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Adsorptive removal of arsenate using inorganic magnetite particles

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ABSTRACT

Arsenic (As)-contaminated drinking water is a serious health problem in many parts of the world. Although magnetite particles have been shown to be effective at removal of arsenic from water, there has been no research to assess the feasibility of using mill scale-derived magnetite for water treatment. This project was conducted to evaluate magnetite particles as a cost-effective and eco-friendly As adsorbent. Mill scale iron waste has been used to prepare magnetite. The synthesized magnetite was fully characterized and used for the removal of arsenate anion from water. Magnetite showed adsorption capacity of 8.612 mg/g for arsenate anion. Desorption studies were done on the adsorbent to check the recyclability. Magnetite was regenerated with no change in arsenic removal efficiency.

Keywords: Adsorption; Arsenic(V); Mill scale; Magnetite; Batch and column tests

1. Introduction

Arsenic (As) contamination in water is considered as one of the most prominent environmental causes of cancer [1] and neurological, dermatological, gastrointestinal, and cardiorenal diseases [2] in the world. This is due to the fact that millions of people around the world favored naturally occurring As-contaminated groundwater over surface water as their source of drinking water [3]. The bulk of the population exposed to As-contaminated water lives in Southern Asian countries, such as Bangladesh, Cambodia, India, Nepal, and Vietnam. In addition, elevated arsenic concentrations found in groundwater have been reported in Latin American countries, such as Argentina, Bolivia, Chile, and Mexico. Due to this arsenic's high nuisance value, World Health Organization has set a provisional guideline limit of 10 μg/L for As in drinking water [4]. This alarming situation requires the development of methods for the removal of As from groundwater for drinking water.

Many technologies have been developed for arsenic removal from water and wastewater. The most

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common are chemical precipitation [5], ion exchange [6], coagulation with alum [7], and *in situ* immobilization and adsorption techniques using various adsorbents such as activated alumina [8]. However, the aforementioned methods and materials are not widely adopted because of technical and economic drawbacks. Alum coagulation for As removal, for example, has been proven to produce toxic sludges and has a low removal As capability [9]. The activated alumina, which is commercially available, needs to be replaced after four to five regeneration, which makes the removal process tedious and costly [10]. Furthermore, ion-exchange process demands high-tech operation and maintenance [11]. From the aforesaid methods, the most convenient is the adsorption techniques that have the potential for regeneration and sludge-free operation. So far, a wide range of adsorbents are available for As removal including hematite and feldspar [12], activated red mud [13], and hydrous zirconium oxide [14]. Most of these adsorbents, however, are inefficient and expensive.

Recently, industrial [15] and agricultural wastes or by-products are utilized for the removal of arsenic from water. Sawdust incorporated with lanthanum oxide was used for the removal of arsenic anions with an arsenate and arsenite adsorption capacity of 28 and 22 mg/g based on the Langmuir isotherm model [16]. Another study [17] has investigated the potential use of rice husk without any pretreatment in the removal of As from aqueous media. In this project, a cost-effective adsorbent for the efficient removal of arsenate anion from water was synthesized from an industrial by-product—mill scale. Mill scale is a steel making by-product from steel hot rolling processes and is basically composed of wustite (FeO), maghemite (γ-Fe_2O_3), and magnetite (Fe_3O_4) with variable oil and grease contents. It contains significant amounts of iron oxides that can be used in removing soluble arsenic, for it is known that iron oxides are important arsenic adsorbents [18].

Magnetite particles have been widely applied for the adsorption of arsenic [19] and phosphate [20] ions. These particles have gained special attention in water treatment because of their unique and novel properties [20]. Among the various methods in preparing magnetite, coprecipitation [21] technique is a convenient way to synthesize magnetite particles from an aqueous Fe^{2+} and Fe^{3+} salt solutions by the addition of base under an inert atmosphere. This process has a relatively low environmental impact for it does not use toxic solvents. The objectives of this project were to (i) synthesize and characterize magnetite from mill scale, and (ii) investigate the adsorption and desorption behaviors of arsenate on these particles. The magnetite particles were synthesized from mill scale via coprecitation method and characterized by X-ray diffraction and scanning electron microscopy coupled with energy dispersive X-ray spectrometer (SEM-EDS). Inductively coupled plasma optical emission spectroscopy was used to study to the adsorption capacity and kinetics of the magnetite for As(V) ions.

2. Materials and methods

2.1. Chemicals

All the reagents used in the experiment were of analytical reagent grade and used as received without further purification. The As(V) stock solution was prepared by dissolving $Na_2HAsO_4{\cdot}7H_2O$ obtained from Wako Pure Chemical Industries, Ltd (Osaka, Japan). Sodium hydroxide (NaOH) and concentrated H_2SO_4 were purchased from OCI Company Ltd (Seoul, Korea). Deionized water was used throughout the experiment.

2.2. Mill scale

Mill scale samples were obtained from "P" iron & steel company (South Korea). It was produced at the surface of a hot-rolled steel during the cooling process and showed a plate shape with a diameter of <20 mm. The collected samples were crushed with a mechanical grinder (HKP-100, Korea Pulverizing Machinery Co. Ltd, Incheon, Korea) in order to minimize as well as homogenize the particle size. The pulverized sample was sieved using a size fractionation apparatus into the following five size fractions: >300 μm, 150–300 μm, 75–150 μm, 45–75 μm, and 25–45 μm.

2.3. Pretreatment of mill scale

Pretreatment process was done to remove impurities present in the pulverized mill scale. The mill scale was heated at 550°C using a muffle furnace for 30 min to remove residual organic matter (oil and grease) and was rinsed with deionized water (MS_R). Additional pretreatment with acid (MS_A) and base (MS_B) were further conducted to activate the surface of the mill scale. The pretreated mill scale was then rinsed with deionized water until the solution pH reached around 7. The detailed pretreatment methods are summarized in Table 1 with references.

2.4. Magnetite synthesis

Magnetite particles were produced via coprecipitation method. The pretreated mill scale and concentrated

Table 1
Pretreatment methods of the mill scale (MS)

Expression	Reagent	Pretreatment methods	Refs.
MS_R	–	Rinsed three times with DI water and dried at 40˚C	–
MS_A	$HCl + H_2O_2$	100 g MS/150 ml-0.7 N HCl sol. → 100 g MS/150 ml-30% H_2O_2 sol.	[22]
MS_B	NaOH	12.5 g MS/200 ml-0.1 N NaOH	[23]

sulfuric acid were mixed in a 2 L jacketed glass reactor equipped with a mechanical stirrer, a temperature sensor, a gas inflow port for N_2 gas, and an exit gas tube. The mixture was stirred at 60˚C for 24 h under inert atmosphere. The resulting mixture is known as the ferrous precursor. The precursor was dissolved in deionized water and to the filtrate, 0.3 N NaOH was added. The black magnetite particles produced was rinsed with deionized water for three times and was dried.

2.5. Instrumental analysis

Scanning electron microscopy coupled with energy dispersive X-ray spectrometer (SEM-EDS) images of the mill scale, before and after its pretreatment process, and the synthesized magnetite particles were obtained using an S-4500 instrument (Hitachi, Tokyo, Japan). Particle size of the particles was analyzed using a SediGraph 5100 Particle Size Analyzer (Micromeritics Japan Limited Liability Company, Chiba, Japan). Powder X-ray diffraction (XRD) patterns were recorded on a Rigaku D/MAX-2500/PC (Rigaku Corporation, Tokyo, Japan) with Cu Kα radiation ($\lambda = 1.5418$ Å), operated at 200 mA at 40 kV. The adsorption capacity and kinetics of the synthesized magnetite particle for arsenate were studied using a dual-view Optima 7300 DV inductively coupled plasma optical emission spectroscopy (ICP-OES) (PerkinElmer, USA) system.

2.6. Batch adsorption tests

Preliminary adsorption tests were performed to evaluate the suitability of the pretreated mill scale for As(V) removal. Adsorption factors including the dosage and average particle size of the mill scale were evaluated. The solid content in the adsorption mixture was 1 g/L (particle size: >300 μm, 150–300 μm, 75–150 μm, 45–75 μm, and 25–45 μm) and the initial As(V) concentration was 1 ppm. The samples were shaken in a mechanical shaker (J-USRC, Jisico, Seoul, Korea) for 12 h at 250 rpm, while maintaining the pH at 6. Time point collections were done at 2.5, 5, 7.5, and 12.5 h. Samples were collected and filtered prior to ICP-OES analysis.

In order to understand the effect of dosage of mill scale on the adsorption of As(V), 1, 2, 5, and 10 g/L of the mill scale was dispersed in an arsenate solution (1 ppm). The samples were placed on a shaker for 12 h at 250 rpm, while maintaining the pH at 6. Samples were collected after 2.5, 5, 7.5, and 12.5 h and filtered prior to ICP-OES analysis.

Arsenic adsorption experiments with the synthesized magnetite were performed in a mechanical shaker. The concentration of the adsorbent was 0.5 g/L, the initial arsenic concentrations were 1 and 5 ppm, and the solution pH was maintained at pH 6. Then the suspensions were stirred at room temperature for 24 h. Effect of solution pH on the adsorption of arsenic was studied with an As(V) concentration of 5 ppm at pH 3–9. The adsorption kinetic study was carried out following the above adsorption experiment at the intervals of time: 1, 2, 3, 4, 6, 12, and 24 h. The arsenic concentration was analyzed using ICP-OES instrument.

2.7. Column adsorption test

Column adsorption test was performed using a magnetite packed-bed column consisting of three divisions. The lower and upper part of the column were both packed with two different sizes of glass beads (1 and 10 mm in diameter) while the middle part was packed with magnetite:garnet (1:1 v/v) mixture, with a packing volume of 98 cm^3 (490 g). To prevent the leaching of the magnetite particles from the middle part of the column, a magnetic bar with screen was placed. Arsenic(V) solution (1 ppm) was pumped into the column with an empty bed contact time (EBCT) of 1–2 h. The column was operated for 133 d.

2.8. Desorption test and recycling of magnetite

Arsenic adsorbed on the surface of the synthesized magnetite particles was desorbed using 0.1 N NaOH and by shaking the mixture for 2 h using a mechanical shaker at 250 rpm. Samples were taken and submitted for ICP-OES analysis to calculate the amount of As(V) desorbed. Five successive cycles of adsorption–desorption, using the same magnetite particles, were monitored to assess the regeneration ability of the synthesized particles. Also, the magnetite's efficiency for re-adsorption of arsenate ions in repeated cycles were monitored and compared. The particles were thoroughly washed with deionized water to neutrality and reconditioned for adsorption in the succeeding cycle after each cycle of adsorption–desorption.

3. Results and discussion

3.1. Characterization of mill scale and magnetite

The physical and chemical characteristics of the pulverized mill scale are presented in Table 2. The mill scale used was an abundant material in the steel manufacturing factory, and its size distribution was as follows: 11% > 300 μm, 25% 150–300 μm, 28% 75–150 μm, 27% 45–75 μm, and 9% 25–45 μm. Because the main components of mill scale are iron oxides (Table 3), mill scale can be used to prepare magnetite via coprecipitation method using mill scale-derived precursors.

The XRD analysis of the mill scale (Fig. 1(a)) showed the presence of the three iron oxides—wustite, hematite, and magnetite. Acid digestion of the raw mill was done to increase the iron content in the products formed. It was reported [24] that the ferrous precursor obtained after the acid digestion contains more of ferrous compound ($FeSO_4{\cdot}H_2O$) and less of ferric compounds ($3Fe_2O_3{\cdot}8SO_3{\cdot}2H_2O$, $FeOHSO_4$ and α-FeOOH).

The XRD analysis of the synthesized particles (Fig. 1(b)) showed the presence of magnetite phase. To further establish the identity of the particle, the obtained X-ray powder diffractogram of the particle was compared with the Joint Committee for Powder Diffraction Standards (JCPDS) card no. 19-0629 for magnetite. The stoichiometry of the magnetite prepared from mill scale was reported [24] to be $Fe_{2.894}O_4$. This stoichiometry is acceptably close and implies that the crystal structure of the synthesized magnetite is slightly distorted. This distortion is the result of the scarcity of Fe ions [24]. The obtained SEM-EDS micrograph of the particles, presented in Fig. 2, further showed that the powder contains aggregated spherical particles of sizes between 10 and 50 μm. These particles were observed to be smaller than the raw mill scale. Also, the particles were regular and uniform suggesting that the powder will show a quality hiding power [25].

Table 3
Composition of the raw mill scale

Component	FeO	Fe_2O_3	Fe_3O_4	Fe^0	Other
Amount (wt%)	59.7	8.3	23.5	3.1	5.4

3.2. As(V) adsorption on mill scale

Fig. 3 presents the test results of arsenate adsorption by mill scale. Mill scale was found to adsorb arsenic. However, arsenate adsorption is significantly affected by the dosage and particle size of the mill scale. Fig. 3(a) shows the time-dependent plot with adsorbent particle size (>300 μm, 150–300 μm, 75–150 μm, 45–75 μm, and 25–45 μm). The percent removal of arsenate increased from 17 to 95 with decrease in particle size of the mill scale. This implies that the smaller the adsorbent particle size, the greater will be the adsorption. This can be explained by the fact that the smaller size, the larger surface area a particle has wherein the arsenic can be adsorbed. The time-dependent plot with dosage of adsorbent (1, 2, 5, and 10 g/L) is shown in Fig. 3(b). The percentage removal was observed to increase with increase in adsorbent dosage. This indicates that the higher the dosage of mill scale, the more the areas wherein arsenate can be adsorbed, the greater the adsorption.

3.3. As(V) adsorption on magnetite

The amount of arsenate adsorbed on the adsorbent are calculated using Eq. (1) and the percentage removal of arsenate being plotted is $100(C_o - C_e)/C_o$.

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

Table 2
Physical and chemical characteristics of the raw mill scale

Physical/chemical properties	Analysis results				
Particle size (μm)	25–45	45–75	75–150	150–300	>300
Weight fraction (%)	9	27	28	25	11
Solution pH (1 g/10 ml)	6.67–6.81		(pH, BET surface area, and pore size of particles (size: >75 μm) were not analyzed)		
BET surface area (m^2/g)	1.34–1.36				
Pore size (cm^2/g)	2.14×10^{-3}–2.18×10^{-3}				

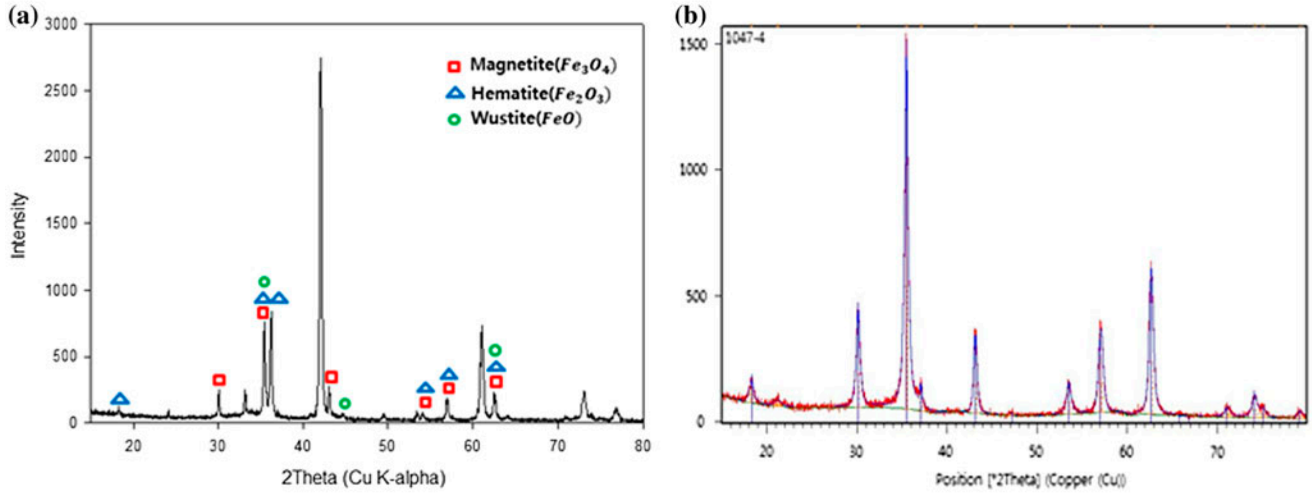

Fig. 1. X-ray powder diffractogram of raw mill scale (a) and magnetite obtained from mill scale (b) (line across in (b)) magnetite (JCPDS 19-0629).

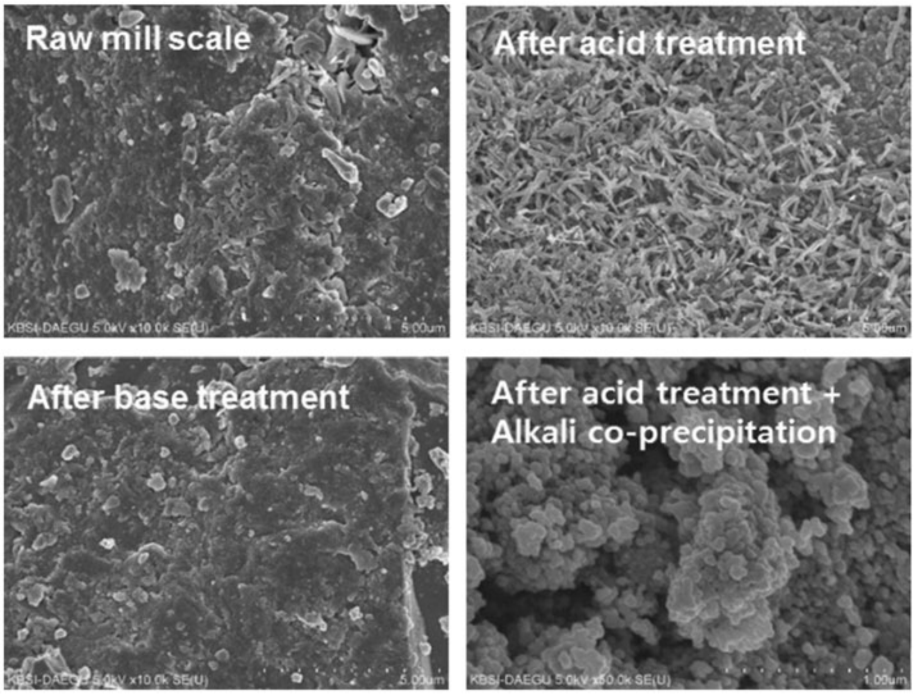

Fig. 2. SEM–EDS images of the mill scale after and before pretreatment processes and synthesized magnetite particles (lower right box).

Here q_e (mg/g) is the adsorption amount at equilibrium condition, C_o (mg/L) is the initial arsenic concentration, C_e (mg/L) is the equilibrium arsenic concentration, V is the volume of aqueous solution, and m is the amount of adsorbent used in the experiments.

3.3.1. Time-dependent studies

To understand the kinetics of arsenic adsorption onto the magnetite surface, data obtained from the experiment were analyzed using both pseudo-first- and pseudo-second-order kinetic models [26], which

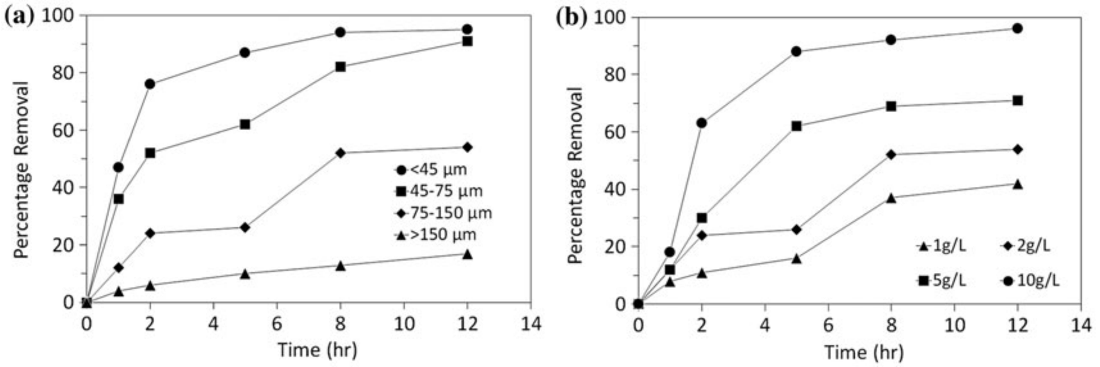

Fig. 3. Time-dependent studies of arsenic removal using raw mill scale with varying particle size (a) and dosage (b) of mill scale. Conditions: (a) [As(V)] = 1 mg/L; adsorbent concentration = 1 g/L; adsorbent particle size: >300 μm, 150–300 μm, 75–150 μm, 45–75 μm, and 25–45 μm and (b) [As(V)] = 1 mg/L; adsorbent concentration = 1, 2, 5, and 10 g/L; adsorbent particle size: 45–75 μm.

were, respectively, presented as follows in Eqs. (2) and (3):

$$\log(q_\mathrm{e} - q_t) = \log\, q_\mathrm{e} - \left(\frac{k_1}{2.303}\right)t \tag{2}$$

Here q_e and q_t (both in mg/g) are the amount of arsenic adsorbed at equilibrium per unit mass of adsorbent at equilibrium and time t, respectively. The rate constant (k_1) was determined experimentally by plotting $\log(q_\mathrm{e} - q_t)$ against t.

The pseudo-second-order model in the linear form is expressed as follows:

$$\frac{t}{q_t} = \frac{t}{q_\mathrm{e}} + 1/(k_2\, q_\mathrm{e}^2) \tag{3}$$

Similarly, the rate constant (k_2) was determined by plotting t/q_t against t.

Time-dependent arsenic removal studies, presented in Fig. 4, showed that magnetite removed 99% of arsenate anion from the solution at an initial concentration of 1 ppm after 6 h. However, at an initial concentration of 5 ppm, magnetite only removed 88% of arsenate anion after 24 h. The big difference in arsenic removal can be attributed to the fact that different rates of adsorption are observed at lower arsenic concentration. In this case, electrostatic attraction is the dominant adsorption mechanism for magnetite at low arsenic concentration (1 ppm).

The calculated kinetic parameters are given in Table 4 and the kinetic plots are presented in Fig. 5. It is clear in Fig. 5 that the adsorption process follows pseudo-second-order reaction. Moreover, the highest obtained arsenate adsorption using magnetite adsorbent was found to be 8.612 mg/g. This value was also found to be higher than the adsorption capacity of mill scale, which was calculated to be 0.455 mg/g. Natural Fe–Mn enriched samples [27], Fe–Mn binary oxide [28], and magnetite [29] have been proven to be

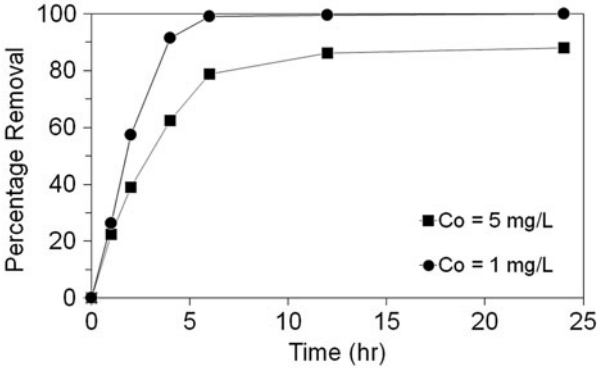

Fig. 4. Time-dependent studies of arsenic extraction using synthesized magnetite with initial arsenic concentration of 1 and 5 mg/L. Conditions: [As(V)] = 1 and 5 mg/L; adsorbent concentration = 0.5 g/L; pH 6.

Table 4
Pseudo-first-order and second-order kinetics parameters for arsenate adsorption on magnetite

		C_o (mg/L)	
Kinetic models	Kinetic parameters	1	5
Pseudo-first-order	q_e (mg/g)	1.5943	8.8436
	k_1 (min^{-1})	0.0084	0.0055
	R^2	0.8763	0.9901
Pseudo-second-order	q_e (mg/g)	2.1756	9.9475
	k_2 (min^{-1})	0.0048	0.0006
	R^2	0.9877	0.9922

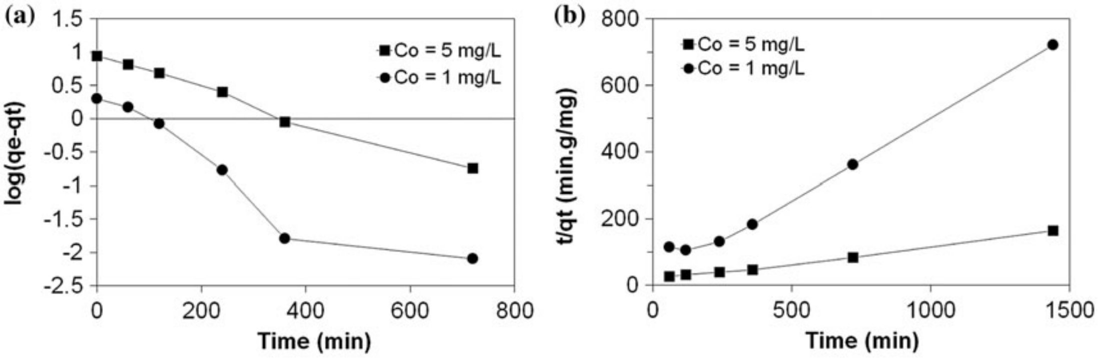

Fig. 5. Pseudo-first-order (a) and pseudo-second-order (b) kinetic plots for arsenate adsorption on magnetite.

Table 5
Adsorption results using various adsorbents

Adsorbent	Adsorption capacity (mg/g)	Refs.
Fe–Mn composite	69.7	[28]
Fe–Mn mineral material	6.7	[27]
Magnetite	1.1	[29]
Magnetite	8.612	Present work

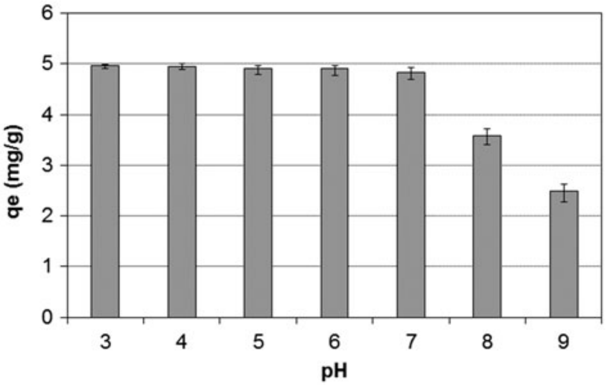

Fig. 6. Effect of pH on As(V) adsorption using magnetite. Conditions: [As(V)] = 5 mg/L; adsorbent concentration = 0.5 g/L; pH 3–9.

promising arsenic adsorbents. The summary of these results are given in Table 5.

3.3.2. Effect of pH

As shown in Fig. 6, the adsorption of arsenate anion was apparently dependent on pH, and the arsenate adsorption was high under acidic conditions. The dominant arsenate species in the range of 3–9 were $H_2AsO_4^-$ and $HAsO_4^{2-}$ [30]. The adsorption mechanism was described to be a mixture of inner-sphere surface complexation and electrostatic interaction [16]. The large dropped on arsenate anion adsorption at pH 8 and 9 is likely due to repulsion of negatively charged surface of magnetite and anionic arsenic species. At pH below PZC of adsorbents, the surface hydroxyl groups are protonated to form OH_2^+ [31]. However, with an increase of solution pH, the surface hydroxyl groups gradually undergo deprotonation that made the adsorbents surface negatively charged. This causes the repulsion with the anionic arsenate and was unfavorable for arsenate adsorption. This observation further implies that electrostatic interaction is the dominant adsorption mechanism.

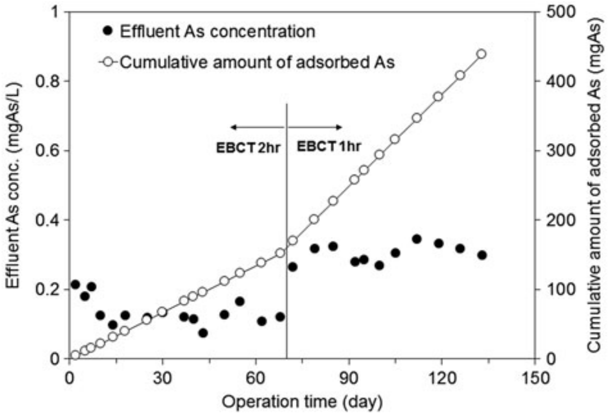

Fig. 7. Time-dependent studies of arsenic removal during the packed-bed column test (input As(V) concentration = 1 mg/L).

3.3.3. Column test

The uptake of arsenate ions are shown in Fig. 7. For a two-hour empty bed contact time (EBCT), the observed arsenate removal was found to be 86.5% which is significantly higher than 69.7%, which is the

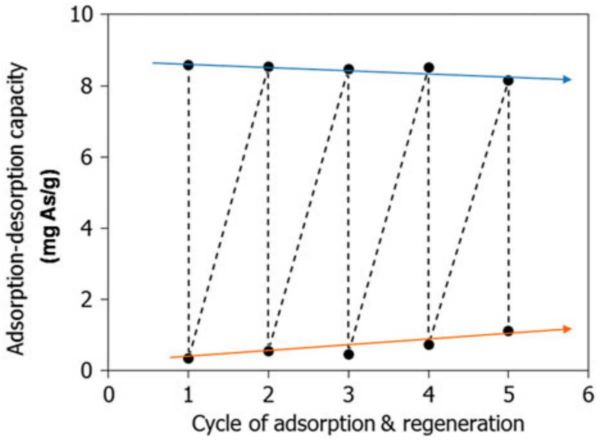

Fig. 8. Desorption and regeneration study of magnetite particles.

obtained arsenic removal after an hour of EBCT. An empty bed contact time of 2 h gave enough time for the As(V) ions to be adsorbed on the synthesized magnetite particles. Furthermore, the cumulative amount of adsorbed arsenate was found to be 439 mg-As after 133 d of operation.

3.4. Arsenic desorption study

Desorption of arsenate anion from magnetite particles was performed using 0.1 N NaOH. The desorption process occurred by the replacement of arsenic anion by OH^- ion on the surface of magnetite. Hence, sodium hydroxide solution was used to desorb the adsorbed arsenic from the adsorbent. Data about the desorption studies were presented in Fig. 8. Magnetite was successfully regenerated without any change in efficiency. Also, the adsorption capacity is expected to decrease by 20% after 10 times of regeneration.

4. Conclusions

In summary, low-cost mill scale was utilized to prepare magnetite. The synthesized magnetite was used as an efficient adsorbent for arsenate anion. The prepared magnetite showed an excellent ability to remove arsenate ions in water. In comparison to similar magnetite materials reported in the literature, the mill scale-derived magnetite showed a relatively higher adsorption capacity. Electrostatic interaction is the observed dominant adsorption mechanism. Furthermore, arsenate adsorption follows pseudo-second-order kinetic model. Magnetite particle shows reused capacity up to five cycles of adsorption.

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