



Synthesis and Characterization of Graphene Single Sheets†

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Graphene sheet has been emerging as a fascinating material with its tantalizing applied foreground. A prerequisite for exploiting most proposed applications for graphene is to seek for a method that readily and simply produces graphene sheets in large quantities. By far, graphene sheets can be prepared by three techniques in general. Among them, oxidation and reduction processing graphene sheets is the most suitable for producing graphene sheets in large quantity in this review. In this report, XRD and FT-IR spectra and transmission electron microscopy and fluorescence microscope analysis indicated that graphene single sheets have been readily synthesized through the chemical processing. Moreover, the crystal structure of the graphene nanosheets was maintained intact after chemical functionalization.

Key Words: Graphene sheets, Oxidation and reduction processing method, Crystal structure.

INTRODUCTION

Graphene, as a single layer of carbon in a two dimensional (2D) lattice, shows many unique physical, chemical and mechanical properties. This unique structure holds great promise for potential applications in many technological fields such as nanoelectronics, sensors, nanocomposites, batteries, super capacitors and hydrogen storage.

Therefore, how to produce graphene sheets in large quantities has been a major obstacle to exploiting most proposed applications. Now graphene can be prepared by three techniques: (i) Micromechanical cleavage method. Novoselov *et al.*^{1,2} made graphene through this method in very limited quantities (ii) Epitaxial growth of graphene films. De Heer *et al.*³⁻⁵ on SiC crystal by heating, was complex on surface texture and it cannot be made into large and uniform thickness styles (iii) Oxidation and reduction processing method, as recently demonstrated by Ruoff *et al.*^{6,7} and Guoxiu Wang *et al.*⁸. They have successfully synthesized graphene sheets in large quantity *via* this oxidation and reduction approach involving, exfoliation and reduction. Therefore, among them, oxidation and reduction processing approach is the most suitable for bulk production of graphene sheets at low cost.

The solution-based route involves chemical oxidation of graphite to hydrophilic graphite oxide, which can be readily exfoliated as individual graphene oxide (GO) sheets by

ultrasonication in water. The resulting reduced graphene oxide (RGO) sheets were prepared by reacting graphene oxide sheets with polyoxyethylene lauryl ether and simultaneously reducing by hydrazine hydrate under hydrothermal conditions. However, during graphite oxidation stage, carboxylic acid will make graphene functional by drawing into some functional groups, such as C-OH and -COOH groups. These functional groups will do harm to crystal structure of graphene. Herein, we study on rapid synthesis of graphene single sheets with the crystal structure intact by oxidation and reduction processing method.

EXPERIMENTAL

Preparation of graphene oxide powder: About 2 g natural flake graphite powder mixed with oxidizing agent (mixture of 26 mL concentrated sulfuric acid (98 %) and 32 mL fuming nitric acid) in 3-neck round-bottom flask was dipped into constant temperature oil baths which is connected with refrigeration plant. Then 4 g potassium permanganate was added and then they were heated in Magnetism Miser. The reaction lasted for 6 h in the temperature of 75 °C then stop heating and reacts in room temperature for 24 h. The product was washed with deionized water until its pH is 5. Then the upper layer suspension was stripped for 6 h and graphene oxide solution was obtained. Finally, the product was washed with

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de-ionized water to remove the acid. The resultant aqueous dispersion was leached and dry for graphene oxide power.

Preparation of reduced graphene oxide power:

Graphene oxide power was reduced with hydrazine hydrate for 1 week at dispersant in existence. The reduced graphene oxide solution was obtained. Finally, the product was leached and dry for reduced graphene oxide power.

Structural and physical characterization of graphene oxide and reduced graphene oxide:

The structure of the pristine graphite and the graphene oxide were analyzed by X-ray diffraction (Rigaku D/max 2200P X-ray diffractometer). The graphene oxide and reduced graphene oxide were characterized by fluorescence microscope (Nikon E400), transmission electron microscopy and high resolution TEM (HRTEM) using a JEO JEM-2010 TEM facility. Fourier transform infrared spectra of graphene oxide and reduced graphene oxide were collected on a BRUKER RQUINOX55 FTIR spectrometer.

RESULTS AND DISCUSSION

XRD analysis: The oxidation made groups entered into the graphite layers so that lattice spacing of graphite oxide changed from 0.335 to 0.587 nm (Fig. 1). The lattice spacing of graphite oxide relates to the degree of oxidation.

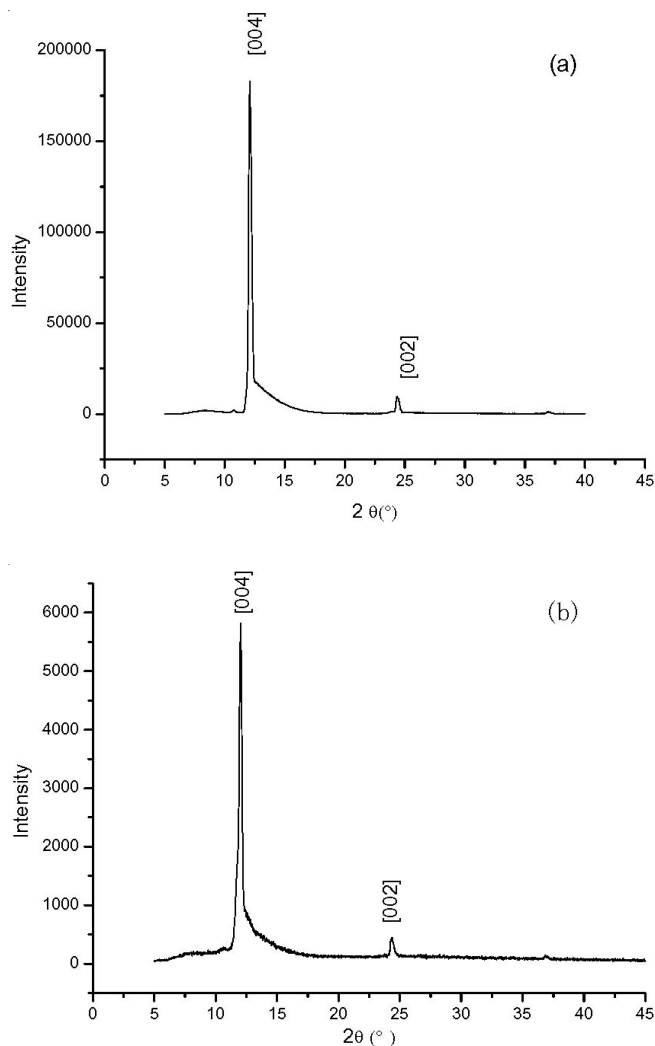


Fig. 1. XRD patterns of pristine graphite powder (a) and graphite oxide (b)

Optical microscope and TEM analysis: From Figs. 2 and 3 we can clearly see graphene oxide and reduced graphene oxide single sheets. They indicate that through the chemical processing method we have successfully synthesized graphene single sheets with the crystal structure intact at low cost.

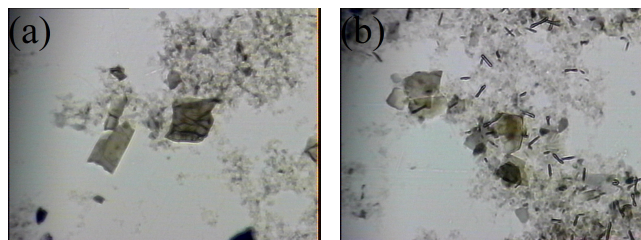


Fig. 2. FM of graphene oxide (x 400)

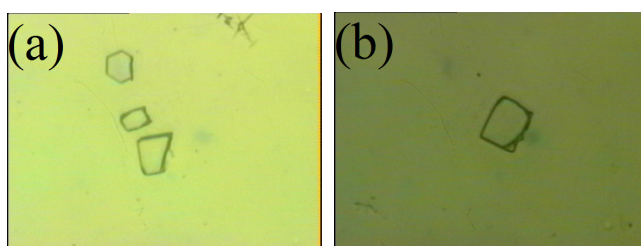


Fig.3 FM of reduced graphene oxide (x 400)

TEM and HRTEM analysis on graphene oxide and chemically converted graphene were also performed in water. From Figs. 4 and 5, it was clearly seen that chemically converted graphene and graphene oxide sheets. Fig. 6 illustrates that the graphitic lattice is clearly in HRTEM image. It demonstrates that chemically converted graphene was maintained the crystal structure intact after chemical modification. The interplanar distance was measured to be 0.45 nm, corresponding to the spacing of the planes.

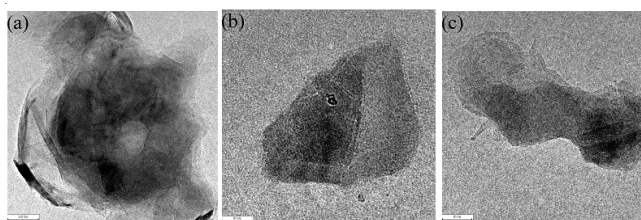


Fig. 4. TEM of graphene oxide

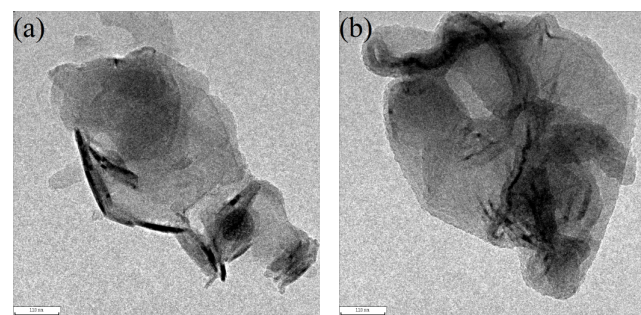


Fig. 5. TEM of reduced graphene oxide

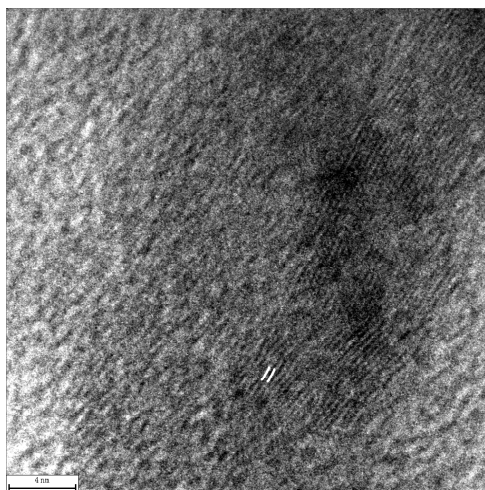


Fig. 6. High resolution image of lattice fringe

Infrared spectroscopic analysis: As shown in Fig. 7, reduction reaction with hydrazine hydrate solution made groups of graphene oxide, such as hydroxyl group (3350 cm^{-1}), carboxyl group (1637 cm^{-1}) and epoxy group (1085 cm^{-1}) decreased obviously.

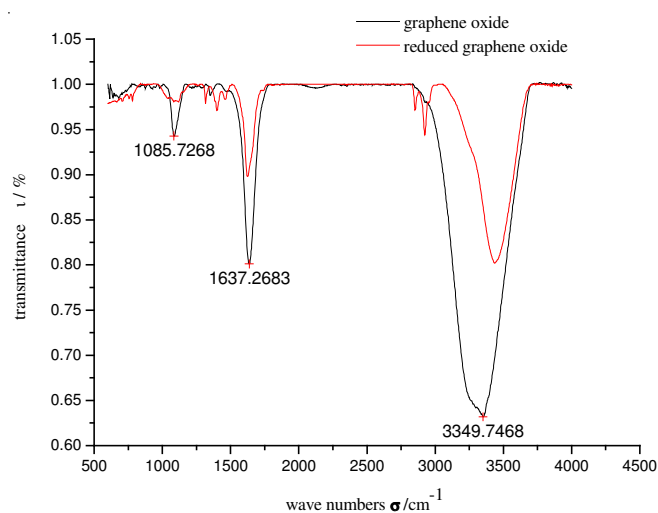


Fig. 7. FT-IR spectra of graphene oxide membrane and reduced by hydrazine hydrate solution

Conclusion

We have demonstrated chemical processing method to be a valuable approach to synthesize graphene single sheets with the crystal structure intact at low cost. The ease of synthesis and the whole of crystal structure make this inexpensive graphene attractive not only for future nanoelectronics, but also for large-scale applications in both conventional technological fields, such as transparent, antistatic coatings and electrochemical devices and emerging areas such as flexible/transparent electronics, high-performance nanocomposites, nanomedicines and bionic materials. It is believed that the work presented here will provide a significant step forward to bringing graphene materials much closer to real world applications.

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