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Featured Letter

A novel and efficient approach to prepare few-layer graphene with high quality

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ABSTRACT

Herein, we proposed a novel approach to prepare the graphene with high quality, which was realized by exfoliating the KOH-etched graphite in H_2O_2 solution under microwave irradiation. The as-prepared graphene exhibits a smooth and transparent feature, and possesses high electrical conductivity of up to 2×10^5 S m^{-1}. Compared with the conventional oxidation-reduction method, the present method is significantly efficient, and, most importantly, does not involve the use of acid and strong oxidizing agent, which will make it more competent in the scalable production of graphene.

1. Introduction

Owing to its unique properties, graphene has become the most focused nanomaterial of the past decade [1,2]. Up to now, many approaches have been developed for preparing graphene, such as micromechanical exfoliation of graphite [3], oxidation-reduction of graphite [4] and liquid-phase exfoliation of graphite [5–7], etc. Among them, the oxidation-reduction is the commonly used method for the industrial production of graphene at present. However, this method involves the use of strong oxidizing agent and strong acid, thereby leading to the structural damage and the inferior quality of graphene as well as the environmentally detrimental impact caused by the effluent of the waste acid.

It is well known that KOH-etching to natural graphite can lead to a larger distance between the graphite layers due to the K ion intercalation [8,9]. It can be anticipated that the larger gallery of graphite interlayers will facilitate the incorporation of other chemically instable species among the graphite layers and thus realize the exfoliation of graphite. Based on the supposition, herein, we proposed a novel and efficient route to prepare graphene. According to this method, natural graphite (NG) is firstly etched by KOH. Subsequently, the resultant KOH-etched graphite is exfoliated into graphene in H_2O_2 solution under 1000 W microwave irradiation (purchased form Midea, China), The resultant graphene exhibits a typical transparent feature and superior electrical conductivity.

2. Experimental

2.1. Sample preparation

High purity natural graphite (NG) and KOH were mixed at weight ratio of 1:10 and ground into a powder mixture. Then the mixture was placed in a horizontal quartz tube furnace and heated to 800 °C at a rate of 10 °C min^{-1} under argon at atmospheric pressure. This treatment was lasted for 1 h. Thereafter, the treated mixture was cooled down to room temperature and washed until pH = 7 and dried, thereby obtaining the KOH-etched graphite. Finally, the etched graphite was dispersed in H_2O_2 solution under ultrasonication for 3 min and then subjected to microwave irradiation at 1000 W for 90 s (commercial EMA34GTQ-SS microwave oven with controlled timer and intensity control, provided by Midea, China), eventually obtaining the graphene.

2.2. Characterization

X-ray diffraction (XRD) powder patterns of the samples were taken on Rigaku D/MAX-2500 powder diffractometer with Cu-Kα radiation (λ = 0.154 nm) operated at 40 kV and 200 mA. Raman spectra were obtained using a Renishaw inVia Raman microscope with a 532 nm laser wavelength. The morphologies of the samples were observed using a Hitachi S-4800 field emission scanning-electron microscopy (SEM) at 15 kV. Transmission electron microscopy (TEM) was performed with a JEOL JEM-2010 at 200 kV. Atomic force microscopy (AFM) was utilized by a DI Nanoscope

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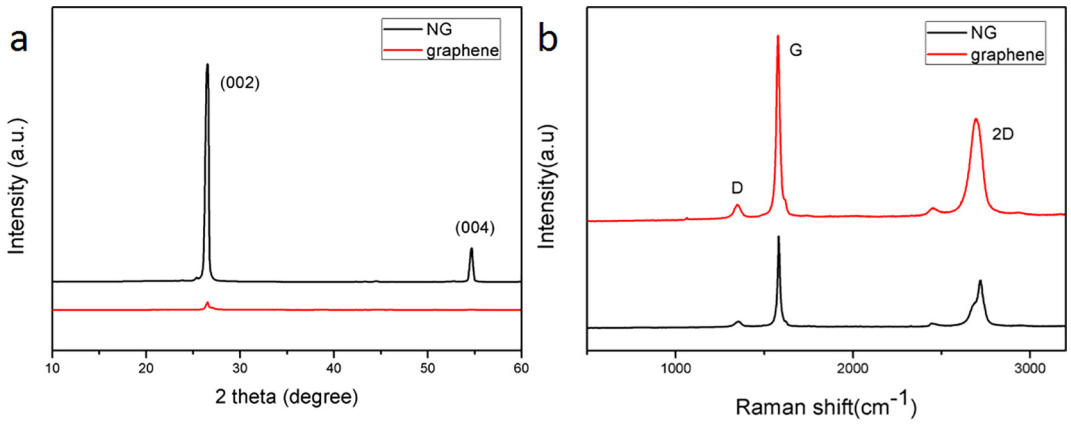

Fig. 1. (a) XRD patterns of graphene and NG; (b) Raman spectra of graphene and NG.

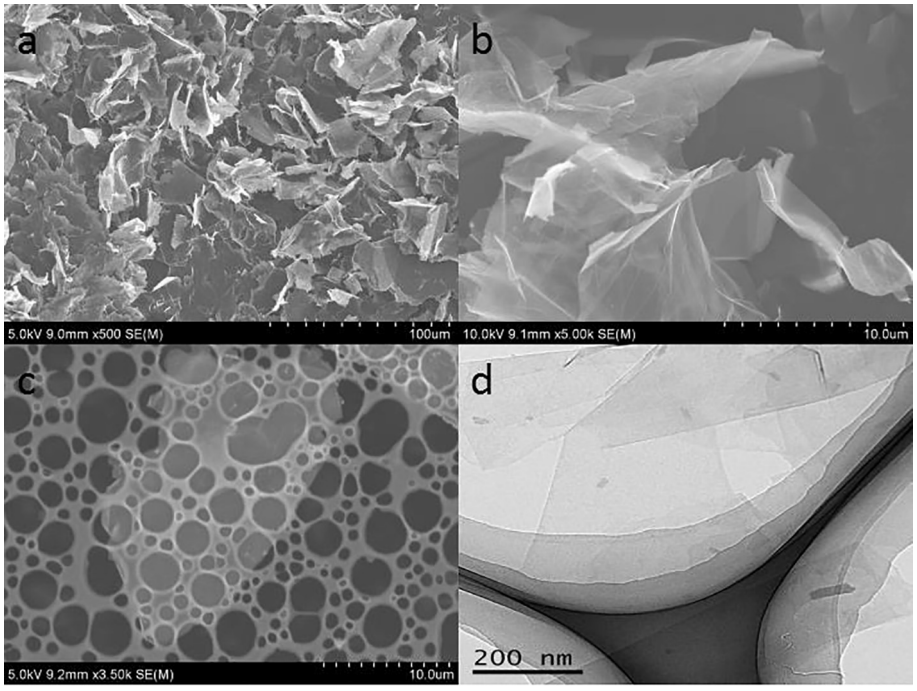

Fig. 2. Images of as-prepared graphene: (a–c) SEM; (d) TEM.

IV (Veeco, USA) microscope with the scan rate of 1 Hz. X-ray photoelectron spectroscopy (XPS) data were recorded on a K-alpha (Thermo VG Corporation, USA) with Al-Kα radiation (1486.6 eV, 15 kV). In addition, the electrical conductivity of the graphene sample was measured by using the standard four-point probe technique (The detailed measurement description is provided in Supporting Information).

3. Results and discussion

Fig. 1a shows XRD patterns of NG and graphene samples, respectively. Two diffraction peaks – (002) and (004) can be observed on both the XRD patterns at the same positions, suggesting that the graphite lattice is retained from graphite to graphene [10]. It is noteworthy that, the relative intensity ratio of (002) and (004) reflections for NG and graphene samples are 28.27 and 70.61, respectively. The markedly lower intensity ratio for the graphene can be considered as an indication of positional disorder in the graphene nanosheet caused by the exfoliation of the graphite layers [11], demonstrating that the as-prepared graphene has few layers in the stack. Fig. 1b shows the Raman spectra of NG and graphene, respectively. Compared with those of NG, the intensities of D- and 2D-band in the Raman spectrum of graphene become obviously higher. This phenomenon was also observed in the graphene obtained by liquid exfoliation of graphite [6], suggesting that the obtained graphene has a structure with few layers in the stack. In addition, the almost symmetrical and single 2D peak centered at 2692 cm^{-1} further demonstrates the few-layer feature [12]. The calculated intensity ratio (I_{2D}/I_G) of 2D (2692 cm^{-1}) and G (1578 cm^{-1}) bands is 0.591, suggesting that the average layer number is ~5 [13]. The few-layer feature of the as-prepared graphene is further identified by AFM measurement (Fig. S1, Supporting Information). Approximately ~7% of the exfoliated sheets are single-layered and ~93% of them are few-layered graphene sheets (layer number <10 layers) with an average layer number of ~5. Furthermore, the peak intensity ratio (I_D/I_G) of D and G bands is calculated as 0.102, indicating the less defect and