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Optimization of electrocoagulation of pistachio processing wastewaters using the response surface methodology

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ABSTRACT

Optimization of electrocoagulation using aluminum electrode in terms of chemical oxygen demand (COD) removal from pistachio processing wastewaters was carried out by using response surface methodology (RSM) and Box–Behnken experimental design. Initial pH, current density, and electrolysis time were selected as independent process variables. The experimental data and model predictions agreed well. Optimization result for the maximum COD removal efficiency was 57.4% at 317 A/m^2 current density, pH 6, and 29 min application time for treatment of pistachio processing wastewaters. The operating cost of the model at the optimized conditions was 2.89 €/m^3.

Keywords: Electrocoagulation; Response surface methodology (RSM); Chemical oxygen demand (COD); Pistachio processing wastewaters; Operating cost

1. Introduction

Pistachio is one of the many popular tree nuts around the world. Turkey is the world's largest producer of pistachios; contributing about 12.00% of world production according to the statistics of Food and Agricultural Organization of United Nations in 2011 [1]. Pistachio is grown around the world in several warm arid climate countries like Iran, USA, Turkey, Syria, and China, as major producers [2]. A large amount of wastewater, 1,000–2000 liters per ton of pistachio, is generated during the manufacturing and processing of pistachio nut. The pistachio processing wastewaters are characterized by extremely high chemical oxygen demand (COD) and total organic carbon concentrations varying between 18–20 and 4–5 g/L, respectively, apart from low pH 4–5, strong odor, and a large amount of dark brown color [3].

Pistachio processing wastewater disposal has become increasingly important due to more stringent legislative requirements for effluent quality. Appropriate treatment of these types of wastewaters is necessary in order to reduce the impact of their discharge related to environmental protection. As such, biological treatment of the pistachio processing wastewater is expected to be very difficult without chemical pretreatment. One of the chemical methods is electrochemically assisted coagulation, electrocoagulation (EC), that can compete with the conventional chemical coagulation process in the treatment of wastewaters [4].

Recently, electrochemical methods, such as EC, electro flotation, and electro oxidation, have received great attention for the effective treatment of organic pollutants [5]. The EC process has been widely and successfully practiced to treat leachate from solid wastes [6,7], textile wastewater [8–10], olive mill wastewater [11,12], dairy wastewater [13,14], poultry slaughterhouse wastewater [15], distillery and fermentation wastewater [16], potato chips manufacturing wastewater [17], yeast industry wastewater [18], tannery wastewater [19], metal plating wastewater [20], paper mill wastewater [21], petroleum refinery wastewater [22], biodiesel wastewater [23], mixed industrial wastewater [24], restaurant wastewater [25], municipal wastewater [26,27], and also drinking water [28,29]. However, there are no studies carried out for the treatment of pistachio processing wastewater by EC methods.

EC process compared to conventional chemical coagulation is very attractive from an economical point of view due to simple and easy installment of equipment, negligible start up time, short reaction time, a reduction or absence of equipment for adding chemicals, reduction of wastewater acidification and salinity, compact treatment facility, and minimal amount of hazardous sludge [30]. EC process efficiency is affected by numerous factors such as pH, applied electric current, the electrolyte concentration, and the electrolysis time [5,18]. The optimization of these factors may significantly increase the process efficiency.

Among the possible approaches for optimization of conventional (one-factor-at-a-time technique) and design of experimental methods, response surface methodology (RSM), factorial design, and mixture design can be stated [31]. In conventional methods of studying a process, optimization is usually carried out by varying single factor while keeping all other factors fixed at a specific set of conditions, which is incapable of reaching true optimum condition due to ignoring of the interactions among variables. This method is also time consuming and ineffective for adequate process optimization [32]. To overcome this difficulty, statistical experimental design technique using the RSM is one of the most popular optimization methods used in recent years.

RSM is widely accepted as a statistical-based method for designing experiments, evaluating the individual and interaction effects of independent variables, and optimizing the process parameters with limited number of experimental run [33]. The application of RSM has demonstrated that this modeling can effectively optimize and predict the EC processes [34–37]. As far as we are concerned, no scientific work has been published dealing with the application of RSM to treat pistachio processing wastewater.

In the present work, RSM is implemented to optimize influencing factors on treatment efficiency of pistachio processing wastewater by using Box–Behnken experimental design.

2. Experimental

2.1. Pistachio processing wastewaters

The composite wastewater sample used in this study was collected from the outlet of an existing physical treatment plant of a pistachio processing factory in Gaziantep, Turkey. The wastewater sample was preserved in the dark at 4℃ in a refrigerator and used without any dilution. The composition of pistachio processing factory wastewater is given in Table 1.

2.2. Experimental setup and procedure

A laboratory-scale reactor, made of Plexiglas, was used in all experiments. Experimental setup is shown in Fig. 1. Two aluminum electrodes were used. Both aluminum cathode and anode were made from plates with the same dimension of 70 × 50 × 3 mm. The total submerged surface area of electrodes (A) of these was $7 \times 10^{-3}\, m^2$ and distance between two electrodes in EC cell was 20 mm in all experiments. The electrodes were connected to DC power supply (GoodWill, Malaysia), with galvanostatic operational options for controlling the current density.

Table 1
Compositions of pistachio processing factory wastewater

Parameter	Value
pH	5.4
COD (mg/L)	23,250
Turbidity (NTU)	2,064
Conductivity (μS/cm)	4,000
Chloride (mg/L)	250
Phenol (mg/L)	2,100
Sulphate (mg/L)	52

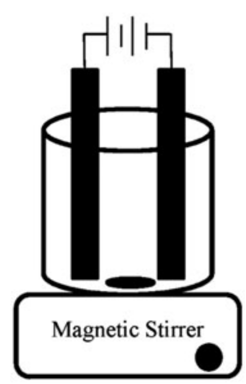

Fig. 1. Schematic diagram of experimental setup.

In each run, 750 mL of the wastewater was placed into the EC reactor. The current density was adjusted to desired value and the EC was started. At the end of the run, the supernatant was centrifuged and then analyzed.

After each experimental run, the electrodes were treated as follows; the EC reactor and electrodes were carefully rinsed twice with 50% (v/v) nitric acid solution for 2–3 min and several times with deionized water to remove impurities and metal hydroxide precipitates from the electrodes and reactor [38]. The electrodes were replaced each time whenever more than 10% of electrode material was lost. Experiments were conducted at room temperature. When the current was passed through wastewater to be treated, there was an increase in temperature but this was negligible due to the relatively low electrolysis time and current density.

2.3. Chemical analysis and operating cost

pH Value, COD, turbidity, conductivity, chloride, phenol, and sulfate analysis were conducted according to the procedure of Standard Methods by using Electrometric method (4500-H^+ B), Closed Reflux Titrimetric Method (5220 C), Nephelometric Method (2130 B), Laboratory Method (2510 B), Argentometric Method (4500-Cl^- B), Direct Photometric Method (5530 D), and Turbidimetric Method (4500-SO_4^{2-} E), respectively [39]. Merck analytical quality chemicals were used in experiments. All studies and tests were performed at room temperature. The pH of the solution was adjusted with 1 M NaOH or HCl. Turbidity, conductivity, and pH of the samples were measured by using Hach 2100P turbidimeter and a WTW 340i multiparameter, respectively.

The operational cost for EC process can be analyzed by taking the price of electricity, chemical reagents, sludge disposal, labors, maintenance, and equipment into account. But, in the present work, cost of electricity and cost of electrode have been considered as the main strands for the determination of the economically feasible optimum condition since the balance issues are negligible when compared with these [40]. Then, operation cost for EC process can be calculated as follows:

$$\text{Operation cost} = a \times C_{\text{energy}} + b \times C_{\text{electrode}} \tag{1}$$

The a and b are given for the Turkish market for July 2013, and defined as follows: a is the electrical energy price 0.0838 €/kWh and b is the electrode material price 1.776 €/kg Al. The electrical energy consumption was calculated in Equation 2; whereas, the electrode consumption (kg Al/m^3) was calculated using equation (3) and Faraday's Law [41].

The electrical energy consumption was calculated by using the following equation:

$$C_{\text{energy}} = \frac{I \times V \times t}{v} \tag{2}$$

where W is specific energy consumption, I is current density (A), V is applied potential difference (V), t is reaction time (h), and v is solution volume (m^3).

According to the Faraday's law, as current density increases, the rate of Al^{+3} ions passing from soluble aluminum electrodes to the solution increases. In other words, an increase in current density also increases the flocking ion rate and therefore causes more flocks to form in the reactor [42].

$$\Delta M = \frac{M \times I \times t}{n \times F} \tag{3}$$

where ΔM represents the solved aluminum rate (g), M is the molecular weight of aluminum (26.892 g/mol), n is the number of electrons, F is the Faraday constant (F = 96,487 C/mol), I is the current (A), and t is the reaction time.

2.4. Experimental design and data analysis

The design expert 8.0.7.1 software is used for the statistical design of experiments and data analysis and performed in duplicate. The effects of three important operating variables such as initial wastewater pH (X_1), current density (X_2), and time of electrolysis (X_3) on the COD removal efficiency were investigated. These selected operating variables are commonly used for the prediction of response and understanding treatment mechanisms of EC in many studies [8,10,20].

Table 2
Experimental range and levels of the independent variables used in RSM

Independent process variables code	Real values of coded levels		
	−1	0	1
Initial pH	3	6	9
Current density (A/m^2)	143	357	571
Application time	15	30	45

Table 3
The actual design of experiments and responses for EC

Exp. No.	pH (x_1)	CD (x_2) (A/m^2)	t (x_3) (min)	COD removal (%)	Energy consumption kWh/m^3	Energy consumption kWh/kg COD	Electrode consumption kgAl/m^3	Electrode consumption kg Al/kg COD	Operation cost €/m^3	Operation cost €/kg COD
1	9	357	45	49.6	54.13	3.52	0,918	0.060	5.41	0.469
2	6	571	15	47.7	35.13	2.38	0.617	0.042	3.55	0.320
3	3	357	45	46.8	51.63	3.56	1.144	0.079	5.60	0.515
4	6	357	30	59.1	33.92	1.85	0.785	0.043	3.74	0.272
5	3	357	15	34.8	15.71	1.46	0.374	0.035	1.75	0.216
6	9	143	30	39.2	8.13	0.67	0.363	0.030	1.18	0.130
7	6	143	45	44.8	12.30	0.89	0.362	0.026	1.48	0.142
8	3	143	30	31.4	8.47	0.87	0.227	0.023	0.98	0.135
9	6	357	30	58.9	33.83	1.85	0.655	0.036	3.52	0.257
10	6	571	45	60.6	107.40	5.72	1.748	0.093	10.62	0.754
11	6	357	30	59.1	32.92	1.80	0.585	0.032	3.33	0.243
12	9	571	30	50.3	71.20	4.57	0.927	0.059	6.66	0.569
13	6	357	30	58.7	31.25	1.72	0.622	0.034	3.28	0.240
14	6	357	30	58.9	33.83	1.85	0.682	0.037	3.56	0.260
15	3	571	30	47.3	67.73	4.62	0.911	0.062	6.38	0.580
16	9	357	15	41.7	17.38	1.34	0.390	0,030	1.89	0.195
17	6	143	15	36.1	4.38	0.39	0.147	0.013	0.56	0.067

In the present work, the Box–Behnken experimental design with three replicates of the center point was used to find the relationship between the response functions and variables (Table 2). A total of 17 experiments were conducted. A significant advantage of Box–Behnken statistical design is that it is a more cost-effective technique compared with other techniques, such as central composite design, three-level factorial design, and D-optimal design, which require fewer experimental runs and less time for optimization of a process. [43]. After preliminary studies, process variable ranges were determined. The study ranges were chosen as initial wastewater pH 3–9, current density 143–571 A/m^2, and operating time 15–45 min for the EC process using Al electrodes. The actual design of experiments and responses for EC is given in Table 3.

RSM postulates the functional relationship between the controllable input parameters and the obtained response surfaces [44]. If all variables are assumed to be measurable, the response surface can be expressed as below:

$$y = f(x_1, x_2, x_3 \ldots, x_k) + \varepsilon \tag{4}$$

where y is the response of the system and x_i is individual variables. For evaluation of experimental data, the response variable was fitted by a second-order model in the form of quadratic polynomial equation.

$$y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \beta_{ii} X_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} X_i X_j + \varepsilon \tag{5}$$

where $X_1, X_2, \ldots, X_k$ are the input factors which influence the response y; β_0, β_{ii} $(i = 1, 2, \ldots, k)$, β_{ij} $(i = 1, 2, \ldots, k; j = 1, 2, \ldots, k)$ are unknown parameters and ε is a random error. The β coefficients should be determined in the second-order model. The analysis was focused on verifying the influence of individual parameters on percentage of COD removal.

The quality of the fit quadratic polynomial model was expressed by determination of R^2 coefficient, adjusted R^2, and coefficient of variation (CV) and its statistical significance was checked by the Fisher's F-test. Three-dimensional plots with the respective contour plots were obtained from the results of the experiments.

3. Results and discussion

3.1. Statistical analysis

Box–Behnken experimental design was employed to determine the simple and combined effect of pH, current density, and electrolysis time on COD removal efficiency for the EC process in which aluminum electrodes were used. Optimization process involves three steps; performing the statistically designed experiments, estimating the coefficients in the proposed model and predicting the response of process, and checking the validity of the model. In the optimization step of the design, Expert 8.0.7.1 software program, desired goal for each variable and response, should be chosen.

Experimental data were analyzed by using the response surface regression procedure. Significant model terms are desired to obtain a good fit of model. For evaluation of experimental data, the response variable was fitted by second-order model in the form of quadratic polynomial model. Application of RSM offers, in terms of actual factors, an empirical relationship between COD removal efficiency (Y) and the variables (X_1, X_2, X_3) has been expressed by the following second-order polynomial equation:

$$\begin{aligned} Y(\%) = {} & -53.19629 + 16.18146X_1 + 0.13308X_2 \\ & + 1.76106X_3 - 1.86916 \times 10^{-3}X_1X_2 \\ & - 0.022778X_1X_3 + 3.27103 \times 10^{-4}X_2X_3 \\ & - 1.16472X_1^2 - 1.39914 \times 10^{-4}X_2^2 - 0.023256X_3^2 \end{aligned} \tag{6}$$

Table 4 shows the variance analysis (ANOVA) of regression parameters belonging to the predicted response surface quadratic model for COD removal.

As it can be seen from Table 4, the fisher's F-test with a very low probability value (p-value = < 0.0001) demonstrates a very high significance for the regression model when COD removal is considered. Values of $p > F$ are less than 0.05 which show that the model terms are significant. The p-values were much lower than 0.05. The other criterion used for evaluating the model was lack of fit test which is performed for comparing the residual with pure error achieved from the replicated design points at the central level of variables. In this case, if the F-value of lack of fit is lower than tabulated value or the related p-value is greater than 0.05 (at the significance level of 95%), the regression will adequately be significant [45]. The ANOVA results indicated that the p-value of lack of fit test was 0.0532, which confirms the insignificant lack of fit. R^2, the coefficient of determination, which ranges between 0 and 1, indicates the goodness of fit of a regression model. It shows the proportion of the total variance of the dependent variable explained by the regression model. High R^2 indicates that the quadratic model explains all of the variations of the dependent variable. When R^2 is 0, it indicates that the quadratic model explains none of the dependent variable's variance. In many applications, a higher R^2 value is preferred to a lower one. In this study, value of R^2 was 0.9996. The value of the adjusted determination coefficient (Adj. $R^2 = 0.999$) was also very high, which indicates a high significance of the quadratic model [46]. At the same time, a relatively lower value of the coefficient of variation (CV (%) = 0.63) indicates improved precision and reliability of the conducted experiments. The adequate precision value is a measure of the "signal to noise ratio" for the responses. A ratio which is greater than 4 was considered as adequate model discrimination [47]. The adequate precision for the COD removal response was 123.967, which is well above 4. The predicted model can be used to navigate the design space defined by Box–Behnken.

The actual and predicted COD removal plot is shown in Fig. 2. Actual values are the measured response data for a particular run, and predicted

Table 4
ANOVA results for the response surface model for COD removal efficiency

Source	Sum of squares	d*f*	Mean square	*F* value	*p*-value
Model	1480.53	9	164.50	1786.69	*<0.0001*
A-pH	*52.53*	1	*52.53*	*570.55*	*<0.0001*
B-CD	*369.92*	1	*369.92*	*4017.75*	*<0.0001*
C-time	*215.28*	1	*215.28*	*2338.20*	*<0.0001*
AB	*5.76*	1	*5.76*	*62.56*	*<0.0001*
AC	*4.20*	1	*4.20*	*45.64*	*0.0003*
BC	*4.41*	1	*4.41*	*47.90*	*0.0002*
A2	*462.66*	1	*462.66*	*5025.06*	*<0.0001*
B2	*172.87*	1	*172.87*	*1877.54*	*<0.0001*
C2	*115.28*	1	*115.28*	*1252.07*	*<0.0001*
Residual	*0.64*	7	0.092		
Lack of fit	*0.53*	*3*	*0.18*	*6.34*	*0.0532*
Pure error	*0.11*	*4*	*0.028*		
Cor total	1481.18	16			

values are evaluated from the model and generated by using the approximating functions.

According to Fig. 2, the observed points on plot reveal that the actual values are distributed relatively near to the straight line. Therefore, prediction of experimental data is quite satisfactory.

In addition, plots of normal probability and residual vs. predicted value for COD removal efficiency are illustrated in Fig. 3. As seen in Fig. 3(a), the normality assumption was relatively satisfied as the points in the plot form fairly straight line. The reliability of the model was also examined with the plot of residuals vs. fits in Fig. 3(b). As a result, Fig. 3 shows that the model is adequate to characterize COD removal efficiency by RSM. Residuals are considered as unexplained variations by model and they will occur based on a normal distribution, if the model is a good predictor [48]. The perturbation plot compared the effect of all the factors at a particular point in the design space as shown in Fig. 4. A perturbation plot at the center point (pH:6, current density 357 A/m^2, and electrolysis time 30 min) was obtained to show the relative effect of three chosen variables as "one factor at a time" on COD removal. The perturbation plot indicated that pH (A) has the most influential effect (steepest slope) on COD removal followed by electrolysis time (*C*); whereas, current density (*B*) has the least effect on COD removal when compared to each other comparing each of them.

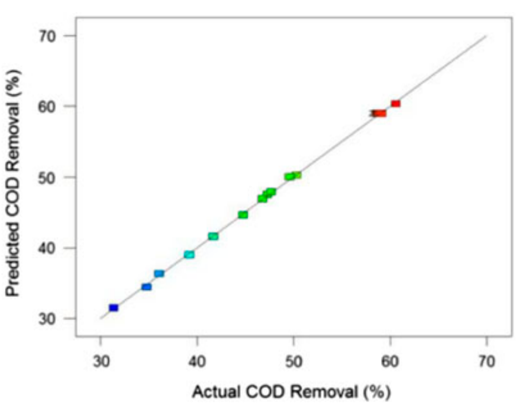

Fig. 2. The actual and predicted plot for COD removal.

The surface response and contour plots of the quadratic model belonging to COD removal with one varible kept at central level and the other two varying within the experimental ranges are shown in Figs. 5–7.

The 3D surface graph, initial pH vs. applied current density in Fig. 5, shows that a significant mutual interaction occurs between applied current density and inital pH for COD removal as a response. It is clear from the figure that the COD removal reduces at low initial pH and applied current density. pH of electrolyte plays an important role in the pollutant removal. There are two primary theories regarding the exact mechanism by which coagulants actually cause the removal of colloids in wastewater. One theory involves neutralization of the surface charge on the particle and the other mechanism is often referred to as the "sweep floc" theory. At pH higher than 6, formed amorphous $Al(OH)_3(s)$ (sweep flocs) flocs with the minimum solubility within the pH range 6.5–7.8 had a large specific surface area that can absorb some soluble organic compounds [18]. The maximum COD removal efficiency is achieved in relatively neutral medium.

The current density determines the coagulant dosage rate. By increasing current density, anodic dissolution of aluminum electrode was accelerated; this resulted in a greater amount of coagulant for the removal of pollutants. Bubble generation rate increases and the bubble size decreases with the increasing current density. In addition, increasing electrolysis time results in an increasing amount of aluminum hydroxide flocs. Both of them had positive effects on COD removal efficiency for treating pistachio processing wastewaters.

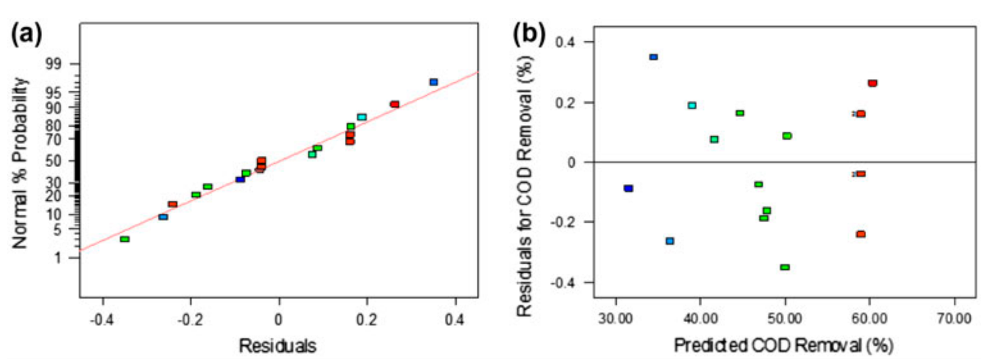

Fig. 3. (a) Normal (%) probability plot and (b) residual vs. predicted plot for COD removal efficiency (%).

In Figs. 6 and 7, it can be observed that the COD removal efficiency increases at the center of the region, which involves the interaction between applied current density with initial pH and electrolysis time with initial pH. When electrolysis time and current density are increased, COD removal efficiency is increased as shown in Fig. 7.

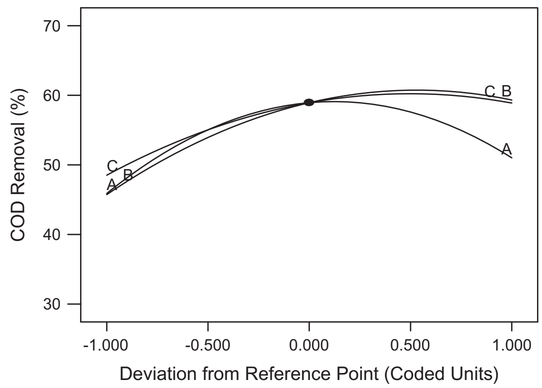

Fig. 4. Perturbation plot, A (pH), B (current density), and C (electrolysis time).

3.2. Process optimization

In this study, the main purpose of the process optimization is to confirm the optimal values of independent variables for treatment with EC process from the models obtained by experimental data. Optimization process was performed to determine the optimum value of COD removal efficiency with minimum operating cost.

Optimization result for the maximum COD removal efficiency was 57.4% at 317 A/m^2 current density, pH 6, and 29 min electrolysis time for treatment of pistachio processing wastewaters. The operating cost of the model at the optimized conditions was 2.89 €/m^3. EC process can satisfy effluent limits for COD removal as a pre-treatment step.

A verification of the results using the set of optimized parameters was accomplished by carrying out the experiments including the optimized variables. The experiments were performed in duplicate. The average COD removal obtained through the experiment was 58%. This experimental finding was in close agreement with the quadratic model prediction.

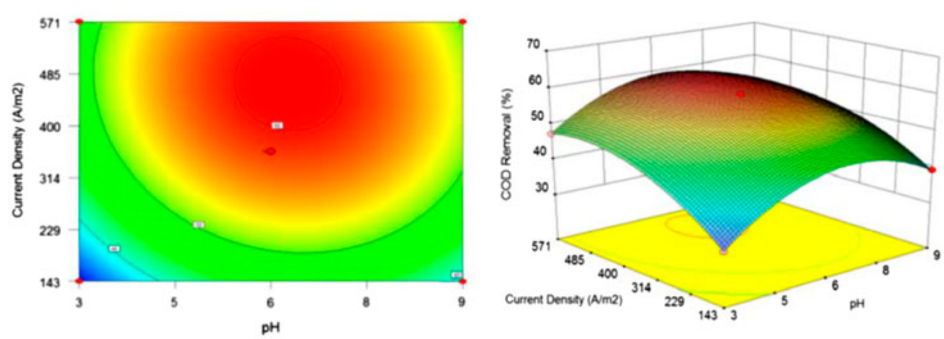

Fig. 5. The effect of applied current density and initial pH on COD removal (electrolysis time: 30 min).

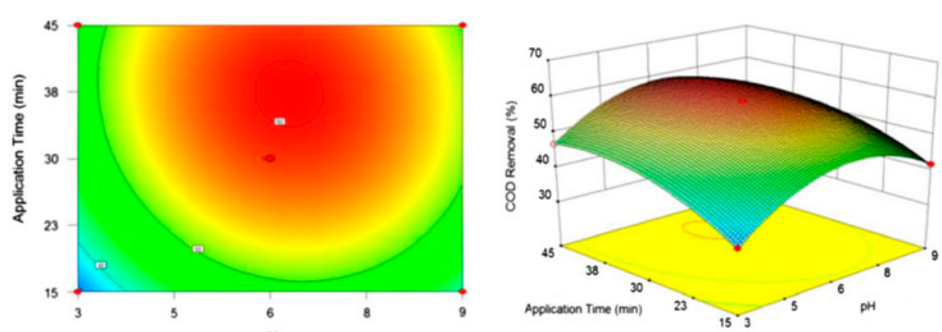

Fig. 6. The effect of electrolysis time and initial pH on COD removal (current density: 357 A/m^2).

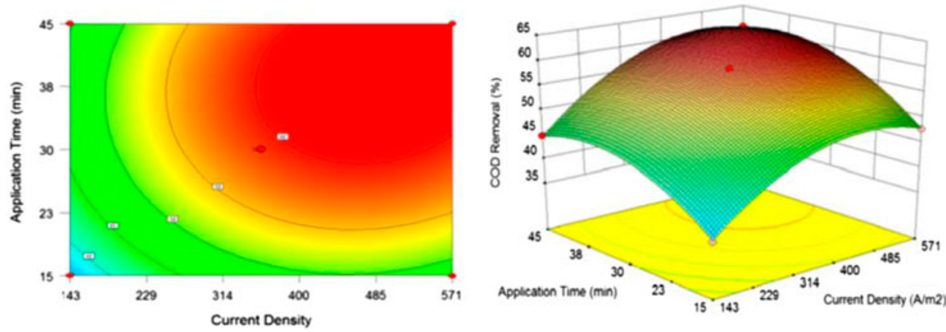

Fig. 7. The effect of electrolysis time and current density on COD removal (initial pH: 6).

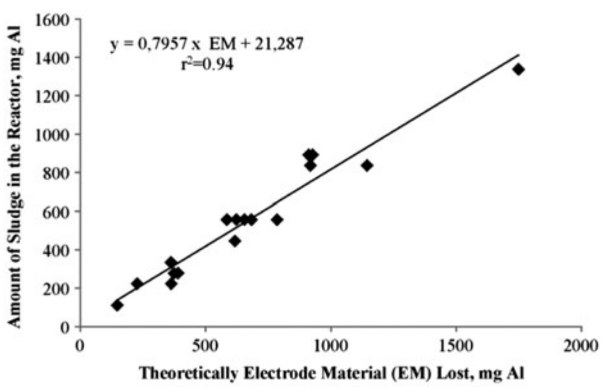

Fig. 8. Relationship between the amount of sludge in the reactor and theoretically electrode material lost.

3.3. *Sludge production*

The loss of electrode material can theoretically be calculated by using Faraday's Law. But, EC is too complex to be described by Faraday's empirical relationship. For this reason, it was preferred to determine the amount of dissolved Al experimentally by weighing the electrodes before and after EC.

The sludge samples were collected from the reactor at the end of the each run. Sludge samples were filtered by a Whatman No. 41 filter paper before the sludge analysis. The dry weight of the sludge was determined after drying in the oven at 1,05 ℃ for 24 h.

Fig. 8 illustrates the relationship between the mass of $Al(OH)_3$ sludge formed and mass of Al electrode lost during EC.

From Fig. 8, theoretical electrode material lost and amount of sludge in the reactor have good correlation. And it was also figured out from the results that theoretical amount of sludge production was a bit less than the measured one. Similar findings were reported by Arslan-Alaton [49].

4. Conclusions

All results obtained indicate that an EC process can be used for COD removal from pistachio processing wastewaters. The Box–Behnken design was successfully used to develop a mathematical model for predicting COD removal. The value of $R^2 > 0.99$ for the obtained quadratic model indicates high correlation between observed and predicted values by the mathematical model. Numerical Optimization using RSM led to the optimum operating conditions as 317 A/m^2 current density, pH 6, and 29 min electrolysis time for treatment of pistachio processing wastewater yielding COD removal with 57.4% efficiency. The operating cost of the model at the optimized conditions was 2.89 €/m^3. Removal efficiency can also be improved by changing to oxidizing electrodes or coupling with some other treatment process like ozonation or Fenton reaction.

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