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Arsenite removal from groundwater in a batch electrocoagulation process: Optimization through response surface methodology

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ABSTRACT

In this study, influences of seven process variables such as initial pH (pH_i), applied current (i), operating time (t_{EC}), initial As(III) concentration (C_o), diameter of Fe ball anode (d_p), column height in the electrocoagulation (EC) reactor (h) and airflow rate (Q_{air}) for removal of As(III) from groundwater by a new air-fed fixed-bed EC reactor were evaluated with a response surface methodology (RSM). The proposed quadratic model fitted very well with the experimental data for the responses. The removal efficiencies and operating costs were determined to be 99% and 0.01 \$/m³ at the optimum operating conditions (a pH_i of 8.5, 0.05 A, 4.94 min, d_p of 9.24 mm, h of 7.49 cm, Q_{air} of 9.98 L/min for 50 µg/L). This study clearly showed that the RSM in the EC process was a very suitable method to optimize the operating conditions at the target value of effluent As(III) concentration (10 µg/L) while keeping the operating cost to minimal and maximize the removal efficiency.

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groundwater;
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Introduction

Arsenic contaminations in groundwater are a problem of global concern due to the results of natural and/or anthropogenic sources from several parts of the world.^[1] Anthropogenic sources of arsenic include various industrial activities, fossil fuel combustion, mining, smelting, land filling, pesticides, herbicides and fertilizers. Excess amount of arsenic in drinking water causes many health problems such as liver and skin cancers, black foot disease, diffused and spotted melanosis, diffused and spotted keratosis, Bowen's disease and gangrene.^[2,3] The World Health Organization (WHO)^[4] and the United States Environmental Protection Agency (US-EPA)^[5] have reduced the maximum contamination level in drinking water from 50 to 10 µg/L due to the toxicity of arsenic.

The most common arsenic species in the natural waters are inorganic arsenate (As(V)) and arsenite (As(III)). As(III) species in the pH range of 4–10 are neutral in charge, while As(V) species are negatively charged. Therefore, the removal efficiency with any conventional technologies for elimination of arsenic from the groundwater is often much lower for As(III) than that for As(V).

Conventional treatment plants may employ several methods for removal of arsenic from water. Commonly used processes for the removal include coagulation/filtration^[6], lime softening,^[7] adsorption,^[8–10] ion exchange^[11] and membrane filtration.^[12] The coagulation/filtration technology is simple; only common chemicals are used, installation costs are small and can be easily applied to large water volumes. However, this technology has been found to be not as efficient for As(III), and pre-oxidation of As(III) to As(V) using some oxidizing chemical agents like chlorine, potassium permanganate and hypochlorite is necessary for a better removal efficiency.^[13] The lime softening is efficient to treat water containing arsenic with high hardness at pH >10.5, but it is required for pre-oxidizing of As(III), pH adjustment and high coagulant dose. The adsorption results in a low rate of removal efficiency for As(III). The ion exchange is also effective for the removal of As(V) except for the operating cost due to resin type and resin regeneration. Currently, the available membranes such as microfiltration, nanofiltration and reverse osmosis are more expensive than the other arsenic removal options because of high electrical consumption, relatively high capital and operating cost and the risk of membrane fouling. In recent years, electrocoagulation (EC) as an alternative treatment technology is potentially

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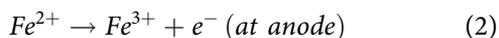
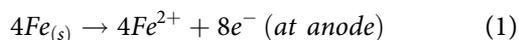
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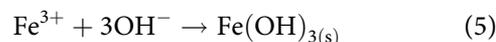
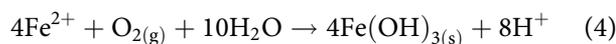
an effective tool for the treatment of water containing both As(III) and As(V). It could have the removal efficiency up to 93–99.9%.^[14–19] In an EC reactor, metals such as aluminium or iron are often used as sacrificial anode electrodes. When a direct current is applied, the sacrificial anodes dissolve, and metallic cations such as Al^{3+} , Fe^{2+} or Fe^{3+} are generated by the oxidation of the sacrificial anodes. Generally, the metallic cations released from the anode are gradually hydrolysed and spontaneously form a range of polymeric coagulant species or metal hydroxides that absorb or coprecipitate with the dissolved pollutants. Simultaneously, the bubbles at the cathode capture and float the suspended solids, resulting in additional removal of contaminants.^[14,15] Considering cost and removal efficiency in the EC process, sacrificial iron electrodes are advantageous and more efficient than aluminium anodes. Different electrode materials have different electrochemical characteristics, and appropriate electrode materials can improve treatment efficiency significantly. Plate and rod types of Al or Fe electrodes for the arsenic removal have some disadvantages namely, time consuming (changing and maintenance of the EC reactor) and accommodate a limited number of plate and rod types of electrodes with low surface areas. For this reason, the air-fed EC reactor using iron ball anodes was designed to eliminate the above problems.^[20,21] The new EC reactor has specifications of compactness, easy to use and accommodating more anode electrodes with higher surface areas. The objective of this study was to investigate combined effects of operating parameters on arsenite removal by the EC process in a batch mode operation. The Box-Behnken experimental design was applied to optimize the independent seven important operating variables such as pH, current density, operating time, anode ball size, initial As(III) concentration, column height in the EC reactor and air flow rate in order to increase the As(III) removal efficiency and minimize the operating cost.

Arsenite removal mechanism in the EC

When a charge applies through an external electrical power source, $\text{H}_{2(\text{g})}$ production occurs together with OH^- release at the cathode in the EC process. The main anode (Eqs. 1–2), cathode (Eq. 3) and hydrolysis (Eqs. (4–6)) were shown in the following equations:^[15]



When introducing oxygen to the process (Eq. (4)), Fe^{2+} is oxidized rapidly.^[22]



Atmospheric oxygen is the most readily available oxidizing agent and many treatment processes such as Fe (II) removals prefer oxidation by air. But, air oxidation of As(III) is a very slow process and can take weeks for oxidation to occur.^[23] As(III) removal mechanism in the EC process with Fe plate electrodes was reported to be oxidation of As(III) to As(V) and surface complexation with iron hydroxides.^[24] The generated H_2 gas at the cathode in the EC process helps both the flotation of flocculated particles and assured turbulence in the EC reactor. In the air-injected EC reactor using iron ball anodes, the drag force of the airflow towards the surface created turbulent conditions in the reactor, thus promoting the coagulation/flocculation process.^[20,25] As(III) oxidation followed with As(V) adsorption onto the metal hydroxides/oxyhydroxides occurs in As(III) removal by EC. As(III) is of higher mobility and solubility, and more toxic than As(V). Moreover, As(V) adsorption capacity is 3–20 times than that of As(III) in EC process. Therefore, pre-oxidation of As(III) and subsequent adsorption of As(V) are the best way to remove As(III) by EC.^[26]

Material and methods

Experimental

The groundwater (GW) obtained from a well situated in the province of Kocaeli in Turkey was stored in a 5-tonne high-density polyethylene container. All the chemical reagents were of analytical grade. The chemical analyses of the groundwater were determined according to Standard Methods.^[27] The characterizations of GW were determined as a pH of 7.6, electrical conductivity of 1055 $\mu\text{S}/\text{cm}$, dissolved oxygen (DO) of 6.86 mg/L, total alkalinity of 260 mg CaCO_3/L , total dissolved solids of 528 mg/L, total organic carbon of 4.6 mg/L, 24 mg/L of nitrate, 94.2 mg/L of sulphate, 10.2 mg/L of silicate, 127 mg/L of chloride, 0.12 mg/L of total iron, 0.006 mg/L of aluminium, 22 mg/L of sodium, 152 mg/L of calcium, 15 mg/L of magnesium, respectively. Sodium arsenite salt (NaAsO_2) was used to prepare stock arsenite solutions. The simulated groundwater solutions were prepared daily by dissolving sodium arsenite.

Construction details of the air-fed EC reactor were reported in the earlier study.^[20] The groundwater sample (0.95 L) containing As(III) was placed in the EC reactor. pH of the solutions was adjusted by adding either 0.1 N

NaOH or 0.1 N H₂SO₄. The anode and cathode were connected to a digital DC power supply (Agilent 6675A; 120 V and 18 A). An air-fed diffuser was attached underneath the reactor and the air was fed continuously at different rates in the reactor to maintain uniform shaking. The electrical current was adjusted to a desired value by the DC power supply and the experimental operation was started. The samples at different operating times taken from the EC reactor were filtered using a 0.45 μm millipore membrane and As(III) concentration was measured by inductive coupled plasma with optical emission spectrometer (PerkinElmer Optima 7000 DV ICP-OES). The detection limit of this study was 0.1 μgAs/L and analysis of the duplicates was within 2% of errors. All the experiments were repeated three times and the average data was reported.

Experimental design and statistical analysis

The Box-Behnken experimental design method was used to determine the effects of major operating variables on As(III) removal and to find the combination of variables resulting in maximum the removal efficiency. The main advantage of the design method is to reduce the number of experimental stages required to considerate multiple parameters and their interactions. The design expert software (trial version 10) is used for the statistical design of experiments and data analysis. Seven important operating parameters: initial pH (pH_i), applied current (*i*), EC time (*t*_{EC}), size of Fe ball anode (*d*_p), initial As(III) concentration (*C*_o), height of Fe anode in the reactor (*h*) and air flow rate (*Q*_{air}) were chosen as the independent variables and designated as *x*₁, *x*₂, *x*₃, *x*₄, *x*₅, *x*₆ and *x*₇, respectively. The ranges and levels of independent variables in the study are presented in Table 1.

The coded values of *x*₁, *x*₂ and *x*₃ in Table 1 were set as 3 levels: -1 (minimum), 0 (central) and +1 (maximum). Responses: effluent As(III) concentration (*y*₁: *C*_f), arsenic removal efficiency (*y*₂: *R*_e), energy consumption (*y*₃: *ENC*), electrode consumption (*y*₄: *ELC*), operating cost (*y*₅: *OC*) and arsenic adsorption capacity (*y*₆: *q*_e) in the EC process were determined with three dependent parameters. Results of the responses are presented in the supplementary materials (Tables S1 and S2).

FTIR spectra of groundwater containing arsenite

The FTIR (Perkin Elmer 100) spectra were recorded in the range of 4000–400 cm⁻¹. Two peaks were observed at 3300 cm⁻¹ from OH stretching and around 1637 cm⁻¹ from water bending vibration. One peak at 790–910 cm⁻¹ was corresponded to the As-O symmetric stretch. As

Table 1. Experimental range and levels of the independent process variables.

Experimental variables	Unit	Coded (<i>x</i> _{<i>i</i>})	Variables levels		
			-1	0	+1
Initial pH (pH _i)	(-)	<i>x</i> ₁	6.5	7.5	8.5
Applied current (<i>i</i>)	(A)	<i>x</i> ₂	0.05	0.10	0.15
EC time (<i>t</i> _{EC})	(min)	<i>x</i> ₃	1	3	5
Size of Fe ball anode (<i>d</i> _p)	(mm)	<i>x</i> ₄	5.0	7.5	10.0
Initial As (III) concentration (<i>C</i> _o)	(μg/L)	<i>x</i> ₅	20	35	50
Height of anode in the reactor (<i>h</i>)	(cm)	<i>x</i> ₆	2	5	8
Air-fed flow rate (<i>Q</i> _{air})	(L/min)	<i>x</i> ₇	2	6	10

(III)-O in H₃AsO₃ was observed at 796 cm⁻¹ for the EC operating time of 10th and 15th. As the EC process was continued, peak of As(III) disappeared due to its oxidation to As(V) and As(V)-O peaks in H₂AsO₄⁻ and HAsO₄²⁻ were observed at 841 cm⁻¹, 895 cm⁻¹ and 916 cm⁻¹. Lepidocrocite phase at 1024 cm⁻¹, magnetite at 570 cm⁻¹ and FeOOH stretching peak at 513 cm⁻¹ were observed (Fig. 1).

Results and discussion

The response surface methodology (RSM) modelling results

In the present study, the RSM was applied to investigate the effect of seven independent variables on the removal efficiency and operating cost of As(III) from the groundwater. The design method led to reduce the number of experiments and arranged them with various combinations of independent variables. The matrix for the removal efficiencies of As(III) is presented in Table S1. When the variables were in the range, the maximum removal efficiency and minimum operating cost at the effluent concentration of ≤10 μg/L were 82.37% and 0.015 \$/m³ for run 44, 92.51% and 0.021 \$/m³ for run 57, 80.20% and 0.021 \$/m³ for run

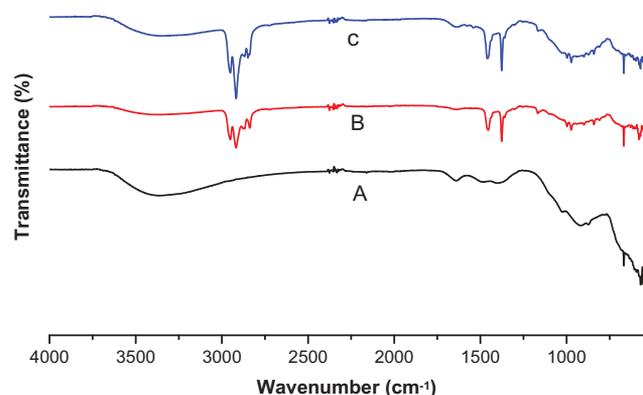


Figure 1. FTIR vibrations and their corresponding wavenumbers observed at different operating times (A: 1 min, B: 5 min and C: 3 min) for removal of As(III) from the groundwater in the EC process using Fe ball anodes.

55 in the concentration range of 20–50 $\mu\text{gAs(III)/L}$ (Tables S1 and S2). The removal efficiencies and effluent concentrations were 91.52% and 1.6 $\mu\text{g/L}$ for 20 $\mu\text{gAs(III)/L}$ at a pH_i of 7.50, 0.15 A, 5 min, d_p of 7.50 mm, h of 80 mm in the EC reactor and Q_{air} of 6.0 L/min (Tables S1 and S2).

Table 2 presents the response parameters obtained from analyses of variance (ANOVA). The quadratic model fitted very well to the experimental data due to the lowest value of standard deviation, the highest values of the correlation (R^2) and adjusted R^2 (Adj R^2) coefficients. The R^2 gives the proportion of the total variation in the response variables accounted for the predictors (x 's) included in the model. The values of R^2 and Adj R^2 were 0.913–0.964 and 0.796–0.916 for As(III) removals. Values of Adj R^2 represented high significance of the model. Only terms found statistically significant were included in the model. F -values for the removal efficiency, energy consumption, electrode con-

there was only a 0.01% chance that a “model F -value” could occur due to noise. The coefficient of variance (CV) is the ratio of the standard error of estimate to the mean value of observed response and considered reproducible when it is not $>10\%$. In this work, values of the CVs for the removal efficiency and operating cost were 18.40 and 27.96 which indicated good precision and reliability for the experiments. Adequate precision (AP) measures the signal-to-noise ratio, and the ratio >4.0 is desirable. For the present study, AP values for the removal efficiency and operating cost at low As(III) concentrations were 10.4 and 16.0 indicating an adequate signal. The model provided accurate description of the experimental data referring a successful correlation among seven independent parameters.

The quadratic regression model for predicting the optimum values of the removal efficiency and operating cost in terms of coded factors (x_1 – x_7) was presented in Eqs. 7 and 8 for low As(III) concentrations:

$$R_e(\%) = +685.09 - 145.91 \times \text{pH}_i + 449.47 \times i - 8.38 \times t_{EC} - 53.33 \times d_p + 3.80 \times C_o - 20.34 \times h + 17.39 \times Q_{\text{air}} - 73.84 \times \text{pH}_i \times i + 2.35 \times \text{pH}_i \times t_{EC} + 4.40 \times \text{pH}_i \times d_p - 0.311 \times \text{pH}_i \times C_o + 3.76 \times \text{pH}_i \times h - 0.34 \times \text{pH}_i \times Q_{\text{air}} - 34.31 \times i \times t_{EC} + 63.14 \times i \times d_p - 3.08 \times i \times C_o - 15.50 \times i \times h - 29.94 \times i \times Q_{\text{air}} - 0.206 \times t_{EC} \times d_p + 0.25 \times t_{EC} \times C_o - 0.158 \times t_{EC} \times h - 0.639 \times t_{EC} \times Q_{\text{air}} - 0.0065 \times d_p \times C_o - 0.872 \times d_p \times h - 1.014 \times d_p \times Q_{\text{air}} - 0.044 \times C_o \times h - 0.046 \times C_o \times Q_{\text{air}} + 0.371 \times h \times Q_{\text{air}} + 7.71 \times \text{pH}_i^2 + 822.71 \times i^2 + 0.068 \times t_{EC}^2 + 1.72 \times d_p^2 - 0.035 \times C_o^2 + 0.336 \times h^2 - 0.131 \times Q_{\text{air}}^2 \quad (7)$$

$$OC(\$/m^3) = -0.458 + 0.151 \times \text{pH}_i - 0.997 \times -0.0172 \times t_{EC} - 1.51 \times 10^{-3} \times d_p - 1.12 \times 10^{-3} \times C_o - 0.021 \times h + 0.013 \times Q_{\text{air}} - 0.022 \times \text{pH}_i \times i + 8.22 \times 10^{-4} \times \text{pH}_i \times t_{EC} - 6.82 \times 10^{-4} \times \text{pH}_i \times d_p + 1.10 \times 10^{-4} \times \text{pH}_i \times C_o - 1.70 \times 10^{-3} \times \text{pH}_i \times h - 1.84 \times 10^{-3} \times \text{pH}_i \times Q_{\text{air}} + 0.378 \times i \times t_{EC} - 3.16 \times 10^{-3} \times i \times d_p + 2.87 \times 10^{-3} \times i \times C_o + 0.024 \times i \times h + 0.023 \times i \times Q_{\text{air}} - 2.32 \times 10^{-3} \times t_{EC} \times d_p + 7.76 \times 10^{-6} \times t_{EC} \times C_o - 4.59 \times 10^{-4} \times t_{EC} \times h + 5.10 \times 10^{-4} \times t_{EC} \times Q_{\text{air}} + 4.87 \times 10^{-4} \times d_p \times C_o + 1.27 \times 10^{-3} \times d_p \times h - 7.16 \times 10^{-4} \times d_p \times Q_{\text{air}} + 1.55 \times 10^{-4} \times C_o \times h + 2.32 \times 10^{-5} \times C_o \times Q_{\text{air}} + 3.33 \times 10^{-4} \times h \times Q_{\text{air}} - 8.62 \times 10^{-3} \times \text{pH}_i^2 + 1.96 \times i^2 + 1.74 \times 10^{-3} \times t_{EC}^2 + 5.31 \times 10^{-4} \times d_p^2 - 2.01 \times 10^{-5} \times C_o^2 + 1.25 \times 10^{-3} \times h^2 + 2.26 \times 10^{-5} \times Q_{\text{air}}^2 \quad (8)$$

sumption and operating cost for 20–50 $\mu\text{gAs(III)/L}$ were 5.96, 19.91, 7.78 and 10.48, respectively. The large F -value indicated that most of the variation in the response was explained by the regression equation, and the terms in the model had a significant effect on the response. Values of $\text{Prob} > F$ for the responses were less than 0.0001 and the model was significant for low As(III) concentration. The p -values < 0.0001 meant that

Positive and negative signs in front of the terms refer to a synergistic effect and antagonistic effect, respectively. Negative effects were observed based on the interactions between x_1 and x_2 , x_5 , x_7 ; x_2 and x_3 , x_5 , x_6 , x_7 ; x_3 and x_4 , x_6 , x_7 ; x_4 and x_5 , x_6 , x_7 ; x_5 and x_6 , x_7 for the removal whereas x_1 and x_3 , x_4 , x_5 ; x_3 and x_5 ; x_6 and x_7 showed positive interaction effects on the removal (Eq. 7). In addition, similar trends were also obtained for the

Table 2. Analysis of ANOVA from the response surface quadratic model (y) for the removal of As(III).

Responses	R^2	Adj- R^2	S.D.	CV	PRESS	F-value	Prob> F	AP
y_1 : C_f ($\mu\text{g/L}$)	0.949	0.881	3.68	26.13	2071.2	13.84	<0.0001	15.2
y_2 : R_e (%)	0.889	0.740	11.49	18.40	20910.5	5.96	<0.0001	10.4
y_3 : ENC (kWh/m^3)	0.964	0.916	0.012	20.76	0.030	19.91	<0.0001	22.4
y_4 : ELC (kg/m^3)	0.913	0.796	0.003	32.93	0.002	7.78	<0.0001	13.4
y_5 : OC ($\text{\$/m}^3$)	0.934	0.845	0.016	27.96	0.050	10.48	<0.0001	16.0
y_6 : q_e ($\mu\text{g As/mg Fe}$)	0.891	0.731	1.380	33.27	340.94	5.75	<0.0001	12.7
q_e ($\mu\text{g As/C}$)	0.886	0.732	0.40	33.25	28.56	5.74	<0.0001	12.9

operating cost between x_1 and x_2 , x_7 ; x_2 and x_4 , x_7 ; x_5 and x_6 ; x_4 and x_5 ; x_5 and x_6 , x_7 ; x_6 and x_7 for the negative interactions, and x_1 and x_3 , x_4 , x_5 , x_6 ; x_2 and x_3 , x_5 , x_6 ; x_3 and x_4 , x_5 , x_7 ; x_4 and x_6 , x_7 for the positive interactions at 20–50 $\mu\text{gAs(III)/L}$ (Eq. 8). Moreover, effects of the independent variables for the removal at low concentrations evaluated with perturbation graph (Fig. 2). The perturbation graph provided interaction of the independent experimental variables. The highest effects for the removal efficiency and operating cost were observed with pH, operating time, column height and iron ball size at low concentrations. The removal efficiency also increased slightly with increase in current and airflow rate, and decreased with increase in concentration (Fig. 2).

Optimization of operating conditions

Numerical optimization is a function to evaluate the combination of all process parameters. The limits given in the range of each variables and responses provided by the surface and contour plots were used in the optimization procedure. The optimum operating conditions for the maximum removal efficiency and minimum operating cost for 20 $\mu\text{g/L}$ and 50 $\mu\text{g/L}$ in the

groundwater were determined from the model as follows:

- Optimization results at 50 $\mu\text{g/L}$ were C_f of 4.17 $\mu\text{g/L}$, R_e of 99%, ENC of 0.01473 kWh/m^3 , ELC of 0.00214 kg/m^3 , OC of 0.01 $\text{\$/m}^3$ and q_e of 8.53 $\mu\text{gAs/mgFe}$ or 2.47 $\mu\text{gAs/C}$ when the optimum operating parameters or variables were set as a pH_i of 8.50, 0.05 A, 4.47 min, d_p of 9.73 mm, h of 7.48 cm, Q_{air} of 7.04 L/min.
- Optimization results at 20 $\mu\text{g/L}$ were C_f of 0.34 $\mu\text{g/L}$, R_e of 99%, ENC of 0.01131 kWh/m^3 , ELC of 0.00211 kg/m^3 , OC of 0.01 $\text{\$/m}^3$ and q_e of 3.81 $\mu\text{gAs/mgFe}$ or 1.10 $\mu\text{gAs/C}$ when the variables were set as a pH_i of 7.9, 0.05 A, 4.71 min, d_p of 8.69 mm, h of 7.13 cm, Q_{air} of 8.53 L/min.

Dissolved oxygen concentration in groundwater was 6.86 mg/L at 0 L/min; values of DO decreased to 4.9 mg/L at 15 min and 3.6 mg/L at 20 min with increase in the operating time during the EC process. Values of DO increased from 6.86 to 8.1 mg/L with increase in values of Q_{air} from 0 to 6 L/min at 20 min for the removal of As(III) from groundwater whereas the value of DO without air decreased from 6.86 to 3.1 mg/L at the same operating time. Concentrations of DO increased with increase in flow rate as compared to no Q_{air} (Table S1).

Effects of independent variables

Effect of charge loading and pH on As(III) removal

Charge loading is the most important parameter for controlling the reaction rate within the EC reactor and it may serve as a design parameter for the process.^[28] The charges transferred in electrochemical reactions for a given amount of water treated are defined as charge loading which is calculated as the applied current multiplied by the operating time in the EC process:

$$q \text{ (C/L)} = \frac{i \times t_{\text{EC}}}{v} \text{ or } q \text{ (F/m}^3\text{)} = \frac{i \times t_{\text{EC}}}{F \times v} \quad (9)$$

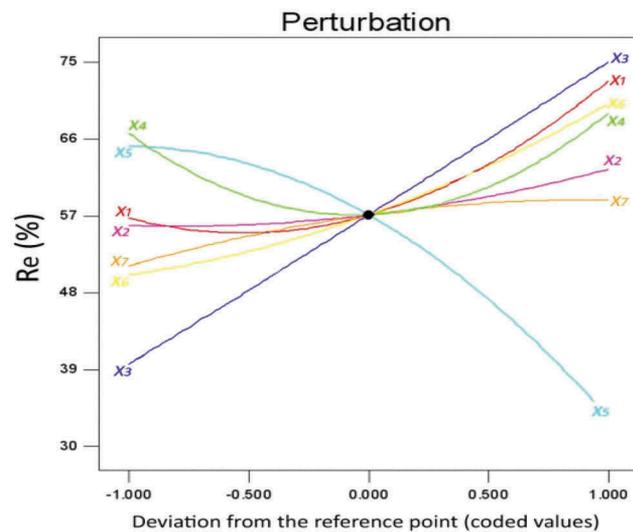


Figure 2. Perturbation plots for the removal efficiency at low As(III) concentrations with respect to the independent variables.

where q is the charge loading (C/L or F/m³ water), F is the Faraday's constant (1 F = 96487 Coulomb) and v is the solution volume (L or m³) in the EC reactor. In the EC process, the charge loading depends on values of t_{EC} and i . As t_{EC} and i increased, amount of anodic dissolution of electrodes increased (*i.e.*, dissolved amount of Fe or coagulant). Moreover, removed arsenic per unit adsorbent (q_e ; mg Fe of electrochemically generated in the EC process) increased from 2.56 $\mu\text{gAs/mgFe}$ (or 0.74 $\mu\text{gAs/C}$ at a pH_i of 6.5) to 5.02 $\mu\text{gAs/mgFe}$ (or 1.45 $\mu\text{gAs/C}$ at a pH_i of 8.5) at charge loading of 18 C (Tables S2 and S3). The average arsenic removed per C or mg Fe is called arsenic removal capacity and given in Eq. 10

$$RC = \frac{(C_o - C_t)}{q} \text{ or } RC = \frac{(C_o - C_t) \times v}{C_{Fe}} \quad (10)$$

where RC (μg removed As/C or μg removed As/mg Fe) is arsenic removal capacity, C_o and C_t are initial and at time t arsenic concentrations ($\mu\text{g/L}$) in solution and C_{Fe} (mg) is electrochemically dissolved iron concentration defined by

$$C_{Fe} = \frac{i \times t_{EC} \times M_{Fe}}{z \times F} \quad (11)$$

where M_{Fe} (55850 mg/mole) is the molecular weight and z is the number of electrons involved in the oxidation/reduction reaction ($z_{Fe} = 2$). Arsenic removed per charge or arsenic removal capacity were 0.93 $\mu\text{gAs/LC}$ at a pH_i of 6.5 (Table S3); it was calculated to be 1.82 $\mu\text{gAs/LC}$ at a pH_i of 8.5 showing that value of RC was increased twofold. Effluent arsenic concentration reduced to under 10 $\mu\text{g/L}$ at low current (0.05 A) and high pH_i (a pH_i of 8.5). Values of adsorption capacity were 4.18 $\mu\text{g As/mg Fe}$ or 1.21 $\mu\text{gAs/C}$ at a pH_i of 6.5 and 9.94 $\mu\text{g As/mg Fe}$ or 2.88 $\mu\text{g As/C}$ at a pH_i of 8.5 indicating that the removal efficiency increased with increase in pH_i and t_{EC} . The removal efficiency of As(III) increased from 38.91% at a pH_i of 6.5–92.51% at a pH_i of 8.5 for 0.05 A and from 84.37% at a pH_i of 6.5 to 95.45% at a pH_i of 8.5 for 0.15 A.

Final pH_f values were 6.95–7.95 for a pH_i of 6.5 and 8.82 for a pH_i of 8.5. The increased pH in the EC process was attributed to the formation of hydrogen gas at the cathode. As(III) removal efficiency increased also with decrease in the diameter of the anode material because total surface area of iron ball anodes increased (0.13188 m²). Similar results for q_e were obtained. As (III) removal efficiencies at 0.05–0.15 A for 20–50 $\mu\text{g/L}$ were 80.20% and 57.80% (runs of 10 and 55) and 44.20% and 38.32% (runs of 42 and 36) at the same experimental conditions, respectively (Table S1).

Effects of current and operating time on As(III) removal

Current and operating time are also two important parameters on performance of the EC process since these two parameters affect amount of coagulant generated electrochemically in the process.^[29,30] As(III) removal efficiency increased from 37.95% (C_f of 12.41 $\mu\text{g/L}$) to 62.25% (C_f of 6.95 $\mu\text{g/L}$) when the applied current increased from 0.05 to 0.15 A at 20 $\mu\text{gAs(III)/L}$ (runs of 27 and 29, conditions: a pH_i of 7.5, t_{EC} of 3 min, d_p of 7.5 mm, h of 5 cm, and Q_{air} of 2 L/min). In addition, values of charge loading and experimentally dissolved amount of iron increased from 9 C to 27 C and from 4.66×10^{-5} mole to 13.99×10^{-5} mole. The amounts of removed As per mg Fe or charge as Coulomb were calculated as 2.33 $\mu\text{g/mg}$ (or 0.67 $\mu\text{g/C}$) for 0.05 A and 1.34 $\mu\text{g/mg}$ (or 0.39 $\mu\text{g/C}$) for 0.15 A (Tables S2 and S3). As the current increased from 0.05 A to 0.15 A, the energy consumptions increased from 0.01528 to 0.10491 kWh/m³. The operating costs were also increased from 0.037 to 0.072 $\$/\text{m}^3$. As a result, an applied current of 0.15 A (0.072 $\$/\text{m}^3$) was required for removal of the groundwater containing 20 $\mu\text{gAs(III)/L}$ in the EC process. As seen in Table S1, effluent concentration of <10 $\mu\text{g/L}$ could not be met at any operating conditions for concentration >50 $\mu\text{g/L}$. Either increasing in the applied current (0.15 A) or the EC time (5 min) was adequate to make it possible. As (III) removal efficiencies at 1–5 min (Table S1) increased from 46.83% (C_f of 18.61 $\mu\text{g/L}$) to 95.91% (C_f of 1.43 $\mu\text{g/L}$) at 0.05 A and from 66.47% (C_f of 11.73 $\mu\text{g/L}$) to 94.08% (C_f of 2.07 $\mu\text{g/L}$) at 0.15 A (runs of 1 and 62, and 59 and 41). The above results showed that the removal efficiencies increased when both applied current and EC time (Fig. 3(a)) and pH_i (Fig. 3(b)) increased. However, 5 min of EC time was required to reach the recommended the effluent concentration. In this case, operating cost and amount of sludge at an operating time of 5 min were 0.034 $\$/\text{m}^3$ and 0.0188 kg/m³ for 0.05 A and 0.213 $\$/\text{m}^3$ and 0.0188 kg/m³ for 0.15 A (Tables S1 and S2). As the EC time and applied current increased, the arsenic adsorption capacity decreased (Fig. 4(a)) whereas the operating cost increased with the same operating parameters (Fig. 4(b)).

Comparison of arsenic removal by EC process

Table 3 presents comparison of this study with the other studies in the literature based on batch and continuous EC processes, water types (natural groundwater and synthetic water samples), arsenic species (As(III)

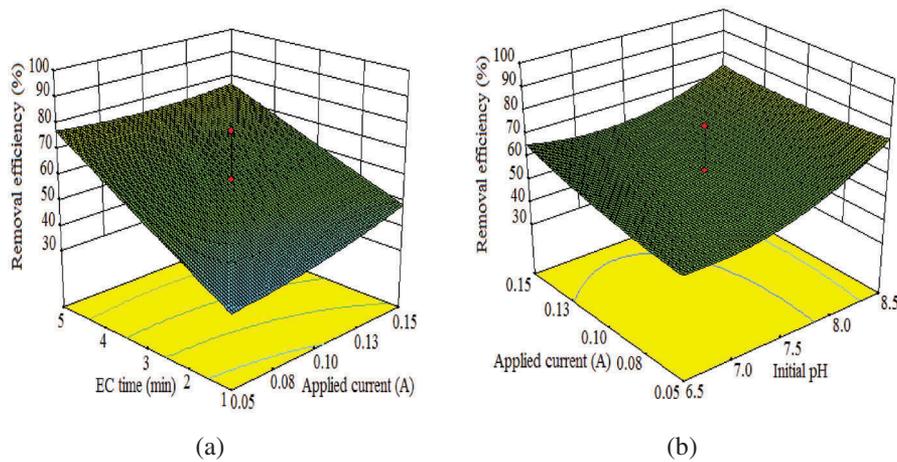


Figure 3. Three-dimensional contour plot for As(III) removal efficiency vs (a) i - t_{EC} and (b) i -pH_i variables.

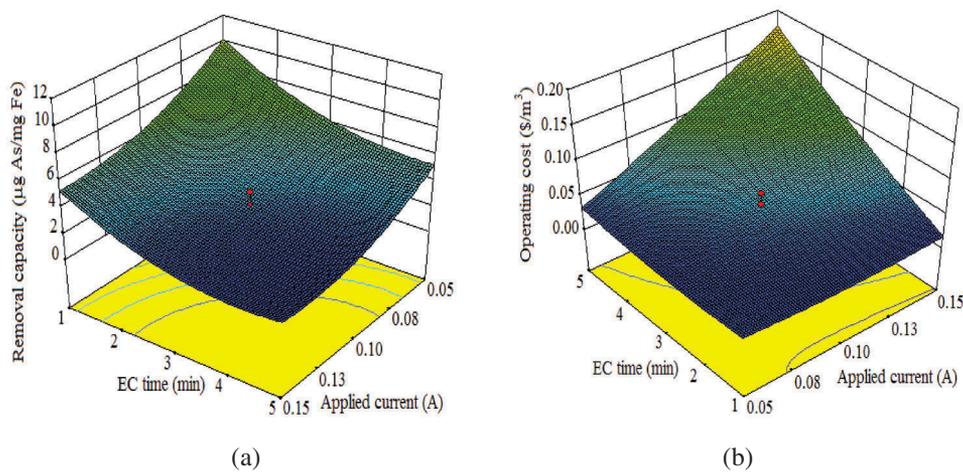


Figure 4. Effect of EC time and applied current on (a) As removal capacity and (b) operating cost.

and As(V)) and concentrations, electrode types (Al, Fe, Zn and Cu, etc., or/and hybrid combinations of the electrodes) and electrode shapes (plate, rod and ball).

It was obvious from Table 3 that optimization of the process conditions using Fe plate electrodes for the removal efficiency at different operating parameters was performed to meet the permissible WHO limit value. Generally, the removal efficiencies of 85.0–99.9% were observed for As (III) or As(V) concentrations in the range of 0.005–130 mg/L (Table 3). Arsenic removal efficiencies and operating costs from real natural groundwater samples containing arsenic concentration of 5.4–760 μg/L were 85.0–99.9% and 0.0020–1.04 \$/m³, respectively. For the groundwater samples, removal efficiencies of 97.6–99.9% and operating costs of 0.0020–0.22 \$/m³ at initial arsenic concentrations of 25–760 μg/L were obtained for continuous EC in a pilot scale using Fe plate electrodes.^[25,37] Treatments of synthetic and real groundwater samples using Fe ball anodes were reported in the literature.^[20,21] The removal

efficiencies and operating costs for both studies in a batch EC were >99.2% and 0.031–1.55 \$/m³ at 50–285 μgAs(V)/L. The results from the above study revealed that the removal efficiency and operating cost for the groundwater containing ≤ 50 μgAs(III)/L was 99% and 0.01 \$/m³ which satisfied the recommended WHO limit value. Operating costs for the arsenic removals meeting the permissible WHO value were 1.21 \$/m³ for coagulation-filtration, 1.20 \$/m³ for granulated ferric hydroxide/oxide, 3.20 \$/m³ for activated alumina, 3.40 \$/m³ for ion exchange, 3.72 \$/m³ for reverse osmosis and 0.054 \$/m³ for combined arsenic and iron removal by air oxidation-filtration, respectively.^[55] The operating cost for the removal in the EC process was calculated as 0.01 \$/m³ which was more economical compared to the literature values (Table 3). In addition, conventional removal techniques used for the removal of arsenic were presented in the introduction. EC turns out to be an environmentally friendly and effective method for the removal of arsenic

Table 3. Comparison of this study with the literature studies.

EC reactor type and water type	Electrode type	Optimum operating conditions	R_e (%)	References
BECR and SW	Fe-Al hybrid plate	$pH_i = 7, j = 0.47 \text{ A/dm}^2, t_{EC} = 20 \text{ min}, Q_{air} = 0.00\text{--}0.64 \text{ L/min}, C_o = 500 \mu\text{g/L As(III)}, \text{ and } OC = 0.0782 \text{ \$/m}^3$.	99.9	[31]
BECR and GW	Fe plate	$pH_i = 7.6, i = 0.3 \text{ A}, A_s = 285 \mu\text{g/LAs(V)}, t_{EC} = 6 \text{ min}, OC = 0.101 \text{ \$/m}^3$ (Fe plate),	96.9 (Fe plate)	[20]
	Fe ball	$t_{EC} = 20 \text{ min}, \text{ and } OC = 1.55 \text{ \$/m}^3$ (Fe ball).	99.3 (Fe ball)	
BECR and SW	Fe ball	$pH_i = 7.2, i = 0.5 \text{ A}, t_{EC} = 1.2 \text{ min}, C_o = 150 \mu\text{g/LAs(V)}, d_p = 5 \text{ mm}, h = 4.8 \text{ cm}, Q_{air} = 9.9 \text{ L/min}, \text{ and } OC = 0.031 \text{ \$/m}^3$.	99.2	[21]
CECR and GW	Al plate	$j = 6 \text{ mA/cm}^2, A_s = 134 \mu\text{g/L}, Q_w = 0\text{--}1 \text{ L/min}, u = 0.91 \text{ cm/s}, \text{ and } ENC = 1.19 \text{ kWh/m}^3$.	92.6	[32]
BECR (pilot) and GW	Fe plate	$q = 400 \text{ C/L}, U = 2.1 \pm 0.1 \text{ V}, A_s = 266 \pm 42 \mu\text{g/L}, \text{ and } OC = 0.84\text{--}1.04 \text{ \$/m}^3$.	>98.2	[33]
BECR or CECR and SW	Hybrid Fe-Al plate	$pH_i = 7, j = 2.5 \text{ A/m}^2, C_o = 150 \mu\text{g/LAs(V)}, Q_w = 0.05 \text{ L/min} (t_{EC} = 20 \text{ min}), t_{EC} = 1 \text{ min}$ (BECR) and 3 min (CECR), $q_e = 97.27 \text{ mg/g}, \text{ and } OC = 0.00202 \text{ \$/m}^3$ (BECR) and 0.0091 $\text{\$/m}^3$ (CECR).	>95.80 (BECR) 96.07 (CECR)	[34]
BECR and SW	Fe plate	$pH_i = 4, j = 0.54 \text{ mA/cm}^2, t_{EC} = 45 \text{ min}, C_o = 100 \text{ mg/L As(III)}, \text{ stirring speed} = 150 \text{ rpm}, \text{ and } ENC = 0.52 \text{ kWh/m}^3$.	>99.9	[35]
BECR and SW	Fe plate	$j = 5.64 \text{ A/m}^2, t_{EC} = 5 \text{ min}, C_o = 112.3 \mu\text{g/L As(III)}, ENC = 0.0156 \text{ kWh/m}^3, ELC = 0.0509 \text{ kg/m}^3, \text{ and } OC = 0.067 \text{ \$/m}^3$.	93.9	[36]
CECR (pilot) and GW	Fe plate	$j = 0.02\text{--}100 \text{ mA/cm}^2, t_{EC} = 5\text{--}90 \text{ min}, d = 3 \text{ cm}, A_s = 80\text{--}760 \mu\text{g/L}, \text{ and } OC = 0.22 \text{ \$/m}^3$	97.6–99.9	[37]
BECR and SW	Fe plate	$pH_i = 7, Fe^{3+} = 4 \text{ mg/L}, C_o = 100 \mu\text{g/LAs(V)}, \text{ operating time} = 26 \text{ h}, OC = 0.066 \text{ \$/m}^3$ (CC-MF: chemical coagulation-microfiltration). $pH_i = 7, d = 0.4 \text{ cm}, Fe^{3+}$ dosage = 4 mg/L (anodic), $t_{EC} = 16 \text{ h}, \text{ and } OC = 0.12 \text{ \$/m}^3$ (EC-MF).	97 (CC-MF) 95 (EC-MF)	[38]
BECR and SW	Hybrid Al, Fe or Zn plates	$pH_i = 5\text{--}8, C_o = 2\text{--}5 \text{ mg/L As(V)}, U = 3 \text{ V}, \text{ and } t_{EC} = 12 \text{ min}$ (Al-Fe pairs). $pH_i = 7, C_o = 2 \text{ mg/L As(V)}, U = 3 \text{ V}, T = 30 \text{ }^\circ\text{C}, \text{ and } t_{EC} = 10 \text{ min}$ (for Fe-Zn pairs).	99.9 (Al-Fe) 98.8 (Fe-Zn)	[39, 40]
CECR and GW	Al plate	$pH_i = 5, j = 5.78 \text{ mA/cm}^2, A_s = 5.4 \pm 16.4 \mu\text{g/L}, DOC = 9.31 \pm 0.51 \text{ mg/L}, d = 2.8 \text{ cm}, A/V = 0.248 \text{ cm}^2/\text{cm}^3, Q_w = 4.3 \text{ L/h}, \tau_{EC} = 62.8 \text{ min}, U = 17 \text{ V}, ELC = 0.066 \text{ kg/m}^3, \text{ and } ENC = 1.7 \text{ kWh/m}^3$.	85 (As) 77 (DOC)	[41]
BECR-1 and SW	Fe rod (BER-1)	For BECR-1: $pH_i = 7, C_o = 100\text{--}1000 \text{ mg/L As(III) or As(V)}, t_{EC} = 30\text{--}90 \text{ min}$ for As(III) and	90.1–99.1 (BECR-1)	[42]
BECR-2 and GW	Fe plate (BER-2)	15–45 min for As(V), $Q_{air} = 60 \text{ mL/min}, i = 22 \text{ mA}, U = 12 \text{ V}, \text{ and } ENC = 0.5 \text{ kWh/m}^3$. For BECR-2: $i = 2 \text{ A}, U = 12 \text{ V}, A_s = 449\text{--}677 \mu\text{g/L}, ENC = 0.72\text{--}0.78 \text{ kWh/m}^3, \text{ and } OC = 0.11 \text{ \$/m}^3$.	96.2–99.9 (BECR-2)	
BECR and SW	Fe or Al plate	$pH_i = 6.5$ (for Fe) and 7 (for Al), monopolar series electrode connection mode, $j = 2.5 \text{ A/m}^2, C_o = 150 \mu\text{g/L As(III)}, t_{EC} = 2.5 \text{ min}$ (for Fe) and 4 min (for Al), $OC = 0.0047 \text{ \$/m}^3$ (for Fe) and 0.0064 $\text{\$/m}^3$ (for Al).	94.1 (Fe) 93.5 (Al)	[15]
BECR and SW	Fe plate	$i = 3 \text{ A}, t_{EC} = 120 \text{ s}, C_o = 1.18 \text{ mg/L As(III)}, \text{ and } A = 57 \text{ cm}^2$.	98.56	[43]
BECR and GW	Fe plate	$j = 45 \text{ A/m}^2, t_{EC} = 6.5 \text{ min}, A_s = 133 \mu\text{g/L}, Q_r = 3 \text{ L/min}, Q_{air} = 1.5 \text{ L/min}, ENC = 0.606 \text{ kWh/m}^3, ELC = 0.113 \text{ kg/m}^3, \text{ and } q_e = 128.33 \mu\text{g/g}$	>92.5	[44]
BECR and SW	DSA	$pH_i = 7, C_o = 1000 \mu\text{g/LAs(III)}, j = 40 \text{ A/m}^2, \text{ and } t_{EC} = 40 \text{ min}$.	99	[45]
BECR and SW	Fe plate	$pH_i = 7, j = 0.2 \text{ A/dm}^2, t_{EC} = 15 \text{ min}, C_o = 0.5 \text{ mg/L As(V)}, q_e = 30.844 \text{ mg/g}$.	98.6	[16]
BECR and SW	Fe rod	$pH_i = 6.5, C_o = 0.05 \text{ mg/L As(III) and As(V)}, Q_r = 500 \text{ mL/min}, i = 28 \pm 2 \text{ mA}, t_{EC} = 20\text{--}30 \text{ min}, \text{ and } q_e = 1\text{--}12 \mu\text{gAs(III)/mg Fe or } 10\text{--}80 \mu\text{gAs(V)/mg Fe}$.	80–95	[22]
BECR and SW	Fe rod	$j = 5.2 \text{ mA/cm}^2, t_{EC} = 10 \text{ min}, \text{ Electrolyte} = 0.01 \text{ M NaCl}, C_o = 10 \text{ mg/L As(III) or As(V)}, \text{ and } q_e = 86.78 \text{ mg As/g Fe}$.	99.8	[46]
CECR and GW	Fe plate	$j = 45 \text{ A/m}^2, A_s = 133 \mu\text{g/L}, Q_w = 0.875 \text{ L/min}, \tau_{EC} = 1.6 \text{ min}, t_{EC} = 0.75 \text{ min}, ENC = 0.103 \text{ kWh/m}^3, \text{ and } ELC = 0.022 \text{ kg/m}^3$.	99	[17]
CECR and GW	Fe plate	$54 \text{ C/L}, t_{EC} = 240 \text{ min}, j = 1.98 \text{ A/m}^2, C_o = 38.15 \mu\text{g/L}, OC = 0.113 \text{ \$/m}^3$	96	[47]
BECR and SW	Fe or hybrid Al-Fe plate	$pH_i = 6\text{--}10, j = 0.48 \text{ A/dm}^2, t_{EC} = 10 \text{ min}, C_o = 10 \text{ mg/L As(III) or As(V)}, \text{ Fe-Fe and Al-Fe}$.	>99.9	[48]
BECR and SW	Al plate	$pH_i = 7, j = 10 \text{ A/m}^2, t_{EC} = 95 \text{ min}, d = 1 \text{ cm}, C_o = 550 \mu\text{g/L}, OC = 0.357 \text{ \$/m}^3$.	98.51	[49]
BECR and GW	Fe ball	$pH_i = 7.5, 0.3 \text{ A}, t_{EC} = 14 \text{ min}, d_p = 7.5 \text{ mm}, h = 5 \text{ cm}, C_o = 200 \mu\text{g/L}, Q_{air} = 6 \text{ L/min}, OC = 0.612 \text{ \$/m}^3$.	96	[50]
BECR and SW	Fe, Zn or Cu plate	$j = 1.5 \text{ mA/cm}^2, pH_i = 6.1\text{--}6.4, t_{EC} = 40 \text{ min}$ (for Fe or Zn anodes) and 60 min (for Cu-Zn and Cu anodes), $C_o = 68\text{--}130.5 \text{ mg/L As(V)}$.	93 (Fe or Zn) >73 (Cu-Zn) >67 (Cu)	[51]
CECR or BECR and SW	Fe plate (CECR) cylindrical Fe (BECR)	$C_o = 101 \text{ mg/L}, j = 1.2 \text{ A/dm}^2, A/V = 6.5 \text{ m}^2/\text{m}^3, Q_w = 3 \text{ L/h}, \tau_{EC} = 90 \text{ min}$ (CECR). $C_o = 105 \text{ mg/L}, j = 1.2 \text{ A/m}^2, t_{EC} = 120 \text{ min}, A/V = 6.5 \text{ m}^2/\text{m}^3$ (BECR).	>98	[18]
CECR (pilot) and GW	Fe plate	$i = 5 \text{ A}, U = 20\text{--}30 \text{ V}, A_s = 25\text{--}300 \mu\text{g/L}, pH_i = 7, \tau_{EC} = 1 \text{ min}, Q_w = 30 \text{ L/min}, \text{ and } OC = 0.002 \text{ \$/m}^3$.	99	[25]
BECR and SW	Fe plate	$pH_i = 7, j = 1.53 \text{ mA/cm}^2, t_{EC} = 60 \text{ min}, q = 100 \text{ C/L}, C_o = 2 \text{ mg/L As(V) or As(III)}, \text{ and } q_e = 150 \text{ mg As(V)/g Fe or } 250 \text{ mg As(III)/g Fe}$.	>99.5	[24]
BECR and SW	Al-Fe plate	$pH_i = 4, j = 30 \text{ mA/cm}^2, t_{EC} = 60 \text{ min}, C_o = 13.4 \mu\text{gAs(III)/L}$	99.6	[52]
BECR and SW	Al-Al plate	$pH_i = 4, j = 30 \text{ mA/cm}^2, t_{EC} = 60 \text{ min}, C_o = 13.4 \mu\text{gAs(III)/L}$	97.8	[53]
BECR and SW	Fe-Fe plate	$pH_i = 2.4, j = 30 \text{ mA/cm}^2, t_{EC} = 60 \text{ min}, C_o = 13.4 \mu\text{gAs(III)/L}$	99.6	[54]
BECR and SW	Fe plate	$pH_i = \geq 3, t_{EC} = 15\text{--}20 \text{ min}, C_o = 0.5\text{--}5 \text{ mg/L As(III)}, \text{ electrical gradient} = 200 \text{ V/m}, \text{ and } q_e = 0.45\text{--}90.91 \text{ mg/g Fe}$ (for $C_o = 0.5\text{--}100 \text{ mg/L As}$)	>99	
BECR and GW	Fe plate	$i = 3 \text{ mA/cm}^2, t_{EC} = 20 \text{ s}, d = 3 \text{ mm}, A_s = 131, A/V = 0.466 \text{ cm}^2/\text{cm}^3, U = 4.54 \text{ V}, Q_{air} = 1.6 \text{ L/min}, \text{ and } Q_r = 2.5 \text{ L/min}, ENC = 0.059 \text{ kWh/m}^3, \text{ and } ELC = 0.0096 \text{ kg/m}^3$.	>92.4	

A/V : the total active electrode area/volume of treated water in the reactor ratio; BECR: batch EC reactor; CECR: Continuous EC reactor; C_o : initial arsenic concentration; DSA: Dimensionally stable electrode; ELC: electrode consumption; ENC: energy consumption; GW: groundwater; i : applied current; j : current density; OC: operating cost; q : charge loading; Q_{air} : air flow rate; Q_r : recirculation rate; Q_w : water flow rate; q_e : adsorption capacity; R_e : removal efficiency; SW: synthetic water sample; t_{EC} : operating time, τ_{EC} : residence time in EC reactor, u : the mean linear flow rate.

from groundwater compared with the techniques covered in this study.

Conclusions

In this study, 62 experiments suggested by the experimental design method for As(III) removal in the batch EC process were conducted to construct the a quadratic model. The effects of some operational parameters like charge loading, operating time, current, As(III) concentration on the removal were evaluated in detail. Very high regression coefficient between the variables and the responses indicated for excellent evaluation of experimental data by the quadratic regression model. When the operating variables in range, the minimum operating cost and maximum removal efficiency were determined to be 99% and 0.01 \$/m³ at 20 µg/L and 50 µg/L. The results indicated that the mathematical approach in the EC process could be effectively used for the treatment of groundwater containing As(III).

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References

- [1] Ravenscroft, P.; Brammer, H.; Richards, K. (2009) *Arsenic Pollution: A Global Synthesis, RGS-IBG Book Series*; A John Wiley & Sons, Ltd. Publication: U.K.
- [2] McClintock, T.R.; Yu, C.; Jochen, B.; John, T.O.; Julio, N.; Valentina, O.; Edda, V.L.; Habibul, A.; Faruque, P. (2012) Arsenic exposure in Latin America: biomarkers, risk assessments and related health effects. *The Science of the Total Environment*, 429: 76. doi:10.1016/j.scitotenv.2011.08.051.
- [3] Ng, J.C.; Wang, J.; Shraim, A. (2003) Review: a global health problem caused by arsenic from natural sources. *Chemosphere*, 52: 1353. doi:10.1016/S0045-6535(03)00470-3.
- [4] WHO (World Health Organization). (2004) *Guidelines for Drinking Water Quality, Recommendations*, Vol. 1, 3rd Ed.; WHO Press, Geneva, Switzerland.
- [5] USEPA (The United States Environmental Protection Agency). (2000) *Arsenic treatment technology evaluation handbook for small system*, EPA 816-R-03-014. National Academy Press, Washington, D.C.
- [6] Baskan, B.M.; Pala, A. (2010) A statistical experiment design approach for arsenic removal by coagulation process using aluminum sulfate. *Desalination*, 254: 42. doi:10.1016/j.desal.2009.12.016.
- [7] Song, S.; Lopez-Valdivieso, A.; Hernandez-Campos, D. J.; Peng, C.; Monroy-Fernandez, G.M.; Razo-Soto, I. (2006) Arsenic removal from high-arsenic water by enhanced coagulation with ferric ions and coarse calcite. *Water Research*, 40: 364. doi:10.1016/j.watres.2005.09.046.
- [8] Mohan, D.; Pittman, C.U. (2007) Arsenic removal from water/wastewater using adsorbents: a critical review. *Journal of Hazardous Materials*, 142: 1. doi:10.1016/j.jhazmat.2007.01.006.
- [9] Kadouche, F.S.; Drouiche, N.; Messaoudi, K.; Messaoudi, B.; Lounici, H. (2016) Competitive adsorption of toxic metals on bentonite and use of chitosan as flocculent coagulant to speed up the settling of generated clay suspensions. *Chemosphere*, 165C: 87.
- [10] Demim, S.; Drouiche, N.; Aouabed, A.; Semsari, S. (2013) CCD study on the ecophysiological effects of heavy metals on Lemna gibba. *Ecolog Engineering*, 57: 302. doi:10.1016/j.ecoleng.2013.04.041.
- [11] An, B.; Steinwinder, T.R.; Zhao, D. (2005) Selective removal of arsenate from drinking water using a polymeric ligand exchanger. *Water Research*, 39: 4993. doi:10.1016/j.watres.2005.02.007.
- [12] Elcik, H.; Cakmakci, M.; Sahinkaya, E.; Ozkaya, B. (2013) Arsenic removal from drinking water using low pressure membranes. *Industrial Engineering Chemical Researcher*, 52: 9958. doi:10.1021/ie401393p.
- [13] Guan, X.; Dong, H.; Ma, J.; Jiang, L. (2009) Removal of arsenic from water. Effects of competing anions on As (III) removal in KMnO₄-Fe(II) process. *Water Processing Researcher*, 43: 3891. doi:10.1016/j.watres.2009.06.008.
- [14] Song, P.; Yang, Z.; Xu, H.; Huang, J.; Yang, X.; Wang, L. (2014) Investigation of influencing factors and mechanism of antimony and arsenic removal by electrocoagulation using Fe-Al electrodes. *Industrial Engineering Chemical Researcher*, 53: 12911. doi:10.1021/ie501727a.
- [15] Kobya, M.; Ulu, F.; Gebologlu, U.; Demirbas, E.; Oncel, M.S. (2011) Treatment of potable water containing low concentration of arsenic with electrocoagulation: different connection modes and Fe-Al electrodes. *Separation and Purification Technology*, 77: 283. doi:10.1016/j.seppur.2010.12.018.
- [16] Vasudevan, S.; Lakshmi, J.; Sozhan, G. (2010) Studies relating to removal of arsenate by electrochemical coagulation: optimization, kinetics, coagulant characterization. *Separation Science and Technology*, 45: 1313. doi:10.1080/01496391003775949.
- [17] Garcia-Lara, A.M.; Montero-Ocampo, C.; Martinez-Villafane, F. (2009) An empirical model for treatment of arsenic contaminated underground water by electrocoagulation process employing a bipolar cell configuration with continuous flow. *Water Science and Technology*, 60: 2153. doi:10.2166/wst.2009.641.
- [18] Hansen, H.K.; Nunez, P.; Raboy, D.; Schippacasse, I.; Grandon, R. (2007) Electrocoagulation in wastewater containing arsenic: comparing different process designs. *Electrochimica Acta*, 52: 3464. doi:10.1016/j.electacta.2006.01.090.
- [19] Boudjema, N.; Drouiche, N.; Abdi, N.; Grib, H.; Lounici, H.; Pauss, A.; Mameri, N. (2014) Treatment of Oued El Harrach river water by electrocoagulation noting the effect of the electric field on microorganisms. *Journal Taiwan Institute Chemical Engineering*, 45: 1564. doi:10.1016/j.jtice.2013.10.006.

- [20] Kobya, M.; Ozyonar, F.; Demirbas, E.; Sık, E.; Oncel, M.S. (2015) Arsenic removal from groundwater of Sivas-Şarkışla Plain, Turkey by electrocoagulation process: comparing with iron plate and ball electrodes. *Journal Environment Chemical Engineering*, 3: 1096. doi:10.1016/j.jece.2015.04.014.
- [21] Sık, E.; Kobya, M.; Demirbas, E.; Oncel, M.S.; Goren, A.Y. (2015) Removal of As(V) from groundwater by a new electrocoagulation reactor using Fe ball anodes: optimization of operating parameters. *Desalina Water Treatment*, 56: 1177. doi:10.1080/19443994.2014.951691.
- [22] Lakshmanan, D.; Clifford, D.A.; Samanta, G. (2010) Comparative study of arsenic removal by iron using electrocoagulation and chemical coagulation. *Water Research*, 44: 5641. doi:10.1016/j.watres.2010.06.018.
- [23] Pierce, M.L.; Moore, C.B. (1982) Adsorption of arsenite and arsenate on amorphous iron hydroxide. *Water Resource*, 16: 1247.
- [24] Kumar, P.R.; Chaudhari, S.; Khilar, K.C.; Mahajan, S.P. (2004) Removal of arsenic from water by electrocoagulation. *Chemosphere*, 55: 1245. doi:10.1016/j.chemosphere.2003.12.025.
- [25] Parga, J.R.; Cocke, D.L.; Valenzuela, J.L.; Gomes, J.A.; Kesmez, M.; Irwin, G.; Moreno, H.; Weir, M. (2005) Arsenic removal via electrocoagulation from heavy metal contaminated groundwater in La Comarca Lagunera Mexico. *Journal of Hazardous Materials*, 124: 247. doi:10.1016/j.jhazmat.2005.05.017.
- [26] Song, P.; Yang, Z.; Zeng, G.; Yang, X.; Xu, H.; Wang, L.; Xu, R.; Xiong, W.; Ahmad, K. (2017) Electrocoagulation treatment of arsenic in wastewaters: A comprehensive review. *Chemical Engineering Journal*, 317: 707. doi:10.1016/j.cej.2017.02.086.
- [27] APHA (American Public Health Association). (1998) *Standard Methods for the Examination of Water and Wastewater*, 19th Ed.; American Public Health Association, Washington, D.C..
- [28] Kobya, M.; Demirbas, E.; Ulu, F. (2016) Evaluation of operating parameters with respect to charge loading on the removal efficiency of arsenic from potable water by electrocoagulation. *Journal Environment Chemical Engineering*, 4: 1484. doi:10.1016/j.jece.2016.02.016.
- [29] Lakshmanan, D.; Clifford, D.A.; Samanta, G. (2009) Ferrous and ferric ion generation during iron electrocoagulation. *Environmental Science & Technology*, 43: 3853.
- [30] Kobya, M.; Gebologlu, U.; Ulu, F.; Oncel, S.; Demirbas, E. (2011) Removal of arsenic from drinking water by the electrocoagulation using Fe and Al electrodes. *Electrochimica Acta*, 56: 5060. doi:10.1016/j.electacta.2011.03.086.
- [31] Song, P.P.; Yang, Z.H.; Xu, H.Y.; Huang, J.; Yang, X.; Yue, F.; Wang, L.K. (2016) Arsenic removal from contaminated drinking water by electrocoagulation using hybrid Fe-Al electrodes: response surface methodology and mechanism study. *Desalination and Water Treatment*, 57: 4548.
- [32] Alcacio, R.; Nava, J.L.; Carreno, G.; Elorza, E.; Martinez, F. (2014) Removal of arsenic from deep well by electrocoagulation in a continuous filter press reactor. *Water Sciences Technology: Water Supply*, 14: 189.
- [33] Amrose, S.E.; Bandaru, S.R.S.; Delaire, C.; van Genuchten, C.M.; Dutta, A.; DebSarkar, A.; Orr, C.; Roy, J.; Das, A.; Gadgil, A.J. (2014) Electro-chemical arsenic remediation: field trials in West Bengal. *The Science of the Total Environment*, 488–489: 539. doi:10.1016/j.scitotenv.2013.11.074.
- [34] Kobya, M.; Akyol, A.; Demirbas, E.; Oncel, M.S. (2014) Removal of arsenic from drinking water by batch and continuous electrocoagulation processes using hybrid Al-Fe plate electrodes. *Environment Progress Sustain Energy*, 33: 131. doi:10.1002/ep.11765.
- [35] Can, B.Z.; Boncukcuoglu, R.; Yilmaz, A.E.; Fil, B.A. (2014) Effect of some operational parameters on the arsenic removal by electrocoagulation using iron electrodes. *Journal Environment Health Sciences Engineering*, 12: 2. doi:10.1186/2052-336X-12-95.
- [36] Kobya, M.; Demirbas, E.; Gebologlu, U.; Oncel, M.S.; Yildirim, Y. (2013) Optimization of arsenic removal from drinking water by electrocoagulation batch process using response surface methodology. *Desalination and Water Treatment*, 51: 6676. doi:10.1080/19443994.2013.769700.
- [37] Amrose, S.E.; Gadgil, A.J.; Srinivasan, V.; Kowolik, K.; Muller, M.; Huang, J.; Kostecki, R. (2013) Arsenic removal from groundwater using iron electrocoagulation: effect of charge dosage rate. *Journal Environment Sciences Health A*, 48: 1019. doi:10.1080/10934529.2013.773215.
- [38] Molgora, C.C.; Dominguez, A.M.; Avila, E.M.; Drogui, P.; Buelna, G. (2013) Removal of arsenic from drinking water: a comparative study between electrocoagulation-microfiltration and chemical coagulation-microfiltration processes. *Separation and Purification Technology*, 118: 645. doi:10.1016/j.seppur.2013.08.011.
- [39] Ali, I.; Gupta, V.K.; Khan, T.A.; Asim, M. (2012) Removal of arsenate from aqueous solution by electrocoagulation method using Al-Fe electrodes. *International Journal Electro Sciences*, 7: 1898.
- [40] Ali, I.; Khan, T.A.; Asim, M. (2012) Removal of arsenate from groundwater by electrocoagulation method. *Environmental Science and Pollution Research*, 19: 1668. doi:10.1007/s11356-011-0681-3.
- [41] Mohora, E.; Roncevic, S.; Dalmacija, B.; Agbaba, J.; Watson, M.; Karlovic, E.; Dalmacija, M. (2012) Removal of natural organic matter and arsenic from water by electrocoagulation/flotation continuous flow reactor. *Journal of Hazardous Materials*, 235–236: 257. doi:10.1016/j.jhazmat.2012.07.062.
- [42] Wan, W.; Pepping, T.J.; Banerji, T.; Chaudhari, S.; Giammar, D.E. (2011) Effects of water chemistry on arsenic removal from drinking water by electrocoagulation. *Water Research*, 45: 384. doi:10.1016/j.watres.2010.08.016.
- [43] Majumder, C.; Gupta, A. (2011) Prediction of arsenic removal by electrocoagulation: model development by factorial design. *Journal Hazard.; Toxic, and Radioactive Waste*, 15: 48. doi:10.1061/(ASCE)HZ.1944-8376.0000065.
- [44] Garcia-Lara, A.M.; Montero-Ocampo, C. (2010) Improvement of arsenic electro-removal from underground water by lowering the interference of other

- ions. *Water, Air, & Soil Pollution*, 205: 237. doi:10.1007/s11270-009-0069-x.
- [45] Zhao, X.; Zhang, B.; Liu, H.; Qu, J. (2011) Simultaneous removal of arsenic and fluoride via an integrated electro-oxidation and electrocoagulation process. *Chemosphere*, 83: 726. doi:10.1016/j.chemosphere.2011.01.055.
- [46] Lakshminathiraj, P.; Prabhakar, S.; Raju, G.B. (2010) Studies on the electrochemical decontamination of wastewater containing arsenic. *Separation and Purification Technology*, 73: 114. doi:10.1016/j.seppur.2010.03.009.
- [47] Mohora, E.; Roncevic, S.; Agbaba, J.; Zrnic, K.; Tubic, A.; Dalmacija, B. (2018) Arsenic removal from groundwater by horizontal-flow continuous electrocoagulation (EC) as a standalone process. *Journal Environment Chemical Engineering*, 6: 512. doi:10.1016/j.jece.2017.12.042.
- [48] Deniel, R.; Bindu, V.H.; Prabhakararo, A.V.S.; Anjaneyulu, Y. (2008) Removal of arsenic from wastewaters using electrocoagulation. *Journal Environment Sciences Engineering*, 50: 283.
- [49] Thakur, L.S.; Mondal, P. (2017) Simultaneous arsenic and fluoride removal from synthetic and real groundwater by electrocoagulation process: parametric and cost evaluation. *Journal Environment Manual*, 190: 102. doi:10.1016/j.jenvman.2016.12.053.
- [50] Şık, E.; Demirbas, E.; Goren, A.Y.; Oncel, M.S.; Kobya, M. (2017) Arsenite and arsenate removals from groundwater by electrocoagulation using iron ball anodes: influence of operating parameters. *Journal Water Pro Engineering*, 18: 83. doi:10.1016/j.jwpe.2017.06.004.
- [51] Maldonado-Reyes, A.; Montero-Ocampo, C.; Solorza-Feria, O. (2007) Remediation of drinking water contaminated with arsenic by the electro-removal process using different metal electrodes. *Journal Environment Monitoring*, 9: 1241. doi:10.1039/b708671g.
- [52] Gomes, J.A.G.; Daida, P.; Kesmez, M.; Weir, M.; Moreno, H.; Parga, J.R.; Irwin, G.; McWhinney, H.; Grady, T.; Peterson, E.; Cocke, D.L. (2007) Arsenic removal by electrocoagulation using combined Al-Fe electrode system and characterization of products. *Journal of Hazardous Materials*, 139: 231. doi:10.1016/j.jhazmat.2005.11.108.
- [53] Pinisakul, A.; Polprasert, C.; Parkpian, P.; Satayavivad, J. (2002) Arsenic removal efficiency and mechanisms by electro-chemical precipitation process. *Water Science and Technology*, 46: 247. doi:10.2166/wst.2002.0250.
- [54] Martinez-Villafane, J.F.; Montero-Ocampo, C.; Garcia-Lara, A.M. (2009) Energy and electrode consumption analysis of electrocoagulation for the removal of arsenic from underground water. *Journal of Hazardous Materials*, 172: 1617. doi:10.1016/j.jhazmat.2009.08.044.
- [55] Ahmed, F. (2004) *Arsenic contamination of groundwater in South and East Asian countries*, The World Bank, Volume I and II Policy and Technical Reports, Report no. 31303, World Bank, Washington, D.C.