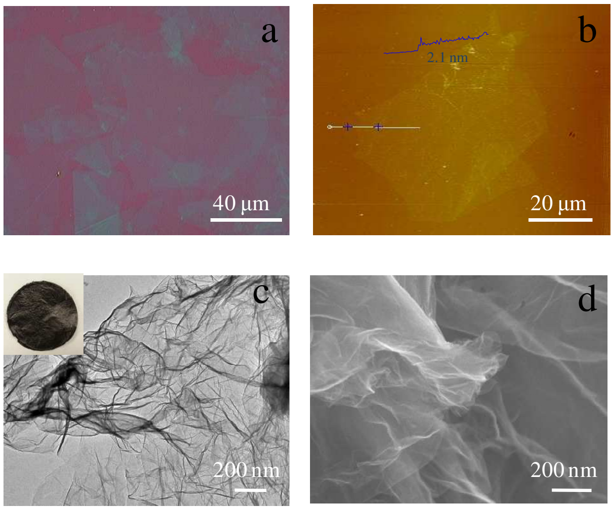

Fig.2 (a) Optical image of the MGG sheets (b) AFM images of MGG (c) High magnification TEM micrograph of MGG (d) High magnification SEM micrograph of MGG

Raman spectroscopy has been widely used to probe the structural properties of graphene in the past decade, such as defects,[30, 31] doping [32], edge[33, 34] stacking[35], and crystalline quality[36]. Here we employed Raman spectroscopy and FTIR to further investigate the quality of the graphene products prepared in our work. Figure 3a compares Raman spectra of EG, GGO and MGG, where EG is the staring material (i.e. commercial expanded graphite), GGO is the intercalation intermediate obtained from the second step, resulting from the oxidation of microwave-treated EG (see supporting information) while MGG is the final product, microwave-giant-graphene. The two most intense bands for each sample are G band at ~1580 cm^{-1} and D band at ~1350 cm^{-1}. The intensity ratio between D and G bands (I_D/I_G), which is normally used to evaluate the crystalline quality of graphene materials, is high (1.16) for the starting material EG. It increases to 1.39 for the GGO sample due to the oxidation and intercalation. But the D band intensity decreases dramatically (I_D/I_G = 0.85) for MGG after the second microwave exfoliation process, suggesting the restoration of sp2-hybridized carbon network via microwave-heating. FTIR spectra in Fig.3b shows remarkably strong peaks at 1109 (epoxy C-O), 1223 (carboxyl OH), 1383 (carboxyl C-O), 1623 (aromatic C=C), 1732 (carbonyl and carboxyl C=O) and 3500 (O-H groups) cm^{-1} respectively[37], for GGO. These oxygen-containing functional groups are characteristic of GO, resulting from the microwave-assisted oxidation and intercalation of EG with concentrated sulfuric acid and $KMnO_4$. These peaks are weak both in EG and MGG spectra, suggesting that GGO can be effectively deoxygenated/reduced by microwave-assisted treatment.[38]

Based on the above observations the repeated microwave treatments play important roles in the synthesis of giant graphene sheets from expended graphite. The first radiation leads to the increase of the interlayer distance as well as the edge opening of graphene sheets in EG, in favour of subsequent solvent intercalation and oxidation by concentrated sulfuric acid and $KMnO_4$. This helps the effective expansion and exfoliation of EG into giant graphene oxide (GGO). More than 90% of the graphene oxide sheets yielded was dispersed stably in water as observed by optical microscope. A video in supporting information showed that a lot of thin-layered, large-area GGO sheets were uniformly displayed on Si substrate everywhere. Significantly lower amount of chemicals (sulphuric acid, $KMnO_4$) were used, as compared to conventional exfoliation without using microwave. Centrifugation process for separating the ultra-thin graphene oxide sheets from the thick ones is not necessary. The second microwave heating can effectively trigger deoxygenation of GGO and heal the defected carbonnetwork, resulting in giant graphene sheets with high quality. This is a facile, high efficiency and low cost method to simultaneously achieve the exfoliation and reduction of graphite oxide. The giant graphene samples were characterized by XPS and four-point probe conductivity measurements, and compared to conventional reduced GO under identical conditions. The results are compared to those of conventional rGO under identical conditions. XPS wide scan spectra of MGG and conventional reduced GO (i.e. rGO) are displayed in Fig.S4 with a higher C/O ratio (2.6) for MGG than 1.7 of conventional rGO. For 4-point probe measurements a GGO and a conventional GO sheet were deposited on SiO_2/Si substrates, reduced by annealing in H_2/Ar at high temperatures (800 ℃) and then cooled to ambient (see SEM image in Fig. 3c). The reduced giant-graphene sheet was 2 nm in thickness. As shown in Figure 3d the reduced giant graphene sample (MGG) is more conductive (~408 S/cm) than conventional rGO (350 S/cm), indicating that the giant graphene samples prepared in our work have fewer defects and thus better conductivity than conventional rGO.

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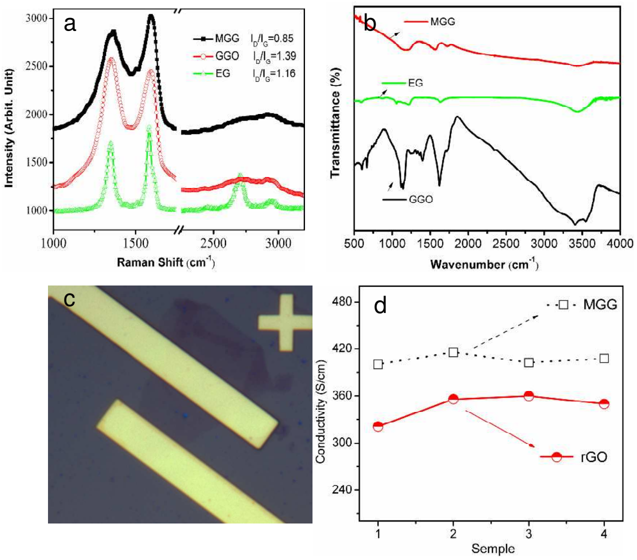

Fig. 3 (a) Raman spectra of EG, GGO and MGG; (b) IR spectra of EG, GGO and MGG. (c) SEM image of reduced giant graphene (MGG) and conventional reduced GO (rGO) on Si substrate for 4-point measurements; (d) Plot of the mean conductivities for MGG and rGO sheets.

The high quality MGG is very suitable for the energy storage applications. In this paper we studied MGG sample as the electrode material both for supercapacitor and Li-ion battery. (see supporting information) The performance of our MGG-based supercapacitor was measured in a two-electrode symmetrical supercapacitor cell. Figure 4a shows the cyclic voltammetry (CV) curves of MGG at scan rates of 10–100 mV/s over the voltage range from 0 to 1 V. All these curves display a rectangular shape, suggesting that the as-prepared MGG exhibits excellent capacitive behavior. A specific capacitance of 221F/g was calculated from the CV curve at a constant rate of 10 mV/s, using formula listed in the supporting information. This value is obviously higher than the value reported elsewhere using similar method[39]. With increasing the charge/discharge rate to 100 mV/s, the specific capacitance still shows good performance (164 F/g). The MGG supercapacitor cell was also measured by galvanostatic charge/discharge curves which give a specific capacitance of up to 189 F g^{-1} at a constant current of 1 A g^{-1}, and a capacitance of 136 F g^{-1} can be obtained even when the supercapacitor is operated at rate of 2A g^{-1}. The charge/discharge curves show symmetrical triangle, which is characteristic of an excellent supercapacitor with good electrical conductivity. Furthermore, the MGG exhibits excellent cyclability. It can retain as high as 96% of its initial capacitance over 5000 cycles even under a rate of 100 mV/s. (see Fig.4c) The slight capacitance fluctuation of the device in the first several hundreds of cycles may be attributed to the change in effective interfacial area between MGG nanostructure and electrolyte with the increase of reaction time.[40] We also measured the impedance of the MGG sample and conventional rGO. As shown in Fig. S5 inset the Nyquist impedance plot of MGG has smaller loop and steeper slope (of nearly 60°) than conventional rGO, indicating lower contact resistance and higher charge transfer for MGG than conventional graphene. The excellent electrochemical properties of MGG can be attributed to the big size of giant graphene sheets. For MGG films, the large area of MGG sheets results in low inter-sheet contact resistance due to a small amount of inter-sheet junctions, which is helpful to increase conductivity of electrode materials[41] and remarkably improve the capacitance characteristics.

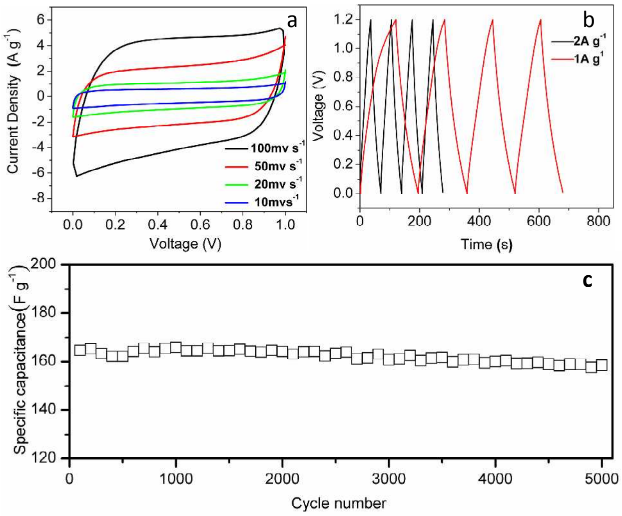

Fig. 4(a) CV curves of MGG for different scan rates. Rectangular shapes indicate the ideal double layer capacitive behaviour. (b) Galvanostatic charge/discharge curves of MGG–based supercapacitor under different constant currents. (c) The specific capacitance values calculated from CV curves at a scan rate of 10 mVs^{-1} during the 5000 cycles

The electrochemical performance of MGG as the Li-ion battery electrode is demonstrated in Fig.5. The initial 3 discharge and charge curves of MGG are displayed in Fig.6a at current density 100 mA g^{-1} in the voltage window of 0.005-3.0 V vs.Li^+/Li. The MGG delivers 1383 and 775 mAh g^{-1} capacities for the first discharge and charge, respectively, corresponding to an initial irreversible capacity loss of 680 mAh g^{-1}. The big initial irreversible loss of MGG can be mainly ascribed to the formation of the solid electrolyte interface due to the high specific surface area of graphene. The second and third cycles show minor difference between charge and discharge, corresponding to 100% columbic efficiency. With an increase in the cycle number, the reversible capacity decreases slowly from 829 mAh g^{-1} in the 2nd cycle to 641 mAh g^{-1} at the end of 80th cycle at current of 100 mA g^{-1}. This rate of the capacity decay with respect to the cycle number is reasonably small, similar to that has been normally observed for carbon materials[42]. Moreover, the MGG electrode also exhibits excellent rate performance (Figure 5b). As the current density increases from 0.1, 0.5 to 1 A g^{-1}, the electrode shows good capacity retention, varying from 670 to 480, and 290 mA h g^{-1}, respectively. Remarkably, when the current density returns to 0.5A g^{-1}, the charge capacity maintain at about 490 mA h g^{-1} which is much higher than the theoretical capacity of commercial graphite. After 100 cycles, the capacity of graphene electrodes increases and become more stable as shown in Fig 5b. In order to test the stability under high rate, after 180 cycles we tested the battery at rate of 0.5 A g^{-1} for 300 cycles, the results show that the performance of MGG electrode is excellent, indicating a reversible charge capacity as high as 400 mAh g^{-1} after 300 cycles（see Fig5c）. Compared to the small-sized thermally reduced graphene, the large-sized graphene sheet in MGG not only can reduce the inter-sheets resistance and hence contact resistance of the electrode, but also can enhance the conductivity due to lower structural defects. Both the effects can improve the

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Li ion diffusion and electron movement, reducing polarization of the electrode during the charging and discharging processes, which lead to improved stability and rate performance of the battery.

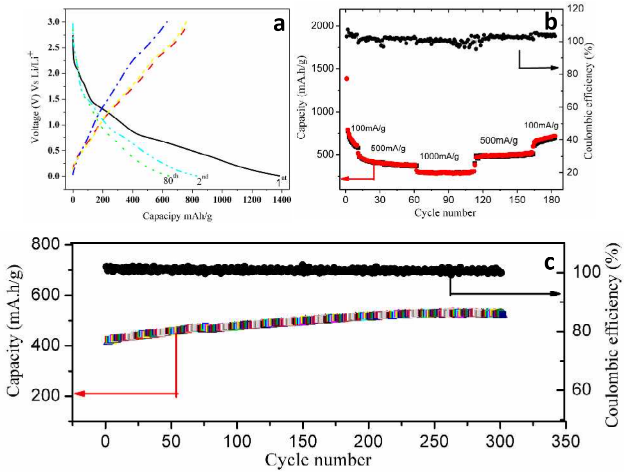

Fig. 5(a) charge and discharge curves of MGG at a current density of 100 mA g^{-1}; (b) capacity of MGG electrode cycled at various current densities: 100, 500 and 1000mA$g^{-1}$and (c) capacity retention of MGG at current density of 500 mA g^{-1} for 300 cycles

Conclusions

We have developed a novel method to prepare the MGG with high crystalline quality by using repeated exfoliations of expandable graphite under microwave irradiation. Compared to the traditional complicated method for preparing the large size graphene, both the sonication and centrifugation are not included in this newly developed technique, and the amount of hazardous chemical oxidants is greatly reduced. The supercapacitor cell using the as-prepared MGG as the electrodes exhibits remarkable performance, and yields a specific capacitance value of 221 F/g in the two-electrode symmetrical system, which is almost two times that of commercial carbon supercapacitor. When used as the Li ion battery electrode, the MGG also exhibits excellent properties in terms of high reversible specific capacity, high rate capability and long cycle life for lithium ion battery. It can be charged and discharged under high current density, achieving a reversible charge capacity as high as 400 mAh g^{-1} at 0.5 A g^{-1} after 300 cycles, which is much higher than that of commercial graphite anode. In short, this newly reported method provides a simple, efficient and low cost route for producing giant graphene materials in large scale. The MGG prepared by this method have a high potential for supercapacitor and Li ion battery applications.

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Notes and references

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† Electronic Supplementary Information (ESI) available: Experimental details,photoimages, additional SEM, AFM and Nyquist plot have been included here.

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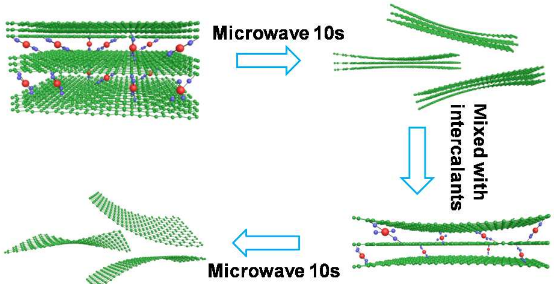

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