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Preparation of a Stable Graphene Dispersion with High Concentration by Ultrasound

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With unique structure and extraordinary electronic, thermal, and mechanical properties, graphene fascinates the scientific community. Due to its hydrophobic feature, preparation of a stable and highly concentrated graphene dispersion without the assistance of dispersing agents has generally been considered a challenge. Chemical reduction of graphene oxide (GO) is one of the most important methods for preparing a graphene dispersion. The aggregation of graphene sheets is a key reason to destabilize the resulting dispersion during conversion of aqueous GO dispersion to graphene. In this study, by replacing mechanical stirring with ultrasonic irradiation, the aggregation of various intermediates is effectively suppressed during the process of reduction of GO. Hence, a stable graphene dispersion with a high concentration of $1 \text{ mg} \cdot \text{mL}^{-1}$ and relatively pure graphene sheets are achieved, and the as-prepared graphene paper exhibits a high electric conductivity of $712 \text{ S} \cdot \text{m}^{-1}$. Ultraviolet–visible absorption spectroscopy and X-ray photoelectron spectroscopy show that ultrasound is the essence of enhancing chemical reaction rate. Fourier transformed infrared spectra and Raman spectra indicate that ultrasound has less damage to the chemical and crystal structures of graphene.

1. Introduction

Graphene has attracted much attention from chemical, physical, and material scientific communities due to its extraordinary electrical, thermal, and mechanical properties and potential applications.^{1–5} At present, there have been many methods reported to prepare graphene, such as micromechanical exfoliation,⁶ chemical vapor deposition,⁷ and epitaxial growth,⁸ while graphene prepared by these methods has a common disadvantage of difficulty in processing. It is well-known that hydrophobic graphene sheets, which have a high specific surface area, are prone to form irreversible aggregation through van der Waals interactions. Like many other nanomaterials, a key challenge in the preparation and processing of massive graphene sheets is aggregation which restrains graphene from exhibiting outstanding properties in that most of the unique ones are only associated with an individual sheet. Aggregation makes graphene completely unsuitable for a large scale application in composites. Hernandez et al.⁹ measured the dispersibility of graphene in 40 different solvents, and the best dispersibility is only $8.5 \mu\text{g} \cdot \text{mL}^{-1}$ in cyclopentanone. The poor dispersibility is a major obstacle to processing the bulk-quantity graphene.

For the utilization of graphene by solvent-assisted techniques, such as spin-coating,¹⁰ layer-by-layer assembly,¹¹ and filtration,¹² the ability to prepare a graphene solution or dispersion will be critical. Although the method of solvent exfoliation has potential for preparing a graphene dispersion, the critical disadvantages, such as low yield (1–10%), harsh requirements for solvent, and poor stability of the graphene dispersion, make many applications completely impractical.^{13–16} Even though the concentration of the graphene dispersion is enhanced to $1.2 \text{ mg} \cdot \text{mL}^{-1}$, a time-consuming sonication process (longer than 460 h) is necessary.¹⁷ Generally, the method of the chemical reduction of graphene

oxide (GO) is both scalable, affording the possibility of preparing a graphene dispersion, and versatile in terms of being well-suited to chemical functionalization.¹⁸ Unfortunately, aggregation will occur in the process of converting GO to graphene. Initially, a graphene dispersion was obtained by the attachment of the surfactant stabilizers or polymers on the sheets,^{19–21} while the presence of foreign stabilizers was undesirable for most applications in that it could decrease the purity and decline the electrical, thermal, and mechanical properties of graphene. Recently, Li et al.²² have demonstrated that a homogeneous aqueous graphene dispersion can be obtained by controlling reduction of the GO dispersion and maintaining the pH at about 10, while agglomeration occurred as concentrations greater than $0.5 \text{ mg} \cdot \text{mL}^{-1}$. Yan et al.²³ found that a graphene dispersion can even be achieved at 165°C in a mixture of *N,N*-dimethylacetamide and water via microwave; the concentration of the dispersion is $0.56 \text{ mg} \cdot \text{mL}^{-1}$, and the electric conductivity of the graphene paper is only $200 \text{ S} \cdot \text{m}^{-1}$. At present, it is very difficult to prepare both a highly concentrated and stable graphene dispersion as well as a high electric conductivity of graphene.

However, on the basis of the fact that new chemistry can be generated by using ultrasonic irradiation in preparing nanomaterials due to its cavitation and vibration effect,²⁴ we are motivated to investigate the ability of ultrasonic irradiation to suppress aggregation, especially in the process of the chemical reduction of GO. By far, a stable graphene dispersion which exhibits both good dispersity and high concentration is obtained at low temperature (65°C) under ultrasonic irradiation, and then as-prepared graphene has a high electric conductivity ($712 \text{ S} \cdot \text{m}^{-1}$).

2. Experiments

Preparation of a GO Dispersion. All chemicals except natural flake graphite were of analytical grade from Xi'an Chemical Reagent Factory. The graphite oxide was prepared from natural flake graphite (200 mesh, Shandong Nanshu) by

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the well-known Hummers method.²⁵ A 10 g portion of graphite, 5 g of sodium nitrate, and 230 mL of sulfuric acid (98%) were mixed and strongly stirred at 0 °C for 15 min in a 1000 mL reaction flask immersed in an ice bath. Then, 30 g of potassium permanganate was added slowly to the above mixture and the rate of addition was controlled carefully to prevent the temperature of the suspension from exceeding 20 °C. After the dispersion was stirred continuously for 2 h, the ice bath was then removed and the temperature of the suspension was brought to 35 °C. At the end of 30 min, 460 mL of water was added slowly to the suspension for 40 min. The suspension was diluted by 1.4 L of warm water (40–60 °C) and treated with some H₂O₂ (30%) to reduce residual permanganate to soluble manganese ions until the gas evolution ceased. The resulting suspension was filtered, washed with water, and dried in a vacuum oven at 50 °C for 24 h to obtain graphite oxide. The graphite oxide was dispersed in water to create a 1 mg·mL⁻¹ GO dispersion and exfoliation of graphite oxide to GO was achieved by ultrasound of the dispersion using an ultrasonic cleaning bath (model ACQ-600, Shaanxi Ultrasonic Instrument Company) for 2 h, using a power of 250 W (determined by the electrical method) and operated at 27 kHz, the GO dispersion was obtained.

Preparation of a Graphene Dispersion. The chemical reduction of the GO was carried out using ultrasound treatment. Ammonia solution (5% in water) was added to the GO dispersion to increase the pH to 10, 100 mL of the GO dispersion was heated in a water bath at 65 °C under a water-cooled condenser and a nitrogen-filled drybox, and the dispersion was processed using an ultrasonic cleaning bath in the whole reaction. A 0.14 g portion of hydrazine (50%) was added to the dispersion at a temperature of 65 °C, a graphene dispersion was collected by filtration with a 200 mesh net at different reaction times, such as 10, 20, 40, 60, 70, and 120 min, and the sample was defined as UG_{time}, like UG₁₂₀, indicating that the graphene dispersion was collected at 120 min. After the reaction, the suspension was filtered through a microspore filter (50 mm in diameter, 0.2 μm pore size, Shanghai), and the graphene paper was washed copiously with water and dried in a vacuum oven at 25 °C. For comparison, all graphene papers and GO papers were also prepared by the same method.

As a control, graphene was also prepared by a similar procedure with mechanical stirring instead of ultrasonic irradiation, and the sample was denoted as MG_{time} here.

Characterization. Dried graphene powder (ca. 10 mg) and solvent (ca. 5 mL) were added into a centrifugal cuvette to be ultrasonicated for 2 h. The suspension was subjected to 30 min of centrifugation at 3000 rpm. After the supernatant was decanted, the cuvette with sediment was dried with no solvent existing. The concentration of supernatant could be calculated as

$$C_g = \frac{w_1 - w_2}{V}$$

in which the w_1 is the weight of dried graphene, w_2 is that of the sediment, and V is the volume of solvent. C_g can be used to describe the dispersibility of graphene in a certain solvent.

Ultraviolet–visible (UV–vis) absorption spectra were recorded using a 1 cm path length quartz cuvette with water as the reference on a U-2001 UV/Vis Spectrophotometer (Hitachi). Fourier transformed infrared (FT-IR) spectra of all of the samples were recorded between 500 and 4000 cm⁻¹ from KBr pellets by a Bruker Tensor 27 Spectrophotometer. When the

electric conductivity of graphene was lower than 10⁻² S·m⁻¹, the volume resistance of free-standing graphene paper (100.0 mm in diameter and 2 μm in thickness) prepared by vacuum filtration was measured at ambient temperature employing a CS2676C Insulation Resistance Tester (Nanjing Changsheng Instrument Co. Ltd., China). When the electric conductivity of graphene was higher than 10⁻² S·m⁻¹, the electric conductivity of free-standing graphene paper (50.0 mm in diameter and 2 μm in thickness) prepared by vacuum filtration was measured at ambient temperature employing the standard four-probe technique with a SZ85 Digital Multimeter (Suzhou Telecommunication Factory China). Morphology was determined on a JEM-3010 Transmission Electron Microscope (TEM). For TEM examination, the dried samples were ultrasonically dispersed in water and cast onto TEM grids. Raman spectra were recorded from 200 to 2000 cm⁻¹ on a Confocal Raman Microprobe (HORIBA Jobin Yvon) using a 633 nm helium–neon laser. X-ray photoelectron spectroscopy (XPS) measurements were performed using an X-ray Photoelectron Spectrometer (AXIS ULTRA, Kratos Manchester, U.K.) with monochromated Al Kα radiation ($h\nu = 1486.6$ eV).

3. Results and Discussion

It is hard to prepare a stable graphene dispersion with high concentration under common mechanical stirring in that the inter- or intra- π – π stacking of graphene with a mount of aromatic carbon leads to aggregation.²⁶ The shear action of the mechanical stirring is beneficial to dispersing the graphene sheets, whereas it is not intense enough to suppress aggregation of graphene sheets through van der Waals interactions. Since being prepared in the middle twentieth century, graphite oxide has been mainly produced by acid treatment of graphite, as the Hummers method. The graphite oxide containing lots of hydrophilic oxygen-containing groups, such as carboxyl, hydroxyl, and epoxide, very easily forms a well-dispersed aqueous GO dispersion. As recently demonstrated by Rouff and his co-workers, the reduction of GO by the chemical method (using hydrazine which is an effective reductant) has produced electrically conducting graphene. Unfortunately, as reduction proceeds, the yellow-brown of the GO dispersion gradually turns black and the graphene sheets aggregate into irregular particles in the case of mechanical stirring, which severely restricts the exploration of further applications. It is found that a stable and homogeneous graphene dispersion could be obtained at a reaction time no longer than 40 min. A small quantity of particles emerge in the dispersion and the gradually agglomerating particles are too big to pass the 200 mesh net (the net is utilized to filtrate MG_{time} for removal of the big particles due to agglomeration) when the reaction time reaches more than 40 min until 90 min. After the reaction of 90 min, the graphene dispersions obtained by filtration with the 200 mesh net are quite transparent and the color approaches achromaticity. Because most of the intermediates agglomerate into irregular big particles which are filtrated by the 200 mesh net, the concentration of the as-filtrated graphene dispersion is quite low. The same volumes of graphene dispersions (1 mg·mL⁻¹) with different reduction levels are diluted to 0.05 mg·mL⁻¹ graphene dispersions (Figure 1a). The results that the reduction of GO dispersion with a concentration greater than 0.5 mg·mL⁻¹ leads to aggregation in the presence of mechanical stirring in our experiments are consistent with those found by Li et al.²² In the case of mechanical stirring, the homogeneous dispersion is obtained before the reaction of 40 min, and yet the electric conductivity of graphene is below 18.1 S·m⁻¹. The conflict

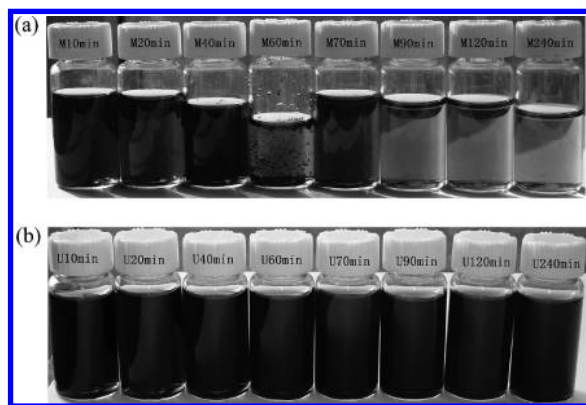


Figure 1. Photographs of MG_{time} (a) and UG_{time} (b) dispersion. The concentration of the graphene dispersion is $0.05 \text{ mg} \cdot \text{mL}^{-1}$.

between a high concentration of a stable graphene dispersion and a high electric conductivity of graphene is hardly solved in preparation of graphene using chemical reduction of GO under mechanical stirring, which is the major obstacle to their potential applications.

To solve the problem, ultrasound is introduced in the process of reduction of GO instead of mechanical stirring, and then a stable graphene dispersion with high concentration ($1 \text{ mg} \cdot \text{mL}^{-1}$) is successfully obtained; graphene paper prepared from UG_{120} has a high electric conductivity which is similar to that of graphite and higher than that of graphene paper prepared from MG_{120} . In the presence of ultrasonic irradiation, the chemical reduction process is traced and the yellow-brown of the GO dispersion gradually turns black, indicating that GO is gradually reduced to UG_{time} . The graphene dispersions show the excellent dispersity and homogeneity in the whole reaction of reduction of GO under ultrasonic irradiation. After 40 days, the aqueous hydrazine-reduced graphene dispersions at different reaction times are still stable and homogeneous (Figure 1b). The results show that ultrasound is efficient to assist preparation of the high quality graphene dispersion in that the ultrasonic irradiation not only prevents the agglomeration of graphene particles but also suppresses the aggregation of graphene sheets.

In the mechanical stirred system, the morphology of the aggregated graphene is observed, while in the case of the ultrasonic irradiation, the aggregation of graphene sheets is effectively suppressed and flat film-like graphene sheets are exhibited. The graphite oxide in water is ultrasonically exfoliated to single layers or a few layers of GO which are flat, thin, and transparent in Figure 2a. The GO sheets corrugate together with a size in the range of tens to thousands of square nanometers. With the continuing of the chemical reduction of GO, there is lots of corrugation, scrolling, and agglomeration on the edge of graphene in the mechanical stirred system (Figure 2b) and no obvious aggregation is observed in the TEM image of UG_{120} (Figure 2c), indicating that the ultrasound is very important for stabilizing graphene dispersions and maintaining the flat graphene sheets. The HRTEM image of UG_{120} shows very thin sheets with a thickness of less than 4 nm (layers of most graphene < 5) and very small lateral size compared with GO, which is beneficial in forming a stable graphene dispersion. The selected area electron diffraction (SAED) images of UG_{120} and MG_{120} , as shown in the insets in Figure 2b and c, suggest that graphene is partially restored to the ordered crystal structure. No obvious diffraction spot is visible in the SAED of MG_{120} , while the well-defined diffraction spots confirm the crystal structure of the graphene nanosheets obtained via reduction of GO under ultrasonic irradiation, indicating that the thin flat graphene film

with high quality is obtained by ultrasound. At the same time, the SAED image (see the inset in Figure 2c) is also indicative of random stacking of graphene sheets, as the reaggregation of graphene is effectively suppressed rather than completely prevented in the presence of ultrasound. Though slight reaggregation occurs, it is not sufficient to destroy the stability of a graphene dispersion. As compared with the mechanical stirring, unexfoliated graphite oxide sheets are gradually exfoliated to form GO under ultrasonic irradiation, which effectively suppresses the self-assembly and aggregation of graphene sheets, and a high concentration of a stable graphene dispersion is also achieved. These are probably attributed to the cavitation effect of ultrasonic irradiation that is induced by the formation, growth, and implosive collapse of bubbles in the liquid.²⁷ Ultrasound provides acoustic cavitation near the graphene surface, which can weaken the van der Waals interactions among graphene sheets, rip the platelets apart, and restrict the aggregation of graphene sheets to thoroughly disperse in water, suggesting that ultrasound supplies a qualification to prepare a high concentration of graphene dispersion. At the same time, the as-prepared graphene which is prepared from U_{120} through successive filtration, dryness, and comminution can be dispersed in water with a concentration of $0.5 \text{ mg} \cdot \text{mL}^{-1}$ due to graphene with a few hydrophilic oxygen-containing groups, which is supported by the FT-IR spectra of UG_{120} (Figure 5). It is suggested that graphene can readily disperse in water to generate a high concentration of stable graphene dispersion without any stabilizers. Of great significance is that the successful formation of a stable graphene dispersion with high concentration enables the use of conventional low-cost solution-phase processing techniques to create new graphene-based materials and devices.

During the chemical reduction of GO in ultrasonic irradiation, the reaction is accelerated, and as a result, the electric conductivity of the product, graphene, is enhanced. It is found that the reduction process from GO to graphene is very sensitive to reaction temperature. UV-vis spectra, in which the absorption peaks of GO and graphene are ca. 230 and 268 nm, respectively, and electric conductivity measurements are employed to monitor the degrees of reduction of GO. The absorption peak of the product does not reach 267 nm in UV-vis spectra until the reaction maintains 60 min at 65°C , and the electric conductivity is only $700 \text{ S} \cdot \text{m}^{-1}$. Once the temperature is enhanced to 95°C , the absorption peak of the product reaches 267 nm in UV-vis spectra when the reaction time is 7 min and the electric conductivity has already approached $4000 \text{ S} \cdot \text{m}^{-1}$. It is clearly proved that the temperature is an important factor in the process of chemical reduction of GO. It is well-known that transient cavitations form short-lived cavities (bubbles) that end in catastrophic collapse.²⁸ This occurs when the cavity is vapor filled rather than gas filled which leads to the release of large localized amounts of energy and results in rapid temperature increase of the surrounding environment that provide the energy for preparing the graphene, and also the reaction rate of reduction GO is enhanced. This is supported by the redshift of UV-vis spectra and the increase of electric conductivity. In UV-vis spectra, the absorption peak of the GO at 230 nm gradually redshifts to 268 nm with continuing reaction under ultrasound (Figure 3), suggesting that the electronic conjugation within the graphene sheets is restored upon hydrazine reduction. Little increase in absorption is found after 120 min, indicating completion of the reduction within that period. The wavelength of the absorption peak of UG_{time} redshifts 1–2 nm compared with that of MG_{time} with the same reaction time (Supporting Information Figure S1), hinting that the conversion from GO

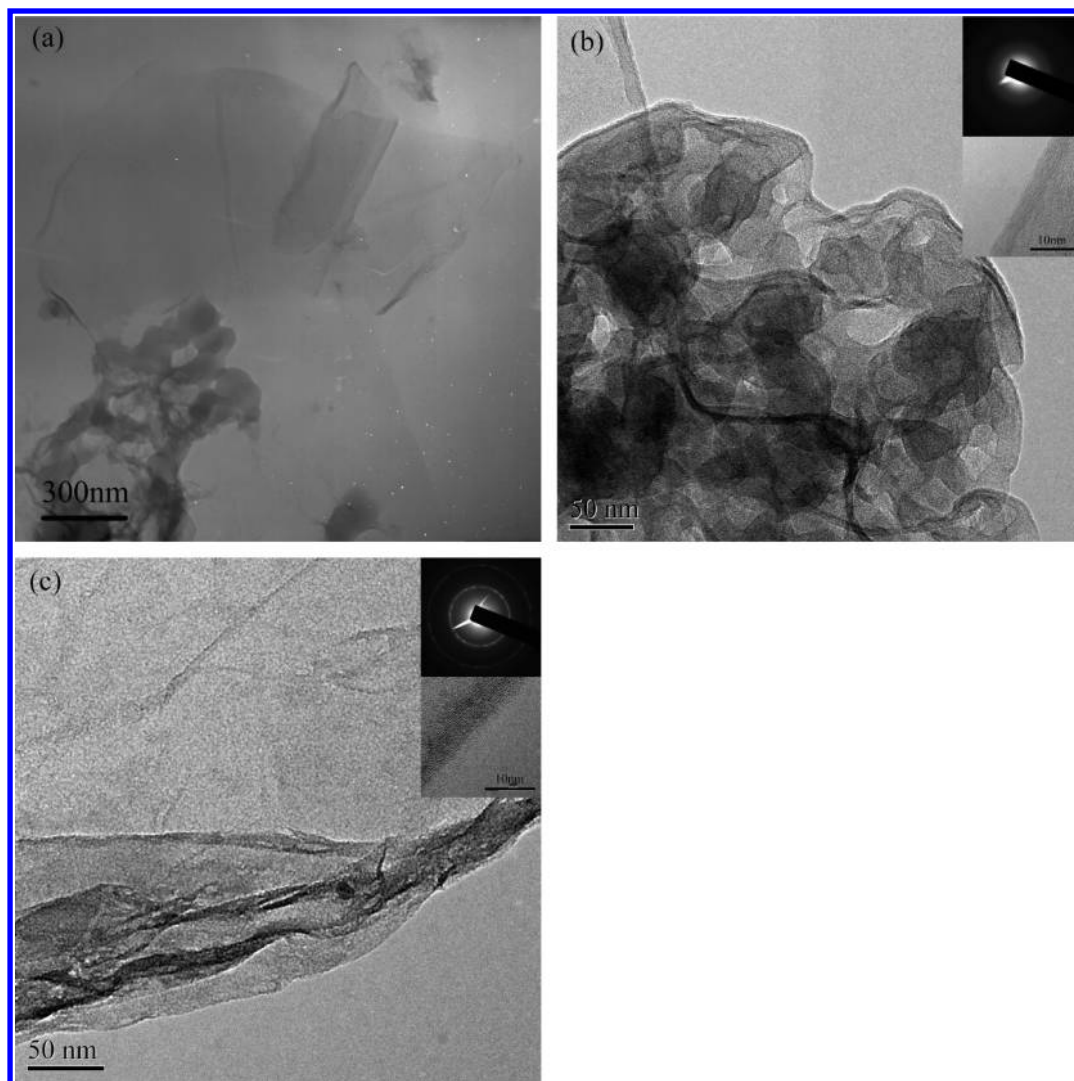


Figure 2. TEM images of GO (a), MG₁₂₀ (b), and UG₁₂₀ (c). The insets in panels b and c are SAED and HRTEM.

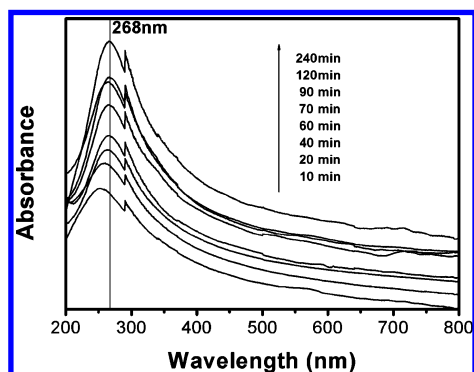


Figure 3. During the reduction of GO, UV-vis absorption spectra showing the change of the GO dispersion as a function of reaction time under ultrasonic irradiation.

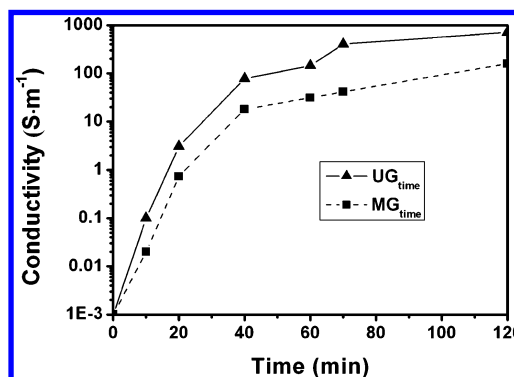


Figure 4. Electric conductivity of UG_{time} and MG_{time}.

to graphene under ultrasonic irradiation is quicker than that under mechanical stirring. It is found that the electric conductivity of UG_{time} is far higher than that of MG_{time} in the same reaction time (Figure 4). The electric conductivity of MG₁₂₀ is close to $159 \text{ S} \cdot \text{m}^{-1}$, while the electric conductivity of UG₁₂₀ has already reached $712 \text{ S} \cdot \text{m}^{-1}$. It implies that the ultrasonic reduction route is more effective than the mechanical stirring route in repairing the sp^2 network and enhancing the electric conductivity of graphene.

FT-IR spectra of UG₁₂₀ (c) and MG₁₂₀ (d) are shown in Figure 5 and no significant difference is observed between UG₁₂₀ and MG₁₂₀. Figure 6 shows the FT-IR spectra of graphite (a), GO (b), UG₁₀ (c), UG₂₀ (d), UG₆₀ (e), UG₉₀ (f), and UG₁₂₀ (g). The spectrum of graphite oxide illustrates O-H (carboxyl) at 1400 cm^{-1} and O-H at 3448 cm^{-1} originated from carboxylic acid and the O-H stretching mode of intercalated water, C=O in carboxylic acid and carbonyl moieties at 1730 cm^{-1} , C-O (epoxy or alkoxy) at 1052 cm^{-1} and C=C at 1627 cm^{-1} assigned to skeletal vibrations of unoxidized graphitic domains or contribution from the stretching deformation vibration of

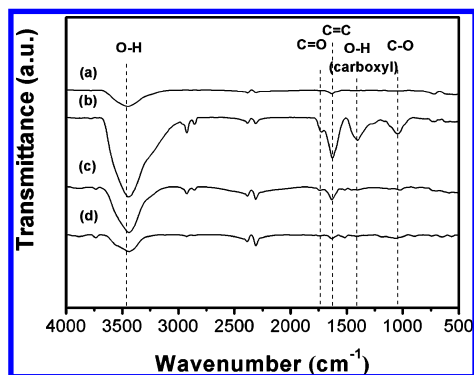


Figure 5. FT-IR transmittance spectra of graphite (a), GO (b), MG₁₂₀ (c), and UG₁₂₀ (d).

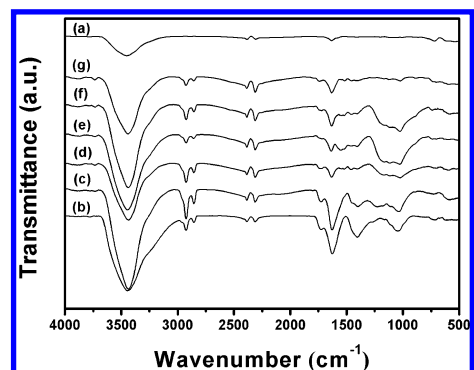


Figure 6. FT-IR transmittance spectra showing the change of the GO dispersion as a function of reaction time: graphite (a); GO (b); UG₁₀ (c); UG₂₀ (d); UG₆₀ (e); UG₉₀ (f); UG₁₂₀ (g).

intercalated water.²⁹ After the GO is chemically treated by hydrazine, the peaks for oxygen functional groups gradually disappear and the FT-IR spectra of UG_{time} are more and more similar to that of graphite, demonstrating most of the oxygen-containing groups of GO are removed and a high quality of graphene can be achieved under ultrasonic irradiation. However,

a bit of epoxide groups and hydroxyl groups remain after reduction, which results in dispersibility of graphene.

GO is more thoroughly reduced and graphene owns fewer oxygen-containing groups under ultrasonic irradiation than those under mechanical stirring, which is proved by C1s XPS spectra of GO (a), MG₁₂₀ (b), and UG₁₂₀ (c) in Figure 7. The C1s XPS spectrum of GO clearly indicates a considerable degree of oxidation with four components that correspond to carbon atoms in different functional groups: the nonoxygenated ring C (284.5 eV), the C in C—O bonds (286.2 eV), the carbonyl C (C=O, 287.8 eV) and the carboxylate C (O—C=O, 289.0 eV).³⁰ Although the C1s XPS spectrum of UG₁₂₀ also exhibits the same oxygen functionalities, their peak intensities are much smaller than those in GO, indicative of removal of oxygen-containing groups, which is accompanied by recovery of sp² carbon networks. In addition, there is an additional component at 285.9 eV corresponding to C—N,³¹ indicating the deoxygenation by the reduction process as well as nitrogen in corporation. By tracing the removal of the oxygen-containing groups of GO, the C/O ratio changes from 2.9 for GO to 8.48 for UG₁₂₀, which is higher than 6.38 for MG₁₂₀, suggesting that the reduced degree of GO under ultrasonic irradiation is higher than that under mechanical stirring. Furthermore, it is proved that ultrasound can accelerate the reaction and thoroughly remove the oxygen-containing groups of GO. It is a potential method to combine the ultrasonic irradiation and the chemical reduction in the future to prepare a high concentration of a stable graphene dispersion.

The crystal structure of graphite is damaged during the process of converting graphite to graphite oxide; however, graphene is partially restored to the ordered crystal structure and some defects are repaired via chemical reduction of GO. Raman spectroscopy is a powerful nondestructive tool to distinguish order and disorder in the crystal structure of carbon. Figure 8 shows Raman spectra of graphite (a), GO (d), MG₁₂₀ (b), and UG₁₂₀ (c). The G band is usually assigned to the E_{2g} phonon of C sp² atoms, while the D band is a breathing κ -point phonon of A_{1g} symmetry.³² The Raman spectrum of the pristine graphite in Figure 8 displays a strong G band at 1580 cm⁻¹, a

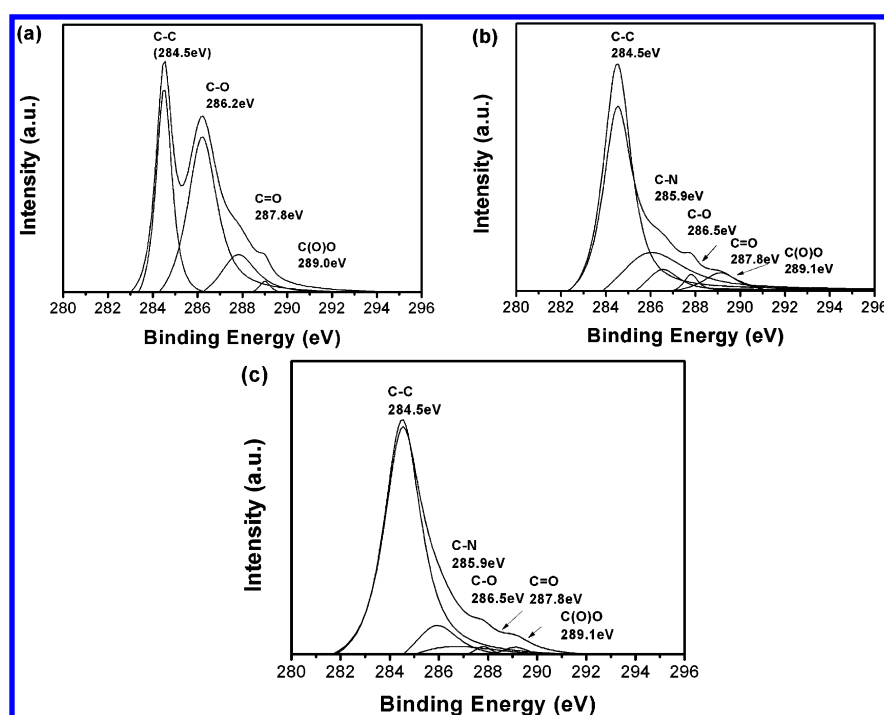


Figure 7. C1s XPS spectra of GO (a), MG₁₂₀ (b), and UG₁₂₀ (c).

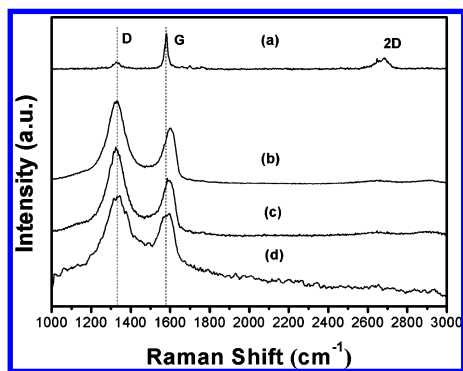


Figure 8. Raman spectra of graphite (a), MG₁₂₀ (b), UG₁₂₀ (c), and GO (d).

weak D band at 1332 cm⁻¹, and a middle 2D band at 2700 cm⁻¹. The typical features of GO in the Raman spectrum are the G band at 1597 cm⁻¹ and the D band at 1334 cm⁻¹. As compared with GO, in the Raman spectra of the UG₁₂₀ and MG₁₂₀, the G bands are narrowed and respectively shift to 1590 and 1596 cm⁻¹, which approach the G band of graphite. At the same time, the intensity of the D band at 1328 cm⁻¹ increases substantially, possibly due to the extensive oxidation and ultrasonic irradiation.³³ The intensity ratio (I_D/I_G) of the D band to the G band is correlative with the average size of sp² domains. The I_D/I_G ratio of UG₁₂₀ is 2.73, and the I_D/I_G ratio of MG₁₂₀ is 2.75 (Supporting Information Figure S2), indicating that ultrasound is able to recover aromatic structure by repairing defects and increasing the number of smaller graphite domains,³⁴ but UG₁₂₀ always has quite a disorder degree. In addition, there is a significant change in the 2D band of UG₁₂₀ and MG₁₂₀ compared with graphite in Raman spectroscopy. The weak and broadened 2D peaks show that the UG₁₂₀ and MG₁₂₀ still possess some defects which can be further eliminated by annealing. Raman spectra, as well as our hitherto mentioned studies, suggest that less crystal structural change takes place in UG₁₂₀ compared with MG₁₂₀. UG₁₂₀ has a small quantity of the oxygen-containing groups, which usually lead to a little disordered structure existing in graphene; however, it is beneficial to dispersion of graphene and further modification for novel materials.

4. Conclusions

In summary, stable graphene dispersions with high concentration are prepared by chemical reduction of GO in the presence of ultrasonic irradiation, instead of the conventionally employed mechanical stirring. The self-assembly and aggregation of the graphene sheets into irregular shaped particles as occurred in the mechanical stirred system are suppressed due to the cavitation and vibration effect of ultrasound. The as-prepared graphene could be dispersed in water to form a dispersion which is useful for the further modification of graphene and preparation of novel materials. Ultrasonic irradiation can accelerate the process of chemical reduction of GO, and then a highly electrically conductive graphene with less damage in chemical and crystal structures could be obtained. In comparison with other methods for preparation of graphene dispersions, this process represents a conveniently and potentially scalable route, which enlarges the scope of application of graphene.

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Supporting Information Available: The position of the functional absorption peak of UG_{time} and MG_{time} in UV-vis spectra and Raman characteristics and fitting curves of the MG₁₂₀ and UG₁₂₀. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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