

## Arsenic removal from drinking water by electrocoagulation using iron electrodes

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**Abstract**—Arsenic removal from drinking water was investigated using electrocoagulation (EC) followed by filtration. A sand filter was used to remove flocs generated in the EC process. Experiments were performed in a batch electrochemical reactor using iron electrodes with monopolar parallel electrode connection mode to assess their efficiency. The effects of several operating parameters on arsenic removal such as current density ( $1.5\text{--}9.0\text{ mA cm}^{-2}$ ), initial arsenic concentration ( $50\text{--}500\text{ }\mu\text{g L}^{-1}$ ), operating time ( $0\text{--}15\text{ min}$ ), electrode surface area ( $266\text{--}665\text{ cm}^2$ ), and sodium chloride concentrations ( $0.01$  and  $0.02\text{ M}$ ) were examined. The EC process was able to decrease the residual arsenic concentration to less than  $10\text{ }\mu\text{g L}^{-1}$ . Optimum operating conditions were determined as an operating time of  $5\text{ min}$  and current density of  $4.5\text{ mA cm}^{-2}$  at pH of  $7$ . The optimum electrode surface area for arsenic removal was found to be  $266\text{ cm}^2$  taking into consideration cost effectiveness. The residual iron concentration increased with increasing residence time, and maximum residual iron value was measured as  $287\text{ }\mu\text{g L}^{-1}$  for electrode surface area of  $266\text{ cm}^2$ . The addition of sodium chloride had no significant effect on residual arsenic concentration, but an increase in current density was observed.

Key words: Arsenic, Electrocoagulation, Drinking Water, Iron Electrodes, Iron Concentration

### INTRODUCTION

Arsenic is found in the atmosphere, soils, rocks, natural waters, and organisms [1]. The presence of elevated levels of arsenic in groundwater has become a major concern, especially in Bangladesh, India [2–5], and several other countries such as United States [6–9], China [10], Australia [11], Czech Republic [12] and New Zealand [13]. According to Human Development Report 2006 Beyond Scarcity: Power, Poverty and Global Water Crisis by the United Nations Development Programme, arsenic contaminated water creates risks for million of people in some countries including Turkey in the world [14]. Especially in Western Turkey, high arsenic concentrations in groundwaters have been found related to the dissolution of some minerals in the colemanite boron formations. The observed enrichment of arsenic in groundwaters also results from both hydrothermal and evaporitic conditions, with some redistribution of both elements during diagenesis, and rock/mineral water interaction [15–18].

The U.S. Environmental Protection Agency (USEPA) reduced the maximum contaminant level (MCL) for arsenic in drinking water from  $50\text{ }\mu\text{g L}^{-1}$  to  $10\text{ }\mu\text{g L}^{-1}$  [19]. According to the last edition of the World Health Organization (WHO) Guidelines for Drinking-Water Quality (2006),  $10\text{ }\mu\text{g L}^{-1}$  was established as a provisional guideline value for arsenic. MCL was also lowered to  $10\text{ }\mu\text{g L}^{-1}$  in Turkey by Turkish Standards 266-Water Intended for Human Consumption.

Arsenic can occur in the environment in several oxidation states, but in natural waters is mostly found in inorganic form as oxyanions of trivalent arsenite [As(III)] or pentavalent arsenate [As(V)]. The relative concentrations of each are controlled by the redox poten-

tial (Eh) and pH [20]. Organic arsenic forms may be produced by biological activity, mostly in surface waters, but are rarely quantitatively important [1]. The forms of arsenic present are dependent on the type and amounts of sorbents, pH, redox potential, and microbial activity [9]. Arsenate is relatively easy to remove from water, since it bears a negative charge in natural waters above pH 2.2, and is electrostatically attracted to the positive charge on metal hydroxide surfaces [21]. Under reducing conditions at pH less than about pH 9.2, the uncharged arsenite (As(III)) species will predominate [1]. Therefore, As(III) is less efficiently removed than As(V), so pre-oxidation is necessary for better removal [22].

A variety of treatment processes have been developed for arsenic removal from water, including chemical precipitation or coagulation, adsorption, lime softening, ion exchange, and membrane separation [23–29]. These methods are mainly effective for arsenate removal. Although precipitation–coprecipitation with ferric and aluminum salts is one of the conventional methods for arsenic removal, handling and disposal of the waste sludge is a significant problem of this process [30]. These treatment technologies take considerable time, require an extensive set-up, and are not economically applicable in small community systems. These processes generate a considerable quantity of secondary pollutants (chloride, sulfate in the coagulation-precipitation) and large volumes of sludge or waste which pose serious environmental problems [31]. Since chemical coagulation consists of three stages, including rapid mixing, slow mixing, sedimentation, and/or filtration, it requires extensive land usage. Continuous chemical addition and mixing of chemicals in this process causes additional operating cost and chemical handling problems [32]. In addition, the usage of different types of oxidants for oxidation of arsenite to arsenate for better removal efficiency increases the amount of used chemicals and operation costs significantly. Moreover, these reagents decrease the water quality because

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of the residues and by-products formed [33].

Recently, EC has had a more prominent role in drinking water treatment because it provides some significant advantages, such as being quite compact and having easy operation and automation, no chemical additives, high velocities and reduced amount of sludge [34,35]. Therefore, EC is an alternative to using chemical coagulants for arsenic removal, and thus may be beneficial for communities with better access to electricity than to chemicals. EC systems supply in-situ generation of coagulant [36–38]. Moreover, EC is the most promising process for arsenic removal due to high arsenic removal efficiency, feasible for small scale requirements, cost-effective, and no need for handling chemicals [39]. Because EC systems use a sacrificial anode generally made of iron or aluminum rather than corrosive iron or aluminum salt solutions, EC units can be constituted as a portable plant for use in rural, remote, and small residential areas [40]. Several previous studies have reported arsenic removal from water and wastewater by electrocoagulation [32–34, 36,37,39–41]. When a direct current is applied between two electrodes, metal ions such as  $\text{Fe}^{2+}$  and  $\text{Al}^{3+}$  that can contribute to coagulant formation are released by anode oxidation. With iron electrodes, the  $\text{Fe}^{2+}$  released can subsequently be oxidized in solution to produce an  $\text{Fe(III)}$  hydroxide or oxyhydroxide [36,38]. In the EC process, the electrodes are consumed and the coagulant is generated and precipitated. No liquid chemical is added, alkalinity is not consumed, and pH adjustment is not needed [40].

The major objective of this study is to investigate removal efficiencies of arsenate from tap water by EC process followed by filtration using iron electrodes and to reduce the energy and electrode consumption by shortening the treatment time for arsenic removal, increasing at the same time the knowledge about the mechanisms involved in the process. A sand filter was used to remove flocs generated in the EC process. The effects of important operating parameters such as initial arsenic concentrations, electrolysis time, current density, electrode surface area, and sodium chloride concentrations on percent removal of arsenate were investigated.

## MATERIAL AND METHODS

### 1. Materials

The chemical composition and some properties of the tap water used in this study are listed in Table 1. Analytical reagent chemi-

**Table 1. Chemical and physico-chemical initial characteristics of the tap water used for the experiments**

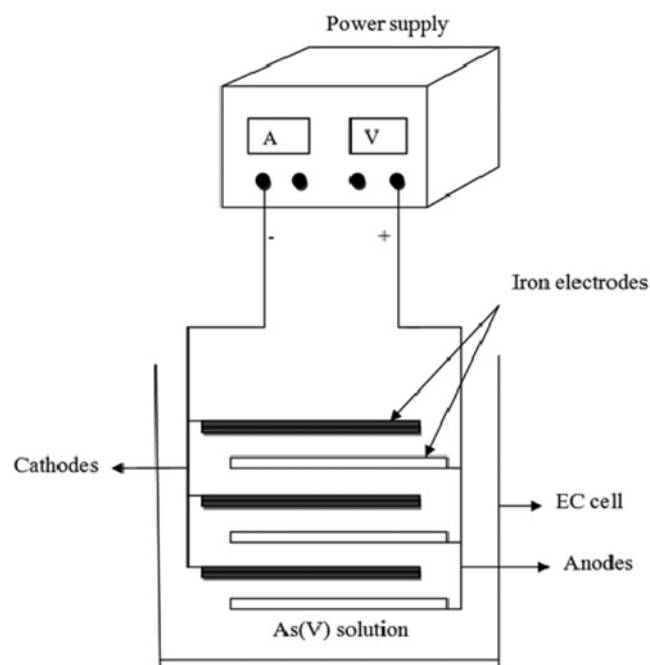
Components	Concentration
pH	8.2
Turbidity, NTU	0.1
Chloride, $\text{mg L}^{-1}$	46
Nitrate, $\text{mg L}^{-1}$	3
Iron, $\text{mg L}^{-1}$	0.0343
Aluminum, $\text{mg L}^{-1}$	0.012
Manganese, $\text{mg L}^{-1}$	0.0141
Sodium, $\text{mg L}^{-1}$	23
Conductivity, $\mu\text{S cm}^{-1}$	463
Sulfate, $\text{mg L}^{-1}$	36

cals and ultrapure grade water were used to prepare all stock solutions. Stock  $\text{As(V)}$  solution of  $2 \text{ mg L}^{-1}$  was prepared with deionized water by using the sodium arsenate ( $\text{Na}_2\text{HAsO}_4 \cdot 7\text{H}_2\text{O}$ ). Arsenic working solutions were freshly prepared by diluting arsenic solutions with deionized water. All measurements were made at ambient temperature ( $25 \pm 1^\circ\text{C}$ ). The solutions were stored at  $4^\circ\text{C}$  in the refrigerator. The pH of the arsenic-containing solutions was adjusted by adding aliquots of 1 M HCl or 1 M NaOH and kept nearly constant in the range of 6–8. To investigate the effect of salt on arsenic removal, 0.01 M NaCl ( $0.58 \text{ g L}^{-1}$  and  $1,400 \mu\text{S cm}^{-1}$ ) and 0.02 M NaCl ( $1.16 \text{ g L}^{-1}$  and  $2,500 \mu\text{S cm}^{-1}$ ) were added into the solutions.

### 2. EC Reactor and Experimental Procedure

The EC experiments were carried out in batch mode using a 2 L plastic reactor with horizontally placed iron electrodes spaced by 1.2 cm. Iron plates with the dimensions  $0.3 \text{ cm} \times 19 \text{ cm} \times 3.5 \text{ cm}$  were connected a DC power supply (0–9.2 V, 0–6 A) in monopolar parallel connection mode. A schematic of the experimental setup is shown in Fig. 1. Different numbers of electrodes (2–6) were used to determine the effects of electrode surface area. Before each experiment, the electrodes were abraded with sand paper to remove scales and then cleaned with 1 M HCl and ultrapure water. All the experiments were performed in duplicate to evaluate test reproducibility under identical conditions and the arithmetic average result of the two experiments was reported in this study.

In each run, 1 L of sample water containing various concentration of  $\text{As(V)}$  was added into the EC reactor. The anodes and cathodes were connected to the positive and negative outlets of a DC power supply. The current, residence time or voltage was held constant at desired values. The samples were taken from the EC reactor and allowed to settle for 30 min. Then the effluent was passed through a filter column filled with quartz sand of 0.2 mm particle size for the removal of suspended particles. Filtration rate was adjusted to  $7 \text{ mL min}^{-1}$ . An aquarium pump with a diffuser was used during



**Fig. 1. The diagram of the experimental equipment.**

the filtration process to provide oxidation of  $\text{Fe}^{2+}$  species generated by EC process to  $\text{Fe}^{3+}$  species. The finished water was analyzed for the residual concentrations of arsenic and iron.

### 3. Analytical Procedure

The arsenic in the influent and effluent aqueous solutions was determined by the hydride generation procedure coupled with inductively coupled plasma-atomic emission spectrometry (HG-ICP-AES), also known as inductively coupled plasma-optical emission spectrometry (HG-ICP-OES) (Optima 2100 DV). Hydride generation involves the production of volatile hydrides upon a chemical treatment with a strong reducing agent, typically sodium borohydride ( $\text{NaBH}_4$ ). The sodium borohydride instantaneously converts As(III) to arsine gas at room temperature, while the reduction of As(V) to arsine occurs relatively slowly. Therefore, a total arsenic determination requires a preductant such as KI to convert all arsenic to the +3 oxidation state prior to the arsine formation step [42]. In this study, the sample water (20 mL) was first mixed with 6 mL HCl (10%) and 2 mL of reducing agent (5% KI and 5% ascorbic acid), and allowed to react for 30 min at dark place to reduce As(V) to As(III). Then, 10 mL solution was taken for the analysis of As(III) concentrations. The iron concentrations were also determined by use of the HG-ICP-AES. Each analysis of arsenic and iron concentrations was duplicated. The arithmetic average of the two analyses results was reported in this study.

## RESULTS AND DISCUSSION

### 1. Effect of Electrolysis Time on Arsenic Removal

The effect of electrolysis time on the residual arsenic concentration was studied for a current density of  $4.5 \text{ mA cm}^{-2}$ , initial arsenic concentration of  $100 \mu\text{g L}^{-1}$ , and pH of 7. The time dependence of arsenic removal by EC process is shown in Fig. 2. It can be seen that the increase in residence time improved the arsenic removal efficiency. About 90% of the removal efficiency was achieved within 3 min of the process and residual As(V) concentration was reduced to  $10 \mu\text{g L}^{-1}$ , which is the recent guideline value of WHO.

This can be explained by the main anode and cathode reactions that occurred in the EC process with iron electrode. The anode and cathode reactions can be described as follows [39,40]:

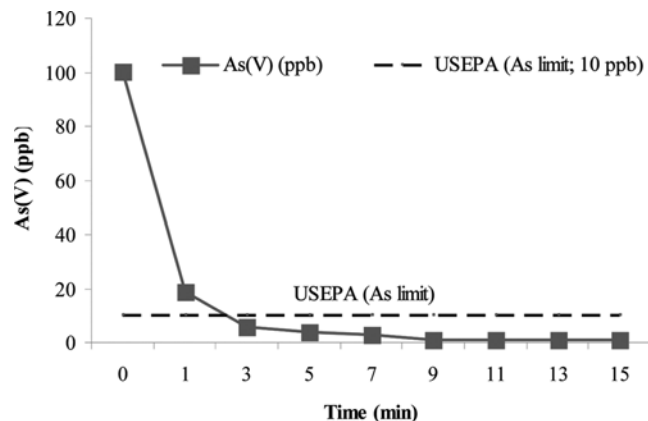


Fig. 2. Effect of residence time on arsenic removal efficiency (initial arsenic concentration of  $100 \mu\text{g L}^{-1}$ , current density of  $4.5 \text{ mA cm}^{-2}$ ).

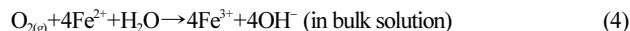
Anode reactions:



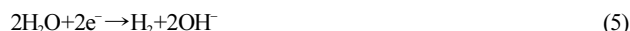
In the above two-step process, iron is first oxidized to ferrous ion, and then oxidized to ferric ion. The single step oxidation of iron to ferric ion can be shown as follows [39,40]:



The second step of iron oxidation reactions can be accelerated by aeration [39]:



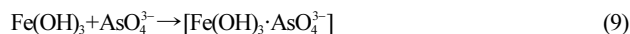
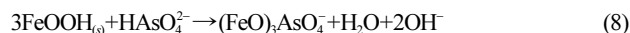
Cathode reaction [39,40]:



The  $\text{OH}^-$  ions in the cathode migrate towards the ferric ion for the formation of iron hydroxide [39]:



At the beginning of the process, arsenic removal was rapid and later it decreased gradually. Arsenic ions were more abundant at the beginning of the EC process, and the generated iron hydroxides due to corrosion of the anode at that time formed complexes with arsenic, and therefore rapid removal of arsenic was observed [43]. Coprecipitation of arsenic with occurred iron hydroxides is expressed in the following equations [39,40]:



However, as the experiment proceeded, the aqueous phase arsenic concentration continued to be reduced and simultaneously hydrous ferric oxides concentration increased [43]. When the initial arsenic concentrations were higher, more iron oxides and electrolysis time were needed to decrease the arsenic concentrations. Therefore, optimum electrolysis time was determined as 5 min. During the arsenic removal by EC process using iron electrodes, although a similar trend was observed by other researchers [32,34,36,40], the shortest electrolysis time was achieved for reducing arsenic concentration below  $10 \mu\text{g L}^{-1}$  in this study. Because the consumption amount of electrical energy determines the cost of the process, obtaining the shortest operation time reduces the cost of the process.

### 2. Effect of Current Density on Arsenic Removal

In all electrochemical processes, current density is one of the important parameters among the various operating variables for controlling the reaction rate within the electrochemical reactor, and it strongly influences the performance of electrocoagulation [44,45]. It is well known that current density determines coagulant dosage rate, bubble production rate, size and growth of flocs, which can influence removal efficiency of the pollutions in the EC process with different electrode configurations [31,44].

Faraday's law describes the relationship between current density and the amount of anode material that dissolves in the solution. It is given as

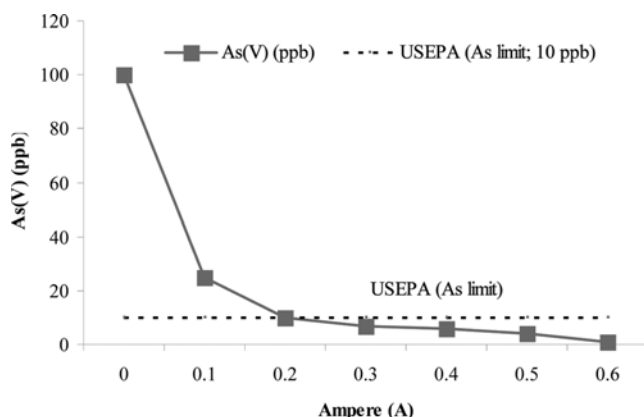


Fig. 3. Effect of current on arsenic removal efficiency (initial arsenic concentration of  $100 \mu\text{g L}^{-1}$ , electrolysis time of 5 min).

$$m = \frac{M \times i \times t_{EC}}{Z \times F} \quad (1)$$

where,  $m$  (kg Fe electrode per  $\text{m}^3$  treated potable water) is the theoretical amount of ion produced per unit surface area by current density  $i$  passing for a duration of time  $t_{EC}$  (s).  $Z$  is the number of electrons involved in the oxidation/reduction reaction; for Fe,  $Z=2$ .  $M$  is the atomic weight of anode material, for Fe,  $M=55.85 \text{ g mol}^{-1}$ ; and  $F$  is the Faraday's constant ( $96.486 \text{ C mol}^{-1}$ ) [46].

Generally, metal removal increases with increasing iron dosages in chemical coagulation. Therefore, arsenate removal by EC is governed by the formation of metal-hydrous ferric oxide complexes [46]. Higher current is expected to generate larger amounts of iron, which in turn will trap more pollutant and enhance removal efficiency [47]. Therefore, the effect of current density on the removal efficiency of As (V) was studied at  $1.5\text{--}9.0 \text{ mA cm}^{-2}$  at an initial concentration of  $100 \mu\text{g L}^{-1}$  at pH 7.0 for 5 min of operation time. Fig. 3 illustrates the effect of the current density on the arsenate removal efficiency. The results showed that as current density increases, the removal of arsenate also increases. This is consistent with the results reported by several authors [48,49]. The rate of reduction in the arsenic concentration was almost sharp in the early stages of the process and decreased to gradual reduction in later part of electrolysis. This behavior was explained as follows: as the current density increased, the oxides and hydroxides dosages increased too, favoring the arsenic removal [44]. The amount of solubilized iron increases with the current density because of Faraday's law [45]. The arsenic removal efficiencies were changed in the range of 75–99% and residual As(V) concentration was reduced to  $10 \mu\text{g L}^{-1}$  at current density of 0.3 A or  $4.5 \text{ mA cm}^{-2}$ . Flores et al. (2013) showed that complete arsenate removal by EC process was obtained at a current density of  $5 \text{ mA cm}^{-2}$  [50]. Similarly, Zhao et al. (2011) also noted that the initial arsenic concentration of  $1 \text{ mg L}^{-1}$  can be reduced below the values of  $10 \mu\text{g L}^{-1}$  at current density of  $4 \text{ mA cm}^{-2}$  and within 40 min [34]. Some researchers showed that arsenic concentration can be decreased below the MCL at a low current density of  $0.25 \text{ mA cm}^{-2}$ , but it can require long electrolysis time such as 30 min [39,40].

However, the cost of the process is determined by the consumption of the sacrificial electrode and the electrical energy. Although increasing current density and operating time enhances the efficiency

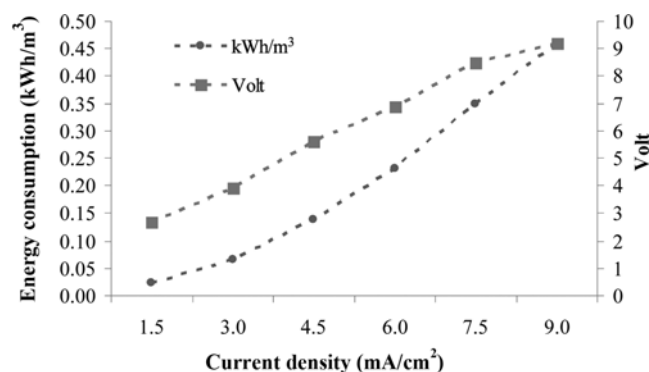


Fig. 4. The energy consumption at various current densities for EC.

of EC, it raises the cell voltage, energy consumption and operating costs [44]. The energy consumption is controlled by the applied current and voltage. The electrical energy required for the removal of arsenic was calculated by the following equation:

$$E = \frac{U \times I \times t_{EC}}{V} \quad (2)$$

where  $E$  is energy consumption ( $\text{kWh m}^{-3}$ ),  $U$  is voltage (V),  $I$  is ampere (A),  $t_{EC}$  is residence time (h), and  $V$  is volume of water in EC reactor ( $\text{m}^3$ ). Fig. 4 illustrates the energy consumption as a function of the current density and the cell voltage. As shown, the applied highest current density of  $9.0 \text{ mA cm}^{-2}$  caused the highest energy consumption; this is in part due to the increase in cell voltage, since when current density increases progressively from  $1.50$  to  $9.0 \text{ mA cm}^{-2}$ , cell voltage increases from  $2.7$  to values of  $9.2 \text{ V}$ . However, the optimum current density of  $4.5 \text{ mA cm}^{-2}$  obtained from the study produced a relatively small increase of energy consumption. In this study, the obtained sufficient arsenic removal efficiency was provided at current density of  $4.5 \text{ mA cm}^{-2}$  with energy consumption of  $0.45 \text{ kWh m}^{-3}$ . Although Flores et al. (2013) obtained the highest arsenic removal efficiency at almost the same current density, energy consumption of the system was calculated as  $3.9 \text{ kWh m}^{-3}$  because of long electrolysis time [50].

Residual arsenate concentrations were decreased below  $10 \mu\text{g L}^{-1}$ , which is the maximum contaminant level of arsenic in drinking water determined by USEPA and WHO, for current density of  $4.5\text{--}9 \text{ mA cm}^{-2}$ . Beyond  $4.5 \text{ mA cm}^{-2}$  the removal efficiency remains almost constant. The increase in current density value from  $4.5$  to  $9.0 \text{ mA cm}^{-2}$  for arsenate removal also increases the power required to achieve the about same removal efficiency. Considering this cost factor, all further experiments were carried out at  $4.5 \text{ mA cm}^{-2}$ .

### 3. Effect of Initial Concentration on Arsenic Removal

To evaluate the effect of initial arsenate concentration, experiments were conducted at varying initial concentration from  $50$  to  $500 \mu\text{g L}^{-1}$ . Fig. 5 shows the effect of the initial arsenic concentration as a function of operating time. As can be seen, the initial arsenic concentration increased from  $50$  to  $500 \mu\text{g L}^{-1}$ , and the obtained highest removal efficiency increased from about 84 to 95%. In the EC process, the rate of arsenic removal is proportionate to the influent concentration. When the initial arsenic concentrations are higher, more iron oxides are needed to decrease the dissolved arsenic concentrations [36]. It appears that the rate of removal is sharp in the begin-

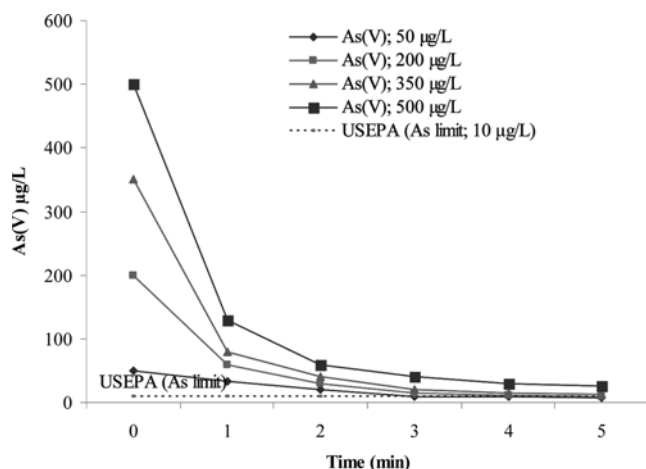


Fig. 5. The effect of initial arsenic concentration on the arsenic removal efficiency (electrolysis time of 5 min, current density of  $4.5 \text{ mA cm}^{-2}$ ).

ning of the EC process, and afterwards the slope of the curve decreases. As expected, the arsenic removal efficiency increases with the increase in initial arsenic concentration. This is because adsorption capacity onto metallic hydroxide flocs was limited, which is consistent with the results reported by several authors [31,44].

#### 4. Effect of Electrode Surface Area on Arsenic Removal

Design of EC cell is important to achieve maximum arsenic removal efficiency. The electrode area influences current density, in addition to bubble production and bubble path length [51]. In this study, different electrode surface areas were obtained by changing the electrode numbers. The effect of the electrode surface area was studied by adding electrodes to the reactor from  $n=2$  to  $n=6$ . To investigate the effect of the electrode surface area on arsenic removal, the EC process was operated with initial arsenate concentration of  $100 \mu\text{g L}^{-1}$ , pH of 7, and at a constant voltage of 9.1. Fig. 6 shows the residual arsenate concentration as a function of operation time for each electrode number used. As shown, the number of electrodes is reduced progressively from six to four and two when the required operation time to reduce arsenic concentration below  $10 \mu\text{g L}^{-1}$  is increased from about 2.5 min to over the five. This is associated with the increased electrode number generating larger amounts of iron; therefore, higher arsenate removal is produced. However, the cost of the process is determined by the consumption of the electrode besides the consumption of electrical energy. Therefore, the optimum number of electrodes was obtained from the study as two, taking into consideration cost effectiveness. Residual iron concentrations during the experiments are also shown in Fig. 6. The results demonstrated that after arsenate removal by EC process residual iron concentration increased with increasing residence time. Maximum residual iron values were measured as 287, 272, and  $246 \mu\text{g L}^{-1}$  for electrode numbers of two, four, and six, respectively. Because the maximum contaminant level of iron in drinking water is  $300 \mu\text{g L}^{-1}$  according to USEPA, all of electrode numbers are found effective and reliable considering residual iron and arsenate concentration.

#### 5. Effect of Sodium Chloride Concentration on Arsenic Removal

Usually, NaCl is added to increase electrolytic conductivity. In this study, the conductivity of the water was adjusted to the desired

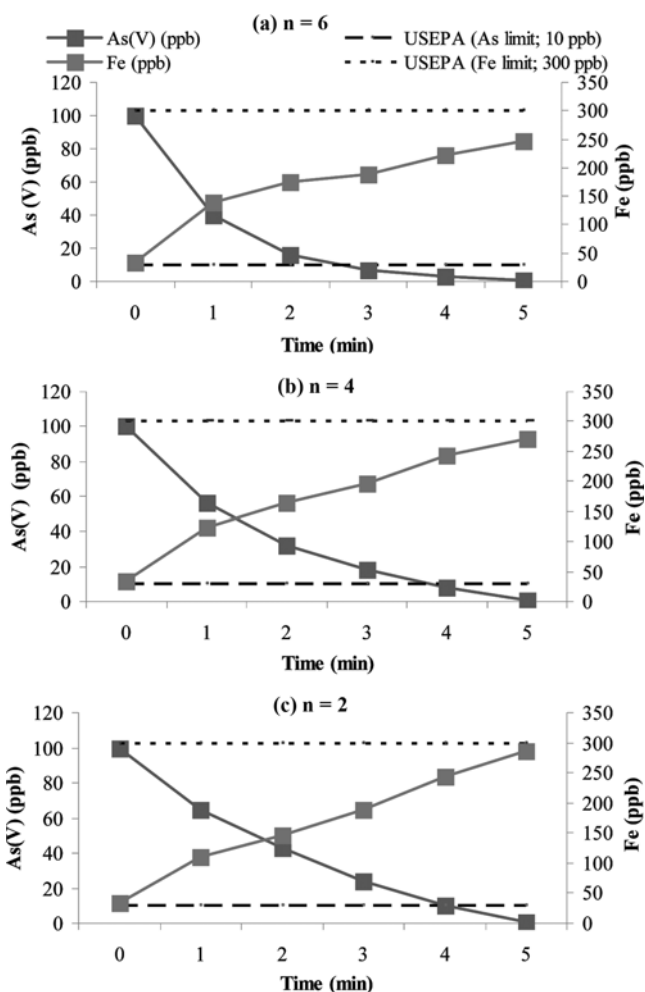


Fig. 6. Effect of electrode numbers on arsenic removal efficiency and residual iron concentration (a) electrode number=6, (b) electrode number=4, and (c) electrode number=2 (initial arsenic concentration of  $100 \mu\text{g L}^{-1}$ , electrolysis time of 5 min, current density of  $4.5 \text{ mA cm}^{-2}$ ).

levels by adding an appropriate amount of NaCl. When investigating the supporting electrolyte's influence on arsenic removal efficiency, a series of experiments were performed with different initial arsenic concentration and residence time as shown in Table 2.  $0.01 \text{ M NaCl}$  ( $1,400 \mu\text{S cm}^{-1}$ ) and  $0.02 \text{ M NaCl}$  ( $2,500 \mu\text{S cm}^{-1}$ ) were added into the solutions. Because the Turkish standard for conductivity of drinking water is given as  $2,500 \mu\text{S cm}^{-1}$ , the addition of NaCl was adjusted either less or equal to this limit value.

Ihos et al. (2005) added  $0.01 \text{ M NaCl}$  as supporting electrolyte, Hansen et al. (2007) added 10 g of NaCl into arsenic solution to increase electrical conductivity in the solution and to minimize the passivation of the anode [52,53]. Gomes et al. (2007) added 0.8 g NaCl into arsenic solution to avoid excessive ohmic drop and prevent the formation of the passivation layer on electrodes [54]. Larue (2003) reported that the addition of chloride salts decreased the energy consumption due to solution conductivity affecting the cell voltage and current efficiency [55]. Furthermore, more energy is required for overcoming a high ohmic resistance between an anode and a cathode when electrical conductivity of the solution is low. Bayramoglu et al. (2007) reported that increasing electrical conduc-

**Table 2. The effect of salt on arsenic removal efficiency (463  $\mu\text{S cm}^{-1}$ =no salt addition, 1,400  $\mu\text{S cm}^{-1}$ =0.58 g L<sup>-1</sup> salt addition, 2,500  $\mu\text{S cm}^{-1}$ =1.16 g L<sup>-1</sup> salt addition)**

Salinity ( $\mu\text{S cm}^{-1}$ )	Time (min)	Initial As concentration ( $\mu\text{g L}^{-1}$ )	Final As concentration ( $\mu\text{g L}^{-1}$ )	Current density (mA cm <sup>-2</sup> )	Efficiency (%)
1400	15	10	<1	4.80	>90.0
1400	15	500	<1	4.50	99.8
463	15	255	<1	3.30	99.6
463	9	10	2	1.35	80.0
1400	9	255	<1	6.30	99.6
463	9	500	<1	3.30	99.8
2500	9	500	<1	8.40	99.8
1400	3	500	<1	6.90	99.8
2500	3	255	<1	4.50	99.6
1400	3	10	<1	4.50	>90.0

tivity caused an increase in the current density at constant cell voltage, or a decrease in the voltage at constant current density [56].

The experiments showed that electrical conductivity was increased greatly with NaCl added. An increase in current density was observed, which was mentioned by Bayramoglu et al. It can be seen from Table 2 that the addition of salt (NaCl) had no significant effect on residual arsenic concentration. The standard value for the arsenic in drinking water given by WHO and USEPA was achieved at the end of every experiment.

### CONCLUSIONS

EC was evaluated as a remediation tool for arsenic removal from aqueous solutions in a batch EC reactor with iron electrodes. Batch EC studies were performed to evaluate the influence of various experimental parameters such as electrolysis time, initial arsenic concentration, current density, electrode surface area, and sodium chloride concentration on the removal of arsenic from drinking water. The EC process was able to decrease the residual arsenic concentration to less than 10  $\mu\text{g L}^{-1}$  (below the limit set by the WHO). The removal mechanism of arsenic by the EC process might be co-precipitation and adsorption with metal hydroxides generated in the process.

Optimum operating time and value of current density for arsenic removal were found to be 5 min and 4.5 mA cm<sup>-2</sup>, respectively. The increase in current density value from 4.5 to 9.0 mA cm<sup>-2</sup> for arsenate removal also increases the power required to achieve the about same removal efficiency. The results showed that the shortest electrolysis time of arsenic removal by EC process using iron electrodes was achieved. Accordingly, the calculated energy consumption of the system was caused to decrease the operation cost. These findings have importance for EC applications of arsenic removal from water and reveal that EC process is cost-effective and efficient arsenic removal method for future evaluations. The initial arsenic concentration increased from 50 to 500  $\mu\text{g L}^{-1}$ , the obtained highest removal efficiency increased from about 84 to 95%. The results showed that the rate of arsenic removal was proportionate to the influent concentration. The optimum electrode surface area for arsenic removal was found to be 266 cm<sup>2</sup> (n=2), taking into consideration cost effectiveness. The results demonstrated that residual iron concentration increased with increasing residence time and maximum

residual iron value was measured as 287  $\mu\text{g L}^{-1}$  for electrode surface area of 266 cm<sup>2</sup>. The addition of salt (NaCl) had no significant effect on residual arsenic concentration, but an increase in current density was observed. This study showed that electrocoagulation has the potential to treat arsenic-contaminated water to standards required for drinking water and can be considered a robust technology for arsenic removal.

### ACKNOWLEDGEMENT

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### REFERENCES

1. P. L. Smedley and D. G. Kinniburgh, *Appl. Geochem.*, **17**, 517 (2002).
2. M. A. Ali, *International Review for Environmental Strategies*, **6**, 329 (2006).
3. R. Bhattacharyya, D. Chatterjee, B. Nath, J. Jana, G. Jacks and M. Vahter, *Mol. Cell. Biochem.*, **253**, 347 (2003).
4. M. A. Halim, R. K. Majumder, S. A. Nessa, Y. Hiroshiro, M. J. Uddin, J. Shimada and K. Jinno, *J. Hazard. Mater.*, **164**, 1335 (2009).
5. C. F. Harvey, K. N. Ashfaq, W. Yu, A. B. M. Badruzzaman, M. A. Ali, P. M. Oates, H. A. Michael, R. B. Neumann, R. Beckie, S. Islam and M. F. Ahmed, *Chem. Geol.*, **228**, 112 (2006).
6. D. C. Cory and T. Rahman, *Ecol. Econ.*, **68**, 1825 (2009).
7. S. S. Farias, V. A. Casa, C. Vazquez, L. Ferpozzi, G. N. Pucci and I. M. Cohen, *Sci. Total Environ.*, **309**, 187 (2003).
8. A. M. Sancha, *Journal of Health Population and Nutrition*, **24**, 267 (2006).
9. S. Wang, and C. N. Mulligan, *Sci. Total Environ.*, **366**, 701 (2006).
10. T. Yuan, Q. Luo, J. Hu, S. Ong and W. Ng, *J. Environ. Sci. Heal. A.*, **38**(9), 1731 (2003).
11. S. J. Appleyard, J. Angeloni and R. Watkins, *Appl. Geochem.*, **21**, 83 (2006).
12. P. Drahota, J. Rohovec, M. Filippi, M. Mihaljevic, P. Rychlovsky, V. Cervený and Z. Pertold, *Sci. Total Environ.*, **407**, 3372 (2009).
13. J. Gregor, *Water Res.*, **35**, 1659 (2001).
14. B. Ross-Larson, M. Coquereaumont and C. Trott, (Eds.), *Human Development Report Beyond Scarcity: Power, Poverty and Global*

- Water Crisis*, Human Development Report, Palgrave Macmillan, Retrieved September 26, 2012, from <http://hdr.undp.org/en/media/HDR06-complete.pdf> (2006).
15. M. Çolak, Ü. Gemici and G. Tarcan, *Water Air Soil Poll.*, **149**, 127 (2003).
  16. M. Çöl and C. Çöl, *Hum. Ecol. Risk Assess.*, **10**, 461 (2004).
  17. M. Doğan and A. U. Doğan, *Environ. Geochem. Hlth.*, **29**, 119 (2007).
  18. Ü. Gemici, G. Tarcan, C. Helvacı and A. M. Somay, *Appl. Geochem.*, **23**, 2462 (2008).
  19. Y. Lee, I. Um and J. Yoon, *Environ. Sci. Technol.*, **37**, 5750 (2003).
  20. K. Moore, *Treatment of arsenic contaminated groundwater using oxidation and membrane filtration*, Master of Applied Science in Civil Engineering, University of Waterloo (2005).
  21. R. Johnston, H. Heijnen and P. Wurzel, *United Nations synthesis report on arsenic in drinking water; chapter 6: safe water technology* (2001), Available at [www.cepis.ops-oms.org/bvsacd/who/arsin.pdf](http://www.cepis.ops-oms.org/bvsacd/who/arsin.pdf).
  22. M. Fujimoto, *The Removal of arsenic from drinking water by carbon adsorption*, Master of Science, Department of Civil and Environmental Engineering, Michigan State University (2001).
  23. S. R. Wickramasinghe, B. Han, J. Zimbron, Z. Shen and M. N. Karim, *Desalination*, **169**, 231 (2004).
  24. E. O. Kartinen and C. J. Martin, *Desalination*, **103**, 79 (1995).
  25. T. Viraraghavan, K. S. Subramanian and J. A. Aruldoss, *Water Sci. Technol.*, **40**, 69 (1999).
  26. U. S. EPA, *Arsenic treatment technologies for soil, waste, and water*, U.S. EPA/National Service Center for Environmental Publications, Cincinnati (2002).
  27. A. Zouboulis and I. Katsoyiannis, *Sep. Sci. Technol.*, **37**, 2859 (2002).
  28. B. An, T. R. Steinwinder and D. Zhao, *Water Res.*, **39**, 4993 (2005).
  29. M. M. Gholami, M. A. Mokhtari, A. Aameri and M. R. A. Fard, *Desalination*, **200**, 725 (2006).
  30. K. Banerjee, G. L. Amy, M. Prevost, S. Nour, M. Jekel, P. M. Gallagher and C. D. Blumenschein, *Water Res.*, **42**, 3371 (2008).
  31. M. Kobya, U. Gebologlu, F. Ulu, S. Oncel and E. Demirbas, *Electrochim. Acta*, **56**, 5060 (2011).
  32. E. Mohora, S. Roncevic, B. Dalmacija, J. Agbaba, M. Watson, E. Karlovic and M. Dalmacija, *J. Hazard. Mater.*, **235-236**, 257 (2012).
  33. E. Lacasa, P. Canizares, C. Saez, F. J. Fernandez and M. A. Rodrigo, *Sep. Purif. Technol.*, **79**, 15 (2011).
  34. X. Zhao, B. Zhang, H. Liu and J. Qu, *Chemosphere.*, **83**, 726 (2011).
  35. P. K. Holt, G. W. Barton, M. Wark and C. A. Mitchell, *Colloids Surf. A.*, **211**, 233 (2002).
  36. W. Wan, T. J. Pepping, T. Banerji, S. Chaudhari and D. E. Giammar, *Water Res.*, **45**, 384 (2011).
  37. P. R. Kumar, S. Chaudhari, K. C. Khilar and S. P. Mahajan, *Chemosphere.*, **55**(9), 1245 (2004).
  38. D. Lakshmanan, D. A. Clifford and G. Samanta, *Environ. Sci. Technol.*, **43**(10), 3853 (2009).
  39. M. Kobya, A. Akyol, E. Demirbas and M. S. Oncel, *Environmental Progress Sustainable Energy*, DOI:10.1002/ep.11765.
  40. X. Zhao, B. Zhang, H. Liu and J. Qu, *J. Hazard. Mater.*, **184**, 472 (2010).
  41. N. Balasubramanian and K. Madhavan, *Chem. Eng. Technol.*, **24**(5), 519 (2001).
  42. T. Guerin, N. Molenat, A. Astruc and R. Pinel, *Appl. Organomet. Chem.*, **14**, 401 (2000).
  43. P. R. Kumar, S. Chaudhari, K. C. Khilar and S. P. Mahajan, *Chemosphere.*, **55**, 1245 (2004).
  44. M. Kobya, F. Ulu, U. Gebologlu, E. Demirbas and M. S. Oncel, *Sep. Purif. Technol.*, **77**, 283 (2011).
  45. S. Vasudevan, J. Lakshmi and G. Sozhan, *Sep. Sci. Technol.*, **45**, 1313 (2010).
  46. K. Thella, B. Verma, V. C. Srivastava and K. K. Srivastava, *J. Environ. Sci. Heal. A.*, **43**, 554 (2008).
  47. N. S. Kumar and S. Goel, *J. Hazard. Mater.*, **173**, 528 (2010).
  48. J. F. Martinez-Villafane, C. Montero-Ocampo and A. M. Garcia-Lara, *J. Hazard. Mater.*, **172**, 1617 (2009).
  49. S. Chaudhari, P. R. Kumar, K. C. Khilar and S. P. Mahajan, *Chemosphere.*, **55**, 1245 (2004).
  50. O. J. Flores, J. L. Nava, G. Carreno, E. Elorza and F. Martinez, *Chem. Eng. Sci.*, **97**, 1 (2013).
  51. P. Holt, G. M. Barton and C. Mitchell, *Electrocoagulation as a wastewater treatment*, The Third Annual Australian Environmental Engineering Research Event, Sydney (2006).
  52. M. Ihos, A. Negrea, L. Lupa, M. Ciopec and P. Negrea, *Chem. Bull.*, **50**(64), 1 (2005).
  53. H. K. Hansen, P. Nunez, D. Raboy, I. Schippacasse and R. Grandon, *Electrochim. Acta*, **52**, 3464 (2007).
  54. A. G. J. Gomes, P. Diada, M. Kesmez, M. Weir, H. Moreno and J. R. Parga, *J. Hazard. Mater.*, **139**, 220 (2007).
  55. O. Larue, E. Vorobiev, C. Vu and B. Durand, *Sep. Purif. Technol.*, **31**, 177 (2003).
  56. M. Bayramoglu, M. Eyvaz and M. Kobya, *Chem. Eng. J.*, **128**, 155 (2007).