Journal of Colloid and Interface Science 530 (2018) 46–51

Contents lists available at ScienceDirect

Journal of Colloid and Interface Science

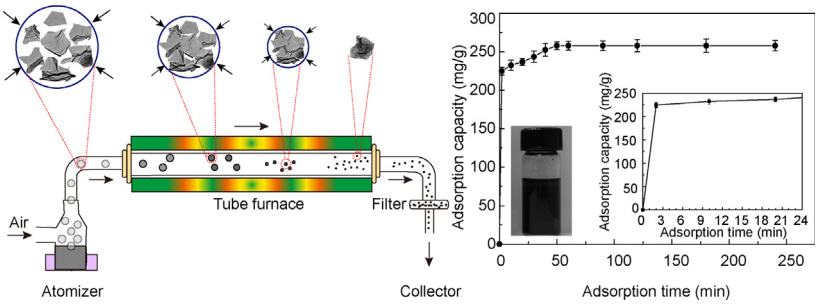
journal homepage: www.elsevier.com/locate/jcis

Crumpled graphene balls as rapid and efficient adsorbents for removal of copper ions

Deqiang Chen [a], Xiang Liu [a], Huali Nie [a,b,*]

[a] College of Chemistry, Chemical Engineering and Biotechnology, Donghua University, Shanghai 201620, PR China
[b] Jiangsu R&D Center of the Ecological Textile Engineering & Technology, Yancheng Polytechnic College, Yancheng, Jiangsu Province 224000, PR China

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 31 May 2018
Accepted 21 June 2018
Available online 27 June 2018

Keywords:
Crumpled graphene balls
Copper ions
Adsorption
Water treatment

ABSTRACT

A novel, three-dimensional (3D), self-supporting material—the crumpled graphene ball—was developed using an aerosol capillary approach. The resultant crumpled-graphene-ball architecture showed a self-supporting, 3D network microstructure with plenty of ridges and wrinkles. Due to their unique structural characteristics, the 3D balls exhibited a rapid adsorption rate and superior adsorption capacity toward the copper ion (Cu^{2+}). It was noted that the adsorption capacity for Cu^{2+} reached about 224.56 mg/g within 2 min. A high adsorption capacity, fast adsorption kinetics, excellent regeneration and reusability characteristics, and the ease of materials processing make these crumpled graphene balls ideal candidates for heavy metal ion decontamination in practical application.

© 2018 Elsevier Inc. All rights reserved.

1. Introduction

Removal of heavy metal ions from wastewater has attracted popular attentions for the environmental protection and human health [1,2]. Many methods remove the ions from water, including ion exchange [3–6], chemical precipitation [7], electrochemical treatment [8], membrane filtration [9], and adsorption [10–12]. Among these methods, adsorption is one of the most recommended physicochemical treatment processes due to its high efficiency, simple operation, fast response time, and low cost [13,14]. The common adsorbents for copper ions are clay minerals [15,16], activated carbons [17–19], chelating materials [20], graphene [21–23] and chitosan/natural zeolites [24], etc. However, these absorbents have often suffered from limited efficiency at low metal concentrations, low adsorption rates, or conditional dependence; they can also be hard to produce [25]. Therefore, a novel adsorbent is needed to improve adsorption efficiency.

Graphene, used as a new type of two-dimensional (2D) materials [26], has a variety of unique and desirable properties. Many researchers use it and its modified products as adsorbents to remove heavy metal ions [27–29]. Unfortunately, 2D graphene

* Corresponding author at: College of Chemistry, Chemical Engineering and Biotechnology, Donghua University, Shanghai 201620, PR China.
E-mail address: niehuali@dhu.edu.cn (H. Nie).

https://doi.org/10.1016/j.jcis.2018.06.051
0021-9797/© 2018 Elsevier Inc. All rights reserved.

sheets are easily agglomerated [30], causing the specific surface area to become smaller and the adsorption capacity to decrease. It has been proven that the construction of three-dimensional (3D), graphene-based materials [31–34], is one of the most effective ways to further optimize the properties of 2D graphene for water purification. However, the process of preparing these graphene-based, 3D materials has thus far been complex or polluted.

The aerosol-assisted synthesis method is a convenient and simple technique for synthesizing a variety of inorganic and/or organic materials with a narrow-sized tribute ion and controllable morphology and composition [35]. Furthermore, the aerosol method is easy to scale up and is compatible with continuous on-line production. A crumpled-ball structure was first prepared with graphene-based materials using an aerosol capillary approach [36]. Compared to the methods that covalently bind a graphene oxide sheet to other support materials, the procedure described in this study has many advantages. First, producing crumpled graphene balls is a non-toxic, facile process. Scientists can avoid time-consuming chemical synthesis and the use of toxic reagents. Second, no support materials are needed. A self-supporting graphene ball is easily prepared by capillary compression in rapidly evaporating aerosol droplets. Switching from graphene oxide sheets to graphene balls is not only a good solution for the aggregation phenomenon but also introduces desirable characteristics. The balls have a high specific surface area, high stability, and strong mechanical properties. Such a graphene ball has been successfully applied in ultracapacitors [37], electrocatalysts [38], batteries [39], oil adsorbents [40] and lubricant additives [41]. In this study, we will extend the application of crumpled graphene to water purification for the first time.

Given the urgent demand of water purification, combined with the inherent merits of graphene-based macrostructure, herein, we have reported a crumpled graphene ball with a self-supporting, 3D network microstructure applied for heavy metals adsorption. The as-prepared crumpled-graphene-ball macrostructure possessed a plenty of ridges and wrinkles, benefiting its sorption in heavy metals. The adsorption studies revealed its superior capacity in copper ions (Cu^{2+}) contaminated waters. Different experimental parameters such as adsorbent concentration, pH and contact time were systematically studied. The uptake stability of Cu^{2+} from aqueous solution and the regeneration of crumpled graphene balls were carried out by adsorption and desorption process.

2. Experimental section

2.1. Chemicals

$CuSO_4{\cdot}5H_2O$ (Mw = 249.68 g/mol, purity > 99%) from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China, were used as copper source. Natural graphite powders were bought from Aladdin Industrial Inc. All other reagents were analytical grade without additional purification and obtained from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). All solutions were prepared with ultra-pure water (18.2 MΩ cm).

2.2. Preparation of crumpled graphene balls

The graphene oxide was prepared according to the modified Hummers synthesis [42], and the graphene oxide sheets were compressed into crumpled graphene balls using an aerosol capillary approach [36]. In a typical synthesis procedure, an aqueous dispersion of micrometer sized graphene oxide sheets (1.0 mg/mL) was nebulized to generate aerosol droplets [43] and flown through a vacuum tube furnace preheated at 400 °C. Rapid evaporation causes shrinkage of the droplets, first concentrating the graphene oxide sheets and subsequently compressing them into crumpled balls of submicrometer scale. The products were filtered with a filter (pore size: 0.22 μm) and dried at room temperature for 5 h, then putted into vacuum dryer at 60 °C for 24 h.

2.3. Characterizations

The concentration of Cu^{2+} was determined using a UV-2102PC spectrophotometer (UNICO Instrument Co. Ltd., Shanghai, China). The microstructure and morphology of the samples were characterized by scanning electron microscopy (SEM, JSM-5600LV, Japan) and transmission electron microscopy (TEM, JEM-2000, Japan). Fourier transform infrared spectrometer (FTIR) spectra were collected on Nicolet Nexus 670 spectrometer (Thermo Fisher, USA) over 4000–500 cm^{-1} at a resolution of 2 cm^{-1}; 32 scans were taken. The presence of Cu^{2+} ions on the surface of crumpled graphene balls is documented by energy dispersive X-ray spectrometer (EDS, Oxford, IE 300X). The surface chemistry was analyzed by X-ray photoelectron spectroscopy (XPS) (Thermo ESCALAB 250XI). The XRD patterns were obtained by X-ray diffractometer D/Max-2550, Rigaku, Japan) with the Cu Kα radiation at 40 kV and 30 mA in the range of 5–90°.

2.4. Adsorption experiments

All of the experiments were conducted in triplicate using a batch equilibration approach at a constant temperature (25 °C), and the glass tubes containing the crumpled graphene balls and Cu^{2+} solution were shaken in a thermostatic shaker bath, operating at 200 rpm. The batch experiments of Cu^{2+} adsorption on crumpled graphene balls were carried out by adding 10.0 mg of crumpled graphene balls to 20 mL of Cu^{2+} aqueous solution (150 mg/L) in 25 mL glass tubes. The initial pH of the solution was adjusted from 2 to 6 by adding different concentrations of HCl or NaOH solutions (0.01–1.0 mol/L) to study the effect of pH on adsorption. Subsequently, crumpled graphene balls were separated through a centrifuge at the speed of 10,000 rpm for 5 min. The filtrate was collected for Cu^{2+} analysis, and the concentration of residual $CuSO_4$ solution was analyzed spectrophotometrically at 740 nm using Na_2EDTA reagent as a complexing agent.

The q_t (mg/g) is the adsorption amount of heavy metals onto crumpled graphene balls at time t and calculated by Eq. (1):

$$q_t = \frac{(C_0 - C_t)V}{m} \tag{1}$$

where C_0 and C_t (mg/L) are the Cu^{2+} concentrations at initial stage and time t, respectively. V (L) is the volume of Cu^{2+} solution and m (mg) is the mass of added adsorbent. The removal efficiency (E) of adsorbent on Cu^{2+} is evaluated by Eq. (2):

$$E(\%) = \frac{C_0 - C_f}{C_0} \times 100\% \tag{2}$$

where C_0 and C_f are the initial and final equilibrium concentration of Cu^{2+} (mg/L) in aqueous solution, respectively.

2.5. Desorption and regenerated studies

To study the regeneration of crumpled graphene balls, seven consecutive cycles of adsorption-desorption process were carried out. For each cycle, 20 mL of 150 mg/L Cu^{2+} solution was adsorbed by 10 mg crumpled graphene balls for 3 h, and then desorbed with 20 mL of 0.01 M Na_2EDTA solution. After adsorption or desorption had reached equilibrium, crumpled graphene balls were separated through a centrifuge at the speed of 10, 000 rpm for 5 min, and the supernate was collected for metal concentration measurements.

Download English Version:

https://daneshyari.com/en/article/6989662

Download Persian Version:

https://daneshyari.com/article/6989662

Daneshyari.com