



Research paper

Evaluation of the mixed oxides produced from hydrotalcite-like compound's thermal treatment in arsenic uptake



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ABSTRACT

In this work, hydrotalcite-like compounds (prepared by a simple and environmentally-friendly method) were evaluated as adsorbent's precursors for arsenic (V) removal in water. Materials were characterized by X-ray powder diffraction, N₂ adsorption-desorption at −196 °C and thermogravimetric analyses. It was revealed by the XRD characterization that the calcined samples exhibited the characteristic reflections of the MgO and spinel crystalline phases. The thermal treatment increase gave rise to a specific surface area increment achieving a maximum at 550 °C. The effect of adsorbents' activation, reaction time, adsorbent amount, pH and initial arsenic concentrations were evaluated. Results showed that removal efficiency of arsenate increased with the increment of the annealing temperature reaching a maximum removal with the calcined sample at 550 °C. The adsorption isotherms can be well described by Freundlich model and adsorption kinetics of arsenate was adjusted by the pseudo-second-order, indicating that chemisorption was the limit step. It was demonstrated that removal capacity depends on the initial arsenic concentration where more than 98% removal was achieved with initial concentrations below 250 µg/L.

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1. Introduction

Arsenic (As) has been included in Group I carcinogen by the International Agency for Research on Cancer (IARC). In this sense, it is well known that the continuous ingestion of waters with high As concentrations induces the appearance of arsenicosis. Long-term drinking As-containing water exposure causes skin, lung, bladder, and kidney cancer as well as pigmentation changes, skin thickening (hyperkeratosis) neurological disorders, muscular weakness, loss of appetite and nausea (Mohan and Pittman, 2007). Considering the toxic effects of As, the World Health Organization (WHO) recommends a maximum concentration of 10 µg/L (10 ppb) of As in drinking water (WHO, 2004). However, many countries have established higher maximum As concentration limits. For instance, in Mexico the maximum As concentration limit is 25 µg/L. Although in Mexico less rigorous normativity is applied compared to other countries, As in groundwater has become a serious problem in regions such as Hidalgo (up to 1100 µg/L) and La Comarca Lagunera (0.24–1 mg/L) (Parga et al., 2005), San Luis Potosí (Martínez-Villegas et al., 2013), Guanajuato (50 µg/L) (Flores et al., 2013) and Baja California Sur (up to 450 µg/L) (Wurl et al., 2014), among many others. Moreover, this problem demands immediate

attention considering that in Mexico, ~75% of the total population relies on groundwater for drinking (Alarcón-Herrera et al., 2013).

In the group of the available technologies for As removal (oxidation, phytoremediation, coagulation–flocculation, ion-exchange, electrochemistry and membrane technologies), adsorption has attracted much attention due to the following advantages: (a) it usually does not need a large adsorbent volume and additional chemicals, (b) it is easy to set up as a point of entry/point of use As removal process and (c) it does not produce harmful by-products and can be more cost effective. In this context, adsorption process has been used most widely because of its easy operation and handling, high removal efficiency, and low cost (Singh et al., 2015).

Among the reported adsorbents employed for arsenic removal (see Mohan and Pittman, 2007 and references therein), a number of studies have focused on the layered double hydroxide (LDH) materials (in particular hydrotalcite and hydrotalcite-like compounds) (Goh et al., 2008, 2009; Violante et al., 2009; Grover et al., 2010; Palmer and Frost, 2010; Liang et al., 2013;) since these compounds offer a large interlayer surface to host diverse anionic species with the additional advantage of being potentially recyclable (Dadwhal et al., 2009).

It is worth mentioning that although distinct synthesis methods have been developed to prepare such LDH, the coprecipitation method is generally preferred (Gillman, 2006; Bujdosó et al., 2009; Wang et al., 2009; Dadwhal et al., 2011; Türk and Alp, 2014). Nevertheless, it is important to recognize that this method presents environmental, process and economic drawbacks restricting their large-scale production

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(Valente et al., 2008, 2009). An alternative is given by the use of raw materials which do not introduce undesirable anions into the process, adjusting the process variables such as pH, aging temperature and time. Valente et al. (2008, 2009) developed a sustainable method for hydrotalcite and hydrotalcite-like compounds' synthesis where the raw materials are insoluble oxides and materials are obtained under mild conditions and short synthesis times. Moreover, a minimal amount of water is required, raw materials are cheaper, readily available and washing or purifying of the final product is not necessary. All these issues make this method a sustainable and potentially viable method to produce high amounts of sorbents which could fulfill the growing demand of As sorbents.

It is important to remark that even though Valente et al. (2009) demonstrated that their materials exhibited similar physicochemical characteristics compared to those prepared by coprecipitation, it is necessary to evaluate them to ensure that the produced materials can be used in applications such as As removal.

Thus, in this work arsenic (V) adsorption was selected in order to prove our material's uploading capacity. It has been reported that MgAl LDH synthesized by conventional procedures can remove As(III) and As(V) species, where monolayer adsorption equilibrium (Langmuir) describes the uploading process and relates the homogeneity of the adsorbate-adsorbent system (Wan et al., 2012). However, our experimental adsorption data was fitted to the Freundlich isotherm which is widely used to describe adsorption on a surface with a heterogeneous energy distribution (Jiménez-Cedillo et al., 2013). In the same way, adsorption data of this work revealed adsorbents' heterogeneous surface.

Therefore, a MgAl LDH with a nominal Mg/Al molar ratio of two was synthesized and evaluated in As (V) removal. The effect of various factors such as adsorbents' activation temperature, reaction time, adsorbent amount, pH and initial As concentrations were tested.

2. Experimental section

2.1. Materials

MgO (96.57%), boehmite HiQ-10 (99.89%) and HNO₃ (66.1%) were purchased from Industrias Peñoles (México), Engelhard Corporation (USA) and J. T. Baker Chemicals (México), respectively.

2.2. Hydrotalcite-like compound synthesis

MgAl with a nominal molar ratio of two was prepared considering the general formula $[M^{2+}_1 - xM^{3+}_x(OH)_2](NO_3)_{x/n} \cdot mH_2O$, according to the procedure described by Valente et al. (2008, 2009). The sample was prepared as follows: 15 g of MgO were dispersed at 5000 rpm in 73 mL of deionized water for 30 min (A). 11 mL of HNO₃ were dissolved separately in 263 mL of deionized water (B1). Then, 12.1 g of boehmite were added to (B1) and the mixture was dispersed at 5000 rpm for 30 min (B2). Then, (A) was added to (B2) and the slurry was dispersed for 30 min at 8000 rpm. The slurry was aged at 80 °C for 6 h at 350 rpm. After aging, the sample was only filtered to remove the excess of water and dried at 100 °C overnight. After drying the samples were calcined under a static air atmosphere for 2 h at the indicated temperature.

2.3. Analytical methods

The X-ray diffraction patterns of the solid samples were acquired in a D8 Bruker Discover Series 2 diffractometer with CuK_α radiation. The samples were measured between 5 and 70°, with a 2θ step of 0.04° and a counting time of 0.6 s per point. The crystalline phases were identified by means of the JCPDS (Joint Committee of Powder Diffraction Standards) database. Average crystal sizes were determined by Scherrer equation: $L_{(hkl)} = K\lambda / (B(\theta)\cos\theta)$ where K is the shape factor (a value of 0.9 was used), L is the average crystal size,

λ is the average wavelength of CuK_α radiation ($\lambda = 1.5418 \text{ \AA}$), $B(\theta)$ is the Full Width at Half Maximum (FWHM), θ is the diffraction angle and hkl are Miller indices.

Thermogravimetric (TG) analysis was developed on a TGAi 1000 Series System (Instrument Specialists, WI, USA) which was operated under a nitrogen flow at a heating rate of 10 °C/min from 25 to 800 °C. In the determination, ~40 mg of finely powdered dried sample was used.

Specific surface areas were obtained from N₂ adsorption-desorption at -196 °C in a ChemiSorb 2750 (Micromeritics, GA, USA) using the one-point BET method. Prior to the analysis the samples were thermally treated for 1 h at 120 and 300 °C for the fresh and calcined samples, correspondingly.

2.4. Adsorption measurements.

A Perkin-Elmer Precisely Model 400 spectrophotometer AA analyst equipped with a Hydride Generator System (MHS 15) with an EDL 2 lamp of 400 mA was used to determine As(V) concentration. The spectrophotometer was previously calibrated, based on Perkin-Elmer, USA standards. The reagents utilized were Merck trademark, purity ≥ 98%. Aqueous solutions of NaOH (0.25 mol/L and HCl (0.15 mol/L) were used for NaBH₄ (0.8 mol/L) preparation. The gases used were nitrous oxide (N₂O, 6 L/min), acetylene (C₂H₂, 2.5 L/min) and Argon as carrier. The determinations were conducted within a purging time of fifty-second pre-reacting and forty-second post-reaction, respectively. Samples were measured by triplicate and the relative standard deviation was calculated giving a value of 0.5% which was used to plot error bars in Fig. 5.

2.5. Batch sorption experiments

As(V) solutions used for the adsorption experiments were prepared by dilution from a 1000 mg/L ICP standard solution (As(V) in 5% HNO₃ purchased from Perkin Elmer) using deionized and decarbonated water. To evaluate the operational variables, several experimental conditions such as i) calcination temperature, ii) mass of the adsorbent, and iii) initial pH solution, were tested. In all these experiments the initial As(V) concentration was 1000 µg/L, adsorption temperature 25 °C, 2 h of contact time between the adsorbate and adsorbent, 0.05 g of adsorbent and 0.05 L of solution, unless otherwise noted. Thus, the effect of the calcination temperature over the adsorption of As(V) by the LDH was evaluated from 450 to 650 °C; the influence of the mass of the adsorbent was evaluated from 0.01 to 0.05 g of LDH and the impact of the initial solution pH was determined from 6 to 10. The solution pH was adjusted with a 0.1 M solution of NaOH and 1% v/v of acetic acid for basic and acid solutions, respectively.

Adsorption kinetics was followed in a jacketed, magnetically stirred batch reactor charged with 1000 µg/L of aqueous As solution. The temperature was stabilized at 25 °C and then the adsorbent was added to the solution. The solution volume was 0.05 L and the adsorbent's mass was 0.05 g to keep a mass-volume ratio of 1. Once adsorption was completed, the samples were filtered and the residual As(V) concentration in the solution was determined.

The amount of As(V) adsorbed at any time $q(\mu\text{g/g})$, was calculated from Eq. 1.

$$q = (C_0 - C_t) * V / m \quad (1)$$

where C_0 is the initial As concentration (µg/L), C_t is the As concentration at time "t" (µg/L), m is the mass of the adsorbent (g) and V is the volume of As solution (L).

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