

Journal of Hazardous Materials 121 (2005) 61-67

*Journal of* Hazardous Materials

www.elsevier.com/locate/jhazmat

# Removal of arsenic from water by zero-valent iron

Sunbaek Bang<sup>1</sup>, George P. Korfiatis, Xiaoguang Meng\*

Center for Environmental Systems, Stevens Institute of Technology, Hoboken, NJ 07030, USA

Received 18 December 2003; received in revised form 4 August 2004; accepted 9 January 2005 Available online 18 March 2005

#### Abstract

Batch and column experiments were conducted to investigate the effect of dissolved oxygen (DO) and pH on arsenic removal with zerovalent iron [Fe(0)]. Arsenic removal was dramatically affected by the DO content and the pH of the solution. Under oxic conditions, arsenate [As(V)] removal by Fe(0) filings was faster than arsenite [As(III)]. Greater than 99.8% of the As(V) was removed whereas 82.6% of the As(III) was removed at pH 6 after 9 h of mixing. When the solution was purged with nitrogen gas to remove DO, less than 10% of the As(III) and As(V) was removed. High DO content and low solution pH also increased the rate of iron corrosion. The removal of arsenic by Fe(0) was attributed to adsorption by iron hydroxides generated from the oxic corrosion of Fe(0). The column results indicated that a filtration system consisting of an iron column and a sand filter could be used for treatment of arsenic in drinking water. © 2005 Elsevier B.V. All rights reserved.

Keywords: Arsenic; Iron; Adsorption; Groundwater; Filtration

# 1. Introduction

Elevated arsenic concentrations are found in groundwater due to anthropogenic activities and natural processes. Anthropogenic activities include mining, use of arsenical pesticides, herbicides and crop desiccants, release of industrial effluents, and disposal of chemical waste [1,2]. The release of arsenic from natural processes can be caused by the reduction of iron hydroxides and the oxidation of pyrite minerals including orpiment (As<sub>2</sub>S<sub>3</sub>) and realgar (As<sub>2</sub>S<sub>2</sub>) [3,4]. Naturally occurring arsenic in drinking water supplies may affect more than 100 million people worldwide, including countries like Bangladesh, India, China, Chile, Argentina, Mexico, Hungry, Taiwan, Vietnam, Japan, New Zealand, Germany, and the United States [5–11].

Arsenic can be concentrated in liver, kidney, lung, and skin tissues by ingestion of arsenic in drinking water [12]. In order to minimize the health risk, the United States Environmental Protection Agency (USEPA) adopted a new maximum contaminant level (MCL) for arsenic of 0.01 mg/L on January 22, 2001 [13]. It is estimated that the new arsenic standard may affect about 3000 community water systems (CWSs), currently serving up to 11 million people in the United States [14]. The new arsenic MCL will become effective by January 2006 [14].

Fe(0) has been used for the treatment of halogenated organic compounds by reductive dehalogenation [15,16] and for the removal of nitrate [17,18]. Recent researches have also shown that Fe(0) has effectively removed inorganic contaminants such as chromate, uranyl, copper, cadmium, aluminum, zinc, nickel [19,20] and metalloids such as selenocyanate [21]. The mechanism of the metal removal includes reductive precipitation, complexes with iron oxides, or electrochemical reduction [19–21].

Numerous technologies have been developed to remove arsenic from water. The USEPA recommends several technologies as the best available technology (BAT) candidates for arsenic removal [22]. These technologies include ion exchange, activated alumina, reverse osmosis, modified coagulation/filtration, modified lime softening, electrodialysis reversal, and oxidation/filtration [22]. Several research groups have investigated removal of arsenic using Fe(0) [23–29].

<sup>\*</sup> Corresponding author. Tel.: +1 201 216 8014; fax: +1 201 216 8303.

E-mail address: xmeng@stevens-tech.edu (X. Meng).

<sup>&</sup>lt;sup>1</sup> Current address: Department of Environmental Science and Engineering, Gwangju Institute of Science and Technology (GIST), Gwangju 500-712, South Korea.

<sup>0304-3894/\$ -</sup> see front matter © 2005 Elsevier B.V. All rights reserved. doi:10.1016/j.jhazmat.2005.01.030

Batch experiments, X-ray absorption spectroscopy (XAS), and X-ray photoelectron spectroscopy have been used for studying the kinetics and chemical reactions between arsenic and Fe(0) [23–26]. As(V) and As(III) were detected in the reacted Fe(0) generated under anoxic conditions. The results indicate that the arsenic species are removed by Fe(0) through adsorption on iron oxides, although thermodynamic considerations predict that As(V) and As(III) can be reduced to As(0) by Fe(0). Reduction of As(V) to As(III) was observed after 30 and 60 days of reaction with Fe(0) [25].

Column filtration experiments have been conducted by a few investigators to evaluate the effectiveness of Fe(0) for arsenic removal [24,27–29]. Melitas et al. [24] reported that the removal rates of arsenic were up to 10 times faster near the inlet end of the iron column than near the effluent end. The faster removal was attributed to rapid oxidation of Fe(0) by small amounts of dissolved oxygen (DO) in the influent. On the other hand, Ramaswami et al. [30] reported that the presence of air in batch reactors hindered arsenic removal by Fe(0). The column filtration experiments reported in the literature were all conducted under anoxic conditions and at relatively long hydraulic contact time. The effect of DO content on arsenic removal by Fe(0) filters has not been investigated systematically.

DO content in groundwater ranges from zero to several mg/L. If As(V) and As(III) removal is due to adsorption by iron oxide, high DO content will increase the rates of iron oxidation and subsequently improve the removal of arsenic by the Fe(0) column. Long hydraulic retention time may simulate in situ treatment of groundwater with permeable Fe(0) barriers. In drinking water treatment, a hydraulic retention time of less than 10 min is usually required due to the high flow rate and space limitations in the treatment facilities. High DO content in influent water may be necessary for efficient removal of arsenic by Fe(0) filters at short hydraulic retention time.

The objectives of this study are to study the effects of DO and pH on arsenic removal using Fe(0), and to determine the efficiency of Fe(0) filings for arsenic removal from water. The knowledge gained from this study will aid in the understanding and development of arsenic removal processes using Fe(0).

## 2. Materials and methods

# 2.1. Materials

All chemicals used in this study were analytical grade and all stock solutions were prepared with deionized water (DI) from a Milli-Q water system. The As(III) and As(V) stock solutions were prepared by dissolving NaAsO<sub>2</sub> (Fisher, Fairlawn, NJ) and As<sub>2</sub>O<sub>5</sub>·3H<sub>2</sub>O (Aldrich, Milwaukee, WI) in DI water, respectively. Fe(0) filings (100 and 40 mesh) and filter sand (0.35–0.45 mm) were obtained from the U.S. Metals Inc. (Mentune, IN) and Ricci Bros Sand Inc. (Port Norris,

Table 1	
Chemical characteristics of the arsenic-contaminated of	proundwater

			8	
Total As	Total SiO <sub>2</sub>	Total Fe	pН	DO (mg/L)
(iiig/L)	(IIIg/L)	(119/12)		(1115/12)
85	560	0.2	12	8

NJ), respectively. The Fe(0) filings contained approximately 95% iron, 1.2% carbon, 1.2% silicon, 1.2% manganese, 0.2% copper, 0.2% chromium, 0.2% nickel, 0.05% phosphorus and 0.05% sulfur. The Brunauer–Emmett–Teller (BET) surface area analysis indicated a surface area of  $0.55 \text{ m}^2/\text{g}$  for 100 mesh and  $0.169 \text{ m}^2/\text{g}$  for 40 mesh Fe(0) filings. Arsenic-contaminated groundwater used in the study was obtained from a superfund site in Tacoma, Washington [31]. Chemical characteristics of the groundwater are listed in Table 1. Arsenic in the groundwater sample exists primarily in the As(V) form. The high DO content in the groundwater samples is due to exposure to air during storage and transportation.

#### 2.2. Batch experiments

In the batch experiments, 1 g/L of Fe(0) filings (100 mesh) were placed in conical beakers containing 200 ml of arsenicspiked aged tap water. The solutions were open to the air or purged with nitrogen gas during continuous mixing with a magnetic stirring bar. For the anoxic systems, the solution was purged with nitrogen gas for 20 min to remove the oxygen before the addition of Fe(0) filings. During the anoxic experiments, the solution was continuously purged with nitrogen gas. The solution pH was controlled to the desired values by adding hydrochloric acid or sodium hydroxide. At different mixing times, 1.5 ml of samples were taken and centrifuged for 10 min to separate the solution from the precipitate for analysis of soluble arsenic. The samples used for analysis of total iron and arsenic concentrations were prepared by adding concentrated nitric acid into 2 ml of suspension samples to dissolve the precipitates.

### 2.3. Column experiments

Column experiments were conducted using acrylic columns packed with Fe(0) filings. The arsenic-spiked tap water or contaminated groundwater was passed through columns upward at various flow rates using a peristaltic pump. Four column filtration tests were conducted. In the first column experiment, the DO content in the influent solution was varied from 0.9 to 5.5 mg/L by purging nitrogen gas into influent water to evaluate the effects of DO on arsenic removal by Fe(0). The influent As(V) concentration was 90  $\mu$ g/L and 150 g of iron filings (100 mesh) was used in a column of 2.5 cm diameter and 17.8 cm length. The flow rate was 30 ml/min. In the second column experiment, the pH of the influent solution was adjusted to between 4 and 5 with hydrochloric acid to check the effect of pH on the removal of arsenic using Fe(0). Arsenic-contaminated groundwater

was used in this column test. An influent solution containing 85 mg/L of As(V) and 600 g of iron filings (100 mesh) was used in a column (5.1 cm in diameter and 17.8 cm in height). The flow rate was set to 200 ml/day. In the third column experiment, the spiked tap water contained 100 mg/L of As(V) and 50 mg/L of As(III). The flow rate was 700 ml/day and 400 g of Fe(0) filings (100 mesh) was used in a column. In the fourth column experiment, an iron column with an inside diameter of 2.5 cm was conjugated with a sand filter that had a diameter of 7.6 cm. The sand column was packed with 61 cm of filter sand with a grain size of 0.35–0.45 mm. Arsenic-spiked tap water was pumped through the iron column in an upward flow and then through the sand filter in a downward flow. The flow rate in the iron column was maintained at  $0.44 \text{ m}^3/\text{m}^2 \text{min} (10.8 \text{ gpm/ft}^2)$ . The influent and effluent solution pHs were measured during all column experiments. Effluent solutions after the iron column and the sand filter were collected and acidified with nitric acid for analysis of total arsenic and iron.

### 2.4. Analysis

The arsenic concentration in the solution was determined with a graphite furnace atomic absorption spectrometer (GFAAS, Varian Zeeman SpectraAA-400 and Varian Zeeman 220Z). Iron analysis was performed using a GFAAS and inductively coupled plasma (ICP, Varian Liberty 200 and Varian VISTA-MPX). In all experiments, pH was measured with an Orion pH meter (290A and 410A) and the pH meter was calibrated with three buffers (pH 4.0, 7.0, and 10.0) daily.

# 3. Results and discussion

# 3.1. Effects of DO and pH on arsenic removal by Fe(0) in batch experiments

The effect of DO on the removal of arsenic was evaluated by comparing the experimental results obtained under oxic and anoxic conditions (Fig. 1). During the reaction, the solution pH was controlled at 6. Greater than 82.6% of the As(III) was removed after 9 h of mixing when the solution was open to the air, while less than 4% of the As(III) was removed under anoxic conditions during 9 h of reaction. Similar trends were observed in As(V) solutions. Greater than 99.8% of the As(V) was removed under oxic conditions after 9 h of reaction. On the other hand, less than 9% of the As(V) was removed when the solution was purged with nitrogen gas. The results in Fig. 1 also indicate that As(V) removal by Fe(0) is faster than As(III) removal under oxic conditions.

The ineffective removal of arsenic under anoxic conditions was due to the lack of ferric hydroxide formation and slow kinetics of electrochemical reduction of As(V) and As(III)to As(0) by Fe(0). The small amount of As(V) and As(III)removed under anoxic conditions (Fig. 1) was attributed to adsorption on the ferric hydroxide layer on the iron filings.

Fig. 1. Effect of dissolved oxygen on arsenic removal by Fe(0). As(III) and As(V) = 100 mg/L; Fe(0) content = 1 g/L (100 mesh); pH 6.

Because the industrial-grade nitrogen gas used to purge the solution contained low content of oxygen, small amount of ferric hydroxide could be formed on the surface of the iron filings during the anoxic experiments.

Fe(0) can be oxidized to Fe(II) and Fe(III) when it is exposed to oxygen or water. When the solution is purged with nitrogen gas to exclude oxygen, only Eq. (1) occurs in the solution. Since only soluble Fe(II) was generated gradually, no precipitates were observed in the solution purged with nitrogen gas during 9h of mixing. Under anoxic conditions, iron oxide such as magnetite (Fe<sub>3</sub>O<sub>4</sub>) can be formed slowly on the Fe(0) particle surface [26]. The reduction of As(III) and As(V) to As(0) by Fe(0) is thermodynamically favorable [24]. However, Farrell et al. [23] and Manning et al. [26] reported no measurable reduction of As(V) to As(III). Su and Puls [25] observed the reduction of As(V) to As(III) on the Fe(0) surface after 30 days. The investigators did not observe the reduction of As(V) to As(0).

$$Fe^{0} + 2H^{+} = Fe^{2+} + H_{2}$$
(1)

In the presence of oxygen, large amounts of ferric hydroxide precipitate were formed rapidly from Fe(0) filings as described by Eqs. (2), (3), and (4). As(V) and As(III) were mostly removed by the iron hydroxide precipitate through adsorption. Because iron hydroxide has a higher adsorption capacity for As(V) than for As(III) at neutral pH, As(V) removal was greater than As(III) [32].

$$2Fe^{0} + O_{2} + 4H^{+} = 2Fe^{2+} + 2H_{2}O$$
 (2)

$$4Fe^{2+} + 4H^+ + O_2 = 4Fe^{3+} + 2H_2O$$
(3)

$$Fe^{3+} + 3H_2O = Fe(OH)_3 + 3H^+$$
 (4)

The effect of pH on As(V) removal from arsenic solution using Fe(0) is illustrated in Fig. 2. The test was conducted by mixing 0.2 g of Fe(0) filings (100 mesh) with 200 ml of solution containing 100 mg/L of As(V). The pH of the solutions was controlled at 6, 7, and 8 during the mixing with





Fig. 2. Removal of As(V) by Fe(0) at various pH. As(V) = 100 mg/L; Fe(0) content = 1g/L (100 mesh); mixing in ambient air.

Fe(0) in ambient air. The removal of As(V) occurred rapidly at pH 6. Greater than 99.8% of the As(V) was removed after 9 h of reaction. However, only 55.5 and 2% of the As(V) was removed from the solution during 9 h of mixing when the pH of the solutions was maintained at 7 and 8, respectively.

The corrosion rate of Fe(0) filings in the As(V) solution at various pHs is presented in Fig. 3. Fe(0) corrosion was much faster at pH 6 than at pH 7 and 8. Specifically, the total iron concentration in the suspension increased to approximately 113 mg/L at pH 6 after 9 h of mixing. Less than 1.5 mg/L of total iron was detected in the As(V) solution when the solution pH was 7 and 8. The total iron was mainly in iron hydroxide precipitate form because the Fe(0) filings were not suspended in the solution. The formation of iron hydroxides from Fe(0) filings decreased dramatically when the solution pH increased from 6 to 8.

As shown in Figs. 2 and 3, As(V) removal is directly proportional to the amounts of ferric hydroxide precipitate formed. The extent of As(V) removal decreased from 99.8



Fig. 3. Concentration of iron generated from Fe(0) filings during reaction at various pHs. As(V) = 100 mg/L; Fe(0) content = 1 g/L (100 mesh); mixing in ambient air.

Table 2	
Pseudo-first-order rate constant ( <i>k</i> ) and half-life $(t_{1/2})$	

Condition	pН	As species	$k ({\rm h}^{-1})$	$t_{1/2}$ (h)	$R^2$
Oxic		As(III)	1.75E - 01	3.96	0.964
	6	As(V)	3.24E - 01	2.14	0.945
	7	As(V)	7.95E - 02	8.72	0.980
	8	As(V)	NA	NA	NA
Anoxic		As(III)	NA	NA	NA
	6	As(V)	1.78E - 02	38.93	0.830

to 2% when the total iron concentration in the suspension decreased from 113 to less than 0.2 mg/L. In addition to increased Fe(0) corrosion at low pH, the water pH directly influences the adsorption of arsenic by iron hydroxides and oxides. It is well known that the extent of As(V) adsorption decreases when the pH increases from about 5 to 10 [32].

The data in Figs. 1 and 2 can be described by a pseudofirst-order reaction (d[As]/dt = -k[As]) with  $R^2$  values ranging from 0.83 to 0.98. The pseudo-first-order reaction constants (*k*) and the calculated half-lives ( $t_{1/2}$ ) are summarized in Table 2 for arsenic removal by Fe(0) under various conditions. The rate constants could not be determined for As(III) removal at pH 6 under anoxic conditions and for As(V) removal at pH 8 under oxic conditions because less than 3% arsenic removal occurred. At pH 6 and under oxic conditions, the half-lives for As(V) and As(III) removal were 2.14 and 3.96 h, respectively. Under anoxic conditions, the half-life for As(V) removal increased to 38.93 h. The half-life for As(V) removal increased from 2.14 to 8.72 h when the pH increased from 6 to 7.

Su and Puls [25] have reported the kinetics results for arsenic removal using four types of Fe(0). The half-lives for As(V) removal using Fe(0) products from Fisher, Peerless, Master Builders, and Aldrich Fe(0) were  $8.98 \pm 1.12$ ,  $19.9 \pm 1.1, 28.3 \pm 0.3$ , and  $131 \pm 9$  h, respectively. They concluded that the surface area of Fe(0) was not related to the kinetics of arsenic removal by Fe(0) because Fisher Fe(0) had the least surface area but it had the greatest removal rate. The rates of arsenic removal are affected by many factors such as DO, pH, types of Fe(0) products, chemical composition of the water, and intensity of mixing.

# 3.2. Effects of DO and pH on arsenic removal by Fe(0) columns

A column experiment was conducted to investigate the arsenic removal at different DO contents. The influent As(V) concentration was 90  $\mu$ g/L and a relatively short empty bed contact time [i.e., EBCT (Fe bed volume/flow rate) = 1.6 min] was used to simulate filtration treatment of water containing low level of arsenic. Effluent concentrations of arsenic are shown in Fig. 4 as a function of filtration time. The effects of DO were investigated by changing the DO concentration of the influent solution from 0.9 to 5.5 mg/L during the column experiment. When the influent DO was 0.9 mg/L, the total effluent arsenic, which included soluble and particulate arsenic



Fig. 4. Effect of dissolved oxygen content on arsenic removal by iron column. As(V) =  $90 \mu g/L$ ; Fe(0) content = 150 g (100 mesh); column diameter = 2.5 cm; iron bed height = 11.6 cm; flow rate = 30 ml/min (1.45 gpm/ft<sup>2</sup>).

forms, increased approximately from 4 to  $62 \mu g/L$  within 24 h. The concentration of the effluent arsenic decreased with increasing DO content in the influent solution. Arsenic was reduced to about  $15 \mu g/L$  when the influent DO increased to 4.3–5.5 mg/L. The arsenic concentration increased again when the influent DO was reduced to 0.9 mg/L. The dramatic effect of DO on the removal of arsenic in the column test was consistent with that observed in the batch experiments.

Lackovic et al. [27] observed effective removal of arsenic from an anoxic landfill leachate containing  $300 \mu g/L$ of As(III). The main difference between their filtration conditions and those used in Fig. 4 was the hydraulic contact time of the water in the Fe(0) columns. A very long hydraulic contact time of several hours was used in their filtration test. Due to the slow formation rate of iron oxides at low DO content, a long contact time is required to achieve sufficient removal of arsenic.

Another column experiment was conducted to investigate the effects of pH on the arsenic removal from contaminated groundwater containing 85 mg/L of As(V) (Fig. 5). The influent solution pH was adjusted to 5 and a long EBCT (i.e., 23 h) was used in the filtration test. The long hydraulic contact time simulated an in situ treatment of contaminated groundwater with a permeable iron barrier. The effluent arsenic concentration and pH decreased gradually in the first 31 days of the test. The effluent pH and arsenic concentration achieved a relatively steady state between the 13th and the 34th day of filtration. The effluent pH was between 7.1 and 7.7, and the effluent total arsenic concentration was about 3.4 mg/L, representing approximately 96% removal of As(V). After 34 days of filtration, the influent pH was reduced to 4 and the effluent arsenic was reduced to approximately 0.1 mg/L. Meanwhile, the effluent pH decreased to about 6 and the effluent total iron concentration increased from about 0.03 to 15 mg/L.

Decreasing the pH could increase the corrosion rate of iron and also reduce the adverse effect of coexisting silicate  $(SiO_2 = 560 \text{ mg/L})$  in the contaminated water on As(V) ad-



Fig. 5. Effect of pH on arsenic removal from contaminated groundwater by iron column. As(V) = 85 mg/L; Fe(0) content = 600 g (100 mesh); column diameter = 5.1 cm; iron bed height = 11.3 cm; flow rate = 200 ml/day.

sorption by iron hydroxide. Su and Puls [33] have studied the effect of silicate on arsenic removal by Fe(0) filings. When the solution pH was 10.12 in the presence of 6 mg/L of SiO<sub>2</sub>, less than 0.2 mg/L of arsenic was removed from 2 mg/L of arsenic solution [33]. Competitive adsorption of silicate significantly reduces As(V) adsorption by iron hydroxides [32]. The adverse effect of silicate on As(V) removal decreases with decreasing pH.

The third column test was conducted continuously for 200 days to determine the capacity of the Fe(0) filings for arsenic removal. The influent solution contained 100 mg/L of As(V) and 50 mg/L of As(III) and influent and effluent pH were approximately 7 and 8, respectively. The EBCT in the iron column containing 400 g of Fe(0) filings was 4.46 h. The results in Fig. 6 show that arsenic was reduced from 150 mg/L in the influent to less than about 20 mg/L in the first 100 days of the filtration test. The effluent arsenic concentration started to increase after 150 days of continuous filtration. It increased to about 120 mg/L after 190 days of the test.



Fig. 6. Removal of arsenic by iron column. Total As = 150 mg/L (100 mg/L of As(V) + 50 mg/L of As(III)); Fe(0) content = 400 g (100 mesh); column diameter = 3.8 cm; iron bed height = 13.6 cm; flow rate = 700 ml/day.

The increasing arsenic concentration coincided with the decrease in the effluent iron concentration from about 20-0.3 mg/L. The slow release of iron indicated that the Fe(0) filings became much less reactive possibly due to the formation of a thick iron oxide layer on the iron particle surface.

During 200 days of filtration, approximately 18,000 mg of arsenic was removed by 400 g of Fe(0) filings (i.e., 45 g As/kg Fe) based on mass balance calculations. Two layers were formed in the iron bed due to an upward flow used in the column test. The top layer contained fine particles with a gray color, and the lower layer contained black and coarse iron particles. Total digestion analysis of the spent solids determined that the arsenic content in the top layer was 175 g/kg of dry solid. The solids in the coarse iron had a much lower arsenic content (i.e., 12 g/kg). These results indicated that more arsenic was associated with fine iron particles than coarse iron particles, which could be attributed to the high surface area and high iron oxide content of the fine particles.

# 3.3. Treatment of As(V) by Fe(0) and sand filtration in series

If Fe(0) is used for the treatment of drinking water, the treated water has to meet the MCL for arsenic and the secondary drinking water standard for iron (i.e., 0.3 mg/L). The process has to be able to remove arsenic at short EBCT. The EBCT of adsorptive filters used for drinking water treatment is usually a few minutes. In this experiment, a sand column was connected to an iron column containing 40 mesh iron filings for the removal of iron hydroxide precipitates and adsorbed arsenic. Spiked tap water containing 100 µg/L As(V) and approximately 6 mg/L of DO was pumped through the iron column at an EBCT of about 1 min and then through the sand column. Naturally occurring arsenic in groundwater is usually less than 100 µg/L in the United States.

The arsenic concentration in the effluent of the iron column fluctuated between 20 and  $60 \mu g/L$  (Fig. 7). Approximately 50% of the As(V) was removed by the iron column. The sand column further reduced the arsenic concentration to less than 5  $\mu g/L$ . No breakthrough occurred during the treatment of nearly 34,000 bed volumes of water. Activated alumina (AA) usually treats less than 10,000 bed volumes of arsenic contaminated groundwater [34]. Driehaus et al. [35] reported that approximately 35,000 bed volumes of water containing 16–17  $\mu g/L$  of As(V) were filtered by granular ferric hydroxide (GFH) before the effluent arsenic increased to 10  $\mu g/L$ . Fe(0) filings, AA, and GFH costs were about \$0.20/lb, \$0.41/lb, and \$4.60/lb, respectively.

The total iron concentration in the effluent of the iron column was between 3 and 6 mg/L (Fig. 8). The sand column reduced the total iron concentration to less than 0.3 mg/L. In the presence of DO Fe(0) was oxidized to form ferric hydroxide in the iron column. Approximately 50% of the total arsenic in the influent passed through the iron column and was removed by the sand filter because the arsenic was adsorbed on the surface of the ferric hydroxide precipitate. The results indicate



Fig. 7. Effluent total arsenic from iron column and sand filter as a function of bed volume. As(V)=100  $\mu$ g/L; Fe(0) content=1000 g (40 mesh); iron column diameter=2.5 cm; iron bed height=46.3 cm; sand column diameter=7.6 cm; sand bed height=61 cm; flow rate=220 ml/min (10.82 gpm/ft<sup>2</sup>).



Fig. 8. Effluent total iron from iron column and sand filter as functions of bed volume. As(V) =  $100 \mu g/L$ ; Fe(0) content = 1000 g (40 mesh); iron column diameter = 2.5 cm; iron bed height = 46.3 cm; sand column diameter = 7.6 cm; sand bed height = 61 cm; flow rate = 220 ml/min (10.82 gpm/ft<sup>2</sup>).

that a cost-effective filtration process can be developed using Fe(0) filings to remove arsenic from water. Aeration or/and pH adjustment pretreatments may be required for treatment of groundwater with low DO content and high pH.

## 4. Conclusions

Batch and column experimental results demonstrated that arsenic was effectively removed from arsenic-spiked solutions and arsenic-contaminated groundwater using Fe(0) filings. The presence of DO and low pH increased the rate of iron corrosion and arsenic removal in both batch and column experiments. The removal of arsenic was mainly attributed to adsorption on iron hydroxides produced through the oxidation of Fe(0) by dissolved oxygen. A filtration system consisting of an Fe(0) filter and a sand filter can be used to remove arsenic and iron from contaminated water at short EBCT in the presence of DO and in a near neutral pH range.

### Acknowledgements

The authors would like to thank the Hazardous Substance Management Research Center for the support of this research. We would also like to thank Maria Pena for assistance with the BET analysis.

#### References

- S. Chakravarty, V. Dureja, G. Bhattacharyya, S. Maity, S. Bhattacharjee, Water Res. 36 (2002) 625.
- [2] P.L. Smedley, D.G. Kinniburgh, Appl. Geochem. 17 (2002) 517.
- [3] X.G. Meng, G.P. Korfiatis, C. Jing, C. Christodoulatos, Environ. Sci. Technol. 35 (2001) 3476.
- [4] R.T. Nickson, J.M. McArthur, P. Ravenscroft, W.G. Burgess, K.M. Ahmed, Appl. Geochem. 15 (2000) 403.
- [5] X.G. Meng, G.P. Korfiatis, C. Christodoulatos, S. Bang, Water Res. 35 (2001) 2805.
- [6] S.P. Pande, L.S. Deshpande, P.M. Patni, S.L. Lutade, J. Envrion. Sci. Health A32 (1997) 1981.
- [7] P.L. Smedley, D.G. Kinniburgh, Appl. Geochem. 17 (2002) 517.
- [8] H. Kondo, Y. Ishiguro, K. Ohno, M. Nagase, M. Toba, M. Takagi, Water Res. 33 (1999) 1967.
- [9] S.J. Mclaren, N.D. Kim, Environ. Pollut. 90 (1995) 67.
- [10] M.R. Jekel, Water Supply 14 (3/4) (1996) 50.
- [11] A.H. Welch, D.B. Westjohn, D.R. Helsel, R.B. Wanty, Ground Water 38 (2000) 589.
- [12] M.M. Wu, T.L. Kuo, Y.H. Hwang, C.J. Chen, Am. J. Epidemiol. 130 (1989) 1123.
- [13] The U.S. Environmental Protection, Federal Register 66 (2001) 6975.

- [14] The U.S. Environmental Protection Agency, Technologies and Costs for Removal of Arsenic from Drinking Water, EPA 815-R-00-028, USEPA, Washington, DC, 2000.
- [15] B.R. Helland, P.J.J. Alvarez, J.L. Schnoor, J. Hazard. Mater. 41 (1995) 205.
- [16] J. Chen, S.R. Al-Abed, J.A. Ryan, Z. Li, J. Hazard. Mater. 83 (2001) 243.
- [17] C. Huang, H. Wang, P. Chiu, Water Res. 32 (1998) 2257.
- [18] S. Choe, Y. Chang, Y. Hwang, J. Khim, Chemosphere 41 (2000) 1307.
- [19] T.E. Shokes, G. Möller, Environ. Sci. Technol. 33 (1999) 282.
- [20] K.J. Cantrell, D.L. Kaplan, T.W. Wietsma, J. Hazard. Mater. 42 (1995) 201.
- [21] X.G. Meng, S. Bang, G.P. Korfiatis, Water Res. 36 (2002) 3867.
- [22] The U.S. Environmental Protection Agency, Implementation Guidance for the Arsenic Rule, EPA 816-D-02-005, USEPA, Washington, DC, 2002.
- [23] J. Farrell, J. Wang, P. O'Day, M. Conklin, Environ. Sci. Technol. 35 (2001) 2026.
- [24] N. Melitas, J. Wang, M. Conklin, P. O'Day, J. Farrell, Environ. Sci. Technol. 36 (2002) 2074.
- [25] C. Su, R.W. Puls, Environ. Sci. Technol. 35 (2001) 1487.
- [26] B.A. Manning, M.L. Hunt, C. Amrhein, J.A. Yarmoff, Environ. Sci. Technol. 36 (2002) 5455.
- [27] J.A. Lackovic, N.P. Nikolaidis, G.M. Dobbs, Environ. Eng. Sci. 17 (2000) 29.
- [28] N.P. Nikolaidis, G.M. Dobbs, J.A. Lackovic, Water Res. 37 (2003) 1417.
- [29] C. Su, R.W. Puls, Environ. Sci. Technol. 37 (2003) 2582.
- [30] A. Ramaswami, S. Tawachsupa, M. Isleyen, Water Res. 35 (2001) 4474.
- [31] P.E. Mariner, F.J. Holzmer, R.E. Jackson, H.W. Meinardus, Environ. Sci. Technol. 30 (1996) 1645.
- [32] X.G. Meng, S. Bang, G.P. Korfiatis, Water Res. 34 (2000) 1255.
- [33] C. Su, R.W. Puls, Environ. Sci. Technol. 35 (2001) 4562.
- [34] The U.S. Environmental Protection Agency, Arsenic Removal from Drinking Water by Ion Exchange and Activated Alumina Plants, EPA 600-R-00-088, USEPA, Cincinnati, OH, 2000.
- [35] W. Driehaus, M. Jekel, U. Hildebrand, J Water SRT-Aqua. 47 (1998) 30.