



Research article

Facilitative capture of As(V), Pb(II) and methylene blue from aqueous solutions with MgO hybrid sponge-like carbonaceous composite derived from sugarcane leafy trash



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ABSTRACT

Enhancing the contaminant adsorption capacity is a key factor affecting utilization of carbon-based adsorbents in wastewater treatment and encouraging development of biomass thermo-disposal. In this study, a novel MgO hybrid sponge-like carbonaceous composite (HSC) derived from sugarcane leafy trash was prepared through an integrated adsorption-pyrolysis method. The resulted HSC composite was characterized and employed as adsorbent for the removal of negatively charged arsenate (As(V)), positively charged Pb(II), and the organic pollutant methylene blue (MB) from aqueous solutions in batch experiments. The effects of solution pH, contact time, initial concentration, temperature, and ionic strength on As(V), Pb(II) and MB adsorption were investigated. HSC was composed of nano-size MgO flakes and nanotube-like carbon sponge. Hybridization significantly improved As(V), Pb(II) and methylene blue (MB) adsorption when compared with the material without hybridization. The maximum As(V), Pb(II) and MB adsorption capacities obtained from Langmuir model were 157 mg/g, 103 mg/g and 297 mg/g, respectively. As(V) adsorption onto HSC was best fit by the pseudo-second-order model, and Pb(II) and MB with the intraparticle diffusion model. Increased temperature and ionic strength decreased Pb(II) and MB adsorption onto HSC more than As(V). Further FT-IR, XRD and XPS analysis demonstrated that the removal of As(V) by HSC was mainly dominated by surface deposition of MgHAsO₄ and Mg(H₂AsO₄)₂ crystals on the HSC composite, while carbon π - π^* transition and carbon π -electron played key roles in Pb(II) and MB adsorption. The interaction of Pb(II) with carbon matrix carboxylate was also evident. Overall, MgO hybridization improves the preparation of the nanotube-like carbon sponge composite and provides a potential agricultural residue-based adsorbent for As(V), Pb(II) and MB removal.

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1. Introduction

Industrial processes often include a finishing procedure which generates waste (Wang and Yang, 2016). Heavy metals and organic substances released from these wastes can contaminate water and pose serious threats to living organisms (Çoruh et al., Çoruh et al., 2013; Tansel and Rafiuddin, 2016; Pecorini et al., 2017, Çoruh et al., 2013; Tansel and Rafiuddin, 2016; Pecorini et al., 2017). Cases in point include extremely toxic arsenic (As) and lead (Pb). For

example, in June 2008, Yangzonghai Lake, a large plateau lake in the Yunnan Province, China, was polluted due to the illegal discharge of As by chemical manufacturers resulting in concentrations higher than 0.12 mg/L making water from the lake unusable for drinking, fishery and irrigation (Chen et al., 2015). Increasing Pb and salinity in Thar Jath, South Sudan seriously affects human and livestock health (Pragst et al., 2017). Recently, it was reported that approximately 1/6 of the total farmland area in China had been contaminated with heavy metals including Pb and As due to the development of mine exploration and metallurgy industry (Lahori et al., 2017).

Various physical, chemical and biological approaches have been explored to meet allowable limits of contaminants in discharged effluent (Deveci et al., 2016). Among these methods, adsorption is

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considered to be one of the most effective methods for removing pollutants because it is operationally easy and cost-effective (Vu et al., 2017). Activated carbon has been the most commonly used adsorbent (Tansel and Surita, 2016), however newer and superior carbonaceous materials such as carbon nanotubes and graphene oxide are being developed as adsorbents for heavy metals (Ihsanullah et al., 2016; Peng et al., 2017) but a disadvantage of these materials is their cost. The search for highly effective and economical adsorbents is driving research and development (Ihsanullah et al., 2016).

A new, promising material is sponge-like carbonaceous material derived from organic feedstock that is thermally decomposed under oxygen-limited conditions (Clough et al., 2013; Lu et al., 2016; Shang et al., 2015). It acts as both the adsorbent matrix and adsorbent for water with dissolved substances (Vardon et al., 2013). Some types of these carbonaceous sponges have been successfully utilized as matrices in the production of high-performance Li–Se batteries (Zhang et al., 2015) and for the adsorption of tetracycline (Zhu et al., 2014). Although these materials feature stable structure, low cost, environmental friendliness, and high adsorption capacities for organic substances and cationic metals (Lee et al., 2017; Zhu et al., 2014), their surfaces are predominantly negatively charged, therefore limiting the adsorption of anionic pollutants. Hence, their physicochemical properties must be modified to increase the adsorption of negative ions if they are to be used on an industrial scale.

One such modification is treatment with chemical reagents. For example, composites of magnetite and orange peel carbon (Chen et al., 2011), MnO₂ particles grafted swine manure carbon (Yu et al., 2017), MgO particles impregnated into alga carbon (Jung and Ahn, 2016), and Mg-accumulated within tomato tissue carbon (Yao et al., 2013) are all synthesized through the addition of chemical reagents and have shown excellent phosphate sorption ability. Rice husk carbon material soaked with salts of Ca²⁺ and Fe³⁺ also exhibits high removal efficiency for arsenate and chromate (Agrafioti et al., 2014). Recently, a Fe₂O₃-carbon foam was used as an adsorbent for the removal of Cu(II), Ni(II) and Cr(VI) ions from metal-plating wastewater (Lee et al., 2017). Focusing on anionic adsorbent with a biomass-based carbon sponge material would seem to yield a nearly universal adsorbent. Even with the above studies, however, research on such modifications of biomass-based sponge carbon material through impregnation with metals is limited. In particular, same as the iron and aluminum oxides particle, the MgO particle impregnation often endows the carbon material a positive surface as well (Jung and Ahn, 2016; Yao et al., 2013). However, the simultaneous adsorption of cationic metal ions, anionic contaminants and organic pollutants using MgO hybridized with sponge-like carbon materials has not been fully investigated yet.

Herein, a kind of MgO particle hybrid with sponge-like carbon material was prepared by using an integrated adsorption and pyrolysis method. The sample feedstock was made from readily available sugarcane harvest residue based on our previous study (Jeong et al., 2012) and the adsorption of negatively charged arsenate (As(V)), positively charged Pb(II), and the organic pollutant methylene blue (MB) were evaluated. Elemental analysis, BET-N₂ surface area analysis, XRD, SEM, TEM, XPS, and FT-IR analysis were performed for sample characterization and adsorption mechanisms were investigated.

2. Materials and methods

2.1. Materials

Sugarcane trash (mainly leaves) collected from the Louisiana

State University Agricultural Center Sugar Research Station at St. Gabriel, LA was used as the feedstock biomass. The leafy trash was cut into pieces smaller than 5 cm and sequentially washed with tap and deionized water (DI, 18.2 MΩ cm) to remove dust, followed by drying at 55 °C overnight. The dry leafy trash was crushed to pass through a <0.12 mm sieve before pyrolysis. Magnesium acetate, sodium hydroxide, hydrochloric acid, nitric acid, sulfuric acid, hydrogen peroxide, and methylene blue (MB; C₁₆H₁₈N₃SCl) of analytical grade were purchased from Fisher Scientific. Working solutions of Pb(II) and AsO₄³⁻ were prepared by diluting 1000 mg/L standard stock solutions (Fisher Scientific). All chemical solutions were prepared by using DI water.

2.2. Adsorbents preparation

The MgO hybrid sponge-like carbonaceous material (HSC) was produced using a modified adsorption and pyrolysis combined method as described by Liu et al. (2013). In brief, about 50 g of leafy trash powder and 1000 mL of magnesium acetate solution were mixed in a glass flask with continuous shaking on a constant-temperature (23 ± 0.2 °C) oscillator for 24 h. The mixture was then heated at 80 °C to remove supernatant and dried at 105 °C for 6 h. Thereafter, the dried mixture was transferred into a porcelain crucible and placed in a muffle furnace under N₂ flow at 550 °C as described elsewhere (Jeong et al., 2012). After pyrolysis, the resulting product was gently crushed and passed through a <0.12 mm nylon sieve. The sugarcane non-hybridized sponge-like carbonaceous material (SSC) was also made as a comparison.

2.3. Adsorbents characterization

Total C, H and N contents in the different carbon foams were measured using an elemental analyzer (Elementar Analysensysteme GmbH, Germany), and total Mg content was determined by inductively coupled plasma-atomic emission spectrometry (ICP–AES, SPECTRO Plasma 3200, Germany) after samples incineration and digestion with H₂SO₄–H₂O₂. The BET surface area (*S*_{BET}), total pore volume (*V*_{tot}) and pore size distribution of the carbon samples were characterized based on N₂ adsorption at 77 K using a V-Sorb 2800P analyzer (App-one, China). The microscopic features and morphology were evaluated by using a field emission gun scanning electron microscope (FEG–SEM, JEOL 6335F, Japan) and a transmission electron microscope (JEOL 200CX TEM, Japan). A computer-controlled X-ray diffractometer (XRD, Philips Electronic Instruments), a Fourier transform infrared (FT-IR) spectroscopy (Nicolet 5700, USA) and an X-ray photoelectron spectroscopy (XPS, Kratos, UK) were also used to characterize the samples.

2.4. Adsorption performance tests

The effects of solution pH on adsorption were investigated in batch experiment. In doing so, a series of 20 mL 50 mg/L As(V), Pb(II) or MB solutions were adjusted with 0.1 mol/L HNO₃ or NaOH solution to desired pH values before mixing with 0.05 g of SSC and HSC in centrifuge tubes on a shaker platform operating at 120 rpm for 24 h at room temperature. The initial solution pHs were 3.0–10.0 for As(V), 3.0–5.0 for Pb(II) and 3.0–10.0 for MB. After 24 h shaking, the supernatants containing As(V) and Pb(II) were filtered through 0.22 μm nylon membrane filters (GE cellulose nylon membrane) and quantified using an Atomic Fluorescence Spectrometer (AFS-9700, Beijing Haiguang Instrument, China) and an ICP–AES (SPECTRO Plasma 3200, Germany), respectively. The mixtures for MB adsorption experiments were centrifuged at 12,000 rpm for 15 min, and the supernatants were collected

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