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Novel Fe/glass composite adsorbent for As(V) removal

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Abstract

An effective adsorbent for arsenic removal was synthesized by hydrothermal treatment of waste glass powder (GP), followed by loading Fe(III) oxyhydroxide onto the surface of waste glass powder. The ≡Si–O–H group was formed on the surface of GP and the specific surface area of GP powder was slightly increased after hydrothermal treatment. FeOOH was loaded onto the surface of hydrothermally treated waste glass powder (HGP) by the hydrolysis of $FeCl_3$. The formation conditions of FeOOH were also investigated. The ability of this new adsorbent for arsenic (As(V)) removal was evaluated. The results indicated that the highest removal efficiency was about 97% for 1 mg/L As(V) solution at pH 6 and keeping time 2 h.

Key words: arsenic removal; adsorbent; iron oxide; hydrothermal treatment

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Introduction

Arsenic contamination in aqueous system, which resulted in human poisoning and death, is a global problem. Recent reports from Taiwan (Tseng *et al*., 1968; Chen *et al*., 1994), Chile (Borgono *et al*., 1977), China (Zhang and Chen, 1997), Bangladesh (Nickson *et al*., 2000), Vietnam (Berg *et al*., 2001), India (Jiang, 2001; Dixit and Hering, 2003) and the western United States (Roberts *et al*., 2004) indicated that many of the natural water resource have been contaminated by arsenic. Therefore, the arsenic concentration of 0.01 mg/L has been recommended by World Health Organization (WHO, 1996) as a guideline for drinking water. This level was also adopted by some industrialized countries (EPA, 2001). In China, the maximum arsenic level of 0.05 mg/L has been permitted in groundwater supplies for drinking purposes (GB/T14848-93). To meet such a low permit level, it is required developing a effective novel technology for arsenic contaminated drinking water at a reasonable cost.

Arsenic exists in natural water system as well as in the industrial wastewater as the major form of inorganic species. In natural water, the arsenate (As(V)) is dominant in surface water, while arsenite (As(III)) in the groundwater. Since As_2O_3 is widely used, such as pesticides, the glass industry, and the copper refining industry (Saha *et al*., 1999; Ferguson and Gavis, 1972). As(III) is expected to be the main form of arsenic pollutant and is known to be more toxic, mobile, and difficult to be removed than As(V) (Stronach *et al*., 1997; Zhang and Itoh, 2005). Therefore, for common methods As(III) is always be oxided before removing (Zaw and Emett, 2002).

Many technologies such as coagulation (Gregor, 2001), ion exchange, and adsorption (Ghurye *et al*., 1999) have been employed for arsenic removal. The adsorption technique is becoming more popular because of its simplicity, potential of regeneration, and low cost. It is reported that iron oxides including oxyhydroxides and hydroxides, such as amorphous hydrous ferric oxide (FeOOH), goethite (α-FeOOH) and hematite (α-Fe_2O_3), are promising effective adsorbents for As(III) and As(V) removal (Ferguson and Gavis, 1972; Piece and Moore, 1982; Cornell and Schwertmann, 1996; Reed *et al*., 2000; Altundogan *et al*., 2000). Among these iron compounds, FeOOH has the highest adsorption capability due to its large surface area. However, FeOOH available as a fine powder is difficult to be separated from aqueous solution after the adsorption process. In addition, amorphous FeOOH is easy to transform into crystalline iron oxides during the preparation process, resulting in a decrease of its surface area and removal capacity for arsenic. To solve these problems, many researchers attempted to load the iron oxide on the sand surface (USEPA, 1999; Thirunavukkarasu *et al*., 2003) and load amorphous FeOOH on the surface of municipal solid waste melted slag (Zhang and Itoh, 2005).

In the present study, we developed a technology to prepare adsorbent for As removal from contaminated water,

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which involves two steps: hydrothermal treatment of glass powders and the loading of FeOOH on HGP via chemical processes.

1 Materials and methods

1.1 Materials

The $FeCl_3 \cdot 6H_2O$ and NaOH powder (analytical grade, Wako Pure Chemical Industries, Ltd., Japan) were used to prepare $FeCl_3$ and NaOH solution, respectively. Arsenic-containing solution with an As(V) concentration of 1 mg/L was prepared from the standard As(V) solution (Wako Pure Chemical Industries, Ltd., Japan)).

A commercially available glass powder (GP) produced by used glass bottles, was purchased from Toyo System Plant Co., Ltd., Japan. In this work, the glass powder with the particle size of 250–350 μm was used to prepare adsorbent.

1.2 Hydrothermal treatment of glass powder

A mixture of the raw GP (15 g) and the distilled water (30 mL) was transferred into a Teflon container with 40% volume filling, and then sealed by a stainless steel autoclave. The autoclave was put into an oven for heating and hydrothermal reaction was conducted at 180°C for 2 h. Hydrothermally treated waste glass powder (HGP) and the raw GP were observed by scanning electron microscopy (SEM S530, Hitachi, Japan). Nitrogen adsorption behavior of the adsorbent was measured using a NOVA 1200 (Quanta Chorome, USA), and the specific surface area of the absorbent was calculated based on the BET model.

1.3 Formation of β-FeOOH

The formations of β-FeOOH were carried out in a water bath (70°C, 2 h) by the hydrolysis of $FeCl_3$ solution with the addition of HGP. The effects of solution pH, solution concentration, treating temperature, and time on arsenic removal were examined to determine the optimum conditions for preparing effective arsenic absorbents. For each 1 g powder sample, 1, 2, 5, 10 mL $FeCl_3$ solution with the concentrations of 0.01, 0.1, 0.5 and 1 mol/L was added, respectively, and treated at 70°C for 2 h in the water bath. After cooling, filtering and drying at the room temperature in vacuum, the Fe(III) oxyhydorxide-loaded glass powder (IGP) was obtained, which was used as an absorbent for arsenic removal in this study.

The amount of the loaded iron on the IGP was also determined by inductively coupled plasma (ICP) spectrometry (SPS7000A, Seiko, Japan). Sample powder 0.5 g was dissolved in 30 mL 6 mol/L HCl with an ultrasonic stirring for 10 min.

The hydrolyzed productions were identified by powder X-ray diffraction (XRD, RTP-300RC, Rigaku Co., Japan) with Cu K_α radiation (40 kV and 100 mA). The surface of the IGP was observed by SEM.

1.4 Arsenic removal test

The synthesized absorbent IGP 1 g was putted into the 10 mL 1 mg/L As(V) standard solution and reacted at different pH. Then treated at different temperatures for different time in a water bath incubator. The arsenic concentration was determined by atomic absorption spectrometry (AAnalyst200, Perkin-Elmer, Japan). The arsenic removal efficiency (R) was calculated using the following Eq. (1):

$$R = \frac{C_{\mathrm{Asb}} - C_{\mathrm{Asa}}}{C_{\mathrm{Asb}}} \times 100\% \qquad (1)$$

where, C_{Asa}, C_{Asb} are the arsenic concentration after and before the adsorption through the synthesized absorbent.

2 Results and discussion

2.1 Hydrothermal treatment of glass powder

In this work, GP was hydrothermally treated at 180°C for 2 h according to a previous work (Yanagisawa *et al.*, 2006) and used as absorbent after loading Fe compounds through the hydrolysis of $FeCl_3$ solution. Figure 1 shows XRD patterns of the original glass powder and HGP. A broad peak, ranging from 10° to 40°, indicates GP amorphous property. After hydrothermal treatment, the powder still mainly consisted of amorphous materials. It is supposed that Na^+ were dissolved under hydrothermal condition by the following Reaction (2).

$$\equiv\mathrm{Si{-}O{-}Na^+} + H_2O \longrightarrow \equiv\mathrm{Si{-}O{-}H} + \mathrm{NaOH} \qquad (2)$$

The ≡Si–O–H group was formed on the surface of the glass powder.

Figure 2 shows SEM micrographs GP and HGP surfaces. After the hydrothermal treatment, the morphology of the GP surface was changed greatly. It was getting rough. Many fine flocky bulges were formed on the GP surface, which might be crystalline calcium silicate and sodium silicate. The change of glass powder surface resulted in the variation of specific surface area of GP. It was checked by BET. The results showed that the specific surface areas for GP, HGP, and IGP were 1.04, 1.24, and 40.59 m^2/g, respectively.

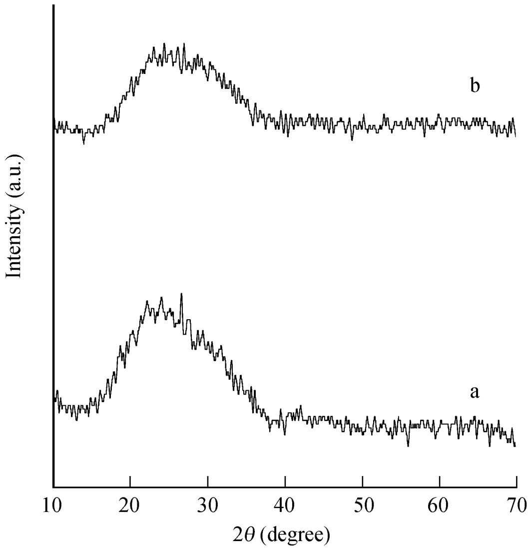

Fig. 1 X-ray diffraction (XRD) patterns of the used glass powder (a) and hydrothermally treated glass powder (b) at 180°C for 12 h.