

Arsenic and iron removal from water using constructed soil filter – a novel approach

Pravin Nemade, * Avinash M. Kadam and Hariharan S. Shankar

Department of Chemical Engineering, Indian Institute of Technology, Bombay, Powai, Mumbai, Maharashtra, India

Received 9 September 2007; Accepted 16 May 2008

ABSTRACT: Arsenic poisoning is a global problem. It is known that groundwater in Bangladesh and West Bengal predominantly contains As(III) and concentrations of Fe(II) which are often too low for removal of arsenic in optimum quantities. To eliminate arsenic from drinking water, several basic methods such as coagulation, adsorption, ion exchange, filtration, membrane processes, are reported. To deal with this issue on a small to large scale, we present a novel approach to remove arsenic and iron from water by constructed soil filter (CSF).

In CSF, As (III) is oxidized to As(V) by media via natural oxidation and subsequently, arsenic is co-precipitated with iron. Here water containing arsenic, iron, and phosphate is passed through the CSF media at the rate of 100 ml/min. Results show a mean residence time of 8.6 min and a holdup of 1.85 l. Results of six runs show that with initial As(III) of 300 μ g/l and iron concentration of 5–10 mg/l, residual arsenic levels below 10 ppb and residual iron <0.30 mg/l in water can be achieved.

In CSF, no chemicals are needed for the oxidation of As(III) to As(V). The oxidation takes place due to the presence of various oxides such as iron, manganese, aluminum, and the microbial diversity in the media of soil bioreactor. Such a novel technology is also applied for purification of water and wastewater. © 2008 Curtin University of Technology and John Wiley & Sons, Ltd.

KEYWORDS: arsenite; oxidation; constructed soil filter; adsorption; hydrous ferric oxide

INTRODUCTION

Arsenic is a highly toxic chemical constituent thereby posing epidemiological problems to human health worldwide. In Bangladesh and India, drinking water drawn from underground sources has been responsible for widespread arsenic poisoning affecting nearly 100 million people.^[1] Arsenic enters the environment through both natural and processes such as weathering of arsenic containing minerals, geothermal water, weathered sedimentary rocks, and anthropogenic processes including application of organo-arsenical pesticides and wastes from metallurgical, glassware, mining, and ceramic industries.^[2] Inorganic arsenic is predominantly present in natural waters ranging from 0.20 to 3.7 mg/l in India.^[3] In acid mine drainage the normal range of the concentration of arsenic in Carnoules Creek, France has recently been reported as 0-250 mg/l.

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). As understood from the literature,^[3-5] the pH factor is a main trigger for arsenic release from the solid-iron oxide phase. As(III) is mostly present as un-ionized species in the pH of normal groundwater. As(V) is present as negative ion. At natural pH conditions, arsenite exists in solution as H₃AsO₃ and H₂AsO₃⁻, while arsenate is present as H₂AsO₄⁻, and HAsO₄²⁻. Under high pH (>9) conditions, arsenite is more strongly bound to soil components than arsenate. In addition, arsenite is more toxic than arsenate and tends to be more mobile in the environment.

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, Nepal, Poland, Hungary, Belgium, Chile, Argentina and North Mexico.^[5] Manifestation of high 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, and central nervous system, and lead to diseases like skin lesions, hyperkeratosis, and hyperpigmentation.^[6,7] Due to its toxic effects on

^{*}*Correspondence to*: Pravin Nemade, Department of Chemical Engineering, Indian Institute of Technology, Bombay, Powai, Mumbai, Maharashtra, India. E-mail: npravin@iitb.ac.in

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human health, recently the United States Environmental Protection Agency (EPA) has lowered the maximum contaminant level (MCL) of arsenic in drinking water from 50 to 10 μ g/l.

Many researchers have put forward their efforts to alleviate the problem of arsenic in water bodies. 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 arsenic removal by conventional coagulation^[7,8] and the removal rate achieved was found to be up to 99% for As(V), but only up to 65-85% for As(III). Therefore, oxidation of As(III) to As(V) is considered a critical step for effective arsenic removal. Other methods include adsorption on alumina,^[9] anion exchange and reverse osmosis (RO). Some recent treatment technologies based on oxidation and adsorption of arsenic are green sand filtration,^[10] iron oxide-coated sand,^[11] manganese dioxide-coated sand,^[12] ferruginous manganese ore,^[13] ferrihydrite,^[14] clav minerals.^[15] iron oxide-coated cement (IOCC)^[16] and zero-valent iron.[17]

Table 1 shows a review of the maximum adsorption capacities of different adsorbent materials. Due to the treatment capacity of the coagulation process, the high cost of resins and the problem of membrane fouling, adsorption using low cost adsorbents has been proved to be a suitable alternative, offering more reliable and efficient removal of complex, inorganic and organic metals than many other conventional methods.

Numerous studies have shown that phosphates exhibit a high affinity for adsorption and co-precipitation in the presence of iron (oxyhydr) oxides.^[31,32] Arienzo *et al.*^[33] evaluated the extent of removal of As(III) from tap water by adsorbing colloidal HFO produced during the electrochemical peroxidation (ECP) and assessed the effects on As removal and found >98% removal of As(III) in HFO. From the above discussion, it is clear that As (III) removal is favored by oxidation to As(V) followed by removal via suitable methods. Accordingly, it is necessary to investigate the treatment approaches that would consistently provide drinking water with arsenic levels less than 10 µg/l.

Iron is found in surface and ground waters at varying concentration levels, usually up to 3–4 mg/l, but in some cases up to 15 mg/l.^[34] In West Bengal, India, owing to iron-rich soil, groundwater invariably contains dissolved iron or Fe(II) ranging from 2 to 10 mg/l.^[3] When present, even at low concentrations it can be linked to aesthetic and operational problems such as bad taste and color, staining, as well as deposition in the water distribution system leading to incidence of high turbidity.^[35] Also, iron promotes the growth of certain types of chlorine-tolerant microorganisms in water distribution systems, thus increasing the costs for cleaning and sterilizing in addition to odor and taste problems. The permissible limit for iron in drinking water recommended by WHO standard is 0.3 mg/l.

Ferrous ion is oxidized in air according to the following reaction:

$$Fe^{2+} + (1/4) O_2 + H^+ \leftrightarrow Fe^{3+} + (1/2) H_2O$$
 (1)

The state of iron in water depends, above all, on the pH and the redox potential. By increasing the pH content, dissolved iron (Fe²⁺ or Fe³⁺) hydrolyzes to form precipitates. The ferrous ion hydrolyzes to produce an array of mononuclear species of FeOH⁺ to Fe(OH)₄²⁻ between pH 7 and 14. The ferric ion (Fe³⁺) hydrolyzes much more readily than the ferrous ion. The rates of ferrous iron oxidation by air increase with pH and about 90% conversion may be achieved in a few minutes at a pH rate of 7. As constructed soil filter (CSF) is an oxygen reservoir (>5.0 mg/l) which oxidizes Fe(II) to Fe(III) forming Fe(OH)₃ and removes

	Adsorption capacity (mg/g)		
Adsorbent	As(III)	As(V)	References
Mesoporous alumina	47.2	121.37	[18]
Iron (III)-loaded orange waste	68.17	68.62	[19]
Nanocrystalline titanium dioxide	59.94	37.46	[20]
Bead cellulose-loaded iron oxyhydroxide	99.6	33.2	[21]
Activated alumina	3.48	15.9	[22]
Waste Fe(III)/Cr(III) hydroxide	_	11.02	[23]
Ferrihydrite	5.63	11.59	[24]
Iron filings	1.5	0.6	[25]
Polymetallic sea nodule	0.69	2.85	[26]
Granular ferric hydroxide	0.11	0.16	[27]
Iron oxide-coated sand	_	0.008	[28]
Polymeric Al-/Fe-modified montmorillonite	19.11	21.23	[29]
Ferruginous manganese ore	0.537	_	[30]

Table 1. Maximum adsorption capacities for As(V), As(III) and DMA adsorption by different adsorbents.

To eliminate arsenic and iron simultaneously so as to comply with WHO drinking water guidelines, a robust method is required. The present study provides this option via a novel concept called CSF.^[36] In CSF, fundamental chemical reaction of nature such as respiration, photosynthesis and mineral weathering are integrated and synergized as shown in Fig. 1. In CSF, respiration serves to bring about oxidation of organic and inorganic substances, and reduces the oxygen demand substantially. Mineral weathering enables these reactions to occur at required rates, while photosynthesis serves as a bio-indicator of process performance. CSF houses an engineered ecology of formulated media containing selected micro- and macro-organisms such as the geophagus worm, namely, Pheretima elongata (k selected organism), ammonia oxidizers, nitrifiers, denitrifiers, proteolytic bacteria, actinomycetes protozoa, naked amoebae, flagellates, and ciliates etc. which are cultured to maintain the required soil microbial ecology.^[37] The media constituents contain mineral oxides of Fe, Mn, Si etc. which could oxidize As(III) to As(V) and are found to be close to a state of complete oxidation of As(III) without using any chemicals^[38] from water containing arsenic, iron, and phosphate. CSF efficiently removes biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids, phosphate, bacteria, and nitrogen.^[37,39,40]

The objective of the present study is to evaluate the performance of the novel CSF system for removal of arsenic and iron from water.

MATERIALS AND METHODS

The experiment was conducted in a bioreactor, an aluminum vessel filled with formulated media and under drain, mounted on a metal frame. The total height of the bioreactor is 30 cm with a diameter of 28.5 cm. The weight of the formulated media is 26.3 kg. The characteristics of the media are shown in Table 2. Table 3 shows the constituents of the formulation along with the height of the constituents in the bioreactor.

Chemicals

Experiments were performed at ambient temperatures ranging from 26 to 28 °C. Stock solutions of arsenite were prepared by dissolving an appropriate quantity of arsenic trioxide, As_2O_3 , (S.D. Fine Chem Ltd., India) in tap water containing 1% (w/w) NaOH and the solution was then diluted up to 1 l. The iron and arsenate stock solutions were prepared from FeSO₄, 7 H₂O, and sodium arsenate, Na₂HAsO₄.7H₂O (Loba Chemie, India). The working solutions containing iron and arsenic were prepared by dissolving appropriate amounts of stock solutions in tap water. The diluted solutions were made from the stock as and when required for calibration purpose.

Tap water analysis shows pH ranging from 7.1 to 7.6 and bicarbonate alkalinity between 40 and 50 mg/l as $CaCO_3$. Dissolved iron, phosphate, and arsenic concentration were not detectable in tap water.



Figure 1. Simplified chemistry of the fundamental natural processes in constructed soil filter (CSF). This figure is available in colour online at www.apjChemEng.com.

Item	Details	Value
Underdrain	Gravel-d _p	25 mm
	White sand $d_{\rm p}$	2 mm
Formulated media	d_{p}	20 mm
	Horizontal hydraulic conductivity	0.9 m/h
	Vertical hydraulic conductivity	1.8 m/h
	% Clay	1.0
	% Silt	52
	% Sand	43
	% Gravel	4
	% Oxidizable organic matter	0.08
	% Total organic matter	0.10
	% Carbon	0.55
	% Hydrogen	0.48
	% Nitrogen	0.22
	C/N	2.40
	Cation exchange capacity (m.e./100 g)	46.20
	Anion exchange capacity (m.e./100 g)	1.9

Table 2.	Characteristics	of the t	formulated	media.
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Constituents	Height (mm)
Media (8–10 mm)	240
Sand (1–3 mm)	20
Thin gravel $(5-8 \text{ mm})$	20
Thick gravel (8–12 mm)	20

Experimental

A laboratory scale bioreactor is fabricated and all tests were conducted in gravity flow mode at normal pH (7.2). A schematic representation of the process is shown in Fig. 2. Water containing arsenic and iron was pumped through a bioreactor with a volume capacity of 20 1 (0.285 m diameter and length of 0.30 m) by a peristaltic pump (Cole Parmer Instrument Company, Ontario). The packed volume had enough headspace to allow for expansion of the medium during backwashing. The flow rate was kept constant at 100 ml/min. All the apparatuses used were washed with tap water for 12 h. The synthetic arsenite As(III) water was prepared in the laboratory. The initial As(III) concentration was kept at 300 µg/l of water. Six runs of down flow column tests were conducted. In each run 25 l of water containing arsenic was passed. Samples were collected at regular time intervals and co-precipitated by FeCl₃ followed by filtration through filter paper and then analyzed for residual As(III) and As(V). Before the commencement of each run, the column was washed with tap water until the residual arsenic levels reached less than $10 \mu g/l$.

Analysis

The residual arsenic in water sample was determined using rapid colorimetric method^[41] with a detection



Figure 2. Schematic of arsenic removal process by constructed soil filter (CSF).

limit of 1 µg/l. The method was used to estimate total Arsenic and As(V) concentrations in treated water samples. Arsenic was determined by double beam UV Spectrophotometer (Shimadzu, Japan) and measurements were made at a wavelength of 880 nm. Total iron analysis was performed by 1, 10 phenanthroline method.^[42] Water temperature, conductivity, and total dissolved solids (TDS) were measured using WTW (Germany) Inolab1 conductivity meter; pH and dissolved oxygen (DO) were measured using WTW (Germany) Inolab1 pH/Oxi meter; and turbidity was measured on WTW (Germany) Turb 550.

RESULTS AND DISCUSSION

Hydrodynamic characteristics of bioreactor were studied. At a flow rate of 100 ml/min, the holdup was 1.85 l

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(11%) (Drainage method) and mean residence time was 8.62 min (potassium bromide tracer method).

Table 4 shows the natural oxidation of As (III) to As (V). A nearly complete conversion of As(III) to As(V) takes place and further arsenic removal by coprecipitation with ferric chloride. As As(III) is nonionic in natural water with pH 6.5 to 8; it gets converted to As(V), after passing through CSF, by the media which is ionic, but does not get adsorbed on media. Table 4 shows the correct mass balance of influent and effluent arsenic in water.

In all experiments, the residual arsenic in water was found to be <10 µg/l in compliance with WHO drinking water standard. Table 4 shows that there is very little adsorption of As (V) in the bed. Almost 99% oxidation of As(III) to As(V) takes place using initial As(III) concentration of 300 µg/l and phosphate 2 mg/l for a flow rate of 100 ml/min. Experimental observations of less removal of As(III) in comparison to As(V) by conventional coagulation,^[43] or HFO are in agreement with the earlier research work.^[8]

Phosphate in water is known to affect As(V) coprecipitation with iron reported elsewhere^[8,31,32] and hence reduces As(V) removal efficiency. It can be seen from the previous study by Kadam^[37,40] that even low phosphates ranging from 0.26 to 0.31 mg/l are effectively removed from water by CSF as shown in Table 4. So, there is no interference of phosphate with arsenic while knocking the As(V) by ferric chloride.

Tables 5 and 6 show the removal of iron with an initial concentration of 5 and 10 mg/l by CSF. The results shows residual iron of <0.3 mg/l in effluent water which is the WHO standard. CSF has its own ecology and oxygen reservoir as effluent DO >5.0 mg/l in water samples.^[37] The concentration of the DO in the liquid phase throughout the filter depth was always ranging from 5.0 to 5.25 mg/l, which is sufficient to oxidize Fe(II) to Fe(III) forming Fe(OH)₃ that was detained on the support material of the filter. The residual iron is <0.30 mg/l which is the permissible limit for drinking water. Tekerlekopoulou *et al.*,^[44] observed iron (1–3 mg/l) removal from water in trickling filter and the residual iron in the outlet was found <0.3 mg/l.

Table 5. Removal of iron having an initial concentration of 5 mg/l from drinking water having a flow rate of 100 ml/min by using a constructed soil filter.

Run time (h)	Residual iron (mg/l)	Total dissolved solids (mg/l)	EC (µS/cm)	pН
2	0.128	113	170	7.45
4	0.09	111	168	7.36
6	0.08	110	163	7.28
8	0.07	109	162	7.20
10	0.06	109	162	7.12

Table 6. Removal of iron from drinking water having an initial concentration of 10 mg/l with a flow rate of 100 ml/min by constructed soil filter.

Run time (h)	Residual iron (mg/l)	Total dissolved solids (mg/l)	EC (µS/cm)	pН
2	0.25	115	170	7.65
4	0.16	112	170	7.46
6	0.13	109	168	7.38
8	0.11	108	165	7.28
10	0.10	107	164	7.22

The effluent is analyzed for different parameters such as pH (6.7 to 7.4); conductivity (100 to 170 μ S/cm), total dissolved solid (107 to 115 mg/l); DO (4.06 to 5.25 mg/l); and turbidity (1 to 5 NTU) which are found to be within permissible limits for drinking water.

APPLICATIONS

CSF is a novel process that works in a soil environment as per the design of carbon cycle and consumes very little energy for the processing of oxidizable organic and inorganic solids and thereby bringing about purification of water. Here, fundamental processes of nature, viz. respiration, mineral weathering, and photosynthesis are engaged to bring about renovation. The process uses a media and culture which houses an engineered ecology

Table 4. Conversion of As(III) to As(V) in CSF and subsequent removal by $FeCl_3$; Initial As(III) = 300 µg/l, initial phosphate concentration = 2 mg/l, flow rate = 100 ml/min.

Run No	Conversion of As(III) to As(V) in CSF (µg/l)	Dose as Fe (mg/l)	Residual As(V) µg/l	Residual phosphate mg/l	Arsenic removal (%)
1	295	30	7	0.31	98.3
2	295	30	6	0.28	98.3
3	296	30	7	0.30	98.6
4	295	30	6	0.26	98.3
5	296	30	7	0.27	98.6
6	296	30	7	0.26	98.6

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of soil, bio-indicator plants, soil organisms containing selected microorganisms such as geophagus worm, Pheretima elongata (k selected organism), which are cultured to maintain the required soil microbial ecology. CSF promotes effective oxidation of As(IIII) to As(V) without the need of an additional oxidation step in a formulated media via natural oxidation containing minerals such as Fe, Mn, Si.

The CSF can be used in large-scale removal of arsenic, iron, and phosphate in polluted areas. In addition to this, CSF can be used for removal of bacteria, and dissolved and suspended solids. CSF might be a novel technology to implement in rural villages or town at large scale as well as for a small community.

CONCLUSION

CSF can also be used for the removal of bacteria, and dissolved and suspended solids from water and wastewater. Based on the results, it is clear that CSF proves to be a novel concept for removal of iron and arsenic from water and offers an alternative method to conventional water treatment plants.

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