

Removal of Arsenic from Aqueous Solutions Using Welding Iron Waste

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Abstract

Background: Contamination of water with arsenic has attracted the researchers' attention as a global problem in recent years and has been observed in some parts of Iran. The purpose of this study is to assess the efficiency of welding iron waste in removing arsenic from aqueous solutions.

Methods: In this study, the effects of different parameters, such as pH (3-9), initial concentration of arsenic (100-3000 µg/l), contact time (5-90min) and adsorbent dose (2.5-20 g/l), were studied. The final concentrations of arsenic were analyzed by atomic absorption.

Results: The results indicated that at pH=3 and fixed dose of 1 g, arsenic removal efficiency of iron waste was 89.73%. By increasing the pH to 7, the removal efficiency increased to 96.44%. Also, an increase in the amount of iron waste from 2.5 to 10g/l, the removal rate increased from about 42.37% to 96.70%. For contact times of 5 and 30 minutes, the removal rate was 9% and 96.62%, respectively. Then, with increasing the contact time to 90 minutes, the removal rate increased to 99.24%. Correlation coefficient of Freundlich and Langmuir isotherms for As(III) was 0.7593 and 0.9979, respectively.

Conclusion: The results of the study showed that welding iron waste has a high potential as an effective, fast and cheap method for removal of arsenate and arsenite from aqueous solutions.

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Introduction

One of the most common pollutants and toxic agents for groundwater is arsenic. In many countries, especially developing ones, arsenic concentrations in the drinking water has exceeded the standards and has received attention as a big problem.^{1,2} Arsenic is a toxic element in drinking water that enters the human body and has harmful health effects. Arsenic enters water supplies from natural and synthetic processes,³ including natural processes, such as earth erosion, drainage water from soil washing and weathering, industrial activities for mining, and smelting of metals from ores, combustion of fossil fuels, application of arsenic-bearing pesticides and plants' wastewater which pollutes the air, soil and water with arsenic.⁴ Arsenic in groundwater is mainly found in the form of arsenate (HAsO_4^{2-} , H_2AsO_4^- , H_3AsO_4),

and arsenite (HAsO_3^{2-} , H_2AsO_3^- , H_3AsO_3).^{5,6} Arsenic is a toxic cumulative substance, and is an inhibitor of SH group enzymes. The trivalent arsenic (arsenite) is usually more toxic than the pentavalent form (arsenate).⁷ Due to the presence of arsenic in drinking water sources, serious health problems have arisen in several countries, including Argentina, Bangladesh, Chile, China, India, Italy, Japan, Mexico, Malaysia, Nepal, Poland, Taiwan, Vietnam, and Iran. Due to the importance of the issue, the US Environmental Protection Agency reduced its MCL standard for arsenic from 50 to 10 ppb in January 2001.^{3,8} The World Health Organization has adopted MCL for arsenic of 10 micrograms per liter.^{6,9} Thus, given the enormous risks of arsenic in water supplies, removing it from water sources is very important. In recent years, various treatment methods have been proposed and used to remove arsenic from

water plants, such as chemical oxidation, dissolved air flotation, surface adsorption, ion exchange, membrane processes, electrical coagulation, chemical coagulation, and biological processes. According to their particular circumstances, water treatment systems may choose one of these methods with regard to their economic and management considerations.^{10,11} While these methods are widely used, they are faced with such problems as the high costs of operation, waste treatment, consumption of large amounts of materials and production of high volumes of sludge.^{12,13} Therefore the use of zero-valent metals (such as Fe⁰) has been studied as an effective method for reducing water contamination in recent years. The use of Fe⁰ has received more attention in this regard because of its frequency, cost-effectiveness, non-toxic nature, quick reaction, and high efficiency and ability to break down contaminants.^{6,7,9,10} The mechanism of removing contaminants by Fe⁰ includes reduction or absorption processes.¹¹ In aerobic conditions, both reduction and absorption processes are involved in arsenic removal; however, due to the greater speed and efficiency of the absorption process, it is introduced as the main factor.¹⁴ Therefore, given the benefits mentioned for using Fe⁰, the main objective of this study is to evaluate the direct use of welding iron waste for removal of arsenic from aqueous solution and the effect of various parameters on the process.

Materials and Methods

This is fundamentally a laboratory scale study. The purpose of this study was to assess the efficiency of welding iron waste in removing arsenic from aqueous solutions. The effects of operating parameters, such as pH,³⁻⁹ initial of arsenic concentration (100-3000 µg/l), contact time (5-90 min), and adsorbent dose (2.5-20 g/L) were studied. This study was carried out at the main laboratory of environmental sciences at Shiraz University of Medical Sciences in 2014.

Preparation of the Adsorbent

The welding iron required for the study was obtained from welding workshops in Shiraz. The filings were obtained from the wastes resulting from lathing operations inoculated with oil and soap. To remove these impurities, the waste iron was washed with distilled water for several times until the residuals of oil and soap were cleaned from the surface. Then, it was put in an oven at a temperature of 100°C for 1 h and the iron waste was passed through a 100 mesh sieve.

Preparation of the Solution

Arsenate and arsenite stock solution (100 mg/l) was prepared via a solution of sodium arsenate (Na₂HAsO₄·7H₂O) and sodium arsenite (NaAsO₂) salts in distilled water. Then, the standard solution

for testing was obtained via diluting the stock solution.

In all experiments, the volume of the solution used in 250-ml Erlenmeyer flask was 50 ml. In all phases of the experiments, different concentrations of arsenate and arsenite were prepared and added to the 250-ml Erlenmeyer flask. pH was set in the desired range by normal HCL and NaCl. Next, a certain amount of welding iron waste (2.5-20 gr/L) was added to the solution and the resulting suspension was immediately placed on a shaker at 200 rpm for a certain period of time (5-90 min). After the contact times, to separate metal waste from each container, 10-ml samples were taken by pipette and centrifuged for 10 minutes at 20 rpm. Finally, the concentration of arsenic remaining in the solution was measured using atomic absorption spectrometry. To investigate the effects of different parameters on the removal efficiency, at each stage, one of the parameters was changed and the others were kept constant and the removal efficiency was calculated.

$$Re(\%) = (C_0 - C_t) / C_0 \times 100$$

Re: arsenic removal efficiency (%)

C₀: initial concentration of arsenic (µg/l)

C_t: concentration of arsenic remaining in the solution (µg/l)

Results and Discussion

The Effect of Initial pH on the Arsenite Removal Efficiency

In this study, as seen in Figure 1, the effect of pH on arsenate removal by welding iron waste in the range of 3-9 with initial arsenite concentration of 500 µg /l and reaction time of 30 minutes was examined. Based on the results presented in this Figure, with an increase in pH, the removal efficiency increased. The results also indicated that at pH=3, the removal efficiency was 89.73%. By increasing the pH to 7, the removal efficiency increased to 96.44%. After that, increasing the pH to 9 did not reveal a significant effect on arsenate removal efficiency which gets close to an almost constant removal efficiency rate. Thus, these results show that the initial pH of the solution is important in achieving maximum removal efficiency. In this study, though the removal efficiency was more than 89% for all the studied pH, the highest removal efficiency was achieved at neutral pH. The iron waste was capable of removing arsenite from water at a wide range of pH. Generally, the pH changes determine the arsenic ionic species and the adsorbent surface charge. This situation will affect the interaction between the adsorbent and the adsorption capacities. In other words, if the surface charge of the adsorbent is positive, its tendency to attract anions increases and electrostatic attraction will happen. Thus, the pH of the solution affects both the adsorbent surface charge and the charge of arsenic species and this

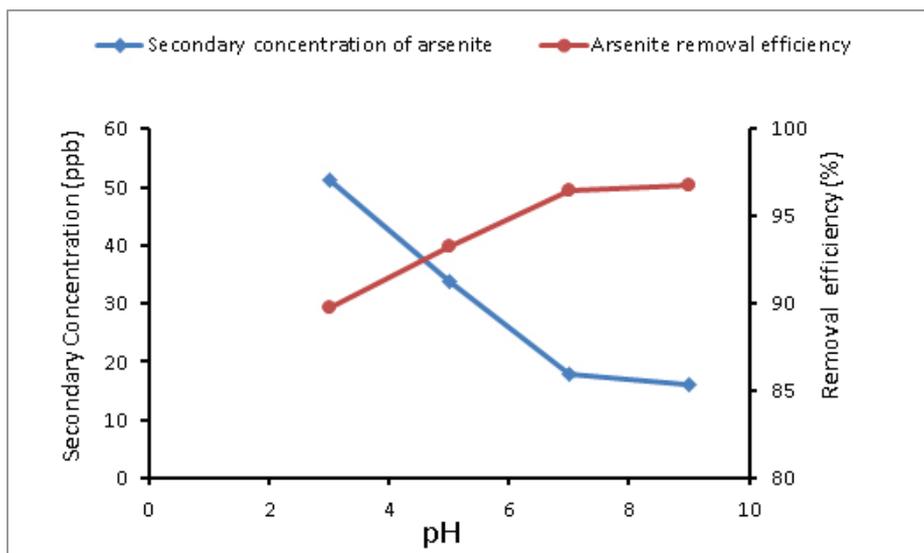


Figure 1: This Figure shows the effect of initial pH on arsenite removal efficiency (initial concentration of arsenite 500ppb, contact time 30 min and welding iron waste dose 10g/L).

condition controls arsenic uptake.^{3,15} According to other researchers, the pentavalent form of arsenic is very active at pH 4 to 9 and is ready to be removed by various methods such as formation of adsorbent complex.^{15,16} At acidic pH, due to the positive ions released from the adsorbent surface, repulsive forces are created between the ions and arsenic; this leads to removal of arsenic and a decrease in efficiency. As pH is increased to 7, the anionic species $H_2AsO_3^-$ increases and thus further adsorption of arsenic to iron particles is expected. The reason for low absorption at low pH is that the only force acting between arsenic species in water and iron particles is the weak vanderwals adsorption force. The maximum adsorption at pH 7 arises from the maximum difference between the energy released from adsorption and that needed for dissociation of the H_3AsO_4 . These results are consistent with those of Deschamps and colleagues' study on

arsenic removal using a natural Fe and Mn enriched sample. The results showed that maximum arsenate and arsenite removal occurred at pH=7.¹⁷ Similarly, in a study conducted by Sperlich and colleagues, the optimum pH for arsenic removal by granular ferric hydroxide was 7-7.6.¹⁸

The Effect of Adsorbent Dosage on the Arsenite Removal Efficiency

Figure 2 shows arsenite removal efficiency at different doses of welding iron waste. At pH=7, contact time of 30 minutes and initial arsenate concentration of 500 µg/l, with increasing doses of iron waste from 2.5 to 10g/l, the removal rate increased from 42.37% to 96.70% and with increasing doses of iron waste to 2, the removal efficiency increased to 98.76%. Figure 3 shows the scanning electron microscope (SEM) image of the

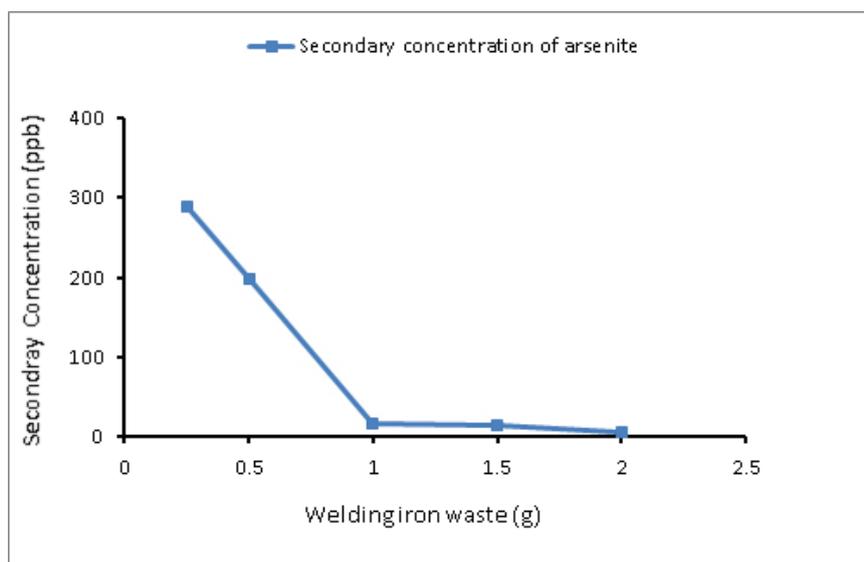


Figure 2: The effect of adsorbent dosage on arsenite removal efficiency (initial concentration of arsenite 500 ppb, contact time 30 min and pH=7).

welding iron waste before and after the adsorption of arsenic. Adsorbent dose is an important parameter affecting the arsenic removal process. As shown in the Figure, arsenite removal follows the dose and increases by an increase in the absorption dose. The reason is that the increase in the amount of iron waste increases the surface active sites of adsorption and the possibility of contact between arsenite and iron waste, leading to higher oxidation and reduction reactions.¹⁹ As a result, better absorption occurs. The test results showed that with the increase in the amount of adsorbent dose from 2.5 to 10g/L, the removal rate maximizes. After that, adding larger quantities of waste iron dose will not have much impact on arsenite removal.

The Effect of Contact Time on the Arsenite Removal Efficiency

The results related to the effect of contact time (5-90 min) at a fixed dose of 10 g/L iron waste and initial arsenate concentration of 500 µg/l are shown in Figure 4. Based on the results presented in this

Figure, with increasing contact time, arsenite removal significantly increased. With increasing contact time from 5 to 30 minutes, the removal rate increased from 9% to 96.62%. Then, with increasing contact time to 90 minutes, the removal rate increased to 99.24%. According to the Figure, it can be concluded that at the beginning of the experiment, increasing the contact time significantly increased arsenite removal. Therefore, removal of arsenite was rapid and it increased rapidly in the first 30 minutes. Then, from 30 to 90 minutes it increased slowly. This is due to more contact between iron waste and arsenite in the first 30 minutes. Therefore, the removal of arsenite increased with the passage of time because of the holes and corrosion on metal surfaces which increased the absorption cross section and the active sites for arsenite adsorption. This way, the products of iron reaction in the aquatic environment and, in turn, the removal efficiency increased.¹⁹ Also, with increasing the contact time, ions in the environment have more time to be absorbed by the iron waste. In other words, with increasing the reaction time, the

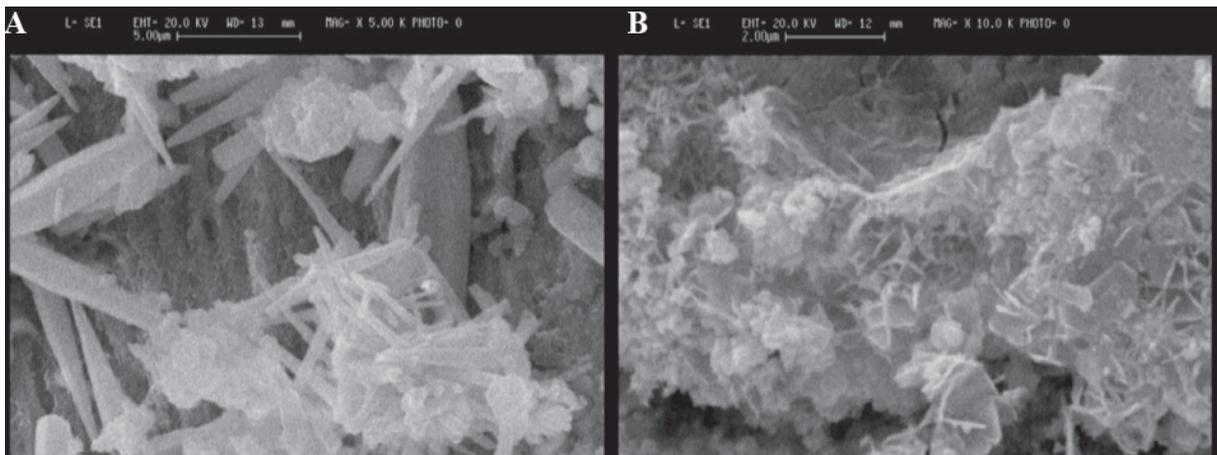


Figure 3: The TEM image of the welding iron waste: a) before the adsorption of arsenic, b) after the adsorption of arsenic.

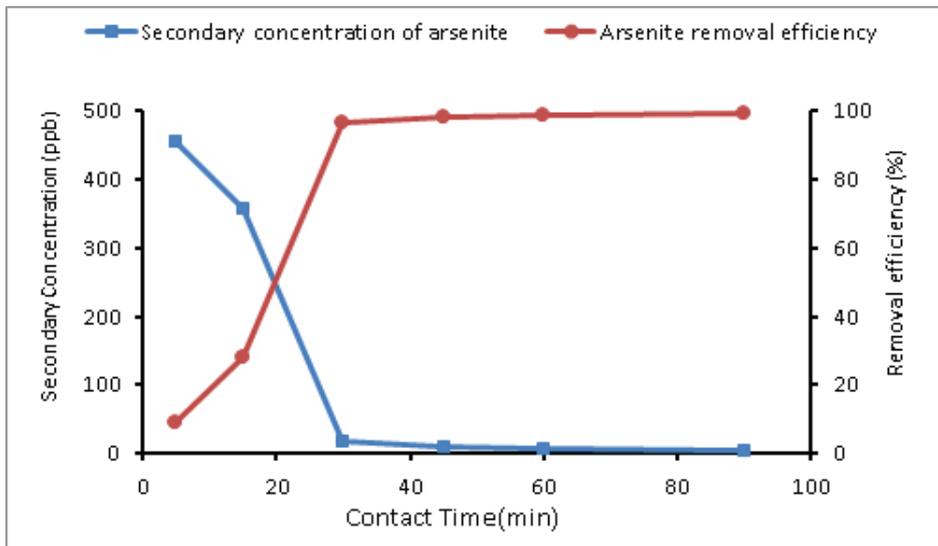


Figure 4: The effect of contact time on the arsenite removal efficiency (initial concentration of arsenite 500 ppb, pH=7 and welding iron waste dose 10 g/L).

contact time between contaminant ions and waste iron increases, and ions have more opportunities to connect to adsorption sites on the particle surfaces. Therefore, in the present study, the highest arsenite removal efficiency occurred in the initial 30 minutes. Ramaswami and colleagues showed in a study that the removal of arsenic by iron in the initial 30 minutes at initial concentration of 2 mg/l was more than 93%.²⁰ Asgari and colleagues conducted a study entitled “the removal efficiency of arsenic from water by Granular Ferric Hydroxide (GFH)”. The results indicated that the best removal time was the initial 30 minutes.²¹ The Langmuir and Freundlich isotherm models were applied to the experimental data. Correlation coefficient of Freundlich and Langmuir isotherms for arsenite was 0.7593 and 0.9979, respectively. The results of the experiments showed that adsorption of arsenite on welding iron waste is compatible with Langmuir model ($R^2=0.9979$). The Langmuir isotherm had the highest correlation to the studied factors. Figure 5 shows the Langmuir adsorption isotherm of arsenite on the welding iron waste. The

maximum adsorption capacity was calculated to be 0.242 mg/g.

The Effect of Initial Concentration of Arsenic on the Removal Efficiency

In Figure 6, the removal efficiencies of arsenate and arsenite at pH=7 and contact time of 45 minutes in different concentrations of arsenate and arsenite (300-3000 µg/l) are shown. As shown in the Figure, with increasing the initial concentrations up to a certain amount, the removal efficiency increased, but further increase reduced the efficiency. Thus, at the initial concentration of 300 micrograms per liter, the removal efficiency of arsenate and arsenite was 92% and 98%, respectively. With increasing the initial concentration to 1500 µg/l, arsenate and arsenite removal increased to 98.92% and 99.66%, respectively. However, with increasing the initial concentration to 3000 µg/l, arsenate and arsenite removal efficiency reduced to 97.17% and 98.59%, respectively. Also, as shown in Figure 7, at pH=7, contact time of 30

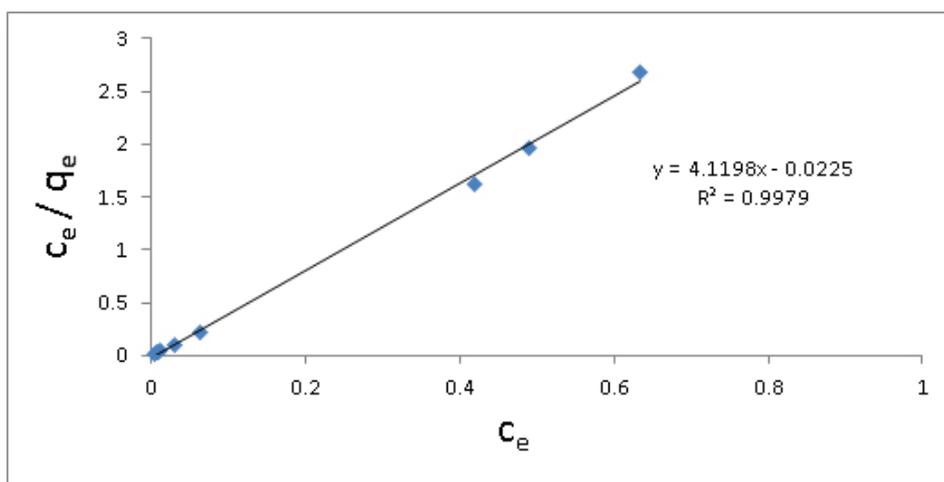


Figure 5: The Langmuir adsorption isotherm of arsenite on the welding iron waste.

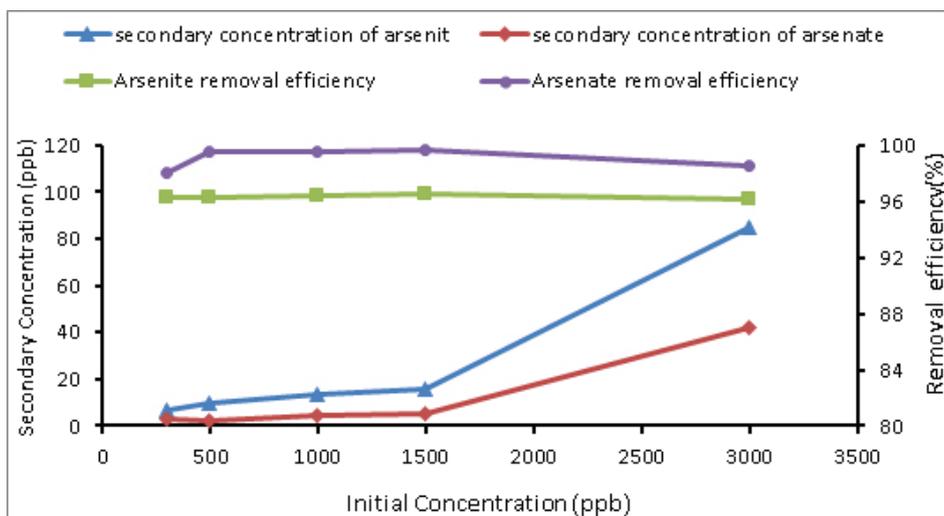


Figure 6: The effect of initial concentration of arsenic on removal efficiency (contact time 45min, pH=7 and welding iron waste dose 10 g/L).

minutes and initial arsenite concentration of 100 in a fixed dose of 10 g/L of iron waste with a mesh less than and greater than 100, the removal of arsenite was respectively 90.09% and 97.80%. These figures increased to 96.54% and 98.87% with increasing the initial concentration to 1500 µg/l. However, with increasing the initial concentration to 3000 µg/l, the removal reduced to 93.24% and 96.67%. The initial concentration of pollutants is one of the important parameters in the removal process. Thus, with increasing the concentrations of arsenate and arsenite at constant amount of iron waste, surface contact increases, leading to faster reaction and removal of arsenate and arsenite. In other words, with increasing concentrations of arsenate and arsenite in the presence of a fixed dose of iron waste, more arsenate and arsenite are removed from the environment, although it is possible that the arsenic concentration in the environment is higher. Investigation of the initial arsenate and arsenite concentrations shows that increased surface area improves the removal rate. Different adsorption behaviors can be due to the fact that at the beginning of adsorption on the adsorbent, there is an equal surface available. For higher concentrations, there will be more adsorption, but then reduction in the available surface reduces the adsorption efficiency for higher concentrations of arsenic. In other words, the removal efficiency increases when the arsenic ions concentration in the solution increases. That is because the increase in concentration of arsenic ions increases the number of contacts between the ions of arsenic and iron waste facilitating the adsorption process.²² Maleki and colleagues investigated Pentavalent arsenic uptake from aqueous solution by wheat straw. The results showed that the amount of arsenic adsorbed increased with increasing initial arsenic concentration.²³ Martinson and Rahmani showed that the initial concentration of arsenic is inversely associated with its removal efficiency.^{6,24} Also according to Figure 6, arsenate removal efficiency was higher than that of arsenite. Another study by Mosaferi and colleagues

on removing arsenic from water using absorption by iron showed that within 2 hours, the removal of As(V) and As(III) was 96% and 94%, respectively.²⁵ These results are consistent with those obtained in the present study. Mosaferi and colleagues investigated the removal of arsenic from drinking water using modified activated alumina. The results showed that the removal of As(III) was negligible compared to that of As(V). For example, at minute 120, the removal of As(V) was 96% and that of As(III) was 16%.²⁵

In this study, the removal of arsenite by iron particles with mesh larger and smaller than 100 was studied. Since the removal of pollutants by iron particles occurs based on surface adsorption or oxidation on the surface of iron particles, the surface area of iron particles is important in pollutant removal efficiency and is positively associated with it.²⁶ Based on the results presented in this study, the smaller the iron particle size, the higher its reactivity with arsenic ion and the higher the removal rate. Thus, the particle size is quite important in adsorption and reaction with contaminants. Smaller particle size increases the surface area and the reactivity of the particles.²⁷⁻³¹ For example, during a period of 30 minutes, at initial concentration of 1500 ppb, with iron particles with mesh of less than and higher than 100, the secondary arsenite concentration was 52.81 and 16.93, respectively. Thus, the results showed that the rate of removal by iron particles with smaller dimensions is much more than large particles. Therefore, the smaller the particles of iron waste, the higher the reaction. The reason is the high specific surface area of the particles and higher activity of their surface centers.²⁷

Conclusion

According to the results of this study, welding iron waste as an affordable and available adsorbent can be a viable option for the removal of arsenic in aquatic environments. It can be produced from discarded metal filings, which is economically important. The

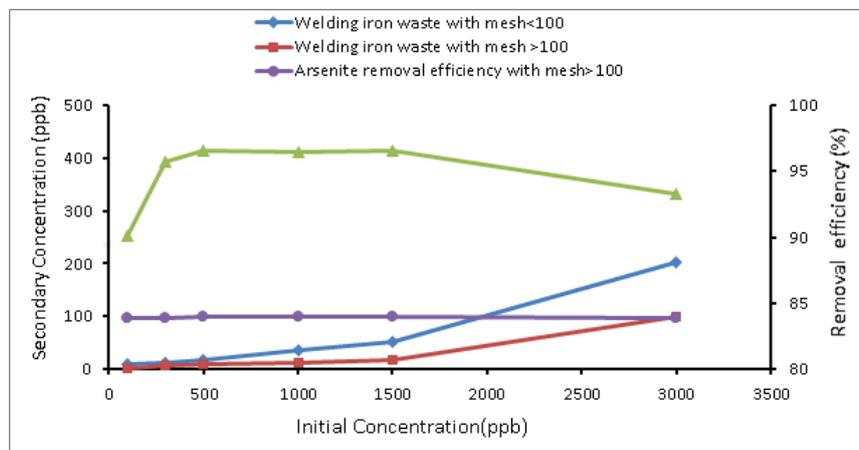


Figure 7: The effect of initial concentration of arsenite on removal efficiency (contact time 30 min, pH=7 and welding iron waste dose 10 g/L).

use of iron waste is easy and lets iron filings stay in the environment. The results of the experiments showed that arsenic removal increased with increasing the contact time and dosage of iron waste. That is because of the increased contaminant's contact with the adsorbent and increased uptake sites. Therefore, the iron waste has a high efficiency for removal of both arsenate and arsenite in the short exposure time of 30 minutes and normal pH of water. Investigating the effect of initial concentration of arsenic shows that at higher concentrations of arsenic, the removal rate increases due to the increased surface area. Therefore, the optimum pH for removal by iron waste is 7 which is close to pH of natural waters. This, in turn, reduces the cost of system administration, because there is no need to adjust pH. This is important since in such a situation there is no need to apply acid and alkaline to increase the decrease in pH values.

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Conflict of Interest: None declared.

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