Bioresource Technology 196 (2015) 355–363

Contents lists available at ScienceDirect

Bioresource Technology

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

Preparation and characterization of a novel graphene/biochar composite for aqueous phenanthrene and mercury removal

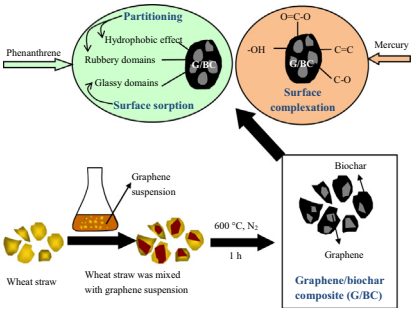
Jingchun Tang [1], Honghong Lv [1], Yanyan Gong *, Yao Huang

Key Laboratory of Pollution Processes and Environmental Criteria (Ministry of Education), Tianjin Engineering Center of Environmental Diagnosis and Contamination Remediation, College of Environmental Science and Engineering, Nankai University, Tianjin 300071, China

HIGHLIGHTS

- G/BC was synthesized by slow pyrolysis of graphene treated wheat straw.
- Adsorption performance of G/BC for contaminants was significantly improved.
- Pseudo second-order model adequately simulated sorption kinetics.
- Dual-model isotherm model adequately represents phenanthrene sorption isotherm.
- Isotherm data of mercury were simulated perfectly by BET model.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 29 May 2015
Received in revised form 12 July 2015
Accepted 15 July 2015
Available online 21 July 2015

Keywords:
Biochar
Graphene
Sorption
Phenanthrene
Mercury

ABSTRACT

A graphene/biochar composite (G/BC) was synthesized via slow pyrolysis of graphene (G) pretreated wheat straw, and tested for the sorption characteristics and mechanisms of representative aqueous contaminants (phenanthrene and mercury). Structure and morphology analysis showed that G was coated on the surface of biochar (BC) mainly through π–π interactions, resulting in a larger surface area, more functional groups, greater thermal stability, and higher removal efficiency of phenanthrene and mercury compared to BC. Pseudo second-order model adequately simulated sorption kinetics, and sorption isotherms of phenanthrene and mercury were simulated well by dual-mode and BET models, respectively. FTIR and SEM analysis suggested that partitioning and surface sorption were dominant mechanisms for phenanthrene sorption, and that surface complexation between mercury and C–O, C=C, –OH, and O=C–O functional groups was responsible for mercury removal. The results suggested that the G/BC composite is an efficient, economic, and environmentally friendly multifunctional adsorbent for environmental remediation.

© 2015 Elsevier Ltd. All rights reserved.

1. Introduction

Oil requirements have been escalated due to the rapid development of industry and the accelerated expansion of urban areas. During the processes of exploration, development, and transportation of oil, the surrounding air, water, and soil can be polluted. Oil is a complex mixture of hydrocarbons, including alkanes, cycloalkanes, polycyclic aromatic hydrocarbons (PAHs), and heavy metals, such as nickel, cadmium, mercury, lead, and chromium (Zhang et al., 2012c). PAHs are persistent, carcinogenic, mutagenic, and mainly responsible for the toxicity of oil to organisms (Zhao et al., 2014). Phenanthrene is a common PAH, which is ubiquitous

* Corresponding author.
E-mail address: yanyangong@nankai.edu.cn (Y. Gong).
[1] These authors contributed equally.

http://dx.doi.org/10.1016/j.biortech.2015.07.047
0960-8524/© 2015 Elsevier Ltd. All rights reserved.

in the environment and is difficult to remove (Kong et al., 2011). Mercury is one of the most harmful heavy metals to humans, and it is pervasive, cumulative, and persistent in both the human body and the environment. Adverse health effects from mercury exposure include oral inflammation, muscle tremors, and mental disorders (Gong et al., 2014). Due to their great threat to the environment and human health, remediation of PAHs and mercury have been widely concerned (Li et al., 2014).

During the past decades, much efforts have been made to develop high-efficiency, environmental friendly, and cost-effective sorbents, such as activated carbons (Brennan et al., 2014), humic acid (Lu and Zhu, 2011), and biochar prepared by various biomass feedstocks (Tran et al., 2015). However, most of current researches mainly focus on the removal of higher concentrations of contaminants (e.g. phenanthrene 1–10 mg/L, mercury > 1 mg/L) (Kong et al., 2011; Gong et al., 2014). It is necessary to develop a novel adsorptive material with low cost and high removal performance for low concentrations of phenanthrene and mercury.

Biochar (BC) is a black carbon produced during the pyrolysis of biomass. It is a stable solid, rich in carbon, porous with high surface area, and can be produced from abundant feedstock materials (Tang et al., 2013). When applied to soils, BC can sequestrate carbon from atmosphere, enhance soil fertility, and remediate soil pollution. It has been reported that BC has a high sorption capacity for various organic and inorganic contaminants, including phenolic compounds, PAHs, organic pesticides, and heavy metals (Tang et al., 2013; Tran et al., 2015). Recently, efforts have been made to prepare BC-based composite with improved properties, new structure, and enhanced adsorption capacity by combining BC with organic and/or inorganic materials through physicochemical methods (Inyang et al., 2014). For example, the resulting MgO–BC nanocomposites (Zhang et al., 2012a), BC/AlOOH nanocomposite (Zhang and Gao, 2013), γ-Fe_2O_3/BC composite (Zhang et al., 2013), nanotube-BC nanocomposites (Inyang et al., 2014), and graphene-coated BC (Zhang et al., 2012b) showed excellent ability to remove phosphate, nitrate, arsenic, and methylene blue, respectively, from aqueous solutions.

Graphene (G) is a member of the carbonaceous nanomaterial family with a two-dimensional single sheet of carbon atoms arranged in a hexagonal network (Ma et al., 2015). It has been proved to be an effective sorption agent for various environmental contaminants, such as PAHs (Wang et al., 2014), sulfamethoxazole (Chen et al., 2014), copper, lead (Ren et al., 2012), and chromium (Setshedi et al., 2015). G/BC composite may obtain physiochemical properties that cannot be achieved by either of the compositions alone. Zhang et al. (2012b) developed a new engineered G-coated BC and investigated its application in aqueous methylene blue removal. Higher maximum sorption capacity achieved with G-coated BC compared to BC (BC: 8 mg/g and G-coated BC: 174 mg/g) was due to the increased sorption sites, which originated from the π–π electron interaction between contaminants and G/graphite. However, the procedure for the preparation of G-coated BC composite was too complex (i.e. 1,3,6,8-pyrenetetrasulfonic acid was used as received), and the effects of G concentration on the sorption capacity and the morphological changes of the composites have not been studied. Moreover, the effectiveness of G-coated BC for treating phenanthrene and mercury contaminated water has not been investigated.

The overall objective of this study was to test the sorption effectiveness and determine the mechanisms of G/BC for the removal of phenanthrene and mercury in aqueous solutions. The specific objectives were as follows: (1) prepare and characterize G/BC in the presence of various concentrations of G, and elucidate the interactions between G and BC; (2) investigate the effectiveness of the composite for the removal of phenanthrene and mercury; (3) determine the effects of G concentration, adsorbent dosage, contact time, and initial contaminants concentration on the removal effectiveness; and (4) acquire further insights into the underlying removal mechanisms.

2. Methods

2.1. Materials

All chemicals used in this work were of analytical grade. G (diameter: 0.5–2.0 μm, thickness: 0.8–1.2 nm) was purchased from Xianfeng nano-material Company (Nanjing, China). Phenanthrene ($C_{14}H_{10}$), calcium chloride anhydrous ($CaCl_2$), sodium azide (NaN_3) were purchased from Jiangtian Chemical Co., Ltd. (Tianjin, China) and mercury nitrate monohydrate ($Hg(NO_3)_2{\cdot}H_2O$) were purchased from Chengdu AI Chemical Reagent Co., Ltd. (Sichuan, China). Wheat straw obtained from Shandong province of China were air-dried and milled into powders of ~2 mm as the feedstock biomass for BC and BC-based composite production.

2.2. Preparation of G/BC composite

A stock solution of G was prepared at 1000 mg/L in deionized (DI) water. The G suspension was sonicated in an ultra-sound homogenizer (KQ-300DE, Kunshan Ultrasonic Instrument Co., Ltd., Jiangsu, China) with an output frequency of 40 kHz for 1 h. The stock solution was then diluted with DI water to a G concentration of 100 and 500 mg/L.

G/BC composite was synthesized following a revised method by Zhang et al. (2012b) (Fig. 1). In brief, 5 g biomass were immersed into 50 mL of the prepared G suspensions and stirred for 1 h. The mixture was then oven-dried at 80 °C for 3 h. The pretreated biomass were pyrolyzed in a tubular furnace (TDRG, Tengda Thermal Technology Co., Ltd., Yixing, China) to produce G/BC composite through slow pyrolysis at a temperature of 600 °C in a N_2 environment for 1 h. The samples were then washed with DI water for several times to remove impurities, oven-dried at 80 °C, and sealed in a glass container before use. Three types of G/BC composite were prepared by varying the mass ratios of G to BC (i.e. 0.1%, 0.5%, and 1%) and the resulting products were designated as G/BC-0.1%, G/BC-0.5% and G/BC-1%, respectively. For comparison, the biomass and G itself were also treated under otherwise identical conditions to produce BC and G.

2.3. Characterization

The surface area, pore size and pore volume of BC, G, and G/BC were determined by the BET adsorption method (ASAP2460, Micromeritics, Atlanta, USA). Zeta potential was determined using a Zetasizer Nano 2S90 (Malvern Instruments, Malvern, UK). pH value was measured by a pH meter (PB-10, Sartorius, Goettingen, Germany). The crystallinities of the samples were identified by X-ray diffractometer (XRD) (D/max-2500, Rigaku, Tokyo, Japan). Scanning electron microscopy (SEM) (Shimadzu SS-550, Shimadzu Corp., Kyoto, Japan) and transmission electron microscopy (TEM) (T-20, Philips, Amsterdam, Holland) were carried out to observe the structures and surface morphologies of the samples. The thermal stability of the samples was tested by thermal gravimetric analysis (TGA) (TG209, Netzsch, Shanghai, China). The amounts of various types of oxygen-containing functional groups were quantified by the Boehm's titration method (Boehm, 1994). Fourier transform infrared spectroscopy (FTIR) measurements (FTS6000, Bio-rad, Beijing, China) were carried out to investigate the interactions between G and BC, and between phenanthrene/mercury and G/BC-1%. The samples after sorption

Download English Version:

https://daneshyari.com/en/article/7073487

Download Persian Version:

https://daneshyari.com/article/7073487

Daneshyari.com