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ORIGINAL ARTICLE

The effects of chemical coagulants on the decolorization of dyes by electrocoagulation using response surface methodology (RSM)

Erick B. Butler[1] · Yung-Tse Hung[2] · Oliver Mulamba[1]

Abstract This study assessed the efficiency of electrocoagulation (ECF) coupled with an addition of chemical coagulant to decolorize textile dye. Tests were conducted using Box Behnken methodology to vary six parameters: dye type, weight, coagulant type, dose, initial pH and current density. The combination of electrocoagulation and chemical coagulation was able to decolorize dye up to 99.42 % in 30 min of treatment time which is remarkably shorter in comparison with using conventional chemical coagulation. High color removal was found to be contingent upon the dye type and current density, along with the interactions between the current density and the coagulant dose. The addition of chemical coagulants did enhanced treatment efficiency.

Keywords Response surface methodology ·
Electrocoagulation · Decolorization · Textile wastewater ·
Chemical coagulation

Introduction

The characteristics of dye wastewater from the textile industry include high color, organic content (Shelley et al. 1976; Tezcanli-Guyer and Ince 2003; Trifi et al. 2011), volatile organic compounds, biological oxygen demand (BOD), chemical oxygen demand (COD), and total suspended solids (TSS) (World Bank Group, United Nations Environment Programme, and United Nations Industrial Development Organization 1999). Wastewater from the industry is generally produced during dyeing. In many cases, wastewater is then a consortium of various dyes that did not affix to the fabrics during the dyeing processes (Joshi et al. 2004). If untreated, discharge from textile plants into surface waters alters the dynamics within the stream, producing unwanted pollution, increased toxicity, and eutrophication (Trifi et al. 2011).

Color removal is one of the most essential parameters that need to be addressed. Color in effluent is not preferred for several reasons (Joshi et al. 2004):

1. The presence of color changes the appearance of any water source such as a stream.
2. The chemical structure of the dyes interrupts biological and photosynthetic activity. Color has the potential of preventing light from penetrating into a water body.
3. Many dyes are hazardous and have the potential of being carcinogenic and mutagenic.
4. Color can impact the efficiency of wastewater treatment.

There are several methods employed to treat textile wastewater. One method of decolorizing dye wastewater is electrocoagulation. It is a technical process in which wastewater is cleaned through radio or short wave frequencies. The technology works as follows—a pair of aluminum or iron metal sheets is connected to a power supply. The anodes oxidize forming metal ions, while the cathodes disassociate water, forming hydroxide (OH^-) and hydrogen gas (H^+). The oxidized metal combines with the disassociated hydroxide forming metal hydroxides (Butler et al. 2011). The addition of metal hydroxides neutralizes and separates the dye from the wastewater and attaches it to

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the complex. The dye hydroxide complex floats to the top of the reactor and is skimmed at high currents (Martinez-Huitle and Brillas 2009). Electrocoagulation replaces the need for a chemical coagulant. Overall, electrocoagulation has the advantage of reducing constituents at low retention times.

Nevertheless, while electrocoagulation is as an alternative to the application of chemical coagulation for wastewater treatment, the prospect of considering the use of chemical coagulant as an additive to improve treatment is an important factor that needs to be considered. A search through the literature concludes that few authors have considered combined electrocoagulation and chemical coagulation (Can et al. 2006; Merzouk et al. 2011; Canizares et al. 2006; Taheri et al. 2013). When it has been employed, all authors with the exception of Taheri et al. 2013 have only used the traditional chemical coagulation jar tests and have not added chemical coagulants directly into the reactor. Therefore, a further study on effects of adding chemical coagulants to an electrocoagulation reaction for treatment of wastewater is warranted.

Response surface methodology (RSM) is a statistical method used to determine the minimum number of experiments conducted to still produce statistically significant results (Wang et al. 2011). Based on the literature in the last decade, authors have preferred to employ RSM to treat textile wastewater by electrocoagulation. Appendix Table 5 provides a summary table considering the parameters and the dye(s) treated. Upon further assessments, it is observed that only two authors considered varying dyes (albeit they are from the same classification) (Körbahti 2007; Murugan et al. 2009), while only one employed combined electrocoagulation/chemical coagulation using only one coagulant (powdered activated carbon) (Taheri et al. 2013). An investigation of several textile dyes is important because the physical and chemical nature of conventional textile dye wastewater can be very complex. Therefore, employing a study on several classifications of textile dyes will contribute to a general understanding of how well one can treat dyes from a given classification.

Box–Behnkenis an example of an RSM technique that assigns a number to three treatments of a parameter— +1 for the highest treatment, −1 for the lowest, and 0 to the center (Tosik and Wiktorowski 2011) in a process known as coding. This computation is expressed in Eq. 1 (Aleboyeh et al. 2008; Gürses et al. 2002; Tir and Moulai-Mostefa 2008; Zaroual et al. 2009; Amani-Ghadim et al. 2013; Taheri et al. 2012):

$$x = X - X_{\text{center}}/(\Delta X) \tag{1}$$

where x = coded data value (−1, 0, 1)

  X = uncoded data point

  X_{center} = center point

ΔX = the distance between the two points

Box–Behnken has been used in designing electrocoagulation experiments to treat color using iron anodes, varying the current density, treatment time, and wash dilution (Prasad et al. 2008). Chavalparit and Ongwandee (2009) found Box–Behnken useful for determining the effects of pH, applied voltage, and reaction time to predict the removals of chemical oxygen demand (COD), oil and grease (O&G), and suspended solids (SS). Zodi et al. (2010) conclude that Box–Behnken is a viable statistical method for optimizing the treatment of industrial wastewater.

As previously stated, little research has been done on using this technique to design an experiment evaluating the impact of chemical coagulation addition to improve the performance of decolorizing dye wastes. The purpose of this experiment is to determine the effects of varying pH, dye weight, current density, applied coagulant, and dose on the color removal of azo dyes (Acid Yellow 11 (AY11) and Acid Orange 7 (AO7)) and nitroso dye Naphthol Green B (NGB) by electrocoagulation using a Box Behnken experimental design.

Materials and methods

Synthetic wastewater was created by weighing and dissolving powder forms of Acid Orange 7 (Santa Cruz Biotechnology, CA, USA), Acid Yellow 11 (Acros Organics, Thermo Fisher Scientific, NJ, USA), and Naphthol Green B (Santa Cruz Biotechnology, CA, USA) into 1 L of distilled water. The wastewater was stirred for homogeneity, and then 25 g/L of sodium chloride (NaCl) and the respective coagulant was added to the solution. The presence of NaCl increases the conductivity and the electric current through the reactor (Körbahti 2007).

The initial pH for each run was adjusted using nitric acid (HNO₃) to decrease the pH of the solution, while sodium hydroxide (NaOH) increases the solution pH. After stirring, a sample was extracted and placed into a test tube vial. The remaining sample was then transferred to an 11.43 cm × 13.97 cm × 16.51 reactor with an average electrode spacing of 12.86 cm. The reactors consist of a setup with a power supply (YiHua DC Power Supply YH-305D, Guangzhou YiHua Electronic Equipment Co., Ltd, Hong Kong) and aluminum electrodes with an average area of 16.05 cm². The power supply was turned on and samples were collected at detention times of 5, 10, 15, 20, and 30 min. Temperature and pH measurements were taken using a portable pH meter (Oakton Waterproof PC Tester 35 Multi-parameter pH Meter, Oakton Instruments, IL) within a few minutes after sample collection. The samples were then transferred to sample vials for color analysis

using a spectrophotometer (Thermo Spectronic Genesys 20 Spectrophotometer, USA). Figure 1 depicts the electrocoagulation setup.

The Box–Behnken design was created using Minitab 16 Statistical Software (Minitab Inc., PA, USA). Six parameters were assigned coded values as computed from Eq. 1—dye weight (20 mg, 33 mg, and 45 mg), pH (4, 7, and 9), current densities (19.38, 25.83, and 31.15 A/m^3), coagulant (alum, ferric sulfate, and ferric chloride), coagulant doses (5, 10, and 15 mg/L), and three types of dye (acid yellow 11, acid orange 7, and naphthol green B). Table 1 provides the coding for the Box–Behnken experiments. Figure 2 is the structural formulas for each textile dye.

A total of 54 experiments were computed by Minitab based on these six parameters. Collected data signifying the color removal after 30 min was entered into Minitab to develop an equation similar to the format of Eq. 2 (National Institute of Science and Technology 2013):

$$y = b_0 + b_1x_1 + b_2x_2 + b_3x_3 + b_{12}x_1x_2 + b_{13}x_1x_3 + b_{23}x_2x_3 + b_{11}x_1^2 + b_{22}x_2^2 + b_{33}x_3^2 \quad (2)$$
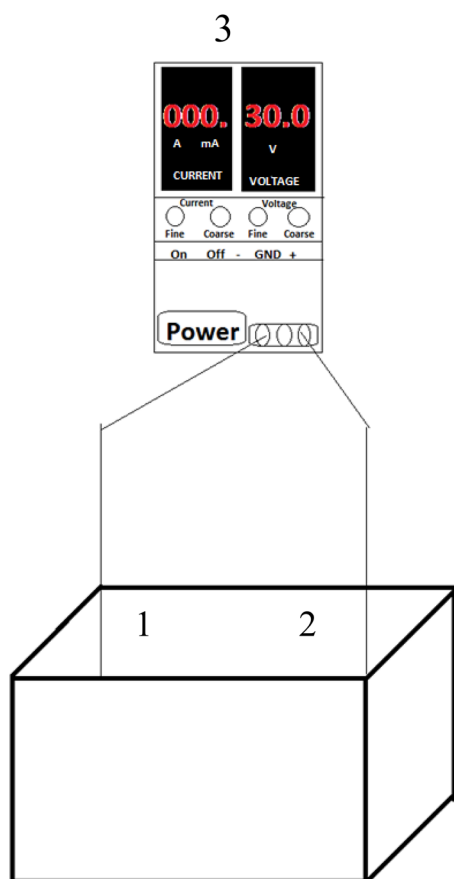
where

y = the resultant variable

x_n = the factors considered within the experiment

b = coefficients

The calculation of color removal is presented below (Nguyen et al. 2012):

$$\text{Color removal} (\%) = \left(\frac{A - A_0}{A_0}\right) \times 100 \quad (3)$$

where A_0 = absorbance at time 0

A = absorbance at time t (5, 10, 15, 20, and 30 min)

Results and discussion

Response surface analysis

Table 2 displays the color removal for the experiment after a detention time of 30 min. All appropriate conditions tested during the experiment along with the resulting color removal values are included in this table. This will provide what is necessary to complete a response surface analysis. The information presented in the table produces valuable information on the effectiveness of the treatment method to remove color. In addition, there are several products created from the data recorded in the table. These products include the estimation of regression coefficients, analysis of variance (ANOVA), predicted response for new design, and surface, residual, and main effects plots. These outputs are of great significance for the parameters analyzed in the experiment. With the exception of the main effects plots, the remaining products will be used in the response surface analysis. According to Table 2, the process decolorized Acid Orange 7 at 98.2 %, Naphthol Green B at 99.42 %, and Acid Yellow 11 at 93.65 %. The results from the study indicate that the combined electrocoagulation-chemical coagulant treatment method is capable of removing color from dyes that represent different dye classifications.

Linear regression model

Beyond the scope of determining color removal efficiencies, one of the main outputs that can be produced from the data in Table 2 is the development of a linear equation to describe the behavior This is accomplished through the use of regression coefficients. The output for this is found in Table 3. The residual plots were assessed to determine data distribution taking into account the Anderson–Darling test ($p < 0.05$). After finding that the data was not normal, a Johnson transformation was used. Equation 4, shown below, is the adjustment applied to each coefficient during the transformation:

Fig. 1 Diagram of electrocoagulation reactor setup (1) location of anode electrode (2) location of cathode electrode and (3) DC power supply

Table 1 Coding parameters for the experimental design

		Coding		
		−1	0	1
Type of dye	x_1	Acid Yellow 11	Acid Orange 7	Naphthol Green B
Initial pH	x_2	4	7	9
Dye weight (mg)	x_3	20	33	45
Current Density (A/m^2)	x_4	18.69	24.92	31.15
Coagulant	x_5	Alum	Ferric Sulfate	Ferric Chloride
Dose (mg/L)	x_6	5	10	15

(a)

(b)

(c)
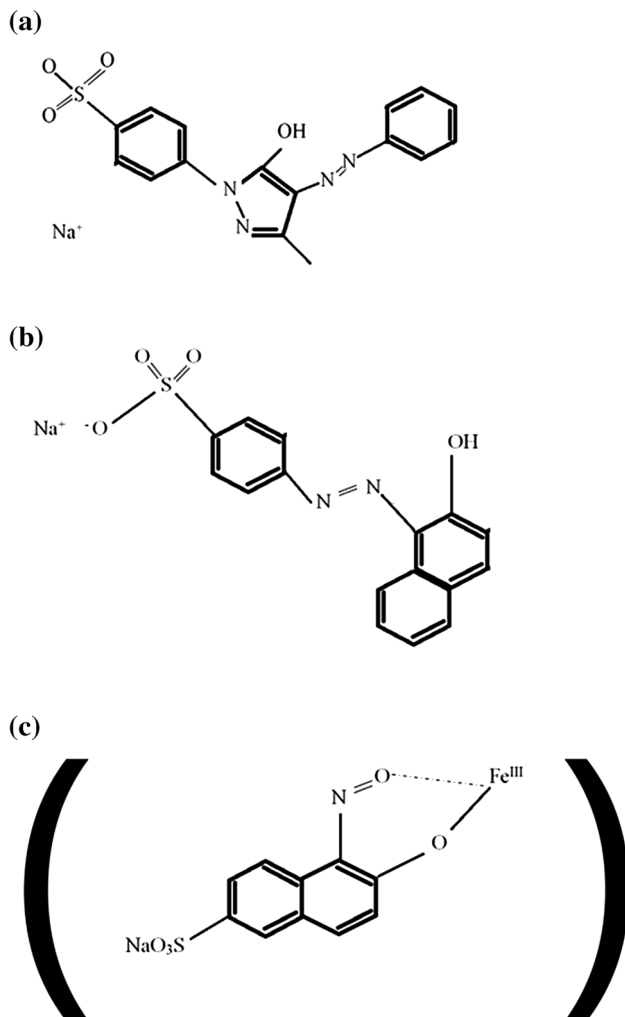
Fig. 2 Structural formulas for each textile dye **a** Acid Yellow 11, **b** Acid Orange 7 and **c** Naphthol Green B

$$x_n = 1.36301 + 0.656222 \times A \sinh((X_n - 98.4463)/0.744691) \qquad (4)$$

where

X_n = the original coefficient of the factor

x_n = the transformed coefficient

Having used the Johnson Transformation, the model fulfilled the criteria of the Anderson–Darling test ($p = 0.303$). After passing the Anderson–Darling test, the regression model was developed. Statistically significant uncoded variables were calculated for the regression model equation and are provided in Eq. 5:

$$y = 0.71481x_1 + 0.6856x_4 - 0.41224x_4x_6 \qquad (5)$$

where

x_1 = dye

x_4 = current density (A/m^2)

x_6 = coagulant dose (mg/L)

The r^2 value for this model was 77.53 % indicating that 22.47 % of the data is not explained by the model (Gürses et al. 2002). The most probable reason for a lower r^2 could be attributed to an unaccounted variable such as time, an important parameter that evaluates the production of Al^{3+} ions from the electrodes, driving the efficiency of decolorization within the system (Daneshvar et al. 2006a).

ANOVA analysis

Statistical analysis was performed by reviewing the regression coefficient (Table 3) and analysis of variable (ANOVA) (Table 4) output tables. When analyzing the transformed data, the data fits the linear model as shown in the ANOVA table.

From the ANOVA table, it has been determined from the p-values that the regression ($p = 0.002$) and linear ($p = 0.000$) models are both statistically significant ($p = 0.000$). The F values were large enough to explain the variability of the data by fitting a linear ($F = 9.34$) and regression ($F = 3.19$) model. While the original data produced has similar p values for both regression and linear, it did not pass the Anderson–Darling test for being normal. There are also no squared interactions that are statistically significant. One thing also to point out is that the lack of fit is not statistically significant in the transformed data, as opposed to the original data (Bhatti et al. 2011). Finally, the F test determined that dye type ($F = 29.18$) and current

Table 2 Box–Behnken design and its associated results

Run order	Dye	pH	Dye weight	Current. Density	Coagulant	Coagulant dose	Color removal (%)	Color removal (transf.)
1	AO7	9	20	24.92	Alum	10	96.38	0.22
2	AO7	9	20	24.92	FeCl$_3$	10	98.82	1.68
3	AO7	7	33	24.92	Fe$_2$(SO$_4$)$_3$	10	97.62	0.74
4	AO7	9	33	24.92	Alum	5	98.2	1.15
5	AO7	9	33	24.92	Alum	15	95.46	−0.01
6	NGB	7	33	18.69	Alum	10	97.35	0.59
7	NGB	7	45	24.92	Fe$_2$(SO$_4$)$_3$	15	97.36	0.59
8	AO7	7	33	24.92	Fe$_2$(SO$_4$)$_3$	10	96.81	0.36
9	NGB	4	33	18.69	Fe$_2$(SO$_4$)$_3$	10	64.87	−1.59
10	AO7	7	33	24.92	Fe$_2$(SO$_4$)$_3$	10	96.9	0.39
11	AY11	7	45	24.92	Fe$_2$(SO$_4$)$_3$	5	92.71	−0.43
12	NGB	7	20	24.92	Fe$_2$(SO$_4$)$_3$	5	97.52	0.68
13	AO7	9	45	24.92	FeCl$_3$	10	92.88	−0.41
14	AO7	7	20	31.15	Fe$_2$(SO$_4$)$_3$	15	95.78	0.06
15	AO7	4	20	24.92	Alum	10	97.82	0.86
16	AY11	7	45	24.92	Fe$_2$(SO$_4$)$_3$	15	88.32	−0.81
17	AO7	7	20	31.15	Fe$_2$(SO$_4$)$_3$	5	97.76	0.82
18	AO7	9	33	24.92	FeCl$_3$	15	97.42	0.62
19	NGB	9	33	18.69	Fe$_2$(SO$_4$)$_3$	10	96.67	0.31
20	NGB	7	20	24.92	Fe$_2$(SO$_4$)$_3$	15	95.43	−0.02
21	AY11	7	33	31.15	FeCl$_3$	10	93.65	−0.32
22	NGB	7	45	24.92	Fe$_2$(SO$_4$)$_3$	5	96.31	0.20
23	AO7	7	45	31.15	Fe$_2$(SO$_4$)$_3$	15	97.14	0.49
24	NGB	9	33	31.15	Fe$_2$(SO$_4$)$_3$	10	*, N/A	*, N/A
25	AO7	4	45	24.92	Alum	10	97.1	0.47
26	AO7	4	45	24.92	FeCl$_3$	10	95.74	0.05
27	AY11	4	33	18.69	Fe$_2$(SO$_4$)$_3$	10	68.24	−1.52
28	AO7	7	45	18.69	Fe$_2$(SO$_4$)$_3$	5	78.37	−1.25
29	AO7	7	45	18.69	Fe$_2$(SO$_4$)$_3$	15	92.93	−0.41
30	AO7	4	20	24.92	FeCl$_3$	10	95.91	0.09
31	AO7	7	33	24.92	Fe$_2$(SO$_4$)$_3$	10	91.59	−0.55
32	AO7	9	33	24.92	FeCl$_3$	5	88.54	−0.79
33	NGB	7	33	31.15	FeCl$_3$	10	99.31	2.01
34	AY11	9	33	18.69	Fe$_2$(SO$_4$)$_3$	10	87.05	−0.88
35	AY11	7	33	18.69	Alum	10	61.75	−1.65
36	AO7	4	33	24.92	Alum	5	82.3	−1.11
37	NGB	7	33	31.15	Alum	10	99.42	2.07
38	AO7	7	33	24.92	Fe$_2$(SO$_4$)$_3$	10	97.76	0.82
39	NGB	4	33	31.15	Fe$_2$(SO$_4$)$_3$	10	98.62	1.51
40	AO7	7	20	18.69	Fe$_2$(SO$_4$)$_3$	15	97.27	0.55
41	AO7	7	20	18.69	Fe$_2$(SO$_4$)$_3$	5	80.46	−1.18
42	AY11	7	20	24.92	Fe$_2$(SO$_4$)$_3$	5	84.68	−1.01
43	AY11	4	33	31.15	Fe$_2$(SO$_4$)$_3$	10	92.4	−0.47
44	AO7	4	33	24.92	Alum	15	95.87	0.08
45	AY11	7	20	24.92	Fe$_2$(SO$_4$)$_3$	15	91.27	−0.58
46	AY11	9	33	31.15	Fe$_2$(SO$_4$)$_3$	10	92.19	−0.49
47	AO7	4	33	24.92	FeCl$_3$	15	97.39	0.61

Table 2 continued

Run order	Dye	pH	Dye weight	Current. Density	Coagulant	Coagulant dose	Color removal (%)	Color removal (transf.)
48	AY11	7	33	18.69	$FeCl_3$	10	68.84	−1.51
49	AO7	7	33	24.92	$Fe_2(SO_4)_3$	10	97.59	0.72
50	NGB	7	33	18.69	$FeCl_3$	10	90.16	−0.67
51	AO7	9	45	24.92	Alum	10	83.5	−1.06
52	AO7	4	33	24.92	$FeCl_3$	5	86.47	−0.92
53	AY11	7	33	31.15	Alum	10	93.38	−0.35
54	AO7	7	45	31.15	$Fe_2(SO_4)_3$	5	97.04	0.45

* Incorrect current density recorded

Table 3 Estimation of regression coefficients from the Minitab output

Term	Coef	SE Coef	T	p
Constant	0.41292	0.2529	1.633	0.115
Dye	0.71481	0.1323	5.402	0
pH	0.15448	0.1323	1.167	0.254
Dye weight	−0.17884	0.1264	−1.414	0.17
Current density	0.68596	0.1323	5.184	0
Coagulant	−0.03397	0.1264	−0.269	0.79
Coagulant Dose	0.19081	0.1264	1.509	0.144
Dye*dye	−0.27015	0.1949	−1.386	0.178
pH*pH	−0.19565	0.2038	−0.96	0.346
Dye weight*dye weight	−0.01587	0.1936	−0.082	0.935
Current density*current density	−0.1572	0.1949	−0.807	0.428
Coagulant*coagulant	0.03586	0.2	0.179	0.859
Coagulant Dose*coagulant dose	−0.29895	0.1936	−1.544	0.135
Dye*pH	0.15245	0.2483	0.614	0.545
Dye*dye weight	−0.02674	0.219	−0.122	0.904
Dye*current density	0.28028	0.1656	1.693	0.103
Dye*coagulant	−0.18761	0.219	−0.857	0.4
Dye*coagulant dose	−0.04451	0.219	−0.203	0.841
pH*dye weight	−0.36814	0.219	−1.681	0.105
pH*current density	−0.32856	0.2483	−1.323	0.198
pH*coagulant	0.07975	0.1549	0.515	0.611
pH*coagulant dose	−0.30791	0.219	−1.406	0.172
Dye weight*current density	0.13647	0.219	0.623	0.539
Dye weight*coagulant	−0.05874	0.219	−0.268	0.791
Dye weight*coagulant dose	0.01344	0.1549	0.087	0.932
Current density*coagulant	0.13714	0.219	0.626	0.537
Current density*coagulant Dose	−0.41224	0.219	−1.882	0.071
Coagulant*coagulant dose	0.36396	0.219	1.662	0.109

$R_2 = 77.53$ %, R_2 (adj) = 53.26 % ($p < 0.1$ = statistically significant)

density ($F = 26.87$) were the most statistically significant parameters.

Regression analysis

The regression output table (Table 3) produced by Minitab contains four statistics represented by column headings in the table—coefficient (Coef.), standard error of the coefficient (SE Coef.), T value (T), and p value (p).

In the regression output (Table 4), the parameters with statistical significance were chosen when $p < 0.1$. From this experiment, it was determined that the regression coefficients that had the most statistical significance were the type of dye ($p = 0$), current density ($p = 0$), and the

Table 4 ANOVA Table from the results of the RSM

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Regression	27	33.1003	33.1003	1.2259	3.19	0.002
Linear	6	23.7094	21.4978	3.583	9.34	0
Dye	1	10.7136	11.1966	11.1966	29.18	0
pH	1	0.3766	0.5229	0.5229	1.36	0.254
Dye weight	1	0.7676	0.7676	0.7676	2	0.17
Current density	1	10.9501	10.3111	10.3111	26.87	0
Coagulant	1	0.0277	0.0277	0.0277	0.07	0.79
Coagulant dose	1	0.8738	0.8738	0.8738	2.28	0.144
Square	6	2.102	2.0131	0.3355	0.87	0.528
Dye*dye	1	0.7399	0.7373	0.7373	1.92	0.178
pH*pH	1	0.0959	0.3536	0.3536	0.92	0.346
Dye weight*dye weight	1	0.1065	0.0026	0.0026	0.01	0.935
Current density*current density	1	0.1746	0.2496	0.2496	0.65	0.428
Coagulant*coagulant	1	0.08	0.0123	0.0123	0.03	0.859
Coagulant dose*coagulant dose	1	0.905	0.9151	0.9151	2.38	0.135
Interaction	15	7.2889	7.2889	0.4859	1.27	0.292
Dye*pH	1	0.1752	0.1446	0.1446	0.38	0.545
Dye*dye weight	1	0.0057	0.0057	0.0057	0.01	0.904
Dye*current density	1	1.4451	1.0998	1.0998	2.87	0.103
Dye*coagulant	1	0.2816	0.2816	0.2816	0.73	0.4
Dye*coagulant dose	1	0.0159	0.0159	0.0159	0.04	0.841
pH*dye weight	1	1.0842	1.0842	1.0842	2.83	0.105
pH*current density	1	0.6717	0.6717	0.6717	1.75	0.198
pH*coagulant	1	0.1018	0.1018	0.1018	0.27	0.611
pH*coagulant dose	1	0.7584	0.7584	0.7584	1.98	0.172
Dye weight*current density	1	0.149	0.149	0.149	0.39	0.539
Dye weight*coagulant	1	0.0276	0.0276	0.0276	0.07	0.791
Dye weight*coagulant dose	1	0.0029	0.0029	0.0029	0.01	0.932
Coagulant*coagulant	1	1.0598	1.0598	1.0598	2.76	0.537
Residual error	25	9.5931	9.5931	0.3837		0.071
Lack-of-fit	20	8.2978	8.2978	0.4149	1.6	0.109
Pure error	5	1.2953	1.2953	0.2591		
Total	52	42.6934				0.317

$R_2 = 78.05$ %, R_2 (adj) $= 54.35$ % ($p < 0.1$ = statistically significant)

interaction between current density and coagulant dose ($p = 0.071$). Gürses et al. (2002) concluded that the type of dye was one of the more important parameters that is statistically significant.

Surface plot analysis

Incorporated within any RSM analysis is the ability to visually represent the effects of various experimental parameters on a desired output. Figures 3a–m is a series of three-dimensional (3D) surface plots that demonstrate the interactions of two variables and their impact on overall color removal. Please note that these figures are based on

the Johnson transformation (Table 2, 9th column) for the color removal.

Another important result is that the experimental error is only (2.5 %) when considering the center point replicates. This value is very comparative of what is in found in literature. Aleboyeh et al. (2008) found the experimental error was (2.18 %) by comparing the color removal of C.I. Acid Red 14 using electrocoagulation. Alinsafi et al. (2005) calculated a 2.8 % experimental error when treating blue reactive dye using electrocoagulation, and Bhatti et al. (2009) determined a 3.02 % experimental error. These values indicate that the results from the experiment are precise.

(a)

(b)

(c)

(d)

(e)

(f)

(g)

(h)
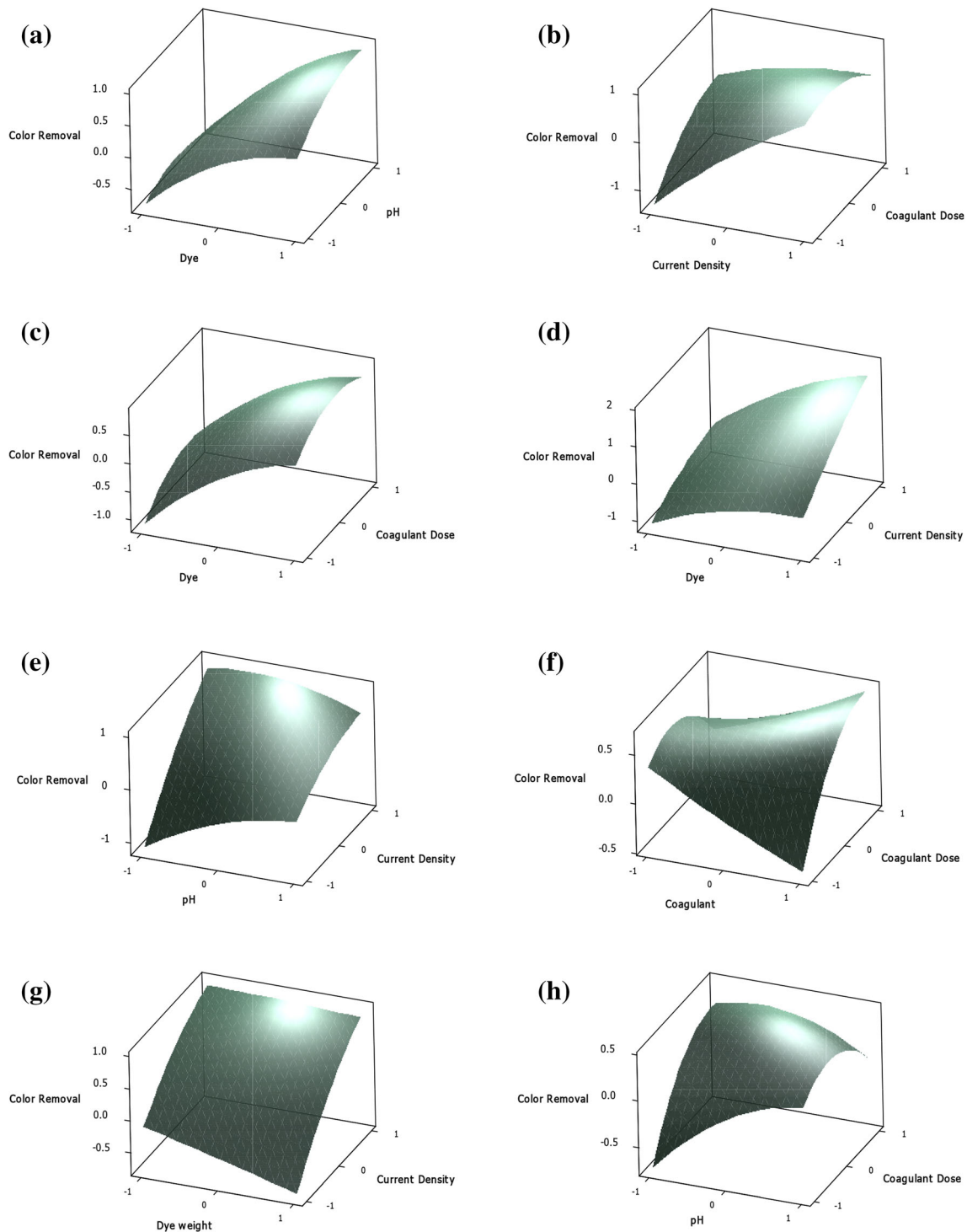
Fig. 3 Surface plots for color removal. **a** color removal vs dye, pH; **b** color removal vs current density, coagulant dose; **c** color removal vs dye, coagulant dose; **d** color removal vs dye, current density; **e** color removal vs pH, current density; **f** color removal vs coagulant, coagulant dose; **g** color removal vs dye weight, current density;

h color removal vs pH, coagulant dose; **i** color removal vs dye, coagulant; **j** color removal vs pH, dye weight; **k** color removal vs pH, coagulant; **l** color removal vs current density, coagulant; **m** color removal vs dye weight, coagulant

The analysis completed by Minitab validates that the combination of electrocoagulation with chemical coagulant addition is capable of removing dyes at different current densities, pH, and coagulant type and doses. With a maximum treatment time of 30 min, color removal ranged from 61.75 to 99.42 % for three different types of dyes. The

electrocoagulation results of Körbahti et al. (2011) required a retention time of 60 min to decolorize Acid Blue 29, Reactive Blue 4, Acid Red 98, and Reactive Red 2 (70.6 to 99.47 %). In comparison to previously documented studies, this work has substantially reduced the treatment time necessary for decolorization of dyes. The treatment time is half of the previously recorded times.
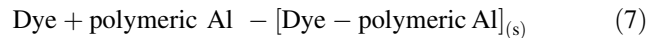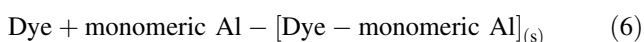
Effects of experimental parameters

It is important to determine the potential reasons as to how each experimental parameter effects the results discovered. Therefore, discussion must be made on the individual effects of current density, initial pH, textile dye type, and coagulation.

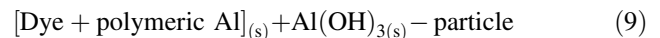### The effect of current density

In electrocoagulation, current density affects the treatment efficiency—an increase in current density will increase the treatment efficiency (Mahmoodi and Dalvand 2013) determining the overall formation of metal hydroxide flocs (Daneshvar et al. 2006b; Karichappan et al. 2014). There are two main reasons to explain this phenomenon. First, the wastewater constituents are neutralized. The intermolecular forces within the solution transition from electrostatic interparticle repulsion to van der Waal's attraction (Wang et al. 2010). This produces more flocs improving treatment efficiency. Second, higher current densities increase the hydrogen bubble production and reduce the size of hydrogen bubbles. This allows agglomerated particulates to float to the surface of the reactor (Mahmoodi and Dalvand 2013). Reviewing the results, it appears that what occurred coincides with these sentiments. At a current density of 31.15 A/m^2, the treatment efficiency range is 92.2–99.4; 82.3–98.6 % of color is removed at a current density of 24.92 A/m^2. Finally, 61.8–97 % of color is removed at 18.69 A/m^2.

The effect of initial pH

In general, the initial pH determines the aluminum hydroxide species present. At a pH less than 4, cationic monomeric species of aluminum [Al(H$_2$O)$_6^{3+}$] is the dominant species. When the pH ranges from 4 to 6, sedimentation is the primary process. At a pH range of 4 to 5, monomeric species of aluminum react with the dye present within the wastewater to form dye-monomeric complexes. Between 5 and 6, polymeric species of aluminum form dye-polymeric complexes (Gürses et al. 2002). Equations 6 and 7 summarize these processes.

$$\text{Dye} + \text{monomeric Al} - [\text{Dye} - \text{monomeric Al}]_{(s)} \qquad (6)$$

$$\text{Dye} + \text{polymeric Al} - [\text{Dye} - \text{polymeric Al}]_{(s)} \qquad (7)$$

However, when the pH was greater than 6.5, dye is adsorbed to the aluminum hydroxide complexes. Equations 8 and 9 describe the resulting constituents formed.

$$\text{Dye} + \text{Al(OH)}_{3(s)} - \text{particle} \qquad (8)$$

$$[\text{Dye} + \text{polymeric Al}]_{(s)} + \text{Al(OH)}_{3(s)} - \text{particle} \qquad (9)$$

In addition, the complexes formed during adsorption vary. When the pH was greater than 9, Al(OH$_4$)$^-$ is the most commonly found species; at a pH greater than 10, it is the only species.

During the experiment, it was found that when the initial pH was 4, the color removal ranges from 64.9 to 98.6 %. At a pH of 7, the range was between 61.8 and 99.42 %. Finally, samples that have an initial pH of 9, 83.5–98.82 % overall, the maximum color removal occurred when the initial pH was at 7. The results from this experiment agreed with the outcomes from previous authors (Murthy et al. 2007; Mahmoodi and Dalvand 2013). Karichappan et al. (2014) explains that the metal hydroxide complexes at this pH have a large surface area which is more conducive to treating wastewater as compared to the presence of Al(H$_2$O)$_6^{3+}$ (Karichappan et al. 2014).

The effect of varying textile dyes

The experiment conducted analyzed the color removal of both azo and nitroso dyes. Azo dyes are one of the most popular dyes present in the textile industry as these dyes represent 50 % of the total dyes present. Azo dyes contain one azo group (–N=N–). Acid Yellow 11 and Acid Orange 7 are examples of low-molecular azo dyes (Hunger 2003). Azo dyes are also anionic and very water soluble (Verma et al. 2012). Körbahti (2007) explains the degradation of both azo and nitroso dyes by means of electrochemical treatment. According to the authors, the products of azo dye degradation include carbon dioxide, nitrates, and sulfates with intermediates of aromatic esters, phenols, aromatic carboxyl acids, cyclic and aromatic hydrocarbons. On the other hand, nitroso dyes consist of o-nitrosophenols or naphthols used on dyeing animal skins and silk (Hunger 2003). Dyes from nitro and nitroso groups are usually degraded into amino groups (Körbahti 2007). It is possible that similar degradation patterns occur within electrocoagulation. Acid Yellow 11, Acid Orange 7, and Naphthol Green B were chosen because they represent a small sample of dyes within these categories. In addition, these dyes have also been decolorized using other treatment methods which allows for a comparison between the chemical coagulation addition of ECF and other treatment methods.

Acid Yellow 11 was decolorized between 61.75 and 93.65 % after 30 min detention time. Four of the six values greater than 90 % occurred when the current density was 31.1 A/m^2. The coagulant that produced the highest treatment efficiency was ferric sulfate, where the pH that produced color removal greater than 90 % was at an initial pH of 7. While electrocoagulation was never a mechanism used for the treatment of Acid Yellow 11, Guo et al. (2006) studied the use of electrochemistry. Xu et al. (2014) degraded Acid Yellow 11 by 90 % in 280 min.

Acid Orange 7 was decolorized 78.37–98.82 % in 30 min retention time. This is very comparable to what was seen in the literature. In many cases Acid Orange 7 has been near or completely decolorized, but at the expense of long retention times—6 h (Ong et al. 2012), 2 h (Wu et al. 2012), and even 50 h (Ong et al. 2008). Acid Orange 7 has been decolorized by Fenton-biological treatment at an optimum pH of 7 (Lodha and Chaudhari 2007), granulated activated carbon (GAC)-biofilm sequencing batch reactor (Ong et al. 2008), wet oxidation (Peng et al. 2008), membrane aerated biological reactor with *Shewanella* sp., and horseradish peroxidase (Gholami-Borujeni et al. 2011).

Naphthol Green B reported the highest treatment removal. When looking at the contour plots (Fig. 3) the dye weight vs dye plot indicated an optimum dye weight of 20 mg. In the current density vs dye plot, the optimum current density was 31.15 A/m^2. When it comes to determining the optimum coagulant, ferric chloride had the highest removal also at the highest current density (31.15 A/m^2), along with having the highest dose (15 mg/L). From the Box–Behnken summary table (Table 3), the coagulant dose is 5 mg/L and pH 9 (FeCl$_3$ = 5 mg/L, pH 9, Color removal = 98.2 %).

When antimony trisulfide was used as a semiconductor, 96.5 % of Naphthol Green B was reduced in 60 min (Ameta et al. 2011). In addition, 10 % Al zinc oxide (ZnO) in sunlight exposure at a treatment time of 6 h (Saber et al. 2012) completely decolorized the dye. Metal hydroxides sludge (MHS) achieved 52 % removal at an optimum pH of 6 (Attallah et al. 2013). Using electrocoagulation, one is capable of having higher treatment efficiency with a shorter retention time as compared with other potential treatment methods.

### The effects of coagulant addition—comparing alum and ferric chloride

From the results, it has been observed that the type of coagulant has an impact on color removal. To better discuss the results, assessments were made based on the initial pH. At an initial pH of 4, ferric chloride performed slightly better in removing AO7. There were two sets of runs that can be compared with similar initial conditions:

1st set

dye = AO7   dye = AO7
coagulant = ferric chloride   coagulant = alum
coagulant dose = 5 mg/L   coagulant dose = 5 mg/L
current density 24.92 A/m^2 current density 24.92 A/m^2
color removal = 86.57%   color removal = 82.30%

2nd set

dye = AO7   dye = AO7
coagulant = ferric chloride   coagulant = alum
coagulant dose = 15 mg/L   coagulant dose = 15 mg/L
current density 24.92 A/m^2 current density 24.92 A/m^2
color removal = 97.39%   color removal = 95.87%

It was found that dye removal can be expected in the acidic range for ferric chloride. This is because the surface charge of the coagulant is positive. A positive surface charge can allow for adsorption and removal of anionic species of dye (Moghaddam et al. 2010). Also, at a pH between 4 and 8, the ferric complexes formed are potentially more insoluble than alum. This provides an opportunity to form more agglomerates. These agglomerates consist of ferric hydroxide complexes that can be easily adsorbed onto colloids (Kim et al. 2004a; Verma et al. 2012).

Coagulant dose also appears to be a significant factor in the removal of color. An increase in coagulant dose tremendously improves color removal regardless of coagulant. Coagulant dose changes the zeta potential. An increase in coagulant dose will increase the zeta potential, thereby reversing the charge of the coagulant. In many cases, the increase will reverse the charge of the coagulant to become positive. With the dye molecule having become positive, the solution becomes stable. It is also possible that the zeta potential during the process is closer to zero improving the removal of color (Kim et al. 2004a).

At a pH of 7, ferric chloride performed better in removing AY11. However, alum performed better when in removing NGB. There are several possible reasons as to why this may have occurred. First, one should consider the initial current density where the first set of experiments occurred at the lowest current density (18.69 A/m^2). As previously stated, optimum treatment occurred when the initial pH was 7. The supplement of coagulant enhanced the formation of aluminum hydroxide complexes, thereby enhancing treatment. In the second set of experiments, the current density was the highest (31.15 A/m^2) providing more binding sites of aluminum hydroxide complexes. Second, the addition of coagulant was suitable for providing more binding sites as compared to the presence of ferric chloride. Third, the difference in the treatment efficiency by ferric chloride is related to the type of dye being treated. Several authors found this to be case when
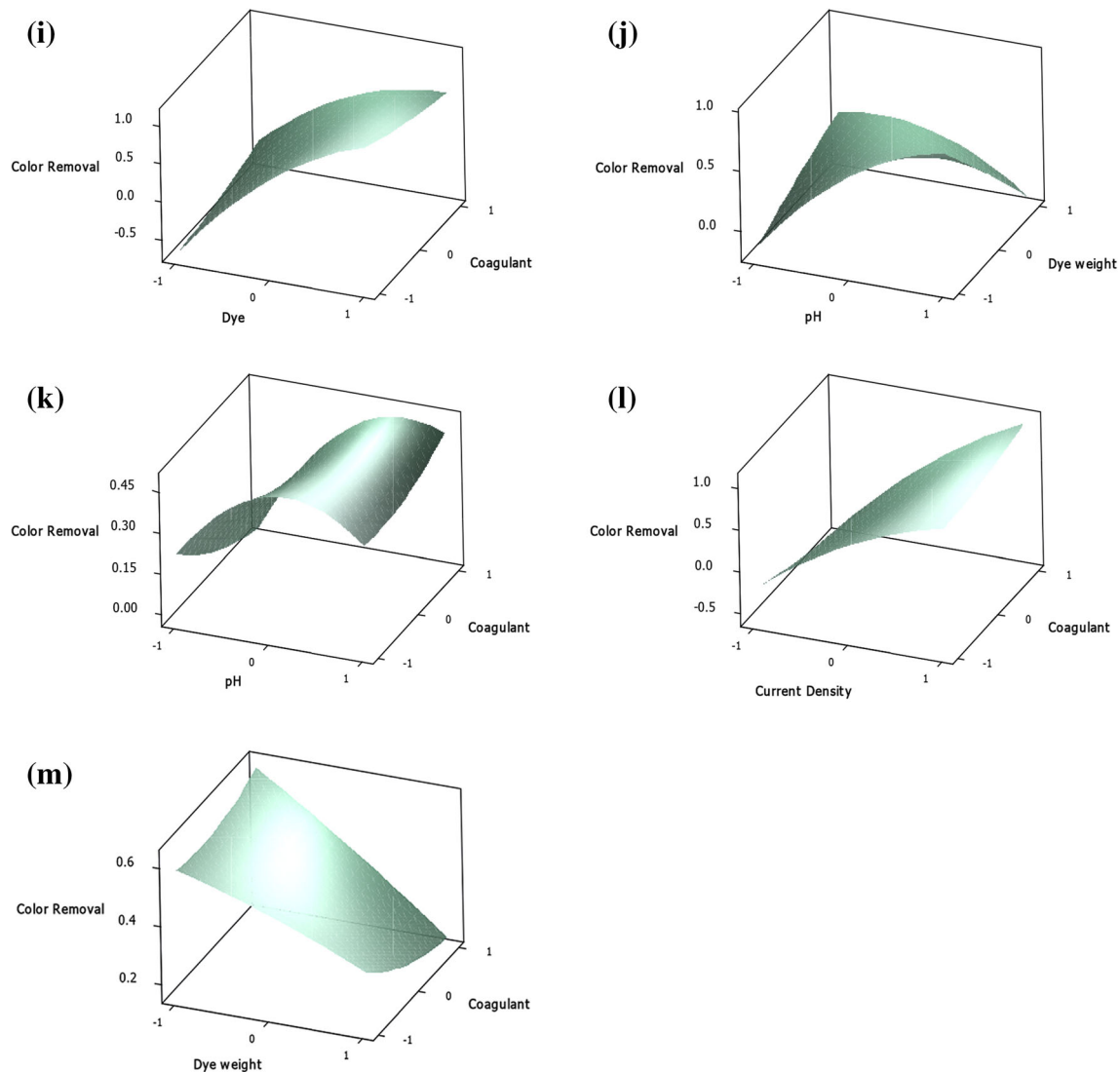
Fig. 3 continued

attempting to optimize the color removal of various textile dyes using ferric chloride (Kim et al. 2004a, b).

At a pH of 9, the addition of ferric chloride (regardless of coagulant dose) reduces color removal as compared to the addition of alum. It is possible that the addition of alum may have shifted the presence of aluminum hydroxide complexes favoring suitable treatment. On the contrary, the addition of ferric chloride may have created an environment for the formation of $Fe(OH)_4^-$, a ferric species formed when the pH is greater than 7. During coagulation, this species reduces treatment efficiency (Kim et al. 2004a).

The effects of coagulant addition—ferric sulfate

Since the experimental design developed had only one run with comparable conditions, the discussion of the effects of

ferric sulfate will be assessed based on initial pH and dye type.

At a pH of 4, it is evident that the current density effects treatment efficiency of both AY11 and NGB. At 18.69 A/m^2, decolorization of AY11 and NGB occurred at 68.24 and 64.87 %, respectively. On the contrary, 92.40 % of AY11 and 98.62 % of NGB was decolorized when the current density was 31.15 A/m^2. The results from adding ferric sulfate to wastewater with an initial pH of 4 indicate that increasing current density increases treatment efficiency.

When the initial pH was 7, decolorization ranged from 78.37 to 97.76 %. The result of 78.37 % color removal occurred when the dye weight was 45 mg, dose of 5 mg/L of coagulant, and a current density of 18.69 A/m^2 with Acid Orange 7 as the dye treated. One the contrary, at a current density of 31.15 A/m^2, regardless of dose or dye

Main Effects Plot for Color Removal
Data Means
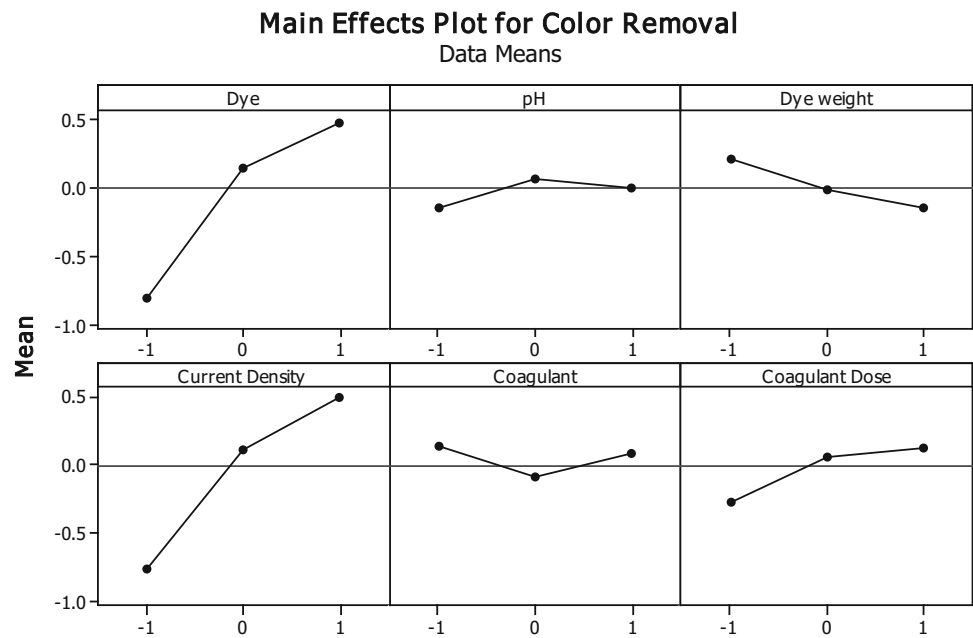
weight, treatment ranged from 95.78 to 97.76 %. Ferric sulfate addition did not have any major trends for removal of NGB or Acid Yellow 11. Decolorization of NGB was greater than 95 % for all runs. It is important to note that the properties of dyes can make certain dyes more susceptible to degradation as compared to others. Finally, at an initial pH of 9, an increase in current density improved treatment for Acid Yellow 11 as treatment was 87.05 % at 18.69 A/m^2 as compared to 92.19 % at 31.15 A/m^2.

Analysis of experimental parameters

The chosen 4–9 pH range within this experiment was suitable for high color removal using aluminum electrodes (Alinsafi et al. 2005). It has been shown that current density is the most prevalent factor within electrocoagulation treatment (Tir and Moulai-Mostefa 2008; Alinsafi et al. 2005). This is because the current density controls not only the amount of coagulant produced, but also the amount of hydrogen gas bubbles formed to allow for the wastewater to aggregate at the top and separate (Tir and Moulai-Mostefa 2008).

The results from this experiment evaluated the variation of three different dye weights, the pH, and addition of three different coagulants at three different doses. The main effects plot (Fig. 4) emphasizes the effects of varying each parameter and its effect on treatment performance. For example, current density and the type of dye had the biggest impact on the treatment performance. As stated in the previous section, the literature has supported this viewpoint, as many authors have found current density as a major influence on the treatment performance, along with

the type of dye when comparing different types of dyes. The moderate r^2 value (72.88 %) can be attributed to a possible variable that has not been considered, such as treatment time, to further explain the model.

Conclusion

The results of this experiment prove that the combined treatment method is capable of treating diverse textile wastewaters in a very short retention time. Electrocoagulation with chemical coagulants was capable of decolorizing dye wastewater between 61.75 and 99.42 % for Acid Yellow 11, Acid Orange 7, and Naphthol Green B. For Acid Yellow 11, the optimum treatment occurred when the pH was 7, dye weight of 32.5 mg, current density 31.15 A/m^2, and ferric sulfate dose of 10 mg/L (93.65 %); Acid Orange 7 had a pH of 9, dye weight of 20 mg, current density of 24.92 A/m^2, and ferric sulfate dose of 10 mg/L (98.82 %); and Napthhol Green B has an optimum pH of 7, dye weight of 32.5 mg, current density of 18.69 A/m^2, and a coagulant alum with a dose of 10 mg/L (97.35 %). From the statistical analysis, it was found that the type of dye, current density, and the interaction between initial pH and current density were statistically significant while dye weight, initial pH, coagulant dose, and coagulant were least important in dye decolorization.

Appendix

See Table 5.

Table 5 Treatment of textile wastewater by electrocoagulation using RSM

Dye(s)	Varying parameters	References
Yellow brown 2GL	Current density	Bhatnagar et al. (2014)
	NaCl concentration	
	Time	
	pH	
Methyl orange	Cell voltage	Pi et al. (2014)
	Time	
	Current density	
	Cell voltage	
	Electrode distance	
	Cyclic period of electrode reversal	
Reactive blue 19	pH	Taheri et al. (2013)
	Dye concentration	
	Concentration	
	Time	
	Powdered activated carbon (PAC) dose	
C.I. reactive red 43	Current density	Amani-Ghadim et al. (2013)
	Electrolysis time	
	pH	
	Chloride concentration	
Acid black 172	pH	Taheri et al. (2012)
	Dye concentration	
	Current	
	Reaction time	
C.I. acid red 14	Current density	Aleboyeh et al. (2008)
	Electrolysis time	
	pH	
Levafix blue CA	Pollution load	Körbahti and Tanyolaç (2008)
	Voltage	
	Electrolyte concentration	
	Reaction temperature	
	Reaction time	
Levafix blue CA	Dye concentration	Körbahti (2007)
Levafix red CA	Current density	
Levafix yellow CA	Electrolyte concentration	
	Electrolysis time	
Drimarene K2LR CDG Blue	Electrolysis time	Alinsafi et al. (2005)
	pH	
	Current density	
Remazol brilliant orange G	Electrolysis time	Gürses et al. (2002)
Remazol brilliant yellow GL	Cell voltage	
Remazol red RB	Current Density	
Remazol blue 3R	Mixing rate	
Acid blue 113	Concentration	Murugan et al. (2009)
	pH	
	Current density	
	Electrolysis time	

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