

Adsorptive removal of arsenate and orthophosphate anions by mesoporous alumina

Mi-Jin Yu, Xiong Li, Wha-Seung Ahn*

Department of Chemical Engineering, Inha University, Incheon 402-751, Republic of Korea

Received 8 September 2007; received in revised form 8 November 2007; accepted 16 November 2007

Available online 3 January 2008

Abstract

A mesoporous alumina was synthesized using aluminum alkoxide in the presence of a cationic surfactant under hydrothermal conditions. The material contains a wormhole-like mesopore structure, and exhibits high surface area ($483 \text{ m}^2/\text{g}$) and pore volume ($0.82 \text{ cm}^3/\text{g}$). The adsorption capacities of the mesoporous alumina for arsenate (61.3 mg/g) and orthophosphate (37.4 mg/g) anions in an aqueous solution were 5.1 and 2.2 times higher, respectively, with initial adsorption rates being 3.8 (1.5 mg/g min) and 1.9 (1.3 mg/g min) times faster than those obtained with an activated alumina. The textural features of larger surface area and pore volume of the mesoporous alumina compared to the activated alumina are believed to be responsible for this enhancement in adsorption behavior.

© 2007 Elsevier Inc. All rights reserved.

Keywords: Mesoporous alumina; Adsorption; Arsenate; Orthophosphate

1. Introduction

The M41S family of mesoporous silicate molecular sieves reported in 1992 by Mobil has opened up new possibilities in the fields of catalysis, sensors, and adsorbents. These materials are synthesized with a self-assembled molecular array of surfactant molecules as a structure-directing template, yielding very sharp and ordered pore distributions of inorganic materials with high surface area and pore volume [1,2]. As an extension of this template-synthesis approach, mesoporous alumina has been prepared in a similar fashion, and is accordingly emerging as an important engineering material. Traditionally, transition alumina or activated alumina has been widely used as catalysts, supports, and adsorbents. However, there is a problem of deactivation in catalysis arising from pore plugging due to coke formation in micropores. It is therefore desirable to produce alumina with mesopores in a narrow pore size distribution. The synthesis of mesoporous

alumina with controlled porosity and high thermal stability has thus been actively pursued [3], and several reviews of the synthesis and characterization of mesoporous alumina were reported [4,5].

In a previous study, the present authors conducted an extensive comparison of synthesis recipes of mesoporous alumina, employing diverse kinds of neutral, anionic, and cationic surfactants [6]. Based on this work, we arrived at the conclusion that the mesoporous alumina prepared using a cationic surfactant is superior in terms of textural properties, including comparatively higher hydrothermal stability. Having established a stable synthesis protocol for mesoporous alumina, we are currently investigating a potential application of the material as an adsorbent for pollutants in air and in aqueous environments.

Arsenic is highly toxic and is a carcinogenic element that exists predominantly in the form of oxyanions in aquatic environments [7], and the removal of arsenic is a very important aspect of wastewater treatment. The most common valence states of arsenic in water are arsenate (As^{+5}) and arsenite (As^{+3}) forms. Generally, As^{+5} is more prevalent in surface water while As^{+3} is more likely to occur in

* Corresponding author. Tel.: +82 32 866 0143; fax: +82 32 872 0959.
E-mail address: whasahn@inha.ac.kr (W.-S. Ahn).

anaerobic ground waters [8]. With almost all of the technologies thus far evaluated, removal of As^{+3} from aqueous solutions is generally poor compared to that of As^{+5} , because the predominant As^{+3} compound is neutral in charge, while the As^{+5} species are negatively charged in a pH range of 4–10.

Concurrently, the presence of orthophosphates in water bodies, resulting from domestic wastewater discharges, agricultural drainage, and urban runoff, poses an environmental problem through the inducement of eutrophication, leading to propagation of aquatic plants, algal growth, and a depletion of dissolved oxygen [9].

The major method for the removal of both arsenate and orthophosphate is adsorption; after being transferred to a solid surface from an aqueous phase, these species are bound either by physical or chemical forces. The adsorbents most often used are activated alumina [10,11], activated carbon [12], goethite [13], and gibbsite [14], as well as other materials [15–17]. However, as a result of lower adsorption capacity and poor adsorption kinetic property, these adsorbents cannot be widely used. Recently, aluminum-impregnated mesoporous silica SBA-15 [2,18] and mesoporous alumina prepared with an anionic surfactant [19] were reported to respectively demonstrate higher adsorption performance than commercial activated alumina.

In this work, we used a mesoporous alumina previously developed by the authors that is prepared with a cationic surfactant for adsorptive removal of arsenate and orthophosphate from drinking water. The adsorption capacities and adsorption kinetics were measured and its effectiveness was critically evaluated against a commercial activated alumina sample (Catapal B, Condea Vista). The pseudo boehmite “Catapal alumina” is among the best catalyst base materials supplied on the market because it can be obtained essentially free from impurities such as sodium [20].

2. Experimental

2.1. Synthesis and characterization of mesoporous alumina

The mesoporous alumina was prepared following the recipe reported by Ray et al. [6]. Aluminum tri-*sec*-butoxide (ASB, Aldrich) was used as the aluminum precursor and cetyltrimethylammonium bromide (CTAB, Aldrich) was used as a cationic structure-directing agent. 2-Butanol (Aldrich) was employed as an organic solvent. CTAB (4.38 g) was dissolved in a solution of 10.18 g ASB in 40.0 g of 2-butanol under stirring. A solution of 1.44 g of water in 5.0 g of 2-butanol was then added slowly with stirring. The molar composition of the mixture was Al:surfactant:water:2-butanol = 1:0.3:2:15. This solution gradually turned into a gel, which was heated in an autoclave at 100 °C for 24 h at static conditions under autogenous pressure. The resulting gel was filtered, washed with ethanol, and dried at room temperature overnight, and then further dried at 100 °C for 5 h. The dried sample was subsequently calcined at 500 °C for 4 h. The mesoporous alumina

showed good structural stability at temperatures to 500 °C. However, further calcinations at higher temperatures resulted in irreversible structural changes [6,21].

The crystallinity of the mesoporous alumina thus prepared was measured by X-ray diffraction using Ni-filtered Cu K α radiation (Rigaku, DMAX 2500). The morphology of the samples was examined by TEM (Philips, CM 200). The BET surface area, pore volume, and average pore diameters were determined by N₂ physisorption at –196 °C using a Micromeritics ASAP 2000 automatic analyzer.

2.2. Adsorption experiments

2.2.1. Adsorption isotherms

Arsenate and orthophosphate adsorption isotherms for the samples were acquired in batch experiments. Sodium hydrogen arsenate heptahydrate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$, Sigma) and sodium dihydrogenphosphate (NaH_2PO_4 , Aldrich) were used as an arsenate and orthophosphate source, respectively. Before the adsorption experiments, mesoporous alumina samples were dried at 200 °C for 4 h. For arsenate adsorption, solid samples weighing between 0.02 and 0.4 g were placed in a series of beakers with 100 mL of solution having 10–70 mg/L arsenate concentration. The sample beakers were stirred at 200 rpm at room temperature for 24 h. After stirring, the suspension was filtered immediately with a 0.45 μm microfilter. The arsenate concentration of the filtered solutions was analyzed by an inductively coupled plasma mass spectrometer (Perkin Elmer, ELAN 6100).

The experimental conditions for obtaining the orthophosphate adsorption isotherms are identical to those for the case with arsenate with the exception of a different initial concentration of orthophosphate (10–250 mg/L). The orthophosphate concentration of the filtered solution was again analyzed using an inductively coupled plasma mass spectrometer.

2.2.2. Adsorption kinetics

Adsorption kinetic experiments of arsenate and orthophosphate were performed in 1 L solutions with 0.4 g sample powders. The initial arsenate and orthophosphate concentrations of the solution were both kept at 10 mg/L, and each solution was stirred at 200 rpm by a magnetic bar. The supernatant samples were taken at various times during an 8 h experiment. The arsenate and orthophosphate concentrations of these samples were measured with an inductively coupled plasma mass spectrometer.

3. Results and discussion

3.1. Physical properties of mesoporous alumina and activated alumina

Fig. 1A shows both the low and wide-angle reflections of the mesoporous alumina and activated alumina samples.

Download English Version:

<https://daneshyari.com/en/article/76669>

Download Persian Version:

<https://daneshyari.com/article/76669>

[Daneshyari.com](https://daneshyari.com)