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Performance Evaluation of Electrocoagulation in Removing COD from Car Wash Wastewater

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Background: Considering the increased demand for cars in different countries during recent years, using car washes for washing vehicles has received a lot of attention. This study aimed to assess removing chemical oxygen demand (COD) from car wash effluent using the electrocoagulation method.
Methods: A reactor with dimensions of 40 cm * 50 cm * 50 cm of Plexiglas with a volume of 90 L equipped with an electric current generator and an electrode was used connected to the DC current generator in the form of Al-Al. The response surface method (RSM) was applied to optimize the factors affecting COD removal in the electrocoagulation process. For this purpose, D-optimal was utilized to optimize the experiments. The effects of measurable factors such as electrolysis time (X_1), current density (X_2), and aeration time (X_3) were examined to check COD removal.
Results: According to the results, the optimal operating conditions for COD removal during electrolysis (30 min) were as follow: the current density was 18.75 A/m^2, and the aeration time of 30 min was 48.51%.
Conclusion: In conclusion, electrocoagulation is, to some extent, a reliable and environmentally compatible technique for car wash wastewater treatment.

1. Introduction

Car wash wastewater is the result of washing different parts of cars, such as the exterior parts of the engine. Car wash wastewater is one of the most polluted with high impurity intensity since it contains sand, particles, oils and greases, surfactants, detergents, phosphates, and solvents. Direct disposal of this wastewater intensifies the pollution of natural waters and increases the additional load. Moreover, it harms the efficiency of wastewater treatment processes due to impurities. Therefore, eliminating environmental pollution in car washes is essential [1]. Consequently, the removal of significant parameters of car wash effluent by various methods and with emphasis on the reuse of wastewater has been considered in several studies [2]. One of the most significant indicators of car wash pollution is COD. Effluent pollution is affected by external materials that enter the water suspended or dissolved, resulting in effluent production and pollution. The higher amount of these materials in wastewater cause more pollution. Therefore, measuring and determining the amount of COD is the main issue in wastewater pollution. According to the International Carwash Association (ICA) reports, important pollutants in car wash wastewater include detergents, COD, oils, fats, heavy metals, etc. [3,4]. Many car washes discharge their wastewater directly into municipal wastewater or absorption wells without pre-treatment. Car wash wastewater is classified in the category of industrial wastewater. Also, the high level of pollution, the variety of pollutants in this type of wastewater, and the large volume of used water make purifying process challenging because of compounds with low degradability. Various methods have been suggested for purifying this type of effluent. Wastewater is typically treated by physical and chemical processes, including chemical coagulation, adsorption on activated carbon [5], ultrafiltration, chemical oxidation, ozonation [6], and biological processes, which often create major problems. Biological purification processes are ineffective in dye removal because dyes are toxic to the organisms in the process and are highly structured polymers with low degradability [7]. The chemical coagulation process is a source of contamination due to adding huge amounts of chemical material and large amounts of sludge [8]. Furthermore, the adsorption process with activated carbon is costly; therefore, its reduction is difficult, and waste disposal costs are high [6]. The charges of ozonation and ultrafiltration are generally more than the coagulation process. Chemical decomposition by oxidizing factors, for example, chlorine which is the most substantial and impressive agent, causes highly toxic compounds, such as organochlorine, which is carcinogenic [5]. Advanced oxidation methods, for example, ozonation, UV/O_3, photooxidation by UV/H_2O_2, UV/TiO_2, and Fenton agent, require adding chemicals and cause secondary contamination [6,7]. In recent years, electrocoagulation (EC) has been capable of dominating the common technologies of wastewater treatment and providing a cost-effective, reliable, and simple method for wastewater treatment without the need to add chemicals [9]. In addition, a lot of research has been carried out to treat dye wastewater using an EC process. The findings demonstrated that dye, COD, turbidity, and soluble solids could be removed considerably under different operating conditions [10-13]. The theory of the EC process is that this process uses direct current with an operating voltage and variable current intensity, which is determined depending on the degree of contamination of wastewater or water, and uses electrodes made of aluminum, iron, and stainless steel. The reactions exerted at the anode and cathode are given below:

$$\text{Anode: } M \rightarrow e^{n+} + ne^- \quad (1)$$
$$\text{Cathode: } 2H_2O + 2e^- \rightarrow 2OH^- + H_2 \quad (2)$$

The physical and chemical passivities in the coagulation chamber are anodic oxidation and cathodic reduction of impurities in the water. Anode metal dissolution and its discharge into the water, resulting in coagulation of colloidal particles, electrophoresis process (in which ions pass through the semiconductor membrane), particle flotation (in which gas bubbles produced at the electrodes cause the flotation of particles), composition of zinc metal ions cathode, production and concentration of acids and bases, other chemical and electrochemical processes [14]. Numerous advantages have been reported for EC, including easy operation and maintenance, fully automatic and continuous operation, low operating cost, wastewater recycling and reuse, low sludge volume, no need for pH control except for additional amounts, no need to add chemicals, efficiency and rapid decomposition of organic matter with a normal efficiency of nearly 90%, reducing the number of treatment plant process units and thus drastically reducing the land surface required by the treatment plant [14-16]. The main disadvantages of EC are hydrogen gas production (which may be a safety risk), the corrosion of the anode and deposition of sludge on electrodes (which is capable of restricting the process), increased amount of aluminum and iron ions at the output, relatively high primary finance costs and exorbitant cost of electricity use in some areas [17-19]. Studies on applying EC in wastewater treatment have progressed during the last decade–the type of electrodes and effective factors that have more attention has been mentioned in recent years. Process parameters (electrolysis time, current density, and aeration time) can significantly affect the separation process in the EC system [20]. RSM is utilized as a statistical technique in experiment design, evaluation of unique and interaction effects of independent variables, and optimization of process parameters provided by a limited number of experimental experiments [21]. Using RSM has proven significantly effective in optimizing and predicting EC processes [22]. Electrolysis time, current density, and aeration time were chosen as three influential factors, and the desired responses were considered COD removal. Therefore, in the present study, the design of the D-optimal experiment is performed to optimize the factors affecting COD removal using the EC method.

2. Materials and Methods

2.1 Description of the experiment

The samples were collected from the car wash effluent. The COD was determined by closed reflux method. All reagents used in this work were of analytical grade and used without any further purification. The initial COD concentration was measured at 348 ppm. The samples were stored at 4°C and used without any dilution. Table 1 shows the values of some quality parameters of car wash effluent.

Table 1: Qualitative characteristics of the car wash effluent sample

Parameter	Value
COD (mg/l)	348
Oil & grease (mg /l)	180
pH	7.5

The present study is a laboratory investigation that was conducted to assay the effect of continuous electricity on detergent contamination in car wash wastewater samples. Further, it examined the effect of electrolysis time, current density, and aeration time on removal efficiency. Effluent samples were collected and transferred to the laboratory to determine COD. The effects of electrolysis time, current density, and aeration time on the EC process are given in Table 1. In order to experiment, a reactor with dimensions of 40 Cm * 50 Cm * 50 Cm made of Plexiglas with a volume of 90 L equipped with an electric current generator and an electrode was utilized (Figure 1). Four aluminum electrodes, each measuring 45 cm * 0.2 cm * 45 cm, were installed at a distance of 2 cm. The electrodes were connected to the DC current generator in the form of Al-Al; two electrodes acted as cathodes and the two electrodes acted as anodes. The dimensions of the aeration chamber were 40 Cm * 40 Cm * 55 Cm, and an air pump was applied for aeration. Ten aeration stones, with a length of 40 cm, were used. The energy-generating device can generate electricity in the amounts used in currents of 30-60 V. Within each experiment, the electrodes were linked to the positive or negative output, and the current density was regulated by a direct current (DC), which was taken from the power supply. Hydrochloric acid weighing 15% and distilled water was used to clean the electrodes. The pond's dimensions were 55 Cm * 75 Cm * 65 Cm. Obstacles were used to allow more water to come into contact with the electrodes. Water entered the reactor and the amount of COD in the treated water was determined with the installed valves. Since the produced sludge was very small compared to other chemical methods, measuring the amount of COD in the effluent and comparing it with standard conditions, the desired water could be utilized for agricultural and industrial purposes. In the present research, factors such as dissolved oxygen and temperature effectively improved COD removal efficiency. The dissolved oxygen concentration was between 2.5-3 mg/l, which was measured using a DO meter after calibrating the device. The minimum amount of dissolved oxygen was 1-1.5 mg/L; in practice, all parts of the aeration tank should be maintained at 1.5 to 4 mg/L. Values more than 4 mg/L did not improve the system's efficiency; however, it caused a sharp increase in the cost of aeration [23]. According to the mentioned points, the range of dissolved oxygen was appropriate.

2.2 Analytical methods

COD removal efficiencies are calculated using the following equation:

$$\% \text{ Removal} = \frac{C_0 - C_i}{C_i} \times 100 \qquad (3)$$

Where C_0 and C_i represent the initial material content and the amount of COD of the EC process, respectively.

2.3 Experimental design

According to the existing scientific research, several studies were conducted on the EC process and COD removal using RSM. Therefore, this study aimed to investigate the effect of process agents on the COD removal efficiency obtained using the EC system with aluminum electrodes. Operational parameters and electrolysis time, current density, and aeration time can significantly impact the efficiency of the EC process. Therefore, in this study, the effect of these usable agents on COD removal was assessed and optimized by RSM. RSM is a statistical technique used to model and analyze the problems in which the response variable is affected by multiple input variables. Its purpose is to optimize this response [24]. Using the estimated parameter, we can determine the variable that produces the most predicted value; thus, it enables the researcher to recognize the variables that have the greatest effect on obtaining the desired answer [25]. The major benefit of RSM is that it is incompatible with examining one factor at a time, examines experiments with fewer numbers, and creates interactions between variables [26]. In separating various pollutants from various chemical [27], physical [28], and biological [29] processes, RSM has been used for optimizing and modeling. The D-optimal design is a suitable option for creating an optimal test design. In this type of test design, the variance related to estimating the coefficients of the model is minimized [30]. In this study, the experimental design was applied by D-optimal with three quantitative variables at three levels to assess the independent and interaction effects of process variables. Operational parameters such as electrolysis time, current density, and aeration time can significantly affect EC performance. Therefore these parameters were investigated and optimized to remove the mentioned materials by the RSM in this study. This plan consisted of 20 tests (each run is the average of 3 tests) by the electrocoagulation process. The designer of the D-optimal test in the RSM with three factors of numerical at the three levels to evaluate the interactive impact of variables of the process, such as electrolysis time (X_1), current density (X_2), and aeration time (X_3) to appraise the efficiency of COD removal was investigated using the EC method (Table 2).

Table 2: Values of actual and coded experiment design in EC process

Variable (unit)	Numerical variable Level		
	-1	0	1
X_1, Electrolysis time (min)	10	20	30
X_2, Current density (A/m^2)	8.75	13.75	18.75
X_3, Aeration time (min)	10	20	30

To describe the procedure, we used the least squares regression method to calculate the mathematical model's parametric estimation [20]. The selected independent variables in the experiments were performed using the following equation:

$$i = 1,2,\ldots, k \; X_i = \frac{X_i - X_0}{\Delta X_i} \qquad (4)$$

x_i actual value; x_0 is the actual values at the center point for an independent variable. Δx_i is the difference between the variable i from the actual values. X_i is considered a partial value of an independent variable; therefore, the information obtained from the above relation is utilized to attain a model's prediction. Equations 5 and 6 express linear and quadratic equations for predicting optimal conditions.

$$Y= \beta_0 + \sum_{i=1}^{k} \beta_i X_i \quad (5)$$

$$Y= \beta_0 + \sum_{j=1}^{k} \beta_j X_j + \sum_{j=1}^{k} \beta_{jj} X_j^2 + \sum\sum_{i<j=2}^{k} \beta_{ij} X_i X_j + e \quad (6)$$

Y is the response, Xi and Xj are variables (i and j are variables from 1 to k), β_0 is a fixed expression, β_i is a linear coefficient, β_{ij} is an interaction factor, β_{jj} is a quadratic factor and k is the number of independent parameters.

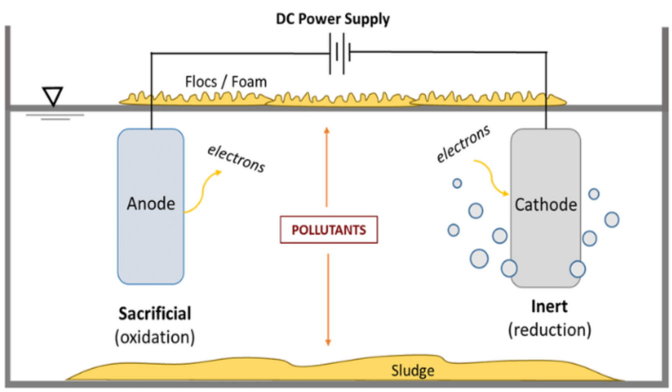

Figure 1: schematic of electrocoagulation reactor and power supply

3. Results and Discussion

3.1 Evaluating the results of COD removal

Based on the design of the experiment, 20 tests were performed. The details of these are presented in Table 3. COD removal percentage ranged from 6.06% - 48.51%. The regression equation was determined by Design Expert 10 and presented as follows:

COD Removal, Y (%)= 20.16+5.23 * A+6.99 * B+6.19 * C-0.57 * AB+2.24 * AC+0.62 * BC-0.53 * A^2-1.18 * B^2+3.92 * C^2 (8)

According to the models of valid regression from variation models, such as linear, reciprocal, quadratic, and cubic models, the data was created from the D-optimal design by two different criteria, including the set of squares of the model and the statistical summary of the model. The results illustrated that the linear model presents R^2, adjusted R^2, predicted R^2, higher F-value, and lower P-value compared to other models (quadratic and reciprocal). Therefore, a linear model was selected to expose the effect of process variables on the EC process in COD removal. The statistical summary of the model executed that the linear model was utilized for the maximum "adjusted R^2" and the predicted values of R^2.

Table3: Design of process experiment using D-optimal design with the response

Run no.	X_1: (min)	X_2: (A/m^2)	X_3: (min)	COD Removal (%)
1	28.1	17.75	20	26.18
2	10	8.75	17.1	6.06
3	30	12.75	21.9	30.3
4	18.1	18.75	21.9	29.24
5	10	18.75	10	21.21
6	30	18.75	10	31.15
7	18.1	12.75	10	20.16
8	10	15.25	30	27.05
9	30	8.75	10	18.01
10	10	14.9	17.6	25.95
11	10	8.75	17.1	9.09
12	28	13.77	12	19.21
13	23	8.75	30	27.25
14	18.1	12.75	10	19.87
15	23.4	8.75	18.6	18.15
16	30	12.75	21.9	28.26
17	30	18.75	30	48.51
18	16.3	11.9	24.2	24.31
19	16.3	11.9	24.2	24.29
20	18.1	18.75	21.9	30.61

3.1.1 variance Analysis (ANOVA)

To present a reliable regression model, ANOVA checked the diversity between the values of the test data and their scatter (F-test). The F-value values for the COD removal efficiency were 10.55, with an acceptable change in their mean values. The p-value was applied to define the statistical association between the response and each variable in the model. Interactions are significant when p<0.05 [31,32]. Under these situations, the step-by-step removal procedure could eliminate variations, including p-values>0.05. The results showed the linear model for COD removal efficiency according to ANOVA of regression of dependent parameters, according to the prediction of surface response. Moreover, the results demonstrated that F-test with a low p-value (p-value=<0.0001) indicated the significance of regression models. As mentioned earlier, designing models is remarkable when the values of prob>F are less than 0.05 [33]. In this study, the p-value was<0.05, indicating that the regression model obtained the ratio of the total variance of the dependent variablesl. Further, the nominal (R2) coefficients illustrated that a total change of 90.47% can be obtained using the mentioned models for COD removal efficiency. Values coefficient of Equivalent nominal (R^2 equal 0.8189) also showed the importance of the COD removal percentage response. The residual values in the assessment normal distribution of data were presented and the model confirmed the ANOVA hypotheses. In contrast, the internal standard residuals with standard deviation were evaluated to separate the actual and predicted value. The percent of natural probability distributions relative to the standard residues for the percentage of COD removal were shown in the normal distribution diagram addresses whether the residues follow the distribution normally; in that state, they establish points in the form of a straight line. Some dispersions even come close to normalized data. Thus, concerning the results, drawn diagrams show that the data are distributed normally.

3.1.2 Comparison of test results and predicted values

The main section of the analytical process analysis is examining the quality of proposed model. The validity of the model indicated the correct approximation that is provided to avoid providing poor or inaccurate analysis. Figure 2 proves the evaluation between the obtained experimental values and the proposed model. This model was presented to conform with the experiential data and the points defined in the proximity of the drawn line. The analyses also displayed that using linear multinomial equations could be favorable for the efficiency of COD removal using the EC process. Also, to ensure the correctness of the obtained equation, four tests with different values of the factors in Table 2 were performed in the laboratory. The coefficient of determination of 0.89 obtained from the laboratory values and the output of the equation showed high accuracy.

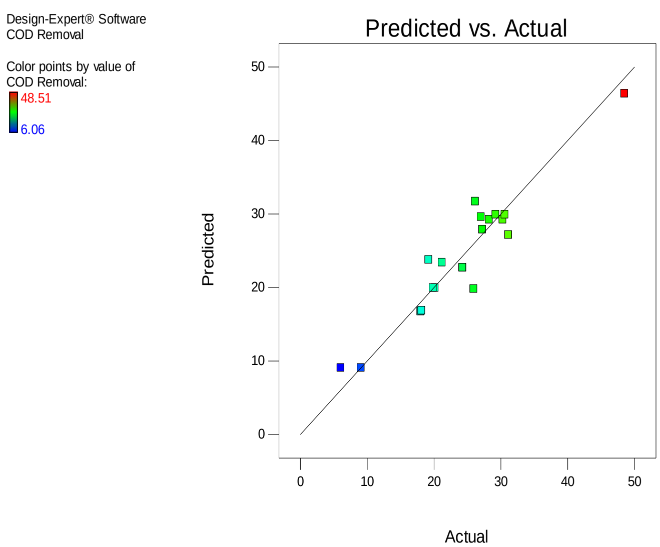

Figure 2: Diagram of comparison of values of predicted and actual for removal of COD

3.1.3 Assessment of influence parameters in the electrocoagulation process

The effect of the three parameters of electrolysis time, current density, and aeration time, which are the main agents for regulating the reaction rate, on the efficiency of COD removal in the EC process was evaluated by the RSM method.

3.1.3.1 Effect of electrolysis time

One of the substantial factors for COD removal in this procedure is the electrolysis time. The tests were performed with a time of electrolysis (X_1) varied from 10-30 min at flow density 11 A/m^2 and aeration time of 20 min. Figure 3 presents that the optimal time takes 30 min. In a similar work, Nawarkar and Salkar (2019) displayed that the generation of cationic metal ion concentration was increased with the time of the electrolysis process involved in removing the contaminant. The increasing rate of reaction time directly affects the contaminant removal [34]. According to Priya and Jeyanthi (2019), the COD removal was increased with time up to 40 min; beyond that, there is no considerable improvement in its performance [3]. The contaminant removal efficiency of the Al-Al electrodes started to fluctuate since the formation of a dissolved form of Al hydroxide flocs that could remove only a few percentages of contaminants [35,36]. The findings also illustrated that the COD removal percentage increased with the enhancement of the electrolysis time because OH-ions at the cathode are regulated by the water electrolysis, which can synthesize with metal ions and produce more clots resulting in increased COD removal efficiency in solution [37].

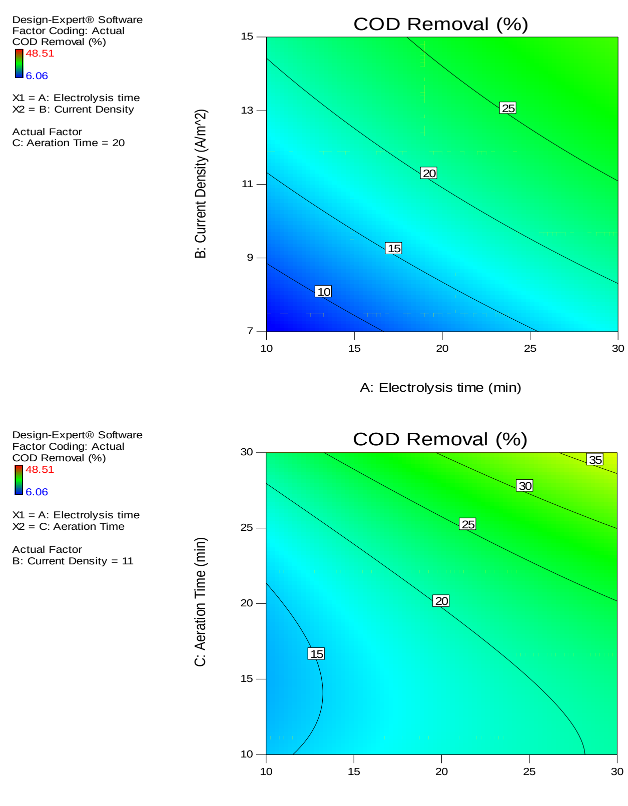

Figure 3: Two-dimensional contour diagrams: (a) Influence of time of electrolysis and current density and (b) Influence of electrolysis time and aeration time on COD removal (%)

3.1.3.2 Effect of current density

In the coagulation of electrochemical processes, one of the major factors that can present a remarkable role in controlling pollutants is the effect of current density. This factor affects the rate of electrochemical reactions coagulation by impacting the number of metal ions removed from the electrode surface. Hence, the optimal current density values of each electrochemical process should be determined [38,39]. Figure 4 is a three-dimensional and two-dimensional contour diagram examining the effect of time of electrolysis and current density on the COD removal percent. The results demonstrated that the COD removal percentage increased by raising the current density. According to Faraday's law, an increase in current density increases the dissolution rate of electrode material [39], which increases the sacrificial anode metal ion concentration and enhances the floc formation. The contaminant removal was improved with increasing current density. These residual flocs were in dis-solved form and reduced the contaminant removal efficiency of the electrode material [35,40]. In addition, the continued increases in current density increase the oxidation reaction rate that supports the phenomena of corrosion and forms the oxide layer on the active surface of the Al anode electrode. Due to the formation of this layer, the passivation effects occurred and reduced EC's removal efficiency [41]. The obtained results are in line with the previous findings. The number of clots increases using anode dissolving, which is highly interdependent on the pollutant and supplies desirable situations for COD removal. However, in many currents, the hydrogen gas creation with more capacity aids in incrementing the flotation of coagulated substances [42].

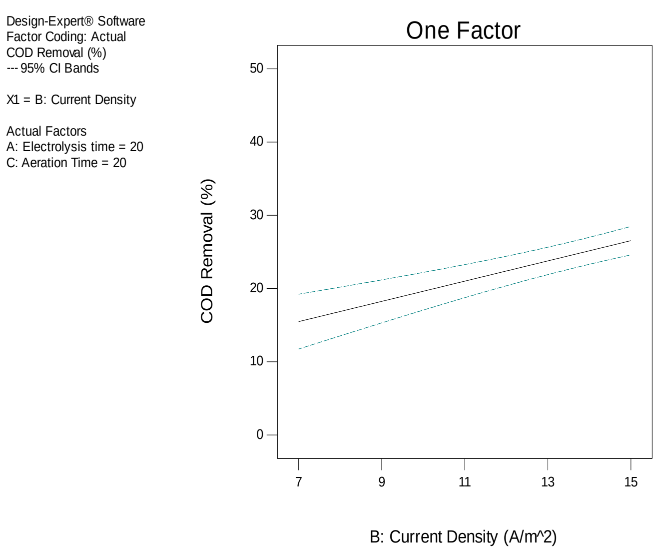

Figure 4: Influence of current density for removal of COD

3.1.3.3 Effect of aeration time

Based on the results, the rate of COD removal reached the highest value during an aeration time of 30 min. Figure 5 shows the results obtained from the effectt of aeration time on the percentage of COD removal by the EC process. The aeration time is changed in the range of 10-30 min. The results of Figure 5 also illustrated that the removal efficiency of COD increases with increasing aeration time, and the maximum removal efficiency of 48.51% at an aeration time of 30 min and current density of 11 A/m², and an electrolysis time of 20 min was acquired.

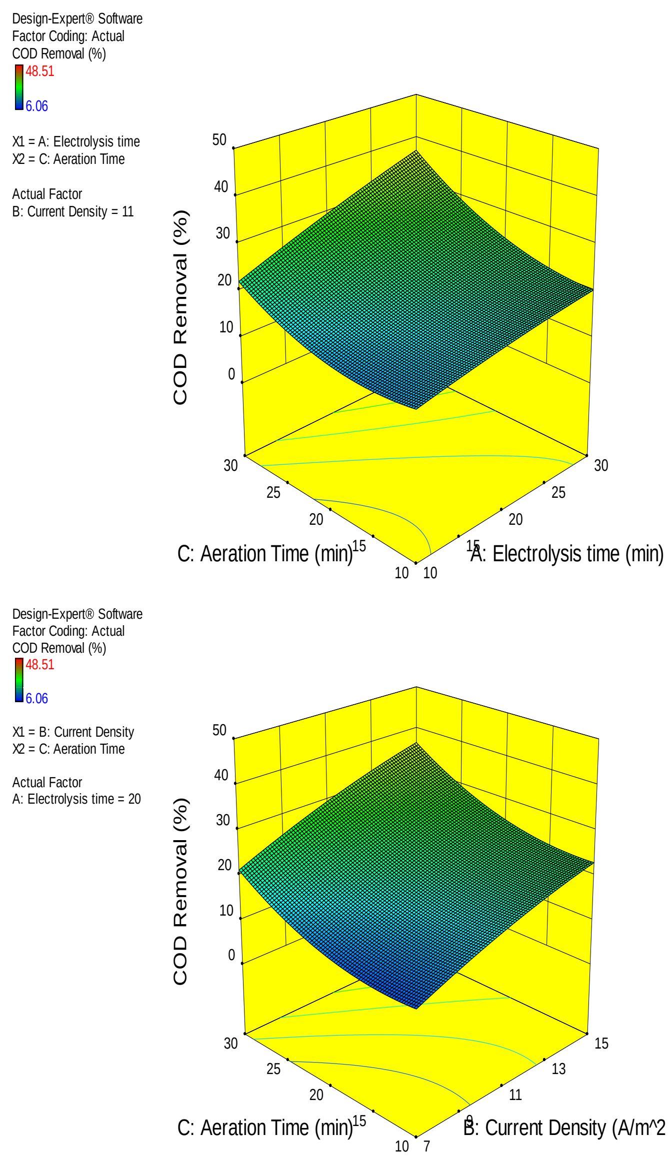

Figure 5: Three-dimensional diagrams of the effect of (a) aeration time and electrolysis time (current density: 11 A / m²) and (b) aeration time and current density (electrolysis time: 20 min) on the percentage of COD removal

4. Conclusion

In this paper, the D-optimal test design is applied to study and optimize process variables such as the time of electrolysis, the density of the current, and aeration time in COD removal from car wash wastewater using the EC method. The F-values for the COD removal efficiency were 10.55, with an acceptable change in their mean values. In removing the COD of this model, the value of R_2 = 0.9047 illustrated that all values are satisfactory, which is sufficient for the model. On the other hand, all values of the -p-obtained<0.05, demonstrating the model's eminent significance. Optimal EC conditions were determined as time of electrolysis (30 min), current density (18.75) A/m^2, and aeration time (30 min), respectively. In this state, the highest COD removal rate was 48.51%. The results showed that the amount of metal ions in the solution increases with increasing current density. The percentage of COD removal efficiency increases when the current density increases from 8.75 A/m^2 to 18.275 A/m^2. The results demonstrated that EC is a reliable method to remove car wash. This investigation creates the basis for further research on other variables and a comprehensive study that is necessary to investigate the application of the electrochemical procedure on a larger scale and in other industries.

Authors' Contributions

Seyed Morteza Moosavirad: Investigation; Carried Out the Experiment; Writing paper; SoftWare; Paper Review; Methodology. Ali Hasanzade-Sabluei: Validation; Formal Analysis; Resources; Administration.

Conflicts of Interest

The Authors declare that there is no conflict of interest.

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