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Optimization of arsenic removal from drinking water by electrocoagulation batch process using response surface methodology

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ABSTRACT

In this investigation, arsenic removal from drinking water using electrocoagulation (EC) in a batch mode was studied by response surface methodology (RSM). The RSM was applied to optimize the operating variables viz. current density (CD, A/m^2), operating time (t_{EC}, min) and arsenic concentration (C_o, μg/L) on arsenic removal in the EC process using iron electrodes. The combined effects of these variables were analyzed by the RSM using quadratic model for predicting the highest removal efficiency of arsenic from drinking water. The proposed model fitted very well with the experimental data. R^2 adjusted correlation coefficients (AdjR^2: 0.93) for arsenic removal efficiency showed a high significance of the model. The model predicted for a maximum removal of arsenic at the optimum operating conditions (112.3 μg/L, 5.64 A/m^2 and 5 min) after the EC process was 93.86% which corresponded to effluent arsenic concentration of 6.9 μg/L. The minimum operating cost (OC) of the EC process was 0.0664 €/m^3. This study clearly showed that the RSM was one of the suitable methods for the EC process to optimize the best operating conditions for target value of effluent arsenic concentration (<10 μg/L) while keeping the OC (energy and electrode consumptions) to minimal.

Keywords: Electrocoagulation; Arsenic removal; Drinking water; Response surface methodology; Optimization; Operating cost

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1. Introduction

Arsenic in natural water source has been a serious concern worldwide. Arsenic concentration in soils and water have become elevated because of several reasons like, mineral dissolution, disposal of fly ash, mine drainage, and geothermal discharge. [1,2] Chronic health effects of arsenic in drinking water include development of various skin lesions, keratoses of the hands and feet. Its ingestion may deleteriously affect the gastrointestinal tract, vascular system and central nervous system [3]. The most serious problems being encountered in many regions of the world such as Argentina, Bangladesh, Chile, India, Mexico, Mongolia, Myanmar, Nepal, New Zealand, Thailand,

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Taiwan, Turkey and Vietnam are high rate of arsenic concentrations when these in ground and surface water exceeds a maximum contaminant level (MCL) of 10 µg/L in drinking water [4,5]. Natural water sources especially in the west regions of Turkey such as Balikesir, Kutahya, Usak, Izmir, and Afyon contain much higher levels of arsenic concentrations in the range 0.05–900 µg/L than the allowed MCL. Lowering of this MCL makes it necessary to find novel technologies to meet the regulations [6–8].

Arsenic species in natural water occur in two oxidation states such as As(III) [H_3AsO_3, $H_2AsO_4^{3-}$, $HAsO_3^{2-}$ and As(V) [H_3AsO_4, $H_2AsO_4^-$, $HAsO_4^{2-}$] and AsO_4^{3-}. Mainly, the concentration of arsenic species depends on redox potential and pH of water. As(III) predominant species under reducing condition, while As(V) found in oxidizing situation As(V) exists as an anion with $H_2AsO_4^-$ to be the predominant species at pH 6 and $HAsO_4^{2-}$ at pH 8, while As(III) is fully protonated and exists as an uncharged molecule (H_3AsO_3). In order to enhance the removal rate of arsenic, it would be necessary to oxidize As(III) to As(V) [9–11].

Different treatment processes including adsorption, precipitation/coprecipitation, membrane filtration and ion exchange resins were used to study the influence of arsenic speciation and concentration, pH and competitive ions on arsenic removal. These processes showed a medium to low As(III) removal efficiency and also required pH regulation as it influences arsenic speciation and surface charge of adsorbents. The literature results on arsenic removal by electrocoagulation/flocculation (ECF) indicated high As(III) removal efficiency without any pH regulation [10,30]. Electrocoagulation (EC) is an alternative process to chemical flocculation (CF). Instead of adding a chemical reagent as ferric chloride, metallic cations are directly generated in the effluent to be treated by applying a current between iron electrodes to dissolve soluble anodes. In a drinking water production plant, CF and ECF are mainly use for colloids and organic matter removal and actually no study deals with the removal of arsenic in the presence of organic matter [12]. In recent years, EC technique has been receiving greater attention for removal efficiency of arsenic as compared to the conventional methods [13–17]. Because the EC is an efficient and cost effective process controlled electrically with no moving parts, thus requiring less maintenance. EC generates metallic hydroxide flocs *in situ* by electrodissolution of soluble anode material. The production of metal cations from the anode and other charged metal hydroxide species may cause neutralization of negatively charged particles. Then, the particles in solution bind together to form flocs, resulting in the removal of pollutant from wastewater. In an EC process with Fe electrode; the anodic reactions Eqs. (1)–(3) occur where iron is firstly oxidized to ferrous ion which depending on anode potential, then oxidizes to ferric ion [18,19]:

$$Fe_{(s)} \rightarrow Fe^{2+} + 2e^- \tag{1}$$

$$Fe^{2+} \rightarrow Fe^{3+} + e^- \tag{2}$$

$$Fe_{(s)} \rightarrow Fe^{3+} + 3e^- \tag{3}$$

As the current is applied, the electrodissolution of the anode is accompanied with the oxidation of water

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \tag{4}$$

The cathode reaction takes place at the cathode and results in the liberation of hydrogen Eq. (5).

$$2H_2O + 2e^- \rightarrow H_{2(g)} + 2OH^- \tag{5}$$

Fe^{2+} is oxidized rapidly if air (or oxygen) is introduced to the process

$$O_{2(g)} + 4Fe^{2+} + 2H_2O \rightarrow 4Fe^{3+} + 4OH^- \text{(in bulk solution)} \tag{6}$$

Generally, Fe^{3+} ions released from anode are gradually hydrolyzed and formed the $Fe(OH)_{3(s)}$ if there is no other reactive species in solution. For Fe electrodes, the rate of the oxidation depends on the availability of dissolved oxygen. Typically at the cathode the solution becomes alkaline with time. The applied current forces OH^- ion migration towards the cathode and combine with hydroxide ions Eq. (7):

$$Fe^{3+} + 3OH^- \rightarrow Fe(OH)_{3(s)} \tag{7}$$

At pH $4 < pH < 7$, iron undergoes hydrolysis according to reactions 6–10 [20]

$$Fe + 6H_2O \rightarrow Fe(H_2O)_4(OH)_{2(aq)} + 2H^+ + 2e \text{ (anode)} \tag{8}$$

$$Fe + 6H_2O \rightarrow Fe(H_2O)_3(OH)_{3(aq)} + 3H^+ + 3e \text{ (bulk of solution)} \tag{9}$$

Fe^{3+} hydroxide begins to precipitate floc with yellowish color

$$Fe(H_2O)_3(OH)_{3(aq)} \rightarrow Fe(H_2O)_3(OH)_{3(s)} \quad (10)$$

Fe causes the evolution of H_2 from cathodic reaction Eq. (5). Rust may also be formed.

$$4Fe(H_2O)_3(OH)_{3(s)} \rightarrow 2Fe_2O_3(H_2O)_{6(s)} + 6H_2O_{(l)} \quad (11)$$

At pH $6 < pH < 9$, precipitation of Fe^{3+} hydroxide Eq. (9) continues. The minimum soluble iron concentration $Fe(OH)_3$ solubility occurs over the pH range of 7–10 and Fe^{2+} hydroxide precipitation also occurs presenting a dark green floc

$$4Fe(H_2O)_4(OH)_{2(aq)} \rightarrow 4Fe(H_2O)_4(OH)_{2(s)} \text{ (bulk of solution)} \quad (12)$$

As removal with the EC using Fe electrodes was formation of a dark green floc. The rate of generation of flocs is easily controlled by applied voltage which leads to minimization of amount of sludge in the EC process. Ferric ions generated by electrochemical oxidation of iron electrode may form monomeric species, $Fe(OH)_3$ and polymeric hydroxyl complexes namely, $FeOH^{2+}$, $Fe(OH)_2^+$, $Fe_2(OH)_2^{4+}$, $Fe(OH)_4^-$, $Fe(H_2O)_2^+$, $Fe(H_2O)_6^{3+}$, $Fe(H_2O)_5(OH)^{2+}$, $Fe(H_2O)_4(OH)_2^+$, $Fe_2(H_2O)_8(OH)_2^{4+}$ and $Fe_2(H_2O)_6(OH)_4^{2+}$ depending on the pH of the aqueous medium [20,21]. These hydroxides/polyhydroxides/polyhydroxyoxide metallic compounds have strong affinity for dispersed particles as well as counter ions to cause coagulation. The arsenic removal occurs also by ligand exchange, arsenate displaces a hydroxyl group of FeOOH giving rise to an insoluble surface complex [22–25]

$$2FeOOH_{(s)} + H_2AsO_4^- \rightarrow (FeO)_2HAsO_4^- + H_2O + OH^- \quad (13)$$

$$3FeOOH_{(s)} + HAsO_4^{2-} \rightarrow (FeO)_3AsO_{4\,(s)}^- + H_2O + 2OH^- \quad (14)$$

where the surface symbols $\equiv$ is used to denote the bonds of the cations with the surface of the solid.

Response surface methodology (RSM), a collection of mathematical and statistical techniques, is commonly used for improving and optimizing processes [26]. It can be used to evaluate the relative significance of several affecting factors in the presence of complex interactions. Optimization of the process variables during wastewater treatment in the EC process can be achieved using the RSM [27–32]. Removal of arsenic from drinking and groundwater has been little investigated in the literature with modeling and optimization of the EC process [33–35].

The objective of this study was to evaluate removal of arsenic from drinking water containing low arsenic concentrations in the EC process using Fe electrodes to meet the permissible limit set by WHO (10 µg/L). The RSM was used to develop a mathematical model to describe the effects and relationships of independent variables for the main process using three operating parameters such as current density, operating time and initial arsenic concentration to maximize arsenic removal efficiency and to minimize operating cost (OC) in relation to energy and electrode consumptions.

2. Materials and methods

2.1. Materials

Stock arsenic solutions of 1,000 mg As/L were prepared according to the EPA standard method by dissolving As_2O_3 in distilled water containing 20% (v/v) KOH and then neutralizing by 20% (v/v) H_2SO_4 to a phenolphthalein end point. The solutions were stored at 4°C in the refrigerator to minimize microbial growth in the sample. The test solutions for different arsenic concentrations were prepared by diluting of stock solution with drinking water before use [25].

2.2. Experimental setup and procedure

The EC experiments were carried out in a batch mode using a 0.80 L Plexiglas reactor (80 mm × 80 mm × 126 mm in dimension) using vertically positioned iron electrodes spaced by 13 mm and dipped in the arsenic solution. Two anodes and two cathodes with dimensions of 50 mm × 73 mm × 2 mm made of iron plate (99.5% purity) were connected to a digital DC power supply (Agilent 6675A model; 120 V, 18 A) equipped with galvanostatic operational options in monopolar parallel connection modes [24]. Total effective area of electrode was 219 cm^2. pH and conductivity of solutions before and after the EC process were measured by a pH meter and a conductivity meter (Hach Lange HQ40), respectively. pH of the solutions was adjusted by adding either 0.1 N NaOH or 0.1 N H_2SO_4. The solution was constantly stirred at 300 rpm (Heidolp 3600) to reduce the mass transport over potential of the EC reactor.

In each run, 0.65 L arsenic solution was placed into the EC reactor. Before each run, organic impurities and oxide layer on electrode surfaces were removed by dipping for 2 min in a solution freshly prepared by mixing HCl (35%) and hexamethylenetetramine aqueous solutions (2.80%) [18]. Current and voltage were held constant at desired values for each run and the experiment was started. The samples at the different operating times taken from the EC reactor were filtered using a 0.45 μm Millipore membrane and arsenic concentration was measured. At the end of the run, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces, dried, and reweighed.

2.3. Experimental design and data analysis

RSM uses an experimental design such as the central composite design (CCD) to fit a model by least squares technique. RSM usually contains three steps: (i) design and experiments, (ii) response surface modeling through regression, and (iii) optimization. The main objective of RSM is to determine the optimum operational conditions of the process or to determine a region that satisfies the operating specifications [26–32]. RSM makes it possible to represent independent process parameters in quantitative form as:

$$y = f(x_1, x_2, x_3, \ldots, x_n) \pm \varepsilon \tag{15}$$

where y is the response (yield), f is the response fuction, ε is the experimental error, and x_1, x_2, x_3,…,x_n are independent parameters. By plotting the expected response of y, a surface, known as the response surface is obtained. The form of f is unknown and may be very complicated. Thus, RSM aims at approximating f by a suitable lower-ordered polynomial in some region of the independent process variables. If the response can be well modeled by a linear function of the independent variables, the function in Eq. (15) can be written as:

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \varepsilon \tag{16}$$

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} x_i x_j + \varepsilon \tag{17}$$

For statistical calculations, the variables X_i were coded as x_i according to the following equation:

$$x_i = \frac{X_i - X_i^x}{\Delta X_i} \tag{18}$$

where x_i is the coded value of the ith independent variable, X_i the actual value of the ith independent variable, X_i^x the actual value of the ith independent variable at the center point, and ΔX_i is the value of step change.

Thus, each response y can be represented by a mathematical equation that correlates the response surface. However, if a curvature appears in the system, then a higher order polynomial such as the quadratic model Eq. (19) may be used.

$$y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 \tag{19}$$

where y is the predicted response surface function (percent As(III) removal), β_0 is the model constant, β_1, β_2, and β_3 are linear coefficients, β_{12}, β_{13} and β_{23} are the cross product coefficients, and β_{11}, β_{22} and, β_{33} are the quadratic coefficients.

The Design Expert 8.0 trial version program (USA) was used for the statistical design of experiments, determination of the coefficients and data analysis. The three important operating variables such as current density (CD: x_1), operating time (t_{EC}: x_2), and arsenic concentration (C_o: x_3) were chosen as the independent variables in the EC process. Since different variables are usually expressed in different units and/or have different limits of variation, the significance of their effects on response can only be compared after they are coded. The independent variables range and levels were set using the RSM model (Table 1). As presented in Table 1, the experimental design involved three parameters (x_1, x_2 and x_3), each at three levels, coded −1, 0, and +1 for low, middle, and high concentrations, respectively. Eight star points (±1), six axial points (±α) and six replicates at the centre point (0) were chosen as experimental points. Experimental independent variables were current density, operating time and concentration were.

The study ranges were chosen as current density of 0.8–9.2 A/m^2, operating time of 1.6–18.4 min and arsenic concentration of 15.9–184.1 μg/L for the EC

Table 1
Experimental range and levels of the independent process variables

Independent variables	Range and levels				
	−α	−1	0	+1	+α
x_1: current density (A/m^2)	0.8	2.5	5.0	7.5	9.2
x_2: operating time (min)	1.6	5	10	15	18.4
x_3: as concentration (μg/L)	15.9	50	100	150	184.1

Table 2
A full factorial design for three independent variables along with responses for removal of arsenic

Experimental run	Independent variables				Responses						
	x_1: CD (A/m^2)	x_2: t_{EC} (min)	x_3: C_o (μg/L)	y_1: C_f (μg/L)	y_2: R_e (%)	ENC (kWh/m^3)	ELC (kg Fe/m^3)	OC (€/m^3)	pH$_{final}$	Current (A)	Voltage (V)
1	5.0	10.0	100.0	4.70	95.3	0.0265	0.0748	0.1023	7.60	0.110	1.07
2	5.0	10.0	100.0	3.90	96.1	0.0274	0.0635	0.0919	7.60	0.110	1.07
3	9.2	10.0	100.0	0.50	99.5	0.0949	0.0676	0.1635	7.95	0.200	1.85
4	7.5	15.0	150.0	2.10	98.6	0.0972	0.0923	0.1915	7.62	0.165	1.58
5	2.5	5.0	150.0	41.92	72.1	0.0035	0.0386	0.0431	7.41	0.055	0.54
6	5.0	18.3	100.0	2.40	97.6	0.0468	0.0994	0.1472	7.43	0.110	1.00
7	7.5	15.0	50.0	1.15	97.7	0.0923	0.0939	0.1872	7.98	0.165	1.50
8	7.5	5.0	150.0	8.70	94.2	0.0312	0.0522	0.0844	7.47	0.165	1.52
9	5.0	10.0	184.0	19.87	89.2	0.0226	0.0683	0.0919	7.56	0.110	0.78
10	2.5	15.0	50.0	2.95	94.1	0.0142	0.0862	0.1014	7.45	0.055	0.74
11	5.0	10.0	100.0	3.40	96.6	0.0274	0.0738	0.1022	7.57	0.110	1.07
12	2.5	15.0	150.0	11.25	92.5	0.0119	0.0712	0.0841	7.47	0.055	0.62
13	5.0	10.0	15.9	0.150	99.1	0.0203	0.0863	0.1076	7.61	0.110	0.79
14	2.5	5.0	50.0	8.40	83.2	0.0042	0.0634	0.0686	7.58	0.055	0.62
15	5.0	1.6	100.0	14.80	85.2	0.0035	0.0415	0.0461	7.55	0.110	0.85
16	5.0	10.0	100.0	3.40	96.6	0.0276	0.0625	0.0911	7.58	0.110	1.07
17	5.0	10.0	100.0	4.20	95.8	0.0274	0.0630	0.0914	7.52	0.110	1.07
18	0.8	10.0	100.0	24.50	75.5	0.0011	0.0564	0.0585	7.23	0.017	0.25
19	5.0	10.0	100.0	3.90	96.1	0.0274	0.0625	0.0919	7.57	0.110	1.07
20	7.5	5.0	50.0	2.90	94.2	0.0338	0.0665	0.1013	7.53	0.165	1.50

process using Fe electrodes (Table 1). In order to study the combined effect of these factors, experiments were conducted at different combination of physical parameters.

In Table 1, the coded values of x_1, x_2 and x_3 were set at five levels: $-\alpha$, -1 (minimum), 0 (central), +1 (maximum) and $+\alpha$. Three dependent parameters were analyzed as responses; effluent arsenic concentration (y_1), arsenic removal efficiency (y_2), electrode (ELC) and energy (ENC) consumptions and OC for removal of arsenic from drinking water in the EC process (Table 2). The quadratic equation model for predicting the optimal conditions could be expressed according to Eq. (19). The actual design of this work was presented in Table 2.

Table 2 showed the CCD in the form of a 2^3 full factorial design with six additional experimental trials (run numbers 1, 2, 11, 16, 17, and 19) as replicates of the central point to find the influencing parameters. A total of 20 experiments were performed (Table 2). The experimental runs were carried out in triplicates. Results of the central points (experimental runs) were used to check the reproducibility of results as per CCD. Analysis of variance (ANOVA) was used for graphical analyses of the data to obtain the interaction between the process variables and the responses. The quality of the fit polynomial model was checked by the determination of coefficient (R^2), and its statistical significance was checked by the Fisher F-test in the same program. Model terms were evaluated by the P value (probability) with 95% confidence level.

2.4. Chemical analysis and OCs

Based on the standard method suggested by APHA [36], an atomic absorption spectrometer (PerkinElmer SIMAA 6000 AAS) equipped with a manual hydride generator at 188.9 nm wavelength was employed to determine the arsenic concentration in the samples. The detection limit for this study was 0.1 μg/L of arsenic and analysis of the duplicates was within 2% of errors.

Laboratory-scale experiments were carried out at room temperature. All the chemical reagents were of analytical grade. The chemical analysis of the drinking water (pH, alkalinity, and the presence of arsenic, iron and phosphate) was carried out according to Standard methods [36]. Chemical analysis of drinking water is performed by using standard methods like Cl^- and NO_3^- by an ion chromatography (Shimadzu HIC-20A), SO_4^{2-} by a turbitimeter (Hach Lange DR 2800), HCO_3^- and total hardness by a titration method, phosphate by a spectrophometer (Perkin Elmer lambda 35 UV/Vis) and conductivity, pH and dissolved O_2 by Hach Lange HQ40d, dissolved organic matter by TOC (Tekmar Dohrmann Apollo 9000), dissolved iron, and manganese, by ICP-OES (PerkinElmer ICP-OES Optima 7000 DV, respectively. Characterizations of drinking water were 85 mg/L of Cl^-, 18 mg/L of SO_4^{2-}, 85 mg/L of HCO_3^-, conductivity of 70 μS/cm (adjusted to 1.7 mS/cm with NaCl and Na_2SO_4), 10 mg/L of NO_3^-, initial pH 7.4 (adjusted to pH 6.5), 6.2 mg/L of dissolved O_2, 6 mg/L of magnesium, and 20 mgCaCO$_3$/L of total hardness, respectively. In addition, dissolved organic matter, dissolved iron, manganese, phosphate and arsenic concentration were not detected in the drinking water [24,25].

The OC of the EC process includes material, mainly electrodes and electrical energy costs, as well as labor, maintenance, sludge dewatering and disposal, and fixed costs. The latter costs items are largely independent of the type of the electrode material [25,37]. In this study, energy, and electrode material costs were taken into account as major cost items in the calculation of the OC (€/m^3):

$$\text{Operating Cost(OC)} = a\text{ENC} + b\text{ELC} + c\text{CC} \tag{20}$$

where ENC (kWh/m^3) and ELC (kgFe/m^3) are consumption quantities for removal of arsenic. "a" and "b" were electrical energy price (0.072 €/kWh) and electrode material price (0.85 €/kg Fe) and prices were provided in Turkish market in June 2011. "c" stands for chemical consumption (CC) such as NaOH and H_2SO_4 for adjustment of desired pH and prices were 0.73 €/kg and 0.29 €/kg, respectively. Costs of electrical energy (kWh/m^3) in Eq. (21) and electrode consumptions (kg Fe/m^3) were calculated from Faraday's Law Eq. (22).

$$\text{ENC(k Wh/m}^3) = \frac{Uit_{\text{EC}}}{v} \tag{21}$$

$$\text{ELC(kg electrode/m}^3) = \frac{it_{\text{EC}}M_w}{zFv} \tag{22}$$

where U is cell voltage (V), i is current (A), t_{EC} is the operating time (s) and v is the volume (m^3) of the wastewater, M_w are molecular mass of iron (56.8 g/mol), z is number of electron transferred ($z_{\text{Fe}} = 2$) and F is Faraday's constant (96,487 C/mol).

3. Result and discussions

The CCD was used to obtain the experimental design matrix for the process optimization. This approach has a limited number of actual experiments performed while allowing probing into possible interaction between these parameters studied and their effects on removal efficiency of arsenic and OC.

3.1. Statistical analysis

In order to determine the optimum conditions for removal of arsenic, the parameters that have greatest influence on the response need to be identified. In the present study, the relationship between three independent variables and responses was established well with the quadratic model. The quadratic regression model for effluent arsenic concentration (y_1), removal efficiency (y_2) and OC including energy and electrode consumptions obtained from the CCD design in terms of coded factors was presented as follows:

$$\begin{aligned} y_1\ (\mu\text{g/L}) = {} & +32.51 - 6.18x_1 - 2.14x_2 + 0.046x_3 \\ & + 0.36x_1x_2 - 0.035x_1x_3 - 0.011x_2x_3 \\ & + 0.35x_1^2 + 0.031x_2^2 + 1.82x10^{-3}x_3^2 \end{aligned} \tag{23}$$

$$\begin{aligned} y_2\ (\%) = {} & +96.03 + 5.51x_1 + 4.40x_2 - 3.72x_3 \\ & - 2.93x_1x_2 + 0.7x_1x_3 + 1.30x_2x_3 - 2.79x_1^2 \\ & - 1.40x_2^2 - 2.47x_3^2 \end{aligned} \tag{24}$$

$$\begin{aligned} \text{ENC (kWh/m}^3) = {} & +0.027 + 0.028x_1 + 0.016x_2 \\ & + 2.31x10 - 4x_3 + 0.013x_1x_2 \\ & + 6.63x_1x_3 + 7.38x10 - 4x_2x_3 \\ & + 8.22x10^{-3}x_1^2 + 2.53x10^{-4}x_2^2 \\ & - 1.18x10 - 3x_3^2 \end{aligned} \tag{25}$$

Table 3
ANOVA of the second-order polynomial equation for arsenic removal

Source	Coefficient estimate	Degree of freedom	Mean square	F value	$Prob > F$	Remarks*
Model intercept	32.51	9	212.33	29.27	<0.0001[b]	Highly significant
x_1: CD	6.18	1	593.50	81.82	<0.0001	Highly significant
x_2: t_{EC}	−2.14	1	311.02	42.88	<0.0001	Highly significant
x_3: C_o	0.046	1	462.63	63.78	<0.0001	Highly significant
x_1x_2	0.36	1	96.40	13.29	0.0045	Significant
$x_1.x_3$	−0.035	1	153.74	21.19	0.0010	Significant
$x_2.x_3$	−0.011	1	113.03	15.58	0.0027	Significant
	0.35	1	113.35	15.63	0.0027	Significant
x_1^2	0.031	1	27.87	3.84	0.0784	Low significant
x_3^2	0.0018	1	67.50	9.30	0.0122	Low significant
Residual	38.11	10	7.25			
Lack of fit	36.88	5	14.26	58.06	0.0002	Highly significant
Pure error	1.23	5	0.25			
Cor total	1080.99	19				
SD	2.69	R^2	0.963			
Mean	8.32	Adj R^2	0.931			
CV	32.36	Pred R^2	0.684			
Press	626.24	Adep precision	20.63			

[a]Response: effluent arsenic concentration (y_1, μg/L).*The values of $Prob > F < 0.500$ indicate that the model terms are significant, whereas the values >0.100 are not significant [35].[b]Model significant.

$$\begin{aligned} \text{ELC (kg/m}^3) = &+0.067 + 4.71x10^{-3}x_1 + 0.016x_2 \\ &- 6.29x10^{-3}x_3 + 1.51x10^{-3}x_1x_2 \\ &+ 2.99x10^{-3}x_1x_3 + 2.81x10^{-3}x_2x_3 \\ &- 1.59x10^{-3}x_1^2 + 1.50x10^{-3}x_2^2 \\ &+ 3.82x10^{-3}x_3^2 \end{aligned} \quad (26)$$

$$\begin{aligned} \text{OC (€/m}^3) = &+0.095 + 0.033x_1 + 0.032x_2 - 5.99 \\ &\times 10^{-3}x_3 + 0.015x_1x_2 + 3.78 \\ &\times 10^{-3}x_1x_3 + 3.68 \times 10^{-3}x_2x_3 + 6.60 \\ &\times 10^{-3}x_1^2 + 1.73 \times 10^{-3}x_2^2 + 2.60 \\ &\times 10^{-3}x_3^2 \end{aligned} \quad (27)$$

where x_1, x_2 and x_3 are the coded values of tested variables. A positive and negative signs in front of the terms refer to a synergistic effect and antagonistic effect, respectively. Table 2 presented how responses changed with independent variables. Experimental runs # 18,11,3 explained changes obtained in Eqs. (23)–(27) related to increasing removal efficiencies of arsenic from 75.5 to 99.5% and ENC, ELC, and OC values with increasing of current density from 0.8 to 9.2 A/m^2, increasing of removal efficiency from 85.2 to 97.6% with time from 1.6 to 18.3 min in experimental runs # 6, 19, 15, and removal efficiencies were increased from 89.2 to 99.1% as concentrations decreased from 184 to 15.9 μg/L in experimental runs # 13, 19, 9, respectively. As represented in the above equations, there were some interaction effects between these parameters. For example, as current density and time were increased, the removal efficiencies were increased. In addition, as current density and concentration were increased, the removal efficiencies were increased with increase in current density and decreased with increase in concentration.

The effects of the studied variables and type of interaction (positive or negative) to the response were illustrated in Tables 3a and 3b. The large value of F indicated that most of the variation in the response could be explained by regression equation. Values of $Prob > F$ less than 0.05 showed that the model terms were significant, whereas the values greater than 0.10 were not significant [38]. The mathematical relationships between energy (ENC, kWh/m^3) and electrode consumptions (ELC, kg Fe/m^3), OC (€/m^3) and variables such as x_1, x_2 and x_3 were obtained. The adequacy of the RSM was justified through ANOVA to assess the "goodness of fit". Only terms found statistically significant were included in the model. The non-significant terms could be reduced by reselect only the significant terms to be included in the model. The ANOVA results for response parameters of Fe electrodes were shown in Table 3.

Table 4
ANOVA for the second-order polynomial equation for arsenic removal efficiency

Source	Coefficient estimate	Degree of freedom	Mean square	F value	$Prob > F$	Remarks*
Model intercept	96.03	9	115.87	30.40	<0.0001[b]	Highly significant
x_1:CD	5.51	1	506.34	132.86	<0.0001	Highly significant
x_2: t_{EC}	4.40	1	262.80	68.96	<0.0001	Highly significant
x_3: C_o	−3.72	1	14.71	3.86	0.0778	Low significant
$x_1.x_2$	−2.93	1	68.45	17.96	0.0017	Significant
$x_1.x_3$	0.70	1	23.12	6.07	0.0335	Low significant
$x_2.x_3$	1.30	1	13.52	3.55	0.0890	Low significant
x_1^2	−2.79	1	104.36	27.38	0.0004	Highly significant
	−1.40	1	23.64	6.20	0.0320	Low significant
x_3^2	−2.47	1	48.92	12.84	0.0050	Highly significant
Residual	38.11	10	3.81			
Lack of fit	36.88	5	7.38	30.03	0.0010	Significant
Pure error	1.23	5	0.25			
Cor total	1080.99	19				
SD	1.95	R^2	0.965			
Mean	92.03	Adj. R^2	0.930			
CV	2.12	Pred R^2	0.727			
Press	295.57	Adep precision	19.58			

[a]Response: arsenic removal efficiency (y_2, %).[b]Model significant.

*The values of $Prob > F < 0.500$ indicate that the model terms are significant, whereas the values >0.100 are not significant [35].

F-values from the ANOVA in Table 4 were 29.3, 30.4 and 116.4 for removal efficiency of arsenic, effluent concentration of arsenic and OC for Fe electrodes, respectively indicating that the model was significant. The P-value was lesser than 0.0001 (99% confidence) indicated that the model was considered to be statistically significant. The R^2 coefficient gave the proportion of the total variation in the response variable accounted for the predictors (x's) included in the model. A high R^2 value, close to 1, was desirable and a reasonable agreement with adjusted R^2. A high R^2 coefficient ensured a satisfactory adjustment of the quadratic model to the experimental data. The values of R^2 for removal efficiency of arsenic, effluent arsenic concentration and OC were 0.965, 0.963 and 0.991, respectively. The values of the adjusted R^2 for removal efficiency of arsenic, effluent concentration of arsenic and OC were 0.930, 0.931, and 0.982, respectively. The value of R^2 indicated that only 0.009–0.065% of the total variable was not explained by the model.

The coefficient of variance (CV) is the ratio of the standard error of estimate to the mean value of observed response (as percentage) and considered to be reproducible when it is not greater than 10%. In this work, the CVs for removal of arsenic and OC were 32.36 and 5.36. Adequate precision (AP) compared the range of the predicted values at the design points to the average prediction error. A ratio of $AP > 4$ was desirable. For the present study, AP values for Fe electrodes used in the EC process were 19.58 for arsenic removal efficiency and 37.1 for OC which indicated an adequate signal. Therefore, quadratic model could be used to navigate the design space.

The lack of fit obtained for this model was <0.0010. The non-significance lack of fit value explained that the quadratic model was valid for the present work when it was higher than 0.05, indicating for both responses insignificant lack of fit for the model. The effect of each variable on the response was the combination of coefficients and variable values as well as contribution of joint effect of variables that could not be observed by conventional experimental methods.

When operating variables were in range, the removal efficiency of arsenic was maximized and the OC was minimized for an initial arsenic concentration of 112.3 μg/L in the RSM (Table 5). The optimal EC conditions found by the response optimization were current density of 5.64 A/m^2, operating time of 5 min, initial concentration of 112.3 μg/L. Final concentration and OC were <6.9 μg/L and 0.0664 €/m^3 at the optimised conditions, and the results were in agree with the recommended arsenic concentration of drinking water (10 μg/L) set by WHO. The desirability function is one of the most frequently used multi-response

Table 5
ANOVA results for the response parameters

Responses	R^2	Adj. R^2	SD	CV	F-value	$Prob > F$	AP
y_1 (μg/L)	0.963	0.931	2.69	32.36	29.3	<0.0001	20.63
y_2 (%)	0.965	0.930	1.95	2.12	30.4	<0.0001	19.58
ENC (kWh/m^3)	0.991	0.983	0.004	11.91	124.6	<0.0001	35.5
ELC (kg Fe/m^3)	0.957	0.918	0.005	6.74	24.7	<0.0001	17.8
OC (€/m^3)	0.991	0.982	0.006	5.36	116.4	<0.0001	37.1

SD: standard deviation, CV: coefficient of variance, AP: adequate precision.

Table 6
The optimization results for removal of arsenic

Name	Goal	Lower limit	Upper limit	Lower weight	Upper weight	Importance
x_1: CD (A/m^2)	In range	2.5	7.5	1	1	3
x_2: t_{EC} (min)	In range	5	15	1	1	3
x_3: C_o (μg/L)	In range	50	150	1	1	3
y_1 (μg/L)	Minimize*	0.50	41.92	1	1	3
y_2 (%)	Maximize	72.1	99.5	1	1	3
ENC (kWh/m^3)	Minimize	0.0011	0.0972	1	1	3
ELC (kg/m^3)	Minimize	0.0386	0.0994	1	1	3
OC (€/m^3)	Minimize	0.0431	0.1915	1	1	3

Optimum results. CD = 5.64 A/m^2, t_{EC} = 5 min, C_o = 112.30 μg/L, C_f = 6.90 μg/L, R_e = 93.86%, ENC = 0.01557 kWh/m^3, ELC = 0.05087 kgFe/m^3, OC = 0.0664 €/m^3, desirability = 0.84.

*The maximum concentration of arsenic for drinking water was targeted as 9.99 μg/L in the model.

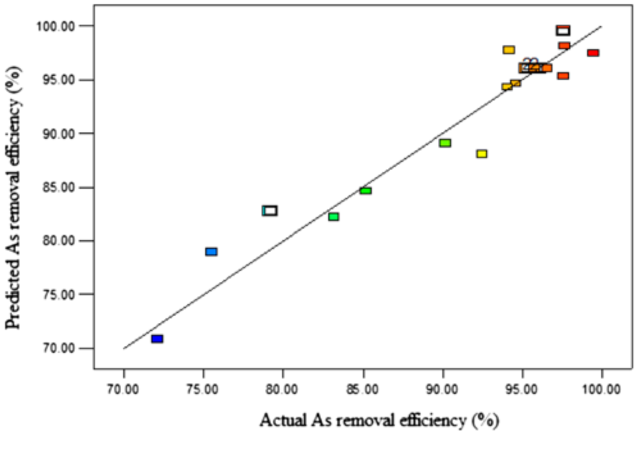

Fig. 1. Comparison of predicted-experimental values using Eqs. (11)–(15) for removal efficiencies of arsenic (R^2 = 0.963).

optimization techniques. The desirability value lies between 0 and 1 and it represents the closeness of a response to its ideal value. In this study, the desirability value obtained from the RSM was 0.84. The arsenic removal at the optimized conditions was found to be 93.86% which confirmed close to the predicted response using the RSM.

The actual and predicted arsenic removal efficiencies for Fe electrodes in the EC process were shown in Fig. 1. Actual values were the measured response data for a particular run, and the predicted values were evaluated from the model and generated by using the approximating functions. It was seen in Fig. 1 that the data points lied close to the diagonal line and the developed model was adequate for the prediction of each response.

The removal efficiencies of arsenic were found to increase with an increase in current density values at any value of initial pH and operating time. Generally, it was known that removal of arsenic increased with increasing Fe dosages in chemical coagulation (CC). It may be inferred from Fig. 2 that higher removal efficiency of arsenic was achieved at higher current density and operating time values. At higher current density, higher dissolution of electrode material (Faraday's law Eq. (22)) with higher rate of formation of iron hydroxides and some polymeric metal complexes resulted in higher removal efficiency of arsenic due to co-precipitation. These soluble species were useless for water treatment. More sludge was also produced from electrodes at higher current density values due to elevated dissolution rate of anode. Removal

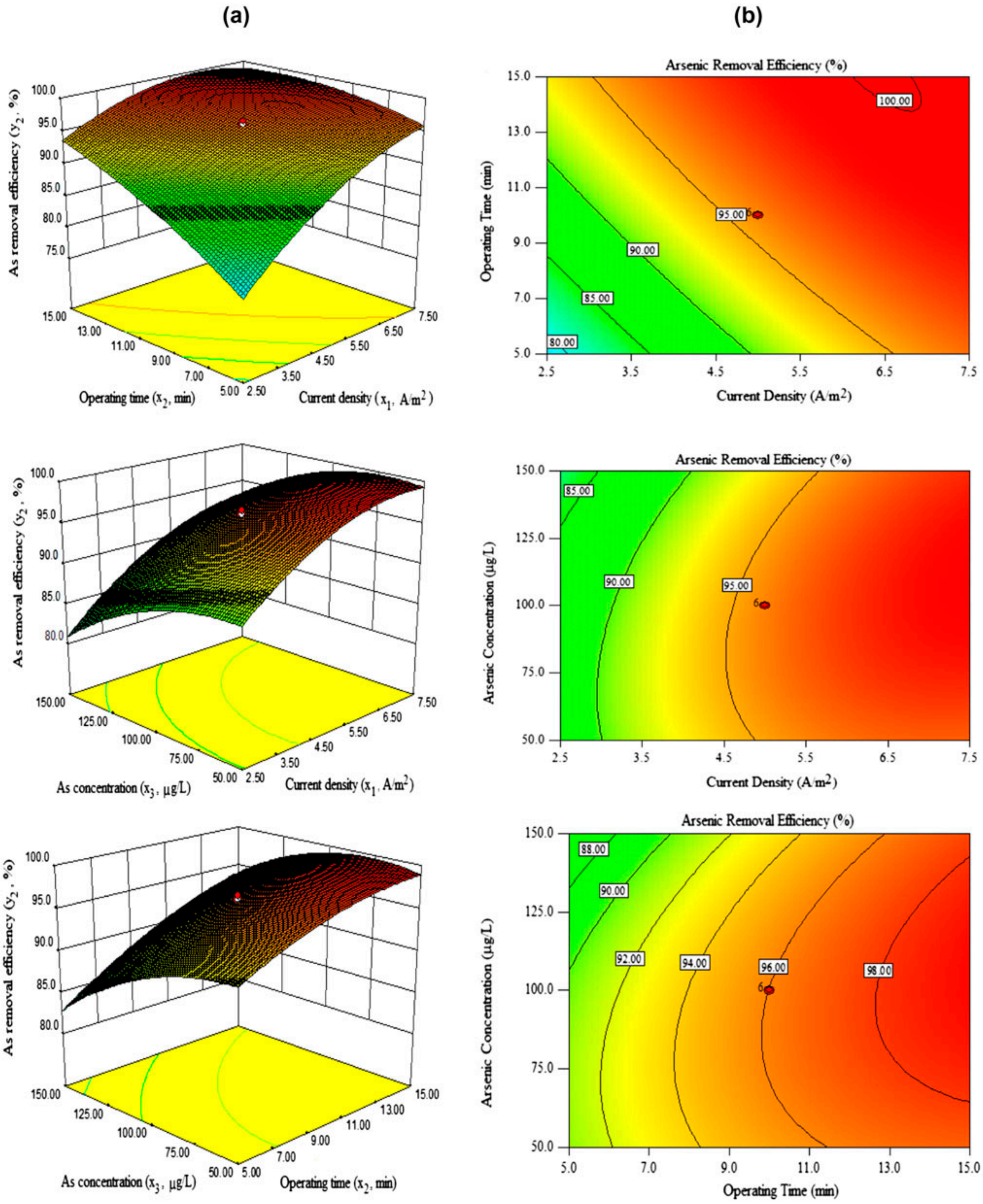

Fig. 2. (a) 3D response surface graphs with Fe electrodes for combined effect of current density–operating time, arsenic concentration–current density and arsenic concentration–operating time on removal efficiencies of arsenic and (b) contour plots of% arsenic removal efficiency representing operating time–current density, arsenic concentration–current density and arsenic concentration–operating time.

efficiencies of arsenic increased with respect to amount of sludge due to sweep coagulation at higher current density and operating time [39–42]. Maximum removal efficiencies of arsenic and OC at optimized operating conditions for Fe electrode in the EC process were 93.86% and 0.0664 €/m^3.

The comparison of EC and CC was done on the basis of iron dose. In the EC process, the amount of iron dissolved was calculated from experimental results. As(III) removals from the EC at 0.8, 5.0, and 9.2 A/m^2 (operating time of 10 min and 100 μg/L) were compared with the CC from the literature (2 mg/L, pH: 6–8 and operating time of 15 min). As removal increased from with increasing iron dosage with two methods. In the EC, removal of As(III) achieved was greater than 78% at 0.8 A/m^2 (36.66 mg Fe/L), 96.1% at 5.0 A/m^2 (43.35 mg Fe/L) and 98.6% at 9.2 A/m^2 (48.94 mg Fe/L) whereas in the CC up to 90–93% of As(III) removal with iron dose of 51.60 mg/L. Removal efficiencies of As(III) are much higher for the EC process than the CC process [10,43].

3.2. Comparisons of this study with literature

There is a limited study in literature about arsenic removal from drinking and groundwater in EC process. Balasubramanian et al. [33] carried out kinetic and statistical modeling for removal of arsenic from aqueous solution through EC using aluminum and mild steel electrodes. Three-level four factorial Box–Behnken experimental design was used in their study and removal efficiencies of arsenic as 86% for Fe electrode and 73% for Al electrode at 100 mg/L, 0.5 A/m^2, pH 7 and 50 min were predicted by the model. Matinez-Villafane and Montero-Ocampo [34] optimized energy consumption in arsenic removal from groundwater with Taguchi method by a continuous EC process. Operating conditions before the removal process were 131 μg/L arsenic concentration, low carbon steel ASTM 1018 electrodes, pH 7.22, 2.5 L/min of water flow rate and air 1.6 L/min of flow rate. After the process, the total energy consumption at optimized conditions (effluent concentration of <10 μg/L, interelectrode distance 3 mm, electrode area-treated volume ratio of 0.466 cm^{-1}, current density of 1.5 mA/cm^2) was 82.21 Wh/m^3. Majumder and Gupta [35] studied removal of arsenic from drinking water by EC using Fe electrodes via factorial design. In their study, predicted removal efficiency of arsenic as 98.6% at 1,180 μg/L, 5.26 A/m^2 (3 A and electrode area of 57 cm^2) and 2 min of operating time was obtained. The effluent concentration of arsenic at the optimum operating conditions was 17 μg/L for the model and 36 μg/L for the experimental value. Comparisons of this study with other works in the literature showed that this study achieved better results (i.e., effluent concentration of 6.9 μg/L and energy consumption of 15.6 Wh/m^3) than the others which were only concerned removal of arsenic at high concentrations.

4. Conclusions

EC was applied for the present investigation to remove arsenic from aqueous solutions. Experiments were carried out in a batch electrochemical reactor using sacrificial iron electrodes. Experimental runs were designed by RSM. The CCD matrix and RSM were applied to design the experiments to evaluate the interactive effects of three most important operating variables: current density (0.8–9.2 A/m^2), initial arsenic concentration (15.9–184.1 μg/L) and operating time (1.6–18.4 min) on the removal efficiency of arsenic in the EC process. The total 20 experiments were conducted in the present study for construction of a quadratic model. Very high regression coefficient between the variables and the response indicated excellent evaluation of experimental data by second order polynomial regression model. The model predicted a maximum removal of arsenic at optimized conditions as 93.86% with an OC of 0.0664 €/m^3. The mathematical approach in the EC process is useful for the treatment of drinking water containing arsenic.

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