Solid-state microwave irradiation synthesis of high quality graphene nanosheets under hydrogen containing atmosphere†

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High quality graphene nanosheets were fabricated within 1 min by solid-state microwave irradiation of a mixture of graphite oxide and graphene nanosheets under a hydrogen atmosphere. The graphene nanosheets in the mixture acted as an effective microwave susceptor under microwave irradiation synthesis, and could provide sufficiently rapid heating for the effective exfoliation of graphite oxide. A hydrogen containing atmosphere played important roles in improving the quality of the graphene nanosheets by increasing the level of reduction and preventing the formation of defects in the graphene nanosheets. The graphene nanosheets thus obtained exhibited a specific surface area of 586 $m^2 g^{-1}$ and an outstanding carbon/oxygen ratio of 18.5.

1. Introduction

Graphene, which is a one-atom thick two-dimensional carbon structure, has attracted considerable attention for potential applications, such as catalyst supports, field effect transistors, sensors, transparent electrodes and electrode materials for energy storage devices owing to its chemical stability, high electrical conductivity and large surface area of over 2600 $m^2 g^{-1}$.[1–8]

Since the first report of the synthesis of graphene by the micromechanical cleavage of graphite, extensive efforts have been made to fabricate single or few layer graphene by epitaxial growth on SiC surfaces, solvothermal synthesis method, liquid phase exfoliation of graphite and chemical reduction or thermal exfoliation of graphite oxide (GO).[1,9–20] Among these, the chemical reduction of GO is used widely for the efficient large scale production of chemically converted graphene nanosheets (GNS).[15–20] However, chemically converted GNS has poor quality compared to pristine graphene due to the presence of oxygen-containing functional groups that are formed during the harsh oxidation of graphite. Therefore, it is essential to improve the quality of GNS with a post treatment, such as thermal annealing.[21,22]

Recently, it was reported that a large quantity of high quality GNS could be fabricated by the thermal exfoliation of GO using a rapid heating process in a preheated conventional furnace or arc discharge.[13,14,23,24] During the thermal exfoliation of GO, oxygen-containing functional groups on GO can decompose into carbon dioxide and the gas evolution leads to expansion between the graphene sheets with efficient exfoliation. Rapid heating is most important for the effective thermal exfoliation of GO. At a heating rate over 2000 °C min^{-1}, the decomposition rate of oxygen-containing functional groups of GO was reported to exceed the diffusion rate of the evolved gases, thereby leading to a sufficiently rapid pressure buildup that can overcome the van der Waals forces between the graphene sheets in GO.[13,14] In addition, the hydrogen containing atmosphere during thermal exfoliation can improve the quality of GNS because hydrogen tends to form H_2O instead of CO_2 from the oxygen-containing functional groups in GO, which prevents the formation of vacancies and defects due to the loss of carbon atoms.[23,24]

Microwaves are used widely as a rapid heating source in the presence of microwave susceptors.[25–34] Carboneous materials, such as graphite, carbon black, carbon nanotube and GNS, can act as effective microwave susceptors.[25–29] Several studies have reported the solid-state microwave irradiation synthesis of exfoliated graphite from graphite intercalation compounds (GICs).[32–34] Exfoliated graphite with a relatively low degree of exfoliation and low specific surface area of ~80 $m^2 g^{-1}$ is obtained when GICs are treated with microwave irradiation. Recently, Ruoff *et al.* reported the solid-state microwave irradiation of GO to prepare GNS with a specific surface area of 463 $m^2 g^{-1}$ and a C/O ratio of 2.75 under an ambient atmosphere using a commercial microwave oven.[35]

This paper reports the fabrication of high quality GNS by solid-state microwave irradiation of a mixture of GO and GNS under a hydrogen atmosphere to improve the quality of GNS. A hydrogen-containing atmosphere plays important roles in improving the quality of GNS during solid-state microwave irradiation synthesis. GNS (10 wt.%) was mixed with GO as a microwave susceptor for more effective rapid heating and exfoliation of GO during solid-state microwave irradiation because GO has a partially disconnected π-system due to various oxygen-containing functional groups, which renders it less effective as a microwave susceptor than GNS under solid-state microwave irradiation.[25–28]

For comparison, GNS was also prepared by a microwave-assisted hydrothermal treatment of GO. Solid-state microwave irradiation synthesis under a hydrogen containing atmosphere

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can produce high quality GNS by promoting the reduction of GNS with the efficient exfoliation from GO.

2. Experimental

Preparation of graphite oxide by Hummers method

The fabrication process of GNS consists of two steps. The first step involves the oxidation of graphite using a modified Hummers method with concentrated sulfuric acid (95%, Samchun Chemical), potassium permanganate (99%, Aldrich) and hydrogen peroxide (35% in water, Junsei) in an ice bath.[17] A mixture containing 4 g of commercial graphite (45 μm nominal particle size, Aldrich) and 100 ml of concentrated sulfuric acid was stirred at room temperature. Subsequently, 14 g of potassium permanganate was slowly added to the graphite/sulfuric acid mixture over a 15 min period and maintained at 0 °C. After stirring for 2 h at 35 °C, a 150 mL excess of distilled water was added to the mixture and stirred for a further 1 h. Hydrogen peroxide (35 wt.% in water, 50 mL) was then added until there was no gas evolution. A yellowish brown solid product was separated by centrifugation and washed repeatedly with distilled water and ethanol until the pH was neutral. The graphite oxide (GO) slurry obtained was dried at 70 °C overnight.

Solid-state microwave irradiation synthesis of graphene nanosheets

For solid-state microwave irradiation synthesis, 90 wt.% of the as-prepared GO powder was mixed with 10 wt.% GNS powder using a ball-miller for 1 h. The resulting GO/GNS mixture was placed into a quartz bottle and covered with a lid inside a glove-box. To examine the effect of the gas atmosphere during the solid-state microwave irradiation synthesis of GNS, the quartz bottle was filled with Ar gas or a H_2/Ar gas mixture (10 : 90 vol.% of H_2/Ar) in the glove box. The assembled quartz bottles were sealed with Paraffin sealing tape for a gas tight seal before being removed from the glove-box. Some quartz bottles of the GO/GNS mixture were assembled in an ambient atmosphere. The GO/GNS mixture in a quartz bottle was then placed inside a microwave oven (Mars 5, CEM) and exposed to microwaves at 1600 W under pulsed irradiation mode for 50 s. (20 s on-time and 5 s off-time for 2 cycles).

For comparison, GNS was also prepared by the microwave-assisted hydrothermal reduction of GO. The as-prepared GO powder (0.1 g) was sonicated in 100 mL of distilled water for 40 min until a homogeneous yellow dispersion was obtained. Microwave vessels (each vessel containing 30 mL of the dispersion) were placed inside a microwave oven and kept at 200 °C under microwave irradiation at a power of 1600 W for 5 to 60 min. The yellowish brown dispersion of GO changed to black after a 5 min reaction time, indicating the reduction of graphite oxide. The black powder was filtered and dried at 70 °C overnight.

Characterization of graphene nanosheets

The morphology of the GO and GNS fabricated by solid-state microwave irradiation synthesis and microwave assisted hydrothermal synthesis was observed by scanning electron microscopy (SEM, Hitachi S-4200). The microstructure of GNS was analyzed by high resolution transmission electron microscopy (HRTEM, JEM-3010, JEOL). The thickness of the GNS was examined by atomic force microscopy (AFM, XE-150, PSIA) in non-contact mode. The surface area of the GNS prepared by different synthesis methods was measured by nitrogen gas absorption using the BET method (ASAP ZOZO, Micromeritics Co.). Elemental analysis (Thermo EA1112, Thermo Electron Corp.) was carried out to determine the quality and reduction level of GNS fabricated by microwave assisted hydrothermal synthesis and solid-state microwave irradiation synthesis. Raman spectroscopy (T64000, Jobin-Yvon) was used to evaluate the structural changes from GO to GNS. Electrical conductivity of graphite, GO and GNS prepared under different conditions was measured using the 2-point probe method combined with the AC impedance method. (See S1 in the ESI†).

3. Results and discussion

Fig. 1 shows photographs of the sequence of the exfoliation process during solid-state microwave irradiation synthesis. In the case of GO powder without GNS, the exfoliation of GO could not be observed under continuous microwave irradiation for as long as 5 min. GO has a partially disconnected π-system due to the presence of oxygen-containing functional groups, which renders it less effective as a microwave susceptor than GNS under solid-state microwave irradiation. Therefore, GO alone might not be heated to a high enough temperature to cause its exfoliation under solid-state microwave irradiation in this study.[25–28] In the case of the GO/GNS mixture (GO : GNS = 90 : 10 wt.%) (Fig. 1a), the explosion of GO in the mixture was observed with huge volume expansion after 10 s microwave irradiation (Fig. 1b) (see video in the ESI†). This suggests that the GO/GNS mixture could be heated rapidly by microwave irradiation in the presence of GNS with its extended π-system as an effective microwave susceptor.[25–28] Within 20 s of microwave irradiation, several intermittent exfoliations of GO/GNS powder occurred and the resulting powder turned from yellowish brown to black. When 0.02 g of the powder mixture was exfoliated in a 30 mL quartz bottle, it occupied the entire volume of the bottle. Upon further microwave irradiation for the next 20 s on the exfoliated GO/GNS powder (Fig. 1c), a white light began to appear probably due to arc evolution from the product powder in the quartz bottle (Fig. 1d).

The temperature could not be measured in this solid-state microwave irradiation process but it was reported that the local temperature of a microwave susceptor could reach as high as 1500 °C instantly when microwave arcing occurred.[29–31] Since exfoliation and subsequently arc evolution could be observed in the GO/GNS powder mixture within 40 s of microwave irradiation, the average heating rate during microwave irradiation was estimated to be over 2000 °C min^{-1} (1500 °C per 40 s) in this study. The solid-state microwave irradiation of a mixture of graphite oxide and GNS can provide a heating rate that is high enough to ensure rapid pressure buildup due to the decomposition of GO that is sufficient to overcome the van der Waals forces between the graphene sheets in GO for uniform exfoliation.[13,14,23]

Fig. 2 shows SEM images of GNS prepared by solid-state microwave irradiation synthesis and microwave-assisted hydrothermal synthesis. As shown in Fig. 2a, the GNS prepared by

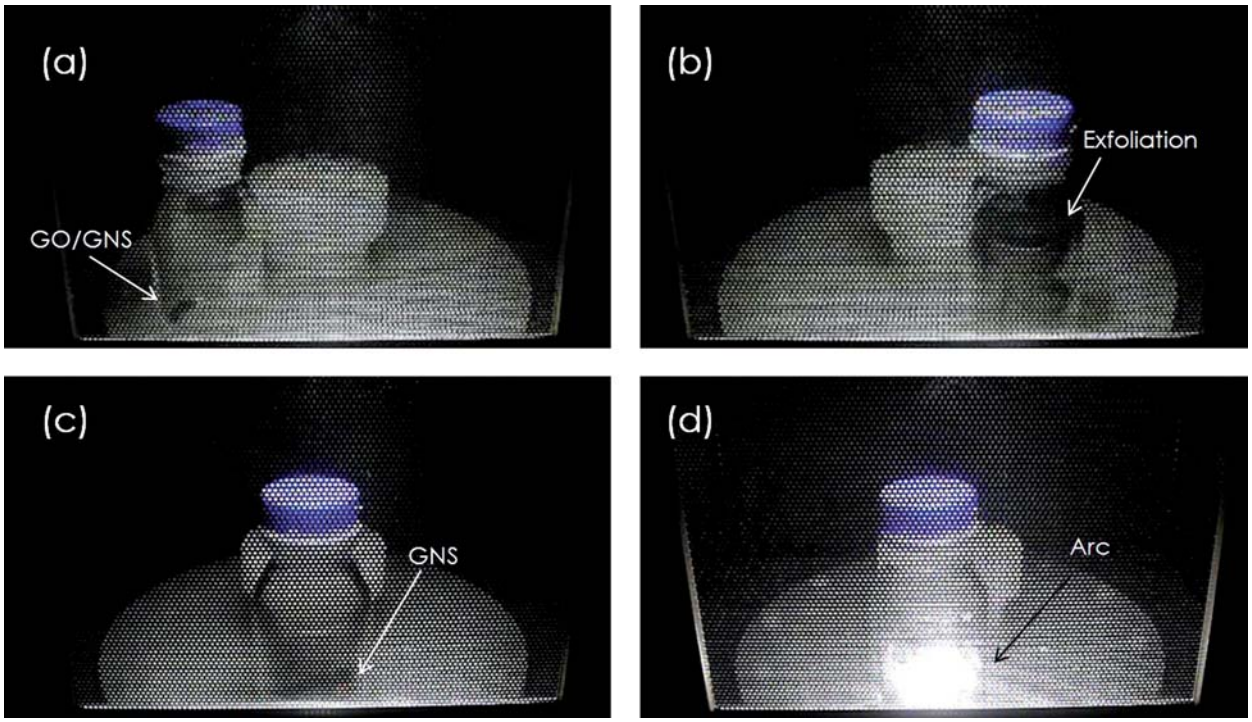

Fig. 1 Photographic images of the sequence of the exfoliation process during solid-state microwave irradiation synthesis. (a) GO/GNS mixture powder, (b) exfoliation of GO/GNS powder, (c) GNS after exfoliation and (d) arc evolution from GNS.

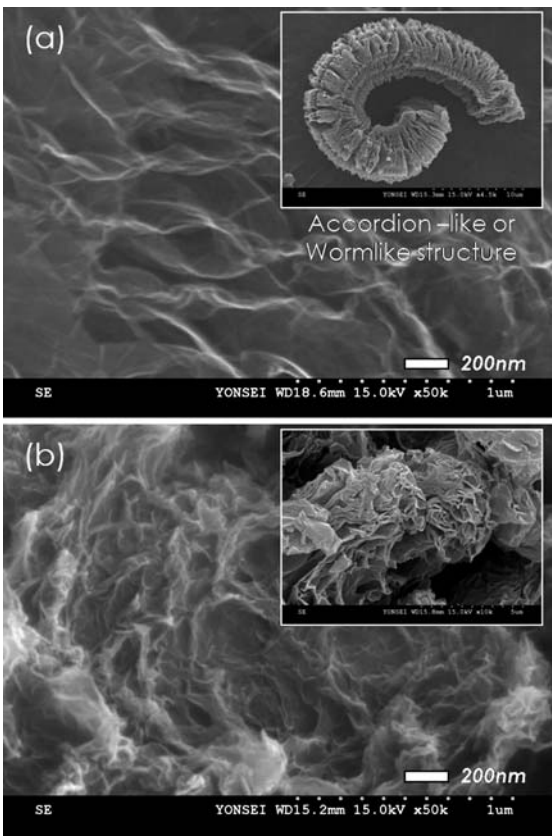

Fig. 2 SEM images of (a) GNS by solid-state microwave irradiation synthesis and (b) GNS by microwave assisted hydrothermal synthesis.

solid-state microwave irradiation synthesis has an accordion or wormlike structure, which is the characteristic morphology of thermally exfoliated GO.[13,14,23] The high magnification SEM image in Fig. 2a shows that most of the GNS was well exfoliated with ultra thin sheets. GNS prepared by microwave-assisted hydrothermal synthesis consists of randomly agglomerated and wrinkled sheets (Fig. 2b), which is the typical morphology of GNS prepared by solution-based reduction syntheses from GO, such as microwave hydrothermal, solvothermal or chemical reduction methods.[15–20,36–38] A comparison of the high magnification SEM images of GNS in Fig. 2 shows that the GNS prepared by microwave assisted hydrothermal synthesis has a more agglomerated and crumpled morphology with overlapping sheets than the GNS prepared by solid-state microwave synthesis.

Fig. 3 shows HR-TEM images of the GNS fabricated by solid-state microwave irradiation synthesis. Observation of the low magnification HR-TEM images (Fig. 3a) indicates that the GNS

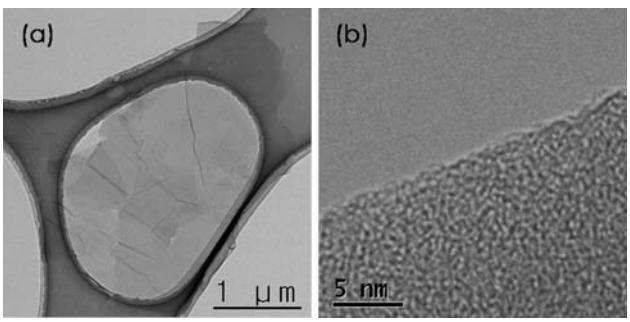

Fig. 3 HR-TEM images of GNS fabricated by solid-state microwave irradiation synthesis. (a) Low magnification and (b) high magnification.

has micrometre-sized flakes with folded and stacked sheets. The high magnification HR-TEM image of GNS (Fig. 3b) shows a folded structure of the GNS edges, which suggests the presence of single or a few layers of GNS. And the electron diffraction patterns indicate the presence of single layer graphene. (See S2 in the ESI†).

AFM was carried out to measure the thickness and number of stacked layer in GNS. Fig. 4 shows representative AFM images of GNS fabricated by solid-state microwave irradiation. A variation of the thickness across the GNS flake was clearly observed, indicating the stacking and folding of GNS. Cross-sectional AFM analysis (Fig. 4a) showed that the height difference was 1.13 nm between (1) and (2) and 2.25 nm between (1) and (3). In a larger GNS flake with a lateral dimension of several micrometres (Fig. 4b), the mean thickness of GNS was 3.15 nm. Since the thickness of a single graphene sheet obtained by AFM was reported to be ~1 nm, it suggests that the GNS prepared in this study contained 1–4 sheets.[13,14,17,23]

The theoretical specific surface area of a graphene monolayer is 2620 $m^2 g^{-1}$. The degree of exfoliation and restacking tendency in GNS can affect the specific surface area of bulk GNS powder. The specific surface area of GNS prepared by solid-state microwave irradiation synthesis and microwave assisted hydrothermal synthesis was measured to be 586 and 382 $m^2 g^{-1}$, respectively. Solid-state microwave irradiation synthesis provides more effective exfoliation of GNS than microwave-assisted hydrothermal synthesis because the as-prepared dry GNS has little possibility of restacking or agglomeration of the exfoliated sheets during synthesis. In the case of the GNS prepared by microwave assisted hydrothermal treatment of an aqueous GO dispersion, there is a strong tendency GNS in the dispersion to aggregate and restack owing to its hydrophobic nature, which might have a negative effect on the specific surface area in its dry state.

Fig. 5 shows the FT-IR spectra of GO and GNS fabricated by solid-state microwave irradiation synthesis and microwave assisted hydrothermal synthesis. GO (Fig. 5a) exhibits the

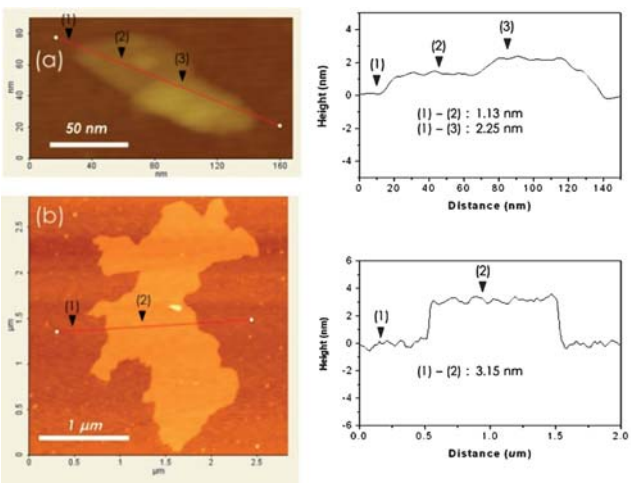

Fig. 4 AFM images and cross-section analysis of GNS prepared by the solid-state microwave irradiation synthesis. (a) GNS with lateral dimension of several tens of nanometres and (b) GNS with lateral dimension of several micrometres.

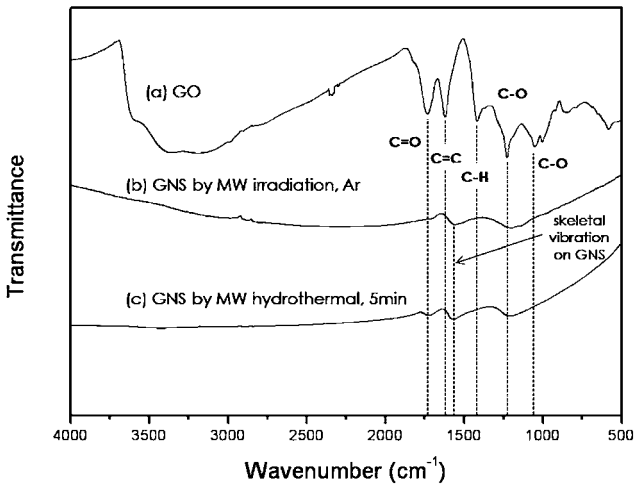

Fig. 5 FT-IR spectra of (a) GO, (b) GNS prepared by solid-state microwave irradiation synthesis and (c) GNS by microwave assisted hydrothermal synthesis.

characteristic peaks for C=O (1731 cm^{-1}), aromatic C=C (1621 cm^{-1}), carboxy C–O (1417 cm^{-1}), epoxy C–O (1226 cm^{-1}) and alkoxy C–O (1052 cm^{-1}).[15,18] For GNS prepared by solid-state microwave irradiation synthesis (Fig. 5b), the peaks for oxygen-containing functional groups were reduced significantly and the peak for C=O (1731 cm^{-1}) was virtually removed. A new peak appeared at 1568 cm^{-1}, which was attributed to the skeletal vibrations of the graphene sheets.[38] For GNS prepared by microwave assisted hydrothermal synthesis (Fig. 5c), the peaks for oxygen-containing functional groups were reduced significantly and the peak at 1568 cm^{-1} for the skeletal vibration of the graphene sheets appeared. This suggests that GO can be reduced without a reducing agent, such as hydrazine and sodium borohydride under microwave-assisted hydrothermal treatment at 200 °C for 5 min.[37,38]

Elemental analysis was carried out to determine the quality and reduction level of GNS fabricated by solid-state microwave irradiation synthesis and microwave assisted hydrothermal synthesis. For a more quantitative analysis of the reduction level of GNS, the carbon/oxygen atomic ratio (C/O ratio) was evaluated and compared for GO and GNS prepared by the two different methods. It was reported that a hydrogen atmosphere plays an important role in improving the quality of GNS during thermal exfoliation and reduction because a H_2 containing atmosphere tends to form H_2O instead of CO_2 from the oxygen-containing functional groups in GO, which prevents the formation of vacancies and defects due to the loss of carbon atoms.[23,24] Therefore, solid-state microwave irradiation synthesis of GNS was performed on a GO/GNS mixture in a quartz bottle filled with an ambient air, Ar gas or H_2/Ar gas mixture.

Table 1 lists the results of elemental analysis of GO and GNS prepared by solid-state microwave irradiation synthesis under different gas atmospheres and microwave assisted hydrothermal synthesis. GO prepared using the Hummers method contained 48.6 wt.% carbon and 45.4 wt.% oxygen, which are similar to the values reported in the literature.[15–20] GNS prepared by microwave solid-state irradiation synthesis in air showed a C/O ratio of 3.6, which is similar to the value of 2.75 for GNS reported by

Table 1 Elemental analysis of GO and GNS prepared by solid-state microwave irradiation synthesis and microwave assisted hydrothermal synthesis under different conditions

Sample	C/ wt.%	O/ wt.%	H/ wt.%	S/ wt.%	N/ wt.%	C/O ratio
GO	48.6	45.4	2.2	3.75	—	—
GNS, MW irradiation, ambient air	70.3	26.2	1.15	2.3	—	3.6
GNS, MW irradiation, Ar atmosphere	84.6	9.85	0.7	4.85	—	11.45
GNS, MW irradiation, H_2 atmosphere	91.8	6.6	1.6	—	—	18.5
GNS, MW hydrothermal, 5 min, 200 °C	78.0	21.2	0.8	—	—	4.9
GNS, MW hydrothermal, 15 min, 200 °C	79.0	20.2	0.8	—	—	5.2
GNS, MW hydrothermal, 60 min, 200 °C	83.5	15.7	0.8	—	—	7.1

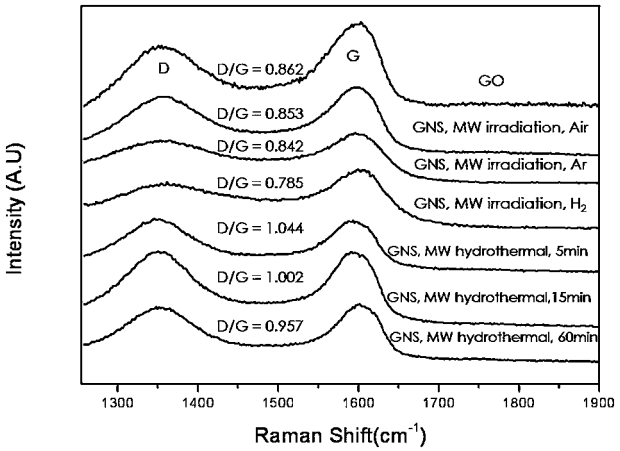

Fig. 6 Raman spectra of GO and GNS prepared by solid-state microwave irradiation and microwave assisted hydrothermal synthesis under different conditions.

Ruoff *et al.* in their study of a solid-state microwave irradiation treatment of GO in ambient atmosphere without a microwave susceptor.[35] However, the GNS prepared by microwave solid-state irradiation synthesis under an Ar atmosphere and H_2/Ar atmosphere showed a C/O ratio of 11.45 and 18.5, respectively. This shows that the C/O ratio of GNS increased in the order of air, Ar atmosphere and H_2/Ar atmosphere for microwave solid-state irradiation synthesis. In addition, a H_2 containing atmosphere is the most effective in promoting the reduction of GNS from oxygen-containing functional groups in GO. It should be noted that GNS with a C/O ratio of 18.5 could be prepared by a solid-state microwave irradiation treatment of a GO/GNS mixture under H_2/Ar atmosphere in as little as 40 s. The C/O ratio of 18.5 in this study is among the highest reported thus far.[13,14,23,24] In the case of the GNS prepared by microwave-assisted hydrothermal synthesis, the C/O ratio of GNS was 4.9, 5.2 and 7.1 for a reaction time of 5, 15 and 60 min, respectively. The C/O ratio of GNS increased gradually with increasing reaction time in the microwave-assisted hydrothermal treatment. Elemental analysis of GNS shows that solid-state microwave irradiation synthesis under an Ar or H_2/Ar atmosphere is far more effective in reducing GO than microwave assisted hydrothermal synthesis.

Raman spectroscopy provides information on the structural properties of carboneous materials, including disorder and defect structures. Fig. 6 shows the Raman spectra of GO prepared using Hummers method, GNS prepared by solid-state microwave irradiation synthesis under different gas atmospheres and microwave-assisted hydrothermal synthesis. All the Raman spectra contained two prominent peaks at 1350 cm^{-1} and 1580 cm^{-1}, which correspond to the D and G bands.[39–43] It was reported that the G band corresponds to the first-order scattering of the E_{2g} mode from the sp^2 carbon domains and the D band originates from the disorder-induced mode associated with structural defects and imperfections.[39–41] Therefore, the intensity ratio of I(D)/I(G) is generally used as a measure to evaluate the quality of the graphitic structures.

In the case of GNS prepared by solid-state microwave irradiation synthesis, the I(D)/I(G) ratio of GNS fabricated in ambient Air, Ar and H_2/Ar atmosphere was 0.853, 0.842 and 0.785, respectively. The I(D)/I(G) ratios of GNS prepared by solid-state microwave irradiation synthesis were smaller than those of GO (0.862). This might be due to a self-healing or defect-healing effect occurring at high temperatures during solid-state microwave irradiation synthesis.[23,42,43] The smaller I(D)/I(G) ratio for GNS prepared under a H_2/Ar atmosphere compared to that under an Ar atmosphere suggests that the hydrogen atmosphere is effective in preventing the formation of vacancies and defects during the decomposition of oxygen-containing functional groups in GO. For microwave-assisted hydrothermal synthesis, the I(D)/I(G) ratio of GNS was 1.044, 1.002 and 0.957 for a reaction time of 5, 15 and 60 min, respectively. The I(D)/I(G) ratio of GNS decreased gradually with increasing reaction time in microwave-assisted hydrothermal synthesis.

Elemental analysis of GNS prepared by microwave-assisted hydrothermal synthesis in Table 1 shows that the C/O ratio increased with increasing reaction time and is associated with an increase in the graphitic sp^2 domains and with an increase in the reduction level of GNS.[44] Regardless of the reaction time during microwave-assisted hydrothermal synthesis, the I(D)/I(G) ratio of GNS was higher than that of GO. It is possible that the graphitic domains created after the reduction process were smaller than that for GO, and the defects once formed during the exfoliation and reduction of GO could barely be removed during the reduction process.[17,41] However, in the case of GNS prepared by solid-state microwave irradiation synthesis, the I(D)/I(G) ratio of GNS decreased from that of GO.

Table 2 shows the electrical conductivity of pellets for graphite, GO prepared by Hummers method, GNS prepared by solid state microwave irradiation under H_2/Ar gas atmosphere, GNS prepared by solid state microwave irradiation under Ar gas atmosphere and GNS prepared by microwave assisted hydrothermal synthesis. The electrical conductivity of the graphite pellet and the graphite oxide pellet was measured to be 1.53×10^3

Table 2 Electrical conductivity of pellets for graphite, GO prepared by Hummers method and GNS prepared by different conditions

Sample	Conductivity/ S m^{-1}	Literature value/S m^{-1}
Graphite	1.53×10^3	2.5×10^{3a}
GO	4.3×10^{-2}	2×10^{-2a}
GNS prepared by solid state microwave irradiation, H_2/Ar gas atmosphere	1.25×10^3	2.4×10^{3a}
GNS prepared by solid state microwave irradiation, Ar gas atmosphere	7.41×10^2	
GNS prepared by microwave assisted hydrothermal synthesis	2.77×10^2	

[a] Conductivity of graphite, graphite oxide and reduced graphite oxide reported in ref. 17.

S m^{-1} and 4.3×10^{-2} S m^{-1}, respectively and these values are within the range of literature values reported previously.[17] GO prepared by Hummers method shows the lowest conductivity of 4.3×10^{-2} S m^{-1} mainly due to the partially disconnected π-system caused by various oxygen-containing functional groups. Electronic conductivity was 1.25×10^3 S m^{-1} for GNS prepared by solid state microwave irradiation under H_2/Ar gas atmosphere, 7.41×10^2 S m^{-1} for GNS prepared by solid state microwave irradiation under Ar gas atmosphere and 2.77×10^2 S m^{-1} for GNS prepared by microwave assisted hydrothermal synthesis. And the C/O ratio was 18.5 for GNS prepared by solid state microwave irradiation under H_2/Ar gas atmosphere, 11.45 for GNS prepared by solid state microwave irradiation under Ar gas atmosphere and 4.9 for GNS prepared by microwave assisted hydrothermal synthesis. It can be clearly seen that the electronic conductivity of GNS pellets increased with increasing C/O ratio, which could be attributed to the degree of restoration of the π conjugated system in GO during the reduction processes.[45] This study shows that solid-state microwave irradiation of GO under H_2/Ar gas atmosphere is far more effective in reducing GO compared with other treatments used in this study. It is noteworthy that the electrical conductivity of 1.25×10^3 S m^{-1} for GNS prepared by solid state microwave irradiation synthesis under H_2/Ar gas atmosphere is of the same order of magnitude as the electrical conductivity of graphite powder (1.53×10^3 S m^{-1}). It demonstrates that solid-state microwave irradiation treatment of the mixture powder of 90 wt.% GO and 10 wt.% GNS under H_2/Ar gas atmosphere can be developed as a procedure to prepare the high quality GNS as evidenced by the C/O ratio and electrical conductivity of GNS.

The electronic conductivity combined with the electron diffraction patterns and the elemental analysis confirmed that the solid-state microwave irradiation treatment of a mixture of GO and GNS under a hydrogen containing atmosphere provides a facile and effective way of fabricating high quality GNS.

4. Conclusions

High quality GNS were fabricated by solid-state microwave irradiation of a mixture of GO and GNS under a hydrogen containing atmosphere.

In this study, the GNS in the mixture acted as an effective microwave susceptor. The GNS prepared by solid-state microwave irradiation synthesis under a hydrogen atmosphere had a high specific surface area of 586 m^2 g^{-1} and a C/O ratio of 18.5. Furthermore, the I(D)/I(G) ratio in the Raman spectra decreased from 0.862 for GO to 0.785 for GNS prepared by solid-state microwave irradiation synthesis under a hydrogen atmosphere, whereas it increased to 0.957 for GNS prepared by microwave-assisted hydrothermal synthesis. A hydrogen atmosphere plays important role in improving the quality of GNS by promoting the reduction of GO and preventing the formation of defects in GNS. Solid-state microwave irradiation synthesis is a rapid, clean, convenient and scalable process that can fabricate high quality GNS.

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