

Linear and Nonlinear Isotherm Modeling of Nitrate Removal from Aqueous Solution by Alternating Current Electrocoagulation

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Abstract

Aims: Nitrates in drinking water which may come from nitrogen fertilizers applied to crops are a potential health risk. The present study was conducted to investigate the application of alternating current (AC) in electrocoagulation (EC) process for nitrate removal from aqueous solution and linear and nonlinear isotherm modeling. **Materials and Methods:** The experiments were performed in pilot scale. The effective parameters including solution pH, the initial concentration of nitrate, total dissolved solids, contact time, and current density were studied. **Results:** The obtained results showed that with increasing solution pH from 3 to 10, the sinusoidal removal efficiency was observed. With increasing current density from 0.5 to 2 A/cm², the nitrate removal efficiency was ascended from 32% to 58%. The optimum electrolyte was 2 g/L of NaCl. With increasing contact time and decreasing initial nitrate concentration, the nitrate removal efficiency was enhanced. In addition, the adsorption NO₃ by AC EC was preferably fitted with Langmuir isotherm. **Conclusion:** The results showed that the EC process could remove the nitrate to less than Iranian standard limit. The solution pH, current density, and contact time were showed the direct effect and initial concentration of nitrate depicted the reverse effect on nitrate removal efficiency.

Keywords: Adsorption kinetics, alternating current, direct current, electrocoagulation, nitrate removal

INTRODUCTION

Humans have altered the nitrogen cycle dramatically over the last half-century, and as a result, nitrate is steadily accumulating in our water. Fertilizer is the largest contributor to anthropogenic nitrogen worldwide; other major sources include animal and human waste, nitrogen oxides from utilities and automobiles, and leguminous crops that fix atmospheric nitrogen. These organic and inorganic sources of nitrogen are transformed to nitrate by mineralization, hydrolysis, and bacterial nitrification.^[1] Pollution of nitrate in surface and underground water resources is as a result of the agricultural activities, leakage from septic tank systems, landfills leachate, and sewage treatment plants.^[2] One of the proven effects in increasing of nitrate is the onset of the blue baby syndrome in infants and/or methemoglobinemia.^[3] This disease occurs when the nitrate ion in digestion system is restored to nitrite ion by bacteria, and hemoglobin converts to methemoglobin by nitrite entrance into the cardiovascular

system, which reduces oxygen transmission and causes death as a result of the patient's suffocation.^[4] Other effects of nitrate on human health include hypertension, excessive bigness, thyroid dysfunction, colon cancer, and increased mortality in children.^[5] It can also be referred to eutrophication phenomena.^[6] Given the importance of nitrate in human health and the protection of consumers against the harms, national and international organizations have proposed laws and guidelines for this type of pollutant in water. The World Health Organization has been developed and proposed the allowance of nitrate in drinking water to be 50 mg/L nitrate and US environmental protection

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agency has been developed and proposed a maximum nitrate release level of 10 mg/L in terms of nitrogen, equivalent to 3.43 mg/nitrate ion.^[7,8] Advanced processes used to remove nitrate from water include ion exchange, membrane processes, and electrochemical processes.^[9-11] However, all of these methods have high costs and waste disposal problems. In recent years, the application of the electrocoagulation (EC) process has been considered due to advantages such as short-term, reduced sludge, low investment costs, ease of use, and the lack of need for chemical agents.^[12] The EC process involves the creation of an electric current of electrodes, most of which are iron and aluminum in a reactor, which forms the coagulant and gas bubbles due to the anode electric oxidation.^[13] The instability mechanisms of this process are the compression of the dual electric layer, adsorption, charge degradation, burst coagulation, and bridging between particles.^[14]

This process has been successfully used in the removal of various pollutants such as turbidity, fluoride, sulfate, hardness, color, diazinon, and arsenic.^[15-18]

In 2002, a study has been carried out with the aim nitrate removal by an electrochemical process and EC by Kopalpar *et al.*, in Turkey and showed that nitrate removal has a direct relationship with pH and potential difference, and removal of nitrate to an allowable concentration has been accomplished at the pH range of 9–11 with efficiency 90%.^[19] Furthermore, a study has been carried out to remove nitrate by electrochemical and EC by DC power by Emamjomeh and Sivakumar shows that highest removal of nitrate is achieved at 90 min of electrolysis and a density of 2.5% with the efficiency of 90%.^[20] In another study to remove nitrate and arsenic from drinking water by Kumar and Goel, the results showed that the maximum nitrate removal efficiency was 84%.^[21]

A further study has been done on the removal of nitrate from Kerman water distribution network using DC power flow by Malkootian *et al.* (2011) that showed the removal efficiency of nitrate was 8.92%.^[14] Also in nitrate removal using DC power flow by Hashim *et al.*, in 2017, the concentration of nitrate was reduced after 55 min of electrolysis at pH of 7 from 100 to 15 mg/L.^[22] In another study by Hossini and Rezaee, the maximum nitrate removal efficiency was achieved 97% at electrolysis time 120 min and flow density of 0.14.^[23] Most reports indicate that using DC power creates an insulating layer on the surface of the cathode and reduces the flow between the anode and the cathode. Therefore, in this study, AC power was used for the EC process. In this study, the effect of different parameters including nitrate initial concentration, contact time, solution pH, current density, and electrolyte dosage on AC/EC process was survived. Furthermore, the linear and nonlinear isotherm modeling in removal of this contaminant was investigated.

MATERIALS AND METHODS

Materials

The NaNO₃ used for preparing the sample solution and chloric acid and sodium hydroxide 0.1N that used to adjust the pH

of the solution was purchased from Merck Co., Germany. The residual NO₃⁻ concentration was determined through cadmium reduction method using UV spectrophotometer (HACH, model DR5000).

Alternating current electrocoagulation setup

A pilot plant with 1.2 L volume and working volume of 1 L was used for removal of nitrate from aqueous solution. The pilot consist of three iron and aluminum electrodes with 10 cm × 10 cm dimensions and thickness of 2 mm and also AC power supply. A regulated alternating current (AC) was supplied from a source (0–3 A, 0–300 V, GOLD STAR Co. Iran) [Figure 1].

Experiments

A stock solution of nitrate 1000 mg/L was supplied using sodium nitrate to prepare the sample and the concentrations of nitrate of (50, 60, 70, and 80 mg/L) were prepared from this solution. A magnetic stirrer at 1000 rpm was used to mix the solutions. At all stages of the experiment, sodium chloride was used to increase solute solids, due to the low electrical conductivity of deionized water, after switching the electric flow with different flows density amperes from the middle of the reactor at intervals time. The extracted samples were filtrated by the paper filter (Whatman No. 42) to remove the flock after 30 min sedimentation time. The experimental conditions in nitrate removal by AC/EC were summarized in Table 1.

RESULTS

Effect of pH changes on removal of nitrate

The effect of pH (3, 4, 5, 6, 7, 8, 9, and 10) was investigated on nitrate removal efficiency. The obtained results of the initial pH changes on nitrate removal amount have been shown in Figure 2. Based on the results of this stage of the experiments, the maximum removal of nitrate at pH = 7, time 60 min, and the initial concentration of 70 mg/l has been 74.2%.

Effect of initial concentration of nitrate

The effect of initial concentration of NO₃⁻ (50, 60, 70, and 80 mg/L) was evaluated and the obtained data are illustrated in Figure 3. The reduses of NO₃⁻ removal efficiency with increasing initial nitrate concentrations are shown in Figure 3. The highest NO₃⁻ removal was 62% and occurred at 60 min, current density of 2A/cm², and solution pH of 7 with 50 mg/L of NO₃⁻ concentration.

Effect of current density

By changing applied current density (1, 1.5 and A/cm²), the NO₃⁻ removal efficiency by AC/EC was studied and the obtained data are shown in Figure 4. As depicted in Figure 4, with increasing applied current density from 1 to 2 A/cm², the NO₃⁻ removal efficiency was enhanced from 32 to 58%. The highest NO₃⁻ removal of nitrate was 58% and obtained at 60 min, current density of 2A/cm², and solution pH of 7 with 70 mg/L of NO₃⁻ initial concentration.

Table 1: Experimental conditions employed in nitrate removal by alternating current electrocoagulation

Variable	Solution pH	NO ₃ ⁻ concentration (mg/L)	Current density (A/cm ²)	Electrolyte dose (g/L)	Contact time (min)
Solution pH	3-10	70	2	2	60
Initial NO ₃ ⁻ concentration	7	50-90	2	2	60
Current density	7	70	1-2.5	2	60
Electrolyte dosage	7	70	2	1-3	60
Contact time	7	70	2	2	10-60

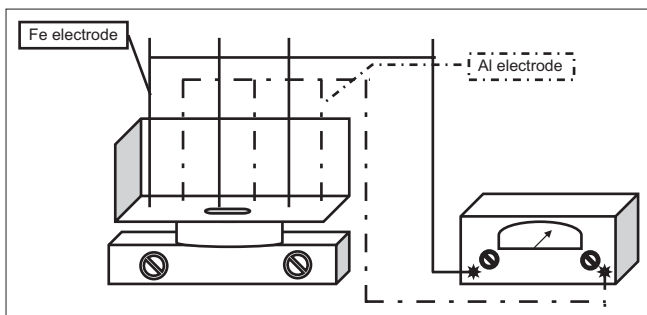


Figure 1: Schematic diagram of the electrocoagulation pilot plant

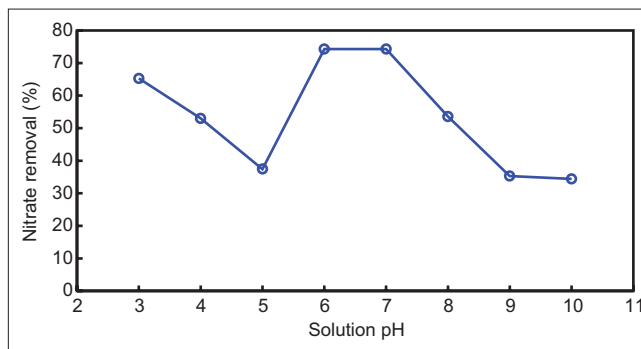


Figure 2: Effect of solution pH on nitrate removal (NO₃⁻ concentration: 70 mg/L, current density: 2 A/cm², contact time: 60 min, and electrolyte dose: 2 g/L)

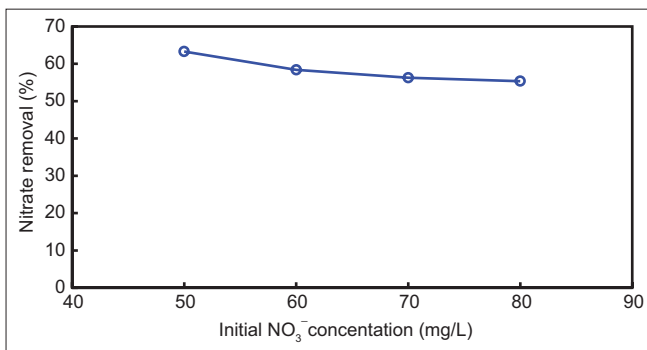


Figure 3: Nitrate removal efficiency as function of initial NO₃⁻ concentration (current density: 2 A/cm², contact time: 60 min, electrolyte dose: 2 g/L, and solution pH: 7)

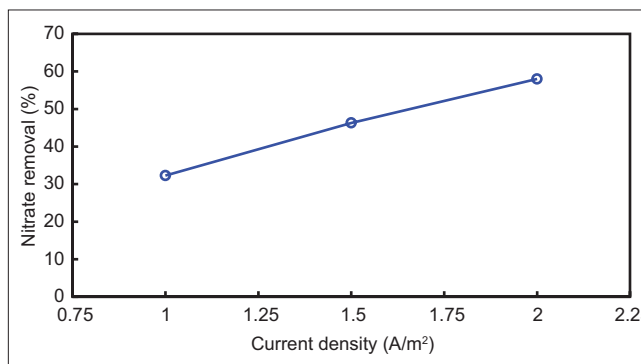


Figure 4: Effect of current density on the removal of nitrate (NO₃⁻ concentration: 70 mg/L, contact time: 60 min, electrolyte dose: 2 g/L, and solution pH: 7)

Effect of the electrolyte does

The concentration of NaCl (1.1.5, 2, 2, 5, and 3) g/L was evaluated, to investigate the effect of solution solids concentration on the removal efficiency in the EC process. The obtained results of concentration changes in NaCl on nitrate removal amount has been shown in graph.^[4] Based on the graph, the efficiency electrolytes is increasing by the concentration of 1–2, and the efficiency is reduced by the concentration above 2 [Figures 5 and 6].

Effect of contact time

Figure 7 shows the results of contact time on nitrate removal by AC/EC. As depicted in Figure 7, when the contact time was increased from 5 to 60 min, the NO₃⁻ removal rose up. The highest removal of NO₃⁻ (67%) was obtained at solution pH of 7, 60 min as contact time and the initial concentration of 70 mg/L with current density of 2 A/cm².

Adsorption isotherms

The NO₃⁻ adsorption capacity by AC/EC was tested with Freundlich and Langmuir isotherm to isotherms constant

estimation. The determination of isotherms constant was done at solution pH of 7, initial NO₃⁻ concentration of 50–80 mg/L, electrolyte does of 2 g/L, current density of 2A/m², and 60 min of contact time.

The Freundlich isotherm is an empirical model relates the adsorption intensity of the sorbent toward adsorbent. The isotherm is adopted to describe reversible adsorption and not restricted to monolayer formation. The nonlinear and linear form of Freundlich isotherm is shown in Equation (2 and 3), respectively.

$$q_e = K_f \cdot C_e^{\frac{1}{n}} \quad \text{Equation (2)}$$

$$\log q_e = \log K_f + \frac{1}{n} \log C_e \quad \text{Equation (3)}$$

where, q_e is the adsorption capacity (mg/g), C_e is NO₃⁻ equilibrium concentration (mg/L) and K_f and n are

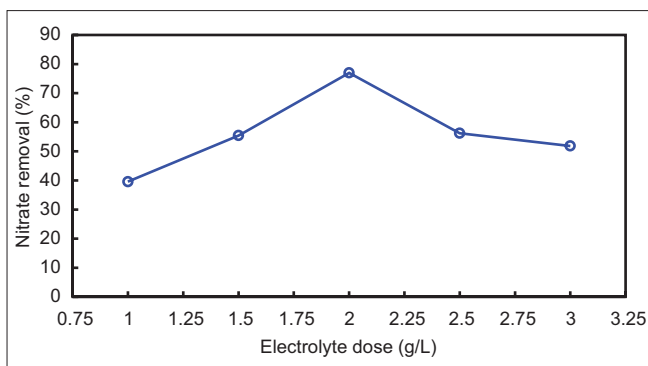


Figure 5: Effect of electrolyte dose on the removal of nitrate (NO_3^- concentration: 70 mg/L, contact time: 60 min, current density: 2 A/cm², and solution pH: 7)

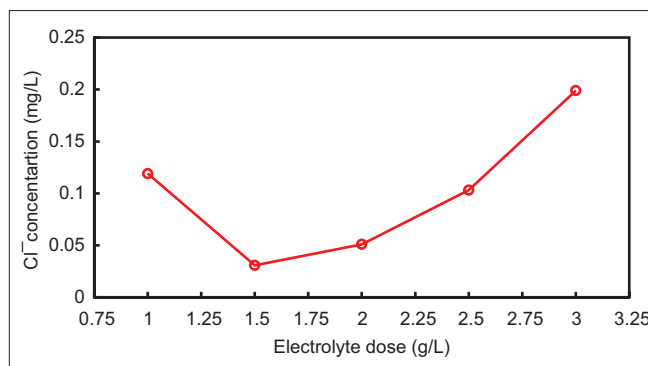


Figure 6: Effect of electrolyte dose on concentration of released Cl^-

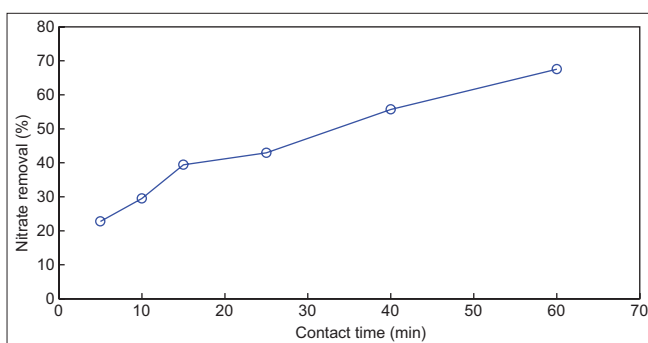


Figure 7: Nitrate removal by alternating current electrocoagulation as function of contact time (NO_3^- concentration: 70 mg/L, current density: 2 A/cm², electrolyte dose: 2 g/L, and solution pH: 7)

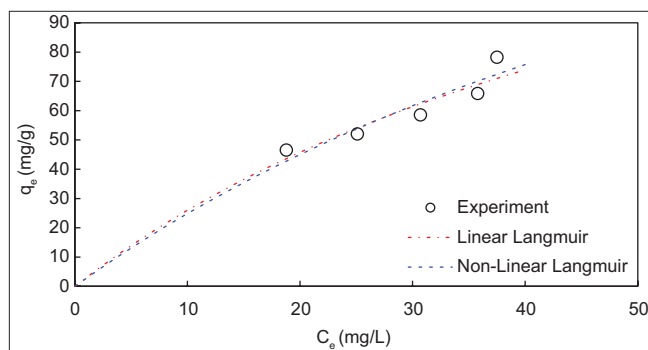


Figure 8: Linear and nonlinear Freundlich isotherm plot of NO_3^- adsorption during alternating current electrocoagulation

the Freundlich constants related to the adsorption capacity and adsorption intensity, respectively. In addition, at the above-mentioned condition, the Freundlich isotherm was applied for NO_3^- adsorption are shown in Figure 8.

The Langmuir isotherm is a valid monolayer sorption on a surface containing a finite number of binding sites. It assumes uniform energies of sorption on the surface and no transmigration of sorbate in the plane of the surface. The nonlinear and linear form of Langmuir adsorption isotherm is depicted in Equation (4 and 5), respectively.

$$q_e = \frac{Q_m \cdot K_L \cdot C_e}{1 + K_L \cdot C_e} \quad \text{Equation (4)}$$

$$\frac{C_e}{q_e} = \left(\frac{1}{K_L \cdot Q_m} \right) + \left(\frac{1}{Q_m} \right) \cdot C_e \quad \text{Equation (5)}$$

Where Q_m is maximum adsorption capacity (mg/g), K_L is sorption equilibrium constant (L/g). Table 2 summarizes the estimated constant values of Langmuir isotherm and Figure 9 showed the linear Langmuir isotherm with the experimental data for NO_3^- adsorption during AC/EC. Furthermore, the results of Freundlich and Langmuir isotherm calculations are summarized in Table 2.

The linearization of isotherm equations using such data transformations will implicitly alter the error structure and may also violate the error variance and normality assumptions of standard least squares. Many isotherm modeling studies showed that nonlinear data fitting methods yield mathematically superior isotherm coefficients compared to linear regression methods. Nonlinear optimization techniques have been applied to determine isotherm constants. An error function is defined to enable the optimization process to determine and evaluate the fit of the kinetic equation to the experimental data. In the present study, the coefficient of determination, r^2 (as error function) was calculated Equation (6).

$$r^2 = \frac{\sum (q_{e,Pre} - \overline{q_{e,Exp}})^2}{\sum (q_{e,Pre} - \overline{q_{e,Exp}})^2 + \sum (q_{e,Pre} - q_{e,Exp})^2}$$

Where $q_{e,Pre}$ is the equilibrium capacity obtained from the isotherm model, $q_{e,Exp}$ is the equilibrium capacity obtained from experiment, and $\overline{q_{e,Exp}}$ is the average of $q_{e,Exp}$.

DISCUSSION

pH is one of the important and effective factors on AC/EC process. The solubility and types of hydroxides in solution were affected by solution pH. Based on Figure 2, maximum removal efficiency was at pH = 7 and the minimum efficiency

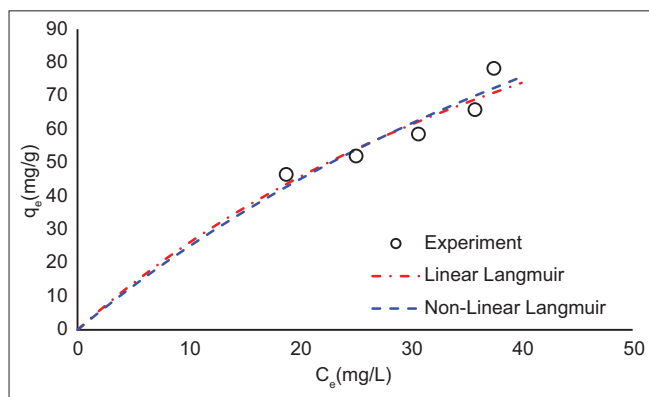


Figure 9: Langmuir isotherm obtained using the linear and nonlinear method for NO_3^- adsorption

Table 2: The estimated isotherms constant by linear and nonlinear approach

Isotherm type	Isotherm constant	Linear approach	Nonlinear approach
Freundlich	K_f	6.16	4.91
	$1/n$	0.67	0.74
	r^2	0.80	0.99
Langmuir	Q_m	192.21	238.42
	K_L	0.016	0.0112
	r^2	0.85	0.99

was at pH = 10. At solution, pH = 7, the dominant aluminum hydroxides species are including $\text{Al}(\text{OH})_3$, $\text{Al}(\text{OH})_3$, $\text{Al}(\text{OH})_4$, AlO , and $\text{Al}(\text{OH})_2$ are present in the solution and led to nitrate adsorption on the surface of hydroxides species.^[25] Furthermore, in the alkaline pH region, the surface charge of flock's comes to a negative charge and coincide with the decreasing with nitrate removal efficiency.^[24] The obtained results in this study are accordance with Vasudevan *et al.* on Cd removal using AC from the water. They reported that at initial Cd concentration of 20 mg/L, the highest removal efficiency was 97.5% and obtained at solution pH of 7 with applying current density of 0.2 A/dm².^[26] Cerqueira *et al.* survived the effects of direct and alternative current on the oily water treatment and depicts that at solution pH of >6, the removal efficiency of oils and greases was 96%.^[27] In addition, Vasudevan *et al.* reported that the highest iron removal efficiency by AC/EC was 99.6% and obtained at a current density of 0.06 A/dm² and solution pH of 7.^[28] Furthermore, Seid Mohammadi *et al.* on removal NO_3^- using EC from the water. They reported that at initial concentration of 100 mg/L, the highest removal efficiency was 88% and obtained at solution pH of 8.^[29] Based on Figure 3, the reduses of NO_3^- removal efficiency with increasing initial nitrate-concentration. When initial concentrations were higher, more metallic hydroxides were needed to reduce NO_3^- concentration. According to Faraday's law, when current density and time are the same, the solution. As a result, ions produced at high initial concentrations NO_3^- are insufficient to reduce all of the NO_3^- ions.^[30]

In all of the electrochemical processes, the current density is the most important parameter for controlling the reaction rate inside the electrochemical reactor. From Figure 4 results, it is found that as current density increases, removal of nitrate also increases. The nitrate removal efficiency is related to amount of produced coagulant ($\text{Al}(\text{OH})_3$), and its concentration can be estimated by Faraday's law according to Equation (1).

$$m = \frac{M \cdot I}{F \cdot z} \cdot t \tag{Equation (1)}$$

where m is mass of the coagulant produced at the anode (g), M is aluminum atomic weight (56 g/mol), I is current density to an electrode, F is the Faraday constant (96,485 C/mol), z is the aluminum valence number (z : 3), and t (s) is total contact time. According to the above equation, the coagulant mass produced is related to applied current density and contact time. As the current density was increased from 1 to 2 a/cm², the amount of coagulation produced enhanced from 0.33 to 0.67 and resulted in higher available active sites for NO_3^- adsorption.^[31] In other stages of the experiment, the current density of 2 A/cm² was used. The obtained results of this study are in line with Vasudevan *et al.* study on the Cd^{2+} removal using AC/EC. They reported that with increasing current density from 0.1 to 0.5 A/dm², the Cd removal efficiency was increased too.^[26] In addition, Keshmirizadeh *et al.* reported that the chromium removal was 20% at applied current density of 40 A/m² enhanced to 99.9% at 230A/m² current density.^[32] According to Figure 5, the efficiency removal is increasing by the concentration of 1–2 electrolyte and the efficiency is reduced by the concentration above 2. Furthermore, Figure 6 shows by increasing the electrolyte dose from 1 to 2 g/L, the amount of chlorine did not meet the full reaction threshold. Therefore, the NO_3^- removal was increased, but after that, the chlorine reaches to the full reaction, reacts with the coagulator, and hence the amount of remained chlorine is increased, which has reduced the efficiency of nitrate removal, due to its competitive specification in the adsorption by the chelate. The obtained results of this stage not confirmed with Keshmirizadeh *et al.* reported that the chromium removal and Malakootian *et al.*, (2011) on the removal efficiency nitrate.^[32] In the EC process, the time of electrolysis affects the treatment efficiency. Figure 7 shows the relation between the reaction time and the efficiency of nitrate removal. As it can be seen in these figure, as the reaction times increases, the removal efficiency increases. According to the Faraday's law, the reaction time is an effective parameter on coagulant produced and determines the nitrate removal efficiency.^[32] The obtained results of this study are consistent with the Cerqueira *et al.* study on treatment of oily water through EC with direct and alternative current, as well as Mansour *et al.* (2012) study on nickel removal from drinking water.^[27,33] The equilibrium relationship between adsorbate concentration in liquid phase and on adsorbent's surface is demonstrates with the adsorption isotherms. The nonlinear models are fitted to the experimental data using the solver add-in function in Microsoft Excel Spreadsheet to adjust the initial calculated constant so that the models can

be the best fit to the experimental data.^[34] Table 2 shows the nonlinear and linear isotherms coefficients for the Freundlich and Langmuir isotherm during nitrate adsorption by AC/EC. As depicted in Table 2, the isotherm constants obtained from the nonlinear methods differed even when compared with the results of linear approach, which had the highest coefficient of determination for any isotherm [Table 2]. The adsorption of NO_3^- was preferably fitting Langmuir adsorption isotherm, the obtained results in this study are accordance with Vasudevan *et al.* on Cd removal using AC from the water.^[26]

CONCLUSION

The present study demonstrated the applicability of alternation current in EC process by Al as a cathode and Fe anode for NO_3^- removal. The effect of solution pH, NO_3^- concentration, current density, electrolyte dose, and contact time was assessed, and also isotherm study of NO_3^- was performed. According to the obtained data, the following items can be concluded:

- It was observed that pH and current density an important role in the removal of nitrate
- The NO_3^- adsorption was preferably fitted with Langmuir adsorption isotherm.

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Nil.

Conflicts of interest

There are no conflicts of interest.

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