Optimization of process variables for treatment of food industry effluents by electrocoagulation

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Abstract

The present study investigates the process optimization for treatment of potato processing industry wastewater by electrocoagulation. Performance of treatability was evaluated by achieved chemical oxygen demand (COD) removal efficiencies. Response Surface Methodology was applied to determine individual and interactive impacts of the selected operation variables (pH, conductivity, current density and duration period) on COD removal. Optimization studies were also performed for energy consumption costs which still remain as the major drawback of the electrocoagulation applications. Interactions between dependent and independent process variables were interpreted by the relevant surface plots. Sufficient correlation coefficients were obtained for the predictive regression models of the optimization. Results of the ANOVA tests showed that both of the models are statistically significant. Impacts of the process parameters on response values have been evaluated numerically by Pareto analysis. Furthermore, validation studies were conducted to confirm the accuracy of the model developed for prediction of COD removal efficiencies.

Keywords: Central composite design, COD removal, electrocoagulation, energy consumption costs, food industry effluents, Pareto analysis.

1. Introduction

Similar to the many other engineering fields, environmental treatment applications involves multifactorial processes. Determination of optimum conditions is essential in order to achieve higher removal efficiencies with lower cost, energy, time, etc. Until recent years, "one at a time" variation of process variables was applied to optimize the treatment processes (Raj and Quen, 2005). This conventional approach proposes examination impacts of one selected parameter by keeping all others constant. So, determination of optimum conditions demands numerous time-consuming tests, especially in case of large numbers of variable. In addition, current techniques are incapable to analyze the interactive effects of multiple variables on targeted responses (Rathinam *et al.,* 2011; Pakvaran *et al.,* 2015).

In order to overcome the mentioned deficiency and shortcomings of conventional optimization technique, factorial design methodologies have been developed. On contrary to "one at a time" approach, these methods provide knowledge about the impacts of several factors at two or more levels (Wang and Wan, 2009). In this way, both individual and interactive impacts of the variables can be analyzed (Dashamiri, 2016). As actual conditions are better represented by taking interactive effects into account, the error in the experimental tests can be minimized.

The factorial design methods are categorized into two major groups as the full factorial and the fractional factorial design. The full factorial design investigates each and every combination of input parameters. Even though this methodology enables the examination of all possible combinations, the number of the required experiments rises by the increase in factor and level numbers. On the other hand, the fractional factorial method involves experiments that make them more beneficial, especially in the case of large number of runs. Taguchi-central-composite and Box–Behnken design are some of the well-known fractional factorial design procedures. Taguchi approach is based on an experimental data transformation method, which is the measure of the present response variations (Elizalde-González and García-Díaz, 2010). In this method, the signal-to-noise ratio is used as a quality characteristic and evaluation measurement tool for the impacts of each selected factor on the responses (Dongxia *et al.,* 2012). Despite this approach's simplicity, the difficulty of taking interactions into account poses a potential problem during application (Tsui, 1996). Thus, selection of levels for the studied factors requires significant experience about the topic.

Considering the mentioned drawbacks of Taguchi, other fractional factorial methods such as central composite (CCD) and Box–Behnken design (BBD) become more favorable. Comparing these two methods, BBD provides the advantage of demanding fewer experiments. On the other hand, CCD enables knowledge about the responses

at extreme conditions which cannot be achieved by using BBD (Gengec *et al.*, 2013; Nikzad *et al.*, 2015).

In experimental design studies, The Pareto analysis is a useful tool used to obtain information for interpreting the results. Percentage effect of factors on the response can be calculated individually by this analysis (Abdessalem *et al.*, 2008). Due to this reason, in recent years it has been applied in optimization of waste water treatment processes involving multi-factors (Anupam *et al.*, 2011).

In this work, CCD method was used to optimize the process variables for treatment of food industry effluents. Electrocoagulation process was applied to remove chemical oxygen demand (COD) from the wastewater. In this process, pH, conductivity, current density and duration period were selected as the system variables. Optimization studies were individually performed for COD removal and electricity consumption costs.

2. Materials and methods

2.1. Experimental set-up and procedure

In this study, electrocoagulation process was performed to remove COD since it is a simple and efficient method for treatment of many pollutants. The wastewater used in the experiments was supplied by a treatment plant of a potato chips manufacturing factory. The samples were taken from the outlet of the anaerobic treatment section. Here, we evaluated electrocoagulation as an alternative to conventional aerobic treatment.

The characterization studies were performed to determine the composition of the studied wastewater. The COD and the total organic carbon (TOC) contents of the sample were measured as 500 and 100 mg/L respectively, and the total suspended solid (TSS) parameter was determined as 145 mg/L. The conductivity and pH of the wastewater was also analyzed as important parameters for electrocoagulation process and they were found to be 3.40 mS/cm and 7.8 respectively. The characterization analysis was conducted according to the Standard Methods (APHA, 2012).

Electrocoagulation tests were performed by using a set-up consisted of a batch electrocoagulation tank and a power source (MERSAN MR 12). Electrocoagulation tank has the dimensions of 15x15x15 cm and the sample volume capacity of 2 L. The scheme of the experimental set-up has been presented in Figure 1.
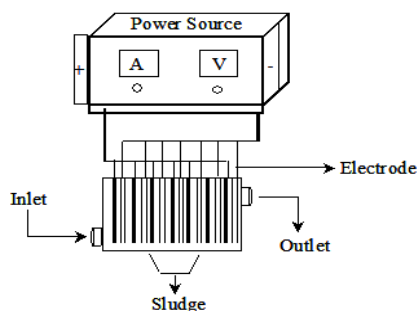

Figure 1. Experimental set-up of electrocoagulation

During experiments Al electrodes with 99.52% purity were used. 14 plates with surface area of 11x16 were placed into the reactor. The electrodes were connected to a direct current power supply.

Before electrocoagulation tests, the electrodes were kept in % HCl for 8 hours. During the experiments, 2 L of the wastewater sample was placed into the electrolytic cell. The pH of the samples were adjusted by using 0.1 N HCl/NaOH whereas conductivity was adjusted by 0.1 N Na_2SO_4. The current density was set to the predetermined value. All the experiments were performed at room temperature.

2.2. Statistical design of the experiments

In this study, central composite design (CDD) was used to evaluate the impacts of the selected independent variables (pH:x_1, conductivity:x_2, current density:x_3 and detention time:x_4) on the response variable (COD removal). With this aim, the experiments were conducted based on four-factor, five-level central composite rotatable design technique. Thus, each independent variable was coded at five levels, from −2 to +2, as shown in Table 1. The determination of the factors used in this study was based on previous experimental studies and literature (Tezcan Un *et al.*, 2009; Veli *et al.*, 2016).

Table 1. Experimental ranges and levels of the studied factors

Parameter	Units	-2	-1	0	+1	+2
pH	-	4	5	6	7	8
Conductivity	mS/cm	3.50	3.75	4.00	4.25	4.50
Current	mA/cm^2	1.01	1.22	1.42	1.62	1.82
Time	min	6	12	18	24	30

Minitab 16 software package was used for experimental design applications. The developed quadratic model with this software provided information about both individual and interactive effects of process variables.

3. Results and discussion

3.1. Results of experimental design procedure

In this work, total 31 experiments were designed using CCD technique. Experimental conditions and obtained response values in each experimental run have been presented in Table 2.

COD removal efficiencies have been calculated with the following simple expression:

$$Removal\,efficiency(\%) = \frac{C_i - C_0}{C_0} \times 100 \qquad (1)$$

where C_i and C_0 denote the initial and final COD concentrations of the samples respectively.

As seen from Table 2, the developed central composite design provided the highest COD removal efficiency of 78% by 23rd run. Coded levels of the pH, conductivity, current density and time were all +1 for this run. Interactions between the independent variables and the response variable have been discussed in Section 3.4 in detail.

Table 2. Design of the experiments by CDD

Run	pH	Conductivity (mS/cm)	Current density (mA/cm^2)	Time (min)	COD removal (%)	Energy cost ($/g COD removal)
1	5	3.75	1.62	12	45	0.96
2	6	4.00	1.42	18	55	0.94
3	7	3.75	1.62	12	59	0.66
4	5	3.75	1.62	24	58	1.44
5	6	4.50	1.42	18	57	0.73
6	6	4.00	1.42	18	56	0.83
7	6	4.00	1.01	18	44	0.50
8	5	4.25	1.62	12	48	0.83
9	5	3.75	1.22	12	27	0.76
10	6	4.00	1.42	18	56	0.83
11	6	4.00	1.42	18	57	0.82
12	7	4.25	1.22	12	45	0.42
13	7	3.75	1.22	24	60	1.03
14	7	3.75	1.22	12	47	0.64
15	7	4.25	1.22	24	68	0.56
16	6	3.50	1.42	18	51	0.98
17	6	4.00	1.82	18	65	1.08
18	5	3.75	1.22	24	28	1.44
19	6	4.00	1.42	18	54	0.86
20	7	3.75	1.62	24	74	1.06
21	7	4.25	1.62	12	57	0.58
22	6	4.00	1.42	18	57	0.82
23	7	4.25	1.62	24	78	0.75
24	5	4.25	1.22	12	35	0.49
25	6	4.00	1.42	18	54	0.98
26	6	4.00	1.42	6	25	0.72
27	8	4.00	1.42	18	60	0.92
28	5	4.25	1.62	24	58	1.37
29	6	4.00	1.42	30	66	1.34
30	5	4.25	1.22	24	56	0.72
31	4	4.00	1.42	18	27	1.52

In this study, an economic analysis of the experiment was also performed for the process. Cost calculations that were made for unit COD removal is based on electricity consumption, since energy is the most expensive item of the electrocoagulation processes. Energy consumptions (C_{energy}) per unit volume (m^3) of the treated wastewater were determined by using the following equation:

$$C_{energy} = \frac{V \times I \times t}{v} \qquad (2)$$

where V shows the applied voltage value (V), I is the current (A), t is detention time (s), and v is the volume of treated wastewater. Calculated C_{energy} values have been multiplied by unit electricity cost in Turkey (0.12 $/Kwh). Finally, the calculated cost has been divided by the amount of removed COD (g) to obtain energy cost per unit COD removal ($/g COD removal).

The highest and lowest energy costs per unit COD removal have been obtained for 31st and 12th experimental runs (Table 2). Considering the corresponding COD removal

efficiencies, it is clearly seen that higher energy cost does not yield higher removal efficiencies. This result indicates the importance of energy cost optimization for this process.

By using data provided by the proposed experimental design, CCD approach develops second order surface model to predict the targeted responses (*y)*. The general expression for the quadratic regression model is given below:

$$y = \beta_0 + \sum_{i=1}^{k}\beta_i x_i + \sum_{i=1}^{k}\beta_{ii}x_i^2 + \sum_{i=1}^{k-1}\sum_{i=1}^{k}\beta_{ij}x_i x_j + \varepsilon \qquad (3)$$

In the equation β_0, (β_1, β_2, β_3), (β_{12}, β_{12}, β_{23}) and (β_{11}, β_{22}, β_{33}) are the intercept, linear, interaction and quadratic regression coefficients, respectively. k represents the number of factors and ε shows the random error of the model (Gengec *et al.,* 2013).

In this study, following regression models were obtained to predict COD removal efficiencies (Eq 4) and energy consumption costs (Eq 5), considering both individual and interactive effects of pH (x_1), conductivity (x_2), current (x_3), and time (x_4) in terms of coded factors.

COD removal (%)=$-512.95 + 70.48 x_1 + 76.88 x_2 + 223.88 x_3 - 3.91 x_4 - 2.47 x_1^2 + 2.55 x_2^2 + 7.11 x_3^2 - 0.06 x_4^2 - 7.75 x_1x_2 - 4.69 x_1x_3 + 0.28 x_1x_4 - 46.25 x_2x_3 + 1.38 x_2x_4 + 0.05 x_3x_4$ (4)

Cost ($/removal g COD)=$0.0191 - 0.571 x_1 + 1.019 x_2 - 0.759 x_3 + 0.195 x_4 + 0.068 x_1^2 - 0.366 x_2^2 - 0.978 x_3^2 + 0.0006 x_4^2 + 0.028 x_1x_2 - 0.247 x_1x_3 - 0.009 x_1x_4 + 1.363 x_2x_3 - 0.036 x_2x_4 + 0.008 x_3x_4$ (5)

The developed models represented by Eq (4) and Eq (5) provided good prediction efficiencies for corresponding resp"onses. R^2 values were determined as 0.95 and 0.92 for COD removal (Figure 2a) and energy costs (Figure 2b) models, respectively.

3.2. Analysis of variance (ANOVA)

Even though high correlation coefficients indicated the appropriateness of the developed predictive models obtained, analysis of variance (ANOVA) was also performed to examine the adequacy and significance of the models.

Statistical parameters obtained from the ANOVA for the models have been presented in Table 3.

Results of the ANOVA tests have confirmed the appropriateness of the obtained quadratic models. The p-values of the models, which are <0.05, indicate statistical significance (Ahmadi *et al.,* 2005; Saini and Kumar, 2016). In addition, F-values of the regression models are significantly higher than tabulated F(2.352) at 95% significance. This result also confirms the accuracy of the models (Hassani *et al.,* 2016).
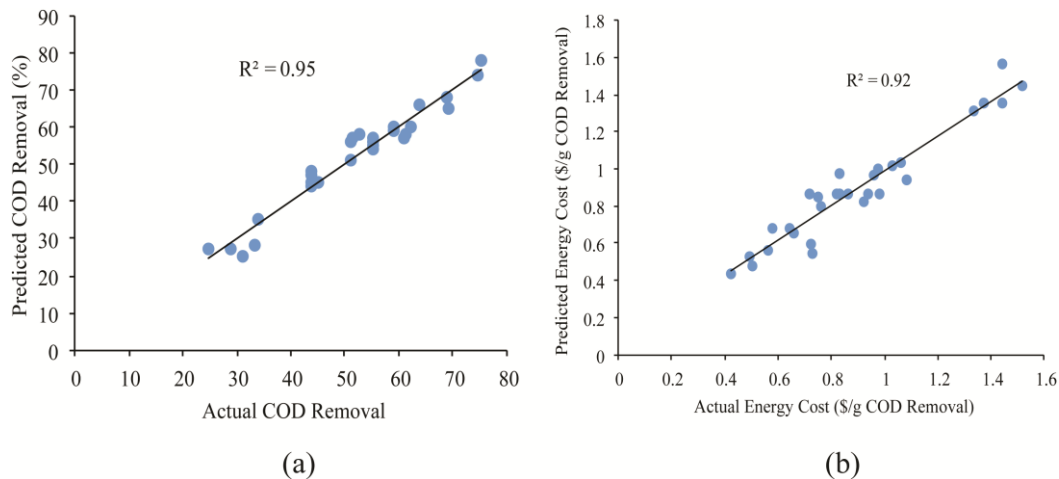
Figure 2. Predictive models for a) COD removal and b) energy costs

Table 3. Analysis of variance for a) COD removal and b) energy consumption costs

Source	DF	Seq SS		Adj SS		Adj MS		F		P	
		a	b	a	b	a	b	a	b	a	b
Regression	14	4974.44	2.341	4974.44	2.341	355.317	0.167	21.76	13.53	0.000	0.000
Linear	4	4420.50	1.908	290.33	0.080	72.581	0.020	4.44	1.62	0.013	0.218
Square	4	280.57	0.227	280.57	0.227	70.142	0.057	4.30	4.60	0.015	0.012
Interaction	6	273.37	0.206	273.37	0.206	45.562	0.034	2.79	2.78	0.047	0.048
Residual error	16	261.30	0.198	261.30	0.198	16.331	0.012				
Lack of fit	10	251.58	0.172	251.58	0.172	25.158	0.017	15.54	4.09	0.002	0.049
Pure error	6	9.71	0.025	9.71	0.025	1.619	0.004				
Total	30	5235.74	2.539								

In this study, the Pareto analysis was also performed to determine the percentage effect of independent variable on the response. The following expression was used for Pareto calculations (Khataee *et al.,* 2010):

$$P_i = \left(\frac{b_i^2}{\sum_{i=1}^{n} b_i^2} \right) \times 100 (i \neq 0) \tag{6}$$

In the equation, b_i represents the regression coefficients in quadratic model equations.

Pareto analysis was applied to both of the models developed for predicating COD removal and energy consumption costs. It was observed that current density has been the most effective factor (79.18%) on the COD removal. It was followed by conductivity (9.34%) and pH (7.85%).

Results of Pareto analysis showed that individual impacts of pH, conductivity, current density and electrolysis period on energy consumption costs are 6.53%, 20.79%, 11.53% and 0.76%, respectively. Among the interactive impacts, current density x conductivity has the highest effect on energy cost with 37.20%.

3.3. Validation of the model

Additional experiments were performed to validate the model. With this aim, 12 different experimental runs were decided which were not used previously (Table 4).

Table 4. Design of the experiments for validation tests

Run	pH	Conductivity (mS/cm)	Current (mA/cm²)	Time (min)	COD Removal (%)	
					Actual value	Predicted Value
1	3	3.5	1.82	10	22	18.84
2	3	3.5	1.82	15	25	21.15
3	3	3.5	1.82	20	28	20.70
4	5	3.5	1.82	10	48	54.64
5	5	3.5	1.82	15	52	59.79
6	5	3.5	1.82	20	58	62.12
7	7	3.5	1.82	10	56	70.71
8	7	3.5	1.82	15	78	78.64
9	7	3.5	1.82	20	78	83.82
10	7	3.5	1.02	10	33	30.80
11	7	3.5	1.42	20	58	62.51
12	7	3.5	1.82	30	89	85.92

COD removal efficiencies were measured and also predicted by using previously obtained predictive model (Eq 4). A high correlation coefficient was determined for the relationship between the predicted and actual COD

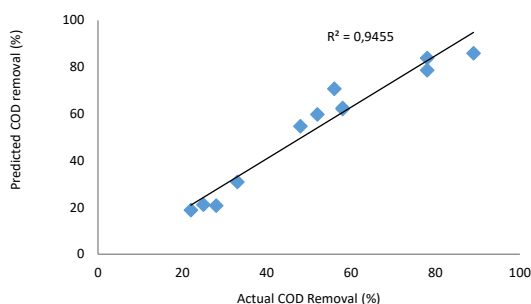removal efficiencies. R^2 value of 0.95 demonstrates the validity of our model (Figure 3).

Figure 3. Validation of RSM model for COD removal

3.4. Response surface plots for COD removal efficiency

In this study, the interactions between dependent and independent process variables were investigated by using response surface plots. In order to examine bivariate interactions, hold values are decided to be as pH=6, conductivity=4, current=1.42 and time: 18 min.

When pH interactions are investigated, it is clearly seen from the corresponding figures that (Figure 4 b,c,d) effective COD removal has been achieved for pH values between 6 and 7.5. This result is in good agreement with the literature. This pH values yielded formation of monomeric and polymeric aluminum complexes such as $Al(OH)_3$, $Al(OH)^{2+}$, $Al_{17}(OH)_{32}^{7+}$ which are effective coagulants improving the performance of electrocoagulation processes (Martinez-Huitle and Brillas, 2009). Impact of pH is more significant especially for lower conductivity conditions (Chen, 2004). Results of our study have also confirmed this knowledge (Figure 4b).

Similar to many other chemical processes, duration period is an important process variable for electrocoagulation. This process starts with the neutralization of particles by ions released from the electrodes and followed by coagulation. Increase in the electrolysis period increases the released ion concentration and in this way, formation of hydroxyl flocks are improved (Daneshvar *et al.,* 2004). In our study, reaction periods longer than 20 minutes provided sufficient COD removal efficiencies (>70%) (Figure 4a, d, e).

In electrocoagulation systems, current density determines the released metal ion dose. Increase in current density yields improvement in ion production and consequently better flocculation efficiencies (Daneshvar *et al.,* 2004). This fact can also be proved by Faraday's Law which is useful for determining maximum dissolved mass (g) of anode material (m) (Gatsios *et al.,* 2015):

$$m = \frac{I \times t \times M_a}{F \times z} \tag{7}$$

In the equation I, t and M_a represent applied current (A), electrolysis period (s) and specific molecular weight of anode material (g/mol) respectively. F shows Faraday's constant (96,485.34 As/mol) and z is the number of electrons in the reaction. As seen from the Eq (7), mass of the dissolved anode material is proportional with both

current density and time. Undoubtedly, high current densities will improve the electrocoagulation efficiencies. In order to achieve a predetermined mass of a certain anode material, adjustment can be made between electrolysis period and current density. In other words efficient treatment can be achieved by applying high currents for shorter periods or vice versa. Considering the fact that electrolysis period and/or current density are the most important parameters influencing the operational costs, it is clearly seen that optimization is mandatory. As seen from Figure 4a, in order to achieve sufficient treatment (higher than 70%) current density should be higher than 1.6. This value can be decreased by increasing electrolysis period.

Conductivity of solution is another important parameter in electrocoagulation processes affecting the current efficiency and energy consumption (Daneshvar *et al.,* 2006). When surface plots involving conductivity is investigated, the positive impact of conductivity can be clearly seen (Figure 4b, e, f).

According to literature, it was observed that 60-89% COD removal efficiency was obtained in EC studies with Al electrode carried out with different contaminant sources and this is consistent with the results obtained in this study (Table 5).

3.5. Response surface plots for energy costs

In this study, energy consumption costs are analyzed by using response surface plots (Figure 5). Similar to previous applications, hold values were determined to be medium ones (pH=6, conductivity=4, current=1.42 and time: 18 min).
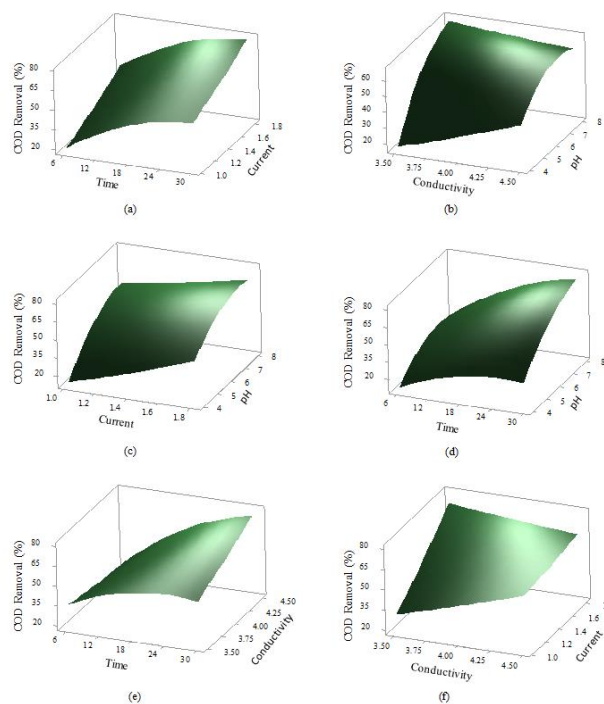

Figure 4. Surface plots for COD removal efficiency as a function of a) current density and time b) pH and conductivity c) pH and current density d) pH and time e) conductivity and time f) conductivity and current density

As seen from Figure 5a, energy consumption costs ($) per unit COD removal (g) can be minimized by adjusting pH to 6-7 and applying low current densities (<1.1 mA/cm^2). It is clearly seen from Figure 5b that high conductivity (>4.25 mS/cm) provides cost-effective treatment options (<0.2 $/g COD removed).

Table 5. Al electrod ile elde edilen COD verimleri

Pollution sources	Current density (A/m^2)	Reaction time (min)	COD removal (%)	Reference
Urban wastewater	200	30	85	Elazzouzi *et al.,* 2017
Real dyehouse wastewater	65	80	77	Kobya *et al.,* 2016
Biologically treated municipal wastewater	75.2	40	86	Chopra and Sharma, 2015
Leachate	200	60	60.5	Kabuk *et al.,* 2014
Egg processing effluent	200	30	89	Sridhar *et al.,* 2014
Domestic wastewater	100	10	72	Ozyonar and Karagozoğlu, 2011
Textile wastewater	100	60	88	Zongo *et. al.,* 2009
Potato chips manufacturing wastewater	15-18	15-30	70-89	This study

The current density and electrolysis period are the most effective parameters on energy consumption cost. Low energy costs can be achieved by decreasing time periods (<15 min) and current density (<1.1 mA/cm^2) that is shown in Figure 5c.
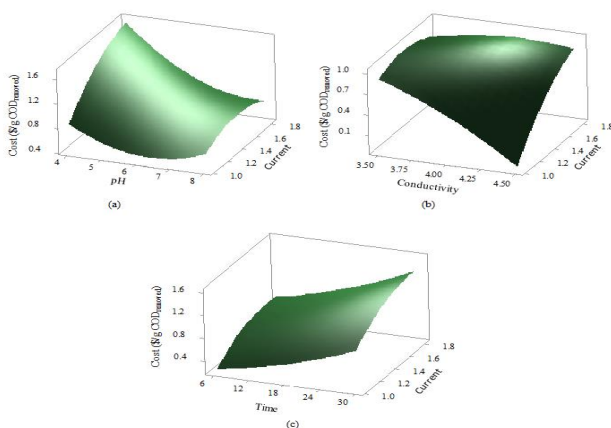

Figure 5. Surface plots for energy consumption costs as a function of a) pH and current density b) current density and conductivity c) current density and time

4. Conclusions

The objectives of this study were to optimize the COD removal efficiencies and energy consumption costs for treatment of food wastewaters by electrocoagulation process. With this aim central composite design method was applied and predictive quadratic models were developed for both of the responses. The accuracy of the models was confirmed by both high R^2 values (0.95 and 0.92 for COD removal and energy cost prediction, respectively) and ANOVA tests. p values obtained from ANOVA analysis were lower than 0.05 for both of the regression equations demonstrating the appropriateness of the models. Accuracy of the model developed for predicting COD removal was also confirmed by the conducted validation tests. R^2 of validation was also sufficient (0.95). In this study, impacts of process parameters on response values were determined numerically by Pareto analysis. Current density was found to be the most effective factor (79.18%) on COD removal whereas current density x conductivity has the highest impact (37.20%) on energy consumption costs.

According to the legal regulations in Turkey (WPCA, 2004) limit value of COD parameter for vegetable and fruit processing sector is set to be 150 mg/L. In order to achieve discharge limit, 70% COD treatment efficiency is targeted for the studied wastewater. Results of the study showed that optimum treatment can be provided by applying 1.5 mA/cm^2 current density for 20 min electrolysis period under original pH (7.8) and conductivity (3.40 mS/cm). Energy consumption cost has been determined as 0.9 $/g COD removal for these conditions.

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