



Adsorption of As(V) from water using Mg–Fe-based hydrotalcite (FeHT)

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ABSTRACT

This paper describes a study of the sorptive removal of arsenate (As(V)) from aqueous solutions by synthetically prepared Mg–Fe-based hydrotalcites (FeHT) as layered double hydroxide (LDH) adsorbents. The synthesis of Fe³⁺-substituted hydrotalcites (FeHT) with the chemical formula [Mg(II)₆Fe(III)₂(OH)₁₆]²⁺[CO₃ × yH₂O]²⁻ was achieved by a co-precipitation method. The reaction products were characterized by powder X-ray diffraction analysis. The influences of solution pH, initial arsenate (As(V)) concentration, and sorbent concentration were investigated in multiple kinetic runs. The adsorption rates and isotherms were investigated in batch experiments. The pseudo-first-order and second-order kinetic models were tested and the latter was found to fit better to the experimental data. Langmuir and Freundlich isotherms were used to describe the adsorption data from equilibrium experiments. The results have shown that FeHT has a high arsenate removal efficiency, with the ability to reduce the concentration of arsenate in the aqueous solution from an initial value of 330 μg/l to <10 μg/l (i.e. below the limit value specified by WHO).

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1. Introduction

Concentrations of arsenic in open ocean seawater are typically in the range of 0.001–0.002 mg/l. Arsenic is widely distributed in surface freshwaters, and concentrations in rivers and lakes are generally below 0.01 mg/l, although individual samples may be as high as 5 mg/l near anthropogenic sources. The arsenic levels in groundwater are typically in the range of 0.001–0.002 mg/l, except in areas with volcanic rock and sulfide mineral deposits where arsenic levels can be as high as 3 mg/l [1].

Arsenic is introduced into water through the dissolution of minerals and ores, from industrial effluents and via atmospheric deposition [2]. Natural sources, such as the dissolution of arsenic-containing bedrock, often contribute significantly to the arsenic content of drinking water and groundwater [3]. Although environmental restrictions and regulations have limited the production and uses of arsenic and its compounds, they are still extensively used in various industries, including metallurgy, agriculture, forestry, electronics, pharmaceuticals and the glass and ceramic industry [4].

Arsenic can exist in different oxidation states in aqueous environments, i.e., +5, +3, 0, –3, but is predominantly present in the trivalent (+3) and pentavalent (+5) states. Inorganic forms of arsenic can exist in natural waters at acidic and under reducing conditions [5]. The distribution of arsenic species (As(III), As(V)) in natural

waters is mainly dependent on the redox potential and pH conditions of the water [6].

The presence of arsenic in drinking water is receiving increasing worldwide attention. The WHO guideline value for As in drinking water was provisionally reduced in 1993 from 0.05 to 0.01 mg/l. The newly recommended value was based on the increasing awareness of the toxicity of As (particularly its carcinogenicity), and on the ability to quantitatively measure As concentrations [7]. The European Union, the United States and many other countries have established a value of 0.01 mg/l arsenic as the maximum contaminant level for total arsenic in potable water. Therefore, the development of removal methods for inorganic arsenic from drinking water is urgent.

Arsenic cannot be destroyed, but can be transformed into different forms or converted into insoluble compounds in the presence of other elements. Therefore, it is essential to develop novel and efficient methods for arsenic removal from drinking water. As a result of the lower safety threshold of 0.01 mg/l, many methods have been developed to remove excessive arsenic from water using techniques such as coagulation, electro coagulation, precipitation, ion exchange, reverse osmosis and adsorption [8].

Among these methods, the adsorption technique is gaining popularity due to its simplicity, potential for regeneration and sludge-free operation [9]. Furthermore, there is potential for adsorption to be an attractive technology if the adsorbent can be made cheaply and ready for use. Many kinds of adsorbents such as modified red mud [4], alumina-modified zeolite [5], natural iron mineral [9], synthetic goethite [10], iron oxide-loaded slag [11] and zero-valent iron [12] have been developed for the removal of

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arsenic. A number of studies have also been done on arsenic sorption onto hydrotalcite minerals [13–23].

Hydrotalcite and hydrotalcite-like compounds have a layered double hydroxide structure and have been known as inorganic anion-exchangers with the chemical formula $M_x^{2+}M_y^{3+}(\text{OH})_{2(x+y)}A_y/n \times m\text{H}_2\text{O}$ ($M^{2+}:\text{Mg}^{2+}, \text{Zn}^{2+}, \dots$; $M^{3+}:\text{Al}^{3+}, \text{Fe}^{3+}, \dots$; where A^{n-} is an intercalated anion. In the case of hydrotalcite the chemical formula is $M^{2+}:\text{Mg}^{2+}, M^{3+}:\text{Al}^{3+}, A^{n-}:\text{CO}_3^{2-}$. Layered double hydrotalcite (LDH) compounds have a cation adsorption property in addition to the anion exchange property due to the presence of surface hydroxyl groups [22]. In addition to the large surface area and high anion exchange capacity of LDHs, their flexible interlayer region, which is accessible to various anionic species and polar molecular species, is another feature that results in the high efficiency for the removal of contaminants [23]. LDHs uptake anions from solution via three mechanisms: (1) adsorption, (2) intercalation by anion exchange, and (3) intercalation by reconstruction of the calcined LDH structure [8].

As has a high affinity for adsorption on iron oxides, which is evident from the observation that As in natural environments often exists on the surface of iron oxides [24]. The iron-based HTs are promising candidates for As removal and some papers have been dedicated to the study of synthetic Mg–Fe carbonate-based HTs [25]. Presently, limited research exists on the FeHT adsorption of arsenic by FeHT materials such as Mg–Fe– CO_3 -HT [24,26]. Nishida et al. [24] found that the As removal ability of Fe^{3+} -HT increased with increasing amounts of Fe^{3+} ions in Fe^{3+} -HT.

In the present study, Mg–Fe– CO_3 -HT was studied for its ability to remove arsenic. The selection of iron-based hydrotalcite-like compounds was based on the high affinity of iron oxides to arsenic. Hydrotalcite-like compounds containing iron (Fe^{3+} -compounds) were synthesized by the co-precipitation method. The synthesized compounds were first characterized based on their physico-chemical structures. Then the adsorption mechanism of arsenic onto the compounds was examined.

2. Material and methods

2.1. Synthesis and characterization of FeHT

Mg–Fe-based hydrotalcites (FeHT) were synthesized as follows: First, samples were prepared at a $\text{Mg}^{2+}/\text{Fe}^{3+}$ ratio of 2.46. 18.3 g of $\text{MgCl}_2 \times \text{H}_2\text{O}$ and 4.24 g of $\text{FeCl}_3 \times 6\text{H}_2\text{O}$ were dissolved in 200 ml of water while the solution was stirred (Solution 1). Next, 32 g of NaOH and 5.72 g of $\text{Na}_2\text{CO}_3 \times 10\text{H}_2\text{O}$ were dissolved into 4 l of water (Solution 2). Solution 1 was added slowly into solution 2 while vigorously stirring. The co-precipitation reaction was always performed at room temperature and at a solution pH of 11.5 maintained by NaOH and HCl. The mixed suspension was centrifuged at a speed of 1300 rpm for 20 min to recover the white solid precipitate. The liquid was removed and the wet cake was collected into a 1-l glass bottle containing fresh water. Following this the solution was rinsed with water to completely remove the Cl^- ions. The solution was filtrated and the wet cake put in the oven for a further 20 h at 80 °C to dry the FeHT sample.

The solid structure of the adsorbent was analyzed by X-Ray Diffractometer (Rigaku DMax-IIIc). The diffractometer was operated with $\text{Co K}\alpha$ radiation source ($\lambda = 1.54059$). Experiments were run at 35 kV and 15 mA power. XRD scans on FeHT were recorded from 20° to 65° (in 2θ) with a 0.05° step width and with a scanning rate of 1° min^{-1} . XRD pattern of the synthesized FeHT shows sharp peaks at lower 2θ values, which are characteristic of HT-like compounds. The (003) and (006) peaks are attributed to basal reflections, corresponding to stacking of the brucite-like sheets. The remaining peaks are attributed to non-basal reflections. The sharp-

ness of the (003) and (006) peaks demonstrates the high degree of crystallinity and layered structure of the sample. These results agree well with the previous research [24].

The BET surface area was measured by a Nova 4000E instrument (Quantachrome Instrument). The particle sizes were measured by a laser granulometer (Mastersizer S, Malvern Instruments SZ-3600). Scanning electron microscope (SEM) micrographs were recorded with a JEOL/JSM-6335F at 20 kV. The mean particle size and specific surface area of the synthesized FeHT were 233 μm and 1.448 m^2/g , respectively. The SEM images of the sample prepared under high base conditions show strong agglomeration of particles in aggregates with a very large distribution of size [27].

2.2. Adsorption experiments

The arsenate (As(V)) stock solution was prepared by dissolving (dried at 60 °C for 30 min and weighed) sodium arsenate ($\text{Na}_2\text{HAsO}_4 \times 7\text{H}_2\text{O}$, Sigma) in pure water. The working solution for all experiments was freshly prepared from the stock solution. Standard acid (0.01 M HCl) and base solutions (0.125 M NaOH) were used for pH adjustments. The pH of the solution was measured with a pH meter (Thermo Orion 5 Star) using a combined glass electrode (Orion 81–75). The pH meter was calibrated with buffers of pH 4.0, 7.0 and 10.0 before any measurement.

All adsorption experiments were done in a temperature controlled (at 25 °C) shaker (Wiggen Hauser SI-100T) for a predetermined contact time at 200 rpm. The tests of As(V) adsorption with FeHT were performed under different initial As(V) concentrations (100–300–500–1000–2000 $\mu\text{g}/\text{l}$), initial pH levels (3–5–7–9–12) and FeHT dosages (0.1–0.5–1–5–10–15 g/l). Adsorption isotherms were obtained by the batch equilibration technique. For the equilibrium studies, 50 ml solution of As(V) of known concentration (300 $\mu\text{g}/\text{l}$) was used at a fixed adsorbent concentration (10 g/l) in a series of 100 ml flasks. The samples were collected at specific time intervals of 15, 30, 60, 120 and 180 min and then centrifuged at 3000 rpm for 15 min.

The adsorption kinetics and isotherms were investigated in batch experiments. Langmuir and Freundlich isotherms were used to describe the equilibrium data. The linearised form below was used to determine Langmuir isotherm parameters [28]:

$$\frac{1}{q_e} = \frac{1}{bQ C_e} + \frac{1}{Q} \quad (1)$$

where q_e is the amount of adsorbed heavy metal per unit FeHT at equilibrium, Q is the saturated monolayer adsorption capacity, b is the binding energy of the sorption system and C_e is the equilibrium concentration of the solution. The slope and intercept of the linear plot of $1/q_e$ versus $1/C_e$ are the values of b and Q respectively.

The Freundlich isotherm (Eq. (2)) is used for modeling the adsorption on heterogeneous surfaces.

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (2)$$

where K_f is the Freundlich constant and n is the adsorption intensity [29]. The linear plot of $\ln q_e$ versus $\ln C_e$ gives a slope of $1/n$ and intercept of $\ln K_f$.

Adsorption kinetics was used to explain the adsorption mechanism and adsorption characteristics of FeHT. The pseudo-first-order (Eq. (3)) and second-order (Eq. (4)) kinetics equations in linear forms are expressed as [30]:

$$\ln(q_e - q_t) = \ln q_e - k_1 t \quad (3)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

where q_t and q_e are the amounts of FeHT adsorbed at time t and at equilibrium, respectively; k_1 and k_2 are the pseudo-first-order and second-order rate constants for the adsorption process.

2.3. Analytical methods

A flow injection hydride generation attachment (FIAS 100) of a PerkinElmer AAnalyst 400 atomic absorption spectrometer was used for the detection of arsenic in water samples before and after each experiment. The instrument was calibrated by arsenic standard solutions of 10, 50 and 100 $\mu\text{g/l}$. For complete reduction of As(V), 10% (KI + ascorbic acid) and 10% HCl solutions were reacted with the samples. Following this procedure, the sample was reacted with 0.4% NaBH_4 and 10% HCl solutions and purged continuously by argon.

3. Results and discussion

3.1. Adsorption of As(V) by FeHT

The speciation of arsenic in aqueous solutions is dependent on the pH of the solution. For pentavalent arsenic, the corresponding stable species and pH values are: H_3AsO_4 (pH 0–2), H_2AsO_4^- (pH 2–7), HAsO_4^{2-} (pH 7–12) and AsO_4^{3-} (pH 12–14) [11]. The effect of pH on the adsorption of arsenate (As(V)) on FeHT was studied with an initial concentration of 300 $\mu\text{g/l}$ of arsenic in solution at different pH values. Ten grams per liter of FeHT was added to each test solution and left to equilibrate for 180 min. Fig. 1 shows the effects of initial pH values on the efficiency of As(V) adsorption by FeHT with an initial As(V) concentration of 300 $\mu\text{g/l}$. The results indicate that the amount of As(V) adsorbed by FeHT was independent of initial pH. The addition of FeHT to water at different pH values was found to have a buffering effect at $\sim\text{pH}$ 9. This can be explained by the amphoteric nature of FeHT. In a low pH medium, the presence of FeHT resulted in an increase in the pH of the bulk solution. On the other hand, in a high pH medium, acid dissociation of FeHT dominated, causing a decrease in the pH of the bulk solution [9]. Therefore, arsenic removal was carried out only at the equilibrium pH. Other authors have also observed the buffering effect of hydrotalcite-like compounds [31,32]. From these results, it is apparent that the maximum adsorption of arsenic on FeHT occurs at around pH 9.0. Chang et al. [26] used Fe- NO_3 -HT and Al- NO_3 -HT for the removal of As(V) from aqueous solutions (433 $\mu\text{g/l}$ As(V)). They reported an As removal of 98% by Fe- NO_3 -

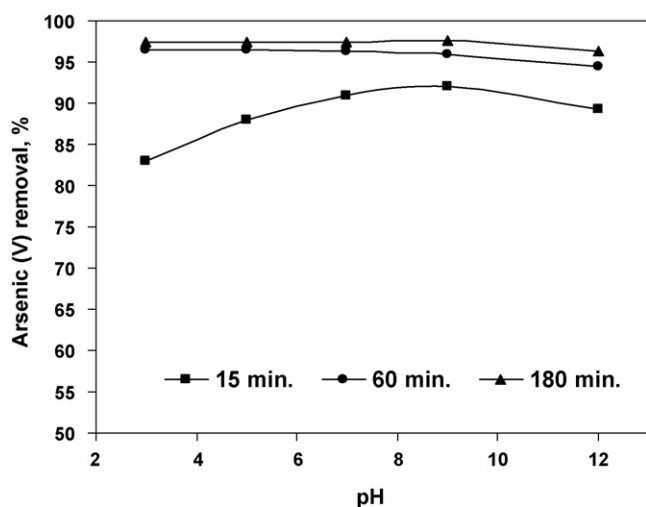


Fig. 1. Effect of pH on As(V) removal by FeHT (T : 25 °C, C_0 : 300 $\mu\text{g/l}$, adsorbent dosage: 10 g/l).

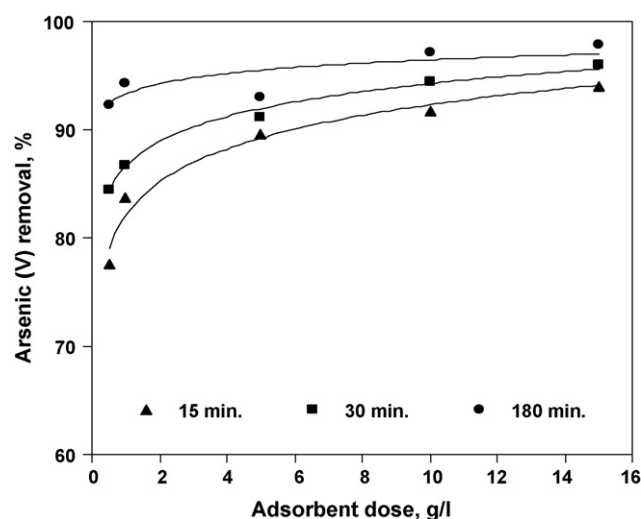


Fig. 2. Effect of adsorbent dosage on As(V) removal by FeHT (T : 25 °C, C_0 : 300 $\mu\text{g/l}$, pH 9).

HT compared with that of 88.2% by Al- NO_3 -HT at pH 8.8. In the current study, 97.2% of As(V) was removed by Fe- CO_3 -HT from the solution initially containing 330 $\mu\text{g/l}$ As(V).

Fig. 2 shows the effect of FeHT dosage on the removal of As(V). The arsenic removal efficiency was increased with the amount of added FeHT. In the subsequent studies, the adsorbent dosage set to be 10 g/l. The effect of different As concentrations was determined after experimental studies were carried out for a range of metal concentrations between 100 and 2000 $\mu\text{g/l}$. The results obtained from the experimental studies are shown in Fig. 3. The adsorption efficiency increased to a certain level, and saturated beyond a certain concentration. Saturation resulted when no more metal ions could be adsorbed on the surface of FeHT where the adsorption occurred. The experimental studies also showed that high efficiency for arsenic adsorption could be obtained over a relatively short period of up to 180 min.

Yang et al. [19] studied the adsorption of As(V) on Al- CO_3 -HT and found that As concentration in solution (20 $\mu\text{g/l}$) was reduced by 71.5% over an equilibrium period of 180 min. Gilman [21] reported the reduction of the concentration of arsenite (432 $\mu\text{g/l}$) by 53.2% over an adsorption period of 60 min when Al- CO_3 -HT

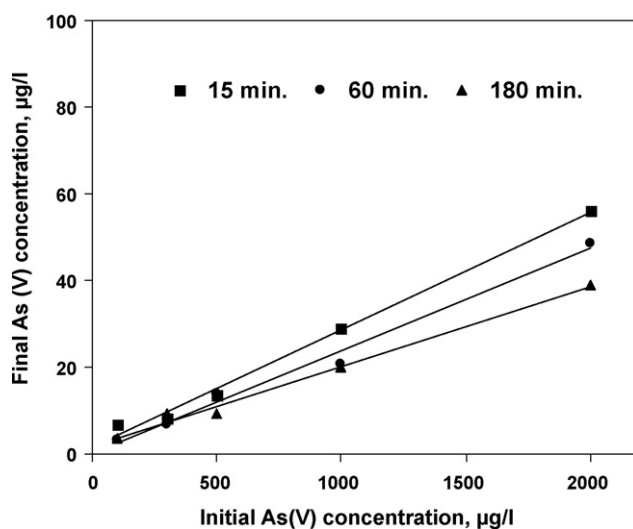


Fig. 3. Final levels of at different initial As(V) concentrations in the range of 100–2000 $\mu\text{g/l}$ (25 °C, pH 9, Adsorbent dosage: 10 g/l).

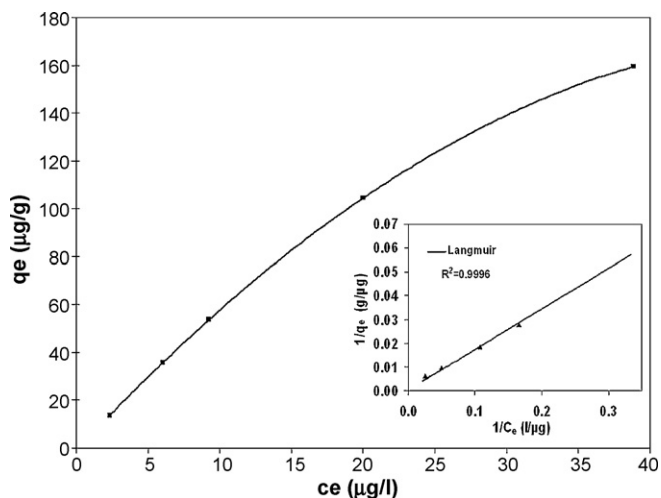


Fig. 4. Equilibrium data and Langmuir isotherm for As(V) adsorption on FeHT at pH 9 and 25 °C.

was used. They noted a slightly lower efficiency for the removal of arsenite by Al–Cl–HT. Nishida et al. [24] also studied the effect of Mg/Fe ratio (2–4) on the adsorptive performance of Fe–CO₃–HT for As. They found that the increase in Mg/Fe ratio adversely influenced the removal of As by Fe–CO₃–HT although it still exhibited a superior performance for As adsorption over Al–CO₃–HT (Mg/Al: 4).

3.2. Adsorption isotherms

An adsorption isotherm describes how adsorbates interact with adsorbents in an adsorption system. The adsorption isotherm for As(V) uptake by FeHT is shown in Fig. 4. It should be noted that when FeHT was used for the removal of As from the solution containing <300 µg/l As(V) it was possible to lower the arsenic concentration below the limit value (i.e. 10 µg/l) recommended by WHO. The Langmuir and Freundlich isotherms were used for analyzing the equilibrium adsorption data of As(V) onto FeHT. The Langmuir isotherm models the single coating layer on the adsorption surface. This model assumes that the adsorption takes place at a specific adsorption surface. The attraction between molecules decreases with increasing distance from the adsorption surface [29]. The Langmuir isotherm fitted to the equilibrium data for the adsorption of As(V) on FeHT are also illustrated in Fig. 4.

The Langmuir parameters, Q and b were found to be 2642.37 and 0.002, respectively with a high correlation coefficient ($R^2 > 0.99$). The sorption isotherm of As(V) ions on to hydrotalcite were determined by Yang et al. [19] and found to be consistent with the Langmuir model. Affinity of As(V) for FeHT can be predicted using the Langmuir parameter b and initial As(V) concentration (C_0) to obtain the separation factor R_L (Eq. (5)) [28]. In the current study, the values of R_L were found to be less than 0.6 with a decreased separation factor at high As(V) concentrations, which indicates a highly favorable adsorption of As(V) on FeHT.

$$R_L = \frac{1}{1 + bC_0} \quad (5)$$

The Freundlich adsorption isotherm is presented in Fig. 5. The Freundlich parameters K_f and n were determined to be 7.00 and 1.134, respectively ($R^2 > 0.99$). The Freundlich isotherm describes the surface heterogeneity and exponential distribution of active sites and their energies [28].

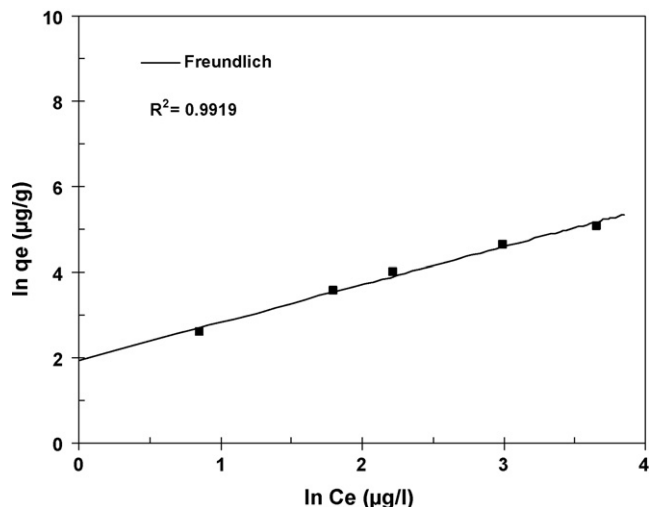


Fig. 5. The plot of $\ln q_e$ versus $\ln C_e$ for the determination of Freundlich isotherm parameters for As(V) adsorption on FeHT.

3.3. Kinetics of adsorption

Adsorption kinetics was used to explain the adsorption mechanism and adsorption characteristics of FeHT. Fig. 6a shows that the initial rate of adsorption of As(V) on FeHT was fairly rapid since it was almost complete over 15 min. The data shown in Fig. 6a

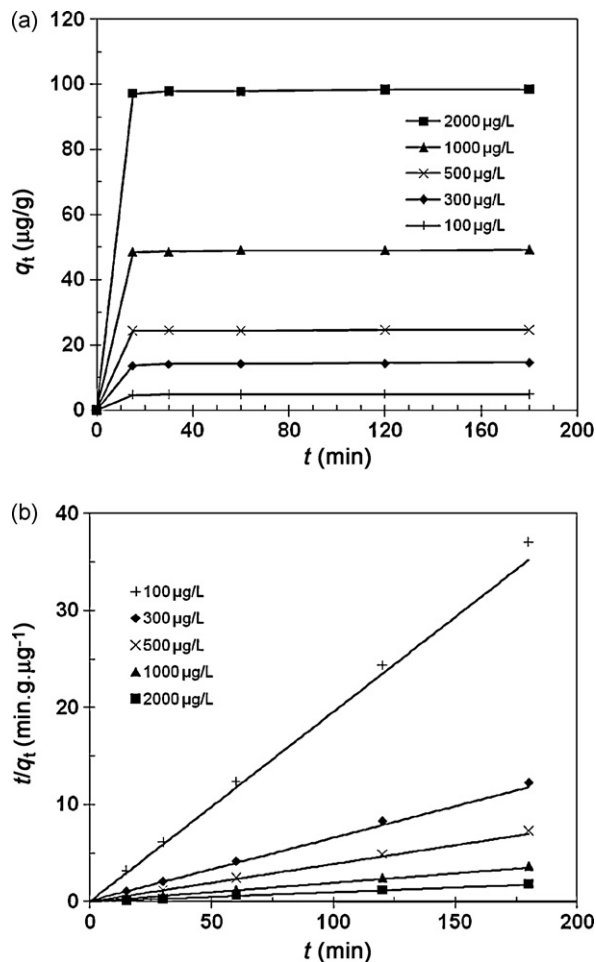


Fig. 6. (a) Time-dependent adsorption As(V) on FeHT at different initial concentrations (T : 25 °C, pH : 9, D : 10 g/l) (b) Linear plot of t/q_t versus t for the pseudo-second-order rate equation for As(V) adsorption onto FeHT.

Table 1

Kinetic parameters of the pseudo-first- and second-order models for the adsorption of As(V) on FeHT at different initial concentrations (T: 25 °C, pH 9, adsorbent dosage: 10 g/l).

Kinetic model	Parameter	Initial As(V) loads ($\mu\text{g l}^{-1}$)				
		100	300	500	1000	2000
Pseudo-first-order	k_1 (min^{-1})	0.004	0.007	0.004	0.003	0.003
	q_e ($\mu\text{g g}^{-1}$)	0.19	1.23	0.70	1.43	2.61
	R^2	0.9658	0.9609	0.9495	0.9649	0.9641
Pseudo-second-order	k_2 ($\text{g } \mu\text{g}^{-1} \text{ min}^{-1}$)	1.11	0.04	0.94	0.06	0.033
	q_e ($\mu\text{g g}^{-1}$)	5.12	15.40	25.87	51.64	103.40
	R^2	0.9998	0.9999	1.00	1.00	1.00

was used for further analysis of the rate of adsorption using the pseudo-first- and second-order kinetic models.

The slope and intercept of the plot of $\log(q_e - q_t)$ versus t were used to determine the first-order rate constant k_1 and equilibrium adsorption density q_e at different initial concentrations of As(V) (Table 1). The correlation coefficients for the pseudo-first-order model were low (i.e. $R^2 < 0.97$). This suggests that the pseudo-first-order equation might not be sufficient to depict the kinetics of As(V) on the FeHT.

The kinetic parameters for the pseudo-second-order model were determined from the linear plot of t/q_t against t (Fig. 6b and Table 1). High correlation coefficients ($R^2 > 0.999$) suggested that the kinetics of adsorption of As(V) on FeHT were best described by the pseudo-second order model. The calculated q_e values also agreed very well with the experimental data. These findings indicate that the adsorption system in our experiments is a second-order system. It is assumed that the adsorption capacity of FeHT was proportional to the number of active sites on its surface. As shown in Fig. 6, the adsorption of As(V) was initially rapid, but, it was then remarkably slow to proceed to the equilibrium. This agreed with the general characteristics of the kinetics of the oxyanion adsorption on LDHs [23]. A few studies have reported on the time-dependent adsorption of As(V) on LDHs [15,16,19]. Although the initial As(V) adsorption process by LDHs was fairly fast, it took 2–3 days to completely reach equilibrium [19]. Goh et al. [23] suggested that the oxyanion adsorption on LDHs such as FeHT occurs through (1) external mass transfer such as boundary layer/film diffusion, (2) interparticle transport within the particle and (3) chemisorption.

4. Conclusion

In this study, Fe^{3+} -substituted hydrotalcites (FeHT) with a chemical formula of $[\text{Mg}_6\text{Fe}_2^{+3}(\text{CO}_3)(\text{OH})_{16} \times 4\text{H}_2\text{O}]$ were synthesized by a co-precipitation method to remove As(V) from water.

FeHT has a high arsenate removal efficiency and it was able to reduce the arsenate concentration (e.g. $\sim 300 \mu\text{g/l}$) down to a final level of $< 10 \mu\text{g/l}$. The results also showed that the amount of adsorption of As(V) by FeHT was only weakly dependent on initial pH. The equilibrium studies indicated that the Langmuir isotherm best described the As(V) adsorption onto FeHT. The adsorption of As(V) was found to occur rapidly during the initial periods; thereafter, it progressed towards equilibrium at a considerably slow rate. The kinetics of adsorption was shown to be suitably described by the pseudo-second-order kinetics model. Finally, the stable arsenic removal efficiency and the capability to reduce As levels below $10 \mu\text{g/l}$ make FeHT a suitable adsorbent for the treatment of As polluted waters.

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References

- [1] WHO, Environmental Health Criteria 224, in: Arsenic and Arsenic Compounds, 2nd edition, World Health Organization, Geneva, 2001.
- [2] P.L. Smedley, D.G. Kinniburgh, A review of the source, behaviour and distribution of arsenic in natural waters, Applied Geochemistry 17 (2002) 517–568.
- [3] J.K. Fawell, E. Ohanian, M. Giddings, P. Toft, Y. Magara, P. Jackson, Arsenic in drinking-water, Background Document for Development of WHO Guidelines for Drinking-Water Quality, 2003.
- [4] H.S. Altundoğan, S. Altundoğan, F. Tümen, M. Bildik, Arsenic removal from aqueous by adsorption on red mud, Waste Management 20 (2000) 761–767.
- [5] A.V. Payan, Arsenic and lead removal from water using tailored zeolites, MSc Thesis, University of Puerto Rico, 1999.
- [6] I.A. Katsouyannis, A.I. Zouboulis, Application of biological processes for the removal of arsenic from groundwater, Water Research 38 (2004) 17–26.
- [7] WHO, Guidelines for Drinking-Water Quality, Volume 1: Recommendations, 2nd editions, World Health Organization, Geneva, 2003.
- [8] D. Mohan, C.U. Pittman, Arsenic removal from water/wastewater using adsorbents—a critical review, Journal of Hazardous Materials 142 (2007) 1–53.
- [9] W. Zhang, P. Singh, E. Paling, S. Delides, Arsenic removal from contaminated water by natural iron ores, Minerals Engineering 17 (2004) 517–524.
- [10] P. Lakshminathiraj, B.R.V. Narasimhan, S. Prabhakar, S.G. Bhaskar Raju, Adsorption of arsenate on synthetic goethite from aqueous solutions, Journal of Hazardous Materials 136 (2006) 281–287.
- [11] F. Zhang, I. Hideaki, Iron oxide-loaded slag for arsenic removal from aqueous system, Chemosphere 60 (2005) 319–325.
- [12] S. Bang, G.P. Korfiatis, X. Meng, Removal of arsenic from water by zero-valent iron, Journal of Hazardous Materials 121 (2005) 61–67.
- [13] F. Kovanda, E. Kovacsova, D. Kolousek, Removal of anions from solution by calcined hydrotalcite and regeneration of used sorbent in repeated calcination-rehydration-anion exchange processes, Collection of Czechoslovak Chemical Communications 64 (1999) 1517–1528.
- [14] G.N. Manju, T.S. Anirudhan, Treatment of arsenic(III) containing wastewater by adsorption on hydrotalcite, Indian Journal of Environmental Health 42 (1) (2000).
- [15] Y.W. You, H.T. Zhao, G.F. Vance, Removal of arsenite from aqueous solutions by anionic clays, Environmental Technology 22 (2001) 1447–1457.
- [16] N.K. Lazaridis, A. Hourzemanoglou, K.A. Matis, Flotation of metal-loaded clay anion exchangers. Part II: the case of arsenates, Chemosphere 47 (2002) 319–324.
- [17] B. Dousovai, V. Machovic, D. Kolousek, F. Kovanda, V. Dornicak, Sorption of As(V) species from aqueous systems, Water, Air, and Soil Pollution 149 (2003) 251–267.
- [18] Y. Kiso, Y.J. Jung, T. Yamata, M. Nagai, K.S. Min, Removal properties of arsenic compounds with synthetic hydrotalcite compounds, Water Supply 5 (2005) 75–81.
- [19] L. Yang, Z. Shahrivari, P.K.T. Liu, M. Sahimi, T.T. Tsotsis, Removal of trace levels of arsenic and selenium from aqueous solutions by calcined and uncalcined layered double hydroxides (LDH), Industrial and Engineering Chemistry Research 44 (2005) 6804–6815.
- [20] L. Yang, M. Dadwhal, Z. Shahrivari, M. Ostwal, P.K.T. Liu, M. Sahimi, T.T. Tsotsis, Adsorption of arsenic on layered double hydroxides: effect of the particle size, Industrial and Engineering Chemistry Research 45 (2006) 4742–4751.
- [21] G.P. Gillman, A simple technology for arsenic removal from drinking water using hydrotalcite, Science of the Total Environment 366 (2006) 926–931.
- [22] Y. Seida, Y. Nakano, Removal of humic substances by layered double hydroxide containing iron, Water Research 34 (2000) 1487–1494.
- [23] K.H. Goh, T.T. Lim, Z. Dong, Application of layered double hydroxides for removal of oxyanions: a review, Water Research 42 (2008) 1343–1368.
- [24] S. Nishida, S. Takesoe, Y. Yamasaki, A. Nakahira, Attempt of arsenic removal in wasted water by inorganic materials, in: 14th International Conference on the Properties of Water and Steam, Japan, 2004.
- [25] O.P. Ferreira, O.L. Alves, D.X. Gouveia, A.G. Filho, J.A.C. Paiva, J.M. Filho, Thermal decomposition and structural reconstruction effect on Mg–Fe-based hydrotalcite compounds, Journal of Solid State Chemistry 177 (2004) 3058–3069.
- [26] W. Chang, H. Jones, J.E. Valsami, H. Garelick, Evaluation of the arsenic(V) removal from water by hydrotalcite-like compounds, in: 7th UK Chinese Association of Resources and Environment (CARE) Annual General Meeting, Greenwich, 2007.

- [27] M.A. Pagano, C. Forano, J.P. Besse, Synthesis of Al-rich hydrotalcite-like compounds by using the urea hydrolysis reaction—control of size and morphology, *Journal of Materials Chemistry* 13 (2003) 1988–1993.
- [28] S. Kundu, A.K. Gupta, Adsorption characteristics of As(III) from aqueous solution on iron oxide coated cement (IOCC), *Journal of Hazardous Materials* 142 (2007) 97–104.
- [29] S. Veli, B. Akyüz, Adsorption of copper and zinc from aqueous solutions by using natural clay, *Journal of Hazardous Materials* 149 (2007) 226–233.
- [30] E. Bulut, M. Özacar, İ.A. Şengil, Equilibrium and kinetic data and process design for adsorption of congo red onto bentonite, *Journal of Hazardous Materials* 154 (2008) 613–622.
- [31] Y. You, G.F. Vance, H. Zhao, Selenium adsorption on Mg–Al and Zn–Al layered double hydroxides, *Applied Clay Science* 20 (2001) 13–25.
- [32] O.P. Ferreira, S.G. Moraes, N. Duran, L. Cornejo, O.L. Alves, Evaluation of boron removal from water by hydrotalcite-like compounds, *Chemosphere* 62 (2006) 80–88.