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Taguchi approach for electrocoagulation for treatment of methyl red dye from textile wastewater by using different connection electrodes

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ABSTRACT

Electrocoagulation (EC) has displayed great potential as an effective and environmentally friendly method to remove dyes from wastewater. This review summarizes the recent developments of dye removal in the EC process including the effects of primary operating parameters, optimization of the EC performance based on Taguchi experimental design (TED). In this study, TED method was applied to determine the optimum operating conditions for the treatment of textile wastewater by electrocoagulation with iron-aluminum and aluminum-iron electrode single. The experimental parameters investigated were electrode connection type; initial concentration: (10–100 ppm); initial pH: (3–11); electrolysis time: (5–60 min); current: (0.2–1 Å); and distance between electrodes: (0.5–2.5 cm). These parameters were varied at five levels to see their effects on the removal efficiency. The results show that the electrocoagulation using Fe-Al electrode single with different connection modes was able to treat the dye wastewater. The maximum color removal percentage (99.5828%) with optimum parameter initial concentration 10 ppm; initial pH: 11; electrolysis time: 60 min; current: 1 Å; and distance between electrodes: 2.5 cm. This review will contribute to a deeper understanding of the Iraqi government to adopt efficient and adequate methods for removing dyes.

Keywords: Methyl red dyes; Electrocoagulation; Taguchi experimental design (TED); Identical electrodes (IE)

1. Introduction

Water is one of the most significant natural resources for living beings on the planet; it is an essential component of life. Freshwater, on the other hand, is scarce in many parts of the planet [1]. Every day, 6,000 people, mostly children under the age of five, die from water-related diseases, according to the United Nations World Water Development Reports. Furthermore, over a billion people do not have access to safe drinking water [1]. As a result, many countries around the world, particularly developing ones, have placed a high priority on protecting freshwater from contamination.

The main sources of contaminated water are industrial, agricultural, and residential activities. In recent decades, the number of dangerous chemicals released into surface waters has grown, rendering it unusable effluent [2].

Many industries that use dyes, such as textiles, printing, paper and pulp, dying, pigments, leather, plastics, rubber, food, and cosmetics, all of which use large amounts of water, produce various toxic organic compounds that are difficult to treat chemically or biologically and can be very dangerous to environmental life, and which are released to the surface of various bodies of waters causing wastewater [3,4].

Textile industries applied one important process called the dyeing process which consumes large quantities of

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water and generate a considerable amount of wastewater containing suspended particles, high chemical oxygen demand concentration, biological oxygen demand concentration, high total organic carbon levels, highly colored synthetic dyes, total suspended solids, and high pH as well as hard degradation materials [5–7]. So management of all types of wastewater can play an important role in the protection of public health and the environment [8].

Dyeing is the technique of imparting color to textiles such as fibers. Dyeing is usually done in a private solution containing dyes and other chemical components. Following dyeing, dye molecules form an uncut chemical link with fiber molecules. Temperature and time control has a significant impact on dyeing [9].

Textile effluent contains a substantial amount of dye, making it one of the most contaminated wastewater types [10]. The direct release of textile effluent into the water supply pollutes the water, affecting marine life (aquatic plants, microbes, fish, and mammals) [11].

There are many problems with dyes such as:

1. Their reflection and absorption of sunlight entering the water is the main environmental problem for dyes. The absorption of light minimizes the photosynthetic behavior of algae and significantly affects the food chain.
2. Due to their brilliance, the presence of very small concentrations of dyes in water (less than 1 mg/L) is highly evident.
3. Due to photostability and high thermal levels, dyes can stay in the environment for a prolonged period.
4. Many dyes and their decomposition products are carcinogenic and death [12].

One of the toxins in water is methyl red. It is used in the cotton industry, which accounts for 50% of the fiber used worldwide. Thanks to their bright colors, great colorfastness, and ease of use, they are widely used in the textile industry [13]. Methyl red, also referred to as C.I. Acid red 2 is an indicator dye that, in acidic solutions, turns red. It is an azo dye and a crystalline powder with a dark red hue. Methyl red (MR) is a mono-azo dye widely used in laboratory experiments, textiles, and other commercial items, but may cause sensitization of the eyes and skin [14] and inflammation of the pharyngeal or digestive tract if inhaled or swallowed. In late, there has been rising interest in developing low-cost means of reducing the amount of MR in wastewater before discharging into the water body, if not fully eliminating it [15].

Methyl red is a pH indicator; it turns pink/red at 4.4 pH and lower, yellow at 6.2 pH and higher, and orange in between as shown in Fig. 1.

Adsorption, ion-exchange, membrane filtration, nanofiltration, photocatalysis, advanced oxidation processes, coagulation, and electrocoagulation methods have all been used to remove colors from wastewater [11].

Despite these technologies being widely accepted and proven processes, practical applications appeared with some restricted and certain limitations due to technical and economic constraints like complex mechanisms, higher spending chemicals, higher costs, and production of large amounts of sludge [16].

The most common plate electrodes in the electrocoagulation process are iron and aluminum, and these two materials have been extensively employed to treat wastewater [17]. The type of coagulant produced was determined by the electrode materials employed. This coagulant has an impact on coagulation and performance processes [18]. The metal electrodes are where the most critical reactions take place. As depicted, anode and cathode reactions for aluminum and iron [17].

Anodic reactions:

Iron electrode:

$$\text{Anode: Fe} \rightarrow \text{Fe}^{2+} + 2\text{e} \tag{1}$$

$$\text{Cathode: } 2\text{H}_2\text{O} + 2\text{e} \rightarrow \text{H}_2 + 2\text{OH}^- \tag{2}$$

$$\text{Overall: Fe}^{2+} + 2\text{H}_2\text{O} \rightarrow \text{H}_2 + \text{Fe}(\text{OH})_2 \tag{3}$$

OR

$$\text{Anode: Fe} \rightarrow \text{Fe}^{3+} + 3\text{e} \tag{4}$$

$$\text{Cathode: } 3\text{H}_2\text{O} + 3\text{e} \rightarrow \frac{3}{2}\text{H}_2 + 3\text{OH}^- \tag{5}$$

$$\text{Overall: Fe}^{3+} + 3\text{H}_2\text{O} \rightarrow \frac{3}{2}\text{H}_2 + \text{Fe}(\text{OH})_3 \tag{6}$$

Aluminum electrode:

$$\text{Anode: Al} \rightarrow \text{Al}^{3+} + 3\text{e} \tag{7}$$

$$\text{Cathode: } 3\text{H}_2\text{O} + 3\text{e} \rightarrow \frac{3}{2}\text{H}_2 + 3\text{OH}^- \tag{8}$$

$$\text{Overall: Al}^{3+} + 3\text{H}_2\text{O} \rightarrow \frac{3}{2}\text{H}_2 + \text{Al}(\text{OH})_3 \tag{9}$$

In general:

$$\text{Anode: M} \rightarrow \text{Mn}^+ + \text{ne} \tag{10}$$

$$\text{Cathode: } 2\text{H}_2\text{O} + 2\text{e} \rightarrow \text{H}_2 + 2\text{OH} \tag{11}$$

$$\text{Overall: Mn}^+ + \text{nH}_2\text{O} \rightarrow \text{nH}^+ + \text{M}(\text{OH})\text{n} \tag{12}$$

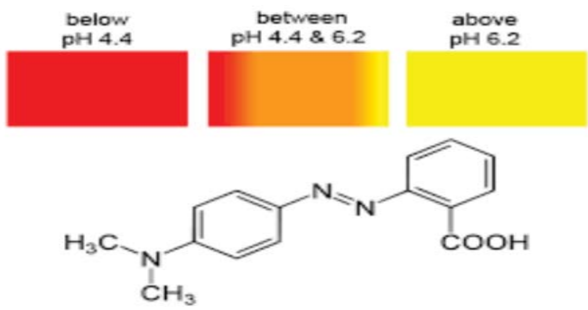


Fig. 1. Methyl red is an indicator that changes color with pH.

The percentage of discoloration was determined using the following formula:

$$\text{Abatement\%} = \frac{C_o - C_{ap}}{C_o} \times 100 \quad (13)$$

where C_o: is the initial concentration of the solution in milliliters per liter, and C_{ap}: is the concentration after treatment in milliliters per liter.

In this method the electrocoagulation (EC) research by two items first is Fe-Al, the second is Al-Fe. The statistical techniques are adopted here design of the experiment using Taguchi experimental design (TED) and analysis by signal-to-noise (S/N) ratio and the tested using analysis of variance (ANOVA) [19,20].

The basic goal encompasses a study using comparing the connection between different electrodes of pure metal to determine which method is more effective by measuring the percentage of the process by which these red dyes are removed from industrial water polluted. The research tests the efficacy of electrocoagulation to extract methyl red dye and to study the effects of different parameters of the experiments such as initial concentration, pH, current density, electrolysis time, the distance between the electrodes and investigate the possible use of TED method technique [21], and find the optimum conditions for improved electrocoagulation efficiency in methyl red dye removal [22]. The electrocoagulation method with methyl red dye was used in a mathematical model with the participation of five factors and five levels instead of the traditional method, and this was not done in previous research.

The novelty of the work design of an electrocoagulation system suitable for dyes removal is by using various pure metal electrodes (Al, Fe) by different strapping and comparing them to determine the effective method, investigating the electrocoagulation efficiency by studying the effect operating conditions by using five parameters and each factor have five levels such as initial methyl red concentration, pH, current, electrolysis time and the distance between the electrodes, selecting the optimum operating condition used for enhancing the efficiency of the electrocoagulation process in addition to using special search dimensions, and this was not done in previous researches.

The mechanism of the electrochemical method is well-known in aquatic systems. This method includes three conceivable mechanisms, that is, electrocoagulation, electroflotation, and electro-oxidation.

The coagulation material is generated at the site by oxidation of the anode's electrolysis with an electrical current between two electrodes in EC. The Fe(OH)*n* and Al(OH)*n* formation with $n = 2$ or 3 is emitted at the anode by using an iron and aluminum anode. Several successive stages are stated by the current theory of EC [23].

A. Oxidation/reduction reactions at the surface of the electrodes.
B. Generation of coagulation agents in the aqueous solution.
C. Adsorption of soluble or colloidal particles of pollutant on the coagulant and removal by sedimentation and/or floatation.

The three stages A, B, and C of the electrocoagulation process are shown in Fig. 2. Metal ions generated at the anode from the dissolving of metal, which directly hydrolyze to polymeric metal hydroxides that are excellent coagulating agents. M metal anodes are used to produce polymeric hydroxides continuously in the anode environment (Fig. 2A). Coagulation occurs when these metal cations combine with the negative particles carried to the anode by electrical movement as illustrated in Fig. 2B [24]. Pollutants in the wastewater stream are treated either by chemical reactions and precipitation or by physical and chemical attachment to colloidal materials resulting from electrode corrosion. Then pollutants are removed by electroflotation, sedimentation, and filtration (Fig. 2C) [25].

EC can treat contaminated water and lower total dissolved solids levels vs. the conventional chemical coagulation process. The lack of significant amounts of chemical additives contributes to this low concentration [26].

EC produces very little sludge and is non-hazardous. Since pH adjustment is the only aspect of EC that requires the addition of chemicals, there is not much to add to the volume of any sludge produced. Also, although most electrodes are sacrificial, they do not oxidize at a rate that adds significant amounts to sludge [17].

2. Material and methods

Methyl red (MR) is an azo dye (known as phenyl}diazenyl]benzoic acid (2-{(E)-[4-(dimethylamino)phenyl]diazenyl}benzoic acid) and is a pH indicator, is yellow at pH greater than 6.2, red at pH below 4.4, and or has been chosen as a model system because of its intense aqueous system color and poor biodegradability due to benzene rings.

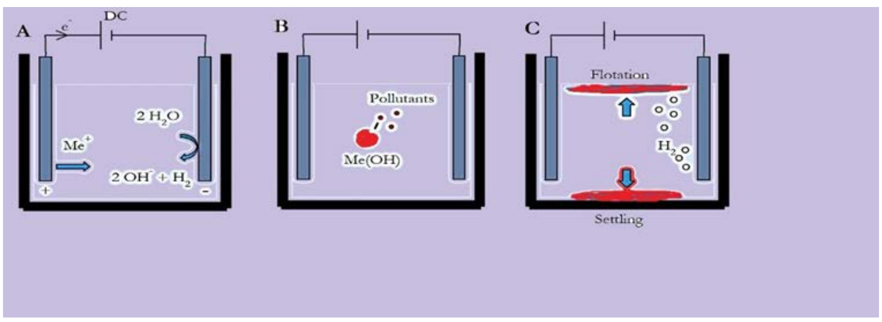


Fig. 2. Mechanism of electrocoagulation [25].

The chemical formula of methyl red is $C_{15}H_{15}N_3O_2$ the properties of methylene blue are listed in Table 1 [27].

2.1. Iron electrodes

The iron plates that were used as electrodes in this research were purchased from local markets. Table 2 analysis of iron electrode properties (Ibn Sina State Company) in Iraq-Baghdad. As shown in the table, the characteristics of the iron plate examined by the State Inspection and Engineering Rehabilitation Company (Ibn Sina State Company) in Iraq are:

2.2. Aluminum electrodes

The aluminum plates used as electrodes in this study were obtained from local markets. As shown in the table, the characteristics of the iron plate examined by the State Inspection and Engineering Rehabilitation Company (Ibn Sina State Company) in Iraq are Table 3. Table 3 analysis of iron electrode properties (Ibn Sina State Company) in Iraq-Baghdad.

2.3. Chemicals

The list of the chemicals used in this study are presented in Table 4.

2.4. Instruments and apparatus

Table 5 instruments and apparatus (Engineering Technical College, Middle Technical University, Baghdad, Iraq)

2.5. Experimental set-up of an electrocoagulation process

The electrocoagulation process uses two electrodes made of aluminum and iron. When the cell is connected in the first stage, the iron is the anode pole, while the aluminum represents the cathode pole (Fe-Al). In the second stage, the cell is reversed (Al-Fe) by digital power supply (Tp-1305EC, 30V/5A) constant current is applied to the electrodes through a wire of (0.2–1 Å) on a fixed area of electrode 150 cm^2, by current density range (0.0026–0.013 A/cm^2). the volume of the cell is 1.8 cm^3. By the spectrum device observation of the absorbance spectrum is carried out in the range of 340–600 nm. The absorbance spectrum indicator solution has a peak at a wavelength of 440 nm for pH 6.3 and 404 nm for pH 7.1.as shown in Fig. 3.

Table 4
Chemicals

Compound	Chemical formula	Specification	Company
Hydraulic acid	HCl	Concentration: 37% w/v Molarity: 12.7 mol/dcm Purity: >95%	Thomas Baker (Chemicals) Limited 4/86, Baharat Mahal, Marine Drive, Mumbai-400 002, India
Sodium hydroxide	NaOH	M.W.: 40.01 g/mol Assay: 98%	Sigma-Aldrich (It is headquartered in Burlington, Massachusetts, United States)
Sodium chloride	NaCl	M.W.: 58.44 g/mol Minimum assay: 99.5%	S.D. Fine Chem Limited Company, Mumbai, India

3. Results and discussion

3.1. Taguchi experimental design

The primary objective behind the Taguchi method is to determine the orthogonal array for experimental design and signal-to-noise ratio for quality. Here, the

Table 1
Physical and chemical properties of methyl red dye

Properties	Values
Chemical formula	$C_{15}H_{15}N_3O_2$
Molar mass	269.304 g/mol
Solubility	Solubility in ethanol
Density	0.791 g/cm^3
Melting point	(179°C–182°C)
UV-Vis (λ_{max})	420 nm

Table 2
Analysis of iron electrode properties

Composition	wt.%
Fe	98.8
C	0.0994
Mn	0.660
S	0.190
Al	0.103
Others	0.1447

Table 3
Analysis of iron electrode properties

Composition	wt.%
Si	0.078
Fe	0.234
Cu	<0.0005
Mn	0.006
Mg	0.0004
Zn	0.014
Ti	0.004
Al	Balance

Table 5
Instruments and apparatus

No.	Device	Model	Origin
1	Digital power supply	Tp-1305EC, (30V/5A)	Ahl Al Kawther, China
2	Magnetic stirrer	Model LMS-2003D	Daihan Labtech, Korea
3	Drying furnaces	Model RWF, Rapid heating (0–250)	Chamber, Japan
4	pH meter	Model Mi150	Martini, Romania
5	Analytical balance	Model: ABS 220-4, with a sensitivity ± 0.0001 g	Kern, Germany
6	Centrifuge	Model: PLS-03	Gemmy Industrial Corp., Taiwan
7	Visible spectrophotometers	Model FG17211901-049	China

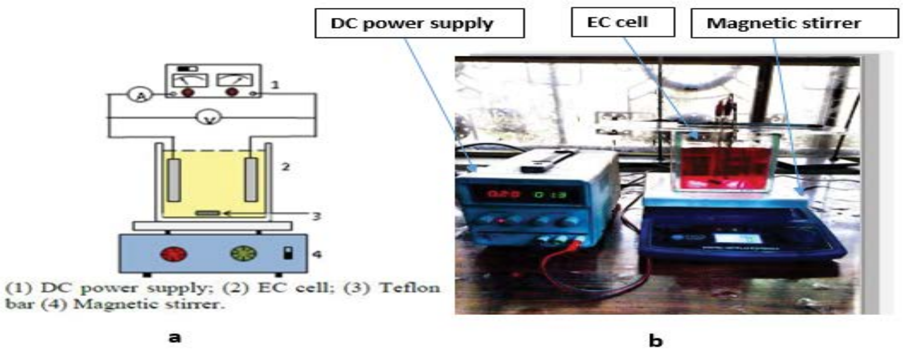


Fig. 3. Simplified experimental set-up of an electrocoagulation cell [18].

orthogonal array consists of controllable factors and experimental combinations of equal probability [28]. The controllable factors considered in this study were pH, time, current, and distance as shown in Table 6.

3.1.1. Residuals normal probability plot for the reduced first model

One of the essential hypotheses for the statistical analysis of the experimental data is their normal distribution. The normal possibility for studentized residuals is elaborated in Figs. 1 and 2. The distribution of residuals is normal if the experimental data follow a straight line. The experimental points in the Fe-Al case are normally distributed with no outliers and located on the normal line spread out between −10 and +10 studentized residuals (Fig. 4). The experimental points in the Al-Fe case are normally distributed with no outliers and located on the normal line spread out between −5 and +5 studentized residuals (Fig. 5).

3.1.2. Signal-to-noise ratio

The S/N was used to measure both the mean value (named "signal" represents the desirable effect) and the standard deviation (named "noise" represents the undesirable effect) of a set of data. Higher S/N ratios are desirable in dye removal. The S/N ratio used for this type of response is defined as follows [29,30].

$$\frac{S}{N} = 10\log_{10}\left[\frac{1}{n}\sum\frac{1}{Y_i^2}\right] \quad (14)$$

Table 6
Orthogonal array for experimental design

Level	Factor				
	I.c	pH	Time	Current	Distance
1	10	3	5	0.2	0.5
2	25	5	10	0.25	1
3	50	7	20	0.5	1.5
4	75	9	40	0.75	2
5	100	11	60	1	2.5

The results of the S/N ratio for various designed experiments after ANOVA calculations are shown in Figs. 6 and 7 in EC by Fe-Al, and Al-Fe, respectively.

Each operational factor had five different conditions (indicated by levels 1, 2, 3, 4, and 5, respectively, as commonly done in the Taguchi method) can be seen in (Table 1). The quality characteristics. In Taguchi method is known as larger-the-better, nominal-the-best, and smaller-the-better. As the objective of this study was to treat dye solution by electrocoagulation, the quality characteristic chosen was larger-the-better.

3.2. Optimization of conditions

The results of the 25 experiments of EC are given in Tables 7 and 8 for Fe-Al and Al-Fe, respectively. The average removals shown in Tables 7 and 8 was the average of two items. The collected data was analyzed using the computer

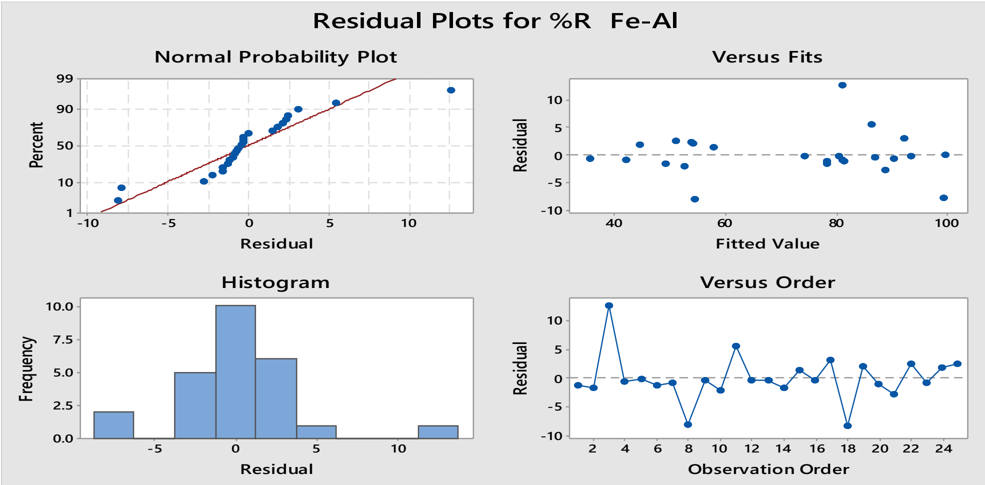


Fig. 4. Normal probability plot of removal dyes by Fe-Al.

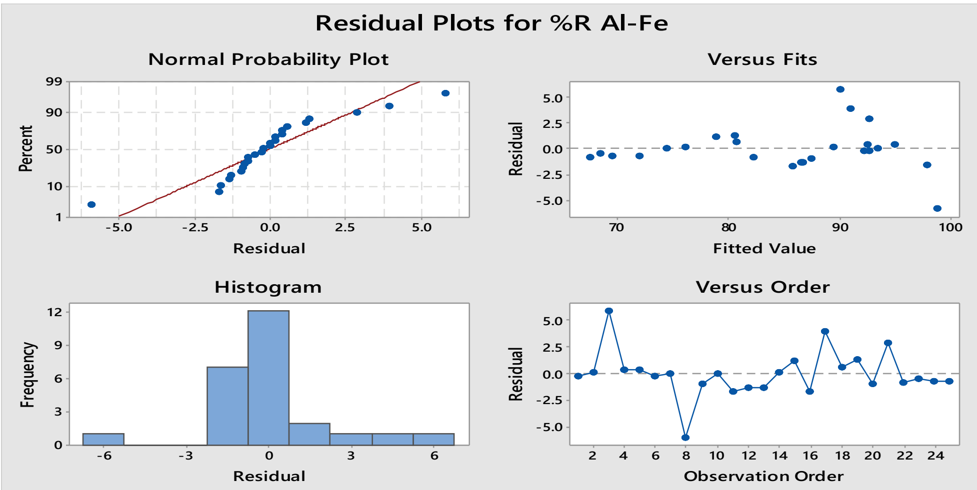


Fig. 5. Normal probability plot of removal dyes by Al-Fe.

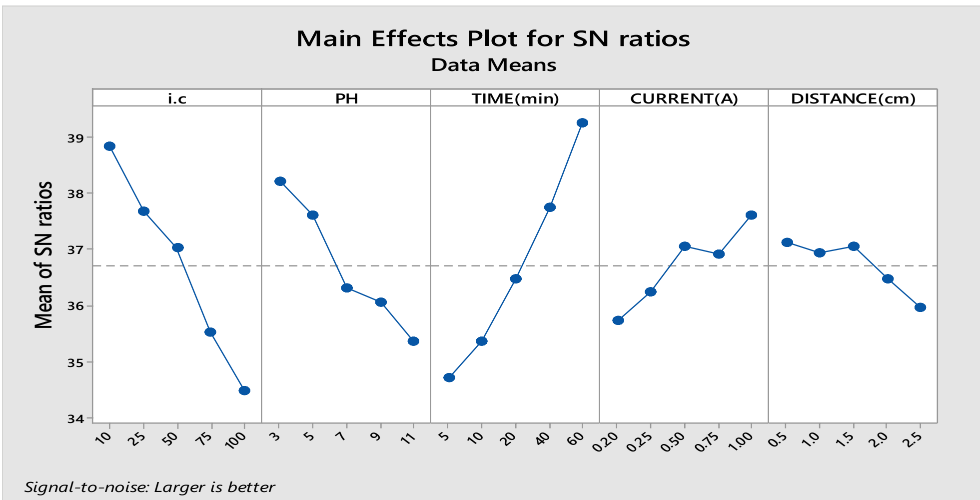


Fig. 6. Main effect plots for S/N ratios of the electrocoagulation process Fe-Al.

Table 7
Dye removal by electrocoagulation through Fe-Al

Exp. No.	I.c (mg/L)	pH	Time (min)	Current (A)	Distance (cm)	%Removal observed	%Removal predicted
1	10	3	5	0.2	0.5	80	81
2	10	5	10	0.25	1	76	78
3	10	7	20	0.5	1.5	94	81
4	10	9	40	0.75	2	89	90
5	10	11	60	1	2.5	99.5	99.6
6	25	3	10	0.5	2	77	78
7	25	5	20	0.75	2.5	80	81
8	25	7	40	1	0.5	91	99
9	25	9	60	0.2	1	93	93
10	25	11	5	0.25	1.5	50	52.7
11	50	3	20	1	1	92	86.5
12	50	5	40	0.2	1.5	80	80.6
13	50	7	60	0.25	2	86.5	86.9
14	50	9	5	0.5	2.5	47.5	49.1
15	50	11	10	0.75	0.5	59	57.8
16	75	3	40	0.25	2.5	74	74
17	75	5	60	0.5	0.5	95	92
18	75	7	5	0.75	1	46	54.5
19	75	9	10	1	1.5	56	54
20	75	11	20	0.2	2	41	42
21	100	3	60	0.75	1.5	85.9	88.7
22	100	5	5	1	2	53	50.9
23	100	7	10	0.2	2.5	35	35.6
24	100	9	20	0.25	0.5	46	44.6
25	100	11	40	0.5	1	56	53.8

Table 8
Dye removal by electrocoagulation through Al-Fe

Exp. No.	I.c (mg/L)	pH	Time (min)	Current (A)	Distance (cm)	%Removal observed	%Removal predicted
1	10	3	5	0.2	0.5	92	92
2	10	5	10	0.25	1	89.5	89
3	10	7	20	0.5	1.5	96	90
4	10	9	40	0.75	2	93	92.5
5	10	11	60	1	2.5	95	95
6	25	3	10	0.5	2	92	92.7
7	25	5	20	0.75	2.5	93	93.
8	25	7	40	1	0.5	92.7	98.7
9	25	9	60	0.2	1	86.5	87.5
10	25	11	5	0.25	1.5	74.5	74.5
11	50	3	20	1	1	96	98
12	50	5	40	0.2	1.5	85	86.7
13	50	7	60	0.25	2	85	86.5
14	50	9	5	0.5	2.5	76	76
15	50	11	10	0.75	0.5	80	78.8
16	75	3	40	0.25	2.5	84	85.7
17	75	5	60	0.5	0.5	95	91
18	75	7	5	0.75	1	81	80
19	75	9	10	1	1.5	81.8	80.5
20	75	11	20	0.2	2	66.7	67.6
21	100	3	60	0.75	1.5	95.6	92.7
22	100	5	5	1	2	81	82
23	100	7	10	0.2	2.5	68	68.5
24	100	9	20	0.25	0.5	68.7	69.5
25	100	11	40	0.5	1	71	72

software package program (MINITAB 17) for the evaluation of the effect of each parameter on the optimization criteria. Figs. 5 and 6 illustrate the S/N ratio averages for each factor at the five different levels and the corresponding response variable. As seen in Figs. 4 and 5 the electrode connection type exhibits large variations; therefore, this parameter has a major effect on the EC process. On the other hand, the current density shows the smallest variation than that of the other parameters. This means that the current density parameter has less influence on the EC process. The pick points in each graph indicated that in the (Fe-Al), the best value of that particular parameter was found as 1st level of initiated concentration (10), 1st level of pH (11), 5th of time (60 min), and 3rd level of current (1 Å), 1st level of distance (2.5 cm). In the (Al-Fe), the best value of that particular parameter was found as 3rd level of initiated concentration (10), 1st level of pH (3), 3rd of time (20 min), and 5th level of current (1A), 2nd level of distance (1 cm). However, electrolysis time differs for Fe-Al and Al-Fe in this study. The optimum electrolysis times were determined as 3rd (60 and 20 min) level for Fe-Al and Al-Fe, respectively.

3.3. Analysis of variance

ANOVA results of the data obtained from EC experiments by Fe-Al and Al-Fe are presented in Tables 3 and 4, respectively. The % R final column in Table 4 shows the percent of contribution (PC, %) of each factor to response. The percent of contribution shows the influence of one factor on the total observed variance in the experiments.

Tables 9 and 10 show the ANOVA result for Fe-Al, and Al-Fe, where terminologies such as DF = degree of freedom, SS = sum of the square, MS = mean square, and PC = percent of contribution. The models of removal dyes by EC method by using two items (Fe-Al, Al-Fe) are significant with a *p*-value (0.000), which is less than the level of significance of 0.05. It is very clear from the data in Table 3 that the type of dopants used and the time have more impact. The highest values of SS (4082.8), MS (4082.63), '*F*' (207.29),

Table 9
Analysis of variance of the data resulted from experimental design for Fe-Al

Source	DF	Adj. SS	Adj. MS	*F*-value	*p*-value	PC%	Rank
Regression	5	9,291.8	1,858.35	94.36	0.000		
I.c (ppm)	1	3,253.1	3,253.05	165.17	0.000	35.01	2
pH	1	1,326.4	1,326.41	67.35	0.000	14.28	3
Time (min)	1	4,082.6	4,082.63	207.29	0.000	43.94	1
Current (Å)	1	470.9	470.86	23.91	0.000	5.07	4
Distance (cm)	1	158.8	158.80	8.06	0.010	1.70	5
Error	19	374.2	19.69				
Total	24	9,666.0				100	

Model summary

S	*R*-sq.	*R*-sq. (Adj.)	*R*-sq. (Pred.)
4.43	96.13%	95.11%	94.20%

%R Fe-Al = 88.41 – 0.3494I.c – 2.575 pH + 0.6265 TIME (min) + 14.36 CURRENT (A) – 3.56 DISTANCE (cm)

Table 10
Analysis of variance of the data resulted from experimental design for Al-Fe

Source	DF	Adj. SS	Adj. MS	*F*-value	*p*-value	PC%	Rank
Regression	5	2,103.31	420.662	72.28	0.000		
I.c (ppm)	1	725.71	725.714	124.70	0.000	34.50	1
pH	1	671.96	671.962	115.46	0.000	31.95	2
Time (min)	1	303.97	303.969	52.23	0.000	14.45	4
Current (Å)	1	385.50	385.502	66.24	0.000	18.33	3
Distance (cm)	1	16.16	16.160	2.78	0.112	0.768	5
Error	19	110.58	5.820				
Total	24	2,213.89				100	

Model summary

S	*R*-sq.	*R*-sq. (adj)	*R*-sq. (pred)
2.41244	95.01%	93.69%	91.69%

%R Al-Fe = 96.42 – 0.1650I.c – 1.833 pH + 0.1710 TIME (min) + 12.99 CURRENT (A) – 1.137 DISTANCE (cm) and PC% (43.94), whereas other factors (pH, current) contribution is very negligible, while the distance has a contributing factor of 1.70%, whereas the current factor is in attaining lower. Table 4 that the type of dopants used and the I.c have more impact. The highest values of SS (725.71), MS (725.714), '*F*' (124.70), and PC% (34.50), whereas other factors' (time, current) contribution is very negligible, while the current has a contributing factor of 4.90%, whereas the distance factor is in attaining lower.

The models of removal of dyes by EC method through items of Fe-Al, and Al-Fe show the value of the determination coefficient (R^2) was 96.13%, 95.01% greater than 95.11%, 93.69%, respectively, which implied that 96.13%, 95.01% of the variations could be explained by the fitted models.

4. Effectiveness of factors on removal dyes by electrocoagulation through Fe-Al

The controllable factors considered in using Fe at EC to remove dyes in this study were pH, time, current, and distance S/N of every factor as:

4.1. Effect of I.c

According to Fig. 6, the best S/N for Fe-Al was found at an I.c of 10. When the I.c was decreased from 100 to 10, the S/N ratio decreased significantly Table 11.

In Fig. 7 the best S/N for Al-Fe was found at I.c of 50. When the I.c was decreased from 100 to 50, the S/N ratio decreased significantly (Table 12).

4.2. Effect of pH

According to Fig. 6, the best S/N for Fe-Al was found at a pH of 11. When the pH was decreased from 11 to 7, the S/N ratio decreased significantly (Table 11).

In Fig. 7 the best S/N for Al-Fe was found at pH of 3. When the pH was decreased from 11 to 3, the S/N ratio decreased significantly (Table 12).

4.3. Effect of time

Fig. 6 shows that the best S/N for Fe-Al was found at the time of 60. When the time was increased from 5 to 60, the S/N ratio decreased significantly (Table 11).

Table 11
Analysis of variance results for experimental responses in electrocoagulation for Fe-Al

Source	DF	Seq. SS	Adj. SS	Adj. MS	*F*-value	*p*-value	PC%	Rank
I.c (ppm)	4	60.077	60.077	15.0193	19.75	0.007	35.24	2
pH	4	27.552	27.552	6.8879	9.06	0.028	16.16	3
Time (min)	4	67.422	67.422	16.8556	22.17	0.005	39.55	1
Current (Å)	4	10.616	10.616	2.6540	3.49	0.127	6.23	4
Distance (cm)	4	4.799	4.799	1.1996	1.58	0.335	2.82	5
Residual error	4	3.042	3.042	0.7604				
Total	24	173.507					100	

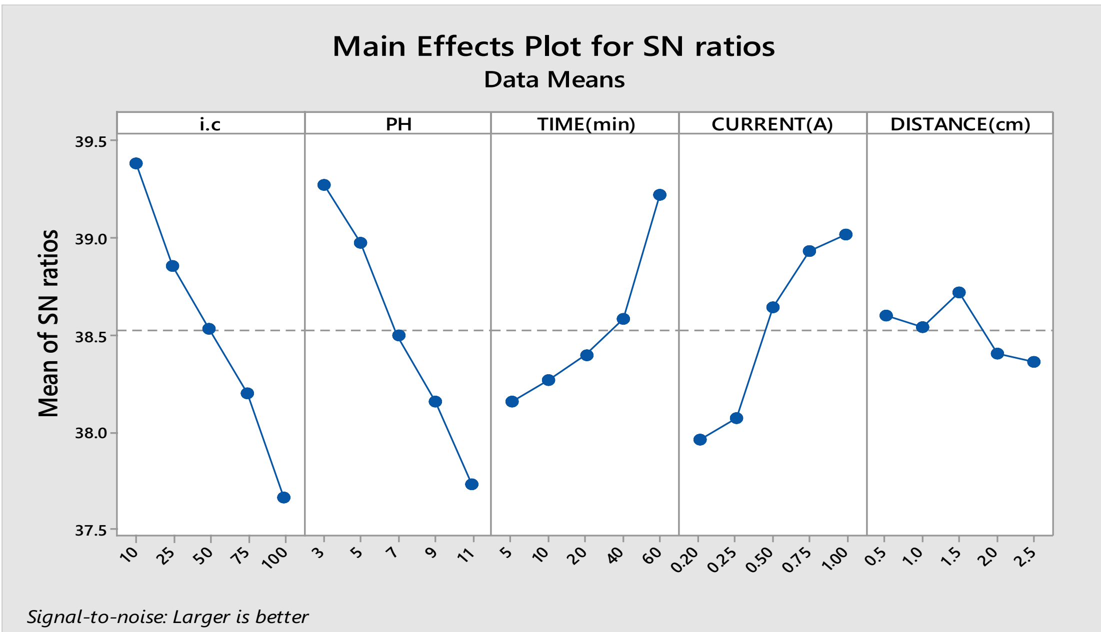


Fig. 7. Main effect plots for S/N ratios of the electrocoagulation process Al-Fe.

Fig. 7 shows that the best S/N for Al-Fe was found at the time of 5. When the time was increased from 20 to 60, the S/N ratio decreased significantly (Table 12).

4.4. Effect of current

Based on Fig. 6 the best S/N for Fe-Al was found at a current of 1. When the current was increased from 0.2 to 1, the S/N ratio decreased non-significantly Table 11.

Based on Fig. 7 the best S/N for Al-Fe was found at a current of 1. When the current was increased from 0.2–1, the S/N ratio decreased significantly (Table 12).

4.5. Effect distance

In Fig. 6 the best S/N was found at a distance of 0.5. When the distance was decreased from 2.5 to 0.5, the S/N ratio decreased non-significantly (Table 11).

In Fig. 7 the best S/N was found at a distance of 1. When the distance was decreased from 1 to 0.5, the S/N ratio decreased non-significantly (Table 12).

5. Contour plot analysis for removal of dye

In the contour plot, the highest values of removal dyes by EC in Fe-Al are in the upper right corner of the plot, which corresponds with high values of both I.c and pH. The lowest values of removal dyes by EC in Fe-Al are in the lower-left corner of the plot, which corresponds with low values of both I.c and pH. The three predictors, time, current, and distance, are not displayed in the plot.

In other meaning, Fig. 8a shows the relation between I.c and pH. It showed that in low pH and I.c, a high amount of dye was removed using EC through Fe-Al.

Table 12
Analysis of variance results for experimental responses in electrocoagulation for Al-Fe

Source	DF	Seq. SS	Adj. SS	Adj. MS	*F*-value	*p*-value	PC%	Rank
I.c (ppm)	4	8.5258	8.5258	2.13144	75.67	0.001	34.22	1
pH	4	7.6558	7.6558	1.91394	67.95	0.001	30.73	2
Time (min)	4	3.5409	3.5409	0.88523	31.43	0.003	14.21	4
Current (Å)	4	4.7655	4.7655	1.19139	42.30	0.002	19.13	3
Distance (cm)	4	0.4255	0.4255	0.10637	3.78	0.113	1.71	5
Residual error	4	0.1127	0.1127	0.02817				
Total	24	25.0262					100	

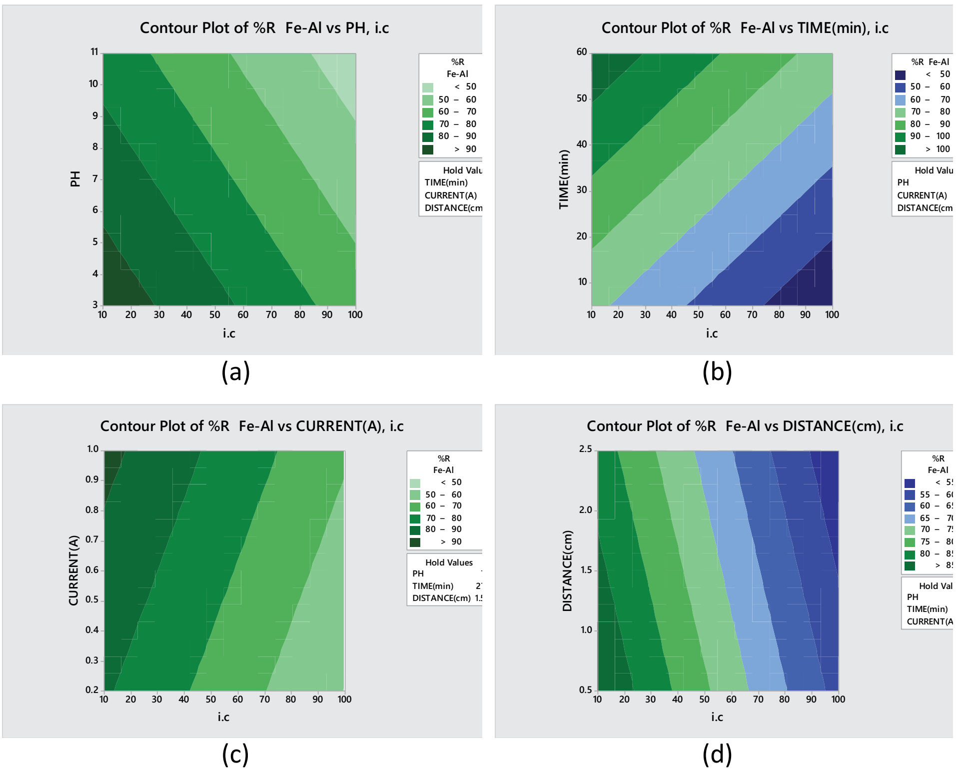


Fig. 8. Contour plots of removal dyes by electrocoagulation through Fe-Al vs. I.c with different study factors.

Fig. 8b shows the relation between I.c and time. It showed that in higher time and low I.c, a high amount of dye was removed using EC through Fe-Al. Similarly, Fig. 8c shows the relation between I.c and current. It showed that in low I.c and high current, a high amount of dye was removed using EC through Fe-Al.

Fig. 8d shows the relation between I.c and distance. It showed that in low I.c and distance, a high amount of dye was removed using EC through Fe-Al.

Fig. 9a shows the relation between pH and time. It showed that in higher time and low pH, a high amount of dye was removed using EC through Fe-Al. Similarly, Fig. 9b shows the relation between pH and current. It showed that in low pH and high current, a high amount of dye was removed using EC through Fe-Al. Fig. 9c shows the relation between pH and distance. It showed that in low pH and distance, a high amount of dye was removed using EC through Fe-Al.

Fig. 10a shows the relation between time and current. It showed that in higher time and current, a high amount of dye was removed using EC through Fe-Al. Similarly, Fig. 10b shows that in higher time and low distance, a high amount of dye was removed using EC through Fe-Al.

Fig. 11 shows the relationship between current and distance. It showed that in higher currents and low distances, a high amount of dye was removed using EC through Fe-Al.

Fig. 12a shows the relation between I.c and pH. It showed that in low pH and I.c, a high amount of dye was removed using EC through Al-Fe. Meanwhile, higher time and current, and low distance had better conditions for dye removal using EC through Al-Fe.

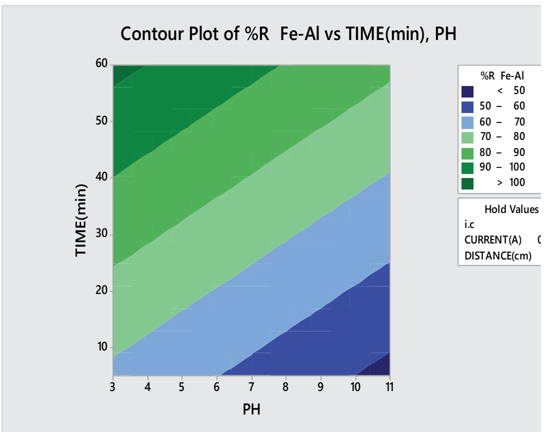
Fig. 12b shows the relation between I.c and time. It showed that in higher time and low I.c, a high amount of dye was removed using EC through Al-Fe. Similarly, Fig. 12c shows the relation between I.c and current. It showed that in higher current and low I.c, a high amount of dye was removed using EC through Al-Fe. Fig. 12d shows the relation between I.c and distance. It showed that in the low distance and I.c, a high amount of dye was removed using EC through Al-Fe.

Fig. 13a shows the relation between pH and time. It showed that in higher time and low pH, a high amount of dye was removed using EC through Al-Fe.

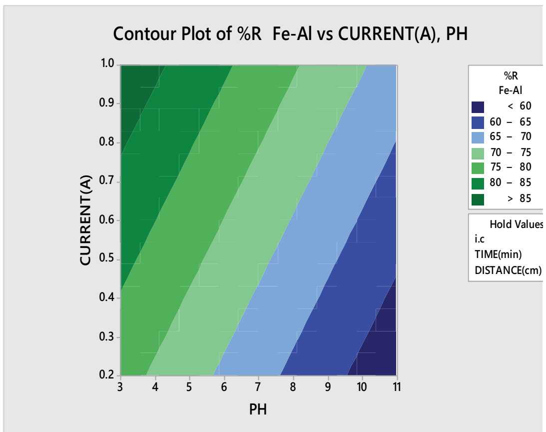
Similarly, Fig. 13b shows the relation between pH and current. It showed that in higher currents and low pH, a high amount of dye was removed using EC through Al-Fe.

Fig. 13c shows the relation between pH and distance. It showed that in low pH and distance, a high amount of dye was removed using EC through Al-Fe.

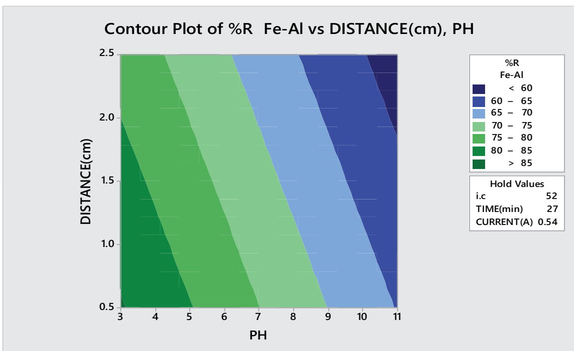
Fig. 14a shows the relation between time and current. It showed that in higher time and high current, a high amount of dye was removed using EC through Al-Fe.



(a)

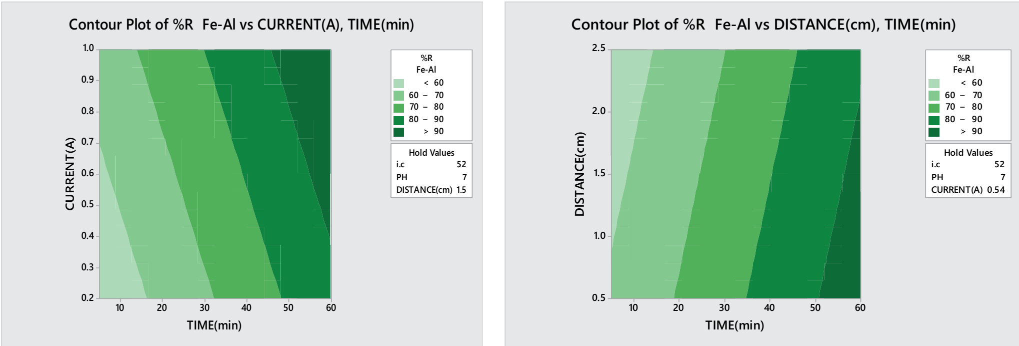


(b)



(c)

Fig. 9. Contour plots for removal dyes by electrocoagulation through Fe-Al vs. pH with different study factors.



(a) (b)

Fig. 10. Contour plots for removal dyes by electrocoagulation through Fe-Al vs. time with different study factors.

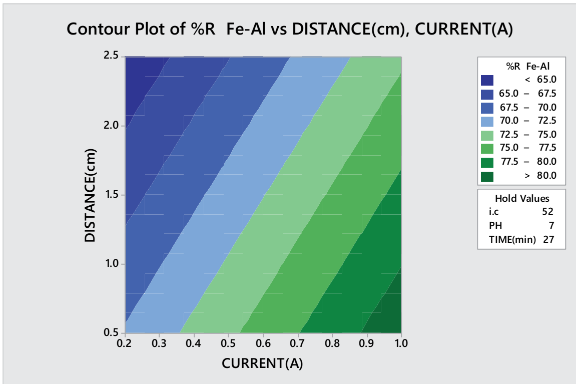
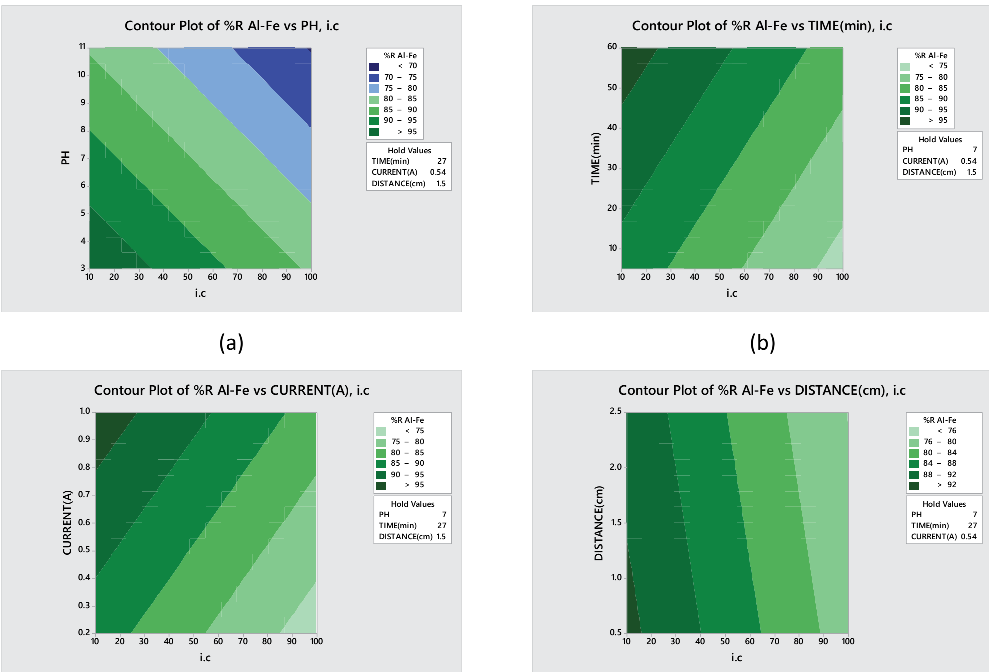


Fig. 11. Contour plots for removal dyes by electrocoagulation through Fe-Al vs. current with distance.



(c) (d)

Fig. 12. Contour plots of removal dyes by electrocoagulation through Al-Fe vs. I.c with different study factors.

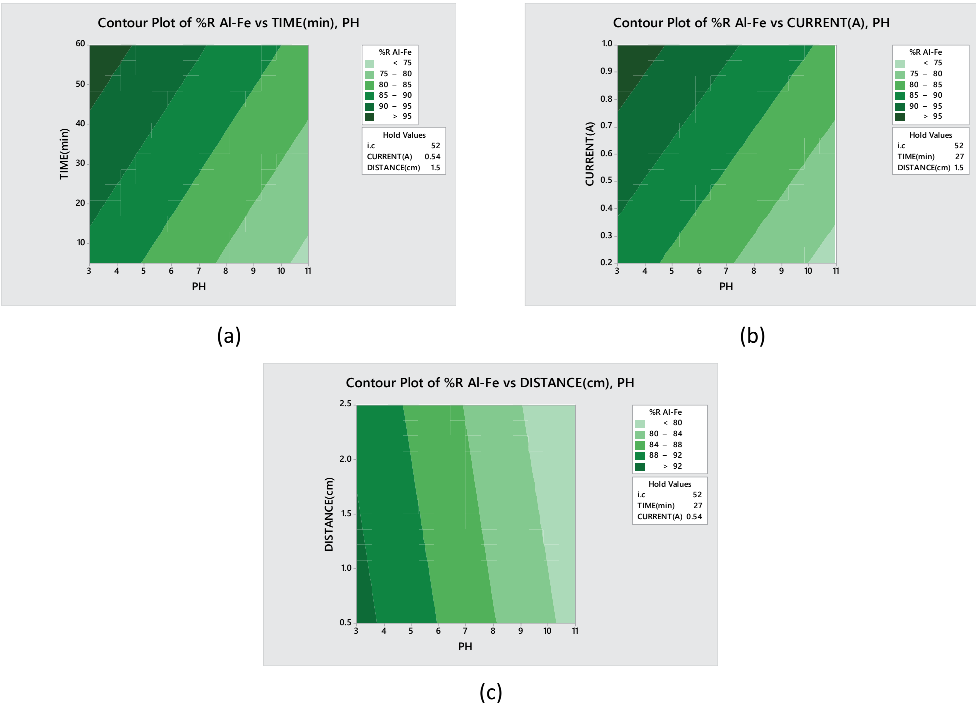


Fig. 13. Contour plots for removal dyes by electrocoagulation through Al-Fe vs. pH with different study factors.

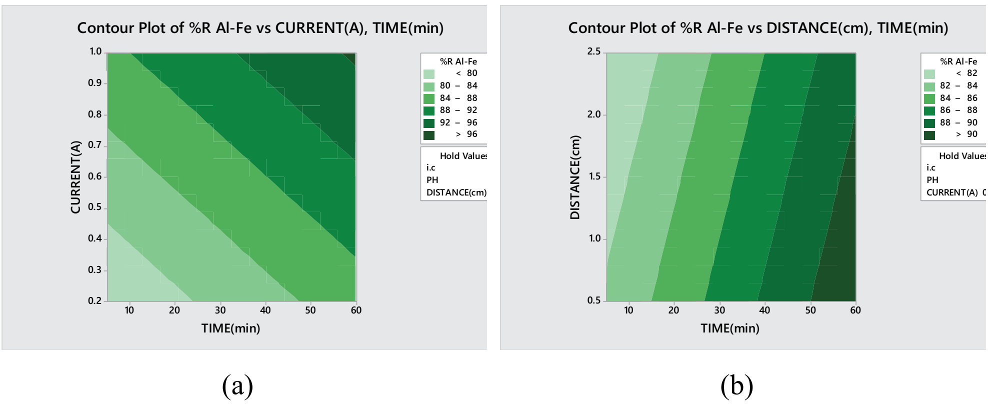


Fig. 14. Contour plots for removal dyes by electrocoagulation through Al-Fe vs. time with different study factors.

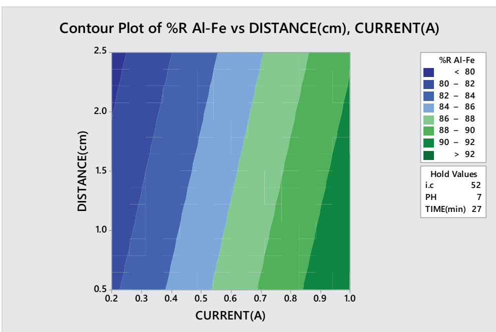


Fig. 15. Contour plots for removal dyes by electrocoagulation through Al-Fe vs. current with distance.

Similarly, Fig. 14b shows that in higher time and low distance, a high amount of dye was removed using EC through Al-Fe.

Fig. 15 shows the relationship between current and distance. It showed that in higher currents and low distances, a high amount of dye was removed using EC through Al-Fe.

6. Conclusion

The experiments were conducted to find how different operational parameters affect methyl red dye removal from textile wastewater. The results confirmed that the removal efficiency of methyl red dye was proportional to the EC through Fe-Al is 99.5828%, I.c: 10, pH: 11, time: 60 min, current: 1, and distance 2.5 cm. % of the contribution of five factors was time is the higher effect in removal dyes for Fe-Al. also. The initial concentration has a higher effect on removing dyes in Al-Fe. It can be concluded that EC by Al-Fe electrodes is less efficient in the performance of Fe-Al electrodes. Furthermore, investigations may determine how this process can affect other types of dye compounds.

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