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Research Paper

Preparation of graphene via liquid-phase exfoliation with high gravity technology from edge-oxidized graphite

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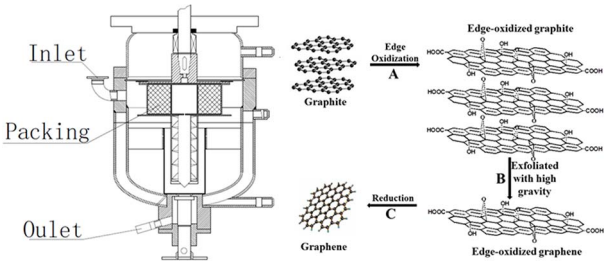
GRAPHICAL ABSTRACT

The process of preparing graphene from edge-oxidized graphite in internal rotating packed bed.



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ABSTRACTS

In this paper, Rotating Packed Bed (RPB) is described to prepare graphene from edge-oxidized graphite (EOG). The concentration of graphene aqueous dispersion can reach 1.00 mg ml − 1 for a long time by the characterization of UV–visible spectrophotometer and the graphene aqueous dispersion can stabilize at room temperature for several months. Significantly, it is verified that the layer numbers of the graphene (R-GN) prepared by the new method is mostly 3–4 layers, the lateral size is double compared to that prepared by ultrasonic method (S-GN). Moreover, it has less defect, specific capacitance can reach 102 F g − 1 at the sweep rate of 20 mV s^{-1}, and the capacitance retention hold 87% (after 10000 cycle numbers) at the sweep rate of 400 mV s^{-1}. These results indicate it is a green and efficient way to prepare graphene.

1. Introduction

Graphene has received tremendous interest since it was first discovered in 2004, owing to its unique two-dimensional atomic crystal structure, remarkable physical and chemical properties [1–3]. Graphene have a great potential applications in many fields, including composite materials [4–7], electronic devices, energy storage and molecular sensors [8,9]. To date, micromechanical cleavage [10], chemical vapor deposition [11], epitaxial growth [12], liquid-phase exfoliation [13,14] and chemical exfoliation (oxidation/reduction) [15] and their variation [16] have been applied to produce graphene. The micromechanical cleavage can prepare mono- and few-layers graphene with high quality [17]. However, it is difficult to be scaled up. Epitaxial growth also suffers from two major drawbacks of high cost and high temperature. Alternatively, the chemical vapor deposition (CVD) can produce graphene over a large area and with a relatively small number of defects [10]. Nevertheless, the processes employed are complex, costly and require harsh conditions. At present, liquid-phase oxidation reduction method is used to prepare graphene in larger scale [14]. In this process, graphite oxides as an important intermedium have a bigger interlayer spacing and weak Van der waals between sheets. Thus graphite oxides are more likely to be exfoliated. The methods of preparing

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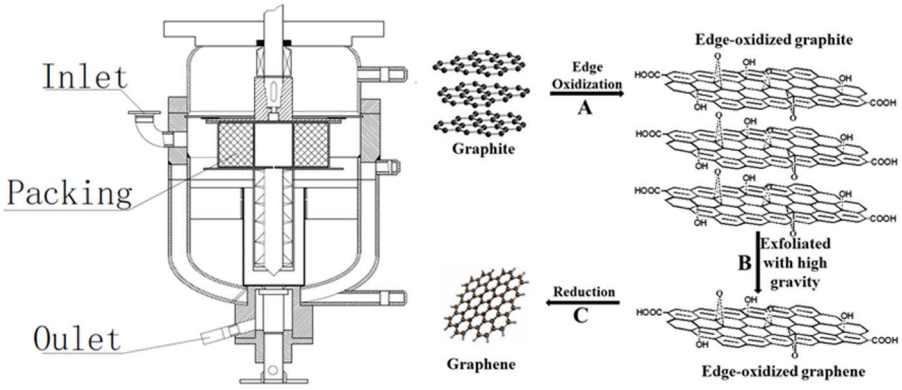

Fig. 1. The exfoliation in Schematic diagram of internal rotating packed bed.

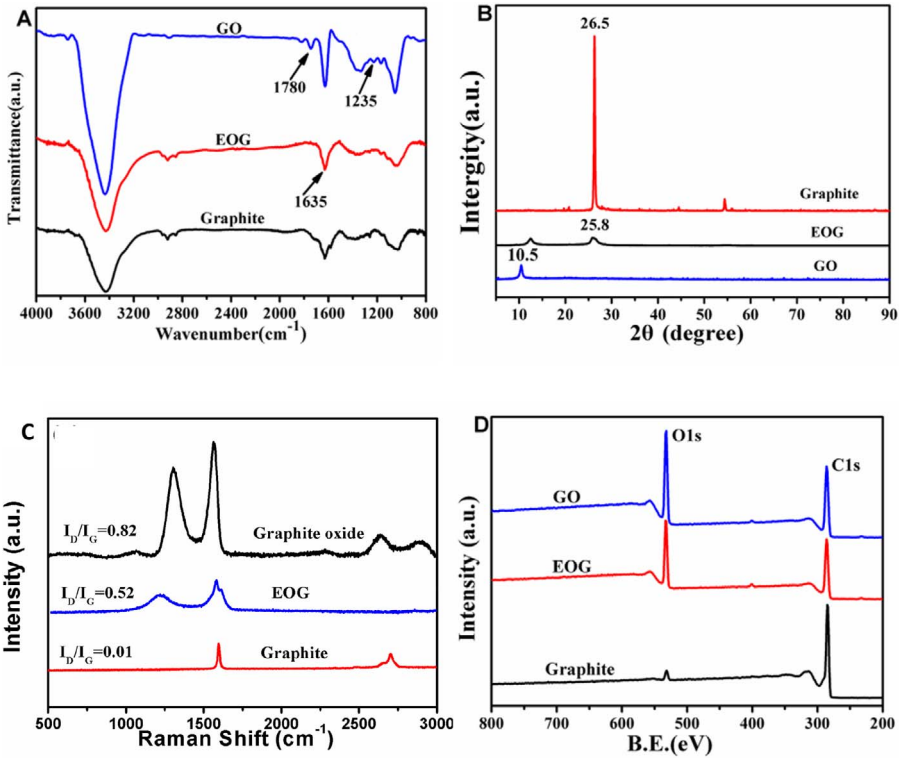

Fig. 2. (A) FTIR spectra of natural graphite, EOG and GO, (B) XRD spectra of natural graphite, EOG, GO, (C) Raman spectra of natural graphite, EOG and GO, (D) XPS survey spectra ofnatural graphite, EOG and GO.

graphite oxide commonly used at present time are the traditional Brodie's method [18,19], the modified Hummer's method [20], staudenmaier's method [21] or their variations [22–25]. These methods have a tremendous potential in large scale production. However, the product also has lower quality due to plentiful defects. Liquid-phase exfoliation of graphite gives the possibility of large-scale production of graphene with high quality [26–32], in which ultrasonic exfoliation method was mainly chosen. However, there are two issues need to be solved. Firstly, graphite can be stabilized in organic solvents, polymer and aqueous surfactant solutions [33]. However, they are usually unenvironmental friendly. And it is difficult to be exfoliated due to small interlayer spacing of graphite. Secondly, sonication is a relatively harsh process which can produce high local temperature (~several thousand K), extreme pressure (~several thousand atm), and rapid heating/cooling rates (~several billion K/s) [34]. These harsh conditions could result in damage to the graphene. Indeed, Polayakova et al. [35] have experimentally evidenced that graphene produced by sonication truly has more oxides and defects than expected. Therefore, finding an environmental friendly method with higher efficiency and fewer defects to prepare graphene is the critical issue.

Recently, some literatures have proved that layered materials can be exfoliated for preparing two-dimensional sheets in the high shear environment. These devices include rotor-stator high shear mixers [36] or kitchen blenders [37]. Its advantage is higher production rates and high quality. High gravity technology (HGT) is generated by the Rotating packed bed (RPB) to create a centrifugal field of gravity environment. It can intensify mass transfer and micro-mixing and create strong, stable and changeable shearing force. We had used it to exfoliate graphite oxide and MoS_2 to prepare graphene oxide [38] and two-dimensional MoS_2 [39]. However, preparing graphene with RPB mainly utilizing its shearing force was seldom reported.

In this paper, we demonstrate RPB as a scalable alternative to ultrasonic method and combine the liquid-phase exfoliation to prepare edge-oxidized graphene (EOGN) from edge-oxidized graphite (EOG), which has not been destroyed the sp^2 character of the carbon basal planes except edges. The EOGN can be stably dispersed in water with a concentration of 1.00 mg ml^{-1} for a long time. Compared with the Brodie method and modified hummer's method, this method shows decreased demand for $KMnO_4$, time saving, and environmental friendly, more importantly, the obtained product has a higher quality compared with that prepared by ultrasonic exfoliation from EOG.