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Optimizing electrocoagulation and electro-Fenton process for treating car wash wastewater

Seyyedali Mirshahghassemi[1], Behnoush Aminzadeh[2*], Ali Torabian[3], Kamelia Afshinnia[1]

[1]Graduate Research Assistant, Center for Environmental Nanoscience and Risk (CENR), Department of Environmental Health Sciences, Arnold School of Public Health, University of South Carolina, Columbia, South Carolina 29208, United States
[2]Assistant Professor, Department of Environmental Engineering, Graduate Faculty of Environment, University of Tehran, Iran
[3]Professor, Department of Environmental Engineering, Graduate Faculty of Environment, University of Tehran, Iran

Abstract

Background: Car wash wastewater contains several contaminants such as organic matter, oil, grease, detergents and phosphates, all of which are harmful for the environment. In this study, the application of electrocoagulation (EC) to treat car wash wastewater has been studied, and the operating parameters optimized. The electro-Fenton (EF) for further contaminant removal was also investigated.

Methods: In EC process, the effect of pH, current density, and the reaction time of the removal efficiency of chemical oxygen demand (COD), phosphate, and turbidity were investigated using the response surface methodology (RSM). The electrochemical cell consisted of four iron electrodes that were connected to a power supply using a monopolar arrangement. In the EF process, the effect of pH, reaction time, and hydrogen peroxide concentration on COD removal efficiency were probed.

Results: The optimum pH, current density, and the reaction time for the EC process were 7.3, 4.2 mA cm^{-2} and 20.3 minutes, respectively. Under these conditions, the COD, phosphate, and turbidity removal percentages were 80.8%, 94.9% and 85.5%, respectively, and the specific energy consumption was 1.5 kWh m-3. For the EF process, the optimum pH, reaction time, current and hydrogen peroxide dosage were 3, 10 minutes, 2 A and 500 mg L^{-1}, respectively. The EF showed higher COD removal efficiency (85.6%) with a lower specific energy consumption (0.5 kWh m^{-3}) and reaction time compared to the EC.

Conclusion: This study shows that both EC and EF can effectively treat car wash wastewater with high removal efficiency within a short reaction time.

Keywords: Electrocoagulation (EC), Electro-Fenton (EF), Response surface methodology (RSM), Car wash wastewater

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***Correspondence to:**
Behnoush Aminzadeh
Email: bamin@ut.ac.ir

Introduction

Water is the most abundant substance on the planet; however, most of it is sea water and very difficult to use in industrial activities. Therefore, it is crucial to take care of the finite water resources. But many industries, such as the car wash industry, consume huge amounts of water and release the wastewater into the environment (1). Car wash wastewater contains various contaminants such as organic matter, oil, grease, detergents, phosphates, and hydrofluoric acid, all of which are harmful for the environment (1,2). Earlier it was reported that the total suspended solids, biochemical oxygen demand (BOD), chemical oxygen demand (COD), and oil and grease concentration in the automobile service stations were in the range of 610–4950 mg L^{-1}, 75–570 mg L^{-1}, 270–1640 mg L^{-1} and 14–420 mg L^{-1}, respectively, which is higher compared to the municipal wastewater (3, 4). For example, for medium municipal wastewater, the concentration of grease and suspended solids is 100 and 200 mg L^{-1}, respectively, figures that are significantly higher compared to the reported data for car wash wastewater.

Different treatment processes such as membrane separation (5-7), chemical oxidation (8), coagulation and activated sludge (2) have been reported for the treatment of car wash wastewater. However, there are some disadvantages associated with these techniques. For example, the membrane processes' main drawback is membrane fouling, which can decrease the efficiency of the process (1). Coagulation generates large amounts of sludge and increases the total dissolved solids in the effluent (9).

Electrocoagulation (EC) is a simple, reliable and cost-effective process that has the potential to be a distinct, cost-effective, and environment-friendly choice for wastewater treatment (10). The EC has been used to treat different types of wastewater including saline wastewater (11), removal of humic acid (12) and arsenic (13).

Another approach to electrochemical treatment is indirect electrolysis, which generates chemical oxidizing agents (e.g., hydrogen peroxide) to react with pollutants (14). The electro-Fenton (EF) process, in which Fenton's oxidation and EC happen simultaneously, shows excellent removal efficiency for treating various wastewater types (15-18). In this process, hydroxyl radical (°OH), a highly oxidizing agent with a high oxidative potential (E° = 2.80 V), reacts rapidly with organic compounds (19). This process is most effective under acidic conditions, in which organic compounds are removed in two stages of oxidation and coagulation (20).

According to Mollah et al, iron as anode electrode can produce iron hydroxide, $Fe(OH)_2$ or $Fe(OH)_3$ (21). Depending on the aqueous pH, ferric ions may form monomeric and polymeric hydroxyl complexes, namely, $FeOH^{2+}$, $Fe(OH)_2{}^{+}$, $Fe(OH)^{-}{}_4$, $Fe_2(H_2O)_8(OH)_2^{4+}$, $Fe(H_2O)_5(OH)^{2+}$ and $Fe(H_2O)_4(OH)_2^{+}$. These suspended solid hydroxides/polyhydroxides/polyhydroxymetallic compounds have a strong affinity with dispersed particles and the counter ions to cause coagulation or adsorption (22-24).

In the EF process, hydroxyl radicals are produced by the reaction between ferrous ions and hydrogen peroxide, which can degrade organic compounds (25).

One advantage of the EF process over the conventional Fenton process is the reduction of ferric ion to ferrous ion, which can reduce iron sludge production (25). Despite intensive scientific research on industrial wastewater treatment by EC and EF, there are few reported publications on the application of these two techniques for treating car wash wastewater. The aim of this study is to investigate the performance of the EC and EF processes using iron electrodes in a batch mode operation for treating car wash wastewater. Studying both the EC and EF processes can lead to a better understanding of the efficiency of these two treatment techniques. Moreover, in this study, the response surface methodology (RSM) has been used to optimize the reaction conditions for EC process.

Methods

Car wash wastewater

The wastewater was collected from the top of a settling tank at the Jamalzadeh Car Wash Company, located in Tehran. Characteristics of the collected wastewater samples for the EC and EF processes are presented in Table 1.

Experimental method

The electrochemical cell consisted of four iron electrodes that were connected to a power supply (Zhaoxin) using a monopolar arrangement. Magnetic stirring at 150 rpm was maintained for a homogenous solution. The batch reactor contained 3.6 L of the wastewater sample. The dimension of the iron plates was 130 × 90 × 1 mm, and the total surface area was 936 cm^2. The pH of the solution was adjusted using NaOH or H_2SO_4. Following the experiment, the solution was collected from the middle of the reactor for further analyses without any post-treatment. COD and phosphate concentrations were measured using DR 5000 (HACH). The pH, total dissolved solid (TDS), and turbidity were measured by the Metrohm 691 pH meter (HACH), SensION378 (HACH) and DR2000 (HACH), respectively. The analyses were carried out according to *Standard Methods for Examination of Water and Wastewater* (26).

Experimental design

The classical or conventional method of experimentation, in which one parameter is changed, while the other parameters are fixed, is time-consuming and does not show the effect of the interactions between the factors (27). On the other hand, the RSM is attractive for its ability to evaluate the relative interaction of parameters. The central composite design (CCD), an experimental design for RSM, was performed using MINITAB software (version 16). The removal percentage of COD, phosphate, and turbidity had been taken as the system response. The levels for the main variables X_i (X_1 [initial pH], X_2 [current density] and X_3 [reaction time]) were coded as x_i according to the following equation:

$$x_i = \frac{x_i - x_0}{\Delta X} \tag{1}$$

where X_i is the dimensionless value of an independent variable, X_i is the real value, X_0 is the real value of the centered point, and ΔX is the step change. Variables, experimental ranges and levels are presented in Table 2. Experimental data were fitted to a second-order polynomial model and regression coefficients were obtained. The generalized second-order polynomial model used in the response surface analysis was:

$$Y_i = b_0 + \sum b_i x_i + \sum b_{ii} x_i^2 + \sum b_{ij} x_i x_j \tag{2}$$

Where b_0, b_i, b_{ii}, and b_{ij} are the regression coefficients for intercept, linear, quadratic and interaction terms,

Table 1. Characteristics of collected car wash samples for EC and EF processes

Parameter	Value
pH	7.2-7.6
Turbidity (NTU)	118-1400
Phosphate (mg L^{-1})	11.4-38.2
COD (mg L^{-1})	610-2619
TDS (mg L^{-1})	209-110
Conductivity (µs cm^{-1})	419-2200

Table 2. Experimental range and the test levels

Variables	Factors	Ranges and levels				
		-1.6	-1	0	1	+1.6
Initial pH	x_1	5.7	6.3	7.3	8.3	9.0
Current density (mA cm^{-2})	x_2	-0.7	1	3.7	6.4	8.1
Reaction time (min)	x_3	10.3	15	22.5	30	34.8

respectively. Y_i is the percentage removal of COD (Y_1), phosphate (Y_2) and turbidity (Y_3). The models' adequacy were also evaluated through the analysis of variance (ANOVA).

Results

Development of regression model equation and validation of the model

Based on the CCD experimental design results, the RSM was applied to develop the polynomial regression equations and find out the relation between the output response (removal efficiency) and the input factors (experimental variables) (Eq. 2). The full quadratic regression model consists of linear, square, and interaction terms. The removal percentage of COD (Y_1,%), phosphate (Y_2, %), and turbidity (Y_3, %) in terms of coded factors are given by the following equations 3, 4 and 5, respectively.

$$Y_1 = 82.673 + 0.086x_1 + 24.803x_2 + 7.913x_3 - 1.117x_1^2 - 16.194x_2^2 - 6.122x_3^2 + 5.575x_1x_2 - 1.12x_1x_3 + 0.200x_2x_3 \quad (3)$$

$$Y_2 = 93.614 - 0.992x_1 + 14.166x_2 + 13.683x_3 - 1.084x_1^2 - 10.387x_2^2 - 9.671x_3^2 - 6.838x_1x_2 + 9.613x_1x_3 - 2.263x_2x_3 \quad (4)$$

$$Y_3 = 83.059 - 5.204x_1 + 21.601x_2 + 5.100x_3 - 4.965x_1^2 - 12.224x_2^2 - 4.890x_3^2 - 2.275x_1x_2 + 0.625x_1x_3 - 3.125x_2x_3 \quad (5)$$

Table 3 presents the ANOVA results for responses. The significance of the second-order regression models was determined by Fisher's variance ratio test (F-test), lack of fit, and coefficients of determination between the experimental and predicted values (R^2). F-value, defined as the ratio of regression's mean square due to the residual error, is greater than the tabulated value (2.352 at 95% significance), showing the desirability of the regression model (28). The *P* value is lower than 0.05 showing that the model is also statistically significant (29). The values of R^2 are greater than 80%, suggesting the desirability of the models (30). The R-squared for COD removal implies that 96.3% of the variations can be explained by the independent variables, and other R-squareds have the same definitions.

Determination of importance of model terms.

The Pareto analysis calculates the percentage of effectiveness of each factor (P_i) on the response (31):

$$Pi = \left(\frac{b_i^2}{\sum b_i^2}\right) \times 100 \quad (i = 0) \qquad (6)$$

Based on the Pareto analysis results, the current density (b_i, CD) has the highest percentage of effectiveness on COD removal (60.8%) and turbidity removal (63.2%) (Figure 1). The pH has the lowest impact, which is in agreement with the results reported by Amani-Ghadim et al (28).

Effect of operating parameters on COD removal

Figure 2 presents the three-dimentional (3D) response surface and two-dimentional (2D) contur plot of COD removal, which are graphical representations of their corresponding regression equation (Eq. 1). The 3D response surface can be used to estimate the removal efficiency, and the 2D contur plot shows the relative effects of any two variables, while the third variable is

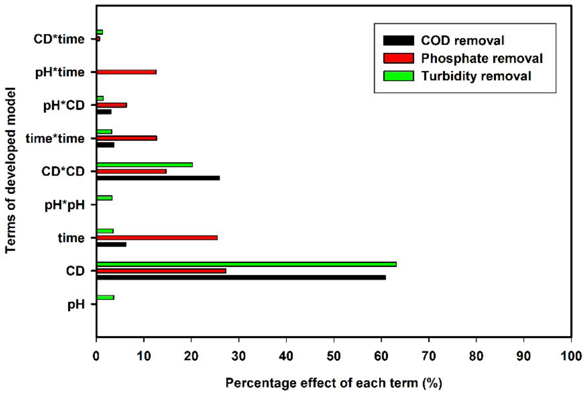

Figure 1. Percentage effect of each model term obtained by Pareto analysis.

Table 3. ANOVA for the RSM model of COD, phosphate and turbidity removals

Source	Degrees of freedom	Sum of squares	Mean square	*F* value	*P* value
		COD removal[a]			
Model (regression)	9	8216.75	912.97	25.97	0.0001
Residual	9	316.42	35.16		
Lack of fit	4	245.63	61.41	4.34	0.07
		Phosphate removal[b]			
Model (regression)	9	7402.65	822.52	13.71	0.0002
Residual	9	540.07	60.01		
Lack of fit	4	475.13	118.78	9.15	0.016
		Turbidity removal[c]			
Model (regression)	9	6505	722.78	11.19	0.001
Residual	9	581.3	64.59		
Lack of fit	4	399.6	99.91	2.75	0.148

[a] R^2 = 96.3%; [b] R^2 = 93.2%; [c] R^2 = 91.8%.

kept constant. To study the effect of pH on COD removal efficiency, the pH was varied between six and eight. The optimum pH for maximum COD removal is 7.3, which provides the maximum flocculation of $Fe(OH)_2$ and $Fe(OH)_3$ (32). Under optimum conditions, the COD removal percentage was 80.8%.

Effect of operating parameters on phosphate removal.

Figures 3 and 4 show the response surfaces and contur plots of phosphate removal. The use of the optimum condition led to a maximum phosphate removal of 94.9%.

Effect of operating parameters on turbidity removal

Figure 5 shows that a decrease in the current density reduces the turbidity removal efficiency. In this study, the turbidity removal percentage was 85.5% under the optimum condition. Figure 5 also illustrates the effect of initial pH on turbidity removal efficiency.

Optimization of the EC process

Table 4 shows the tabulated values for the optimum reaction condition and the expected and observed COD, phosphate, and turbidity removal. In this study, the desired values of COD, phosphate, and turbidity removal, and energy consumption—85%, 95%, 85% and 1.5 kWh m^{-3}, respectively—were defined as the desired target. The energy consumption under optimum conditions was 1.66 kWh m^{-3}. The pH of the car wash wastewater was near the neutral pH, indicating that pH adjustment was not necessary.

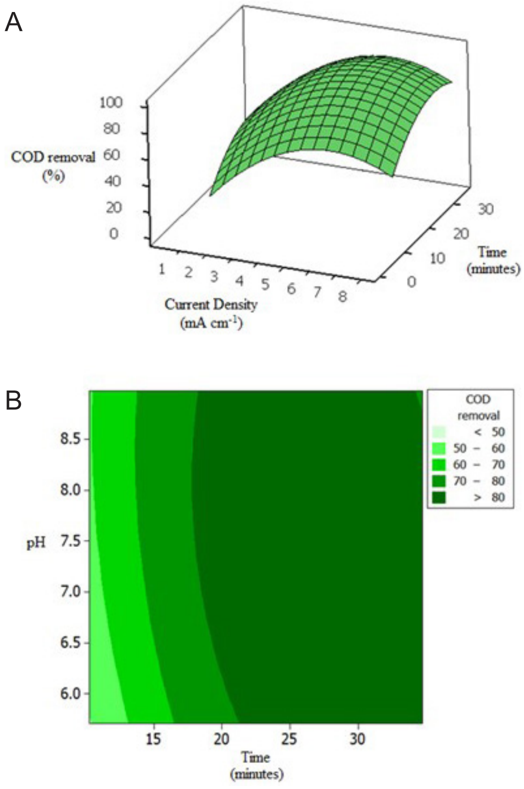

Figure 2. Surface plot and the contour plot for COD removal. (A) pH= 7.3 and (B) current density= 4.2 mA cm^{-1}.

Car wash wastewater treatment by EF

Despite the excellent removal of COD by the EC process, the EF process was performed too. This process is

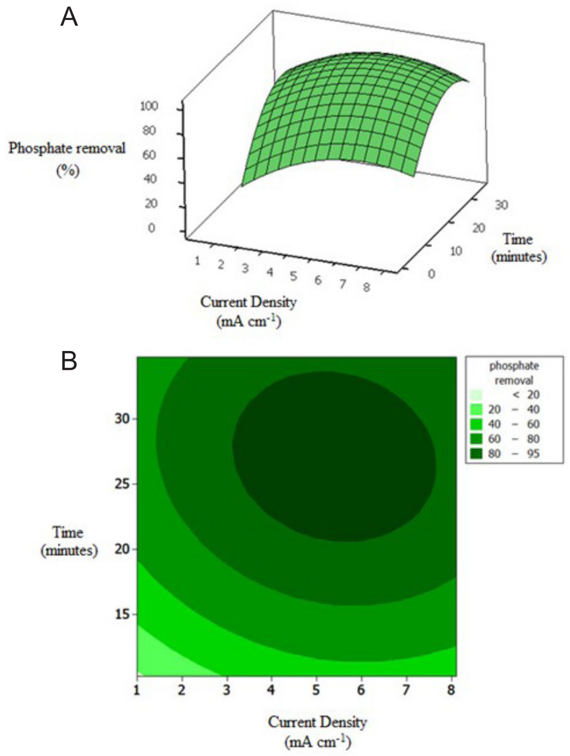

Figure 3. Surface plot and the contour plot for phosphate removal (pH = 7.3).

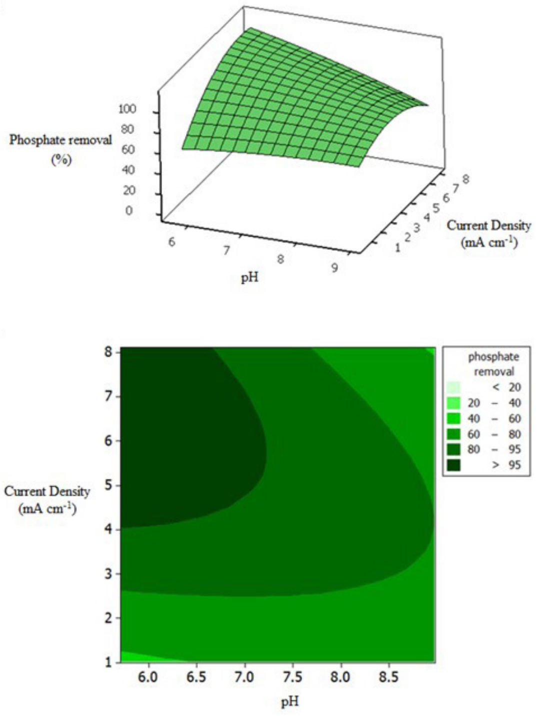

Figure 4. Surface plot and the contour plot for phosphate removal (reaction time = 20 min).

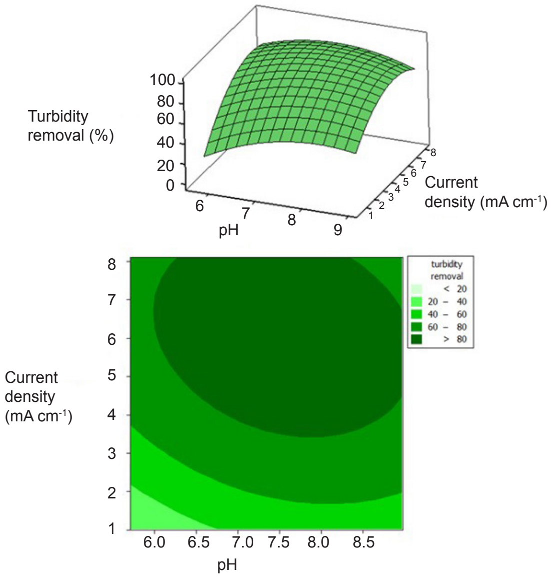

Figure 5. Surface plot and the contour plot for turbidity removal (reaction time = 20 min).

Table 4. Optimum reaction conditions for COD, phosphate and turbidity removal for EC process

Variables	Optimum values
Initial pH	7.3
Current density (mA cm$^{-2)}$	4.2
Reaction time (min)	20.3
COD removal (predicted) (%)	83.8
COD removal (obsereved) (%)	80.8
Phosphate removal (predicted)	91.4
Phosphate removal (obsereved) (%)	94.9
Turbidity removal (predicted) (%)	84.7
Turbidity removal (obsereved) (%)	85.5

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recognized to be most efficient under acidic conditions (33). In this study, the pH range of 2-5 was tested. The maximum COD removal was 90.4% at pH = 3 after 20 minutes of the reaction (Figure 6A).

The effect of hydrogen peroxide concentration on COD removal was studied using the optimum pH. An increase in the H_2O_2 concentration from 100 to 500 mg L^{-1}, COD removal increased from 81.8% to 90.4% (Figure 6B). At a higher concentration of H_2O_2, COD, the removal efficiency decreased, which could be due to the hydroxyl radical scavenging effect of H_2O_2 and the recombination of hydroxyl radicals (34). The optimum conditions for pH, reaction time, current and hydrogen peroxide dosage were 3, 10 minutes, 2 A, and 500 mg L^{-1}, respectively. Under these conditions, a COD removal efficiency of 85.6% with a specific energy consumption of 0.5 kWh m^{-3} was obtained.

Discussion

Our results show that a lower current does not provide enough iron ion to completely destabilize the suspended oxide particles (35). Based on Faraday's law, an increase in the current density and reaction time increases the amount of iron hydroxides, resulting in higher removal efficiency of COD and turbidity via co-precipitation and sweep coagulation (36,37). Similar to the COD removal mechanism, increasing the current density and reaction time can increase phosphate removal efficiency by adsorption into the iron hydroxide, as well as precipitation of metal phosphate. Generally, a higher current density is favourable for pollutant removal. Our study shows that a higher COD and turbidity removal efficiency can be achieved in neutral and weak alkaline solutions, which is in agreement with the results reported by Bayramoglu et al (38). Phosphate removal was higher under acidic conditions, which aligns with results reported by Irdemez et al (39).

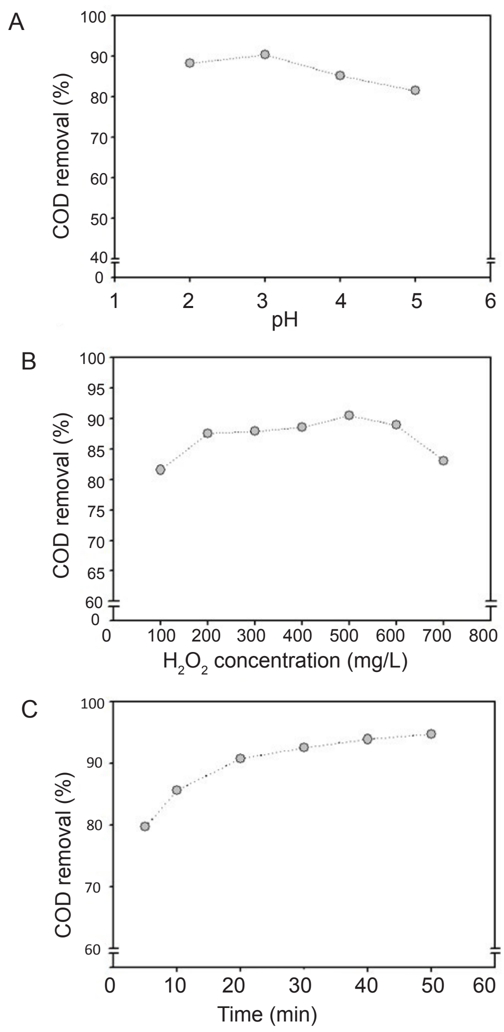

Figure 6. COD removal efficiency as a function (A) pH (current: 2 A, H_2O_2 concentration: 500 mg L^{-1} and reaction time: 20 min) (B) H_2O_2 concentration (current: 2 A, pH: 3 and reaction time: 20 min) and (C) reaction time (pH = 3, current= 2 A and H_2O_2 concentration = 500 mg L^{-1})

For the EF process, a pH greater than 3 decreased the COD removal efficiency. At a higher pH, the oxidation efficiency of EF process may decrease due to the formation of low active $Fe(OH)_{3,}$ which has a lower tendency to react with hydrogen peroxide (40). In the EF process, the hydroxyl radicals are produced by a reaction between ferrous ions and hydrogen peroxide, degrading the organic compounds (25). In addition, by increasing the reaction time, the hydroxyl radicals have a greater chance to react with contaminants, leading to higher COD removal (Figure 6C). Our results showed that the EF process can provide similar COD removal efficiency compared to the EC process with less energy consumption and reaction time. However, a trade-off exists between the complexity of the EF compared to the EC and the needed energy and time, which must be optimized on a case-by-case basis.

Conclusion

An electrochemical cell with four iron electrodes using a monopolar arrangement was used to treat car wash wastewater. The performance of iron electrodes was modelled and optimized using the RSM. The R-squared for COD, phosphate, and turbidity removal was 96.3%, 93.2% and 91.8%, respectively, which satisfies the adjustment of the full quadratic regression model with the experimental data. For the EC process, the optimum pH, current density, and reaction time were 7.3, 4.2 mA cm^{-2} and 20.3 minutes, respectively. Under these conditions, the removal percentage of COD, phosphate, and turbidity was 80.8%, 94.9% and 85.5%, respectively. For the EF process, pH =3 is the optimum pH for COD removal. This study shows that both the EC and EF can effectively treat car wash wastewater with a high removal efficiency within a short reaction time.

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Ethical issues

There were no ethical issues involved in the preparation of this article.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

All authors were involved in the study design, data collection, and article approval.

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