

Article



Arsenic Removal from Highly Contaminated Groundwater by Iron Electrocoagulation—Investigation of Process Parameters and Iron Dosage Calculation

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Abstract: Electrocoagulation (EC) is gaining increased attention for water treatment as it efficiently removes various water contaminants. Therefore, EC was applied to remove arsenic from ground-water of a highly contaminated site in Hamburg, Germany. Groundwater containing 3250 and 14,600 μ g/L arsenic, mainly as Arsenite (As(III)), was treated in three different EC batch reactors using a monopolar parallel electrode-configuration. This study focused on iron EC with constant current densities and variable voltage, to investigate the influence of current density, surface to volume ratio, initial arsenic concentration and water volume on the removal of arsenic and the influences on the groundwater composition. Arsenic removal >99.9% was achieved for configurations with high iron dosage after four hours of EC treatment. German drinking water standard for arsenic (<10 μ g/L) was obtained after around two hours depending on the applied current densities. Arsenic removal efficiency shows independence from current density, surface to volume ratio, initial concentration and water volume, with respect to the calculated iron dosage. Consequently, the dimensioning and regime of efficient operation of the EC reactor for arsenic removal from groundwater can be calculated solely from the iron dosage determined by the applied current.

Keywords: arsenic removal; arsenic contamination; iron electrocoagulation; high arsenic concentration; iron dosage

1. Introduction

Groundwater is the world's most widely used source of drinking water [1] and needs protection and purification, especially in case of contamination. Arsenic in drinking water has been shown to be toxic, mutagenic and carcinogenic [2–4]. In addition to acute arsenic poisoning, the continuous consumption of small amounts of arsenic over a longer period of time leads to chronic diseases [2–5]. Inorganic trivalent arsenic (As(III)) is particularly toxic [2,5]. Based on this, the German Drinking Water Ordinance limits arsenic to <10 μ g/L consensual with the Word Health Organization guideline value [6,7].

Arsenic in groundwater is a severe problem worldwide, especially in densely populated Southeast Asian countries such as Bangladesh [8]. There is an urgent need for simple purification systems. Various techniques are suitable to eliminate arsenic from drinking water, which are thoroughly described in [9–11]: The simplest method is oxidation with atmospheric oxygen, but the effectiveness for the removal of arsenic is <30%. Adsorption processes and chemical coagulation show a removal for pentavalent arsenic (As(V)) of >90%, depending on the material. This method is widely used in developing countries but requires large amounts of coagulants. Despite ion exchange processes not being as effective as chemical coagulation, they are used for removing arsenic in practice. Membrane processes have proven to be very effective. Reverse osmosis in particular shows a removal



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). of up to 90% for As(V) but produces secondary waste [11]. The presence of trivalent arsenic limits the performance of these techniques or a pretreatment to convert As(III) to As(V) is necessary [10,11]. Additionally, these effective methods are expensive and often require complex equipment, chemicals and skilled workers to monitor the process. Often the methods also require pre- and/or post-treatment of the water, such as pH corrections, oxidation or the addition of chemicals (e.g., iron or aluminum salts).

An inexpensive, largely self-sufficient process could be more readily available to small, remote, and financially precarious communities and also simplify drinking water treatment in industrialized countries to make water purification more widely applicable. One promising low-tech and low-cost treatment technique for the elimination of dissolved arsenic from water is electrocoagulation (EC). It provides high removal of both As(III) and As(V) and due to its simple design offers the possibility of fully automatic operation. The efficiency of the in situ produced coagulants is higher than that of added chemical coagulants (e.g., FeCl₃) [10,12]. EC for arsenic removal typically uses either iron or aluminum as an electrode material. Iron electrodes were used in this work because aluminum is suspected to cause neurodegenerative diseases, such as Alzheimer's disease [7,13,14].

Figure 1 shows the relevant processes taking place during iron EC (reactions from [10,15]). The electrochemically driven process starts when a direct current (DC) is applied to the electrodes. Iron dissolves at the anode as a coagulant agent, while a formation of hydrogen gas occurs at the cathode [16]:

anode:
$$Fe(s) \rightarrow Fe^{2+}(aq) + 2e^{-}$$
 (1)

cathode:
$$2H^+(aq) + 2e^- \rightarrow H_2(g)$$
 (2)



Figure 1. Important reactions in iron EC for arsenic removal. $[Fe(OH)_3 \times AsO_4^{3-}](s)$ is representative for the various iron-arsenic-complexes.

According to Faraday's law, the amount of iron dissolved at the anode is equivalent to the amount of charge:

$$Q = I \times t = z \times F \times \frac{m}{M}$$
(3)

Here Q is the electric charge (C), I the applied current (A), t the time during which a constant current is applied (s), z the number of electrons involved in the oxidation (z = 2), F is the Faraday's constant (F = 96,485 C/mol), *m* is the mass of the dissolved iron (g) and *M*

the molecular weight of iron ($M_{Fe} = 55.85 \text{ g/mol}$). Equation (3) can be applied to calculate the concentration c_{Fe} of iron dissolved during EC in the water volume V:

$$c_{Fe} = \frac{I \times t \times M_{Fe}}{z \times F \times V}$$
(4)

The dissolved ferrous iron (Fe²⁺) is oxidized by dissolved oxygen and forms iron (Fe³⁺)-complexes, such as magnetite, goethite, iron hydroxide oxide, and lepidocrocite. In these arsenic is incorporated due to ligand exchange of the hydroxyl group [17,18]. Previous studies have shown the great potential of EC to remove various pollutants from synthetic, waste- and groundwater. It is an effective technique to remove heavy metals (e.g., Cr, Cu, Ni, Zn) [19–21], fluoride [5,22–24], nitrogen compounds [22,25,26] and arsenic [10,15,27,28].

Various process parameters were introduced for iron EC and their influence on arsenic removal and the EC process was investigated. The most important parameters are current density, surface-to-volume (S/V) ratio, initial arsenic concentration and charge load. Current density is often mentioned as a key parameter in EC and can be calculated by dividing the applied current by the anode surface, where the iron is dissolved. It is reported that up to a threshold, a higher current density causes a faster arsenic removal [5,11,15,29]. A smaller current density is supposed to have a higher efficiency with respect to the adsorbed amount of arsenic per iron added [15]. The ratio of the total surface of the electrodes to the treated water volume (S/V ratio) also affects the EC process in terms of runtime and charge load [15,18]. Additionally, the initial arsenic concentration impacts EC efficiency, as a higher initial arsenic concentration is proven to cause a longer EC runtime [15,30]. The charge load is important regarding EC as it is used to calculate the amount of iron added, but in practice the iron production deviates from the calculated amount [31].

The present study focuses on an inexpensive EC batch reactor with extremely high initial arsenic concentrations at a contaminated groundwater plume. The aim is to evaluate the influence of current density, S/V ratio and water volume for a robust set-up as well as to figure out a novel, simple way to calculate the necessary iron dosage to efficiently remove high arsenic concentrations.

2. Materials and Methods

Groundwater sampling and field experiments were performed at an industrial site in Billbrookdeich, Hamburg, Germany. The production of arsenopesticides, among other substances, in the past has been suspected as the reason for the groundwater contamination [32]. The site has a deep zoned arsenic contamination. The groundwater level is c. 3.15 m below the surface and the first aquifer ends c. 25 m below the surface. As(III) content made up c. 93.3–100% of total arsenic. Water samples with different concentrations were taken from different depths. The characteristic properties of the water are listed in Table 1.

The arsenic removal from the groundwater was performed in three different batch reactors: one specially made of plexiglas and two simple HDPE barrels were used (Figure S1). The treated volume was 2.15, 30.0 and 57.5 L (Table 2). Four structural steel (DIN 17100; ST-37-2) plates were monopolar parallels connected to the DC power source. For DC power supply a car battery (U = 12 V) was used in combination with a step-down converter (XL4016, AZDelivery). To prevent passivation of the electrodes, the polarization was reversed every 30 min [29,33]. The electrodes were placed in the water and the immersed dimension were 10 by 10 by 0.30 cm and 30 by 20 by 0.3 cm, respectively. 3D printed spacers guaranteed a constant inter-electrode distance of 1 or 2 cm (Table 2). Two small aquarium air pumps were used for improved oxygen supply and water mixing (AP512, Conrad Electronic SE, Hirschau, Germany). The effect of current density was investigated at 0.35, 0.49, 0.85 and 1.7 mA/cm² and was determined with a multimeter. EC operation time was between 60 and 240 min. The experiments were open to the atmosphere and were performed outside at temperatures between 10-15 °C. Electrodes were cleaned with fine sandpaper and dried before each experiment. A detailed overview of the different EC reactors and process parameters is given in Table 2.

| Characterization | Value | | |
|-------------------------------|--------------------|--|--|
| pН | 6.8–7.4 | | |
| Elect. Conductivity | 1040–1150 (μS/cm) | | |
| Oxygen | 1.92–3.58 (mg/L) | | |
| Temperature | 11.6–13.3 (°C) | | |
| Ca ⁽²⁺⁾ | 121–154 (mg/L) | | |
| Mg ⁽²⁺⁾ | 13.4–14.4 (mg/L) | | |
| Na ⁽⁺⁾ | 55.6–60.7 (mg/L) | | |
| K ⁽⁺⁾ | 8.63–9.64 (mg/L) | | |
| As (total) | 3250–14,600 (µg/L) | | |
| SO_4^{2-} | 172–177 (mg/L) | | |
| Cl- | 77.0–81.7 (mg/L) | | |
| F^- | 0.11–0.18 (mg/L) | | |
| HCO ₃ ⁻ | 249–297 (mg/L) | | |

Table 1. Characterization of the water used in the experiments. HCO_3^- was calculated from measured inorganic carbon.

Table 2. Specification of the used EC Reactors and experimental parameters.

| Specifications | Reactor 1 | Reactor 2 | Reactor 3 |
|--|-----------------------|-----------------------|-----------------------|
| Material | Plexiglas | HDPE | HDPE |
| Treated water volume (L) | 2.15 | 30.0 | 57.5 |
| Immersed electrode dimensions (cm) | 10 	imes 10 	imes 0.3 | 30 	imes 20 	imes 0.3 | 30 	imes 20 	imes 0.3 |
| One electrode effective surface (cm ²) | 209 | 1220 | 1220 |
| Distance between electrodes (cm) | 1 | 2 | 2 |
| Number of electrodes | 4 | 4 | 4 |
| Electrode surface over volume $1 (m^2/m^3)$ | 38.9 | 16.4 | 8.16 |
| Current density (mA/cm ²) | 0.35 | 0.49/0.86 | 0.49/1.72 |
| Applied current (A) | 0.15 | 1.2/2.1 | 1.2/4.2 |
| EC time (min) | 60 | 240 | 180/240 |
| Initial arsenic concentration (μ g/L) | 3250 | 12,700 | 11,400/14,600 |

 $\frac{1}{1}$ S/V ratio.

Water samples were taken at different time steps and on-site parameters pH, temperature, oxygen amount and electric conductivity were measured systematically while the power source was turned off (WTW-Multi 3420, with: WTW Sen-Tix[®] 940 P; WTW Tetra Con[®] 925 IDS; WTW FDO[®] 925, Weilheim, Germany) (Tables S1 and S2). Water samples were filtered through 0.2 μ m syringe filters and cation samples were stabilized with 150 μ L HCl per 15 mL of water. Cations and arsenic were analyzed with ICP-OES (ICP 720 ES, Agilent Technologies Inc., Santa Clara, CA, USA), anions with ion chromatography (ICS-1100, Dionex, Sunnyvale, CA, USA) and HCO₃⁻ was calculated from total inorganic carbon measurements (TOC-VCPH with ASI-V, SHIMADZU Corp., Kyoto, Japan).

3. Results and Discussion

All set ups at different current densities were operating at protective extra-low voltage of at maximum U = 6.9 V.

3.1. Effect on Water Geochemistry

EC can have an influence on the composition of the treated water. On-site parameters were systematically monitored for detailed information on the water quality and to evaluate the necessity of post EC treatment. It is reported that EC affects the water pH [22,29] and that higher current densities have a larger impact on water pH [5,29]. Nonetheless, even at a high applied current of 4.2 A, with visible cathodic hydrogen formation, no critical pH evolution was monitored. Water pH remained in a range between 6.8 < pH < 7.5 at any

time (Table S1) without detectable trends and within the limits specified by the German Drinking Water Ordinance and the World Health Organization [6,7].

Initial water temperature of 11.5–13.3 °C increased during EC treatment up to 15.8 °C (Table S1). It is reported, that the theoretical iron dissolution drops below 40% if the water temperature is <17 °C, which would limit EC from performing under such conditions [29]. Interestingly, no critical limitation of the EC process due to low temperatures was observed, as arsenic was effectively removed. The measured temperature increase over time can be caused by the ambient temperatures (~14 °C) or exothermic reaction of iron (Fe²⁺) oxidation [34]. This stability of EC against low temperatures is important for the treatment of natural waters in temperate climates.

Decreasing electric conductivity was monitored in every experiment. Starting at around 1150 μ S/cm, values dropped by up to 21% after 240 min of EC, indicating coprecipitation (Table S1). The main cations and anions showed a decrease solely in calcium (up to 40.1 wt %; 5.4% of total cation equivalents) and bicarbonate (up to 60.1 wt %; 20.1% of total anion equivalents) (Table S2).

The measured amount of dissolved oxygen indicates whether the injected air was sufficient to oxidize the dissolved iron in order to enhance the EC process [10,17]. Starting at 1.9–2.6 mg/L, the oxygen content increased in experiments with lower current densities (0.35, 0.49, 0.85 mA/cm²) of up to 7.1 mg/L and remained constant at a high current density (1.7 mA/cm²). Current density and the amount of oxygen can be adjusted in order to optimize the ongoing processes and avoid excessive production of iron (Fe²⁺).

3.2. Effect of Current Density on Electrocoagulation Time

Current density controls the operation time needed to remove a specific amount of dissolved arsenic [23,29]. The results of this study show an arsenic removal >99.9% for experiments that ran for 240 min and residual arsenic concentrations <10 μ g/L, with the exception of experiment 0.35 mA/cm²—2.15 L and 0.49 mA/cm²—57.5 L, which still contained 36.6 μ g/L and 601 μ g/L, respectively, at the end of each experiment (Figure 2).



Figure 2. Arsenic concentration reduction as a function of time. Experiments with 0.49 mA/cm²— 57.5 L and 0.35 mA/cm²—2.15 L were not run until $c_{As} < 10 \ \mu g/L$.

Current density is the dominant parameter which controls the process time of removing arsenic from water ($1.72 < 0.86 < 0.49 \text{ mA/cm}^2$), even at different water volumes and S/V ratios. The results show a faster arsenic removal to < 10μ g/L at higher current density (150 min), than at lower current densities (180, 240 min) at similarly high initial arsenic concentrations (Figure 2, detail). Even though under the conditions of constant current density of 0.49 mA/cm², a smaller volume of water and a higher S/V ratio shows faster arsenic removal (Figure 2, yellow cross vs. green square) which is consistent with other studies [15,18].

Confirming the result follow the Faraday's law whereby higher current densities results in higher release of iron per time. That is, with increasing current density, removal of a greater amount of arsenic is achieved in a shorter time.

3.3. Effect of Process Parameters on Arsenic Removal

EC is an effective treatment technique for arsenic removal. The efficiency for the applied process parameters can be evaluated by comparing residual arsenic concentrations as a function of the charge load or iron added [15]. The quantity of iron added is important for the arsenic removal. It is reported that the charge load or calculated amount of iron is not independent from current density and S/V ratio and can deviate from the actual quantity of iron produced [15,31].

However, Figure 3 shows the amount of iron added according to Faraday's law (Equation (4)) for each individual sample while Figure 4a illustrates the residual arsenic concentration as a function of iron added and Figure 4b shows a zoomed-in area of lower residual arsenic concentrations. Within a small difference in all experiments at high initial concentrations, the residual concentrations follow the same pattern and can be expressed by an exponential function. The fitted function considers the samples of all experiments:

$$c_{\rm As} = 16,963 \, [\mu g/L] \times 10^{-0.022 \, [L/mg] \times c_{\rm Fe}} + 5.8 \, [\mu g/L] \tag{5}$$



This implies that the process parameters current density, S/V ratio and water volume have no direct effect on the efficiency of arsenic removal by EC, with respect to the iron dosage. It suggests that the arsenic removal from the investigated groundwater in this study is only dependent on the quantity of iron added by the EC process. Furthermore, the removal of lower initial arsenic concentrations does not linearly depend on the amount



of iron added [30]. Even the residual arsenic concentrations of the experiment with a lower initial concentration of 3250 μ g/L (0.35 mA/cm²—2.15 L) is well described by Equation (5), when the initial arsenic concentration is matched to the function. Thus, it shows no influence of lower initial concentrations on the efficiency of removal process.



Figure 4. Residual arsenic concentrations as a function of iron added. (a) Exponentially fitted to all samples with high initial concentration. $\chi^2_{red} = 29$, fitted with Veusz 3.2 for minimal χ^2 . (b) Zoomed in for residual arsenic concentrations < 1000 µg/L. Experiment 0.35 mA/cm²—2.15 L is referenced to second axis.

This observation of an independent arsenic removal process enables a calculation of the required amount of iron to remove arsenic from this particular water at any individual initial arsenic concentration ($c_{As,Initial}$). If an iron threshold value ($c_{T,Fe}$) is set at a specific residual arsenic concentration, the required amount of iron can be calculated:

$$c_{Fe} = c_{T,Fe} + \frac{\log\left\{\frac{c_{As, \text{ Initial}} - 5.8 \ [\mu g/L]}{16,963 \ [\mu g/L]}\right\}}{0.022 \ [L/mg]}$$
(6)

Based on the results, the residual arsenic reaches < 10 μ g/L at around 200 mg/L iron dosage, which can be set as c_{T,Fe} (Figure 4b). Equation (6) yields a slightly underestimated prediction of the iron amount, caused by an overestimation of the initial arsenic concentration. This preliminary tool to predict the iron amount for an arsenic removal at this contaminated site can help to estimate the costs of an EC treatment and could be improved by incorporation of further data.

4. Conclusions

In this study, EC using iron electrodes for arsenic removal from highly contaminated groundwater was investigated. The batch experiments were performed with different operation parameters and showed a highly effective arsenic removal of up to >99.9% and met national and international drinking water standards of <10 μ g/L. The highest investigated current density was able to remove ~11,400 μ g/L arsenic within 135 min of EC. All monitored on-site parameters remained within an uncritical range. The residual arsenic concentrations with respect to the applied iron dosage followed the same exponential function and were not influenced by:

- Current density (0.35, 0.49, 0.86, 1.72 mA/cm²)
- S/V ratio $(38.9, 16.4, 8.16 \text{ m}^2/\text{m}^3)$
- Water Volume (2.15, 30.0, 57.5 L)

Initial concentration (3250–14,600 μg/L)

This enables a calculation of the iron amount needed to remove arsenic (As(III)) at various initial concentrations. This could be a useful tool also for other arsenic contaminated sites in the future. Further improvements could include a more detailed analysis on how the composition of groundwater affects the EC process.

Supplementary Materials: The following are available online at https://www.mdpi.com/2073-4 441/13/5/687/s1, Figure S1: Experimental set up: (a) Schematic experimental setup. 1: DC power source 2: Measuring sensors 3: Measuring device 4: Iron electrodes 5: Bubble stone 6: Air pump 7: EC cell. (b) Experimental setup of the Plexiglas cell. (c) Small HDPE cell with electrodes. (d) Large HDPE cell with electrodes., Table S1: Time steps of the on-site parameter measurements and the measured values., Table S2: Water sampling time steps and the analyzed composition.

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