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Arsenic Remediation from Drinking Water Using Fenton's Reagent with Slow Sand Filter

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Abstract This paper describes the development of a remediation approach based on the pre-oxidation using Fenton's reagent and the subsequent removal of arsenic (As) through sand filtration from drinking water. The efficiency of the process was carried out including As(III) and As(V) with various concentration ratios up to 3,000 ppb. Efficient removal of As was observed within WHO guideline value of 10 ppb. The recoveries of samples were found to be 98 % ± 2.5 %. The process was applied to field samples, where results show considerable reduction in As concentrations. This process is cost effective for treatment of drinking water with high concentration of As.

Keywords Arsenic remediation · Pre-oxidation · Drinking water · Fenton's reagent

Arsenic is a naturally occurring element present in the environment. As is released to the environment mostly through natural processes, due to the presence of arsenical minerals, volcanic emissions and inputs from geothermal sources, as well as a consequence of anthropogenic activities, such as mining activities, combustion of fossil fuels and use of arsenical pesticides (Jain and Ali 2000). Occurrence of As in ground water much exceeding the tolerance limit (10 ppb) is a global problem and posed an ever-increasing degree of health hazard (Smedley and Kinniburgh 2002). It is a matter of worry that although WHO lowered the guideline value for As from 50 to 10 ppb, some countries like Bangladesh and China still have 50 ppb As in drinking water (Mohan et al. 2007; Pesola et al. 2012). Natural As pollution of drinking water supplies has been reported from over 70 countries, posing a serious health hazard to an estimated 150 million people world-wide (Ravenscroft et al. 2009) and in Asian countries more than 38 million people exposed to ground water with high concentrations of As(Nordstrom 2002).

The application of Fenton's reagent, which has been recently been brought in focus for arsenic removal (Jaafarzadeh et al. 2012). Variety of treatment technologies have been used for the removal of As from water. Advanced oxidation processes (AOPs) are known for their capability to mineralise a wide range of organic compounds. Among AOPs, oxidation using Fenton's reagent (H_2O_2 in the presence of Fe^{2+} catalyst) is an attractive and effective technology for the degradation of a large number of hazardous and organic pollutants because of the lack of toxicity of the reagents, eventually leaving no residues and the simplicity of the technology (Wang 2008). Fenton's reagent is a catalytic oxidative mixture that contains iron ions and hydrogen peroxide, producing extremely reactive and non-selective hydroxyl radicals, as can be seen in

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reactions (1) and (2) (Oliveros et al. 1997; Pang et al. 2009) and Fe(II) is oxidized to Fe(III) which forms a precipitate that gathers all the As(V) (Balarama et al. 2001).

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^- + {}^{\cdot}OH \quad (1)$$

$$^{\cdot}OH + RH \rightarrow \text{oxidation products} \quad (2)$$

Many studies have revealed that the use of a much higher concentration of Fe(II) could lead to the self scavenging of $^{\cdot}OH$ radical by Fe(II) (Chen and Pignatello 1997; Joseph et al. 2000) and induce the decrease in degradation rate of pollutants. In present study the dose of H_2O_2 and $FeSO_4 \cdot 7H_2O$ for preoxidation of As was obtained through jar test and quality of treated water was maintained by passing oxidized water through slow sand filter.

Materials and Methods

Distilled deionized water was used for the preparation of standards. The calibration standards were prepared from preserved (stock) As standard of 1,000 ppm. The sample concentrations were verified using a series of As calibration standards. The standard stocks arsenite [As(III)] and arsenate [As(V)] solutions (1,000 mg/L) were separately prepared from Sodium meta arsenite ($NaAsO_2$) and sodium arsenate ($Na_3AsO_4 \cdot 7H_2O$) and preserved for experimental studies. These stock solutions were used to prepare working solutions with As(III) and As(V) in the ratio 1:1 using natural water. Natural water has been collected from tap water with total As less than 5 ppb. All the experiments were performed at natural pH of the water samples.

Jar test was carried out to optimize the doses of H_2O_2 and $FeSO_4 \cdot 7H_2O$ for Fenton's reagent to remove As. It was performed with 500 mL of As spiked water taken in 1,000 mL beakers. Test water was prepared as shown in Table 1. This solution was stirred slowly for 30 s. Subsequently, beakers were kept stable for 30 min to settle down the flocks. Supernatant was filtered through Whatman filter paper no. 42 and analyzed for As. The experimental results for removal of As shown in Table 1. Table 2 illustrates the results of jar test carried out at different concentrations of As with optimized dose of Fenton's reagent.

Table 1 Removal of As using Fenton's reagent (H_2O_2 + $FeSO_4.7H_2O$) by Jar test

µL H_2O_2 & mg of $FeSO_4 \cdot 7H_2O$	As Conc.[As(III):As(V) (1:1) (c:c)] (ppb)		
	250	500	1,000
	As removal efficiency (%)		
50 & 50	100.00	98.85	96.18
100 & 0	28.34	24.36	25.55
0 & 100	99.70	96.02	93.65

c:c concentration:concentration

Table 2 Removal of As using Fenton's reagent (50 µL H_2O_2 + 50 mg $FeSO_4 \cdot 7H_2O$) by Jar test

As(III):As(V) (1:1) conc. (ppb)	Initial As conc.(ppb)	Residual As conc.(ppb)	As removal efficiency (%)
3,000	3016.28	6.47	99.78
2,000	2044.78	10.17	99.50
100	114.72	ND	100.00
50	51.81	ND	100.00

ND not detected

Batch process was carried out in 15 L of As spiked water taken in a 20 L plastic bucket. Oxidation reaction was carried out by adding H_2O_2 and $FeSO_4 \cdot 7H_2O$ in the range of 1.5 mL/L and 1.5 g/L, respectively. This was stirred slowly for 3 min and kept stable for 30 min. This was allowed to pass through a sand bed which yielded clear water. Samples were collected at regular time interval of 30 min and at the last composite sample were collected. These samples estimated for As and, at the same time Iron, Sulphate, H_2O_2, pH, Conductivity and Turbidity were also measured. For As estimation samples were preserved by adding concentrated hydrochloric acid (HCl) till pH becomes ≤ 2. During the batch process with initial concentration of 100, 500, 1,000, 2,000 and 3,000 ppb, the residual As concentration after treatment was ≤ 10 ppb.

All samples were analyzed in 2 days and analyzed in triplicate. The data analyzes were made with a computer for kinetics by excel software work sheet. The As measurement in all the cases corresponds to total As. Arsenic was estimated by silver diethyldithiocarbamate method using UV–visible Spectrophotometer, following the American Public Health Association (APHA) standard methods (APHA 2005). The detection limit of spectrophotometric method is 1–10 ppb (Rasmussen and Andersen 2002; APHA 2005).

Estimation of residual H_2O_2 was carried out by titrimetric method. 50 mL of sample was taken in 250 mL conical flask and 3 mL sulphuric acid (6 M), two drops of 5 % aqueous manganous sulphate solution were added and titrate with standardized 0.1 M potassium permanganate to the first permanent, faint pink colour. Blank titration was also performed using 50 mL distilled water. For calculation, following formula was used.

$$H_2O_2 Wt.\% = \frac{(A - B) \times N \times 17.007 \times 100}{W \times 1,000} \quad (3)$$

where A is the volume of the permanganate solution used for the titration in millilitre, B the volume of the permanganate solution used for the blank titration in millilitre,

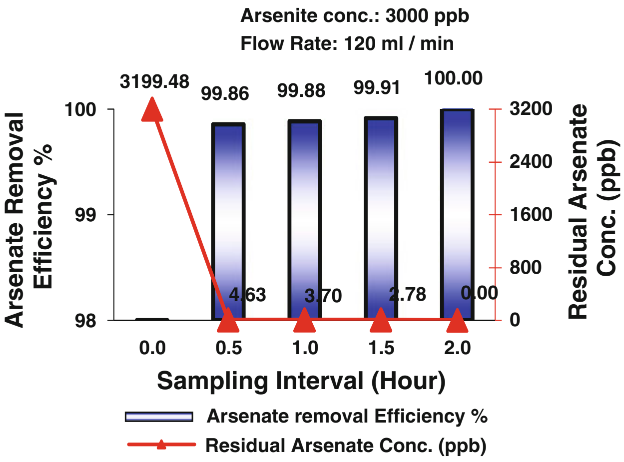

Fig. 1 Removal of arsenate using Fenton's reagent with slow sand filter

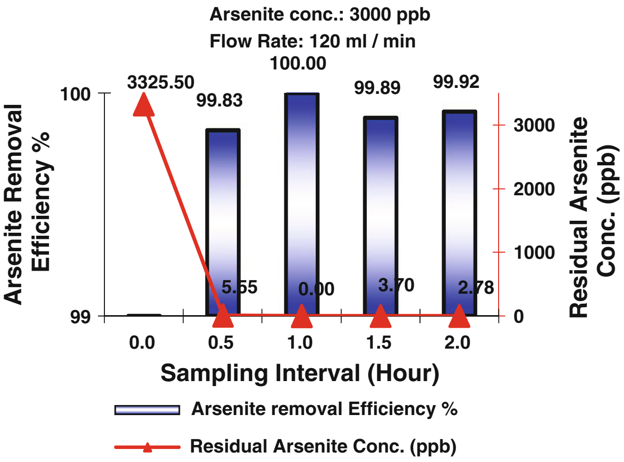

Fig. 2 Removal of arsenite using Fenton's reagent with slow sand filter

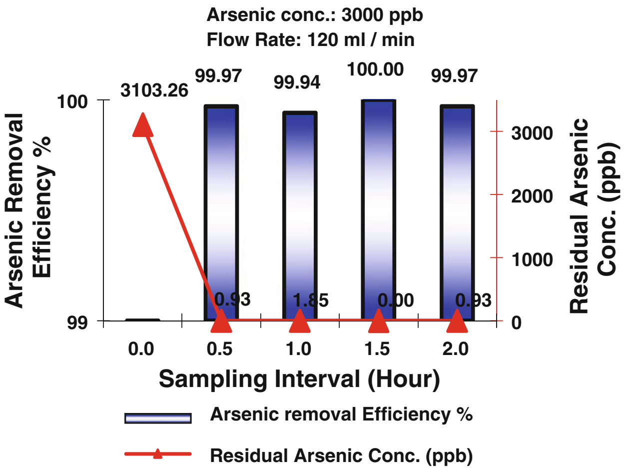

Fig. 3 Removal of arsenic (As III:As V) (3:1) (c:c) (3,000 ppb) using Fenton's reagent with slow sand filter

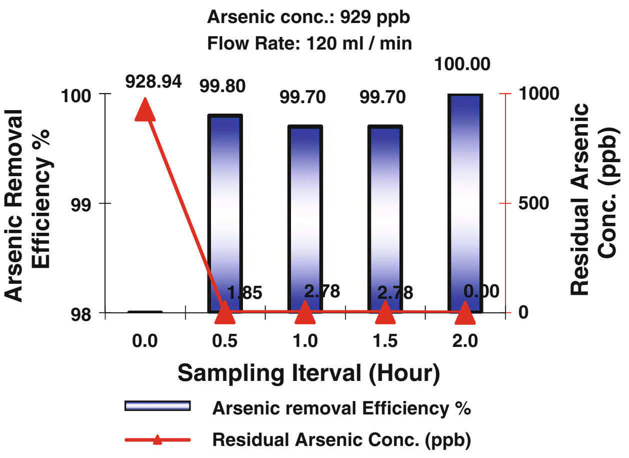

Fig. 4 Removal of arsenic from the ground water sample from Kaudikasa Village using Fenton's reagent with slow sand filter

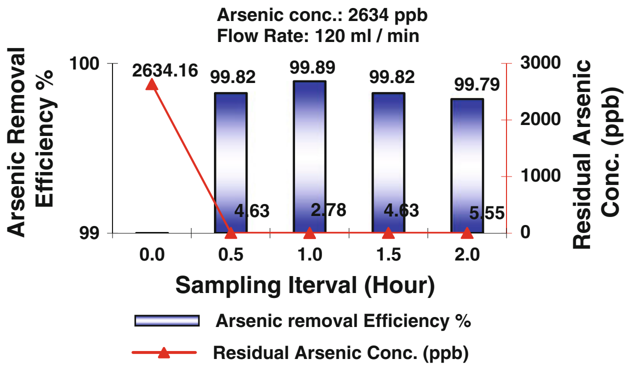

Fig. 5 Removal of arsenic from the ground water sample from Kaudikasa Village using Fenton's reagent with slow sand filter

N the normality of the permanganate solution, and W is the sample weight, in grams.

The following reaction occurs when potassium permanganate solution is added to hydrogen peroxide solution acidified with dilute sulphuric acid.

$$2MnO_4^- + 5H_2O_2 + 6H^+ = 2Mn^{2+} + 5O_2 + 8H_2O$$

For field evaluation of this As removal process, water samples have been collected from a ground water sources in village Kaudikasa of Chawki block of Rajnandgaon district in Chhattisgarh state, India with As concentrations 929 and 2,634 ppb.

Results and Discussion

The feasibility of arsenic removal by pre-oxidation followed by filteration was investigated using Fenton's reagent. Fenton's reagent dose was optimized by jar test. The jar test indicates that addition of 50 μL of H_2O_2 and 50 mg of $FeSO_4 \cdot 7H_2O$ per 500 mL of As spiked water is suitable for As removal. In the view of above Batch process was carried out with initial concentrations of 100, 500, 1,000, 2,000 and 3,000 ppb of As(V), As(III) and As [As(III): As(V) (1:1) (c:c)] each. The batch process after oxidation with Fenton's reagent and 30 min reaction time followed by filtration through slow sand filter was found to be suitable for consistently producing waters with As content $\leq$10 ppb, i.e. capable of removing As lower than the WHO guideline value of 10 ppb. Percent recoveries of samples were found to be 98 % ± 2.5 %. The results for 3,000 ppb of As(V), As(III) and As [As(III): As(V) (1:1) (c:c)] are as shown in Figs. 1, 2 and 3 respectively. Measured values of pH, conductivity, turbidity, iron, sulphate and H_2O_2 of each sample of batch process were found to be within the range of their guideline values of WHO. The removal of total As from drinking water based on the pre-oxidation using Fenton's reagent through slow sand filtration is novel studied than previous studies (Katsoyiannis and Zouboulis 2002; Katsoyiannis et al. 2008).

For field samples the As removal efficiency is good for the batch process with initial As concentrations of 929 and 2,634 ppb. The results of this study are presented graphically in Figs. 4 and 5. The lab study of these samples has shown good arsenic removal efficiency. Hence five arsenic removal units using Fenton's reagent is installed in village Kaudikasa of Chawki block of Rajnandgaon district in Chhattisgarh state, India.

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