Comparison of Arsenic Adsorption by Electrocoagulation and Chemically Generated Hydrous Ferric Oxide (HFO) from Water

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Abstract

In the present study, adsorption of arsenic on hydrous ferric oxide (HFO) generated by chemically and electrochemically (EHFO) was examined and compared for removal of both As (III) and As (V) over a pH range from 4 to 8 with initial arsenic concentration of 0.5 mg/l. The results show maximum adsorption on HFO generated electrochemically than HFO generated chemically ($FeCl_3$). In laboratory HFO was generated by adding $FeCl_3$ to NaOH and EHFO was generated by using iron electrodes dipped into the water with applying current in electrochemical reactor. The arsenic removal and residual iron in treated water has been studied and found that arsenic removal was > 99 % for EHFO for both As(III) and As(V) as compared to HFO, 85% for As(III) and 99 % for As(V). Experiments were carried out with initial arsenic concentration of 0.5 mg/L.

Keywords: Electrochemically hydrous ferric oxide, adsorption; arsenite; arsenate; hydrous ferric oxide.

INTRODUCTION

High concentrations of arsenic are found in groundwater in many regions around the world. Nine districts in West Bengal, India and 42 districts in Bangladesh have arsenic levels in groundwater above the World Health Organization maximum permissible limit of 50 µg /L. Over 100 million people in this region is affected by arsenic contamination in this region alone. In Bangladesh and India, drinking water drawn from underground sources has been responsible for widespread arsenic poisoning [1]. In the United States about 11 million people are affected by the new arsenic drinking water standard getting water from around 3000 community water supply systems. Approximately 42 million people in the US obtain water from their private wells [2].

Arsenic contamination of the ground water occurs by both natural processes - such as weathering of arsenic containing minerals and anthropogenic activities like uncontrolled industrial discharge from mining and metallurgical industries, and application of organo-arsenical pesticides [3]. Inorganic arsenic is predominantly present in natural waters. Arsenate [As (V)] and arsenite [As (III)] are primary forms of arsenic in soils and natural waters [4]. As (III) is more mobile in groundwater and 25 to 60 times more toxic than As (V).

High concentration of arsenic in water has caused symptoms of chronic arsenic poisoning in local populations of many countries like India, Bangladesh, Taiwan, Mongolia, China, Japan, Poland, Hungary, Belgium, Chile, Argentina, Austria and North Mexico [5] [6]. Manifestation of higher doses of inorganic arsenic compounds in the human body leads to the disease called arsenocosis. Arsenic is a carcinogen and its ingestion may deleteriously affect the gastrointestinal tract, cardiovascular system, central nervous system and diseases like skin lesions, hyperkeratosis, and hyper pigmentation [6] [7]. Due to its toxic effects on human health, recently the USEPA has lowered the maximum contaminant level (MCL) for arsenic in drinking water from 50 to 10 µg /L.

Many researchers have put forward their efforts to alleviate the problem of arsenic in water body. A number of treatment options are available with demonstrated efficiency for arsenic removal at least to the level of the current MCL. Such treatment technologies include coagulation [8] [9] [10], adsorption on alumina [11], and reverse osmosis (RO) [10].

Some recent treatment technologies based on oxidation and adsorption are iron oxide coated sand [12], manganese dioxide coated sand [13], ferruginous manganese ore [14], ferrihydrite[15], clay minerals [16], electrocoagulation [17] [18], soil biotechnology [19] and zero-valent iron [20]. From the above literature, it is clear that As (III) removal is favored by oxidation to As (V) followed by adsorption on to adsorbent. It is necessary to investigate the treatment approaches that would consistently provide drinking water with arsenic less than 10 μg/L levels.

Hydrous ferric oxide (HFO) is an important sorbent in both natural and engineered aquatic systems. Table 1 shows literature review on arsenic removal by HFO. [8] examined the arsenic adsorption onto HFO. Ford et al., [21] examined influence of aging on the stability of arsenate co-precipitated with HFO and assessed the transformation rate of HFO to more stable products. Dixit and Hearing [22] studied the comparison of As(III) and As(V) sorption onto iron oxide minerals and HFO and found higher adsorption for As(V) onto goethite than HFO, whereas As(III) binding is similar for HFO and iron minerals.

Davis et al., [23] studied the effect of sorbed silica on arsenate, humic substance removal, and particle stability and found arsenate removal decreased at higher pH and silica concentrations. Nemade et al., [24] examined the effect of silicate and phosphate on As(III) and As(V) removal.

EHFO was generated by electro coagulation with iron electrodes which offers possibility of anodic oxidation and in-situ generation of adsorbents (such as hydrous ferric oxides (HFO), hydroxides of aluminum). During electro coagulation As(III) might be oxidized to As(V) and gets adsorbed on to the metal hydroxides [Fe(OH)$_3$] generated. Therefore, it is expected that electro coagulation would be a better choice for arsenic removal from water. According to Faraday's law, dissolution of electrode is related to the total charge passed. The amount of adsorbent produced in the electrochemical reactor would be proportional to the charge density (total charge passed through the solution) and might be serving as design parameter for EC process.

In electro coagulation, iron dissolves from the anode and hydrogen gas is produced at cathode. Faraday's law can be used to describe the current density (A/ cm^2) and the amount of iron which goes into the solution (g Fe/ cm^2)

$$w = \frac{itM}{ZF}$$

[1]

Where, w = metal dissolving (g /Fe cm^{-2}); i = current density (A cm^{-2}); t = time (sec); M = molecular weight of Fe (55.84); Z= number of electrons involved in the oxidation/reduction reaction (Z=3); F= Faraday's constant, 96500.

Table 1 Previous study on arsenic removal by hydrous ferric oxide (HFO)

Description	Type of arsenic	Procedure for HFO	Removal %	Reference
Amorphous ferric oxide	As(III) & As (V)	Amorphous iron hydroxide was prepared by the addition of 25 ml of 1 M NaOH to 1.665 g of analytical grade Fe$_2$(SO$_4$)$_3$ dissolved in	92 %; 100	[25]

		200 ml of doubly distilled water.		
Hydrous iron oxide (HIO)	As(V)	HIO was prepared by the addition of 0.1 N ferric nitrate stock solution to Milli-Q water at 25 C under a nitrogen atmosphere. 0.1 N $NaNO_3$, solution was used as background electrolyte. HIO was precipitated by dropwise addition of 0.1 N NaOH solution until a pH of about 8.0 was attained.	100 % at pH 4-8	[26]
Hydrous ferric oxide (HFO)	As(III) As(V)	A solution of 0.5 M NaOH was added dropwise with stirring to 150ml of 0.05 M $Fe(NO_3)_3$ in a 300ml, water-jacketed beaker at 25°C. The total volume of 0.5 M NaOH added was 45 ml; the final pH was approximately 8.	< 80 % 100 %	[8]
Hydrous ferric oxide (HFO)	As(V)	0.6 M $NaHCO_3$ in deionized (DI) water: an anoxic solution with an equilibrated concentration of 1.3 or 3.6 mM ferrous iron added as $FeSO_4 7H_2 O$	>99 %	[27]
Crystalline ferric hydroxide	As(V)	Co-precipitates were generated by addition of 1M NaOH to acidified solutions of ferric iron	> 99.4 %	[21]
Hydrous ferric oxide (HFO)	As(III)	Hydrous iron oxide was prepared by electrochemical peroxide (ECP) with direct electric current applied to steel electrode and small addition of H_2O_2	>98%	[28]
Amorphous iron oxide	As(III) As(V)	500 ml of 0.5 M Fe $(NO_3)_3$. 9 H_2O, 60 gm of NaOH pellets were added slowly. The solution was continuously bubbled with nitrogen; centrifuged and washed 3 times with water and dried at 25^0C for 24 hour.	>99 % 100 %	[29]

The objective of the present study was to investigate the As (III) and As (V) adsorption on EHFO and to compare with HFO.

MATERIALS AND METHODS

Reagent preparation

Experiments were performed at ambient temperatures ranging from 26-28^0 C. Stock solutions of arsenate were prepared by dissolving appropriate quantity of arsenic trioxide, As_2O_3, (S.D. Fine Chem Ltd., India) in distilled water containing 1% (w/w) NaOH and the solution was then diluted up to 1 liter with distilled water before use. The arsenate stock solution was prepared from sodium arsenate, $Na_2HAsO_4.7H_2O$ (Loba Chemie, India). The working solutions containing arsenic were prepared by dissolving appropriate amount of arsenic from stock solutions in tap water. Analysis of tap water showed pH ranging from 7.1 to 7.6, bicarbonate alkalinity approximately 40-50 mg/L as $CaCO_3$, and dissolved iron, phosphate and arsenic concentration were below detectable limit.

Experimental plan

Batch experiments were conducted with two iron electrodes placed 0.5 cm apart in a 1.5 L beaker and 1 L of arsenic containing water was added. The total submerged surface area of each electrode was 100 cm^2. Before each experiment, the electrodes were abraded with sand paper to remove scale and

then cleaned with water and 1N H_2SO_4. A direct current by stabilized power supply (0-15 V, 2A) was applied to the terminal electrodes in which electrical current was controlled by a variable transformer. Since most of the arsenic contaminated ground waters have the pH in the range of 6 to 8, the experiments were conducted in this pH range only. The pH of the solution was adjusted by adding either dilute HCl or NaOH. The summary of experimental conditions employed for EHFO is presented in Table 2. Chemical coagulation experiments were for arsenic removal by using ferric chloride ($FeCl_3$) as a coagulant with jar test apparatus as given in the literature [8]. The summary of experimental conditions employed for HFO is presented in Tables 3.

Table 2 Experimental conditions employed in electrochemical hydrous ferric oxide (EHFO)

Experiment Run No.	Arsenic Type	As conc. (mg/L)	Current (mA)	Current Density (mA cm^{-2})	pH	Reactor Volume (mL)
1	As(III)	2	30	0.30	7	1000
2	As(III)	2	30	0.30	8	1000
3	As(V)	2	30	0.30	8	1000
4	As(III)	2	30	0.30	6	1000

Table 3 Experimental conditions employed in chemically generated hydrous ferric oxide (HFO)

Experiment Run No.	Arsenic type	HFO as Fe (mg/L)	pH
5	As(III)	0-87.2	7.0 ± 0.2
6	As(III)	0-87.2	8.0 ± 0.2
7	As(V)	0-65.5	7.0 ± 0.2
8	As(V)	0-65.5	8.0 ± 0.2

Method of analysis

The residual arsenic in water sample was determined using rapid colorimetric method with detection limit 1 μg/L [30]. The method was used to estimate As (III) and As(V) concentrations in treated water samples. Arsenic was determined by UV Spectrophotometer (Shimadzu, Japan) and measurements were made at a wavelength of 880 nm using absorbance cell of 5 cm path length. Total iron analysis was performed by 1, 10 phenanthroline method as described in standard methods [31].

RESULTS AND DISCUSSION

There is no significant effect of pH on arsenic removal and more than 99 % As (III) and As(V) removal were obtained by EHFO over entire pH range of 6 to 8 as shown in Table 3. The residual arsenic concentration in water was less than 10 μg/L, which conforms to the recent guideline of WHO. Initially the arsenic removal is rapid due to presence of more arsenic ions and later it decreased gradually as more HFO generated ions formed complexes with arsenic over almost the entire process. It has been reported that arsenic removal increases with increasing iron dosages in chemical coagulation [8]. Similarly in electrochemical process, arsenic removal would be governed by the formation of hydrous ferric oxides (EHFO) in solution.

Several studies reported arsenic removal by conventional coagulation methods using iron salts (e.g. ferric chloride, ferrous sulfate) and aluminum salts (e.g. alum) [8]. They found that removal achieved was up to 99% for As(V) and the removal was only up to 45–55% for As(III).

Tables 4 and 5 show the performance of EHFO and HFO for As(III) and As(V) at different pH (6, 7 and 8). It is observed that in both methods arsenic removal increased with increasing iron dosage and As(V) removal was always higher than As(III) removal. By EHFO, removal of both As(III) and As(V) achieved was greater than 99%.

In chemical generated HFO adsorption up to 97–99% removal was achieved for As(V), but only 90–93% of As(III) removal was obtained even for iron dose of 51.60 mg/L. Experimental observations of less removal of As(III) in comparison to As(V) by conventional coagulation are in agreement with the earlier research work [8]. Based on the results, it is clear that As(V) removal by HFO and EHFO seem to be the same, but As(III) removal efficiencies of EHFO are much higher than HFO.

Table4 Summary of experimental result by using electrochemical hydrous ferric oxide (EHFO)

pH	Arsenic type	EHFO dose (mg/L)				
		2.08	5.2	11.2	18.36	40.12
		Arsenic removal (%)				
6	As(III)	33	56	89	95	99
	As(V)	21	44	95	98	99
7	As(III)	29	56	89	95	99
	As(V)	45	68	91	97	99
8	As(III)	14	51	70	92	99
	As(V)	35	62	88	95	99

Therefore, oxidation of As (III) to As (V) has been suggested for effective arsenic removal. The comparison of EHFO and HFO has been done on the basis of iron dose. In electrochemical process, the amount of iron dissolved was calculated theoretically by using Faraday's law. The validity of Faraday's law was also ascertained experimentally.

Table 5 Summary of experimental results for chemically generated hydrous ferric oxide (HFO)

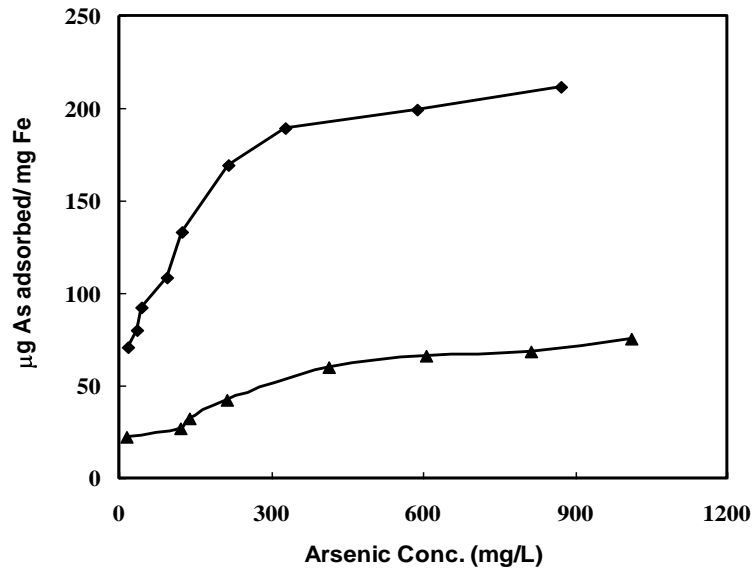| pH	Arsenic type	HFO as Fe (mg/L)				
		3.5	6.9	17.2	25.8	51.6
		Arsenic removal (%)				
6	As(III)	37	41	67	74	98
6	As(V)	73	98	99	99	99
7	As(III)	36	48	68	87	93
7	As(V)	55	94	95	97	99
8	As(III)	22	39	70	84	92
8	As(V)	45	51	92	94	97

Figure 1. Adsorption isotherms obtained for As(III), where, symbols indicate (◆) EHFO; (▲) HFO.
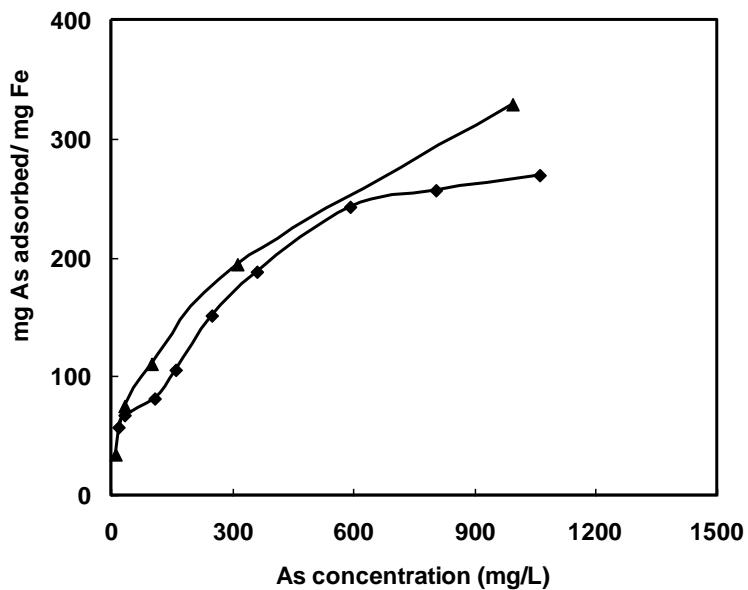

Figure 2. Adsorption isotherms obtained for As(V), where, symbols indicate (◆) EHFO; (▲) HFO.

Fig. 1 and Fig. 2 shows the variation of sorption capacity of As(III) and As(V) with equilibrium arsenic concentration for EHFO and HFO experiments at pH 6. It is evident from the figure that EHFO is more effective for As (III) removal than HFO whereas, As(V) removal by both the processes is nearly the same. The higher As (III) sorptive capacity observed in EHFO in comparison to HF O may be attributed to possible oxidation of As (III) to As(V) during the electrochemical process and formation of nanoscale HFO which increases the surface area of particle and hence results in more arsenic adsorption. The sludge generated by EHFO and HFO is very small and easy to handle. Also the sludge is arsenic rich which can be recovered and used. It can be disposed in buried underground soil pits where it can be mixed with cow dung and can be oxidized by nature cycles.

Effect of phosphate, silica and sulfate on As removal

During the EC process natural water contains some anion and cations among those phosphate finds inhibitory effect and removed from water ultimately competes the arsenic species on the surface of iron oxide such as Lepidocricite. And this effect agrees with the result of previous studies [32], [33], [34]. The presence of 3 mg/L phosphate as P inhibited the removal of As (Fig. 3). As it hampers the conversion of Fe(II) to Fe(III) and observed more in higher concentrations of phosphate.
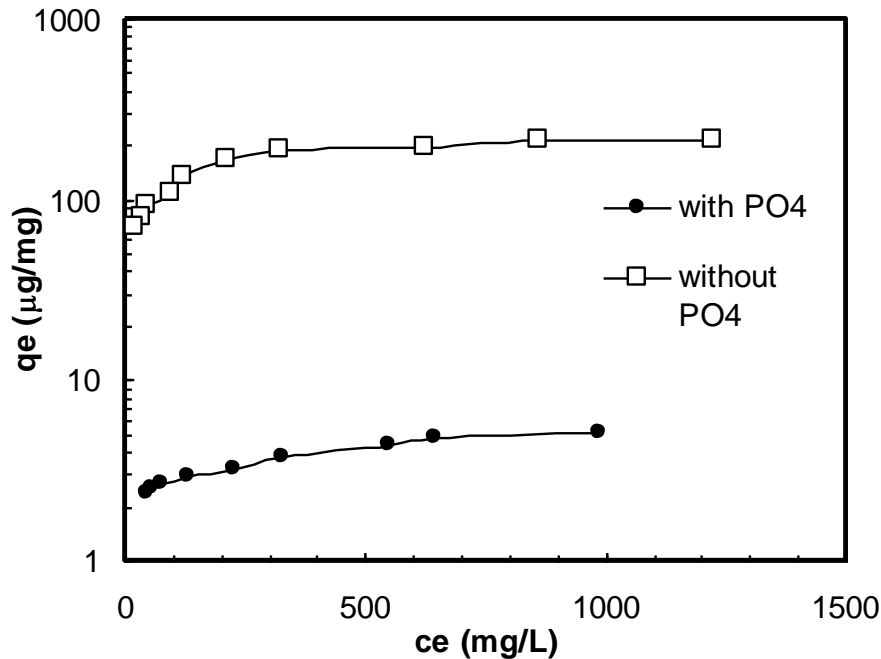

Figure 3. Arsenic Adsorption of HFO in absence and presence of PO$_4$ of concentration 3 mg/L (qe vs. ce)

Dissolved silica of concentration 15 mg/L had no significant effect on arsenic removal (data not shown). [35] Also observed no significant effects of silica on As(V) adsorption to iron hydroxides when silica was present at concentrations as high as 36 mg/L. In a separate study, [23] observed silica inhibition of As(V) adsorption to ferric hydroxide, but only when silica and ferric hydroxide had been pre-equilibrated for 50 days; at shorter contact times, there was much less inhibition. An inhibitory effect of silica was also observed in a study of arsenic removal using a porous iron oxide-based sorbed in packed columns [36]; the inhibition was likely caused by silica polymerization to physically block access to adsorption sites within internal pores of the solid and not through any competitive adsorption phenomenon.

The presence of 10 and 50 mg/L SO$_4$ $^{2-}$ (data not shown) did not affect the removal of As. Because sulfate did not affect lepidocrocite formation and does not adsorb as strongly as As(V) and phosphate, it is not surprising that sulfate did not affect the performance of the electrocoagulation process. Similar observations were made in As adsorption experiments with sulfate concentrations higher than 200 mg/L [34], [37].

Operating Cost

The electricity required for treatment of 50 L was 0.035- 0.04 kW.h or 0.70-0.76 kW h/m^3. The time required to produce sufficient coagulant for arsenic removal depends on the current, which in turn is affected by the conductivity of the water. The electrode spacing and treatment time can be adjusted for the conductivity of a specific groundwater. The operating costs of treatment are primarily those of the electricity. Based on typical electricity costs in India, the operating costs are estimated to be 5.6 Indian Rupees per m^3 of water treated (\$ 0.1/m^3 with a currency conversion of 56.11 Rupees per U.S. dollar) similar result found by [34].

PRACTICAL APPLICATIONS

The point of practical relevance is how much iron is needed for the removal of As(III). [38] reported the arsenite removal capacity of zero valent iron was 7.5 mg As / g Fe. [32] reported that removal of 90% arsenic in phosphate and silicate-rich water (1.9 mg/L P, 18 mg/L Si) required a Fe/As ratio of at least 40, after the As(III) has been oxidized to As(V) with hypochlorite. Similar to these findings, [39] found that 15–18 mg Fe (III) are needed to remove 90% of 500 μg/L As (V) (with 3mg/L P and 20 mg/L Si). As (III) removal under the same conditions required over 80 mg /L Fe(III) [40]. In contrast, addition of a total of 20 mg/L Fe(II) in eight additions every 30 min or 15 mg/L Fe(II) added continuously (this study) was sufficient to remove 90% of As(III) without an added oxidant [40]. Similarly 40 mg of EHFO was needed to remove 99 % of 2 mg/l of As (III) and As (V), which is very less, compared to the processes like HFO.

CONCLUSION

EHFO was able to remove more than 99% (<10 μg/l) of arsenic from arsenic contaminated water and met the drinking water standard of 10 μg/L with iron electrode. Arsenic removal is rapid at higher current densities but when the results of different current density were converted into charge density, arsenic removal correlated well with charge density and hence charge density is a suggested as a design parameter for the process. There is no significant effect of pH on both As (III) and As (V) removal in the pH range 6 to 8. As (III) was more efficiently removed by EHFO than HFO and other conventional methods. As(V) removal performance of both EHFO and HFO was nearly same.

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