

Individual and competitive adsorption of Cr(VI) and phosphate onto synthetic Fe–Al hydroxides

Xin-Hua Wang^a, Fei-Fei Liu^a, Lei Lu^a, Sen Yang^a, Yu Zhao^a, Lin-Bio Sun^b, Shu-Guang Wang^{a,*}

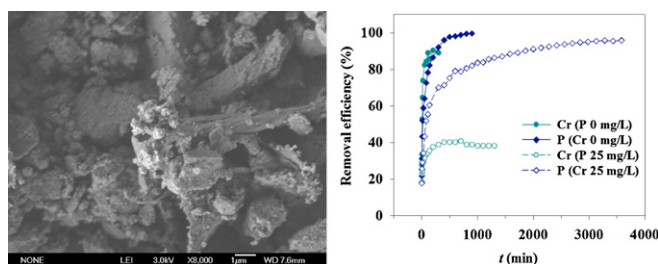
^a Shandong Provincial Key Laboratory of Water Pollution Control and Resource Reuse, School of Environmental Science and Engineering, Shandong University, Jinan 250100, PR China

^b Everbright Water (Jinan) Company Limited, Jinan 250013, PR China

HIGHLIGHTS

- ▶ Fe–Al hydroxides have been successfully synthesized.
- ▶ Individual and competitive adsorption of Cr(VI) and P was investigated.
- ▶ Pseudo-second-order kinetic and Langmuir models fit the individual adsorption.
- ▶ P reduced Cr(VI) adsorption amount, but Cr(VI) decreased P adsorption rate.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 3 October 2012

Received in revised form

27 December 2012

Accepted 22 January 2013

Available online 30 January 2013

Keywords:

Fe–Al hydroxides

Cr(VI)

P

Adsorption

Individual

Competitive

ABSTRACT

Individual and competitive adsorption characteristics of Cr(VI) and phosphate (P) onto synthetic Fe–Al hydroxides were investigated. A series of batch experiments were performed to study the influence of various experimental parameters (pH, ionic strength and contact time) on Cr(VI) and P adsorption. Removal efficiency of Cr(VI) and P decreased with increasing pH and ionic strength. In individual adsorption tests, pseudo-second-order kinetic and Langmuir isotherm models could better describe Cr(VI) and P adsorption on Fe–Al hydroxides. In competitive adsorption experiments, Cr(VI) adsorption was suppressed obviously by P and followed Freundlich isotherm model, revealing that the competitive adsorption of Cr(VI) in the presence of P was heterogeneous. Cr(VI) had no significant influence on P adsorption and the competitive adsorption of P agreed well with Langmuir isotherm model, suggesting that the competitive affinity of P towards the adsorption sites on Fe–Al hydroxides was stronger than that of Cr(VI). The presence of P reduced the adsorption amount of Cr(VI), but the presence of Cr(VI) decreased the adsorption rate of P.

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1. Introduction

With the rapid development of industrialization and urbanization in recent years, significant amount of wastewaters are produced with toxic heavy metals and nutrient substances [1,2]. As one of the priority contaminants, Cr(VI) is generally present in surface and ground waters. Various industries such as electroplating, metal cleaning, leather processing and mining discharge

Cr(VI)-contained wastewaters with varied concentrations (5–220 mg/L) into the aquatic environment [3]. Due to its strong oxidizability and high toxicity, Cr(VI) is always viewed as a carcinogen and mutagen [4,5]. According to US EPA, the maximum level permitted of Cr(VI) in drinking water and inland surface waters were 0.05 and 0.1 mg/L, respectively [6]. Phosphate (P) is one of the essential nutrients for plant growth. However, excess P has drawn great attention during these years as it can cause serious eutrophication in lakes and reservoirs [7]. The main sources of P in water are due to pigments, detergents, electronic industry, mineral processing, and the overuse of P-contained fertilizers [8]. Excessive P can result in the intense

* Corresponding author. Tel.: +86 531 88362802; fax: +86 531 88364513.
E-mail address: wsg@sdu.edu.cn (S.-G. Wang).

accumulation of algae, which leads to the decrease of aquatic life [9]. Besides, large amount of algae may produce toxins and threaten to human health. Therefore, removal of Cr(VI) and P from wastewater before discharge into environment is significantly important from the environmental and healthy point of view.

There are various treatment methods for Cr(VI) removal, i.e. electrocoagulation [10,11], chemical precipitation [12], chemical reduction [13,14], membrane separations [15], ion exchange [16], photocatalytic reduction [17], and adsorption [18–21]. For P removal, numerous methods including chemical precipitation [22], crystallization treatment [23,24], microfiltration [25], membrane bioreactors [26], and adsorption [27–30] have been widely used. Cr(VI) and P can also be removed simultaneously by some novel techniques, such as liquid–liquid extraction [31] and hybrid anion exchange–precipitation [32]. Among these technologies mentioned above, adsorption is one of the most promising methods due to its low operation costs, flexibility of design and high efficiency.

In the recent years, many researchers have devoted their contribution to the development of low-cost and effective adsorbents. As novel and efficient adsorbent, metal hydroxides have been used to remove toxic contaminants [33–35]. Fe or Al hydroxides have great applications in industries due to their unique adsorptive properties for anions and cations. Although Fe or Al hydroxides have been used in water treatment, the application of mixed Fe–Al hydroxides is scarce. Sujana and Anand used Fe–Al hydroxides to remove fluoride and obtained high removal efficiency [35]. Therefore, it is anticipated that other anions such as Cr(VI) and P can also be removed by Fe–Al hydroxides.

In this work, we attempted to synthesize and use Fe–Al hydroxides as an adsorbent to remove Cr(VI) and P from aqueous solution. The objective of this study was to investigate the influencing factors (pH, ionic strength and contact time), kinetics and isotherms of individual adsorption of Cr(VI) and P on Fe–Al hydroxides. Besides, the competitive adsorption characteristics of Cr(VI) and P on Fe–Al hydroxides were also studied.

2. Materials and methods

2.1. Synthesis and characterization of Fe–Al hydroxides

Fe–Al hydroxides used in this study were prepared by a modified co-precipitation method [36]. In brief, solutions of FeCl₃ (0.1 M) in 0.01 M HCl and AlCl₃ (0.1 M) in 0.01 M HCl were mixed together and heated to 65 ± 1 °C. Then, NH₃·H₂O (1:1) was dropped into the mixture under vigorous stirring until the solution pH reached 7.0 ± 0.1. After 2 days of ageing, the precipitate was separated by filtration, washed twice and dried in an oven at 105 °C for 24 h. Finally, the resulting dried product was grinded and sieved to pass 60–100 meshes by standard sieves and stored in a desiccator for further use.

A series of techniques were employed to characterize the adsorbent. The morphology of Fe–Al hydroxides was observed with a scanning electron microscope (SEM) (JSM-6700F, Japan). N₂ adsorption–desorption isotherms were performed at 77 K with a surface area analyzer (JW-BK 122W, China). BET equation and BJH method were used to calculate the surface area and pore size of Fe–Al hydroxides. Fourier transform infrared spectroscopy (FTIR) spectrum was recorded in Avatar 370 spectrometer (Thermo Nicolet, U.S.) within the range of 400–4000 cm⁻¹. The hydrodynamic diameter of Fe–Al hydroxides was measured using Mastersizer 2000 (Malvern, UK). Zeta potential variation with pH was measured with a zeta potential analyzer (JS94H, China). The point of

zero charge (pH_{PZC}) of Fe–Al hydroxides was determined when zeta potential equaled zero.

2.2. Individual adsorption of Cr(VI) and P

All chemicals used in this study were of analytical grade. K₂Cr₂O₇ (Tianjin Kelme Chemical Reagent Co., Ltd.) and KH₂PO₄ (Tianjin Guangcheng Chemical Co., Ltd.) were employed to prepare Cr(VI) and P stock solutions (200 mg/L), respectively. Working solutions of required concentrations were achieved by diluting the stock solution with deionized water.

The effects of initial pH and ionic strength on adsorption were conducted using a series of flasks and a thermostatic shaker. The influence of initial solution pH on Cr(VI) or P adsorption was investigated in the pH range of 3–10 at 25 °C. The pH was adjusted with 0.1 M KOH and 0.1 M HNO₃ and the concentration of Cr(VI) or P was 25 mg/L. The influence of ionic strength on Cr(VI) or P adsorption was examined by 0.01 M and 0.1 M KNO₃. The initial concentrations of Cr(VI) or P were in the range of 10–200 mg/L at pH 4.5.

Adsorption kinetics experiments were carried out by adding 1000 mL Cr(VI) or P solution (25 mg/L, pH 4.5) and 1 g of Fe–Al hydroxides in a beaker. Then the beaker was placed on a magnetic stirrer at a speed of 125 rpm with a temperature control of 25 °C. The stirring time for Cr(VI) and P adsorption was 0–300 min and 0–900 min, respectively. The samples were taken at predetermined time intervals for analysis.

Adsorption isotherm studies were conducted in a thermostatic shaker at 25 °C. Fe–Al hydroxides (0.1 g) and 100 mL of Cr(VI) or P solution (10–200 mg/L, pH 4.5) were transferred into the flasks and shaken at 125 rpm for 24 h. All experiments were conducted in duplicates and the mean values were recorded. Control experiments were simultaneously carried out, confirming that the adsorption of Cr(VI) or P on the flask walls or in the filtration processes were negligible.

2.3. Competitive adsorption of Cr(VI) and P

Competitive adsorption experiments were also conducted at 25 °C. To investigate the effect of P on Cr(VI) adsorption, 10 or 50 mg/L of P was mixed with Cr(VI) solutions (10–200 mg/L), then 1 g/L of Fe–Al hydroxides was added and shaken at 125 rpm for 72 h. The effect of Cr(VI) on P adsorption was also studied in the same way. Briefly, 10 or 50 mg/L Cr(VI) was added to P solutions (10–200 mg/L), then Fe–Al hydroxides (1 g/L) was mixed and shaken at 125 rpm for 72 h. To investigate the competitive adsorption kinetics, Cr(VI) (25 mg/L) and P (25 mg/L) was mixed together and Fe–Al hydroxides (1 g/L) was added to the mixture, then the samples were taken at predetermined time intervals to analyze the concentrations of Cr(VI) and P.

2.4. Analytical methods

All the samples were filtered through 0.45 μm membranes before measurement. The concentrations of Cr(VI) and P were determined spectrophotometrically by diphenyl carbazide and molybdate methods, monitoring the absorbance at 540 nm and 640 nm on a UV–vis spectrophotometer (unico UV-2000) [37], respectively.

3. Results and discussion

3.1. Characterization of Fe–Al hydroxides

SEM images (Fig. 1) show that the surface of Fe–Al hydroxides we synthesized was rough, which was in accordance with other literature [35]. The non-uniform surface of the adsorbent may provide

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