



A novel fabrication of graphene by chemical reaction with a green reductant



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HIGHLIGHTS

- Graphene was successfully prepared via the reduction of graphene oxide used a green reductant.
- Reduction degree of graphene is mainly controlled by temperature and reaction time.
- The mechanism of the reduction of GO by thiourea dioxide was attributed to catalyzing the ring-opening reaction.

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ABSTRACT

In this study, the graphene was successfully prepared via the reduction of graphene oxide (GO) used thiourea dioxide as a new reductant. The graphene oxide after reduction with thiourea dioxide has been characterized by using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FT-IR), X-ray photoelectron spectroscopy (XPS), Raman photoelectron spectroscopy and transmission electron microscopy (TEM). The factors affecting reduction degree of graphene were discussed in the process of reduction, such as reduction time, temperature and the amount of reducing agent. The result shows that reduction degree of graphene is mainly controlled by temperature and reaction time. Finally, it is founded that the stronger reduction capacity of thiourea dioxide was attributed to the synergistic effect of its hydrolysis products, such as HSO_3^- and catalyze the ring-opening reaction.

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1. Introduction

Graphene, a one atomic layer thick carbon atomic layer with sp^2 C—C double-bond and π -electron cloud, has attracted significant attention in potential applications such as supercapacitors [1], gas sensors [2], transparent conductors [3] etc., owing to its excellent properties such as high mechanical strength (~ 125 GPa) and high Young's modulus (~ 1.1 TPa) [4], high thermal conductivity (~ 5000 W/m·K) [5], high intrinsic mobility (2×10^5 $\text{cm}^2/\text{V}\cdot\text{s}$) [6] and its optical transmittance ($\sim 97.7\%$) [7]. Up to now, tremendous efforts, include micromechanical, epitaxial growth, ultrasonic exfoliations of graphite, chemical vapor deposition (CVD), chemical reduction of graphene oxide etc. [8], have been made to develop synthetic methods for graphene. However, compared to all methods mentioned above, the chemical reduction of graphene oxide (GO) was considered as the most promising technique to make graphene in large scale production due to its low-cost and simple.

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Unfortunately, some chemical groups, such as carboxyl ($-\text{COOH}$), carbonyl group ($\text{C}=\text{O}$), hydroxyl group ($-\text{OH}$) and epoxy group ($-\text{CH}(\text{O})\text{CH}-$) are liable to be introduced during the process of oxidation, which decrease the properties of the resulting graphene. Therefore, how to choose reductant plays a crucial role to obtain high quality graphene. As is well known, reducing agents like hydrazine and related derivatives, are always used during the reduction process of GO. But, hydrazine and hydrazine hydrate are highly poisonous and explosive. Hence, a considerable number of studies have sought alternative reagents [9–13]. More recently, a study by Md Moniruzzaman Sk et al. [14] reported that vitamin C could be used as an alternative to hydrazine and has the same ability as hydrazine in the deoxygenation of GO.

Thiourea dioxide (TD, $\text{CH}_4\text{N}_2\text{O}_2\text{S}$), which is also called formamidinesulfinic acid, has been widely used as a strong reductant in the textile printing, paper, photographic and leather processing industries for a long time because of its good and special characteristics, such as safety, nontoxicity [15,16]. Herein, we investigate the preparation method of graphene with a green and friendly TD reducing agent. The influences of dosage, reaction time and temperature of TD were carefully reported. Finally, the possible mechanism of the reduction of GO by TD was also discussed.

2. Materials and methods

2.1. Raw materials

In this work, the raw materials used were natural flake graphite powder (200 mesh, 99.90% pure), which were purchased from Nanjing Xian Feng Nano Materials Technology Co. Ltd. Jiangsu, China. The other reagents, such as NaNO_3 , concentrated H_2SO_4 (98%), HCl (36–37%), KMnO_4 , H_2O_2 (30%), NaOH (pH = 11), thiourea dioxide and so on, were all obtained from Sinopharm Chemical Reagent Co. Ltd. Shanghai, China. All chemicals were used directly without further purification.

2.2. Preparation of the GO

As depicted in Fig. 1, the hummer's method was employed to synthesize graphene oxide. In this work, a typical experiment is carried out as follows: (a) 5 g of graphite powder was mixed with 2.5 g of NaNO_3 and 150 mL of concentrated H_2SO_4 was stirred at ice-bath environment for 30 min. (b) 30 g of KMnO_4 was divided into three times to be added slowly into the mixture and held for 120 min. (c) Raise to 35 °C and hold it for 3 h. (d) Raise to 90 °C and then 200 mL of purified water was added gradually into the system and stirred for additional 30 min. The mixture was filtered under vacuum and washed with HCl and deionized water for many times (≥ 20 times), and then dried at 40 °C for 48 h by vacuum freeze-drying.

2.3. Preparation of the graphene

The graphene was prepared by reduction of the GO with a new green reducing agent thiourea dioxide. A typical experiment is carried out as follows: at first, 20 mg of GO was dispersed into 40 mL of water under the assistant of powerful ultrasound for 2 h to obtain a homogeneous GO suspension solution (0.5 mg/mL). At the same time, thiourea dioxide and NaOH was mixed by 1:3 (vol.%). Then, thiourea dioxide and NaOH mixture was gradually added into the GO solution with agitation. Finally, the obtained mixture solution was separated by high speed centrifugation (10,000 rpm) and washed with ultrapure water for many times (≥ 30 times). Subsequently, graphene was dried at 40 °C for 48 h by vacuum freeze-drying.

2.4. Analytical techniques and characterization

XRD analyses were performed on a Bruker D8 Advance diffractometer with Cu K_α radiation at 40 kV and 15 mA. The patterns were recorded in the range of $2\theta = 5\text{--}60^\circ$ with a scan speed of $8^\circ/\text{min}$. Raman spectroscopy with a Micro Raman LabRAM VIS-633 with a He-Ne laser light (632.8 nm) over the range of $500\text{--}3500\text{ cm}^{-1}$. X-ray photoelectron spectroscopy (XPS) measurements were performed at room temperature on an AXIS ULTRA after plasma cleaning for 300 s in the XPS chamber at a pressure of around 4×10^{-8} torr. The XPS spectra were fitted using the XPS peak 4.1 software in which a Shirley background was assumed. Fourier transform infrared (FT-IR) spectra of graphene oxide and

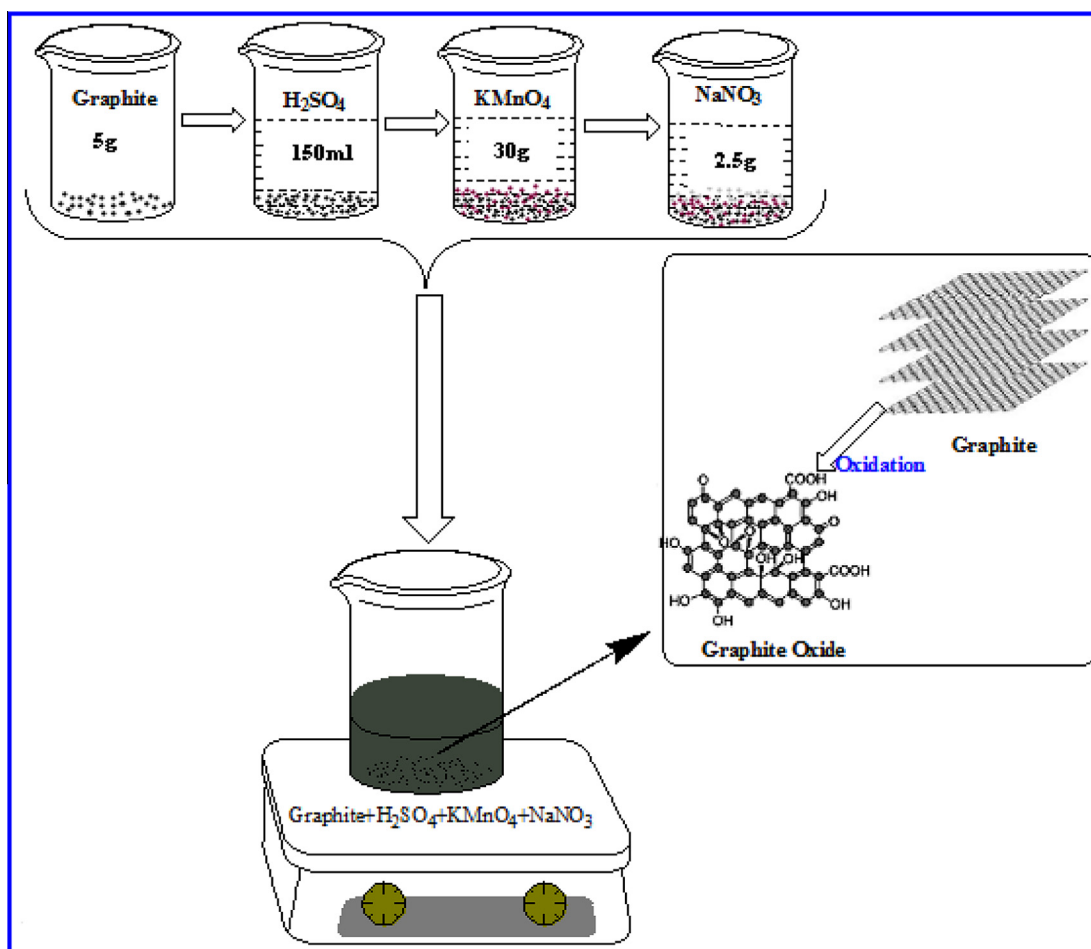


Fig. 1. The schematic procedure for preparation GO.

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