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Green synthesis and characterization of graphene nanosheets

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ABSTRACT

For the first time, we have successfully synthesized graphene nanosheets in the presence of pomegranate juice. In this approach, pomegranate juice was used not only as reductant but also as capping agent to form graphene nanosheets. At first, the improved Hummer method to oxidize graphite for the synthesis of graphene oxide (GO) was applied, and then the as-produced graphene oxide was reduced by pomegranate juice to form graphene nanosheets. Fourier transformed infrared (FT-IR), X-ray powder diffraction (XRD), scanning electron microscopy (SEM), energy dispersive spectrometry (EDS), high resolution transmission electron microscopy (HRTEM), atomic force microscopy (AFM) and raman were used to characterize the samples. The results obtained from the characterization techniques proved high purity of the final products.

1. Introduction

Graphene, a single-atom-thick sheet of hexagonally arrayed sp^2-bonded carbon atoms, promises a diverse range of applications from composite materials to quantum dots [1,2]. Since the discovery of single-layer graphene in 2004, many research groups have developed various methods of synthesizing single-layer graphene, including mechanical exfoliation [3], chemical vapor deposition [4], liquid phase exfoliation [5–8] and epitaxial graphene grown on SiC as a major approach to synthesize single layer graphene [9]. Graphene possesses a unique two-dimensional structure, high conductivity, superior electron mobility and extremely high specific surface area, and can be produced on a large scale at low cost [10].

Recent progress has shown that the graphene can have a profound impact on electronic and optoelectronic devices, chemical sensors, nanocomposites and energy storage [11,12]. At present, chemical conversion of graphite to graphite oxide has emerged to be a viable route to afford graphene-based single sheets via one of three principal methods developed by Brodie [13], Hummers [14], and Staudenmeier [15], respectively. It still retains a layered structure. This reaction occurred by using oxidant including sulfuric acid, nitric acid and potassium permanganate [14]. Compared to pristine graphite, graphene oxide (GO) is highly hydrophilic due to hydroxyl, epoxy, carbonyl and carboxyl groups, so yielding stable dispersion in water. Li et al. showed that the surface charges on graphene oxide are highly negative when dispersed in water by measuring the zeta potential due to the ionization of the carboxylic acid and the phenolic hydroxyl groups. Therefore, the formation of stable graphene oxide colloids in water was attributed to not only its hydrophilicity but also the electrostatic repulsion [16]. Chemical reduction of graphene oxide sheets has been performed with several reducing agents including hydrazine [17–20], and sodium borohydrate [21,22], hydroquinone [23], gaseous hydrogen (after thermal expansion) [24], and strongly alkaline solutions [25]. Thermal reduction is another approach to reduce GO to reduced graphene oxide that utilizes the heat treatment to remove the oxide functional groups from graphene oxide surfaces [26,27].

Recently, great efforts were made to use green and environmentally friendly methods for the synthesis of nano-sized materials. These efforts involve the use of plant or fruit extracts as stabilizer and capping agent to control crystal growth [28]. Here, for the first time, we used pomegranate juice as a natural reducing agent to synthesize graphene nanosheets from GO. This work demonstrates a green and economic method for the preparation of graphene nanosheet.

2. Experimental

2.1. Materials and characterization

All the chemicals used in this work were reagent grade and obtained from commercial suppliers. Powder X-ray diffraction

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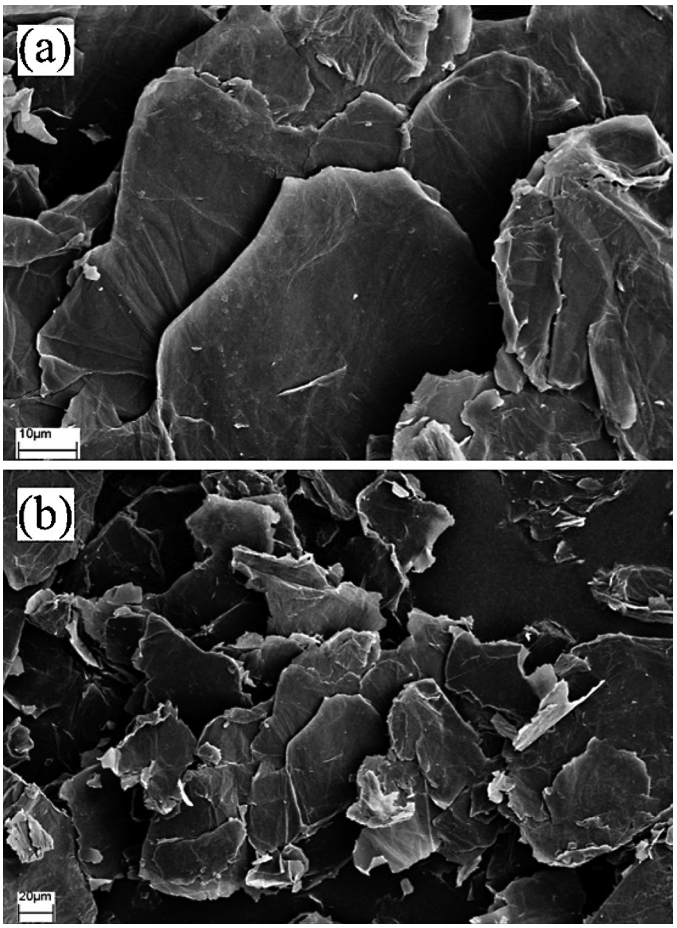

Fig. 1. SEM images of the produced graphene oxide nanosheets.

(XRD) patterns were collected from a diffractometer of Philips Company with X'PertPro monochromatized CuKα radiation (λ = 1.54 Å). A Cam Scan MV2300 scanning electron microscope (SEM) was used to investigate the morphology of the products. The energy dispersive spectrometry (EDS) analysis was studied by XL30, Philips microscope. The Fourier transform infrared spectra were performed using KBr pellets on FT-IR spectrometer (Magna-IR, 550 Nicolet) in the range of 400–4000 cm^{-1}.

2.2. Synthesis of graphene oxide (GO)

In this work, the improved Hummer method to oxidize graphite for the synthesis of GO was applied [29]. First, 3 g of graphite powder, 18 mL of hydrogen nitrate (67–70%), and 46 mL of sulfuric acid (98%) were mixed and strongly stirred in the range of 0–5 °C for 15 min in a 500 mL reaction flask immersed in an ice-water bath. Then 6 g of potassium permanganate was added slowly to the above solution within 15 min. After this, the suspended solution was stirred continuously for 2 h in an ice-water bath and maintained the temperature in the range of 10–15 °C, and then the suspended solution was stirred continuously at 35 °C for half an hour. Subsequently, 138 mL of distilled water was added slowly to the suspension within 10 min, and then the temperature was kept in the range of 95–98 °C for 30 min. Immediately, the suspension was diluted by 200 mL of warm distilled water (40 °C) and treated with 18 mL of H_2O_2 (30%) to reduce residual permanganate to soluble manganese ions. Finally, the resulting suspension was filtered, washed with distilled water and dried in a vacuum oven at 60 °C for 24 h to obtain GO.

2.3. Synthesis of graphene nanosheets from GO

In a typical procedure, 0.1 g of GO was loaded into a 250 mL beaker, and then 40 mL of pomegranate juice was added, yielding an inhomogeneous dispersion. This dispersion was then stirring for 12, 18, and 24 h, respectively. After stirring process, the above dispersion was sonicated for 60 min and finally dried in vacuum oven for 24 h at 60 °C.

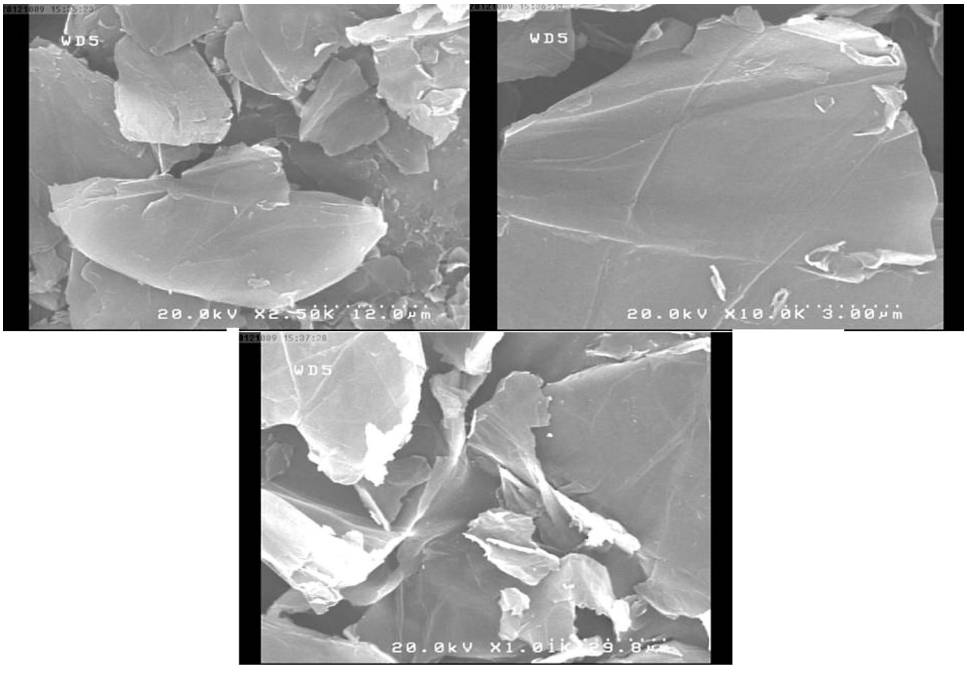

Fig. 2. SEM images of the produced graphene nanosheets after stirring for 12 h.