The Effects Of Operating Parameters On Temperature And Electrode Dissolution In Electrocoagulation Treatment Of Petrochemical Wastewater

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Abstract

This work presents the of study current density NaCl concentration and electrolysis time effects on the solution temperature and electrode dissolution during electrocoagulation removal of turbidity from petrochemical wastewater. In addition, the effects of the parameters on energy consumption were also looked into. To investigate current density effect on solution temperature, experiments were carried out at different values of current density while the concentration of supporting electrolyte was kept at 6.25 g/L. The temperature was measure as a function time for each value of current density. The results showed that current density had a warming effect on reactor temperature, and this effect increased with time. When current density values were 0.0988, 0.1975 and 0.3951A/cm^2 the temperature values were measured to be 27, 29, 46 ^{o}C respectively for 15 minutes electrolysis done. Due to separation problem encountered when experiments were carried out without cooling, to study the combined effect of these operating conditions on electrode dissolution the rector was surrounded by cooling system at 5°C, but solution temperature change was still noted. The results of analysis of statistically generated experiments showed that aluminum electrode dissolution was singly and interactively affected only by current density and electrolysis time. This seems to agree with Faraday's law of electrolysis. The temperature was significantly affected by current density, NaCl concentration, simultaneous variation of current density and electrolysis time.

1. Introduction

The extensive use of industrial products such as petrochemicals has led to discharge of large amount organic compounds into aquatic system which brings about contamination of all environmental resources [1],[2]. Petrochemical wastewater contains groups of compounds that due to their hazardous nature are listed as US EPA and EU priority pollutants [3]. And such pollutants deserve thorough treatment to reduce their concentrations in the wastewater before discharge into water body.

Recently, researchers have revealed electrocoagulation as an attractive and suitable method for the treatment of various kinds of wastewater due to its characteristics such as, ease of operation, energy efficiency, versatility, environmental compatibility and cost effectiveness [4]. Electrocoagulation has been successfully used to treat tap water [5], paper mill wastewater [6], heavy metals containing solution [7],[8], oily water [9], textile wastewater [10] , [11], slaughter house wastewater [4]. Electrocoagulation refers to that process in which electrochemically generated coagulant is responsible for destabilization of pollutants present in the aqueous medium. Generally, in this process three steps are involved, these are electrolytic oxidation of sacrificial electrodes to form coagulant, destabilization of pollutants present in the water or wastewater and agglomeration of the destabilized pollutant to form flocs. Two types of reaction take place during electrocoagulation, anodic reaction refers to dissolution of electrode used which is usually aluminium or iron, and cathodic reaction involves formation of hydrogen gas and hydroxyl group. These reactions lead to generation of coagulant. For the case of aluminum, anodic and cathodic reactions are as follow [12]:

Anodic reaction

$$Al_{(s)} \rightarrow Al^{3+}{}_{(aq)} + 3e^- \tag{1}$$

Cathodic reaction

$$3H_2O + 3e^- \rightarrow \frac{3}{2}H_{2(g)} + 3OH^- \tag{2}$$

In the solution:

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \tag{3}$$

However, efficiency of electrocoagulation depends on current density, electrode material, electrolysis time, temperature, pH, and conductivity of the solution. Current density has been identified as the key operational parameter influencing which pollutant removal mechanism dominates [13]. Current is also said to possibly have warming effect on the system fluid [14]. But this effect may be negligible when experiment is carried out at low value current density. At higher temperature the increase in solubility of precipitates of $Al(OH)_3$ and generation of unstable of flocs can be observed. Consequently these may have adverse effects on the efficiency of the process. Katal and Pahlavanzadeh (2011) has reported a negative effect of temperature on the removal efficiency of an electrocoagulation process used in the treatment of heavy metals containing solution [6]. In most cases where temperature is being considered as an electrocoagulation factor, the process is treated as endothermic process which is achieved by circulating hot water round the reactor especially when low current is applied, whereas it is exothermic by nature. So, it is important to find out what factors contribute temperature change during the process especially when it is carried out at ambient temperature.

In electrocoagulation, electrode dissolution rate becomes important in evaluating removal rate of the process. In other words, removal rate of electrocoagulation is proportional to rate of formation hydro-pollutant-aluminum flocs, for a case where aluminum electrodes are used. And this is dependent of dissolution rate [15]. Another important parameter in electrocoagulation process is energy consumption. This is the only factor that can guarantee the replacement of conventional chemical treatment processes which generate large amount of sludge by clean technologies such as electrocoagulation in industrial wastewater plants, since cost of electrode is low compare to that of energy.

Therefore, this work investigates the effects of operating parameters such as current density, NaCl and electrolysis time on aluminum electrode dose, solution temperature and energy consumption during electrocoagulation treatment of turbidity in petrochemical wastewater. Both conventional and statistical experimental plan approach were used to accomplish the aim of the study.

2. Materials and Methods

The experiments were carried in a Plexiglas made batch reactor with active capacity of 0.8 or 1L, using 2 or 4 aluminium electrodes connected in mono-polar mode to a constant D.C power source. The electrodes were spaced 1.5 cm apart. Prior to each experiment, the electrodes were thoroughly washed and rinsed with distilled water in order to remove any impurities that may negatively affect dissolution. NaCl was added to wastewater as supporting electrolyte. During the experiments the solution was constantly stirred using a magnetic stirrer (Chiltern HS31), to prevent temperature or concentration gradient in the reactor. The electrode dose at specific value of current density was determined using gravimetric method where the weight of electrodes was measured before and after the each experiment. An analytical balance with readability of 0.0001g (Scaltec, SBC 31) was used for the electrode weight measurement. The solution temperature was measured using a thermometer. A real petrochemical wastewater was used for this study to have a picture of how electrode dissolution and temperature of the wastewater under treatment vary with change in operating parameters such as current density, supporting electrolyte concentration and electrolysis time when EC is applied in industrial wastewater treatment, even when no chemical is added. Therefore, pH of the wastewater was not adjusted. The schematic diagram for the experimental set up is given in Figure 1.

Effect of current density on solution temperature was investigated by varying the values of current density between of 0.099 and 0.39 A/cm^2 and measuring the reactor temperature at regular interval of 5 min, for each value of current applied. In all the experimental runs current was held constant. To study combined effect of current density, NaCl concentration and electrolysis time on electrode dissolution and temperature data obtained from a set of statistically designed experiments were used. The effects of these parameters on energy consumption were also studied. With the aid of Design Expert 7.0.0, using central composite design, 20 experiments comprising of 8 factorial points, 6 axial points and 6 were generated. The design matrix in actual units and the experimental results are given in Table 4. And Table 3 presents the actual and coded factors used at five different levels. Also, using the same software regression analyses of the responses were done by choosing quadratic model. Equations (4) and (5) were used to determine the faradaic electrode dose (D_f) and current efficiency (*CE*) respectively. Energy consumption was calculated from Equation (6)

$$D_f = \frac{IMt}{zF} \tag{4}$$

$$CE(\%) = \frac{D}{D_f} \times 100 \qquad\qquad (5)$$

$$E = IVt \qquad\qquad (6)$$

where, I, M, t, z and F are current (in ampere), molecular weight (for Al equals 27 gmol^{-1}), electrolysis time (in seconds), number of electrons oxidized by one mole of metal (z_{Al}= 3) and Faraday Constant (96485.3399 Cmol^{-1}) respectively.

1. DC power supply
2. The wastewater
3. Jacket
4. Stirrer bar
5. Peristaltic pumps
6. Electrodes
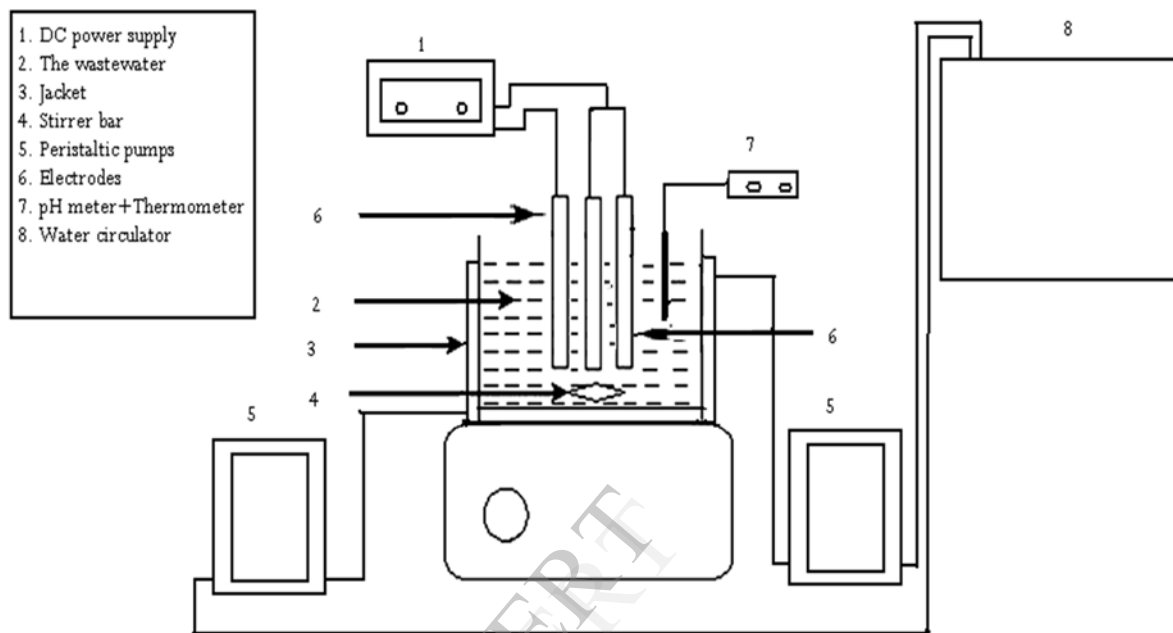7. pH meter+Thermometer
8. Water circulator

Figure 1. Schematic diagram of the experimental set up

3. Results and Discussions
3.1. Electrocoagulation

The results of the experiment confirmed electrocoagulation as an efficient method for abatement of turbidity in petrochemical wastewater. The sample withdrawn after 30 minutes when current density of 0.099 A/cm^2 was applied is given in Figure 2. Though, the solution appeared a bit cloudy. This resulted from excessiveness of electrochemically generated coagulant in the solution. At this current density 10 min treatment time would have given a better result. Besides, extension of treatment time to 90 min for all the values of current density led to formation of unstable floc. This made post treatment separation a bit difficult.
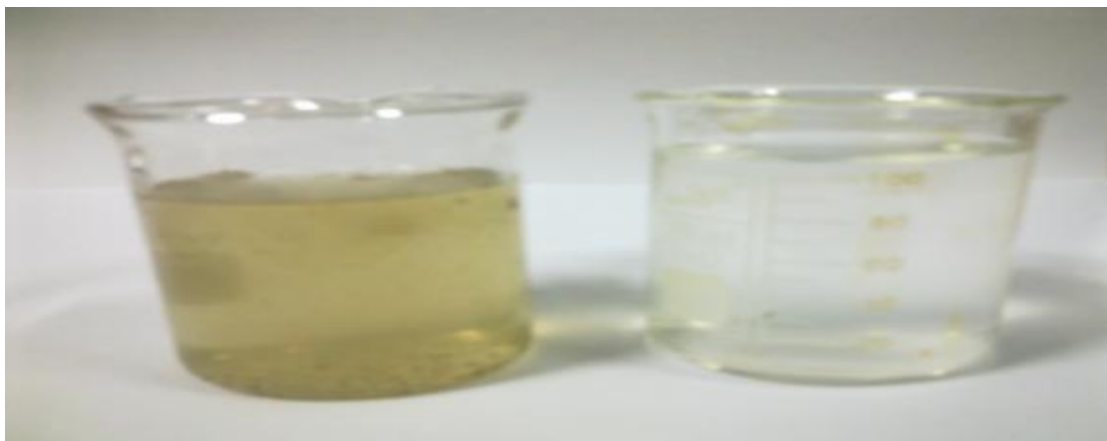

Figure 2. The wastewater before and after electrocoagulation treatment

## 3.2. Effect of operating conditions on solution temperature

As shown in Figure 3-5 temperature varied linearly with time for all the values of current density. It also increased with increase in current density (Table 1). For instance, at the 15 min electrolysis, when current densities of 0.099 A/cm^2, 0.196 A/cm^2 and 0.395 A/cm^2 were applied, the temperature was measured to be 27 ^{o}C, 29 ^{o}C and 46 ^{o}C respectively. Moreover, the results of statistically generated experiments showed that solution temperature change during electrocoagulation was not only affected by current density but also concentration of supporting electrolyte and electrolysis time. Temperature was observed to increase with increase in current density value, at 1.25 g/L and 27 min increasing and decreasing current density by 12.16 mA/cm^2 led to temperature change of 7°C and 0.5 ^{o}C respectively. However, decrease in NaCl concentration gave rise to increase in solution temperature at 18.17 mA/cm^2 27 min where decrease and increase in NaCl concentration by 0.75 g/L brought about 2 ^{o}C and 0°C temperature change respectively. But, the reverse of this was observed at 25.4 mA/cm^2 and 17.09 min, 10.94 and 37.91 min. Also, the effect electrolysis time on this response seems unimportant. For instance, at 10.94 mA/cm^2 and 0.8 g/L, when electrolysis time was 37.91 min and 17.09 the solution temperature was 21 ^{o}C.

Table 1. The effect of current density on solution temperature at different time interval
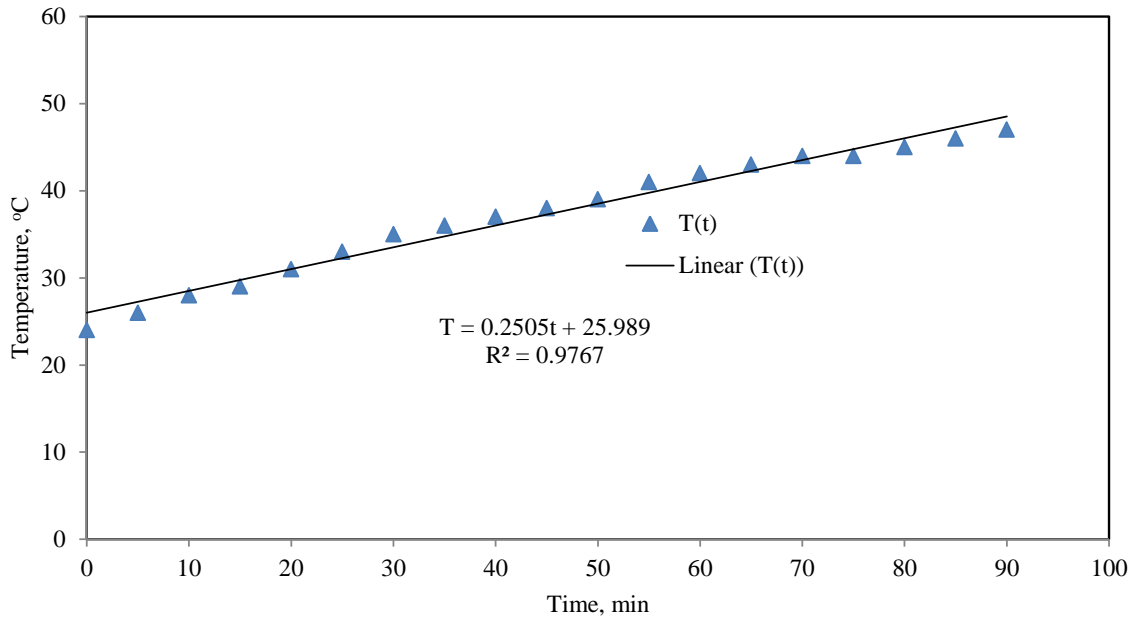
Current density, A/cm^2	Temperature, ^{o}C			
	t= 5 min	t=15 min	t=30 min	t=45 min
0.0988	24	27	30	33
0.1975	26	29	35	38
0.3951	34	46	55	59

Table 2. The effect of current density on electrode dose

Current density, A/cm^2	Electrode dose, g
0.0988	0.6888
0.1975	1.0626
0.3951	2.9891

$$T = 0.193t + 23.737$$
$$R^2 = 0.9926$$
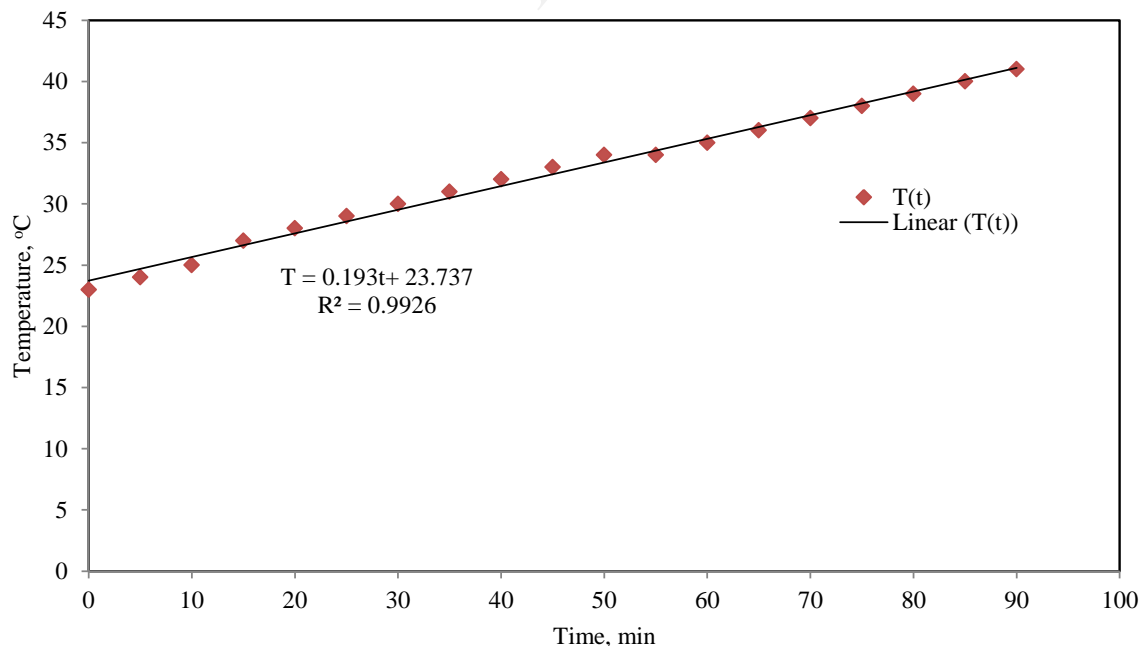
Figure 3. Solution temperature as a function of electrolysis time (current density= 0.099 A/m^2, NaCl concentration=6.25 g/L, pH=6.08)

Figure 4. Solution temperature as a function of electrolysis time (current density= 0.198 A/m^2, NaCl concentration=6.25 g/L, pH=6.08)
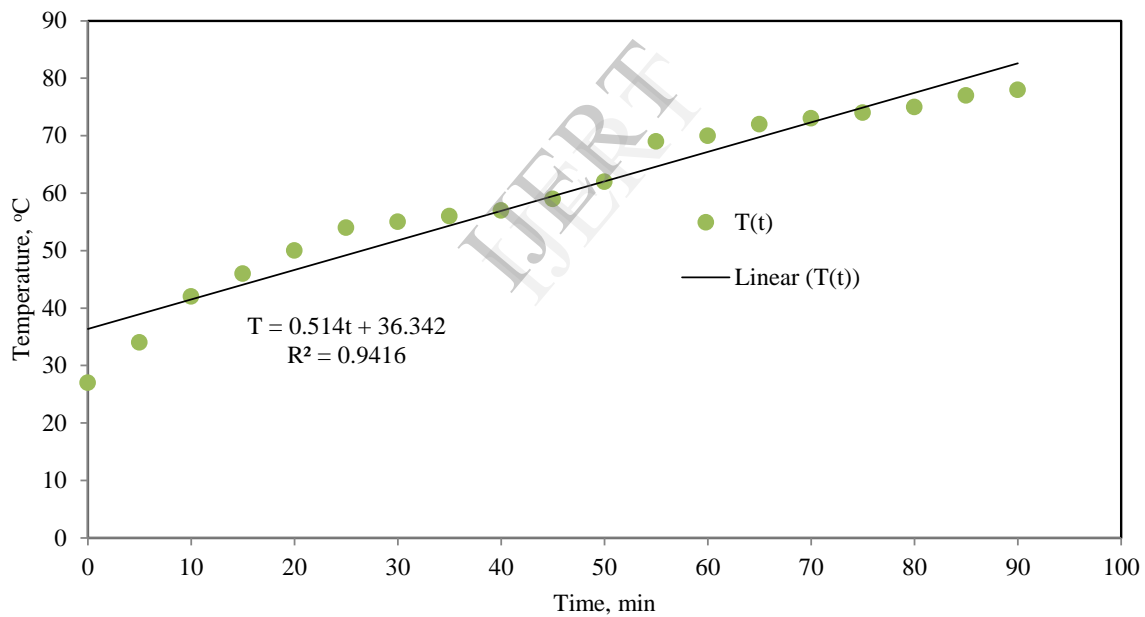
Figure 5. Solution temperature as a function of electrolysis time (current density= 0.395 A/m^2, NaCl concentration=6.25 g/L, pH=6.08)

3.3. Effects of operating conditions on electrode dose

Faraday's Law gives the expression that relates applied current and time with electrode dose. This is given in Equation 4. According to this law electrode dose is expected to increase with increase in value of applied current/current density and electrolysis time. As shown in table 2 electrodissolution of aluminum electrodes increased with increase in current density; at 90 min electrolysis when current density values were 0.099 A/cm^2, 0.198 A/cm^2 and 0.395 A/cm^2 the electrode doses were 0.6888 g, 1.0626 g and 2.9891 g respectively. Looking at Table 4, at the center points ([18.17 1.25 27.5]), the average electrode dose was 0.2205 g. From this point an increasing step change of current density to positive axial point led to delivering of 0.3641 g of aluminum into the solution. While at the negative axial point electrode dose was 0.1638 g. Similarly a step increasing and decreasing changes of electrolysis time from the center point to axial points gave electrode dose of 0.242 g and 0.1886 g respectively. Conversely, similar change of NaCl concentration to the positive and

negative axial point yielded 0.074 and 0.2673 g dose of aluminum respectively. But increasing its concentration from 0.8 to 1.7 g at 25.4 mA/cm^2 and 17.09 min led to increase in electrode dose from 0.1416 g to 0.1672 g. The maximum electrode dissolution of 0.4204 g was obtained at the maximum design point (where current density= 25.40 mA/cm^2, NaCl concentration = 0.8 g and electrolysis time = 45 min. from here, it can be seen that while current density and electrolysis time affect the dissolution in a certain trend, variation of NaCl concentration possesses no significant effect on it. Thus, the results of the experiments agree with Faraday's law of electrochemistry. However, faradaic electrode dose varied significantly for all the experimental runs as indicated by current efficiency values. These are given in Table 4.

Table 3. The factorial levels of the design matrix

Actual factor, unit	Coded factors	Real values of the factors at five different levels				
		-1.8618	-1	0	1	1.8618
Current density, mA/cm^2	x_1	6.005	10.935	18.166	25.397	30.327
NaCl concentration, g/L	x_2	0.5	0.8	1.25	1.7	2
Electrolysis time, min	x_3	10	17.09	27.5	37.9	45

Table 4. The design matrix and the experimental results

	Factors			Responses			
Run no	X_1, mA/cm^2	X_2, g/L	X_3, min	D, g	T, ^oC	E, kWh/m^3	CE, %
1	18.17	1.25	27.50	0.1958	22	25.9646	123.5123
2	10.94	1.70	37.91	0.1354	19	11.7521	102.9297
3	25.40	0.80	17.09	0.1416	24	26.6604	102.8078
4	18.17	1.25	27.50	0.2738	22	25.9646	172.7154
5	10.94	0.80	37.91	0.1697	21	15.2777	129.0042
6	18.17	1.25	27.50	0.2943	22	25.9646	185.647
7	10.94	0.80	17.09	0.0521	21	6.71067	87.85602
8	25.40	1.70	17.09	0.1672	23	21.7385	121.3946
9	18.17	1.25	27.50	0.1886	22	25.9646	118.9705
10	25.40	0.80	37.91	0.4204	26	59.1396	137.5986
11	25.40	1.70	37.91	0.3501	28	54.5904	114.5891
12	10.94	1.70	17.09	0.1918	22	5.2979	323.4316
13	18.17	1.25	45.00	0.242	23	42.4875	93.28959
14	6.01	1.25	27.50	0.1638	18.5	3.11667	313.0184
15	18.17	1.25	27.50	0.2196	22	25.9646	138.5256
16	18.17	0.50	27.50	0.2673	25	23.6042	168.6152
17	18.17	2.00	27.50	0.074	18	23.6042	46.67984
18	18.17	1.25	10.00	0.1288	20	9.44167	223.4324
19	18.17	1.25	27.50	0.1508	23	25.9646	95.12595
20	30.33	1.25	27.50	0.3641	25	51.2417	137.5394

3.4. Effects of operating conditions on energy consumption

Energy consumption is a very important parameter in electrochemical processes in general. It determines the feasibility of their application in industrial wastewater treatment plants. This is because operating cost of the process is more affected by energy cost which is proportional to energy consumption. The results of the experiments showed that the energy consumption was highly affected by current density and electrolysis time. It increased with increase in current density and electrolysis time. For instance, At 1.25 g/L and 27.5 min increasing and decreasing current density by 12.16 mA/cm^2 led to energy consumption of 25.9646 kWh/m^3 and 3.1167 kWh/m^3. Similarly, at 18.17 mA/cm^2 and 1.25 g/L when electrolysis time was increased and decreased

by 17.5 min energy consumptions were 42.4875 kWh/m^3 and 9.4417 kWh/m^3 respectively. Increasing current density can lead excessive evolvement of hydrogen gas which increases energy consumption by reducing electrical conductivity Energy consumption was also affected by NaCl concentration but its effect at axial points was insignificant. In factorial experiments, Energy consumption decreased with increase in NaCl concentration. For instance, at 10.94 mA/cm^2 and 37.91 min, altering NaCl concentration by a factor of 2.215 resulted into reduction of energy consumption by 3.5256 kWh/m^3. Similarly, at 25.4 mA/cm^2 and 37.91 min, when NaCl concentration was increased from 0.8 g/L to 1.7 g/L energy consumption decreased from 59.1396 kWh/m3 to 54.5904 kWh/m^3. This must have been due to the fact that increasing NaCl concentration improves electrical conductivity which in turn reduces resistance. Provided current remains constant, decrease in resistance value will lead to decrease in voltage and energy consumption.

3.5. Statistical analysis

The results of analysis of variance showed that the experimental data fitted well with quadratic polynomial model for the three responses correlation coefficients (R-square) were close to unity. Moreover, for energy consumption model adjusted and predicted R-square value were very close to each other and to 1 (Table 7). This means that the quadratic polynomial model (Equation 9) is statistically significant. Also, the second order polynomial equation developed for electrode dose was significant with p-value less than 0.05 and insignificant Lack of Fit. The model had as its significant model terms x_1, x_3 and x_1x_3 (Table 5). The electrode dose model in terms of coded factors is given in Equation (7). On the other hand, the temperature reduced quadratic polynomial model had significant Lack of Fit but it is still statistically significant with p-value of 0.0005. The terms x_1, x_2, and x_1x_3 significantly affected the model (Table 6). The temperature model in coded units is given in Equation 8.

$$D = 0.2206 + 0.0635x_1 - 0.0194x_2 + 0.0522x_3 - 0.0188x_1x_2 + 0.0501x_1x_3$$
$$- 0.0337x_2x_3 + 0.0149x_1^2 - 0.0181x_2^2 - 0.0129x_3^2 \tag{7}$$

$$T = 22.23 + 2.12x_1 - 0.86x_2 + 0.66x_3 + 1.25x_1x_3 + 0.15x_1^2 \tag{8}$$

$$E = 25.98 + 14.94x_1 - 1.06x_2 + 9.95x_3 - 0.57x_1x_2 + 6.29x_1x_3$$
$$- 0.22x_2x_3 + 0.34x_1^2 - 0.92x_2^2 - 0.087x_3^2 \tag{9}$$

Table 5. ANOVA results for electrode dose

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F
ANOVA for Response Surface Quadratic Model- electrode dose Response: electrode dose, g					
Model	0.14	9	0.016	4.72	0.0118
x_1-Current density	0.055	1	0.055	16.66	0.0022
x_2-NaCl conc	5.12E-03	1	5.12E-03	1.55	0.2418
x_3-Electrolysis time	0.037	1	0.037	11.27	0.0073
x_1x_2	2.82E-03	1	2.82E-03	0.85	0.3778
x_1x_3	0.02	1	0.02	6.06	0.0335
x_2x_3	9.11E-03	1	9.11E-03	2.75	0.128
x_1^2	3.19E-03	1	3.19E-03	0.97	0.3489
x_2^2	4.72E-03	1	4.72E-03	1.43	0.2596
X_3^2	2.39E-03	1	2.39E-03	0.72	0.4148
Residual	0.033	10	3.31E-03		
Lack of Fit	0.018	5	3.66E-03	1.24	0.4103
Pure Error	0.015	5	2.96E-03		
Cor Total	0.17	19			
R-square= 0.8095 Adj R-square= 0.6381					

Table 6. ANOVA results for temperature

ANOVA for Response Surface Reduced Quadratic Model Response: Temperature, ^{o}C					
Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F
Model	90.24	5	18.05	9.06	0.0005
x_1-Current density	61.29	1	61.29	30.76	< 0.0001
x_2-NaCl conc	10.15	1	10.15	5.09	0.0405
x_3-Electrolysis time	5.99	1	5.99	3.01	0.1049
x_1x_3	12.5	1	12.5	6.27	0.0252
x_1^2	0.31	1	0.31	0.16	0.6973
Residual	27.89	14	1.99		
Lack of Fit	27.06	9	3.01	18.04	0.0027
Pure Error	0.83	5	0.17		
Cor total	118.14	19			
R-square = 0.7639 Adj R-square = 0.6796					

Table 7. ANOVA results for energy consumption

ANOVA for Response Surface Quadratic Model Response : Energy consumption, kWh/m^3					
Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F
Model	4750.51	9	527.83	328.16	< 0.0001
x_1-Current density	3048.06	1	3048.06	1895.04	< 0.0001
x_2-NaCl concentration	15.2	1	15.2	9.45	0.0118
x_3-Electrolysis time	1352.92	1	1352.92	841.13	< 0.0001
x_1x_2	2.57	1	2.57	1.6	0.235
x_1x_3	316.39	1	316.39	196.7	< 0.0001
x_2x_3	0.38	1	0.38	0.24	0.6381
x_1^2	1.69	1	1.69	1.05	0.3299
x_2^2	12.25	1	12.25	7.61	0.0202
x_3^2	0.11	1	0.11	0.068	0.7992
Residual	16.08	10	1.61		
Lack of Fit	16.08	5	3.22		
Pure Error	0	5	0		
Cor Total	4766.6	19			
R-Square = 0.9966 Adj R-Square = 0.9936 Pred R-Square = 0.9741					

4. Conclusions

In this paper, the effects of operating conditions on wastewater temperature during electrocoagulation treatment, electrode dose and energy consumption have been investigated. The three responses were highly affected by current density. The increase in current density led to increase electrode dose, solution temperature and energy consumption. But that of energy consumption is not desirable as it leads to high operating cost. While electrode dose and energy consumption were affected by variation of electrolysis time, its single effect on temperature was not significant within a range of 10-45 min, but this was also current density dependent. In the experiments carried out at high current density between 0.099 A/cm^2 and 0.395 A/cm^2 temperature was observed to vary linearly with time. Variation of NaCl concentration significantly affected solution temperature and energy consumption. Increasing NaCl concentration decreased both energy consumption and solution

temperature. In fact, no temperature change was observed at 18.17 mA/cm^2 and 27.5 min when concentration of Nacl concentration was 2 g/L. But, it had no effect on electrode dose.

5. Nomenclature

CE   Current efficiency, %
D   Electrode dose, g
D_f Faradaic electrode dose, g
E   Energy consumption, kWh/m^3
F   Faraday's constant, 96485.3399 Cmol^{-1}
I   Current, A
M   Molecular weight, for Al= 27 g/mol
NaCl   Sodium chloride
T   Temperature, ^{o}C
t   Time, s or min or hour
V   Voltage, volt
X_1 Actual current density, mA/cm^2
x_1 Coded current density
X_2 Actual NaCl concentration, g/L
x_2 Coded NaCl concentration
X_3 Actual electrolysis time, min
x_3 Coded electrolysis time
z   Number of electron oxidized, for Al, z=3

6. Acknowledgement

7. References

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