



Adsorption of Arsenic from Aqueous Solutions by Iron Filings and the Effect of Magnetic Field

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ABSTRACT

Aims Arsenic contamination of natural water resources has become an important environmental problem in the world. The adsorption method by iron filings adsorbent or zero-valent iron was used. The study aimed to evaluate the efficiency of iron filings in arsenite removal from polluted water and to investigate the effect of magnetic field on the process.

Materials & Methods This interventional study was conducted in synthetically polluted water samples with certain arsenic concentrations. With regard to the initial arsenic concentration (0.5 and 2mg/l), iron filings dosages (0, 2.5 and 5g/l), contact times (5, 10 and 15min) and considering the samples before and after magnetic column, 108 samples were prepared. Data was analyzed by paired sample T and one-way ANOVA tests.

Findings The highest mean of removal efficiency at the initial arsenic concentration of 0.5mg/l was seen at the iron filings of 5g/l and 10min contact time (87.7 ± 10.0) and at the initial arsenic concentration of 2mg/l was seen at the iron filings of 5g/l and 15min contact time (86.3 ± 8.4). At the initial arsenic concentration of 0.5mg/l, magnetic field increased the removal efficiency of arsenite at the iron filings dosage of 0g/l and decreased it at the iron filings dosage of 5g/l. The same happened at the initial arsenic concentration of 2mg/l.

Conclusion Arsenic is reduced from the water samples with the iron filings dosage of 5g/l at natural pH. Magnetic field increases the arsenic removal efficiency in the absence of the iron filings and decreases the arsenic removal efficiency in the presence of the iron filings.

Keywords Adsorption; Magnetic field; Arsenite

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Introduction

Today arsenic contamination of natural water resources has become an important environmental problem in the world, and is often referred to as a 21st century calamity [1]. Arsenic is a toxic metalloid and exists in nature in the two organic and mineral forms [2]. Arsenate is the oxidized form and is predominant in the surface waters, while arsenite is a reduced form and is often found in the groundwater. Besides, toxicity and solubility of arsenite is more than that of arsenate. Arsenic pollution in water sources is an outcome of natural and anthropogenic sources [3]. Excessive and prolonged human intake of inorganic arsenic, through drinking water and food, causes arsenicosis, which includes skin disorders, skin cancer, internal organ cancer (bladder, kidney, liver, and lung), arm and leg vascular diseases and diabetes [4]. Arsenate closely resembles the phosphate ion and creates disorder in human energy metabolism while arsenite can link with reactive sulfur atoms present in many human enzymes and cause serious problems [5].

The World Health Organization's (WHO) guideline value for arsenic in drinking water is set as 10µg/l [6]. Regarding the world standard changes of arsenic in drinking water from 50 to 10µg/l, many of the sources of drinking water supply in Asia are among the waters that are contaminated with arsenic [7]. Several reports on the existence of increased levels of arsenic in groundwater have been made in different countries [4].

Arsenic removal depends highly on the composition and chemistry of the polluted water [8]. There are three main methods for arsenic removal from drinking water, including membrane filtration, coagulation-precipitation and adsorption [9]. The removal methods of arsenic need to address both the technological and the socioeconomic considerations [10]. Considerable features of the treatment methods based on adsorption for arsenic removal include its cost-effectiveness, easy operation and maintenance, whereas the other treatment methods do not have these advantages [11]. This adsorbent is low-cost, effective, available and reusable and also does not leave any toxic or harmful chemicals in drinking water [12]. In contact with water and oxygen, iron filings

produce iron oxides, which have the main role in the removal of arsenic compounds [1].

Zeng proposes a method for preparing iron filings as adsorbent for arsenic removal, and the adsorption capacity was reported 0.3mg/g [13]. Lien & Wilkin also have conducted a study on arsenic removal by iron filings [14]. Ramaswami *et al.* have removed the arsenite ion using iron filings and the removal efficiency of 93% was reported [15]. Also, Su & Puls managed to remove arsenite and arsenate by using iron filings and achieved the removal efficiency of 99% [16]. In Iran, Asgari *et al.* have investigated the efficiency of iron filings in arsenate and arsenite removal from drinking water [17]. Water treatment by means of the magnetic field has been recently considered [18]. Ma *et al.* have investigated the arsenic removal via sulfide ions in the magnetic field [19]. Coey & Cass as well, carried out a study on water treatment using the magnetic field [20]. Yean *et al.* have removed arsenic from drinking water by magnetic particles and proposed the magnetic properties of the process [6].

In this study, the adsorption method by iron filings adsorbent or zero-valent iron was used. The study aimed to evaluate the efficiency of iron filings in arsenite removal from polluted water and to investigate the effect of magnetic field on the process.

Materials & Methods

Sampling

This interventional study was conducted in synthetically polluted water samples with certain arsenic concentrations. With regard to the initial arsenic concentration (0.5 and 2mg/l), iron filings dosages (0, 2.5 and 5g/l), contact times (5, 10 and 15min) and also considering the samples before and after magnetic column, which was repeated 3 times, overall of 108 samples were prepared.

All tests were conducted in neutral pH because the pH of the natural waters is close to neutral pH. The iron filings, unlike the other adsorbents, have a high affinity to the reaction with arsenic at the normal pH of water. All the experiments were conducted in the research laboratory in Department of Environmental Health Engineering at Kashan University of Medical Sciences at room temperature (25°C).

Preparation of adsorbent

The required iron filings were prepared in the

turning workshops of Tehran, Iran. The iron filings were passed through a sieve with pores of 2mm and were made wet using deionized water, and ferric hydroxide precipitation was allowed to be formed on the surface of the filings. Iron filings used in this study were heterogeneous and non-uniform in structural geology and existed mostly in semi-cylindrical forms.

Making the magnetic column

A circular magnet with the magnetic field intensity of 0.01T and outer diameter of 7cm and inner diameter of 3cm was primarily placed around a glass column with the height of 28cm and diameter of 1.5cm, and a iron spiral with the length of 6cm was located in front of the magnet inside the column, so that, the sediments on it could be reacted. Further, a switching valve was applied at the end of the column for the discharge velocity adjustment, which the discharge velocity was 2mm/s.

Preparation of the Samples

The synthetically arsenic-polluted water samples were prepared by diluting 0.1N sodium arsenite solution (Merck; Germany) with de-ionized water. In addition, to prevent the oxidation of arsenite to arsenate the required solutions were prepared on a daily basis at 0.5 and 2mg/l concentrations. In the first place, a stock solution with a certain concentration was prepared, and then the samples were prepared by it.

Procedure

The prepared arsenite solutions at 0.5 and 2mg/l concentrations were reacted in contact with the iron filings adsorbent at 0, 2.5 and 5g/l dosages over contact times of 5, 10 and 15 minutes within a beaker with the volume of 100ml on the shaker at the velocity of 400rpm. Half of the sample (50ml) was then passed through the column at the velocity of 2mm/s and a blank (without iron filings) was considered for each sample. The blank was also passed through the column at the same velocity. Then, the samples before and after the column were passed through a filter with pores of 0.45 μ m and their pH was reduced to below 2 using the intact concentrated nitric acid.

Statistical Analysis

The arsenic removal efficiency in any state was calculated and analyzed by SPSS 11.5 software using paired sample T-test and one-way ANOVA test for comparing arsenic

removal efficiency with dosages of iron filings and comparing the initial concentration of arsenic, and the iron filing dosage with the electrical conductivity and also comparing the parameters of the magnetic field with the electrical conductivity.

Findings

The highest mean of removal efficiency at the initial arsenic concentration of 0.5mg/l was seen at the iron filings of 5g/l and 10min contact time (87.7 \pm 10.0) and at the initial arsenic concentration of 2mg/l was seen at the iron filings of 5g/l and 15min contact time (86.3 \pm 8.4; Figure 1). The increase in the arsenic removal efficiency due to the increasing dosage of iron filings, were statistically significant ($p \leq 0.001$).

At the initial arsenic concentration of 0.5mg/l, magnetic field increased the removal efficiency of arsenite at the iron filings dosage of 0g/l and decreased it at the iron filings dosage of 5g/l. The same happened at the initial arsenic concentration of 2mg/l (Figure 1).

Figure 1) Arsenic removal efficiency (%) based on iron filings dosage (0, 2.5 and 5g/l) and contact times (5, 10, 15min) according to initial arsenic concentration (0.5 and 2mg/l) before and after the magnetic column

Parameters	0.5 (mg/l)		2 (mg/l)	
	Before	After	Before	After
0 (g/l)				
5min	34.7 \pm 4.4	42.1 \pm 2.8	41.5 \pm 4.2	43.6 \pm 3.9
10min	36.8 \pm 2.8	49.1 \pm 1.8	43.3 \pm 3.2	50.9 \pm 3.4
15min	38.7 \pm 1.8	55.3 \pm 0.4	46.2 \pm 4.0	58.7 \pm 4.5
2.5 (g/l)				
5min	74.7 \pm 15.1	62.5 \pm 8.0	59.1 \pm 6.9	57.0 \pm 7.4
10min	66.8 \pm 12.3	60.1 \pm 10.6	64.9 \pm 4.3	63.1 \pm 3.8
15min	73.6 \pm 11.7	75.4 \pm 15.3	63.5 \pm 3.4	61.4 \pm 3.8
5 (g/l)				
5min	79.5 \pm 11.9	73.2 \pm 14.6	67.4 \pm 10.2	61.7 \pm 6.2
10min	87.7 \pm 10.0	84.2 \pm 13.4	74.3 \pm 6.4	70.5 \pm 4.9
15min	75.6 \pm 22.9	78.7 \pm 7.7	86.3 \pm 8.4	81.8 \pm 6.1

The interaction of the initial concentration of arsenic and the iron filings dosage on the electrical conductivity changes was statistically significant ($p \leq 0.0001$). There was also a significant relationship between the magnetic field and electrical conductivity ($p \leq 0.0001$).

Figure 2 shows the effects of magnetic field on the arsenic removal efficiency versus contact time based on different iron filings dosages and varies initial arsenic concentrations.

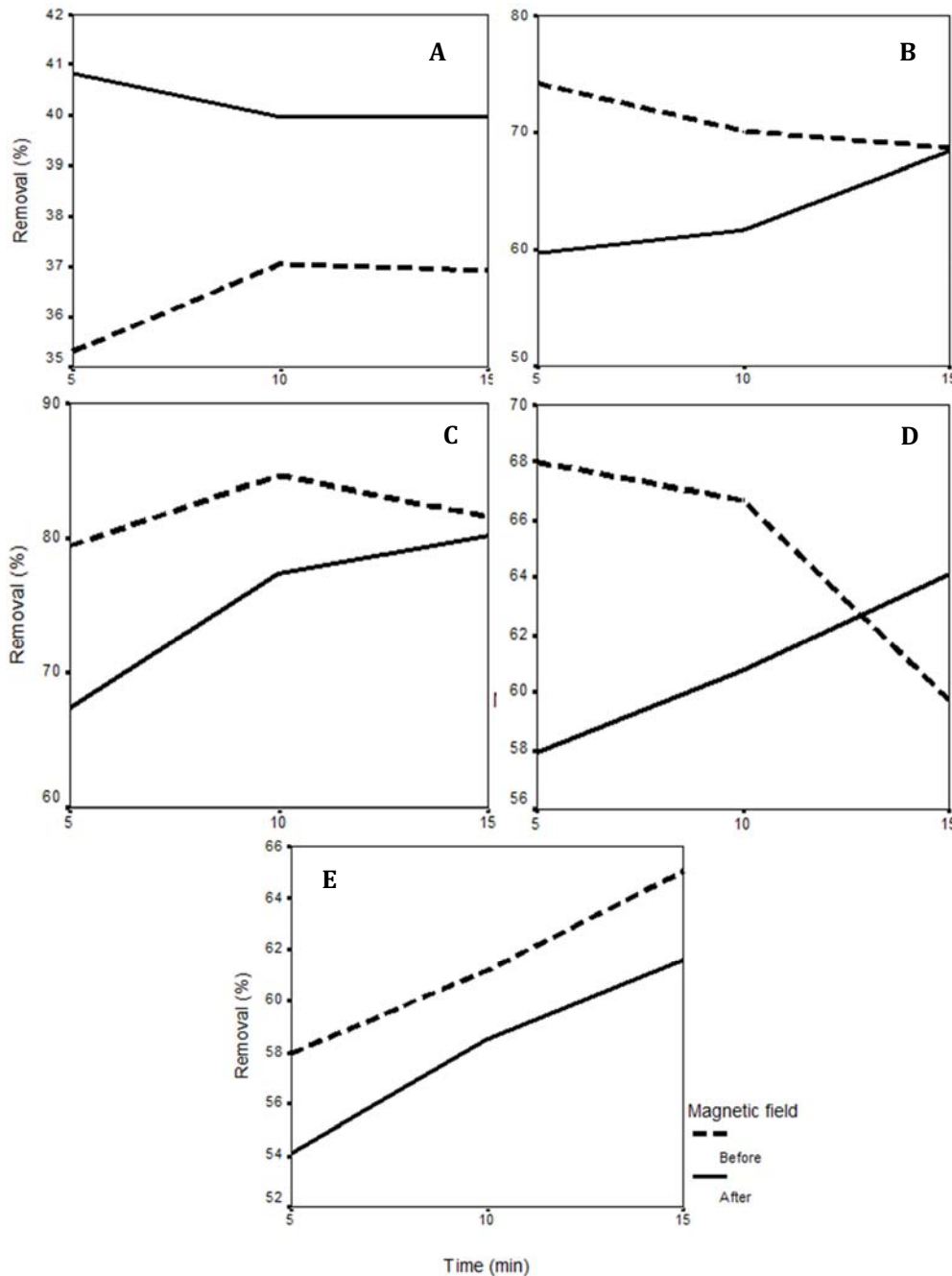


Figure 2) Effects of magnetic field (After is shown with complete line and before with dashed line) on the arsenic removal efficiency versus contact time according to iron filings dosages of 0g/l (A), 2.5g/l (B) and 5g/l (C), and also the initial arsenic concentration of 0.5mg/l (D) and 2/mg/l (E)

Discussion

The aim of this study was to evaluate the efficiency of iron filings in arsenite removal from polluted water and to investigate the effect of magnetic field on the process. The results indicated that with the increase of the initial arsenic concentration, the removal efficiency was also increased that could be

due to the oxidation of arsenite into the insoluble arsenate ion. Via analysis of iron filings by extraction with hydrochloric acid, Hsing *et al.* [14] have shown that almost 28% of arsenic has existed in the form of arsenate, which revealed that oxidation has also been effective in arsenic removal. For the initial arsenic concentration of 2mg/l

and the iron filings doses of 2.5 and 5g/l, the average removal efficiency was reported as 62.5% and 76.1% and for the concentration of 0.5mg/l, it was reported as 71.7% and 80.9%, respectively. The results have demonstrated that with the increase of the initial arsenic concentration, the arsenic removal efficiency decreased. By reducing the concentration of arsenic, the ratio of adsorbent to adsorptive increases and the removal efficiency increases [9]. Zhang *et al.* [21] have reported the adsorption capacity of 16mg/g at the arsenic concentration of 1mg/l, while Lien & Wilkin [14] have reported the adsorption capacity of 7.5 at the arsenic concentration of 50mg/l. Based on the obtained results, with the increase of the iron filings dosage, the arsenic removal efficiency increased as well. With increasing the dosage of iron filings, adsorption sites and surfaces increase and as a result, the reaction between the arsenite ion and ferric hydroxide occurs more. Tyrovola *et al.* [22] have shown that with the increase of the iron filings dose, the removal efficiency of arsenite ion increases.

At the arsenic concentration of 0.5mg/l and the iron filings dosages of 2.5 and 5g/l, the desorption occurred at the contact times of 10 and 15 minutes, respectively and also at the arsenic concentration of 2mg/l and the iron filings dose of 2.5g/l, it occurred at the contact time of 15 minutes. The results showed with the increase of the contact time desorption can occur at various times. In the samples with high arsenic concentration and iron filings, due to ferric hydroxide sites on the iron filings and therefore higher adsorption, the desorption process occurred at longer contact times [9]. In the desorption process, the arsenite ions that are negatively charged and are attached to the ferric hydroxide ions are separated from iron oxides at different contact times and the removal efficiency decreases. Asgari *et al.* [17] have investigated the arsenic removal by using iron filings at the contact times of 5, 10, 15, 30 and 60 minutes. The results of that showed the maximum removal efficiency at the 30 minutes contact time.

The ferric hydroxide ion was formed on the surface of the iron filings. Sodium arsenite (NaAsO_2) reacts with ferric hydroxide ($\text{Fe}(\text{OH})_3$) and forms ferric arsenite ($\text{Fe}(\text{AsO}_2)_3$) on the surface of the iron filings.

Also, ferrous hydroxide ion is formed within the solution and can react with sodium arsenite and ferrous arsenite ($\text{Fe}(\text{AsO}_2)_2$) can be thus formed. Based on the physicochemical Hall Effect, when a multi-atomic ion placed within a fluid passes through the external magnetic field, the bond between the ions is weakened and they are dissociated and form cations and anions. When charged particles are placed in a magnetic field, a force is applied by the magnetic field to the particle, which is called "Lorentz force" [18]. After the blank or control sample (sodium arsenite) passed through the magnetic field, these two ions were dissociated based on the Hall Effect and finally reacted with the ferric hydroxide formed on the metal spiral. When the ferric arsenite ion passed through the magnetic field, the ions were dissociated and were affected by Lorentz force. Arsenite has one negative charge and ferric has three positive charges and since ferric ion has higher charge, more force is applied to it and it attaches to the spring inside the column. As a result, the amount of ferric ion in the outlet column decreased and the sample became less colored. Water discoloration meant that the iron residual was more than the standard amount (0.3mg/l), which can be solved by using lower dosage of iron filings. Arsenite was also affected by Lorentz force and reacted with the ferric formed on the spring, but as the dissociation level of ferric arsenite ion was more than its adsorption, the arsenite level in the outlet column increased. Ferric arsenite is insoluble and was not measured by the device. When the solutions were passed through the column, arsenite separated from ferric and changed into a solution which could be measured. Ma *et al.* [19] have reported that the magnetic field can weaken the bond between the ions. In this test, the fluid velocity was considered as 2mm/s and the intensity of the magnetic field was considered as 0.4T. When the charged particles are placed in a magnetic field, a force is applied by the magnetic field to the charged particle, which is called "Lorentz force". The maximum force happens when the field lines are perpendicular to the direction of the movement of ions.

The iron filings, unlike the other adsorbents, have a high affinity to the reaction with arsenic at the normal pH of water. Ramaswami *et al.* [15] have removed the

arsenite by iron filings at the pH of 7 with the efficiency of 95%. In another study that was conducted by Su & Puls [16], the arsenite and arsenate ions were removed at the pH of 7 using iron filings and the removal efficiency of 99%.

The results showed that the magnetic field reduced the arsenic level of the samples without iron filings but increased the arsenic level of the samples with iron filings.

The results of this study demonstrated that the magnetic field increased the electrical conductivity. When the ions of a solution are exposed to the magnetic field, they are dissociated and the solution forms more ions and thus electrical conductivity increases. Ma et al. [19] have shown that electrical conductivity of the samples before and after the magnetic field were 0.22 and 0.27 $\mu\text{S}/\text{m}$, respectively.

Conclusion

The most efficient removal condition of arsenic from natural pH water is with the iron filings dosage of 5g/l. Magnetic field increases the arsenic removal efficiency in no iron filings and decreases the arsenic removal efficiency with iron filings.

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