

Fe-based layered double hydroxides for removing arsenic from water: sorption–desorption–regeneration

Tuğba Türk, Taha Boyraz and İbrahim Alp

ABSTRACT

Since the presence of arsenic in the waters of the world causes serious health effects on people, it is very important to remove it. Layered double hydroxides have a high surface area and high anion exchange capacity, and because of this feature, it is a potential adsorbent to remove arsenic. For regeneration and reuse of adsorbents, researchers in some limited studies have used agents such as acids and alkalis. Media replacement accounts for approximately 80% of the total operational and maintenance costs. In this paper, an adsorption/desorption/regeneration study was carried out with MgFeHT to determine the desorption properties of the adsorbent and to examine its reusability. The best alkaline desorption solution was determined from two different alkaline solutions: NaOH and KH_2PO_4 . As(V) adsorption capacity of the MgFeHT at different pH (3–12) using the arsenic aqueous solution (with 2,000 $\mu\text{g As(V)/L}$) was evaluated. For the adsorption process, the experimental data are fitted well with the pseudo-second-order kinetic model and the Langmuir model. Moreover, the concentration of 2,000 $\mu\text{g/L}$ arsenic was reduced to below the legal limit determined by the WHO ($<10 \mu\text{g/L}$). The regeneration studies were conducted on the adsorptive media used in the arsenic removal system. The regeneration efficiency of As(V) was maintained 98.5% for four regeneration cycles using 0.5 M NaOH. MgFeHT was successfully regenerated with an aqueous solution of NaOH and was reused with a small loss of sorption efficiency.

Key words | arsenate, desorption, layered double hydroxide (LDH), regeneration, sorption

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HIGHLIGHTS

- Arsenic toxicity is an important health problem.
- Environmental pollution is the biggest problem of industry.
- So, economical, easily available, and reusable adsorbent is necessary.
- Although several studies have been studied on the desorption of layered double hydroxides with arsenic, almost all studies have focused on arsenic removal capacities.
- In this study, it was observed that MgFeHT – as an adsorbent – is reusable and economical.

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INTRODUCTION

Arsenic is among the group 1 cancer-causing compounds by the International Agency for Research on Cancer. Arsenic is naturally present at high levels in the groundwater of countries such as India, Argentina, China, Mexico, Chile, and the USA. Groundwater is the cheaper and abundant source of drinking water in most living areas in the world. Drinking water poses the greatest threat to public health from arsenic (Türk 2017). The source of arsenic for humans is not only drinking water. Especially in areas with high arsenic concentrations, irrigation waters and contaminated soil represent the primary cause of arsenic toxicity (Smedley & Kinniburgh 2002). Prolonged exposure to drinking water containing arsenic causes diabetes, high blood pressure, discoloration of the skin, skin thickening (hyperkeratosis), neurological disorders, muscle weakness, anorexia, and nausea (Zhang *et al.* 2017).

To prevent drinking water pollution with arsenic in many countries around the world and the health effects caused by it, the maximum pollutant level adopted by the World Health Organization (WHO 1993) for arsenic in drinking water has been reduced to 0.01 mg/L in 1993 (Lee *et al.* 2018). The removal of arsenic from waters was achieved with different techniques. The arsenic removal method from underground and surface waters is selected according to the arsenic concentration in the water, its place and purpose, cost, and toxic waste generation. Conventional methods, such as precipitation, coagulation, adsorption and ion exchangers, membrane filtration, and polymer ligand exchangers, are used for arsenic removal (Babaeiveli & Khodadoust 2016; Türk 2016). Among these technologies, adsorption is considered to be one of the low-cost and easily feasible techniques (Yadav *et al.* 2019). Many kinds of synthetic and natural adsorbents have been developed for the removal of arsenic from drinking water. Layered double hydroxides (LDHs) have high arsenic exchange capacities, which are common features with other clays. The main reason for ion exchange in terms of thermodynamics is the electrostatic interaction of positively charged brucite layers and negatively charged anions and a little hydration-free energy (Israëli *et al.* 2000). LDHs are also called anionic clays. Naturally, occurring hydrotalcite is one of the most

representative minerals of the group. The structure of an LDH is basically formed from the main structure brucite $\text{Mg}(\text{OH})_2$ with the isomorphous substitution of divalent cations. To ensure the electroneutrality of the positively charged layers, CO_3^{2-} ions were arrested between the layers. The structure of hydrotalcite is shown as $\text{Mg}_{0.75}\text{Al}_{0.25}(\text{OH})_{2.5}\cdot\text{H}_2\text{O}$. Different anions can be placed in the area between the layers of the LDHs. The prominent features of LDHs are that they are abundant, nature-friendly, and cheap (Chetia *et al.* 2012; Nhat Ha *et al.* 2016). The synthesis of LDHs is relatively easy and inexpensive on laboratory and industrial scales. There are a number of methods used in the synthesis of LDHs, and these include precipitation, restructuring, hydrothermal, and urea hydrolysis methods (Goh *et al.* 2008). Within the scope of the study, modified Fe hydrotalcite (MgFeHT), known as iron-based layered double hydroxide, was used in adsorption/desorption process.

In practice, to remove arsenic in remediation systems, it is desirable to renew and reuse layered double hydroxides. In a successful desorption/regeneration process, the first properties of the sorbent should be recovered. Parameters that have an effect on the desorption efficiency (DE) of arsenic include the surface properties of the adsorbent, the concentration of the ion and desorption solution, the pH, and the duration of the arsenate contact with the desorption solution (Pigna *et al.* 2006).

Although several studies have been studied on the desorption of layered double hydroxides with arsenic, almost all studies on these adsorbents have focused on arsenic removal capacities (Türk & Alp 2010, 2012). Oxyanion-loaded LDHs can be desorbed using various alkaline salt solutions. Caporale *et al.* (2013) performed desorption with LDHs using PO_4 in arsenite desorption. After 48 h of reaction with PO_4 , the Mg-Fe LDH released 25.9% ($\text{PO}_4/\text{As}(\text{III})$ molar ratio). In one study, arsenate desorption was performed with NaOH from akaganéite-type nanocrystalline, and DE was 75% (Deliyanni *et al.* 2003). The desorption of arsenate from maghemite has not been achieved (Yean *et al.* 2005).

Adsorption is often used to remove arsenic in small drinking-water systems. Adsorbent replacement in adsorption technology accounts for about 80% of the total operating

cost. Regeneration is a potential method to reduce the cost of the adsorptive media if the adsorbent can be regenerated and reused without little or no loss of arsenic removal capacity (Sorg et al. 2018). In the present study, we evaluated the capacity of MgFeHT to remove As(V) from polluted aqueous solutions. Specifically, we examined the effect of the desorption by NaOH, KHPO₄, and regeneration of MgFeHT. Thus, the MgFeHT product can be renewed, and an option for cost reduction can be offered.

METHODS

Synthesis of MgFeHT

In this study, the five different synthesis methods for the production of Fe-Hydrotalcite (MgFeHT) were studied. The syntheses were carried out in five different ways by co-precipitation (Table 1). Since adsorption is a surface treatment, adsorption power is an important function of surface properties. Among the surface properties of the adsorbent, the most important parameter affecting the adsorption process is the surface area value (Beyhan 2003). In this study, it is aimed to have a maximum surface area of the adsorbent to be produced. The selection of the material to be used as an adsorbent was made as a result of the characterization studies.

Characterization of MgFeHT

Each synthesis product was characterized by XRD, BET, and grain size analysis. XRD patterns (XRD) were measured by the Rigaku Smart model diffractometer. XRD was recorded between 4° and 65°. The surface area was measured by an ASiQwin™ instrument. In the analysis process, the degassing temperature was determined as 105 °C, and the degassing time was 8 h. To determine the particle size distribution of the materials, grain size distribution analysis was performed in the Master Sizer device.

Batch experiments

The experiments were carried out in a Wise Shake SHO-2D brand orbital mixer, 100 mL flask, at 25 °C and 200 rpm for

a predetermined contact time with MgFeHT. The solution was centrifuged at 4,000 rpm. Arsenic analyses were performed at 193 nm wavelength with the Perkin Elmer-AAS 100 model Hydride Atomic Absorption Spectrometer (HG-AAS). The effect of adsorbent dose was examined with (0.05–0.1–0.5–1–2–3–4–5) g/L MgFeHT (2,000 µg/L As, pH 9, 120 min). pH was studied at (3–5–7–9–12) (2,000 µg/L As and 2 g/L MgFeHT doses). For adsorption isotherm studies (500–5,000 µg/L As), the solution was used at a 2 g/L MgFeHT.

Arsenic desorption and regeneration of MgFeHT

Desorption experiments were examined under three parameters: (i) selection of the best desorption reagent, (ii) determination of the concentration of desorption reagent, and (iii) examination of desorption kinetics. First, the adsorption test for As(V) was performed under optimum adsorption conditions. The 50 mL 2,000 µg/L arsenic solution was shaken with 1 g/L MgFeHT at 200 rpm for 180 min. At the end of the shaking period, the solution was centrifuged for 5 min at 4,000 rpm, and the liquid was analyzed. The solid portion was dried at 60 °C for 2 h. For desorption, 50 mL (0.5–2–4 M) NaOH and (0.1–0.2–0.3 M) KH₂PO₄ solutions were used to dissolve As-MgFeHT. In desorption tests, different As-MgFeHT dosages (1–2–3 g/L) were tested at different times (15–60–120–180 min). Sequential sorption, desorption, and regeneration experiments were performed to evaluate the renewability of MgFeHT (Figure 1). Samples were filtered using a 0.45 mm filter. Then As(V) concentrations were measured with HG-AAS. Adsorption rate (R_{ads}) and desorption rate (R_{des}) are estimated from the following equations:

$$R_{ads}(\%) = 100 \times C_0 - C_i / C_0$$

$$R_{des}(\%) = 100 \times Q_{des} / Q_{ads}$$

where C_0 is the initial As (µg/L) concentration; C_i is the residual concentration of As in the adsorption process (µg/L); Q_{ads} is the amount of As uptake in the adsorption process (µg-As/g MgFeHT); and Q_{des} is the amount of As desorbed in this desorption process (µg-As/g MgFeHT).

Table 1 | Five different synthesis methods for production of Fe-Hydrotalcite (MgFeHT)

Sample	Synthesis method	Fe/Mg molar ratio	pH	Procedure	Stirring condition	Aging time temperature	Washing	Drying time temperature
MgFeHT-1	Co-precipitation	3	10	Solution 1 ^a was dropped by pipette into solution 2 ^b	1,000 rpm – 120 min	24 hours-80 °C	Using warm distilled water until pH was 9.25	2,480 h – 80 °C
MgFeHT-2				Solution 2 ^b was heated to 80 °C then solution 1 ^a directly transferred to solution 2 ^b	Stepwise stirring at 80 °C 750 rpm (first 30 min) and 1,000 rpm (last 90 min)		Using warm distilled water until pH was 10	
MgFeHT-3				Solution 3 was stirred in a magnetic stirrer at 750 rpm then solution 1 was transferred to solution 3 ^c within 2 h; solution 4 ^d was dropped to maintain pH 10	1,000 rpm – 60 min		Using warm distilled water until pH was 9.52	
MgFeHT-4				Solution 1 ^a directly transferred to solution 2 ^b	Stepwise stirring, 1,000 rpm (first 45 min) and 750 rpm (last 75 min)		Using warm distilled water until pH was 9.72	
MgFeHT-5				Solution 1 ^a and solution 2 ^b in the two different burettes were slowly transferred dropwise to the empty beaker in the magnetic stirrer	1,000 rpm – 120 min		Using warm distilled water until pH was 9.29	

^aMgCl₂ and FeCl₃ solution.^bNaOH and Na₂CO₃ solution.^cNa₂CO₃ solution.^dNaOH solution.

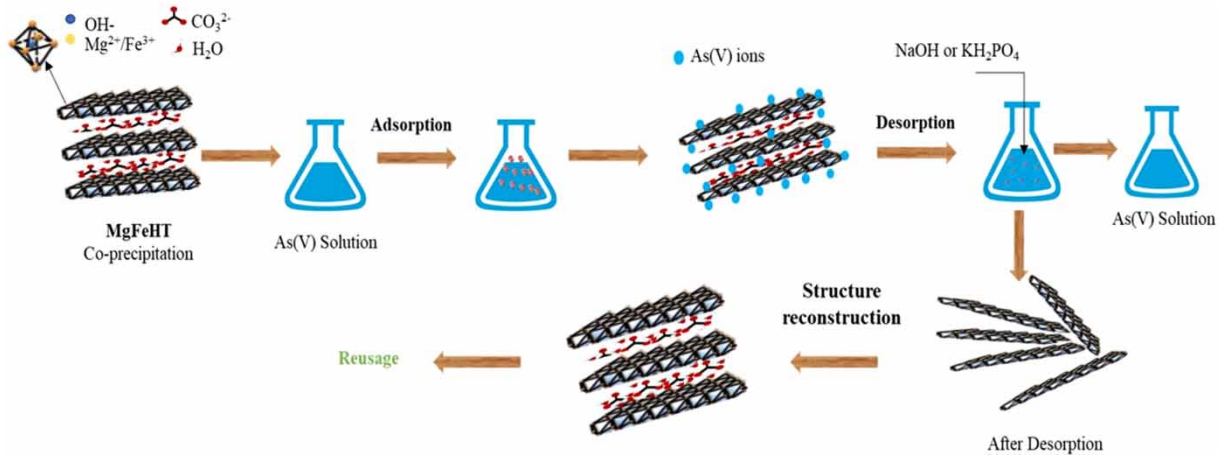


Figure 1 | Schematic representation of the As(V) sorption-desorption-regeneration.

RESULTS AND DISCUSSION

Characterization of MgFeHT

XRD was used to determine the mineralogical composition of the adsorbents. The peaks (003) and (006) belong to the basal reflections in relation to the cluster of the brucite layer. The XRD peaks of the syntheses performed in this study are shown in Figure 2. Other peaks belong to non-basal reflections. The sharpness of the peaks (003) and (006) indicates that the sample has a high degree of crystallinity and a stratified structure. Nishida *et al.* (2004), in their

study, carried out XRD studies for MgFeHT, which they synthesized by the precipitation method. The peaks they obtain are compatible with the peaks obtained in this study.

Grain size is one of the parameters affecting adsorption. The adsorption rate increases, as the grain sizes decrease. However, it has no effect in terms of adsorption capacity (Yang *et al.* 2000). The values obtained as a result of the analysis are shown in Table 2.

The maximum surface area value was evaluated by testing with specific surface area measurements (Table 3). There is an important relationship between the adsorption process and the surface properties of the adsorbent. Among the

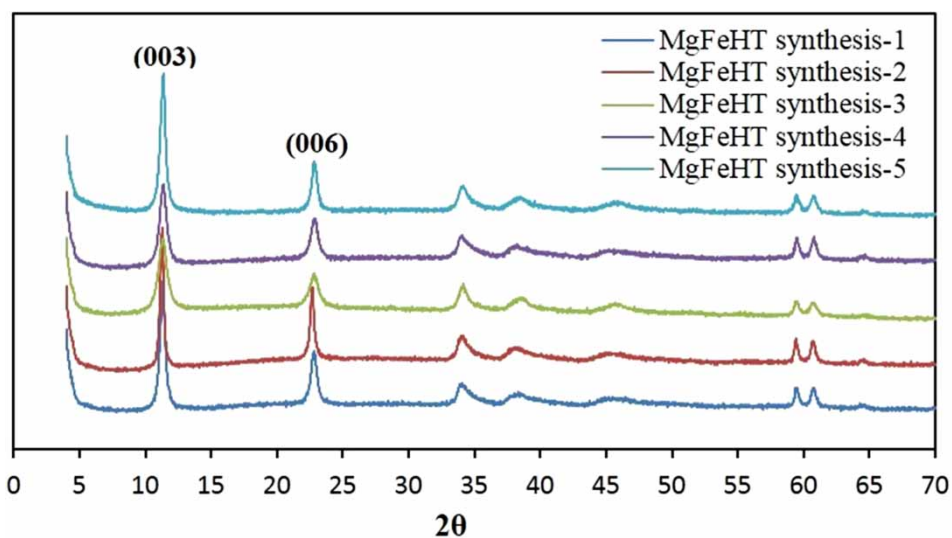


Figure 2 | X-ray diffractogram of MgFeHT synthesis.

Table 2 | Particle size analysis

	Grain size distribution (%), μm		
	D-10	D-50	D-90
MgFeHT-1	3.937	46.968	273.001
MgFeHT-2	4.649	65.710	284.085
MgFeHT-3	3.390	25.305	196.635
MgFeHT-4	3.812	43.665	268.738
MgFeHT-5	7.092	76.439	301.554

Table 3 | BET surface area analysis

Sample	Surface area (m^2/g)
MgFeHT-1	72.83
MgFeHT-2	66.10
MgFeHT-3	97.21
MgFeHT-4	81.10
MgFeHT-5	61.75

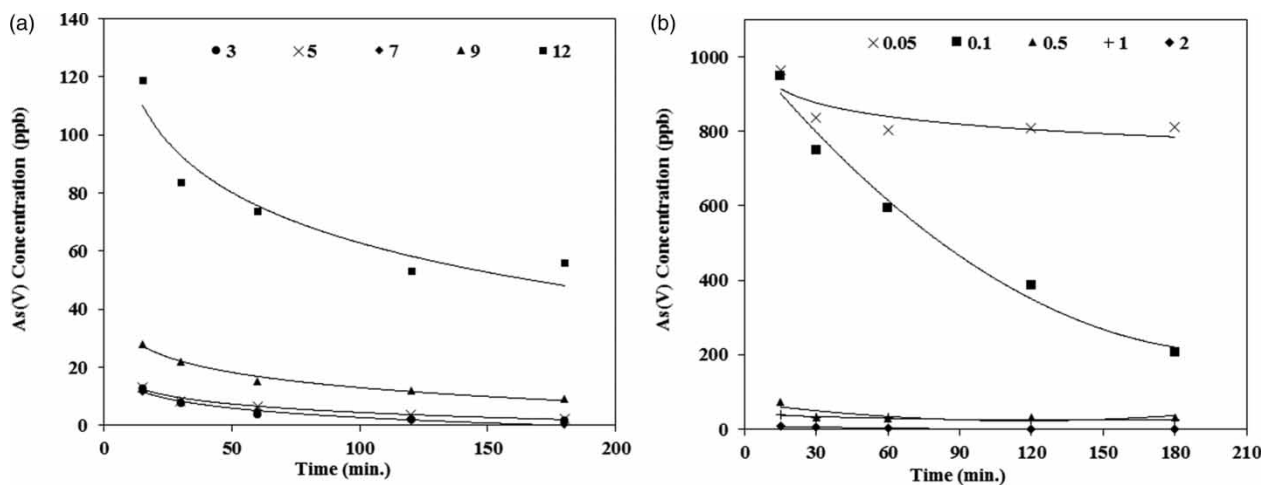
adsorbent surface properties, the most important parameter affecting adsorption is the surface area (Orozco *et al.* 2014). When the surface areas of the synthesis products are examined, it is seen that the production method has an important effect. MgFeHT-3 is the material with the highest specific surface area with a surface area of $97.210 \text{ m}^2/\text{g}$. In addition, when we look at the particle size analysis values of MgFeHT-3, the particle size is lower than the others. The

lower particle size will enable faster adsorption (Table 2). Therefore, it is considered to use MgFeHT-3 in future adsorption/desorption/regeneration process.

Adsorption study of As(V)

pH (3–5–7–9–12) studies with MgFeHT-3 are shown in Figure 3(a). When the results were examined, it was shown that arsenic intake was not affected by the initial pH. Final concentration of As(V) in solution increased from 2.02 to $9.10 \mu\text{g}/\text{L}$ when its pH rises from 3 to 9, possibly due to changes in arsenic types and changes in LDH surface charge (Figure 3(a)). However, when the initial pH was 12, the arsenic concentration in the solution significantly increased to $53.2 \mu\text{g}/\text{L}$. The reason that there is no change between pH 3 and 9 is due to the strong buffer effect seen in layered double hydroxides. This situation has been also observed by many researchers. The value of pH 9 was chosen optimally, and all experiments were carried out at this pH.

In dosage studies with MgFeHT-3, the arsenic concentration in the solution decreased with increasing dosage. However, there was no change after a certain dosage value. Yang *et al.* (2005) determined that as the adsorbent dose increased, the loading capacity increased and there was no change after a certain point. In dosage studies conducted with MgFeHT-3 samples (Figure 3(b)), it was observed that the removal of arsenic increased as the dose

**Figure 3** | Plots of As(V) concentration versus (a) pH and (b) adsorbent dose.

increased. For MgFeHT-3, the optimum adsorbent dosage for As(V) removal was chosen as 2 g/L.

Adsorption isotherms were performed to estimate the adsorption mechanism and to determine the optimum conditions for the maximum removal of As(V) with MgFeHT-3. The Langmuir (Equation (1)) and Freundlich (Equation (2)) isotherm models were used to analyzing adsorption data:

$$\text{Langmuir isotherm equation : } q_e = \frac{Q_m K_L C_e}{1 + K_L C_e} \quad (1)$$

$$\text{Freundlich isotherm equation: } \log q_e = \log K_F + \frac{1}{n} \log C_e \quad (2)$$

C_e is the equilibrium concentration of adsorbate ($\mu\text{g/L}$); Q_m is the maximum capacity ($\mu\text{g/g}$), q_e is the amount of adsorbate at equilibrium ($\mu\text{g/g}$). K_L ($\text{L}/\mu\text{g}$) and K_F [$(\mu\text{g/g})(\mu\text{g/L})^{-1/n}$] are constants, and n is the sorption intensity (Türk et al. 2010). Figure 4(a) and Table 4 show isotherm models. The calculated capacity of MgFeHT-3 was 3,565 $\mu\text{g/g}$ (Table 4). The calculated capacity of MgFeHT-3 was similar to the balance capacity in the kinetic test. MgFeHT-3 showed a good correlation with both Langmuir and Freundlich isotherms. Kinetic studies to remove As(V) with MgFeHT-3 were examined with a solution of 2,000 $\mu\text{g/L}$ As(V) at pH 9 (Figure 4(b)). In kinetic tests, pseudo-first degree (Equation (3)) and pseudo-second degree (Equation (4)) were studied

Table 4 | Langmuir and Freundlich isotherm parameters

Parameter	Value	R^2
Freundlich isotherm		
K_F [$(\mu\text{g/g})(\mu\text{g/L})^{-1/n}$]	217.6	0.98
n	1.45	
Langmuir isotherm		
Q_m ($\mu\text{g/g}$)	3,565.1	0.99
K_L ($\text{L}/\mu\text{g}$)	0.04	

(Ren et al. 2014):

$$\text{Pseudo-first order kinetic: } \ln(q_e - q_t) = \ln q_e - k_1 t \quad (3)$$

$$\text{Pseudo-second order kinetic: } \frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (4)$$

where q_e and q_t are the adsorption capacity at equilibrium and time t ($\mu\text{g/g}$). k_1 (min^{-1}) and k_2 ($\text{g } \mu\text{g}^{-1} \text{min}^{-1}$) are the rate constants. Figure 4(b) shows that the initial rate of the adsorption is very fast. Nonlinear fitted data and rate constants are summarized in Table 5. Here, it is clear that the fitted data follow the pseudo-second-order kinetic model. This model is used in removing pollutants from solution (Beyhan 2003).

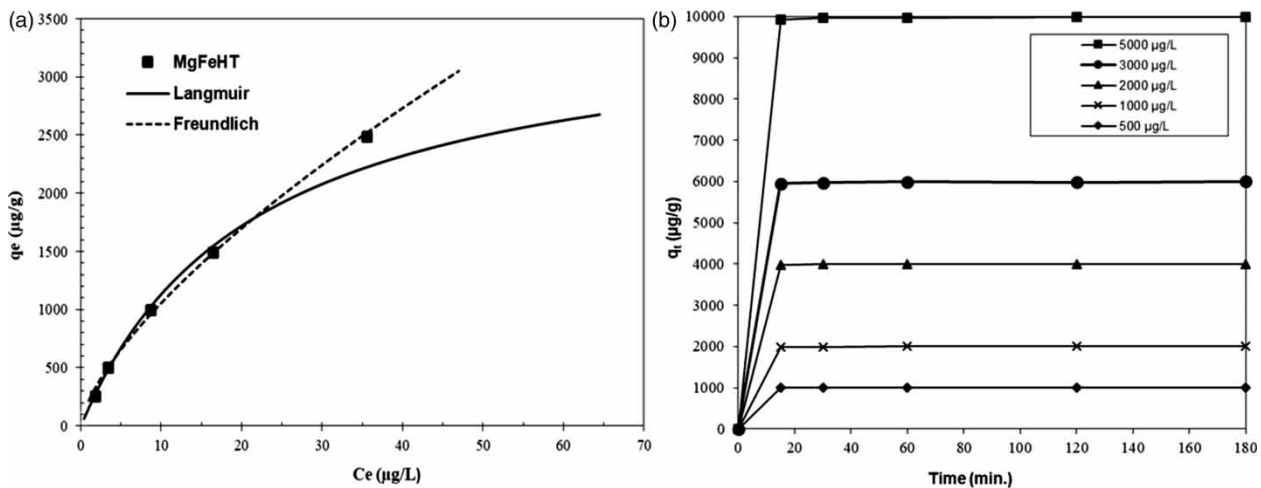


Figure 4 | (a) Isotherm study on MgFeHT-3 (T: 25° C, pH: 9, dosage: 2 g/L, time: 180 min) and (b) kinetic study on MgFeHT-3 (T: 25° C, pH: 9, dosage: 2 g/L).

Table 5 | Kinetic parameters for As(V) adsorption on the MgFeHT-3

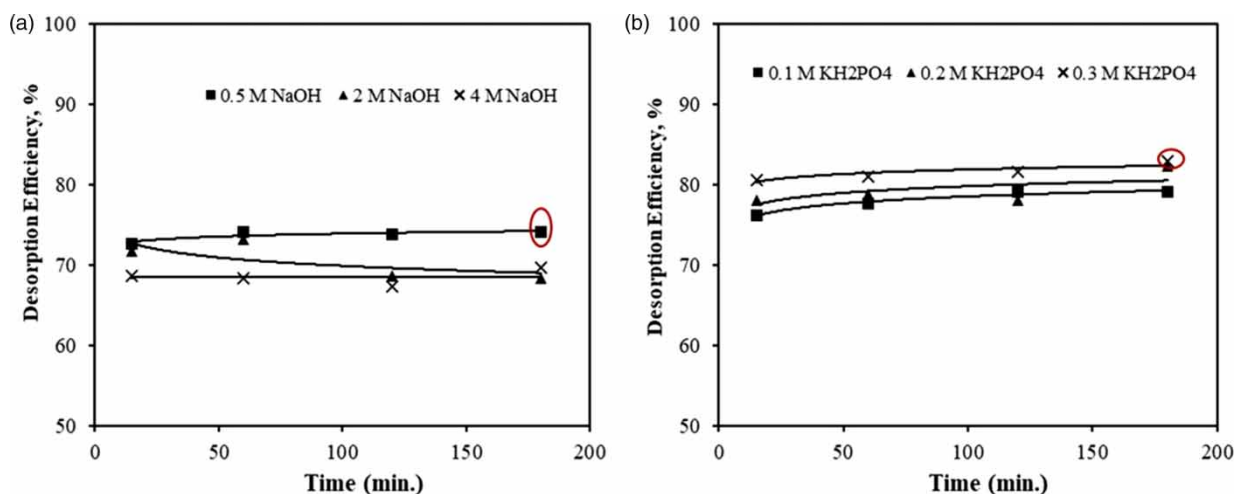
Kinetic model	Variables	First As(V) concentration ($\mu\text{g/L}$)				
		500	1,000	2,000	3,000	5,000
Pseudo-first-order rate equation	k_1	0.028	0.029	0.006	0.006	0.006
	q_e	1.380	11.17	24.47	51.08	133.5
	R^2	0.962	0.959	0.962	0.915	0.930
Pseudo-second-order rate equation	k_2	0.47	0.005	0.003	0.001	0.001
	q_e	999.8	2,000	40,001	5,999	9,984
	R^2	1.000	1.000	1.000	1.000	1.000

Desorption and regeneration

Adsorption/desorption/regeneration study was carried out with MgFeHT-3 as described in the experimental section to determine the desorption properties of the adsorbent and to examine its reusability. NaOH and KH_2PO_4 were tested as desorbents. Some researchers have previously conducted studies on the effect of NaOH on the desorption of anionic pollutants through proton removal, electrostatic repulsion, and ion exchange (Lee et al. 2018). KH_2PO_4 was used in previous studies to remove arsenic from soil aggregates (Min et al. 2008; Zhao et al. 2016). To select the appropriate desorbent solution, different NaOH and KH_2PO_4 concentrations were evaluated depending on the desorption rate of arsenic from As-MgFeHT-3. As-MgFeHT-3 was shaken at different NaOH (0.5–2–4 M) and KH_2PO_4 (0.1–0.2–0.3 M) concentrations (Figure 5). There was little increase in DE with

increasing NaOH concentration. While 69.7% of the DE was obtained with 0.5 M NaOH, 74.1% of the DE was obtained with 4 M NaOH (Figure 5). Increasing DE with increasing NaOH concentration is due to the electrostatic attraction force and the deprotonation surface of LDH between negative charged layered double hydroxides and anions (Zhang et al. 2010). In total, 82.9% of the DE was obtained with 0.3 M KH_2PO_4 (Figure 5). DE increased when the KH_2PO_4 concentration increased. The maximum desorption of As(V) was achieved 74.1 and 82.9% with 0.5 M NaOH and 0.3 M KH_2PO_4 , respectively. However, desorption with KH_2PO_4 is not effective. The reason for this; KH_2PO_4 is due to its tendency to resolve MgFeHT-3. This has been observed by other researchers (Zhao et al. 2016). In their 2016 study, they found that KH_2PO_4 dissolved metal hydroxy oxides such as Fe and Al.

In Figure 6(a), the effect of adsorbent dosages (1–3 g/L) on DE is examined. When 1 and 3 g/L of MgFeHT-3 were used, 74.1 and 52.9% of the DEs were obtained, respectively. It is seen that DE decreases as adsorbent dosage increases (Figure 6). The solid parts obtained after desorption tests were used in regeneration experiments after washing with ultrapure water and drying at 60 °C for 2 h. Regeneration tests were carried out in four cycles. As seen in Figure 7, the regeneration efficiency (RE) of NaOH has decreased slightly from the second regeneration cycle (99.3%) to the fourth regeneration cycle (97.4%). However, the RE of KH_2PO_4 has decreased continuously since the second

**Figure 5** | DE of As(V) by different NaOH and KH_2PO_4 concentrations.

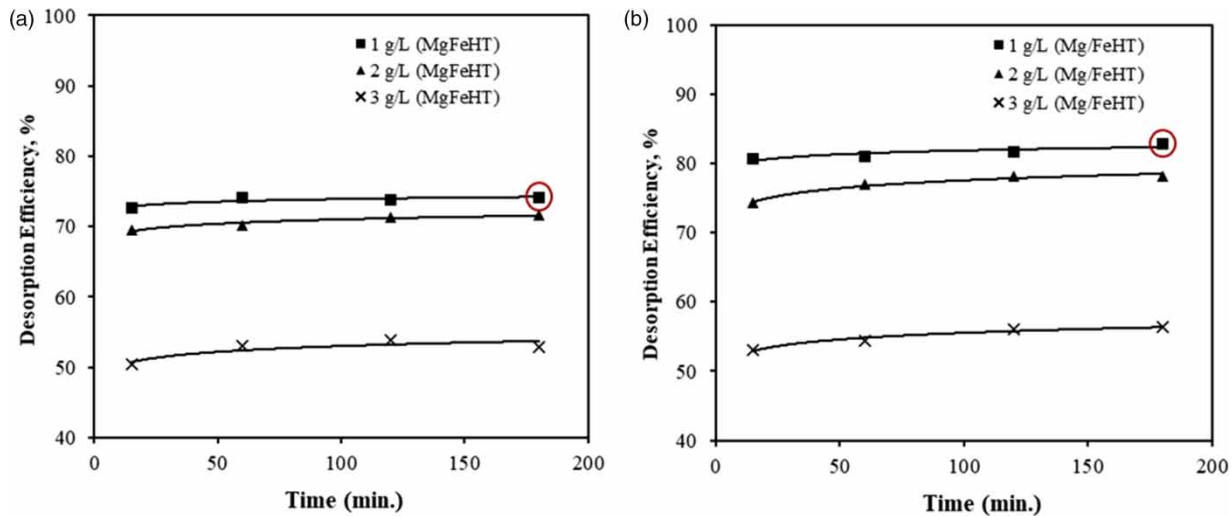


Figure 6 | DE of As(V) by different MgFeHT-3 dosages for using (a) 0.5 M NaOH and (b) 0.3 M KH₂PO₄.

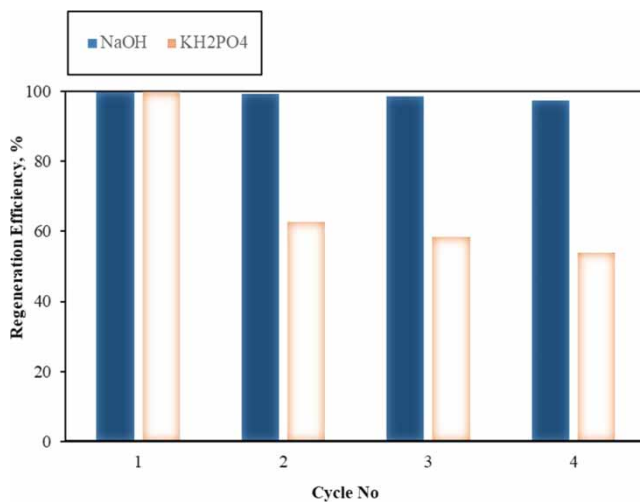


Figure 7 | As(V) regeneration by 0.5 M NaOH and 0.3 M KH₂PO₄ (dosage: 1 g/L, reaction time: 180 min).

cycle. It decreased by 54% at the end of the fourth cycle. The reduction observed after the second regeneration stage may be attributed to the partial dissolution of the LDH sorbent. Therefore, it can be said that the regeneration and reuse of MgFeHT-3 with KH₂PO₄ is negative.

CONCLUSION

In this article, a high surface area of (62–97 m²/g) MgFeHT was produced by the co-precipitation method. In addition, it

has been observed that applying the same synthesis method in different ways affects the surface area of MgFeHT. In adsorption studies with MgFeHT-3, arsenic ion was removed from waters with an initial arsenic concentration of 2,000 µg/L in a 99.5% yield. The arsenic concentration has been reduced below the maximum allowed arsenic concentration in drinking water by the World Health Organization. The desorption of arsenic ions adsorbed by MgFeHT-3 was studied by batch experiments. DE was found to increase with increasing concentration of desorption reagents (NaOH and KH₂PO₄). DE was 74.1 and 82.9%, respectively, when using NaOH and KH₂PO₄. As seen from the results, it was determined that KH₂PO₄ was more effective in removing arsenic from the As-MgFeHT-3 structure. Regeneration studies were conducted on the adsorptive media used in the arsenic removal system. Adsorptive media replacement, which accounts for about 80% of the total operating cost, can be important for small systems. The aim of this research was to determine the renewability of Fe layered double hydroxide products instead of discharge and thus to reduce the operating cost. In regeneration tests conducted to investigate the reusability of the adsorbent, it was found that repeated use of the adsorbent is possible if NaOH is used as the desorption reagent. It was observed that KH₂PO₄ disrupted the FeHT structure and reduced its adsorbent properties and negatively affected arsenic removal performance in repeated use. As a result,

arsenic removal can be achieved at a lower cost with MgFeHT-3, which has a high arsenic removal potential by regenerating an iron-based medium and re-uses the medium.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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