



Electrochemical treatment of acrylic dye-bearing textile wastewater: optimization of operating parameters

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ABSTRACT

The aim of the present study is to investigate the chemical oxygen demand (COD) and color removal efficiency and specific energy consumption during batch electrochemical treatment of synthetic textile wastewater having an initial COD of 3200 mg/L and containing yellow brown 2GL (basic orange 30) acrylic dye. Aluminum (Al) and stainless steel (SS) electrodes have been used as electrodes during the study. A Box–Behnken experimental design has been employed to evaluate the individual and interactive effects of four independent parameters namely: initial pH (pH_0): 4–10, current density (j): 27.78–138.89 A/m², NaCl concentration (w): 0–2 g/L and electrolysis time (t): 10–130 min on the COD and color removal efficiency and specific energy consumed. Pareto analysis of variance showed a high coefficient of determination (R^2) value for COD (0.8815, 0.8995), color (0.9494, 0.8243), and specific energy consumption (0.9331, 0.8805) for Al and SS electrodes, respectively, between the experimental values and the predicted values by a second-order regression model. Maximum COD and color removal efficiencies and minimum specific energy consumed at optimum conditions using Al electrode were 46.87%, 98.61%, and 25.04 kWh/kg COD removed, respectively. Respective values for treatment with SS electrodes were 54.37%, 83.89%, and 30.19 kWh/kg COD removed, respectively.

Keywords: Electrocoagulation; Stainless steel; Aluminum electrode; Textile wastewater; Acrylic dyes

1. Introduction

Textile manufacturing is one of the largest industrial producers of wastewater. It produces \approx 125–150 L of wastewater per kg of textile products. The wastewater from textile processing contains processing bath residue such as color residues, heavy metal ions, and electrolytes generated during preparation, dyeing, finishing, slashing and other operations. Textile waste-

water is well known with its high chemical oxygen demand (COD), strong color, and large amount of suspended solids, variable pH, salt content and high temperature [1–5]. Textile industry wastewater can cause severe pollution if not properly treated before discharge to the water bodies [6].

Among all the treatments proposed in the literature, electrochemical (EC) process is one of the newer methods for the treatment of industrial wastewaters. EC process is a promising technology that can be

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used for the removal of both color and colloidal particles [7–10]. In this method, sacrificial anodes dissolve to produce active coagulant precursors (usually aluminum or iron cations) into solution. Additionally, electrolytic reactions evolve gases (usually as hydrogen bubbles) at the cathode that can enhance the process; this effect is known as electroflotation [11–13]. As compared to traditional chemical method, EC method requires less coagulant and compact equipment. Thus, EC setup can be put in limited space. Amount of sludge formed in EC method is less and that it is easily settleable. Moreover, dosing can be controlled easily by adjustment of current only [14,15]. EC technology has also been recently demonstrated as one of the options for treatment of dye-containing wastewaters [16–27].

Acrylic/cationic dyestuff is used for dyeing acrylic fiber which has been widely used in the manufacturing of blankets, carpets, knitting goods, decorative fabrics, sarees, etc. Acrylic (cationic) dyes, which are present in the textile wastewaters depending upon the type of textile product being produced, can easily interact with negatively charged cells membrane surfaces in living organisms and can enter into cells and concentrate in the cytoplasm. Therefore, it is very essential to develop technologies that can remove cationic dyes from industrial effluents before it is discarded into water bodies [28]. However, only few studies are reported in the literature for the EC treatment of acrylic dyes [28,29]. Yellow brown 2GL (basic orange 30) acrylic dye is used for the dyeing of blankets, carpets, etc. Based on the discussion of the authors with the textile industries using this dye, it was found that though color removal can be obtained by conventional treatment with coagulants such as lime and alum in the industries; however, COD removal was found to be difficult.

Based on this practical problem, the aim of the present work was to evaluate the performance of an EC reactor having aluminum (Al) and stainless steel (SS) electrodes for the treatment of synthetic textile wastewater containing yellow brown 2GL (basic orange 30) acrylic dye.

2. Materials and methods

2.1. Chemicals and materials

All the chemicals used in the study were of analytical reagent grade. Potassium dichromate, sulfuric acid, hydrochloric acid, and NaOH were obtained from Ranbaxy Chemicals Ltd., New Delhi, India. Silver sulfate and mercuric sulfate were obtained from Himedia Laboratories, Mumbai, India. Yellow brown 2GL (basic orange 30) acrylic dye was obtained from Yogesh

Dyestuff Products Pvt., Ltd, Mumbai. The specifications of yellow brown 2GL (basic orange 30) acrylic dye are as follows: molecular formula: $C_{19}H_{24}Cl_2N_5O_2Cl$; molecular weight: 460.78 g/mol; color index: 110,855; light fastness: 6–7; wash fastness: 4–5; dyestuff saturation factor: 0.63 and combination factor: 3.0.

2.2. Synthetic wastewater and its characteristics

Synthetic textile wastewater was made as per the method given in the literature [30]. Simulated textile wastewater was made by mixing following compounds: carboxymethyl cellulose (150 mg/L), starch (1,500 mg/L), acetic acid (500 mg/L), yellow brown 2GL (basic orange 30) dye (250 mg/L), NaOH (660 mg/L), H_2SO_4 (357 mg/L), Na_2CO_3 (1,000 mg/L), $NaHCO_3$ (2000 mg/L), and glucose (2062.5 mg/L). The wastewater was characterized for COD, pH, total solids, suspended solids, dissolved solids, turbidity, conductivity, and color by using the standard methods [31]. The characteristics of simulated wastewater are shown in Table 1.

2.3. Analytical

The analysis of different parameters was carried out using different equipment/instruments. pH and conductivity was measured using a multiparameter digital meter (HACH, USA). Color was measured by a colorimeter (Aqualytic, Germany). The COD of the solution was determined by using a COD/TOC analysis system DR 5000 (HACH, USA).

2.4. Experimental set-up

Circular cross-section lab-scale glass batch reactor of 1 L volume was used for the EC experiments. Al and SS plates of thickness 1.5 and 2.5 mm, respectively, having equal dimension of 9 cm × 10 cm with effective electrode surface area of 180 cm² were used as

Table 1
Characteristics of wastewater used for electrochemical treatment

Characteristics	Value
COD (mg/L)	3,200
Color (platinum cobalt unit)	7,200
pH	9.5
Conductivity (mS/cm)	5.3
Turbidity (NTU)	180
Total solids (mg/L)	7,090
Suspended solids (mg/L)	1,190
Dissolved solids (mg/L)	5,900

electrodes. Two pairs of Al or SS electrodes with 1 cm gap in between the electrodes were used in all experimental runs. Magnetic stirrer was used to agitate the solution and for easy stirring, bottom of electrodes was kept 5 cm above the bottom of the EC reactor. All test runs were performed at $30 \pm 2^\circ\text{C}$. In each run, 0.9 L of wastewater was fed into the EC reactor. The initial pH of the solution was adjusted to the desired value by sodium hydroxide (0.1 N) or HCl (0.1 N) aliquots. The initial conductivity was adjusted by adding sodium chloride. Experiments were conducted for four plate configurations by varying the voltage in the range of 1–15 V. To optimize parameters for reactor performance, the experiments were carried out at varying current density (j : 27.78–138.89 A/m²), pH (4–10), NaCl concentration (w : 0–2 g/L), and electrolysis time (t : 10–130 min). At the end of the EC process, sample was centrifuged, filtered, and analyzed for pH, COD, and color. Iron and Al concentration in solution was determined for optimal conditions. The electrode plates were cleaned manually by abrasion with 15% HCl for cleaning followed by washing with distilled water prior to their reuse. The energy consumption for the removal of 1 kg of COD was calculated in kWh. The average cell voltage during the electrolysis was used for calculating the energy consumption [32].

2.5. Experimental design

In the present study, the 4-factor and 5-level Box–Behnken (BB) experimental design has been applied to investigate the effect of various variables. Percent COD removal, percent color removal, and specific energy consumption have been taken as three responses of the system, while four process parameters, namely, current density (j): 27.78–138.89 A/m², pH: 4–10, time (t): 10–130 min, and NaCl (w): 0–2 g/L are variable input parameters. The factor levels were coded as –2 (low), 0 (central point or middle), and 2 (high) [33]. The variables and their levels are given in Table 2.

A total of 27 experiments have been employed in this work. The actual experimental design matrices for

the treatment with Al and SS electrodes are given in Tables 3 and 4, respectively. The results were analyzed using the coefficient of determination (R^2), pareto analysis of variance (ANOVA), and statistical and response plots. A nonlinear regression method was used to fit the second-order polynomial (Eq. (1)) to the experimental data and to identify the relevant model terms. Considering all the linear terms, square terms, and linear by linear interaction items, the quadratic response model can be described as

$$Y = \beta_0 + \sum \beta_i x_i + \sum \beta_{ii} x_{ii}^2 + \sum \beta_{ij} x_i x_j + \varepsilon \quad (1)$$

where β_0 is the offset term, β_i is the slope or linear effect of the input factor x_i , β_{ii} is the quadratic effect of input factor x_i , and β_{ij} is the linear by linear interaction effect between the input factor x_i and x_j [34,35].

3. Results and discussion

3.1. Box–Behnken analysis

The most important parameters which affect the efficiency of COD and color removal of synthetic textile wastewater by EC method are j , pH, t , and w . In order to study the combined effect of these factors, experiments were performed for different combinations of the physical parameters using statistically designed experiments. The range of values for the input variables are given in Table 2. The results of the COD, color, and specific energy consumed (kWh/kg COD removed) of synthetic textile wastewater onto electrocoagulation were measured according to design matrix and the measured responses are listed in Tables 3 and 4, for Al and SS electrodes, respectively.

By using multiple regression analysis, the responses (COD, color removal, and specific energy (kWh/kg COD removed)) were correlated with the four design factors using the second-order polynomial (Eq. (1)). The quadratic regression model for COD removal (Y_1 , %), color removal (Y_2 , %), and specific energy consumed (Y_3 , %) by Al electrode in terms of coded factors are given as follows:

Table 2
Process parameters and their levels for EC treatment of textile wastewater using BB design

Variable, unit	Factors	Level				
		–2	–1	0	1	2
Current density, j (A/m ²)	X1	27.78	55.56	83.33	111.11	138.89
NaCl concentration, w (g/L)	X2	0	0.5	1	1.5	2
Time, t (min)	X3	10	40	70	100	130
pH	X4	4	5.5	7	8.5	10

Table 3
Full factorial design used for the EC treatment of textile wastewater by Al electrodes

Std	<i>j</i> (X1)	<i>w</i> (X2)	<i>t</i> (X3)	pH (X4)	% COD reduction		% Color reduction		Specific energy, kWh/kg COD removed	
					Y_{exp} (%)	Y_{pre} (%)	Y_{exp} (%)	Y_{pre} (%)	Y_{exp} (%)	Y_{pre} (%)
1	138.89	1	70	7	22.42	24.55	70.41	65.28	25.58	26.03
2	83.33	1	70	7	17.19	16.83	48.35	48.21	12.16	12.11
3	111.11	0.5	120	8.5	37.42	31.81	81.04	81.66	21.33	20.17
4	111.11	1.5	40	8.5	11.66	14.92	22.74	30.00	18.73	16.47
5	111.11	1.5	120	8.5	36.81	36.23	96.71	98.02	17.99	19.76
6	111.11	1.5	120	5.5	38.18	38.58	98.9	102.04	15.96	15.90
7	55.56	1.5	40	5.5	14.2	21.19	16.03	19.87	4.06	4.02
8	83.33	0	70	7	0	6.02	0	10.59	0	4.07
9	83.33	1	10	7	13.54	13.91	15.19	4.03	1.99	4.59
10	83.33	1	70	7	17.19	16.83	48.35	48.21	12.16	12.11
11	83.33	1	70	10	22.03	27.16	81.52	75.54	7.73	10.00
12	55.56	1.5	40	8.5	18.78	14.94	25.46	35.70	2.75	3.16
13	55.56	1.5	120	8.5	22.53	22.91	66.93	71.56	6.84	6.82
14	83.33	1	70	7	17.19	16.83	48.35	48.21	12.16	12.11
15	55.56	0.5	40	8.5	15.55	14.17	35.62	34.07	4.26	2.01
16	55.56	0.5	120	5.5	27.02	23.75	54.02	52.28	7.26	8.12
17	55.56	0.5	120	8.5	17.78	17.96	62.53	56.93	12.1	10.23
18	55.56	1.5	120	5.5	27.84	29.12	65.22	65.71	6.52	6.87
19	27.78	1	70	7	18.38	17.92	46.17	44.42	5.12	6.59
20	83.33	1	70	7	17.19	16.83	48.35	48.21	12.16	12.11
21	83.33	1	130	7	26.04	29.47	77.72	82.85	14.78	14.37
22	111.11	1.5	40	5.5	18.18	17.32	13.15	24.04	11.17	13.43
23	83.33	2	70	7	14.95	10.60	41.59	24.13	10.28	8.12
24	111.11	0.5	40	8.5	19.02	14.68	24.72	26.64	13.38	12.32
25	111.11	0.5	40	5.5	17.27	16.65	25.15	21.89	8.94	7.11
26	111.11	0.5	120	5.5	31.32	33.73	91.62	86.88	14.62	14.15
27	83.33	1	70	4	38.81	35.35	63.34	62.44	5.4	5.05
28	83.33	1	70	7	17.19	16.83	48.35	48.21	12.16	12.11
29	55.56	0.5	40	5.5	21.08	20.00	19.39	19.44	2.99	0.71
30	83.33	1	70	7	17.19	16.83	48.35	48.21	12.16	12.11

$$\begin{aligned}
 Y_1 = & 16.83 + 3.32 j + 2.29 w + 7.78 t - 4.10 \text{ pH} \\
 & + 4.40 j^2 - 8.53 w^2 + 4.86 t^2 + 14.42 \text{ pH}^2 \\
 & - 0.53 j \times w + 10.0 j \times t + 3.86 j \times \text{pH} \\
 & + 3.14 w \times t - 0.43 w \times \text{pH} + 0.036 t \times \text{pH} \quad (2)
 \end{aligned}$$

$$\begin{aligned}
 Y_2 = & 48.21 + 10.43 j + 6.77 w + 39.41 t + 6.55 \text{ pH} \\
 & + 6.64 j^2 - 30.85 w^2 - 4.77 t^2 + 20.78 \text{ pH}^2 \\
 & + 1.72 j \times w + 24.12 j \times t - 9.87 j \times \text{pH} \\
 & + 9.75 w \times t + 1.20 w \times \text{pH} - 7.48 t \times \text{pH} \quad (3)
 \end{aligned}$$

$$\begin{aligned}
 Y_3 = & 12.11 + 9.72 j + 2.02 w + 4.89 t + 2.48 \text{ pH} \\
 & + 4.19 j^2 - 6.02 w^2 - 2.64 t^2 - 4.59 \text{ pH}^2 \\
 & + 3.00 j \times w - 0.28 j \times t + 3.91 j \times \text{pH} \\
 & - 3.42 w \times t - 2.17 w \times \text{pH} + 0.61 t \times \text{pH} \quad (4)
 \end{aligned}$$

The quadratic regression model for COD removal (Y_1 , %), color removal (Y_2 , %), and specific energy consumed (Y_3 , %) by SS electrode in terms of coded factors are given as follows:

Table 4
Full factorial design used for the EC treatment of textile wastewater by SS electrodes

Std	<i>j</i> (X1)	<i>w</i> (X2)	<i>t</i> (X3)	pH (X4)	% COD reduction		% Color reduction		Specific energy, kWh/kg COD removed	
					<i>Y</i> _{exp} (%)	<i>Y</i> _{pre} (%)	<i>Y</i> _{exp} (%)	<i>Y</i> _{pre} (%)	<i>Y</i> _{exp} (%)	<i>Y</i> _{pre} (%)
1	138.89	1	70	7	36.71	38.45	99.45	89.32	18.71	20.41
2	83.33	1	70	7	31.81	31.63	75.40	75.29	8.13	7.84
3	111.11	0.5	120	8.5	46.75	43.15	98.30	99.67	18.97	18.15
4	111.11	1.5	40	8.5	22.37	23.72	55.22	44.95	12.23	13.13
5	111.11	1.5	120	8.5	42.76	44.31	95.05	116.32	19.63	20.04
6	111.11	1.5	120	5.5	42.10	44.17	95.83	95.73	19.65	19.47
7	55.56	1.5	40	5.5	11.80	14.17	0.000	8.37	6.15	6.20
8	83.33	0	70	7	0.000	11.34	0.000	28.51	0.000	5.80
9	83.33	1	10	7	8.52	12.55	0.000	7.89	4.37	6.77
10	83.33	1	70	7	31.81	31.63	75.40	75.29	8.13	7.84
11	83.33	1	70	10	31.64	31.85	79.56	84.72	9.64	10.73
12	55.56	1.5	40	8.5	14.20	15.70	30.08	31.37	5.41	6.22
13	55.56	1.5	120	8.5	27.27	30.62	98.10	103.46	8.62	8.45
14	83.33	1	70	7	31.81	31.63	75.40	75.29	8.13	7.84
15	55.56	0.5	40	8.5	16.39	11.05	14.32	17.17	5.17	3.55
16	55.56	0.5	120	5.5	23.89	21.68	3.89	26.57	12.80	11.12
17	55.56	0.5	120	8.5	28.41	27.04	97.95	67.46	8.49	7.32
18	55.56	1.5	120	5.5	28.65	27.73	67.49	59.46	7.72	9.02
19	27.78	1	70	7	11.36	13.01	4.11	19.94	3.93	4.98
20	83.33	1	70	7	31.81	31.63	75.40	75.29	8.13	7.84
21	83.33	1	130	7	41.48	41.90	99.73	98.21	11.54	13.64
22	111.11	1.5	40	5.5	23.68	24.93	23.33	45.36	11.76	11.97
23	83.33	2	70	7	27.85	19.90	73.61	50.80	10.54	7.50
24	111.11	0.5	40	8.5	22.73	21.49	57.51	50.10	12.99	9.70
25	111.11	0.5	40	5.5	27.21	20.21	52.41	53.62	13.56	11.78
26	111.11	0.5	120	5.5	41.77	40.53	95.75	82.19	22.42	20.81
27	83.33	1	70	4	24.85	28.03	48.95	49.49	11.57	13.23
28	83.33	1	70	7	31.81	31.63	75.40	75.29	8.13	7.84
29	55.56	0.5	40	5.5	11.11	7.04	36.67	-2.72	9.16	6.77
30	83.33	1	70	7	31.81	31.63	75.40	75.29	8.13	7.84

$$\begin{aligned}
 Y_1 = & 31.63 + 12.72 j + 4.28 w + 14.68 t + 1.91 \text{ pH} \\
 & - 5.90 j^2 - 16.01 w^2 - 4.41 t^2 - 1.69 \text{ pH}^2 \\
 & - 2.42 j \times w + 4.26 j \times t - 2.74 j \times \text{pH} \\
 & - 0.81 w \times t - 2.48 w \times \text{pH} + 1.01 t \times \text{pH} \quad (5)
 \end{aligned}$$

$$\begin{aligned}
 Y_3 = & 7.84 + 7.71 j + 0.85 w + 3.44 t - 1.25 \text{ pH} \\
 & + 4.86 j^2 - 1.19 w^2 + 2.36 t^2 + 4.14 \text{ pH}^2 \\
 & + 0.76 j \times w + 3.51 j \times t + 1.14 j \times \text{pH} \\
 & - 1.15 w \times t + 3.23 w \times \text{pH} - 0.44 t \times \text{pH} \quad (7)
 \end{aligned}$$

$$\begin{aligned}
 Y_2 = & 75.29 + 34.69 j + 11.14 w + 45.16 t \\
 & + 17.61 \text{ pH} - 20.66 j^2 - 35.63 w^2 - 22.24 t^2 \\
 & - 8.18 \text{ pH}^2 - 19.34 j \times w - 0.54 j \times t \\
 & - 23.41 j \times \text{pH} + 16.35 w \times t + 3.11 w \times \text{pH} \\
 & + 15.76 t \times \text{pH} \quad (6)
 \end{aligned}$$

Statistical testing of the model was performed with the Fisher’s statistical test (*F*-test) ANOVA, which was conducted to determine the fitness of the second-order polynomial equation with the experimental results. The results of the ANOVA for COD, color, and specific energy consumption by Al and SS electrodes are shown in Table 5. The ANOVA results for the

Table 5

ANOVA for the second-order polynomial model of COD and color removal and specific energy consumption by Al and SS electrode

Source	Sum of squares	Degrees of freedom	Mean square	F-value	P	R ² -values
<i>Al electrode</i>						
<i>COD removal</i>						
Residual	258.63	15	17.24	7.97	0.0001	R ² = 0.8815
Lack of fit	258.63	10	25.86			R ² _{adjust} = 0.7710
Pure error	0.000	5	0.000			
<i>Color removal</i>						
Residual	1032.10	15	68.81	20.09	<0.0001	R ² = 0.9494
Lack of fit	1032.10	10	103.21			R ² _{adjust} = 0.9021
Pure error	0.000	5	0.000			
<i>Specific energy consumption</i>						
Residual	69.96	15	4.66	14.94	<0.0001	R ² = 0.9331
Lack of fit	69.96	10	7.00			R ² _{adjust} = 0.8707
Pure error	0.000	5	0.000			
<i>SS electrode</i>						
<i>COD removal</i>						
Residual	371.11	15	24.74	9.59	<0.0001	R ² = 0.8995
Lack of fit	371.11	10	37.11			R ² _{adjust} = 0.8057
Pure error	0.000	5	0.000			
<i>Color removal</i>						
Residual	6231.75	15	415.45	5.03	0.0018	R ² = 0.8243
Lack of fit	6231.75	10	623.18			R ² _{adjust} = 0.6603
Pure error	0.000	5	0.000			
<i>Specific energy consumption</i>						
Residual	94.63	15	6.31	7.89	0.0001	R ² = 0.8805
Lack of fit	94.63	10	9.46			R ² _{adjust} = 0.7689
Pure error	0.000	5	0.000			

COD, color, and specific energy consumption by Al electrode showed a high *F*-value of 7.97, 20.09, and 14.94, respectively. Respective *F*-values for SS electrode were 9.59, 5.03, and 7.89, respectively. The large value of *F* indicates that most of the variation in the response can be explained by the regression equation, and the terms in the model have a significant effect on the response. The three models gave *R*² values of 0.8815, 0.9494, and 0.9331, respectively, for Al electrode and 0.8995, 0.8243, and 0.8805, respectively, for SS electrode. These values express a good correlation between the observed and the predicted values.

3.2. Optimization analysis

Since there are three responses in this study, therefore, multi-response processes optimization by desirability function approach was used to optimize the EC treatment of textile industry wastewater. One-sided desirability *d_i* is used in the study given by [9,35,36]:

$$d_i = \begin{cases} 0 & \text{if } Y_i \leq Y_{i-\min} \\ \left[\frac{Y_i - Y_{i-\min}}{Y_{i-\max} - Y_{i-\min}} \right]^r & \text{if } Y_{i-\min} < Y_i < Y_{i-\max} \\ 1 & \text{if } Y_i \geq Y_{i-\max} \end{cases} \quad (8)$$

where *Y_i* is response values, *Y_{i-min}* and *Y_{i-max}* is minimum and maximum acceptable values of response *i*, and *r* is a weight and a positive constant used to determine scale of desirability. The individual desirability functions are combined in order to obtain the overall desirability *D*, as follows:

$$D = (d_1 d_2 d_3 \dots)^{(1/k)} \quad (9)$$

where $0 \leq D \leq 1$ and *k* is number of responses.

For EC treatment of textile industry wastewater, responses *Y₁* and *Y₂* are to be maximized while *Y₃* is to be minimized. Since the optimum conditions for responses *Y₁* and *Y₂* are not same, the desirability function approach could be utilized to get the maximum *Y₁* and *Y₂* and minimum *Y₃*, simulta-

neously. The constraints applied for the optimization of various operational parameters is given in Table 6.

3.2.1. Al electrode

For %COD removal (Y_1), the minimum and maximum acceptable values are considered as 11.66% (the minimum experimental value) and 38.81% (maximum experimental value), respectively, in this work. Following equations show the desirability of the individual corresponding responses of %COD removal (Y_1), % color removal (Y_2), and energy consumption (Y_3) of Al electrode:

$$d_1 = \begin{cases} 0 & \text{if } Y_1 \leq 11.66 \\ \left[\frac{Y_1 - 11.66}{38.81 - 11.66} \right] & \text{if } 11.66 < Y_1 < 38.81 \\ 1 & \text{if } Y_1 \geq 100 \end{cases} \quad (10)$$

In a same way, one-sided desirability of Y_2 (d_2)

$$d_2 = \begin{cases} 0 & \text{if } Y_2 \leq 13.15 \\ \left[\frac{Y_2 - 13.15}{98.9 - 13.15} \right] & \text{if } 13.15 < Y_2 < 98.9 \\ 1 & \text{if } Y_2 \geq 98.9 \end{cases} \quad (11)$$

and one-sided desirability of Y_3 (d_3)

$$d_3 = \begin{cases} 1 & \text{if } Y_3 < 1.99 \\ \left[\frac{25.58 - Y_3}{25.58 - 1.99} \right] & \text{if } 1.99 \leq Y_3 \leq 25.58 \\ 0 & \text{if } Y_3 > 25.58 \end{cases} \quad (12)$$

In the above both equations $r=1$. The overall desirability D is calculated by the following equation:

$$D = \sqrt[3]{d_1 d_2 d_3} \quad (13)$$

By using D as a new desirability, the optimum values of operational parameters were found to be $J=97.22 \text{ A/m}^2$, $w=2.0 \text{ g/l}$, $t=120 \text{ min}$, and $\text{pH}_o=4.0$ which produced overall $D=0.981$. To verify the optimization result, three verification run were conducted with the optimized set of operational parameters. The average value of responses Y_1 , Y_2 , and Y_3 were 46.87%, 98.6%, and 25.04 kWh/kg COD removed using Al electrode.

3.2.2. SS electrode

Similarly, according to the above method, overall desirability for SS electrode was calculated by following equations:

$$d_1 = \begin{cases} 0 & \text{if } Y_1 \leq 8.52 \\ \left[\frac{Y_1 - 8.52}{46.75 - 8.52} \right] & \text{if } 8.52 < Y_1 < 46.75 \\ 1 & \text{if } Y_1 \geq 46.75 \end{cases} \quad (14)$$

$$d_2 = \begin{cases} 0 & \text{if } Y_2 \leq 8.52 \\ \left[\frac{Y_2 - 3.89}{99.73 - 3.89} \right] & \text{if } 3.89 < Y_2 < 99.73 \\ 1 & \text{if } Y_2 \geq 99.73 \end{cases} \quad (15)$$

$$d_3 = \begin{cases} 1 & \text{if } Y_3 < 3.93 \\ \left[\frac{22.42 - Y_3}{22.42 - 3.93} \right] & \text{if } 3.93 \leq Y_3 \leq 22.42 \\ 0 & \text{if } Y_3 > 22.42 \end{cases} \quad (16)$$

Thus, the overall desirability for SS electrode was calculated with Eq. (13) by regression analysis, the optimum values of selected variables were $J=111.11 \text{ A/m}^2$, $w=1.0 \text{ g/L}$, $t=105 \text{ min}$, and $\text{pH}_o=8.0$ which produced the maximum $D=0.805$. Correspondingly, the % COD removal, % color removal, and energy consumption were 54.37%, 83.89%, and 30.19 kWh/kg COD, respectively.

3.3. Effects of process parameters

Various reactions take place in the EC reactor with Al and SS as electrode material [12–14,37]. EC process involves the generation of coagulants *in situ* by dissolving Al and iron ions from Al and SS electrodes, respectively, at the anode.



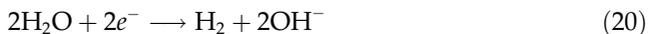
Also, oxygen evolution can compete with Al or iron dissolution at anode via following reaction:



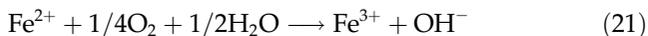
Table 6
Constraints applied for the optimization of EC treatment of acrylic-bearing textile wastewater by Al and SS electrodes

Variables	Objective	Lower limit	Upper limit	Lower weight	Upper weight	Importance
J (A/m^2)	Minimize	27.78	138.89	1	1	3
w (g/l)	Is in range	0	2	1	1	3
t (min)	Minimize	10	130	1	1	3
pH_o	Is in range	4	10	1	1	3

At cathode, hydrogen evolution takes place via the following reaction. It helps in floatation of the flocculated particles out of the water.



For SS electrodes, ferrous ions are oxidized to ferric ions by oxygen generated via Eq. (19) in the aqueous phase



3.3.1. Al electrode

Three-dimensional response surface graphs for all responses by various operational parameters J , w , t , and pH_i are shown in Fig. 1 by Al electrode. Faraday's law describes the relationship between J and the amount of anode material that dissolves in the solution. The COD removal efficiency depends directly on the concentration of Al^{3+} ions produced by the Al electrodes, which in turn as per Faradays law depends upon the t . When the value of t increases, an increase occurs in the concentration of Al ions and their hydroxide flocs. Consequently, an increase in t increases the COD and color removal efficiency (Fig. 1

(a) and (b)). Continuous increase in specific energy consumption as shown in Fig. 1(c) is due to the increase in energy consumption due to increase in J and t .

Fig. 1(d)–(f) shows the effect of w and pH on percent COD removal, percent color removal, and specific energy consumption, respectively. During EC with chloride ions present in the solution, chlorine gas gets generated at anode. Kushwaha et al. [35] have explained that chlorine forms various chlorine species (Cl_2 , HOCl and ClO^-) in the EC reactor depending on the pH of the solution. These chlorine species can indirectly oxidise the organic material present in the wastewater. ClO^- is dominating specie at higher pH and is considered as better oxidant among all chlorine species [38].

Al^{3+} and OH^- ions get generated during EC treatment via electrode reactions (17) and (20), and they react to form various monomeric species, such as $\text{Al}(\text{OH})^{2+}$, $\text{Al}(\text{OH})_2^+$, $\text{Al}(\text{OH})_4^-$, and polymeric species, such as $\text{Al}_6(\text{OH})_{15}^{3+}$ and $\text{Al}_7(\text{OH})_{17}^{4+}$ [39]. The concentration of the hydrolyzed Al species depends on the Al concentration, and the solution pH . Apart from a narrow pH region approximately 5–6, the dominant soluble Al species in solution are Al^{3+} and $\text{Al}(\text{OH})_4^-$ at low and high pH , respectively. It may be seen in

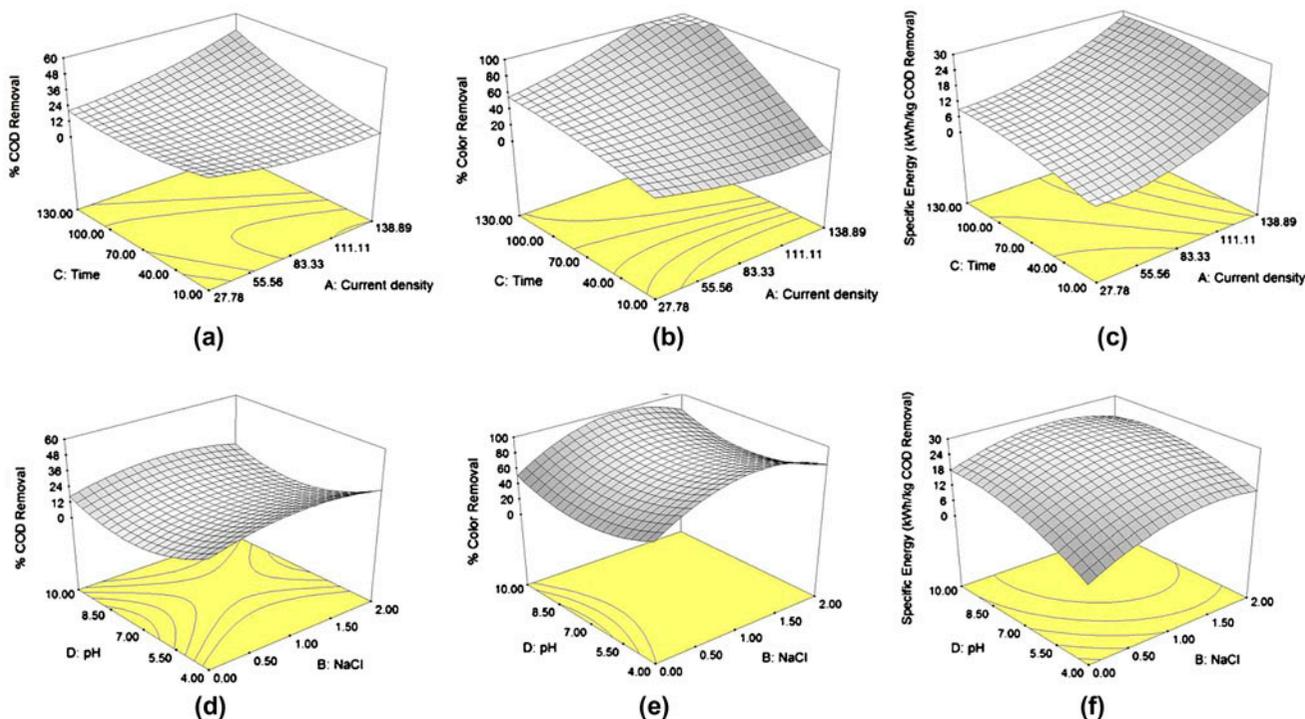


Fig. 1. Effect of various variables for EC treatment by Al electrode. (a) Effect of time and current density on COD removal efficiency, (b) Effect of time and current density on color removal efficiency, (c) Effect of time and current density on specific energy consumption, (d) Effect of pH and NaCl on COD removal efficiency, (e) Effect of pH and NaCl on color removal efficiency, and (f) Effect of pH and NaCl on specific energy consumption.

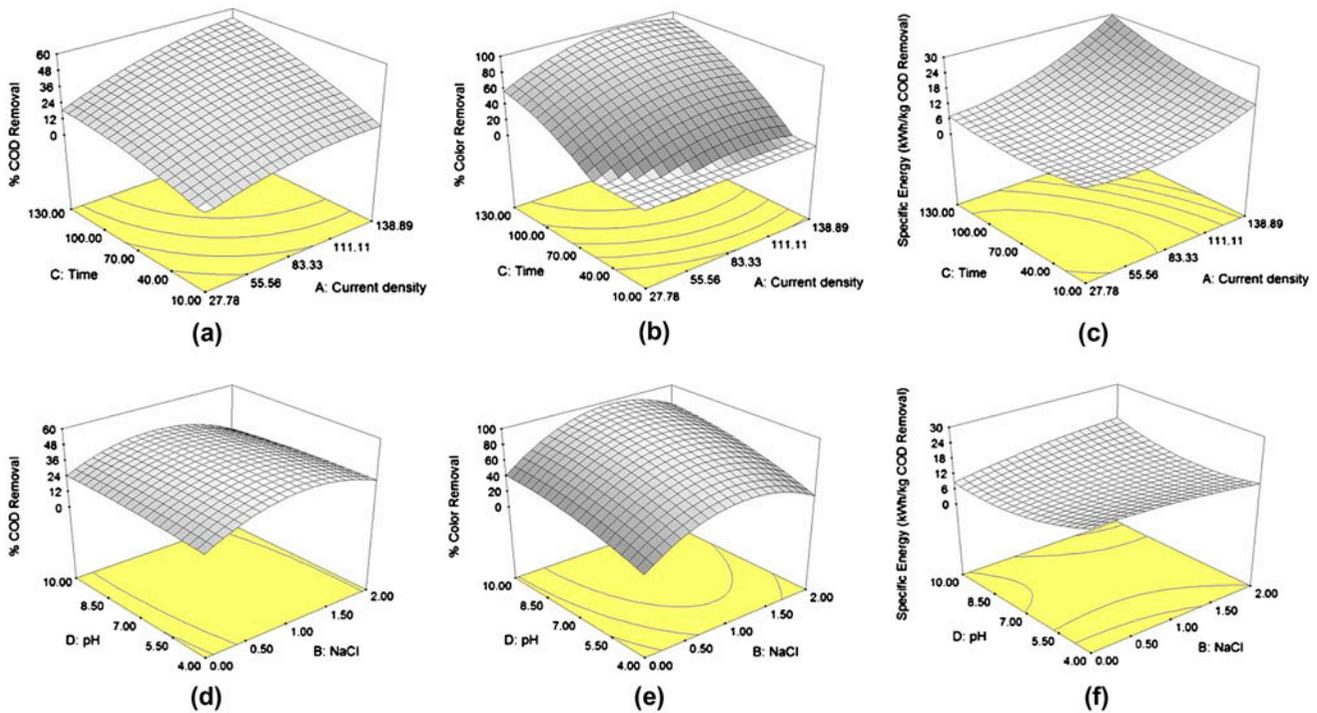


Fig. 2. Effect of various variables for EC treatment by SS electrode. (a) Effect of time and current density on COD removal efficiency, (b) Effect of time and current density on color removal efficiency, (c) Effect of time and current density on specific energy consumption, (d) Effect of pH and NaCl on COD removal efficiency, (e) Effect of pH and NaCl on color removal efficiency, and (f) Effect of pH and NaCl on specific energy consumption.

Fig. 1(d)–(f) that the effect of pH is very marginal on COD and color removal. It seems that combinations of removal mechanisms are operating during treatment by Al electrodes. At lower pH, Al^{3+} ions help in removal process by charge neutralization. In the pH range of 5–6, charge neutralization by monomeric species and sweep coagulation by $\text{Al}(\text{OH})_3$ species help in maintaining the constant COD and color efficiency. At high pH, ClO^- help in organic removal by direct oxidation.

The COD removal efficiency increased with an increase in w . The increase in COD and color removal efficiency with an increase in w is due to an increase in conductivity of the solution which increases the Al dissolution rate and hypochlorite concentration in the reactor which indirectly oxidise the COD. However, at higher w values, excess Al ions cause charge reversal of the negatively charged colloidal particles present in the wastewater which causes marginal decrease in the COD and color removal efficiency [40].

3.3.1. SS electrode

Fig. 2(a)–(c) shows effect of j and t on percent COD removal, percent color removal, and specific

energy consumption, respectively. It can be inferred from all the figures that higher COD removal, color removal, and energy consumption is achieved at higher j value. This is due to the higher dissolution of electrode material with higher rate of formation of iron hydroxides resulting in higher COD and color removal efficiency due to co-precipitation [41] and higher energy consumption occurred due to higher j value and t . Also, increased amount of sludge produced from the electrodes at higher j value enhances the COD and color removal efficiency via sweep coagulation [42]. The COD and color removal efficiency depends directly on the concentration of ions produced by the electrodes which in turn depends upon t . The wastewater used in the present study contains sodium chloride. The chlorides present in the wastewater form chlorine gas at high anodic potential, which goes through successive reactions producing hypochlorite, which causes indirect oxidation.

Fig. 2(d)–(f) illustrates behavior of pH and NaCl dose on percent COD removal, percent color removal, and specific energy consumption, respectively. It is clear from figures that COD and color removal efficiency increased with an increase in NaCl at any

value of pH. The electrical conductivity is directly proportional to the distance between the two electrodes. As NaCl increases, resistance offered by the cell decreases. From Faraday's law, the amount of iron oxidized increases as NaCl increases, and thus, the COD and color removal efficiency generally increases. The results reveal that at $\text{pH} \approx 8$, the COD and color removal efficiency was maximum. It is observed that COD and color removal efficiency increased with an increase in pH from 5 to 8 and then started decreasing for $\text{pH} > 8$. For $\text{pH}_0 < 6$, the protons in the solution get reduced to H_2 , and thus, the proportion of hydroxide ion produced is less and consequently there is less COD and color removal efficiency [43].

Precipitation and adsorption are the two major interaction mechanisms which are applicable at different pH ranges. At low pH values, metal species like Fe^{2+} and Fe^{3+} generated at the anode via electrode reactions (18) and (21) bind to the anionic colloidal particles present in the wastewater, thus, neutralizing their charge and reducing their solubility. This process of removal is termed precipitation. The adsorption mechanism operates at higher pH range (>6) and

involves adsorption of organic substances on amorphous metal hydroxide precipitates. It is evident that COD abatement rates significantly decreased with increasing the $\text{pH} > 7$. Former studies have already revealed that EC works best when the initial reaction pH is in the range of 3–8, so that all forms of metal hydroxide complexes play an active role during the removal of pollutant via EC [44]. Energy consumption graph does not show much effect of pH and NaCl dose; however, it seems at $\text{pH} = 7$ minimum energy consumption is observed. Energy consumption increased again for $\text{pH} > 7$.

Maximum COD using Al and SS electrodes were 46.87 and 54.37%, respectively. Respective color removal efficiencies were 98.61 and 83.89%, respectively. Therefore, SS electrode performed better in terms of COD removal as compared to Al electrodes. However, Al electrode performed better in terms of color removal efficiency. This probably results from the differences in the mechanisms of COD removal for the SS and Al electrodes. The COD removal by Al electrodes is mainly due to charge neutralization by generated Al cations while the COD removal by SS

Table 7
Reported conditions for the electro-chemical treatment of textile wastewaters

Wastewater (ww)	Electrode type	Initial COD (mg/L)	% COD reduction	References
Textile ww containing Levafix Blue CA, Levafix Red CA and Levafix Yellow CA reactive dyes	Fe	950, 690 and 740	32, 37 and 33	[46]
Reactive textile dyes	Al		36	[17]
Simulated acid dyebath ww	Al and SS		40 and 50	[47]
Simulated textile ww	SS as cathode and Fe as anode	3,505	53.5	[23]
Simulated laundry ww	Al	226	62	[48]
Textile ww	Al and Fe	2031	63 and 65	[8]
Industrial ww	Al	1,260	70	[49]
Textile dye ww	SS	3,162	71	[50]
Synthetic solution and textile ww	Al	340	79.7	[51]
Textile dyeing ww	Fe	485	84	[18]
Reactive textile dye solution and a textile ww	SS and Fe	1,200 and 885	89.7 and 80.4	[52]
Textile ww (actual and synthetic)	SS as cathode and titanium covered by a thin film of tantalum, platinum and iridium as anode	405 actual and 281 synthetic	90	[53]
Textile ww	Al	620	90.3	[54]
Synthetic ww	SS and Al	3,200	46.87 and 54.37	Present study

electrodes is due to the combined effect of charge neutralization by Fe cations and sweep coagulation by $\text{Fe}(\text{OH})_3$. However, lower color removal in case of SS electrodes is due to generation of Fe^{2+} and Fe^{3+} ions which impart green and yellow colors to the treated solution. Moreover, $\text{Fe}(\text{OH})_3$ formed also has yellow color [45].

It may be seen in Table 7 that various authors have reported lower COD removal as compared to that obtained in the present study by SS and Al electrodes [46–54]. Similarly, many previous investigators have reported higher removal efficiencies; however, many of these studies used wastewaters which had lower initial COD. Similarly, few researchers have reported higher COD removal efficiencies for wastewaters which had high initial COD also. Thus, it may be interpreted that COD removal efficiencies by EC treatment depends upon several parameters which include type and quantity of dyes and other chemicals present in the wastewater. It also depends upon type of electrode and various operating parameters.

4. Conclusion

The EC degradation of COD, color, and specific energy reduction was investigated in synthetic textile dye wastewater using Al and SS electrodes. Experiments were made as a function of initial pH (pH_0): 4–10, current density (j): 27.78–138.89 A/m², NaCl concentration (w): 0–2 g/L, and electrolysis time (t): 10–130 min. Response surface methodology by the BB model was used to examine the role of 4-factor and 5-level on COD, color, and specific energy consumption efficiency. It was shown that a second-order polynomial regression model could properly interpret the experimental data with coefficient of determination (R^2) value for COD (0.8815, 0.8995), color (0.9494, 0.8243), and specific energy consumption (0.9331, 0.8805) for Al and SS electrodes, respectively, corresponding F-value COD (7.97, 9.59), color (20.09, 5.03), and specific energy consumption (14.94, 7.89) for Al and SS electrodes, respectively. The effect of operating parameters on COD removal, color removal, and specific energy consumed was optimized using response surface methodology and the approximating functions were obtained with satisfactory degrees of fit.

Under specified cost-driven constraints determined for highest desirability, maximum COD and color removal and minimum specific energy of 46.87, 98.61, and 25.04%, respectively, from Al electrode and 54.37, 83.89, and 30.19, respectively, from SS electrode were observed at optimum conditions. This study shows that the BB model is suitable to optimize the experi-

ments for COD, color, and specific energy consumed of synthetic textile wastewater.

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