



Adsorption of arsenic onto an environmental friendly goethite-polyacrylamide composite

Kardia Ramirez-Muñiz^a, Fátima Perez-Rodriguez^b, Rene Rangel-Mendez^{a,*}

^a IPICYT/División de Ciencias Ambientales, Camino a la presa San José 2055 Col Lomas 4ta Sección, C.P. 78216 San Luis Potosí, S.L.P., Mexico

^b CONACYT-IPICYT/División de Ciencias Ambientales, Camino a la presa San José 2055 Col Lomas 4ta Sección, C.P. 78216 San Luis Potosí, S.L.P., Mexico

ARTICLE INFO

Article history:

Received 28 March 2018

Received in revised form 11 May 2018

Accepted 14 May 2018

Available online xxxx

Keywords:

Adsorption

Arsenic

Goethite-P(AAm) composite

Friendly environmental adsorbent

ABSTRACT

Arsenic adsorption in aqueous solutions onto a novel goethite-P(AAm) (Poly(Acrylamide)) composite, in batch and continuous processes, has been studied. This work included the synthesis and characterization of a novel goethite-P(AAm) composite. For the synthesis of the composite, three grades of crosslinking and the goethite content were evaluated. The optimal synthesis conditions were determined by using the response surface methodology. The selected goethite-P(AAm) composite was characterized by swelling, surface area, thermogravimetric analysis (TGA), scan electron microscopy (SEM) and infrared spectroscopy (FTIR). The adsorption capacity of arsenic on the goethite-P(AAm) composite was 1.22 mg g^{-1} , comparable to that of powder goethite. The immobilization mechanism of goethite in Poly(Acrylamide) was attributed to hydrogen bonding between goethite and hydroxyl and amines groups of P(AAm), as well as to acid and basic Lewis interactions. The adsorption isotherms were best fitted by the Langmuir model, and the adsorption process is promoted in acid solution. The column experiments demonstrated that the goethite-P(AAm) composite can supply 400 BV of treated water with less than 25 ppb, this means that only 1.53 kg of composite could be needed to supply the monthly consumption of domestic water for one person. These results suggest the potential application of goethite-P(AAm) composite like a friendly environmental adsorbent.

© 2018 Elsevier B.V. All rights reserved.

1. Introduction

Arsenic in natural water has been a worldwide environmental problem as it provokes toxic effects on biota, and long exposure to this contaminant could lead to health problems, such as cancer [1,2]. The toxicity of arsenic is attributable to the fact that arsenic is similar enough to phosphorus for organism to attempt to replace phosphorus by arsenic, this hinders the essential chemistry process of the phosphorus [3]. According to the World Health Organization guidelines, the recommended limit of arsenic in drinking water is $10 \mu\text{g L}^{-1}$, however, the arsenic concentration in groundwater in different countries around the world goes from 0.5 to $5000 \mu\text{g L}^{-1}$ [4]. Different methods have been employed to remove arsenic, such as ion exchange [5], reverse osmosis [6], coagulation [7], electrocoagulation [8], filtration and adsorption [9]. Among these methods, adsorption has many advantages such as high removal efficient, low cost, easy operation and not require high amount of energy or additional chemicals.

Several materials have been tested for arsenic removal from water, including iron oxides or hydroxides, either natural or synthetics [10]. Gimenez et al. [11] studied the adsorption of arsenic (III and V) onto

different natural iron oxides (hematite, magnetite and goethite) as a function on the different parameters. The kinetics data for the three iron oxides showed that the equilibrium is reached in <2 days. Arsenic adsorption on synthetic goethite and jarosite at acid pH and two ionic strengths was studied [12]. On the other hand Mamindy-Pajany et al. [13] reported the adsorption of arsenic onto commercially available goethite and hematite. The adsorption experiments for these iron oxides showed that there was no effect of the ionic strength on arsenate adsorption suggesting the formation of an inner-sphere surface complex.

It is well-known that iron oxides are good adsorbents for arsenic removal from water, also they are no toxic and have a high natural affinity for arsenic. However, in order to improve their adsorption capacities, the size of iron oxide is crucial. Though, powder materials are difficult to separate from treated water and/or have some drawbacks for their application in packed bed columns. Recently, the development of hybrid adsorbents composed by iron oxide-polymer potentially overcomes the limitations already mentioned [14–18]. Santos et al. [19] synthesized iron oxyhydroxide nanoparticles contained in PVA microspheres. They produced iron nanoparticles simultaneously with PVA three-dimensional network crosslinking. The maximum arsenic adsorption of this composite was 87.18 mg g^{-1} at the pH range 2–5. The use of nanoparticles gives better results to remove arsenic than granular

* Corresponding author.

E-mail address: rene@ipicyt.edu.mx (R. Rangel-Mendez).

adsorbents [20]. However, there are two major challenges to overcome; one is the non-availability of nanoparticles at economically affordable prices and the other is the toxicity and the environmental fate of nanoparticles [21].

Hydrogels, such as P(AAm) (polyacrylamide), consist of three-dimensional flexible polymeric networks that are able to absorb a large amount of water in their swollen state. Specific properties of hydrogels can be obtained by controlling the synthesis parameters such as type of reactor, reaction temperature, reaction time, type and respective amount of ingredients [22]. The aim applications of hydrogels are like drug delivery vehicles, sensors, matrix materials for tissue engineering, adsorbents for environmental applications, and as antimicrobial materials. In addition to these applications hydrogels can be used as support materials for the synthesis of metal magnetic particles and nanostructures [23].

In another hand, the treatment of arsenic contaminated water generated a large quantity of sludge with a high leaching potential [24]. The production of municipal sewage sludge in EU was estimated to be 11.5 million tons in 2010, which is expected to increase to 13.0 million tons by 2020 [25]. Therefore, it is necessary to synthesize effective and environmental friendly novel materials for water treatment.

According to the information in the open literature, to date there is not report of arsenic removal from water by immobilized goethite on a hydrogel (adsorbent material named in this article as goethite-P(AAm) composite). The aim of this work is to synthesize a composite to combine the properties of iron oxide (goethite) and the features of the hydrogel to have an environmental friendly adsorbent with proper mechanical properties, chemical stability and adsorption performance. Selected materials were physically and chemically characterized, and the best adsorbent material was evaluated in batch and column test for arsenic removal: kinetic studies were also carried out in order to understand the adsorption phenomena. Additionally, the effect of pH and ionic strength on arsenic removal was studied.

2. Experimental methods

2.1. Materials

The reagents for the synthesis of hydrogels were acrylamide monomer (AM, 99%), ammonium per sulfate (APS), *N,N'*-Methylenbis (acrylamide, MBA, 99%) and goethite (FeOOH, 20–63% Fe, named in this work as Goethite AD) of analytical grade from Sigma-Aldrich (USA). All reagents were used without further purification.

Sodium arsenate ($\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$) was used for the preparation of arsenic solution, sodium hydroxide (NaOH) and hydrochloric acid (HCl) were used to adjust the solutions pH, all of which were of analytical grade from Sigma-Aldrich (USA). Deionized water ($<11 \mu\text{S}\cdot\text{cm}^{-1}$) was used in all experiments.

2.2. Methods

2.2.1. Preparation of goethite-PAM composite

A mixture of acrylamide and initiator in deionized water were utilized in measured proportions. The monomer concentration was 1.33 g L^{-1} and the ratio of the initiator to monomer was set at 0.66 g L^{-1} [22]. The reaction was purged by nitrogen for 30 min and then conducted under nitrogen atmosphere at $60 \text{ }^\circ\text{C}$ for 1 h. The polymer was washed with acetone, cut in a regular shape and then dried in an oven at $90 \text{ }^\circ\text{C}$ for 12 h. The goethite-P(AAm) (goethite/polyacrylamide) hydrogel was prepared by adding goethite at different concentrations into the P(AAm) via in-situ free radical polymerization [26]. The adsorbents were synthesized at three levels of crosslinking agent (MBA; 2.66, 16.66 and 33.33 g L^{-1}) and different content of goethite AD (FeOOH; 0, 6.66, 16.66, 33.33, 66.66, 200, 233.33 and 266.66 g L^{-1}); these materials were named as G-1 to G-10 and H-1 to

H-3 for goethite-P(AAm) composite and P(AAm)-Hydrogel without goethite, respectively.

2.2.2. Adsorption of arsenic onto goethite-P(AAm) composite

In this work, the batch tests were carried out by placing 25 mg of adsorbent in 20 mL of a solution containing arsenic with an initial concentration of 0.1 to 3 mg L^{-1} . Batch experiments were placed in a chamber at $25 \text{ }^\circ\text{C}$ and $120\text{--}130 \text{ rev}\cdot\text{min}^{-1}$. The pH sample was adjusted to 7 ± 0.2 by adding 0.1 HCl or NaOH until the equilibrium was obtained. The solution was removed from the adsorbent by decantation, then, arsenic and iron concentration were analyzed by inductively coupled plasma atomic emission spectroscopy (ICP-AES), by a Varian spectrometer 730-ES, at a wavelength of 193.696 and 259.94 nm , respectively. The particle size of dryer goethite-P(AAm) composite used in batch experiments were $0.8\text{--}3 \text{ mm}$.

The amount of adsorbate uptake q_e ($\text{mg}\cdot\text{g}^{-1}$), was calculated by the following mass balance:

$$q_e = \frac{V}{m} (C_0 - C_e) \quad (1)$$

where C_0 and C_e are the initial and equilibrium concentrations ($\text{mg}\cdot\text{L}^{-1}$), V is the volume of the solution (L), and m the mass of the adsorbent (g).

The adsorption isotherms of selected adsorbents were fitted by Langmuir model.

2.2.3. Determination of adsorption kinetics of arsenic onto goethite-P(AAm) composite

The adsorption kinetics was performed in 50 mL conical bottom polypropylene tubes. 100 mg of the adsorbent was immersed in 20 mL of arsenic solution of 4.5 mg L^{-1} . These samples were conducted in a thermostatic orbital shaker at $25 \text{ }^\circ\text{C}$ and $120\text{--}130 \text{ rev}\cdot\text{min}^{-1}$. The concentration of arsenic in solution was determinate at different time intervals.

2.2.4. Characterization of the goethite-P(AAm) composite

The swelling behavior of Goethite-P(AAm) composite was performed to verify changes in their size in aqueous medium. Swelling was carried out in triplicate at $25 \text{ }^\circ\text{C}$. % Swelling was calculated by the following mathematical expression:

$$\%S = \frac{(M_t - M_i)}{M_i} \times 100 \quad (2)$$

where, M_i and M_t are the initial mass and the mass at time of 2, 4, 6, 8 24 and 48 h.

Thermal behaviour of goethite-P(AAm) hydrogels was studied using thermal gravimetric analysis (TGA). 50 mg of the samples were placed in a ceramic crucible and heated from ambient temperature to $1000 \text{ }^\circ\text{C}$ under a flow of nitrogen of 100 mL min^{-1} at a heating rate of $10 \text{ }^\circ\text{C min}^{-1}$ on a Thermo Cahn Versa Term.

A Micromeritics Accelerated Surface Area and Porosimetry (ASAP) analyser 2020 was used to determine the specific surface area and pore properties of the goethite-P(AAm) hydrogels at 77 K with nitrogen as the adsorbate. Previously, 0.5 g of sample were frozen at $-40 \text{ }^\circ\text{C}$, followed by lyophilization in a freeze dry system Labconco. Then, the sample was degassed for 12 h, before analysis. The surface area (S_{BET}) of the sample was determined by using the BET equation, in which the cross-sectional area of nitrogen was selected as 0.162 nm^2 .

A FEI-QUANTA 250 scanning electron microscope was used to obtain the SEM image of the goethite-P(AAm) composite.

In order to maintain the structure of hydrogel, the wet sample was frozen at $-40 \text{ }^\circ\text{C}$ for 24 h, followed by lyophilization. Then, the sample was carefully adhered on a thin piece of metal, and then coated with 3 nm gold film. SEM micrographs were taken using a voltage acceleration of 20 to 25 kV.

Download English Version:

<https://daneshyari.com/en/article/7842098>

Download Persian Version:

<https://daneshyari.com/article/7842098>

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