Materials Chemistry and Physics 221 (2019) 47–57

Contents lists available at ScienceDirect

Materials Chemistry and Physics

journal homepage: www.elsevier.com/locate/matchemphys

Preparation and adsorption property of graphene oxide by using waste graphite from diamond synthesis industry

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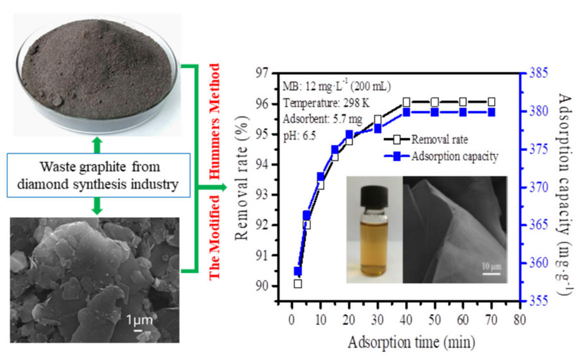
HIGHLIGHTS

- Graphene oxide (GO) was prepared from waste graphite from diamond synthesis industry.
- Structure of GO was a bit different from those prepared from natural flake graphite.
- As-prepared GO shows better adsorption performance for MB than most traditional GO.
- Main mechanism for adsorption of MB onto GO was electrostatic force between charges.

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Waste graphite
Graphene oxide
Preparation
Adsorption
Methylene blue

ABSTRACT

Graphene oxides (GO) were prepared from waste graphite from diamond synthesis industry by using the modified Hummers method. The phase composition, microstructure, specific surface area and pore size distribution of the waste graphite and the as-prepared GO samples were characterized by X-ray diffraction, Fourier transformed infrared spectra, Ultraviolet visible spectroscopy, Raman spectroscopy, field-emission scanning electron microscopy and nitrogen adsorption-desorption tests. The adsorption property of GO to methylene blue (MB) was investigated. The results show that the as-prepared GO has few layers, specific surface area of $116.6\ m^2\ g^{-1}$ and a little different structure from those prepared by using natural flake graphite. It exhibited excellent adsorption performance in MB aqueous solution. Adsorption time, GO dosage, initial concentration, and pH value of MB aqueous solution play important roles but the solution temperature had a little effect on the removal rate of MB and the adsorption capacity of GO. Under the condition of a GO dosage of 5.7 mg, room temperature, initial concentration of $12\ mg\ L^{-1}$, pH value of 6.5 and adsorption for 40 min, the removal rate of MB and the adsorption capacity of GO reached 96.05% and $379.89\ mg\ g^{-1}$, respectively. The adsorption process of MB onto GO met well with a pseudo-second-order kinetic model and Langmuir adsorption isotherm model. The adsorption reaction was spontaneous and purely based on endothermic process.

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https://doi.org/10.1016/j.matchemphys.2018.09.036
Received 18 March 2018; Received in revised form 5 September 2018; Accepted 8 September 2018
Available online 10 September 2018
0254-0584/

1. Introduction

Due to high hardness, good thermal conductivity, high optical transparency, high electrical mobility and other prominent characteristics, synthetic diamond is widely used as abrasives, cutting and polishing tools, optical materials and electronic devices, etc. In general, high pressure and high temperature (HPHT) synthesis is the most commonly used method for diamond synthesis, and graphite with a purity of 99.99% [1] is used as carbon source. Nevertheless, most graphite is not transformed to diamond but remains in tailing, whose component includes graphite, catalytic alloy, diamond, etc. In theory, the graphite in tailing could be recycled for diamond synthesis again. However, the transformation rate and quality of diamonds synthesized from the graphite in tailing are not as high as those synthesized from natural flake graphite. Although a small amount of the graphite in tailing is used to prepare low-value materials, most of it is stockpiled as solid waste, which not only pollutes the environment but also wastes the graphite resources. Consequently, it is meaningful to develop high value-added products from waste graphite in diamond synthesis industry.

With the rapid development of industry, water pollution caused by dye becomes particularly serious [2], which is harmful to human health and the growth of aquatic organisms. Hence, many wastewater treatment techniques, such as co-precipitation, ion exchange and adsorption [3,4], have been carried out to purify wastewater. Due to the simpleness and effectiveness, adsorption method has been widely applied in wastewater treatment up to now. It is well known that adsorbent plays an important role in the adsorption process. Traditional adsorbents include activated carbon [5], zeolite [6] and natural clay [7]. However, each of them has disadvantages of one sort or another, such as high cost of processing, low efficiency, low renewable performance, and so on. Consequently, it is necessary to explore new adsorbents which are inexpensive, high efficiency, etc.

Graphene is a crucial 2-Dimensional material with excellent electronic, thermal, optical, mechanical and absorptive properties. Therefore, it has the potential to be widely applied in the fields of electronic information, new energy, chemical industry, environmental remediation, etc. Although graphene can be prepared by using waste biomass [8], the conventional raw material is natural flake graphite with high purity and high degree of crystallinity [9,10]. Oxidation-reduction method [11] is commonly used to prepare graphene. However, neither monolayer nor few layers graphene can be obtained easily and conveniently by using this method [11]. Moreover, most reduction agents [12,13] are toxic [14,15] and will pollute the environment once discharging into the aquatic environment. All the traditional methods mentioned above increase the cost and risk of graphene preparation. On the other hand, graphene oxide (GO), as the precursor of graphene and a graphene derivative, is rich in oxygen containing functional groups and thus has good dispersibility in aqueous media [16]. Furthermore, GO possesses large specific surface area and adjustable pore size distribution. Thus, it has been directly used for adsorption of organic dye [17–19] and metal ions [20–22]. The modified GO [23,24], GO/Fe_3O_4 composite [24,25] and attapulgite/graphene oxide [26] have exhibited excellent adsorption performance for heavy metal ions or organic compound, and quick separation efficiency.

China has been the biggest synthetic diamond producing country in the world since 2000, and produced 14 billion karat synthetic diamond in 2012 [27]. As a result, a large amount of graphite has been excluded and discarded from synthetic diamond industry in China. On the other hand, exploring new and high-efficiency adsorbents for wastewater treatment is urgently necessary due to the severe water pollution in China. Therefore, GO were prepared from waste graphite from diamond synthesis industry by the modified Hummers method in this study. The adsorption behavior of GO for methylene blue (MB) was explored at different adsorption times, temperatures, adsorbent dosages, initial concentrations and pH values. The adsorption process was discussed with pseudo-first-order kinetic model, pseudo-second-order kinetic model, Langmuir isotherm model, Freundlich isotherm model, etc.

Table 1
EDS analysis of the as-processed WG powder.

element	C	O	Fe	Co	Ni	Total
weight/%	89.15	7.29	1.81	1.42	0.33	100.00
atom/%	93.22	5.67	0.40	0.64	0.07	100.00

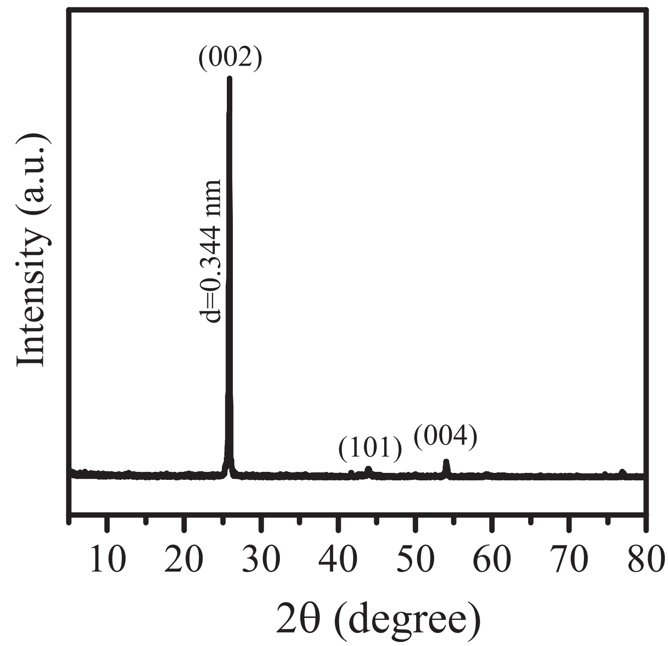

Fig. 1. XRD pattern of the as-processed WG powder.

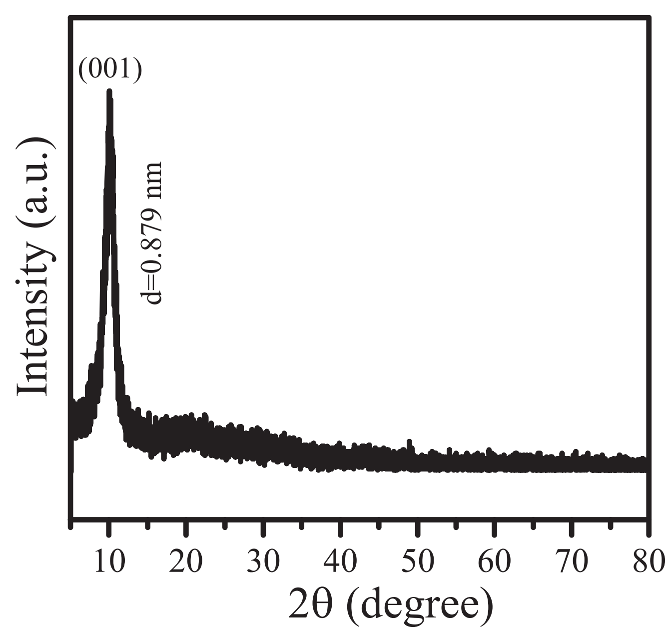

Fig. 2. XRD pattern of GO sample.

2. Experimental

2.1. Materials and chemicals

The original tailing used in this study was discharged from synthetic