



## Research article

# Synthesis of mesoporous bismuth-impregnated aluminum oxide for arsenic removal: Adsorption mechanism study and application to a lab-scale column

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## ABSTRACT

High mobility and toxicity of arsenic [As (III)] limit its removal from an aquatic environment and pose a threat to human health. In this work, batch adsorption experiments were conducted to investigate the adsorption capacity of bismuth-impregnated aluminum oxide (BiAl). Continuous application of As (III) removal was achieved via a lab-scale column reactor. Bismuth impregnation decreased the specific surface area of aluminum oxide and affected its pore size distribution. However, because of its abundant and well-proportioned mesoporous character, it also enhanced its adsorption capacity through the surface complexation of As (III). Batch adsorption experiments demonstrated a suitable Freundlich model and a fitted pseudo-second-order kinetic model for As (III) adsorption. The main mechanism was chemisorption with both bismuth and aluminum atoms; however, physisorption also contributed to arsenic adsorption at the initial stage of the reaction. The Adams-Bohart model better described the breakthrough curves than the Thomas model. BiAl exhibited efficient As (III) adsorption over a wide pH range and could be applied to As (III) removal from wastewater. A high As (III) removal efficiency (91.6%) was obtained at an initial As (III) concentration of 5 mg L<sup>-1</sup> at a flow rate of 1 mL min<sup>-1</sup>. This study indicates the potential for the practical application of BiAl in As (III) removal.

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## 1. Introduction

Arsenic, a widely distributed metalloid element in natural environment, displays superior physical, chemical, and photoelectric properties. Thus, over the past few decades, it has been widely applied to electronic components and photoelectric cells (Song et al., 2017). Unfortunately, because of its teratogenicity, mutagenicity, and carcinogenicity in biological systems, arsenic is toxic to both the ecological environment and human beings (Phung et al., 2017). AsO<sub>2</sub><sup>-</sup> (III) and AsO<sub>4</sub><sup>3-</sup> (V) are the predominant inorganic arsenic species in the aquatic environment. Both species are non-biodegradable and accumulated through the food chain (Bretzler et al., 2017; Mondal et al., 2013; Zhao et al., 2013). Thus, because of the high risk to human health, the World Health Organization recommends that arsenic concentrations exceeding 10 μg L<sup>-1</sup>

should not be discharged into runoffs (Tang et al., 2014; Wang et al., 2010; Zhang et al., 2017). Therefore, it is highly imperative and meaningful to explore high-efficiency, innovative, and cost-effective materials for application to arsenic wastewater treatment.

Adsorption has been widely investigated and proven to be an effective method for wastewater treatment (Boczkaj and Fernandes, 2017; Yang et al., 2017). Previous studies have investigated the arsenic removal efficiency of core-shell Fe@Fe<sub>2</sub>O<sub>3</sub> nanobunches (nanocomposites containing nanoscale zero-valent iron and Fe<sub>2</sub>O<sub>3</sub>) in acid wastewater. They demonstrated that the efficiency of arsenic removal dramatically improved when As (III) was oxidized to As (V) prior to its adsorption by Fe@Fe<sub>2</sub>O<sub>3</sub> (Tang et al., 2017). As (III), which comprises 67–99% of the total arsenic in groundwater, is 100- to 500- folds more toxic than As (V) and more difficult to remove (Giles et al., 2011; Liu et al., 2008). Nonetheless, few works on cost-effective and eco-friendly methods and materials for arsenic (III) treatment have been reported to date.

Traditionally, the adsorption capacity of an adsorbent is considered to be highly related to its specific surface area (Prasanna

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and Kamath, 2009). Biochar possesses a large specific surface area that plays an important role in the adsorption of pollutants in wastewater, and thus, its use in wastewater treatment is widespread (Johansson et al., 2016; Mandal et al., 2017). However, these materials do not remove As (III) efficiently. Many recent studies have proven that the heterogeneous functional groups and vast porous structure enhance the surface chemistry of biochar. Thus, biochar, particularly activated carbon, displays considerable adsorption capability towards inorganic and organic contaminants (Johansson et al., 2016). Numerous efforts have been made to prepare activated carbon, including Fe (III)-modified crop straw biochar and lanthanum-activated carbon, by impregnation with metals and metal oxides (Pan et al., 2015; Wang et al., 2015). The results proved that impregnation is an efficient and effective method to modify the original adsorbent by changing the pore size distribution and the specific surface area of the sorbent and by increasing the adsorption capacity towards certain pollutants. Nevertheless, the smaller pore size of most of these engineered carbons, including biochar, hinders adsorbate diffusion in the biochar matrix. This limits their ability to reduce anionic pollutants such as arsenite and arsenate (Wang et al., 2017). Numerous studies have revealed the excellent ability of aluminum oxide to remove arsenic from wastewater (Duarte et al., 2012; Önnby et al., 2014). Aluminum oxide-related compounds have been used to remove arsenic (V) efficiently, while removal of the more toxic arsenic (III) proved to be difficult. Bismuth, with an electronic configuration of (Xe)  $4f^{14}5d^{10}6s^26p^3$ , is one of the most thoroughly investigated main group elements. It is known as “the wonder metal” owing to its easy involvement in chemical combinations because of the electrons in its p orbital (Sun et al., 2014). Thus, bismuth-related compounds show great potential as adsorbates for inorganic anion contaminants through ligand exchange and electrostatic incorporation. Bismuth oxide has been studied intensively as a potential photocatalyst and electrode material; however, few reports on arsenic removal by bismuth oxide are available to date. In our previous study, we prepared bismuth-impregnated biochar and investigated its arsenic adsorption properties. We reported that while bismuth was mainly responsible for arsenic adsorption (Zhu et al., 2016), its adsorption ability was still limited because of its small pore size distribution (diameter: 2 nm). Impregnation was an effective method to activated biochar and also modified adsorbent. Earlier study has reported the synthesis of mesoporous aluminum and achieved considerable adsorption capacity for arsenic (V) (Han et al., 2013). However, this only worked in an acidic environment and was not so effective for As (III) adsorption. In this work, bismuth-impregnated aluminum oxide was explored as a low-cost but highly efficient material to enhance sorption capacity towards As (III). The specific objectives of this work were: (1) to prepare and characterize bismuth-impregnated  $Al_2O_3$  composites, (2) to test the adsorption capacities of these composites and their potential for practical application in As (III) removal, and (3) to investigate the possible mechanisms involved in As (III) sorption.

## 2. Materials and methods

### 2.1. Preparation of sorbents

A bismuth salt solution was prepared by adding 10 mmol  $Bi(NO_3)_3$  to 0.1 M hydrochloric acid (30 mL). The resulting solution was then diluted to 100 mL with distilled water under ultrasonic treatment. Next, bismuth-modified aluminum oxide was produced as follows: 5 g  $Al_2O_3$  was mixed with 100 mL bismuth salt solution, stirred vigorously at 80 °C for 3 h, and subsequently dried at 105 °C. The afforded precursors were then heated to a set temperature (400, 500, and 600 °C) at a heating rate of 1 °C/min and finally

calcined for 180 min in a furnace. All the prepared materials were washed (threefold) with 0.01 M  $NaHCO_3$  solution and distilled water and labelled as BiAl400, BiAl500, and BiAl600 according to their set temperature. The control was prepared from a mixture of 5 g  $Al_2O_3$  and hydrochloric acid (0.1 M), using the same preparation procedure, at a calcination temperature of 600 °C.

### 2.2. Adsorption experiments

The adsorption capacity of the prepared materials was investigated via adsorption experiments. First, 500 mg/L sodium arsenite stock solution was prepared and diluted to the desired concentrations (below). The effect of the pH on arsenic adsorption was investigated in a centrifuge tube containing 0.1 g adsorbent and 50 mL sodium arsenite solution (10 mg/L). The pH was adjusted to a value ranging from 4 to 12 with hydrochloric acid and sodium hydroxide accordingly.

Adsorption isotherm experiments were conducted as follows: 0.1 g adsorbent was mixed with 50-mL sodium arsenite solutions with different arsenic concentrations ranging from 5 to 500 mg/L under the optimum pH value in the centrifuge tube. All the vessels were shaken at 200 rpm in an oscillator at 25 °C for 2 h. The concentration of arsenic was determined by inductively coupled plasma-mass spectrometry (ICP-MS, 7700x- JP12502215, Agilent Technologies, USA) using  $H_2$  as collision gas. Samples were filtered through a 0.22- $\mu$ m nylon filter and introduced into the analytical column prior to introduction into the ICP-MS model.

Adsorption kinetic experiments were conducted as follows: 0.1 g adsorbent was added to 50 mL arsenic solution under the optimum pH level at an initial concentration of 20 mg/L. The amount of arsenic sorption was investigated at different time intervals (0, 0.5, 1, 2, 5, 10, 30, 60, and 120 min). The mixture was agitated at 200 rpm in an orbital shaker (SHA-C) at 25 °C for 2 h and subsequently allowed to stand for 24 h to assure that equilibrium was reached. The adsorption process varied rapidly in the first 5 min during which adsorption occurred quickly (Ma et al., 2012). Taking into account the possible variation of the adsorption amounts during the initial period of the adsorption process, the sampling time was conducted accurately. Moreover, the samples were immediately filtered through a 0.22- $\mu$ m nylon filter to minimize the short lapses.

### 2.3. Column experiments

Wastewater was collected from Xuanwu Lake, East China. To facilitate comparisons with the results of the static experiments described above, the initial wastewater As (III) concentrations were adjusted to 5.0, 10.0, and 20.0  $mg L^{-1}$  by dilution with water or the direct addition of sodium arsenite. The wastewater exhibited a pH of  $7.9 \pm 0.2$ . The self-designed column reactor consisted of one pump, one clarification tank, one influent tank, and one column (diameter: 6 cm, height: 20 cm) with a 565-mL working volume (Supplementary data, Fig. S1). The column reactor was filled with a mixture of 10 g bismuth-impregnated aluminum and quartz sand (height: 12 cm) and quartz sand at both the top and bottom sides (height for both sides: 2 cm). All the materials used in the reactor were washed thoroughly with 0.05 M HCl and deionized water threefold before packing into the pillar. All the reagents were of analytical grade and purchased from Nanjing Ronghua Company.

### 2.4. Desorption experiments

Desorption tests were carried out to evaluate the usability of the sorbent as described in the adsorption studies above. Six different eluents were selected: distilled water, 3%  $H_2O_2$  solution (w/w),

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