



## Statistical modeling on COD removal from metal-working fluids through electrocoagulation process

K.C. Praveen, C. Arun, N. Balasubramanian, K.V. Radha\*

*Department of Chemical Engineering, Anna University, A.C. Tech Campus, Chennai 600 025, Tamil Nadu, India  
Tel. +91 044 22359124; email: radhavel@yahoo.com*

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### ABSTRACT

The present investigation envisages with the percentage chemical oxygen demand (COD) removal from metal-working fluids through batch electrocoagulation reactor using mild steel anode and stainless steel cathode. Box–Behnken experimental design was used to optimize individual variables and interactions of variables on percentage COD removal. The operating variables are the current density ( $2.0\text{--}2.4 \text{ Adm}^{-2}$ ), the electrolyte pH (6–8), and the electrolysis time (90–110 min). The responses have been critically analyzed with Pareto analysis of variance, response surface methodology, and quadratic model for percentage COD removal. Analysis showed a high coefficient of determination value  $R^2$  (0.9527) for the percentage COD removal and satisfactory prediction for second-order regression model.

*Keywords:* Electrocoagulation; Box–Behnken method; COD removal; Response surface methodology; Current density

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### 1. Introduction

India has large number of industries, namely metal finishing industries, metal processing industries, iron and steel manufacturing industries, electroplating industries, etc. working on metal and its alloy. These industries generate a huge volume of high-strength metal-working fluids (MWFs) annually and that cause serious environmental problems. The effluents from these industries occupy fifth rank with respect to discharging pollutants into local water bodies and sewage systems [1,2]. The MFWs are characterized by a high organic load, dark color, low pH, and low

Biodegradability Index which cannot be treated by biological methods [3]. The MWFs contain various pollutants load (biocides, corrosion inhibitors, extreme pressure and anti wear agents, emulsifiers and surfactants) and have biochemical oxygen demand (BOD), chemical oxygen demand (COD), total organic carbon, and total dissolved solids [4].

Many methods are performed for the last two decades to treat wide spectrum of effluent. Treatment processes that include precipitation, co-precipitation, chemical coagulation, air floatation, adsorption, ion exchange processes, flocculation, membrane processes, biological processes, phyto-extraction, extraction, ozone oxidation, and electrochemical processes are generally practiced in effluent treatment plants [5–8].

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\*Corresponding author.

In chemical treatment processes, alum, ferric chloride, lime, and other ferric compounds are commonly used as chemical coagulants for removing pollutants from wastewater. Major drawbacks of this process for the removal of pollutant by chemical processes are expensive and the chemicals present raise the TDS level in the environment. Lime treatment done results in the increase of pH of the water up to a range of 10–12.

Biological processes are one of the best methods for treatment of any wastewater, but are very slow process for organic degradation. The process depends on the initial concentration of BOD. If BOD of wastewater increases during the process, the aeration rate and oxygen requirement increases resulting in higher capital and energy cost. High organic load in wastewater prevents the growth of micro-organisms, and the process needs constant low organic source of wastewater.

Difficulty in ion exchange methods is foul and precipitate can coat on the surface of media. Other problems in the methods are regeneration and activation of the ion exchange media that makes the process complex and longer. The main drawbacks in membrane processes are high cost of membrane, brine disposal, and fouling.

Electrochemical methods recently gained attention as promising alternative to traditional wastewater treatment. It is an easy and efficient process where the coagulating agents are generated *in situ* by applied current to sacrificial anode without addition of any chemical coagulants. At the cathode, a hydrogen gas bubble evolves, which removes the pollutant from the effluent by floatation.

Electrocoagulation (EC) process is employed in a wide spectrum of industrial wastewater such as electroplating wastewater [9], pharmaceutical wastewater [10], petrochemical wastewater [11], municipal wastewater [12], paper and pulp wastewater [13,14], brackish water [15], portable water, oil mill wastewater, nitrite effluent, textile dyes, agro industries wastewater, laundry wastewater, etc [16]. EC is an ideal technique for effluent treatment because of its versatility, energy efficiency, amenability, cost effectiveness [17], and environmental compatibility [18].

Use of aluminium electrodes in various combinations namely (i) aluminium anode and inert cathodes (ii) inert anode and aluminium cathode (iii) aluminium as both anode and cathode was also investigated using EC that had resulted in the removal of natural organic matter from surface water [19]. But, iron EC is cheaper when compared with aluminium EC [20]. These iron ions have a distinct advantage over aluminium ions because of its harmless property and are not

toxic like aluminum ions [21]. During EC process, a series of active iron ionic species generated that destabilize fine dispersed particles and aggregates to form flock in the solution [22,23].

The dissolved contaminants in the effluent are removed by sorption, coagulation, and other processes by applying electric current [24,25]. EC is not only used for removing COD but also used to treat a wide range of pollutants from various synthetic and industrial effluents [26]. The combined EC-Fenton process increased the removal of organic matter compared to EC only [27]. There are limited works as of now on COD removal from MWFs by electrochemical method.

Response surface methodology (RSM) is a mathematical and statistical based technique used to study the performance of EC systems, and for developing, improving, and optimizing the processes. RSM can be used to evaluate the relative significance of several influencing factors even in the presence of complex interactions.

The objective of the present study is to investigate the percentage COD removal from MWFs by batch EC reactor using iron electrodes. The effect of several parameters namely current density, initial pH, and electrolysis time on percentage COD removal and statistical study were performed for EC process.

## 2. Materials and methods

All the chemicals used in the present work were analytically pure and procured from Ranbaxy Fine Chemical Ltd, India. Distilled water used was of ultra pure quality and the same was used for all stock solutions. The wastewater for investigation was collected from a small-scale metal work industry, Chennai–Ambattur, Tamil Nadu, India. The wastewater was characterized for COD, pH, suspended solids, dissolved solids, and color by using the standard methods [28]. The characteristics of the wastewater collected are shown in Table 1. Sample COD was determined with dichromatic open reflux method. Experiments were carried out until the error was less than 4%. The COD removal percentage was estimated by the following relationship.

$$\text{COD removal \%} = \frac{[\text{COD}]_i - [\text{COD}]_f}{[\text{COD}]_i} \times 100 \quad (1)$$

Table 1  
Characteristics of metal working fluids

Characteristics	Value
pH	3.2
Color	Dark green
COD (mgL <sup>-1</sup> )	3,800
Suspended solids (mgL <sup>-1</sup> )	800
Dissolved solids (mgL <sup>-1</sup> )	1,600
Total solids (mgL <sup>-1</sup> )	2,400

where [COD]<sub>i</sub> is the initial COD concentration (mgL<sup>-1</sup>) and [COD]<sub>f</sub> is the final COD concentration (mgL<sup>-1</sup>) obtained after electrolysis of the effluent.

2.1. Experimental design

Design of experiments is an important tool for modeling and analyzing the effect of process parameters on specific responses. The most important aspect of design of experiment lies in the selection of the control factors. The possible operating parameters can be included to identify nonsignificant variables at the earliest opportunity. In the present research work, the RSM has been used to estimate the relation between COD removal percentage and operating conditions namely electrolyte pH, applied charge density, and electrolysis time. Fig. 1 shows the main effect of each parameter on percentage COD removal. Table 2 shows the parameters and the operating ranges covered for the experiments. The pH, current density, and electrolysis time are denoted as uncoded variables as A1, A2, and A3, respectively. The uncoded variables forms are transformed to coded form: a1, a2, and a3 using the following Eq. [29]:

$$a = \frac{A - \frac{(A_{max} - A_{min})}{2}}{\frac{(A_{max} - A_{min})}{2}} \tag{2}$$

Table 2  
The level and range of variables chosen for EC

Factor	Variables	Unit	Range of coded and uncoded variables		
			-1	0	+1
A	pH	–	6	7	8
B	CD	Adm <sup>-2</sup>	2.0	2.2	2.4
C	Time	min	70	90	110

Note: CD: current density.

The Box–Behnken experimental design of RSM has been selected to find the relationship between the response functions and variables using the statistical software tool. In the Box–Behnken mode, a total number of 17 experiments are used to estimate the COD removal percentage from the effluent. The relationship between the analysis of variance (ANOVA) and the variables has been analyzed using RSM. The quality of the fit of this model is expressed by the coefficient of determination R<sup>2</sup>. The fit of the model is confirmed by means of the absolute average deviation (AAD) defined as

$$AAD = \left\{ \frac{\sum_{i=1}^p \left( \frac{|X_{i,exp} - X_{i,pred}|}{X_{i,exp}} \right)}{p} \right\} \times 100 \tag{3}$$

where X<sub>i,exp</sub> and X<sub>i,pred</sub> are defined as the experimental and predicted responses of pollutant load removal percentage and p is defined as the number of experimental runs.

2.2. EC reactor

The experimental setup of batch EC reactor is schematically represented in Fig. 2. Cylindrical reactor made of Perspex with diameter and height of 6 and

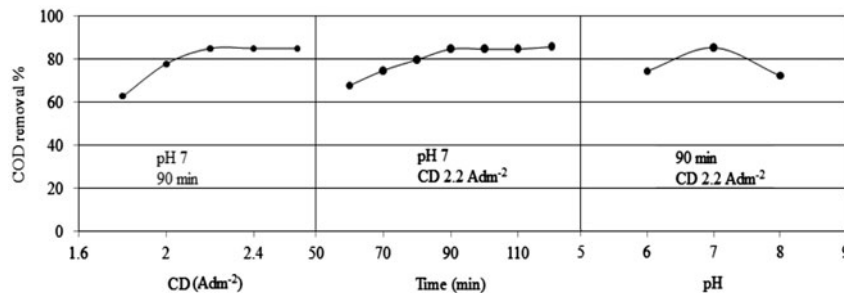


Fig. 1. Main effect plot for the parameters on percentage COD removal.

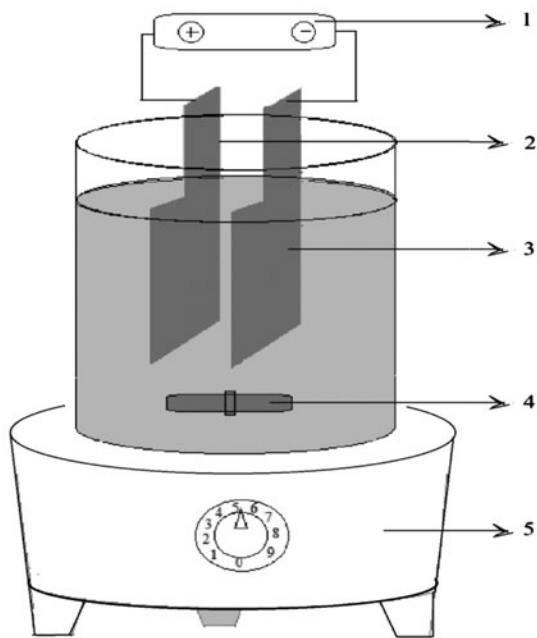


Fig. 2. Experimental setup : (1) DC regulated power, (2) mild steel anodes, (3) stainless steel cathode (4) magnetic pellet and (5) magnetic stirring set-up.

8 cm, respectively. Rectangular flat plates of stainless steel and mild steel (4 cm × 3 cm) were used for cathode and anode, respectively. Two electrodes were placed vertically and parallel into the reactor with an inner gap of 1 cm. The electrode surface was cleaned physically and scraped with sand paper or metal wire brush, followed by 15% HCl acid treatment. Finally, each electrode is washed with distilled water prior to every run. About 300 mL of the wastewater was filled in the reactor which dips 12 cm<sup>2</sup> of the surface of the electrodes. The solution was stirred magnetically at 150 rpm to avoid concentration gradient. The electrodes were connected to DC supply (HIL model 3161) in monopolar mode. All the experiments were performed under potentiostatic conditions at room temperature (28°C). The pH of the solution was adjusted by adding either dilute HCl or NaOH. The samples were collected at regular intervals of time and analyzed for COD.

### 2.3. EC mechanism

During EC process, the coagulants generated *in situ* play an important role in removing the organics from the wastewater. The *in situ* generation of metallic cation flocks takes place at the anode, whereas at the cathode, typically H<sub>2</sub> production

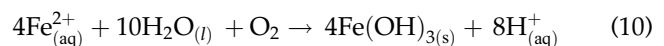
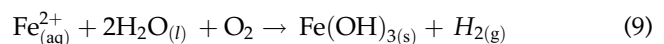
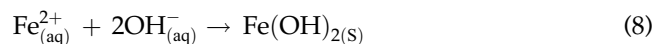
occurs. The generated metallic hydroxides coagulated or get adsorbed with the polluting species present in the effluent. Electrolytic conditions, especially electrolyte pH, applied charge density, and electrolysis time, are influencing the predominance of these mechanisms. Many physiochemical phenomena occur in EC process for the formation of coagulants *in situ*, namely destabilization of the pollutants, particulate suspension, breaking of emulsions, and flocks formation from destabilized pollutants. The EC mechanism can be summarized as follows. When a potential is applied through an external power source, the sacrificial electrode undergoes oxidation as given below [30]:



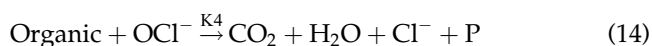
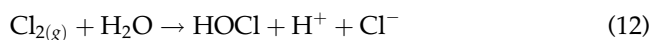
The cathode reactions can be written as:



The bulk reactions for anodes can be written as:



The electrolytic gases generated at the cathode help to float the flocculated particles from the wastewater. The metallic flocks play a major role in the removal of dissolved pollutants in the wastewater by precipitation and/or adsorption mechanism [31]. Other iron hydroxides can also be formed. The monomeric forms of metal hydroxides can get polymerized giving various forms of ferric hydroxo complexes, namely  $\text{Fe}(\text{H}_2\text{O})_6^{3+}$ ,  $\text{Fe}(\text{H}_2\text{O})_5(\text{OH})^{2+}$ ,  $\text{Fe}(\text{H}_2\text{O})_4(\text{OH})_2^{+}$ ,  $\text{Fe}_2(\text{H}_2\text{O})_8(\text{OH})_2^{4+}$ ,  $\text{Fe}_2(\text{H}_2\text{O})_6(\text{OH})_2^{4+}$ ,  $\text{Fe}(\text{OH})_4^{-}$ , etc. The  $\text{Fe}(\text{OH})_n$  formed remains in the aqueous stream as a gelatinous suspension, which can remove the pollutant loads from the effluent [32]. In the electrochemical process, strong oxidizing agents developed *in situ* can take part in destructing the high organic matter to small organic matter present in the waste by oxidizing them:



The hypochlorite ions can oxidize the complex organic molecule to hydrocarbons. This improves the removal percentage of COD in wastewater.

### 3. Results and discussion

#### 3.1. Response surface method

The response of model for percentage removal of COD for the 17 experimental conditions was formulated by the Box–Behnken design as summarized in Table 3. By the design of experimental model, the individual variables ( $a_1, a_2, \dots, a_k$ ) and the response ( $x$ ) can be related as follows [33];

$$x = f(a_1, a_2, a_3, \dots, a_k) + \varepsilon \tag{15}$$

Table 3  
The design of experiment and experimental response for percentage COD removal

Run	Variable factor			Response COD removal%
	A: pH	B: CD	C: Time	
1	8.00	2.40	90.00	83.4
2	8.00	2.00	90.00	68.1
3	7.00	2.00	70.00	75.2
4	6.00	2.20	70.00	64.3
5	7.00	2.20	90.00	85.4
6	6.00	2.20	110.00	76.1
7	7.00	2.40	110.00	85.6
8	7.00	2.00	110.00	78.2
9	6.00	2.40	90.00	75.4
10	7.00	2.20	90.00	85.4
11	7.00	2.40	90.00	85.6
12	8.00	2.20	70.00	74.0
13	7.00	2.40	70.00	83.0
14	6.00	2.00	90.00	67.1
15	8.00	2.20	110.00	79.0
16	6.00	2.20	110.00	76.8
17	7.00	2.00	70.00	75.0

Note: CD: current density.

RSM postulates the functional relationship between the response ( $x$ ) and the independent variables. The first- and second-order RSM can be given as:

$$x = \beta_0 + \sum_{i=1}^k \beta_i a_i + \varepsilon \tag{16}$$

$$x = \beta_0 + \sum_{i=1}^k \beta_i a_i + \sum_{i=1}^k \beta_{ii} a_i^2 + \sum_{i=1}^{k-1} \sum_{j=2}^k \beta_{ij} a_i a_j + \varepsilon \tag{17}$$

The response ( $x$ ) can be mathematically related to the variables as

$$x = \beta_0 + \beta_1 a_1 + \beta_2 a_2 + \beta_3 a_3 + \beta_{11} a_1^2 + \beta_{22} a_2^2 + \beta_{33} a_3^2 + \beta_{12} a_1 a_2 + \beta_{13} a_1 a_3 + \beta_{23} a_2 a_3 \tag{18}$$

and

$$\beta = 1, 2, 3, 4, \dots, k \tag{19}$$

where  $\beta$  and  $k$  are denoted as regression coefficients and variables, respectively.

The Box–Behnken method was used to recover the relationship between the response and variables functions (Table 2). A class of complete factorial designs for the estimation of parameters in a second-order model has been developed (Table 3) using Design-expert V6. The study was focused on estimating the influence of individual parameters on percentage COD removal.

#### 3.2. Effect of various parameters on percentage COD removal

The inferences obtained from the response surfaces to calculate the maximum percentage COD removal with respect to each variable and each variable’s effect on percentage COD removal are discussed below.

##### 3.2.1. Effect of current density and electrolyte pH

Experiments were carried out according to the experimental condition designed by RSM and the results are presented in both surface and contour plots. The combined effect of electrolyte pH and applied charge on percentage COD removal is given in Fig. 3. It has been well established that electrolyte pH is an important parameter and has considerable effect on EC process. From the results it is confirmed that the COD removal rate increases with increase in



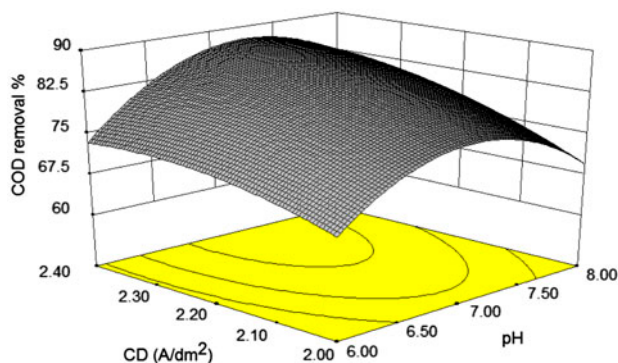


Fig. 3. Combined effects of current density and pH on percentage COD removal.

the electrolyte pH. Further increase in pH from optimal pH has no significant effect on percentage COD removal. The pH increase in EC process was attributed to the generation of hydrogen gas at the cathode electrode. Accordingly, the influent pH was considered as one on the key factor while designing the experimental runs. Experiments were performed under controlled pH conditions by adding NaOH or HCl. From Fig. 4, it is noticed that the percentage COD removal increases from acid to neutral with no improvement on COD removal rate beyond pH 7. This can be explained that the solubility of  $\text{Fe}(\text{OH})_3$  increased beyond the electrolyte pH 7 resulting in the formation of soluble  $\text{Fe}(\text{OH})_4^-$  which does not contribute to the organic removal percentage [34].

### 3.2.2. Effect of electrolysis time and current density

Fig. 5 shows the combined effect of electrolysis time and current density ( $\text{CD A/dm}^3$ ) on percentage COD removal. It can be ascertained from Fig. 5 that the percentage COD removal increased with electrolysis

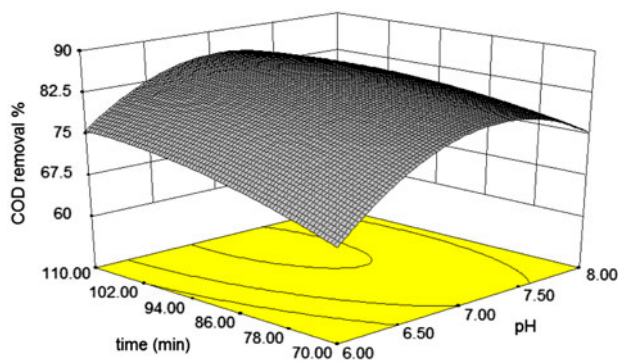


Fig. 4. Combined effects of current density and time on percentage COD removal.

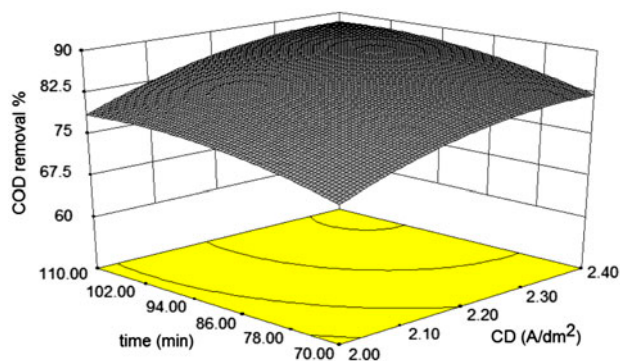


Fig. 5. Combined effects of electrolysis time and current density on percentage COD removal.

time and applied charge density. This can be attributed to the fact that at high current densities, the extent of anodic dissolution increases which increases the hydroxo cationic complexes resulting in increase in the percentage COD removal [35,36]. From Fig. 5, it is observed that the percentage COD removal attains marginal beyond electrolysis time of 90 min and CD of  $2.2 \text{ Adm}^{-2}$ . Further increase in operating parameters of electrolysis time and CD shows no significant effect on the percentage COD removal. This is because the  $\text{Fe}(\text{OH})_3$  flocks are generated *in situ* once the charge is applied and the generated flocks adsorb the organic molecules present in the effluent. This results in the reduction of percentage COD with electrolysis time and attains marginal beyond the optimal condition.

### 3.3. Analysis of variance

Table 4 shows the result of ANOVA performed to estimate the significant effect of variables on the percentage COD removal. From Table 4 it is noticed that the  $F$ -statistics values for the regressions are higher. The large  $F$ -values for the percentage COD removal indicate that the response fits more favorable for the regression model. The associated  $p$ -value is used to determine whether the  $F$ -values are large enough to indicate statistical significance. If  $p$ -values are lower than 0.05, it indicates that the model is statistically significant for the percentage COD removal [25]. The residual error term in the ANOVA table shows the amount of variation in the response data left unexplained by the model. Model fitting with software (Design-expert) suggested that a quadratic model provides the best fit, and the model was found to have significant lack of fit (0.0017). From Table 4, ANOVA indicated that only five of the nine model terms are significant. The reduced quadratic model for

Table 4  
ANOVA of the second-order polynomial equation for the percentage COD removal

Source	Coefficient estimate	Sum of squared	Standard error	DF	Mean square	F-value	p-Value prob > F	Remark
Model	85.02	692.71	1.50	9	76.97	15.65	0.0008	Significant
intercept								
A-pH	2.57	57.03	0.76	1	57.03	11.60	0.0114	Significant
B-CD	4.50	192.91	0.72	1	192.91	39.22	0.0004	Significant
C-Time	2.75	69.63	0.73	1	69.63	14.16	0.0071	Significant
AB	1.77	12.60	1.11	1	12.60	2.56	0.1535	
AC	-1.93	17.44	1.03	1	17.44	3.55	0.1017	
BC	0.24	0.27	1.03	1	0.27	0.055	0.8220	
A <sup>2</sup>	-9.31	317.08	1.16	1	317.08	64.47	<0.0001	Significant
B <sup>2</sup>	-2.43	20.05	1.20	1	20.05	4.08	0.0832	
C <sup>2</sup>	-1.95	13.96	1.15	1	13.96	2.84	0.1359	
Residual		34.43		7	4.92			
Lack of fit		34.16		3	8.54	96.69	0.0017	Significant
Pure error		0.27		4	0.088			
Cor total		727.14		16				

Note: Cortot: Correlationtotal.

percentage COD removal with process parameter such as electrolyte pH (A), applied charge density (B), and electrolysis time (C) in coded factors is given in Eq. (20)

$$\begin{aligned} \text{COD removal \%} = & 85.02 + 2.57A + 4.50B + 2.75C \\ & + 1.77AB - 1.93AC + 0.24BC \\ & - 9.31A^2 - 2.43B^2 - 1.95C^2 \end{aligned} \quad (20)$$

The multiple regression equation for actual factors is given in the Eq. (21)

$$\begin{aligned} \text{COD removal \%} = & -696.45 + 122.07A + 221.99B \\ & + 1.55C + 8.87AB - 0.09AC \\ & + 0.05BC - 9.31A^2 - 60.67B^2 \\ & - 4.86C^2 \end{aligned} \quad (21)$$

Table 5  
Adequacy of the models tested for the percentage COD removal

Sequential model sum of squares						
Source	Sum of squares	DF	Mean square	F-value	Prob > F	Remark
Mean	1.021E + 5	1	1.021E + 5			
Linear	348.40	3	116.13	3.99	0.0324	Suggested
2FI	24.45	3	8.15	0.23	0.8734	
Quadratic	319.86	3	106.62	21.38	0.0006	Suggested
Cubic	34.16	4	8.54	96.69	0.0017	Aliased
Residual	0.26	3	0.088			
Total	1.028E + 5	17	6049.02			
Model summary statistics						
Source	SD	R <sup>2</sup>	Adj-R <sup>2</sup>	Pred-R <sup>2</sup>	PRESS	Remark
Linear	5.40	0.4791	0.3589	0.1417	624.13	
2FI	5.95	0.5128	0.2204	-0.4854	1080.07	
Quadratic	2.22	0.9527	0.8918	0.5503	327.03	Suggested
Cubic	0.30	0.9996	0.9981	-	-	Aliased

3.4. Adequacy of the models for the percentage COD removal

Linear, interactive, quadratic, and cubic models were fitted to the experimental data to obtain the regression equations. To decide about the adequacy of various models to represent removal percentage by two different tests, namely sequential model sum of squares and model summary statistics were carried out and the results are given in Table 5. The cubic model was found to be aliased for percentage COD removal. The model adequacies were checked with  $R^2$  and  $adj-R^2$ . A higher value of coefficient of determination  $R^2$  (0.9527) for percentage COD removal shows that the quadratic model follows the response successfully.

Data were also analyzed to check the normality of the removal percentage. A normal probability plot or dot diagrams for percentage COD removal are shown in Fig. 6. The data points on this plot lay reasonably close to a straight line for percentage COD removal and hence the operating parameters of the analysis are well satisfied.

Fig. 7(a) shows the relationship between the actual and predicted values for percentage COD removal from plating effluent by EC process using mild steel electrodes. It can be ascertained from Fig. 7(a) that the developed model is adequate because percentage COD removal for the prediction of each response is minimum, and the data points lie close to the diagonal line. Error analysis was done to predict the error for actual and predicted data, which gives an  $R^2$  value of 0.9391. Hence, the actual values are highly in line with the predicted values (Fig. 7(b)).

Fig. 8 shows the graphical plot of studentized residuals versus predicted for percentage COD

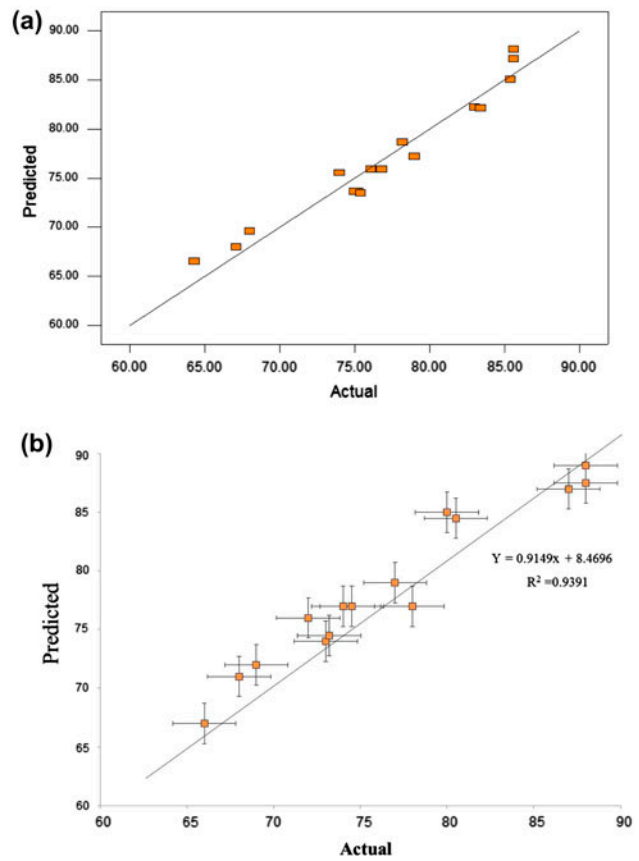


Fig. 7. (a) Scatter diagram of predicted response vs. actual response on percentage COD removal and (b) Error analysis for scatter diagram of predicted response vs. actual response.

removal from the MWFs. The general impression is that the plot should be scatter diagram, suggesting that the variance of original observation is constant for

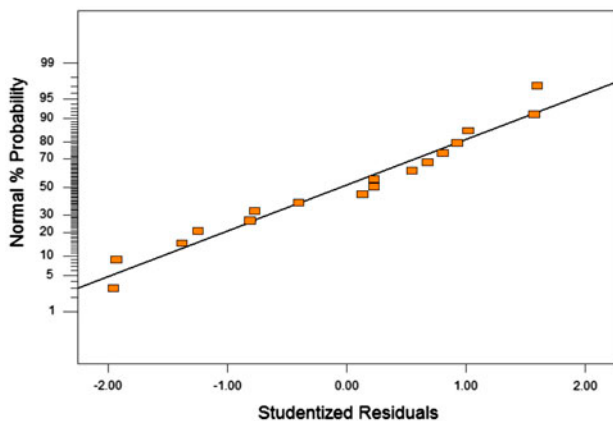


Fig. 6. Normal % probability vs. residual error on percentage COD removal.

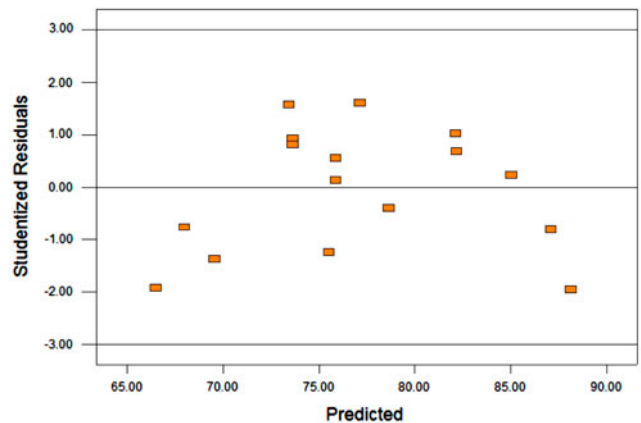


Fig. 8. Predicted percentage COD removal and studentized residual plots.



all values of percentage COD removal (response). Usually, it is important to confirm the model fitness to make sure that it gives sufficient approximation to the actual test.

Interaction plots between operating variables on percentage COD removal are given in Figs. 9–11, respectively. It is evident that percentage COD removal is greatly affected by the interaction of variables. Figs. 9–11 show the interaction between current density and time, pH and time, and pH and current density, respectively. This indicates the difference in apparent percentage COD removal at different levels of pH, time, and current density. Thus, the difference on percentage COD removal results mainly due to the interaction between pH, time, and current density.

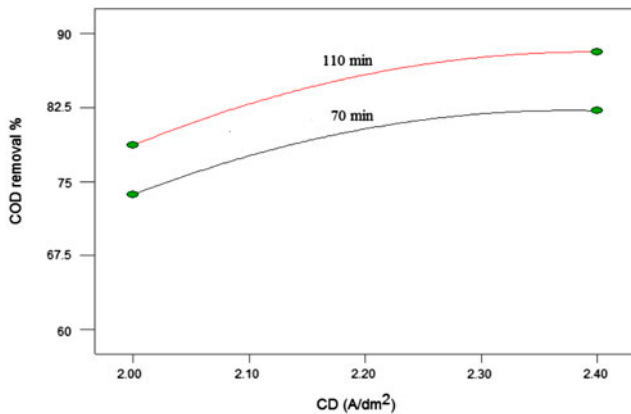


Fig. 9. Interaction effect between current density and time on percentage COD removal.

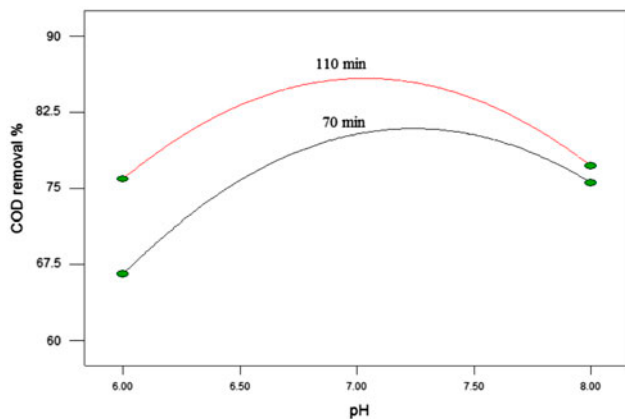


Fig. 10. Interaction effect between pH and time on percentage COD removal.

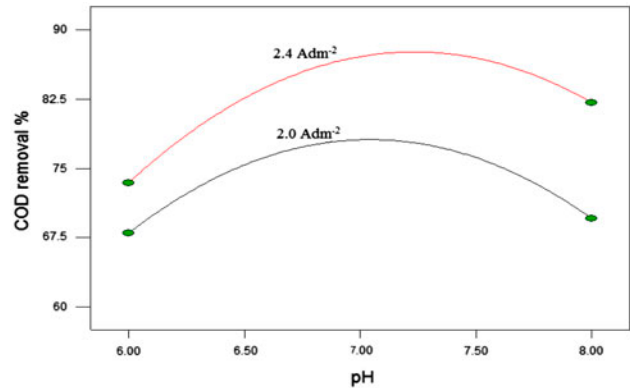


Fig. 11. Interaction effect between pH and current density on percentage COD removal.

#### 4. Conclusions

In the present experimental study, the performance of EC treatment of MWFs was investigated focusing on the effect of operating variables such as electrolyte pH, applied current density, and electrolysis time. From the critical examination it was observed that the percentage COD removal is significantly influenced by the operating variables. The response data were analyzed using ANOVA and RSM through which individual and combined variable effects on percentage COD removal were studied. Three-level three-factor Box–Behnken design was applied in the present investigation. Regression equations have been developed for percentage COD removal and solved by statistical software tool Design-expert. Analysis showed a high  $R^2$  value (0.9527) for the percentage COD removal and satisfactory prediction for second-order regression model.

#### Abbreviations

COD	—	chemical oxygen demand
ANOVA	—	analysis of variance
MWFs	—	metal-working fluids
BI	—	biodegradability index
BOD	—	biochemical oxygen demand
TDS	—	total dissolved solids
EC	—	electro coagulation
NOM	—	natural organic matter
RSM	—	response surface methodology
AAD	—	absolute average deviation
DC	—	direct current

#### Notations

$f$	—	the unknown function of response
$k$	—	the number of independent variables

$p$	—	the number of experimental runs
$a_1, \dots, a_k$	—	coded factor
$A$	—	uncoded variable
$X$	—	the response of the system
$\varepsilon$	—	the statistical error
$\beta_0, \beta_i, \beta_{ii}, \beta_{ij}$	—	regression coefficients

## References

- [1] M. Uğurlu, M.H. Karaoğlu, İ. Kula, Experimental investigation of chemical oxygen demand, lignin and phenol removal from paper mill effluents using three-phase three-dimensional electrode reactor, *Pol. Environ. Stud.* 15(4) (2006) 647–654.
- [2] E. Cséfalvay, V. Pauer, P. Mizsey, Recovery of copper from process waters by nanofiltration and reverse osmosis, *Desalination* 240 (2009) 132–142.
- [3] A. Muszynski, M. Zaleska-Radziwiłł, M. Łebkowska, D. Nowak, Biological and electrochemical treatment of used metalworking fluids: A toxicity-reduction evaluation, *Arch. Environ. Contam. Toxicol.* 52 (2007) 483–488.
- [4] M. Visa, R.A. Carcel, L. Andronic, A. Duta, Advanced treatment of wastewater with methyl orange and heavy metals on TiO<sub>2</sub>, fly ash and their mixtures, *Catal. Today* 144 (2009) 137–142.
- [5] C.P. Nansu-Njiki, S.R. Tchamango, P.C. Ngom, A. Darchen, E. Ngameni, Mercury (II) removal from water by electrocoagulation using aluminium and iron electrodes, *J. Hazard. Mater.* 168 (2009) 1430–1436.
- [6] N. Meunier, P. Drogui, G. Mercier, J. Franc, O. Blais, Treatment of metal loaded soil leachates by electrocoagulation, *Sep. Purif. Technol.* 67 (2009) 110–116.
- [7] H.R. Mortaheb, H. Kosuge, B. Mokhtarani, M.H. Amini, H.R. Banihashemi, Study on removal of cadmium from wastewater by emulsion liquid membrane, *J. Hazard. Mater. B* 165 (2009) 630–636.
- [8] D. Reddithota, A. Yerramilli, R.J. Krupadam, Electrocoagulation: A cleaner method for treatment of Cr(VI) from electroplating industrial effluents, *Indian J. Chem. Technol.* 14 (2007) 240–245.
- [9] J. Yi-zhong, Z. Yue-feng, L. Wei, Experimental study on micro-electrolysis technology for pharmaceutical wastewater treatment, *J. Zhejiang Univ. Sci.* 3 (2002) 401–404.
- [10] A. Dimoglo, H.Y. Akbulut, F. Cihan, M. Karpuzcu, Petrochemical wastewater treatment by means of clean electrochemical technologies, *Clean Technol. Environ. Policy* 6 (2004) 288–295.
- [11] A.A. Bukhari, Investigation of the electro-coagulation treatment process for the removal of total suspended solids and turbidity from municipal wastewater, *Bioresour. Technol.* 99 (2008) 914–921.
- [12] P.A. Soloman, C. Ahmed Basha, M. Velan, N. Balasubramanian, Electrochemical degradation of pulp and paper industry waste-water, *J. Chem. Technol. Biotechnol.* 84 (2009) 1303–1313.
- [13] N. Daneshvar, H. Ashassi, R. Rakhshaei, Pretreatment of brackish water using DC-electrocoagulation—method and optimization, *Iran. J. Chem. Chem. Eng.* 21 (2002) 13–20.
- [14] M. Vepsäläinen, J. Selin, P. Rantala, M. Pulliainen, H. Särkkä, K. Kuhmonen, A. Bhatnagar, M. Sillanpää, Precipitation of dissolved sulphide in pulp and paper mill wastewater by electrocoagulation, *Environ. Technol.* 32(12) (2011) 1393–1400.
- [15] P.A. Soloman, C. Ahmed Basha, M. Velan, N. Balasubramanian, P. Marimuthu, Augmentation of biodegradability of pulp and paper industry wastewater by electrochemical pre-treatment and optimization by RSM, *Sep. Purif. Technol.* 69 (2009) 109–117.
- [16] K. Jüttner, U. Galla, H. Schmieder, Electrochemical approaches to environmental problems in the process industry, *Electrochim. Acta* 45 (2000) 2575–2594.
- [17] S. Yi, Y. Ma, X. Wang, Y. Jia, Green chemistry: Pre-treatment of seawater by a one-step electrochemical method, *Desalination* 239 (2009) 247–256.
- [18] S. Vasudevan, J. Jayaraj, J. Lakshmi, G. Sozhan, Removal of iron from drinking water by electrocoagulation: Adsorption and kinetics studies, *Korean J. Chem. Eng.* 26(4) (2009) 1058–1064.
- [19] I. Kabdasli, T. Arslan, I. Arslan-Alaton, T. Olmez-Hanci, O. Tünay, Organic matter and heavy metal removals from complexed metal plating effluent by the combined electrocoagulation/Fenton process, *Water Sci. Technol.* 61 (2010) 2617–2624.
- [20] E. Lacasa, P. Cañizares, C. Sáez, F. Martínez, M.A. Rodrigo, Modelling and cost evaluation of electrocoagulation processes for the removal of anions from water, *Sep. Purif. Technol.* 107 (2013) 219–227.
- [21] M. Yousuf, A. Mollah, Robert Schennach, Jose R. Parga, David L. Cocke, Electrocoagulation (EC)—Science and applications, *J. Hazard. Mater. B* 84 (2001) 29–41.
- [22] M.F. Ni'am, F. Othman, J. Sohaili, Z. Fauzia, Electrocoagulation technique in enhancing COD and suspended solids removal to improve wastewater quality, *Water Sci. Technol.* 56 (2007) 47–53.
- [23] V.P. Malkin, Industrial ecology-electrolytic effluent treatment, *Chem. Pet. Eng.* 39 (2003) 1–2.
- [24] M.M. Emamjomeh, M. Sivakumar, Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes, *J. Environ. Manage.* 90 (2009) 1663–1679.
- [25] M. Kumar, F.I.A. Ponselvan, J. RamMalviya, V.C. Srivastava, I.D. Mall, Treatment of bio-digester effluent by electrocoagulation using iron electrodes, *J. Hazard. Mater. B* 165 (2009) 345–352.
- [26] P.K. Holt, G.W. Barton, C.A. Mitchell, The future for electrocoagulation as a localized water treatment technology, *Chemosphere* 59 (2005) 355–367.
- [27] M. Vepsäläinen, M. Pulliainen, M. Sillanpää, Effect of electrochemical cell structure on natural organic matter (NOM) removal from surface water through electrocoagulation (EC), *Sep. Purif. Technol.* 99 (2012) 20–27.
- [28] APHA, Standard Methods for Examination of Water and Wastewater, 17th ed., American Public Health Association, Washington, DC, 1989.
- [29] B. Deniz, I.H. Boyaci, Modeling optimization I: Usability of response surface methodology, *J. Food Eng.* 78 (2007) 836–845.
- [30] N. Balasubramanian, Toshinori Kojima, C. Srinivasakannan, Arsenic removal through electrocoagulation: Kinetic and statistical modelling, *Chem. Eng. J.* 155 (2009) 76–82.

- [31] K.S. Parama Kalyani, N. Balasubramanian, C. Srinivasakannan, Decolorization and COD reduction of paper industrial effluent using electro-coagulation, *Chem. Eng. J.* 151 (2009) 97–104.
- [32] K. Chithra, N. Balasubramanian, Modeling electrocoagulation through adsorption kinetics, *J. Model. Simul. Syst.* 2 (2010) 124–130.
- [33] K. Rajeshwar, J. Ibanez, *Environmental Electrochemistry Fundamentals and Applications in Pollution Abatement*, Academic Press, London, 1997.
- [34] R. Saravanathamizhan, N. Mohan, N. Balasubramanian, V. Ramamurthi, C. Ahmed Basha, Evaluation of electro oxidation of textile effluent using response surface technique, *Clean Soil Air Water* 35 (2007) 355–361.
- [35] K. Chithra, N. Balasubramanian, Modeling electrocoagulation through adsorption kinetics, *Chem. Eng. J.* 155 (2009) 76–82.
- [36] N. Balasubramanian, K. Madhavan, Arsenic removal from industrial effluent through electrocoagulation, *Chem. Eng. J.* 24 (2001) 519–521.