



Simple electrocoagulation set up using cloth filter for arsenite removal from aqueous solution

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ABSTRACT

Electrocoagulation is an emerging water treatment technology that has been applied successfully to treat various wastewaters such as potable water, urban wastewater, heavy metal laden wastewater, restaurant wastewater, and colored water. Electrocoagulation offers possibility of anodic oxidation and in situ generation of adsorbents (such as hydrous ferric oxides, hydroxides of aluminum) on which As(III) might be oxidized to As(V) gets adsorbed on to the metal hydroxides (HFO) generated. Therefore, electrocoagulation would be a better choice for arsenic removal from water. In the present study an attempt has been done to develop a simple process employing electrocoagulation with iron electrodes and separation of HFO has been achieved by cloth filtration. Arsenic laden water is passed through a double layer cotton cloth in 2 L volume beaker at flow rate 44ml/min with current 0.2 ampere at pH 7.0. Results show that arsenic levels below 50 µg/L would be achieved which is drinking water standard in India and Bangladesh.

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Introduction

Arsenic (As), a common trace inorganic contaminant in drinking water and well known toxic element around the world is extremely detrimental to human beings. Recent epidemiological evidences on arsenic carcinogenicity suggest that the standard of 50 ppb may not be sufficiently protective to human health (Pontius et al. 1994). Therefore, the U.S. EPA has reviewed the benefits and costs associated with implementing a MCL of 3–20 ppb and recently they have finalized a new U.S. standard of 10 ppb (U.S. EPA 2001). Arsenic found in groundwater by leaching from geological formations and natural weathering of arsenic-bearing rocks/minerals, although in certain areas, high arsenic concentrations are caused by industrial waste discharges and application of arsenical herbicides and pesticides (Smedley and Kinniburgh, 2002). Arsenic exists in multiple oxidation states (+5, +3, 0 and -3); arsenate As(V) and arsenite As(III) are the most common inorganic forms of arsenic in aquatic environments. Arsenate species (AsO_4^{3-} , HAsO_4^{2-} and H_2AsO_4^-) are considered to be soft acid and mostly stable in oxygen rich environments. Whereas, arsenite species (AsO_3^{3-} , $\text{AsO}_2\text{OH}^{2-}$, $\text{As}(\text{OH})_4^-$ and $\text{As}(\text{OH})_3$) are stable in moderate reducing environments such as underground water. Furthermore, As(III) has higher toxicity and more mobility, which need to be oxidized to better adsorbing As(V) (Mohan and Pittman, 2007; Onnby et al., 2012). The surface charge properties of As(V) make it easier to remove from water (Onnby et al., 2012).

Chronic health effects of arsenic include development of various skin lesions such as hyper pigmentation (dark spots), hypo pigmentation (white spots), and keratosis of the hands and feet. Skin cancers and internal cancers lung, kidney, liver, and bladder can appear due to high arsenic exposure. Long term exposure to high levels of arsenic may cause serious health problems including skin, cardiovascular, neurological, renal and respiratory diseases in humans (Bissen and Frimmel, 2003).

Adsorption on low-cost media is an attractive means for trace metal removal from water: Adsorption-based processes are reliable and efficient for removal of complex inorganic and organic metals than many conventional treatment methods (Benjamin et al. 1996). Amorphous iron oxide or ferrihydrite is a common coating of subsoil particles and has a high capacity for different anions including arsenic (Pierce and Moore 1982). Hydrous ferric oxide (Hearing et al., 1996; Nemade et al., 2007) and constructed soil filter (Nemade et al., 2009). Iron containing salts have also been used for coating of quartz sand for the removal of arsenic from groundwater (Joshi and Chaudhuri 1996). Electrocoagulation has been reported to offer various advantages over conventional coagulation in conjunction with other processes, such as dissolved air floatation (Pouet and Casmick, 1995), electrofloatation (Chen et al. 2000). Treating various types of wastewater such as domestic grey water reuse, laundry wastewater, decolourization of dye (Can et al. 2003). From the literature reported, the electrolytic oxidation of iron rods followed by hydrolysis was expected to result in production of the solid iron hydroxides, necessary for contaminant removal (Chen et al., 2000; Mills, 2000; Balasubramanian and Madhavan, 2001; Mollah et al., 2004; Kumar et al., 2004; Parga et al., 2005; Kobya et al., 2006). During the experiment, it was found that the formation of solid iron hydroxides was erratic at pH 6.5, 7.5, and soluble iron was present in the treated water at pH 6.5 due to slow oxidation rate at low pHs.

EC has been reported to be efficient in treatment of potable water (Vik et al., 1984; Holt et al., 2002), urban waste water (Pouet and Grasmick, 1995; Kobya et al., 2006), heavy-metal contaminated waters (Mills, 2000), turbidity and dye/color (Can et al., 2003) have been removed by EC. Some studies have reported EC to be effective in removing arsenic from water and waste water (Balasubramanian and Madhavan, 2001; Kumar et

al., 2004; Parga et al., 2005; Hansen et al., 2007). Arienzo et al. (2002) and Kumar et al. (2004) reported simultaneous oxidation and removal of As(III) during EC.

In the present study arsenic is removed by electrocoagulation an innovative process using iron as an electrode and cotton cloth as filter which is cheaply available and is non toxic to human being. As(III) was chosen for this study due to its predominant occurrence in the Bengal Delta Basin, in India.

Materials and Methods

Preparation of samples

The experiments were performed at Environmental engineering laboratory, at ambient temperatures ranging from 26 to 28°C. The chemicals were analytical reagent grade and were used without any further purification. All glassware were cleaned with water and 1 N H₂SO₄ and then rinsed with distilled water. Stock solutions of arsenite were prepared by dissolving appropriate quantity of arsenic trioxide, As₂O₃, (S.D. Fine Chem Ltd, India) in distilled water containing 1% (w/w) NaOH and the solution was then diluting up to 1 l with distilled water before use. The working solutions containing arsenic were prepared by dissolving appropriate amount of arsenic from stock solutions in tap water. Tap water was tested for the pH, alkalinity, and the presence of arsenic, iron and phosphate. It was found that the pH of the water varied from 7.2 to 7.5, bicarbonate alkalinity was approximately 45–50 mg/l as CaCO₃, the dissolved iron, and phosphate and arsenic concentration were not detectable in tap water.

Procedure

Preliminary experiments were conducted with iron electrode. Two electrodes of same material were placed 0.5 cm apart in a 2 l beaker and arsenic containing water was added in this beaker. The total submerged surface area of each electrode was 36 cm². Before each experiment, the electrodes were abraded with sand paper to remove scale and then cleaned with successive rinses of water and 1 N H₂SO₄.

A direct current by stabilized power supply (0–15 V, 2 A) 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 6–8, the experiments were conducted in this pH range only. The pH of the solution was adjusted by adding either dilute HCl or NaOH.

Table 1. Experimental Conditions Employed in Continuous Mode Electrocoagulation

Expt Run No.	Arsenic Type	As Conc(mg/l)	Current (mA)	Current Density (mA/cm ²)	pH	Flow rate ml/m in	Reactor volume (ml)
1	As(III)	1	200	1.66	7±0.2	44	2000
2	As(III)	1	240	2.00	7±0.2	44	2000
3	As(III)	2	200	1.66	7±0.2	44	2000

Analytical methods

The residual arsenic in water sample was determined using molybdenum blue complex/ rapid method (Dhar et al., 2004). The method was used to estimate As (III) and As (V) concentrations in treated water samples to assess the efficiency of the oxidation step and the subsequent removal of arsenic. Spectrophotometric measurements were made at a wavelength of 865 nm using absorbance cells of 5 cm path length for arsenic determination. Calibration curve for total arsenic was prepared using solutions containing As (III) dissolved in distilled water. All solution preparation and analyzed as per APHA, 1998

Results and Discussion

In any electrochemical process, electrode material has significant effect on the treatment efficiency. The electrode material for drinking water treatment should also be non-toxic to human health. Hence iron was chosen as electrode material as these are non-toxic and readily available instead of aluminum.

A study (Fig. 1) was conducted to evaluate the arsenic removal by the electrocoagulation using 2 litre glass beaker and double cotton cloth to assess the feasibility of this process in domestic filters in arsenic affected areas. For arsenic concentration up to 2 mg/L was treated by EC cloth filter with 44ml/min flow rate for 3-4 hours. This result shows that removal of arsenic is found below 50 ppb. This finding is significant for arsenic removal in Bengal Delta Plain (BDP) areas. Hydraulic retention time (HRT) is calculated by = Volume of Reactor / flow rate = 2000 ml / 44 ml = 45 minutes. Initially, within 45 to 50 minutes arsenic removed 90 % further HFO was generated and residual arsenic concentration was below 50 µg/L as shown in Fig. 1.

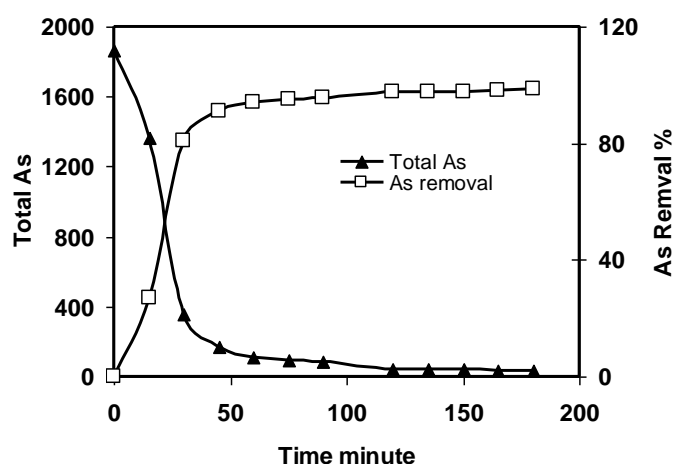


Figure 1. As = 2 ppm, double layer cloth; Current = 0.2 Amp, Volume =2 lit. in beaker, pH=7 flow rate 44 ml /min (Run No. 3)

Theoretically iron dissolved in to the solution at HRT of 45 minute is 104 mg removing 90 % total arsenic from the water treating 1.98 lit water at a flow rate of 44 ml/min. After 120 minute total arsenic removed is found below 50 µg/L at this stage adsorbing capacity is achieved 96 µg/mg at equilibrium arsenic concentration of 45 µg/L.

In electrocoagulation, iron dissolves from the anode and hydrogen gas is developed at cathode. Faraday's law can be used to describe the current density (A/ cm²) and the amount of iron which goes into the solution (g Fe/ cm²) (Vik et al., 1983)

$$w = \frac{itM}{ZF} \quad (1)$$

where,

w = metal dissolving (g Fe/Fe cm²)

i = current density (A cm²)

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.

Fig. 2 and Fig. 3 present the results of experimental run no. 3. The figure shows the variation of total as, and total As removal with charge passed in solution. It can be seen from the figure that arsenic removal was 95.5% and the residual arsenic

concentrations obtained were less than 50 $\mu\text{g/L}$ after 100 coulombs per liter (C l^{-1}) was passed.

Effect of Current Processing Time

The Fig. 4 shows that up to 85–90% of the initial concentration decreased within 15–25 min of the process and the residual arsenic concentration in water was less than 50 $\mu\text{g l}^{-1}$ at pH 7, which in the recent guideline value of WHO. At the beginning of process the arsenic removal is rapid and later it gradually decreased over the entire process examined due to arsenic ions which are more abundant at the beginning of the EC process, and the generated iron hydroxides due to corrosion of the anode at that time will form complexes with arsenic and therefore rapid removal of arsenic was observed. As the experiment proceeds the aqueous phase arsenic concentration goes on reducing and simultaneously hydrous ferric oxides concentration increases, so the curves are nearly the remains horizontal at the end of experiment.

The pH increase in electrocoagulation is attributed to the formation of hydrogen gas at the cathode (Vik et al., 1984; Chen et al., 2000). Also a slight increase in pH may be expected because of sorption reactions of As(V) and As(III), which release OH^- groups from sorbents as a result of ligand exchange (Arienzo et al., 2002). Ratna Kumar et al., (2004), reported that there is no effect of pH on both As(III) and As(V) removal up to 97–99%. Therefore, the filter runs at pH 7.0.

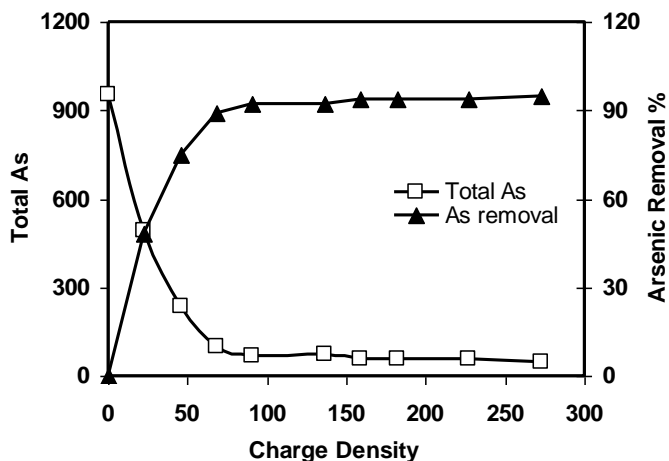


Figure 2. As = 1 ppm, double layer cloth; Current = 0.2 Amp, Vol. 2 Lit., pH = 7 flow rate 44 ml/min, in Beaker

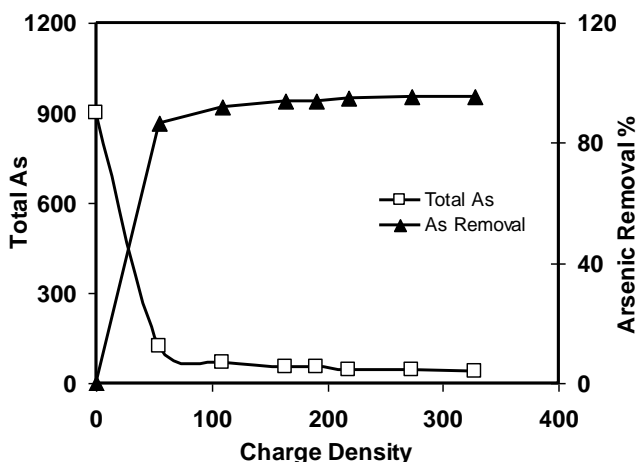


Figure 3. As = 1 ppm, double layer cloth; Current = 0.24 Amp, Volume 2 lit, pH = 7 flow rate 44 ml/min

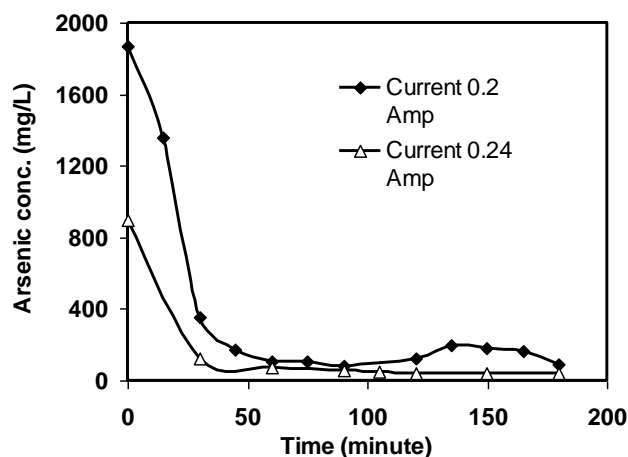
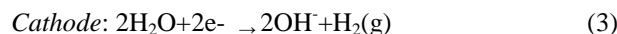


Figure 4. Effect of current density on arsenic removal, (experimental run no. 2, 3), symbols indicate (Δ) As = 2 mg/L and Current = 0.24 Amp, and (\blacksquare) As = 2 mg/L and current 0.2 Amp, double layer cloth; Volume 2 lit, pH = 7, flow rate 44 ml/min.

Change in pH during Electrocoagulation

The faster Fe^{2+} oxidation observed during EC was attributed to a temporary elevation in pH during Electrocoagulation/electrolysis, which did not occur in chemical coagulation process. During batch EC, the pH increased locally in the electrolytic cell/reactor over the period of electrolysis and then slowly decreased reported by (Lakshamanan et al., 2009, 2010). More specifically, the pH increased from initial pHs of 6.5, 7.5, and 8.5 to 7.0 ± 0.1 , 8.1 ± 0.2 , and 9.0 ± 0.1 , respectively, during electrolysis. However, after electrolysis, the pH decreased, and the final pH after 2 min of mixing remained unchanged for initial pH 7.5, slightly increased for initial pH 6.5, and slightly decreased for initial pH 8.5. A similarly sharp increase and then decrease in pH has been reported during Al-EC by Holt et al. (2002). When the EC current stops and Fe^{2+} generation ceases, the Fe^{2+} oxidation by dissolved oxygen (DO) continues to consume the available hydroxides resulting in a pH decrease. See the following reactions given by (Lakshamanan et al., 2009; 2010).



At pH 6.5, the oxidation of Fe^{2+} is slow and the hydroxides produced at the cathode are not fully utilized, which results increase in initial pH. At pH 8.5, in addition to $\text{Fe}(\text{OH})_3(\text{s})$, precipitation, CaCO_3 can precipitate, which leads to a pH decrease. Furthermore, because $\text{Fe}(\text{OH})_3$ is amphoteric, some $\text{Fe}(\text{OH})_4^-$ formation is likely at pH g 8.5, which is a possible reason for a small decrease in pH as OH^- ions are consumed to form iron hydroxide complexes (Lakshamanan et al., 2009; 2010).

From the observation during experiment the characterization of sludge deposited and analyzed for different oxyhydroxide generated and was identified as lepidocrocite (g- FeOOH) (Stumn and Morgan, 1996) by its XRD pattern. Also it will not be affected by water chemistry and by As(III) and As(V) in water, which is well reported by (Wan et al., 2011).

Conclusions

Since As (III) is more mobile and more toxic than As (V), it is advantageous to convert As (III) to As(V). Also due to poor As(III) removal from water by many conventional processes, oxidation of As(III) to As(V) as the first step is recommended and then subsequent removal of As(V).

Electrocoagulation process was able to remove 99% of arsenic from arsenic contaminated water and met the drinking water standard of 10 µg/L with iron electrode. As(III) was more efficiently removed in electrocoagulation than conventional methods.

The current study has undergoing further modification for the feasibility of this process for arsenic removal in small villages. As electrocoagulation process is very cheap and simple can be use in any household in developing countries like India, Bangladesh and other parts of world, Due to its robustness and convenience in preparation EC filter may become a viable and cost-effective alternative as a filter medium for arsenic removal from contaminated drinking water.

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