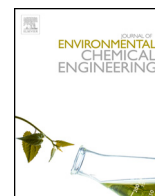




Contents lists available at ScienceDirect

## Journal of Environmental Chemical Engineering

journal homepage: [www.elsevier.com/locate/jece](http://www.elsevier.com/locate/jece)

## Competitive adsorption of As(V) with co-existing ions on porous hematite in aqueous solutions

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## ARTICLE INFO

## Article history:

Received 27 February 2015

Accepted 17 May 2015

## Keywords:

Arsenic(V)

Adsorption

Porous hematite

Co-existing ions

## ABSTRACT

Effects of co-existing ions on the adsorption of As(V) in aqueous solutions by porous hematite have been investigated in this work. The porous hematite was prepared by the goethite concentrate which was concentrated from the iron oxide tailings. The co-existing ions include  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{HCO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$  anions and  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ . The arsenic aqueous solutions with these co-existing ions were simulated using chemical reagents and deionized water. The adsorption isotherm and kinetics were all examined. The experimental results showed that  $\text{HCO}_3^-$  and  $\text{PO}_4^{3-}$  anions inhibited the As(V) adsorption, whereas  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  ions have promoting effects on the As(V) adsorption. Furthermore, the ionic strength has no significant effect on the arsenate adsorption to the porous hematite. These results may be attributed to competitive adsorption of As(V) on porous hematite in aqueous solutions.

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## Introduction

Arsenic is a toxic element in the form of inorganic arsenite, arsenate and organic species. Arsenic mainly originates from weathering reactions, bio-logical activities, geochemical reactions, volcanic emissions and also from some anthropogenic sources including mining, glass processing, insecticides, pesticides and landfill leaching [1]. Exposure of arsenic in drinking water probably brings the greatest threat to human health [2]. It can cause cancers, blackfoot and hyperkeratosis diseases [3]. The World Health Organization established the maximum arsenic level in drinking water as 0.20 mg/L in 1958, 0.05 mg/L in 1963, reduced to 0.01 mg/L in 1993 [4].

Various methods of arsenic removal, such as precipitation-coagulation, membrane separation, ion exchange, lime softening and adsorption [5] on iron oxides or activated alumina, have so far been reported. However, arsenic adsorption using iron oxides as adsorbents has been a promising method because of its ease and sludge-free of operation, low-cost regeneration [6]. Especially, the iron oxide minerals including goethite, hematite, siderite, limonite, ferrihydrite and magnetite, are originated from the tailings [7]. The utilization of such iron oxides tailings as adsorbents plays a significant role in the aspects of economics and environment.

However, iron oxides tailings have a limited adsorption capacity because of their small surface area. The goethite concentrate could be transformed into porous hematite around 300 °C with the increasing

rate of 5 °C/min and the residence time of 3 h. The specific surface area was increased from 14 to 121 m<sup>2</sup>/g after calcination [8]. The adsorption capacity of As(V) on porous hematite in aqueous solutions has been greatly improved to 8.94 mg/g compared to the value of 1.07 mg/g from the siderite-goethite concentrate [9]. It also has been found that the adsorption capacity of hematite which was made from fine powder of magnetite at 1200 °C for 24 h, was 0.65 mg/g with a specific surface area of 3.75 m<sup>2</sup>/g [10]. Recent experimental results have shown that arsenic in water was difficult to be removed by natural iron oxides because of the co-existing ions in aqueous solutions. It was reported that the arsenic adsorption by hematite could be reduced significantly in the presence of  $\text{SO}_4^{2-}$  and  $\text{PO}_4^{3-}$  [11]. However, Zhang et al. [12] pointed that the presence of  $\text{Cl}^-$  and  $\text{SO}_4^{2-}$  both had a slightly positive effect on As(V) adsorption on the iron ore. Furthermore, it has been found that the ionic strength had no effect on the arsenate adsorption on the hematite [13]. Hence, to date, the effects of co-existing ions on the arsenic adsorption on porous hematite still remain unclear.

In this work, the adsorption of As(V) in aqueous solutions has been investigated by porous hematite which was thermally modified from the goethite concentrate. The porous hematite was originated from naturally abundant goethite but with low grade iron. Such application for the natural goethite as adsorbents after treated may enhance the value of resource utilization and have great potential on various sources of cheap adsorbents. In particular, the effects of the co-existing ions in aqueous solutions on the arsenic(V) adsorption isotherm and adsorption kinetics are discussed in order to understand the adsorption mechanisms of those competitive ions.

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## Materials and methods

All chemical reagents were analytical grade, including sodium arsenate ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ), sodium chloride ( $\text{NaCl}$ ), sodium bicarbonate ( $\text{NaHCO}_3$ ), three sodium phosphate ( $\text{Na}_3\text{PO}_4$ ), sodium sulfate ( $\text{Na}_2\text{SO}_4$ ), sodium nitrate ( $\text{NaNO}_3$ ), magnesium chloride hexahydrate ( $\text{MgCl}_2$ ), potassium chloride ( $\text{KCl}$ ), calcium chloride ( $\text{CaCl}_2$ ). The arsenate solutions were prepared using  $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$  (Beijing Chemical Works, China). Other chemicals mentioned above (Sinopharm Group Chemical Reagent Company, China) were used for the simulation of the co-existing ions in aqueous solutions. Deionized water was employed through all the preparation processes.

### Preparation of the adsorbent

The original goethite tailings were obtained from Huangmei Concentration Plant (Hubei province, China). The goethite concentrate was then separated from such tailings by the high gradient magnetic separator after being grinded in the ball mill. At last, the porous hematite was prepared by calcinating the goethite concentrate in a muffle furnace (Vulcan 3-550 PD, Addlestone, UK). The calcination temperature was increased from 20 to 300 °C with a rate of 5 °C/min, then remained at 300 °C for 180 min and cooled to the room temperature. The final product was so-called the porous hematite which was used as the adsorbent through all the adsorption experiments.

### Adsorption of As(V)

0.1 g of adsorbent was first put into a flask. Then, 100 mL arsenic aqueous solution with an initial As(V) concentration of 10 mg/L was added into the flask for adsorption. 0.01 mol/L NaCl was also added as an electrolyte. Next, the arsenic(V) solution with the adsorbent inside was settled in a shaker (SHA-B, Guohua, China) at 125 rpm and 25 °C for 24 h. Until the adsorption equilibrium was reached, the solution was centrifuged at 6000 rpm for 5 min. In the end, the centrifuged supernatant was collected and analyzed using an atomic fluorescence spectroscopy (AFS-8220 Beijing Titan, China). The adsorption capacity of arsenic(V) was calculated by the following equation:

$$q = \frac{V_0(C_0 - C_E)}{m} \quad (1)$$

where  $q$  is the adsorption capacity of the adsorbent, mg/g;  $V_0$  is the initial volume of As(V) solution, L;  $C_0$  and  $C_E$  are the initial and the equilibrium arsenic(V) concentration, respectively, mg/L;  $m$  is the mass of the adsorbent used in the adsorption experiments, g. The adsorption isotherm can be derived from the adsorption capacity versus the initial As(V) concentration.

For the adsorption kinetics analysis, 2 mL of the solution was collected and filtered by a cellulose acetate membrane (0.22  $\mu\text{m}$  pore size), at a given adsorption interval time for the As(V) concentration measurements. Removal ( $\Phi$ ) of As(V) was determined as follows:

$$\Phi = \frac{C_0 - C}{C_0} \times 100\% \quad (2)$$

where  $C$  is the As(V) concentration at a given adsorption time,  $\mu\text{g/L}$ .

### Characterization

The crystal structures of the original goethite concentrate and the prepared porous hematite were recorded by X-ray diffraction (D8 Advance, Bruker, Germany) with Cu-K $\alpha$  radiation. The particle

size was determined using a laser particle analyzer (Mastersizer 2000, Malvern, UK). The chemical composition of the adsorbent was analyzed using X-ray Fluorescence (Axios advanced, PANalytical B.V., Netherlands). The specific surface area and pore properties of the goethite concentrate and the porous hematite were measured using a gas adsorption analyzer (F-sorb3400, Gold Aipu Technology Co., Ltd., China) at 77 K with nitrogen as the adsorbate. Zeta potential was also determined by a Zeta Probe analyzer (Nano-ZS90, Malvern, UK).

The concentrations of As(V) before and after adsorption were collected and diluted at different times for the data evaluation by Atomic Fluorescence Spectrometry (AFS-8220 Beijing Titan, China).

## Results and discussion

### Characterization of the adsorbent

The chemical composition of the goethite concentrate was given with values of 52.75 wt% Fe, 2.97 wt% Si, 4.10 wt% Ba, 0.88 wt% Al, 0.14 wt% Ca and 0.09 wt% As. Furthermore, X-ray diffraction (XRD) patterns of the as-received goethite tailings, goethite concentrate and porous hematite were compared in Fig. 1, in which G, H, Q labeled on the characteristic peaks represented goethite, hematite and quartz, respectively. Compared with the original goethite tailings, goethite concentrate contains less quartz and more goethite. However, there were no goethite peaks but hematite peaks, which indicated that the goethite concentrate had completely transformed into hematite at 300 °C. In addition, the pore-volume characterization of goethite and porous hematite are listed in Table 1. The specific surface area was increased from  $8.891 \pm 0.045 \text{ m}^2/\text{g}$  to  $73.458 \pm 1.39 \text{ m}^2/\text{g}$ . The micropores volume was increased approximately four times. The formation of micropores and the increased surface area was mainly attributed to the dehydration and dehydroxylation of the goethite concentrate [8], which may be proved by the AFM observation in Fig. 2(b).

SEM and AFM images in Fig. 2 confirmed the porous morphology of the prepared hematite by calcination. It could be seen that the adjacent micropores were merged into mesoporous or macroporous. Furthermore, the pore size of the porous hematite became smaller compared to that of the goethite. The particle size distribution curve of the porous hematite was illustrated in Fig. 3. As it can be seen,  $D_{50}$  and  $D_{90}$  size was determined 34.526 and 235.332  $\mu\text{m}$ , respectively. Weighted mean of the particle size was 77.860  $\mu\text{m}$ .

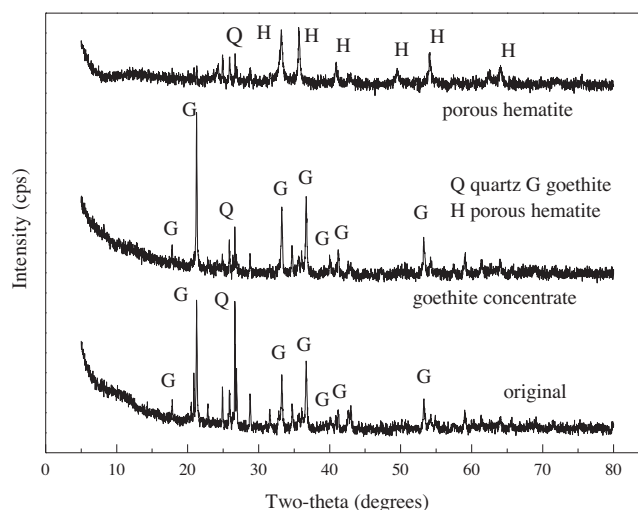


Fig. 1. X-ray diffraction patterns of the goethite concentrate and porous hematite (cps, counts per second).

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