

# Electrochemical Removal of Methylene Blue from Aqueous Solutions Using Taguchi Experimental Design

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Taguchi optimization method ( $L_{32}$  orthogonal array) was applied as an experimental design to determine optimum conditions for methylene blue dye removal from aqueous solutions by electrocoagulation (EC). Various electrocoagulation parameters such as initial pH, time of electrolysis, concentration of dye, electrodes gap, applied current, solution temperature, amount of supporting electrolyte, design and materials of electrodes were investigated. The results have been analyzed using signal-to-noise (S/N) ratio and analysis of variance (ANOVA). The amount of electrolyte parameter has been found to be the most significant parameter on the color removal. The study shows that the Taguchi's method is suitable to optimize the experiments for dye removal.

*Key words:*

Taguchi method, methylene blue, electrocoagulation, decolorization

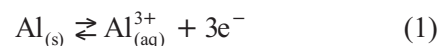
## Introduction

Wastewater from textile industry has a high environmental impact and therefore needs to be treated before being discharged into the environment or reused.<sup>1,2</sup> The treatment methods of dyeing wastewater are biological treatment, chemical coagulation, activated carbon adsorption, ultrafiltration, ozonation and electrocoagulation (EC).<sup>3</sup> EC is a simple and efficient electrochemical method for the purification of many types of water and wastewater. This technique is characterized by its simple equipment, easy operation, and decreased amount of sludge. The coagulant of EC process is generated by electrolytic oxidation of an appropriate anode material that leads, at an appropriate pH, to the insoluble metal hydroxide capable of removing a large variety of pollutants.<sup>4</sup> These metal hydroxide species neutralize the electrostatic charges on suspended solids and oil droplets to facilitate agglomeration or coagulation and resultant separation from the aqueous phase.<sup>5,6</sup> A growing research interest is reported on the treatment of various wastewater types: paper industry wastewater,<sup>7–10</sup> landfill leachate,<sup>11–14</sup> electroplating wastewater,<sup>15</sup> tannery effluent,<sup>16</sup> laundry wastewater,<sup>17,18</sup> latex particles.<sup>19</sup> EC process has also been widely used to decolorize various structurally different dye containing solutions such as disperse, reactive and acidic dyes.<sup>20–25</sup>

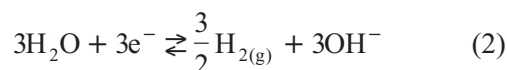
The mechanism of EC is extremely dependent on the chemistry of the aqueous medium, especially

its conductivity. The mechanism of generating ions by EC can be explained with the examples of iron and aluminum, which was used as both the anode and cathode in this study. In the case of aluminium, the main reactions are:<sup>26,27</sup>

anodic reactions:



cathodic reactions:



$\text{Al}^{3+}$  and  $\text{OH}^{-}$  ions generated by electrode reactions (1) and (2) react to form various monomeric species such as  $\text{Al}(\text{OH})^{2+}$ ,  $\text{Al}(\text{OH})_2^{+}$ ,  $\text{Al}(\text{OH})_3^{4+}$ ,  $\text{Al}(\text{OH})_4^{-}$ , and polymeric species such as  $\text{Al}_6(\text{OH})_{15}^{3+}$ ,  $\text{Al}_7(\text{OH})_{17}^{4+}$ ,  $\text{Al}_8(\text{OH})_{20}^{4+}$ ,  $\text{Al}_{13}\text{O}_4(\text{OH})_{24}^{7+}$ ,  $\text{Al}_{13}(\text{OH})_{34}^{5+}$ , which finally transform into  $\text{Al}(\text{OH})_{3(s)}$  according to complex precipitation kinetics.<sup>27–29</sup>

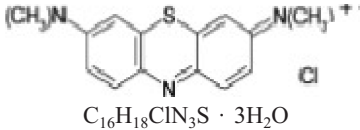


Freshly formed amorphous  $\text{Al}(\text{OH})_{3(s)}$  “sweep flocs” have large surface areas, which is beneficial for a rapid adsorption of soluble organic compounds and trapping of colloidal particles. Finally, these flocs are removed easily from aqueous medium by sedimentation or  $\text{H}_2$  flotation.<sup>26,28</sup>

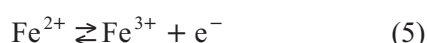
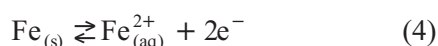
When iron is used as anodes, the following main reactions occur in the EC at different pH values:

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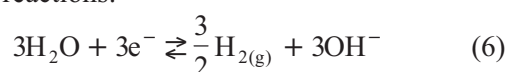
Table 1 – Properties of MB dye

Dye	Structure	$\lambda_{\max}$ (nm)	M.W.	C.I.
Methylene blue [3,7-bis(dimethylamino) phenazathionium chloride trihydrate]	 $C_{16}H_{18}ClN_3S \cdot 3H_2O$	658	373.9	52015

anodic reactions:



cathodic reactions:



Similarly, ferric ions generated by electrochemical oxidation of iron electrode may form monomeric species  $Fe(OH)^{2+}$ ,  $Fe(OH)_2^{+}$ ,  $Fe(OH)_3^{+}$ ,  $Fe(H_2O)_5(OH)^{2+}$ ,  $Fe(H_2O)_4(OH)_2^{+}$ ,  $Fe(OH)_3$ , and  $Fe(OH)_4^{-}$ , and polymeric species  $Fe_2(H_2O)_8(OH)_4^{2+}$ ,  $Fe_2(H_2O)_6(OH)_4^{2+}$ , depending on the pH of the aqueous medium.<sup>28–30</sup>

In the research and development stage, the Taguchi method has been found effective by means of improved productivity, which brings along obtaining high quality items at low costs. Also, this method has been found applicable in a wide range of industrial fields worldwide.<sup>31,32</sup> The main purpose in using statistical experimental design is to provide maximum and reliable information by making fewer possible runs. For this purpose, Taguchi is the preferable technique among statistical experimental design methods since it uses a special design of an orthogonal array (OA) to study the effective parameters with a small number of experiments.<sup>31</sup> This method helps researchers to determine the possible combinations of factors and identify the best combination. However, in industrial settings, it is extremely costly to run a number of experiments to test all combinations. The Taguchi approach developed rules to carry out the experiments, which further simplified and standardized the design of experiments (DOE), along with minimizing the number of factor combinations that would be required to test the factor effects. The greatest difference between the Taguchi methods and classical methods is that in Taguchi's experiments, orthogonal arrays are used to assure the reproduction of the effects of parameters. Another difference is that various types of "signal to noise" (S/N) ratios are used in a Taguchi study in order to measure variability around the target performance.<sup>33–36</sup>

In this study, the color removal from methylene blue solution in an electrocoagulation reactor was investigated. The effect of experimental parameters

such as initial pH, time of electrolysis, concentration of dye, electrodes distance, current density, solution temperature, amount of electrolyte, design and materials of electrodes on the color removal were investigated using an  $L_{32}$  ( $7^4 \times 1^2$ ) orthogonal array. The Taguchi experimental design method was used to determine optimum color removal conditions for maximizing the treated aqueous solutions.

## Materials and methods

### Experimental system

All the chemicals used in this study were reagent grade. Methylene blue (MB) was prepared from Merck (Darmstadt, Germany). The properties of MB are shown in Table 1. Dye solutions in purified water were prepared having 50, 100, 150 and 200 mg  $L^{-1}$  individual concentration. To increase solution conductivity and study the effect of electrolyte amount, NaCl was added to the solutions. Concentrations of NaCl in the solutions were 2, 4, 8, 12 g  $L^{-1}$ . The electrocoagulation experiments were conducted in a batch reactor (Fig. 1). Used were 5 cm  $\times$  5 cm rectangular iron and aluminum plate electrode pairs having submerged surface area of 25 cm<sup>2</sup>, and 1 cm and 2 cm electrode spacing. The electrodes were disposed vertically in the cell. The volume of treated aqueous dye solutions was 1000 mL in an 1100 mL glass electrolytic reactor.

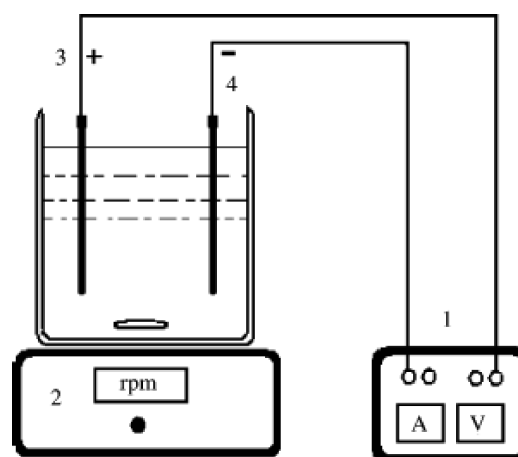


Fig. 1 – Schematic diagram of the electrocoagulation cell: (1) digital DC power; (2) magnetic bar-stirrer; (3) anode; (4) cathode

DC potential was applied from DC power supply (Micro 7145, China). Cell current density was varied by changing the impressed cell voltage (0.1, 0.5, 1 and 2 A). A Sunwa Electronics multimeter, model-YX-360TR-EB (Japan) was used for measuring the current and the potential between the two electrodes. The solution was agitated with a magnetic stirrer hot plate (Tarsons MC02, India) during electro-processing at 200 rpm. Electrodes were washed with dilute HCl between experiments. Experiments were conducted at variable temperature (25, 40, 50 and 70 °C). The pH was adjusted using 0.2 mol L<sup>-1</sup> NaOH and HCl solutions.

### Chemical analysis

A Shimadzu (S 2000, Japan) spectrophotometer was used for recording the spectra and measuring the absorbance of dye solutions at 658 nm as  $\lambda_{\max}$  and generating the calibration plot. Calibration curves were plotted between absorbance and concentration of the dye solution. The calculation of color removal efficiency after electrocoagulation treatment was performed using this formula:

$$\text{Dry removal efficiency (\%)} = \frac{\gamma_0 - \gamma}{\gamma_0} \cdot 100 \quad (7)$$

where  $\gamma_0$  and  $\gamma$  are concentrations of dye before and after electrocoagulation in mg L<sup>-1</sup>. All the samples were allowed to settle for 30 minutes before any analysis was performed. The conductivity measurement was performed using a conductometer (Hana, HI8733). pH measurements were performed with a Metrohm 691 pH meter using a combined glass electrode.

### Statistical analysis

The parameter design phase of the Taguchi method generally includes the following steps: (1) identify the objective of the experiment; (2) identify the quality characteristic (performance measure) and its measurement systems; (3) identify the factors that may influence the quality characteristic, their levels and possible interactions; (4) select the appropriate OA and assign the factors at their levels to the OA; (5) conduct the test described by the trials in the OA; (6) analysis of the experimental data using the signal-to-noise (S/N) ratio, factor effects and the ANOVA (analysis of variance) to see which factors are statistically significant, and find the optimum levels of factors; (7) verify the optimal design parameters through confirmation experiment.<sup>37</sup>

The Taguchi method uses the S/N ratio to measure the quality characteristic deviating from the desired value. The S/N ratios are different according to the type of characteristic. In the case that the bigger characteristics are better, the S/N ratio is defined as:<sup>38</sup>

$$S/N = -10 \log \left( \frac{1}{n} \sum_{i=1}^n \frac{1}{y_i^2} \right) \quad (8)$$

where  $y_i$  is the characteristic property,  $n$  is the replication number of the experiment. The unit of S/N ratio is decibel (dB), which is frequently used in communication engineering.

An analysis of variance (ANOVA) was applied to the data in order to conduct an analysis of the relative importance of each factor more systematically. ANOVA was performed to see whether the process parameters were statistically significant or not. The following equations were used:

$$SS_T = \left[ \sum_{i=1}^N Y_i^2 \right] - \frac{T^2}{N} \quad (9)$$

$$SS_A = \left[ \sum_{i=1}^{k_A} \left( \frac{A_i^2}{n_{A_i}} \right) \right] - \frac{T^2}{N} \quad (10)$$

$$v_T = N - 1 \quad (11)$$

$$v_A = k_A - 1 \quad (12)$$

$$\sigma_A = \frac{SS_A}{v_A} \quad (13)$$

where  $T$  is the sum of all observations,  $N$  is the total number of observations (in this case 32),  $A_i$  is the sum of observations under the  $A_i$  level,  $n_{A_i}$  is the number of observations under the  $A_i$  level,  $k_A$  is the number of levels of the factor A,  $SS_T$  is the total sum of squares,  $SS_A$  is the sum of squares for factor A (this equation is similar for the factors B and etc),  $v_T$  is the total degrees of freedom,  $v_A$  is the factor A degrees of freedom and finally  $\sigma_A$  is the variance for the factor A.<sup>39</sup>

Table 2 – Variables and their values corresponding to their levels investigated in the experiments

Variables	Levels			
	1	2	3	4
A: Distance of electrodes (cm)	1	2		
B: Temperature (°C)	25	40	50	70
C: pH	4.5	7.3	10	3.1
D: Concentration (mg L <sup>-1</sup> )	50	100	150	200
E: Electrode material and design	Fe, series	Fe, parallel	Al, series	Al, parallel
F: Time of electrolysis (min)	8	12	25	35
G: Current (A)	0.1	0.5	1.0	2.0
H: Amount of electrolyte (NaCl) (g L <sup>-1</sup> )	2	4	8	12

The variables chosen for this investigation were initial pH, time of electrolysis, concentration of dye, electrodes distance, current, solution temperature, amount of electrolyte, electrode material and design. The variables investigated and their levels were summarized in Table 2. In order to opti-

mize the dye-removal efficiency process, experimental parameters and their levels were investigated as shown in Table 3. The experimental design, based on standard OA<sub>32</sub> orthogonal array, was conducted to change the settings of the various process parameters.

Table 3 – Experimental variables, their levels and results of conducted experiments corresponding to L9 experimental plan

Experiment number	Variables and their levels								Dye removal efficiency (%)
	A	B	C	D	E	F	G	H	
1	1	1	1	1	1	1	1	1	34.16
2	1	1	2	2	2	2	2	2	35.21
3	1	1	3	3	3	3	3	3	35.00
4	1	1	4	4	4	4	4	4	38.92
5	1	2	1	1	2	2	3	3	18.44
6	1	2	2	2	1	1	4	4	15.34
7	1	2	3	3	4	4	1	1	58.62
8	1	2	4	4	3	3	2	2	45.49
9	1	3	1	2	3	4	1	2	53.57
10	1	3	2	1	4	3	2	1	78.33
11	1	3	3	4	1	2	3	4	5.11
12	1	3	4	3	2	1	4	3	31.72
13	1	4	1	2	4	3	3	4	24.10
14	1	4	2	1	3	4	4	3	56.58
15	1	4	3	4	2	1	1	2	29.62
16	1	4	4	3	1	2	2	1	34.44
17	2	1	1	4	1	4	2	3	27.75
18	2	1	2	3	2	3	1	4	16.26
19	2	1	3	2	3	2	4	1	98.28
20	2	1	4	1	4	1	3	2	91.31
21	2	2	1	4	2	3	4	1	97.76
22	2	2	2	3	1	4	3	2	78.72
23	2	2	3	2	4	1	2	3	37.32
24	2	2	4	1	3	2	1	4	16.66
25	2	3	1	3	3	1	2	4	15.36
26	2	3	2	4	4	2	1	3	27.63
27	2	3	3	1	1	3	4	2	92.31
28	2	3	4	2	2	4	3	1	94.51
29	2	4	1	3	4	2	4	2	96.14
30	2	4	2	4	3	1	3	1	88.15
31	2	4	3	1	2	4	2	4	20.11
32	2	4	4	2	1	3	1	3	24.03

Table 4 – ANOVA table for the dye removal

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Distance of electrodes	1	3354.6	3354.6	3354.55	187.20	0.000
Temperature	3	66.6	66.6	22.20	1.24	0.352
pH	3	55.6	55.6	18.52	1.03	0.423
Concentration	3	169.2	169.2	56.40	3.15	0.079
Electrode material and design	3	1506.1	1506.1	502.02	28.02	0.000
Time of electrolysis	3	896.1	896.1	298.69	16.67	0.001
Current density	3	5792.9	5792.9	1930.98	107.76	0.000
Amount of electrolyte (NaCl)	3	16099.9	16099.9	5366.63	299.49	0.000
Residual Error	9	161.3	161.3	17.92		
Total	31	28102.2				

DF: degrees of freedom; Seq SS: sequential sum of squares; Adj SS: adjusted sum of squares; Adj MS: adjusted mean squares; F: F-value; p: p-value

## Results and discussion

### Decolorization performance evolution

The collected data were analyzed by using Minitab® 14 (trial version) Statistical Software for the evaluation of the effect of each parameter on the optimization criteria. In order to determine the effective parameters and their confidence levels on the color removal process, an analysis of variance was performed. A statistical analysis of variance (ANOVA) was performed to see which process parameters were statistically significant. *F*-test is a tool to see which process parameters have a significant effect on the dye removal value. The *F*-value for each process parameter is simply a ratio of the mean of the squared deviations to the mean of the squared error.<sup>35,40</sup> The color removal from the dye solutions was investigated in different experimental conditions.

The ANOVA assumptions, which had to be checked in order to test the hypothesis, were satisfied, so the ANOVA results shown in Table 4 are reliable. Since a confidence level of 95 % is used for significance of the factors, the *p*-value less than 0.05 indicates that the corresponding factor is significant. Table 4 shows that five factors are significant with *p*-value equal to zero, whereas parameters B, C, D are not significant. In the present study, the most significant factor was the amount of electrolyte with 299.49 *F* ratio, and other important factors based on *F* were, respectively, current, distance of electrodes, design and material of electrodes, time of electrolysis, initial concentration of dye, pH and temperature. Thus, the initial concentration of dye, pH and temperature were less significant parameters for the EC process. Table 5 provides information about proportionality of influential factors with regard to ANOVA results.

Table 5 – Summarization of factor effects

Factors	Significance level	Relation
A: Distance of electrodes	Significant	Direct
B: Temperature	Less significant	Direct
C: pH	Less significant	Direct
D: Concentration	Less significant	Indirect
E: Electrode material and design	Significant	Direct
F: Time of electrolysis	Significant	Direct
G: Current	Very significant	Direct
H: Amount of electrolyte (NaCl)	Highly significant	Indirect

Fig. 2 is a plot of the main factors' effects on dye removal. This plot is used to visualize the relation between factors and the output response. Fig. 2 shows an increase in R with an increase in the level of electrode distances (A). Meanwhile, Fig. 2 shows that the better electrode material and design (E) is the Al electrode with parallel design.

pH is known to affect the structural stability of dyes and thus, its color intensity. However, MB is stable against variation of pH, therefore, the dye removal efficiency is independent of pH which can be seen in Fig. 2. The same effect as pH is shown for temperature (B).

An increase in the levels of factors such as dye concentration (D) and concentration of NaCl (H) results in a decrease in the dye removal % (R). At higher initial dye concentrations, the amount of dye in the solution is higher. Intermediate products formed from the degradation of dyes increase the resistance by blocking the electrode active sites, and thus, decrease R. In addition, at higher concen-



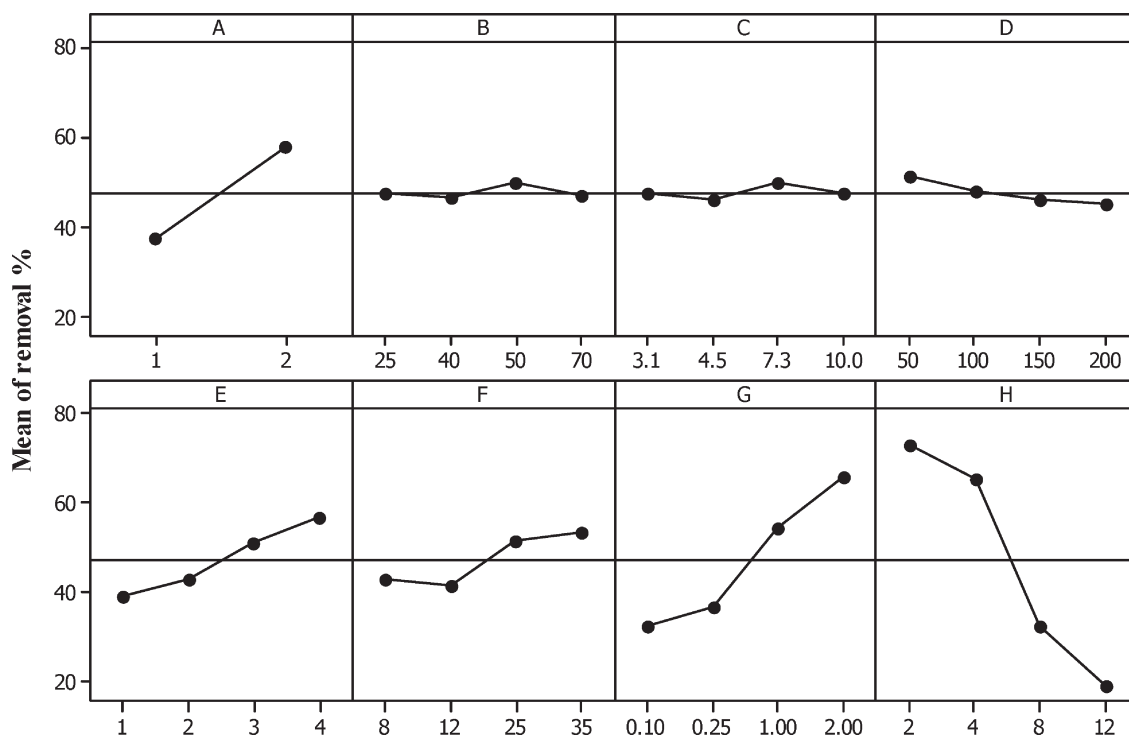


Fig. 2 – Illustration of factor effects on the decolorization

trations, the amount of electrode dissolved is not enough to remove all the dye thus causing a decrease in  $R$ . Therefore, lower dye concentrations caused higher  $R$ .

Fig. 2 indicates an increase in time (F) from 8 to 25 minutes increased  $R$  and after that the  $R$  approached plateau. The dye removal efficiency depends directly on the concentration of ions produced by the electrodes which in turn depends upon time. When the value of time increases, an increase occurs in concentration of Fe ions and their hydroxide flocs.

The effect of current (G) on the removal of dye is shown in Fig. 2.  $R$  increases with an increase in current from 0.1 to 2 A. As the current increases, the efficiency of electrode ionic species production on the anode increases, therefore, there is an increase in floc production in the solution, and hence an improvement in the MB removal efficiency.

High concentrations of chloride ions and salts in water can improve the performance and effectiveness of the EC process. Sodium chloride is easily available at a reasonable cost, and the chloride species avoid inhibition phenomena at the surface of sacrificial anodes. Sodium chloride was chosen to increase the electrical conductivity of the wastewater to a suitable level. An increase in  $R$  with an increase in the NaCl concentrations up to  $2 \text{ g L}^{-1}$  was experimentally shown in our laboratory, however,  $R$  decreases with further increase in electrolyte concentrations (higher than  $2 \text{ g L}^{-1}$ ) as

shown in Fig. 2. Greater ionic strength generally causes an increase in current density at the same cell voltage or a decrease in the cell voltage with an increase in the cell solution conductivity at a constant current density. With an increase in NaCl dosage (H), the conductivity increases with a decrease in electrical resistance and energy consumption. The addition of NaCl enhanced the  $R$  in comparison to that obtained without the addition of NaCl. Addition of NaCl in excess of  $2 \text{ g L}^{-1}$  decreased the  $R$  (Fig. 2). This may be due to the irregularity in electrode dissolution at higher electrolyte concentrations.

Since the most significant factor that varies dye removal during this process is the amount of electrolyte, two-factor effect plots of the amount of electrolyte with other seven factors are provided in Fig. 3 to inform audiences how variations of those factors affect the influence of the amount of electrolyte on the decolorization.

#### Determination of the optimal parameters

Table 6 shows the S/N ratios obtained for each factor level. Combination of optimal factor levels based on this strategy is defined in Table 7. In continuation, analysis of variance was employed to define the important factors affecting the amounts of S/N ratios. Table 8 represents the results of ANOVA for S/N ratio. The Table shows that the amount of electrolyte with a zero  $p$ -value and an  $F$  ratio equal to 299 (Table 4) is the most

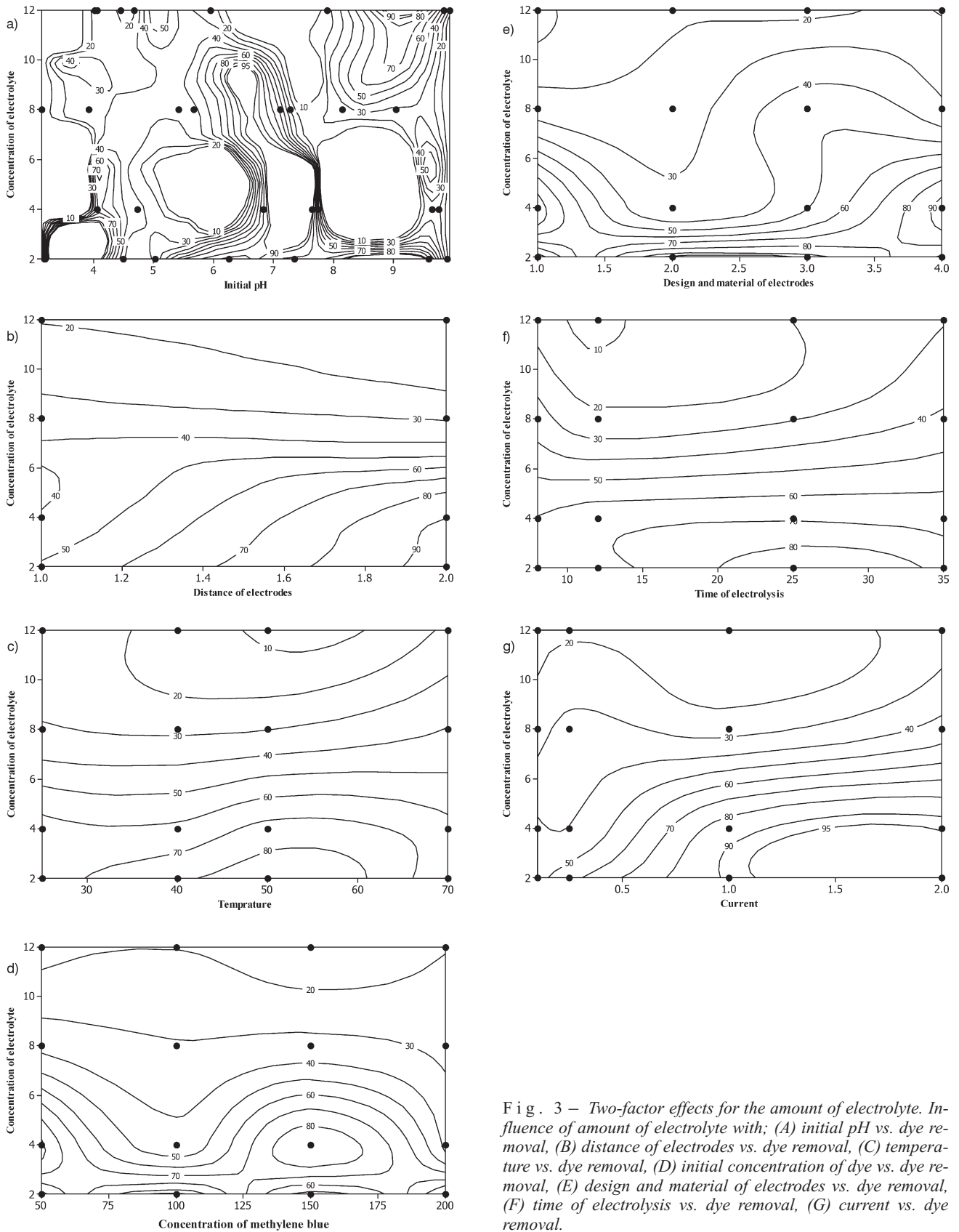


Fig. 3 – Two-factor effects for the amount of electrolyte. Influence of amount of electrolyte with; (A) initial pH vs. dye removal, (B) distance of electrodes vs. dye removal, (C) temperature vs. dye removal, (D) initial concentration of dye vs. dye removal, (E) design and material of electrodes vs. dye removal, (F) time of electrolysis vs. dye removal, (G) current vs. dye removal.

influential parameter that varies the S/N ratio, and the most important factor affecting the sensitivity of the process versus the noise factors. Para-

meters with p-value above 0.05 are not significant and cause less variation in sensitivity of the process.

Table 6 – *S/N analysis for factor levels*

Level	Distance	Temp	pH	Concentration	Electrode material and design	Time	Current density	Amount of electrolyte
1	30.09	32.06	32.06	32.24	29.13	30.89	29.39	36.59
2	33.09	31.39	31.28	31.91	30.67	29.24	30.35	35.53
3		31.00	32.18	31.57	32.57	32.56	31.70	29.75
4		31.89	30.81	30.62	33.98	33.65	34.90	24.47
Delta		1.05	1.37	1.62	4.85	4.40	5.51	12.12

Table 7 – *Optimum factor levels*

Factor	Level
Distance of electrodes	2
Temperature	1
pH	3
Concentration	1
Electrode material and design	4
Time of electrolysis	4
Current density	4
Amount of electrolyte (NaCl)	1

Table 8 – *ANOVA table for the S/N ratio*

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Distance of electrodes	1	72.00	71.995	71.995	35.67	0.000
Temperature	3	5.51	5.514	1.838	0.91	0.474
pH	3	10.17	10.174	3.391	1.68	0.240
Concentration	3	11.75	11.752	3.917	1.94	0.194
Electrode material and design	3	108.77	108.768	36.256	17.96	0.000
Time of electrolysis	3	89.42	89.421	29.807	14.77	0.001
Current density	3	138.71	138.711	46.237	22.91	0.000
Amount of electrolyte (NaCl)	3	755.87	755.874	251.958	124.83	0.000
Residual Error	9	18.17	18.166	2.018		
Total	31	1210.37				

### Modeling of predictor function

The relationship between variables was characterized by a mathematical model called regression equation. Since it is assumed that the factors and the response are linearly related to each other, the regression equation is as follows:

$$y = \beta_0 + \beta_1 x_1 + \dots + \beta_k x_k + \varepsilon \quad (14)$$

where  $y$  is the response and  $\beta_i$  is the regression coefficient for  $i$ th regressor factor. The least square method is used to estimate the regression coefficients.<sup>41,42</sup> Table 9 shows the regression coefficients and the derived regression equation is as follows:

$$\begin{aligned} \% \text{ removal} = & 18.4 + 20.5 A + 0.0055 B + \\ & + 0.155 C - 0.0396 D + 6.09 E + \\ & + 0.471 F + 17.6 G - 5.75 H \end{aligned}$$

Table 9 – *Coefficients for the regression analysis*

Predictor	Coefficient	SE coefficient	T	P
Constant	18.422	7.873	2.34	0.028
C: pH	0.1548	0.4629	0.33	0.741
A: Distance of electrodes	20.477	2.456	8.34	0.000
B: Temperature	0.00546	0.07512	0.07	0.943
D: Concentration of dye	-0.03963	0.02197	-1.80	0.084
E: Design and material of electrodes	6.088	1.098	5.54	0.000
F: Time of electrolysis	0.4714	0.1148	4.11	0.000
G: Current	17.565	1.631	10.77	0.000
H: Concentration of electrolyte	-5.7459	0.3197	-17.97	0.000
S = 6.94620	$R^2 = 96.1 \%$	$R^2_{\text{adj}} = 94.7 \%$		



Analysis of variance was derived to examine the null hypothesis for the regression presented in Table 10. The result indicates that the estimated model by the regression procedure is significant at the  $\alpha$  level of 0.05.  $R$ -Squared ( $R^2$ ) amount was calculated to check the goodness of fit. The  $R^2$  value indicates that the predictors explain 96.1 % of the response variation. Adjusted  $R^2$  for the number of predictors in the model was 94.7 %. Both values show that the data are fitted well.

Table 10 – ANOVA table for the regression analysis

Source	DF	SS	MS	F	P
Regression	8	26992.4	3374.1	69.93	0.000
Residual error	23	1109.7	48.2		
Total	31	28102.2			

#### Confirmed experiments

The purpose of the confirmation experiments is to validate the conclusions drawn during the analysis phase. The confirmation experiment is performed by conducting a test with specific combination of the factors and levels previously evaluated. In this study, after determining the optimum conditions and predicting the response under these conditions, a new experiment was designed and conducted with the optimum levels of EC parameters. The final step is to predict and verify the improvement of the performance characteristic.

Table 11 shows the results of confirmation tests for decolorization. This Table evaluates the predicted % removal and the actual % removal. Actual % removal is achieved through applying a combination of the optimum level of each control parameter. Optimization with S/N ratios from initial setup to optimum condition indicates a growth equal to 6.91 dB, and Table 11 shows that the Taguchi method has obviously increased the % removal in obtained optimum condition.

Table 11 – Results of the confirmation experiments

	Initial setup A <sub>1</sub> B <sub>3</sub> C <sub>1</sub> D <sub>1</sub> E <sub>2</sub> F <sub>1</sub> G <sub>1</sub> H <sub>1</sub>	Optimal condition	
		Prediction	Experiment
Level		A <sub>2</sub> B <sub>1</sub> C <sub>3</sub> D <sub>1</sub> E <sub>4</sub> F <sub>4</sub> G <sub>4</sub> H <sub>1</sub>	
Decolorization (%)	44.70	99.13	97.00
S/N ratio	33.01	39.92	39.92

## Conclusion

In this paper, the Taguchi experimental design method has been used to determine the optimum working conditions for the removal of dye from aqueous solutions by EC. Identified were the most significant factors in the removal process. Optimum levels for many different parameters can be discovered simultaneously with the Taguchi method. A significant advantage of the Taguchi method is the reduction in time and cost. In this study, the most important parameter affecting the color removal was the amount of electrolyte. The following factor level settings have been identified to yield the best combination: amount of electrolyte – level 1, current – level 4, distance of electrode – level 2, design and material of electrodes – level 4, time of electrolysis – level 4, initial concentration of dye – level 1, initial pH – level 3 and temperature – level 1.

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## List of symbols

- EC – electrocoagulation
- OA – orthogonal array
- DOE – design of experiments
- ANOVA – analysis of variance
- S/N – signal to noise ratio
- $\gamma_0$  – concentrations of dye before electrocoagulation, mg L<sup>-1</sup>, in eq. (7)
- $\gamma$  – concentrations of dye after electrocoagulation, mg L<sup>-1</sup>, in eq. (7)
- $y_i$  – characteristic property in eq. (8)
- $n$  – replication number of experiment in eq. (8)
- $SS_T$  – total sum of squares in eq. (9)
- $T$  – sum of all observations in eq. (9)
- $N$  – total number of observations in eq. (9)
- $A_i$  – sum of observations under the  $A_i$  level in eq. (9)
- $n_{A_i}$  – number of observations under the  $A_i$  level in eq. (10)
- $k_A$  – number of levels of the factor A in eq. (10)
- $SS_A$  – sum of squares for factor A in eq. (10)
- $\nu_T$  – total degrees of freedom in eq. (11)
- $\nu_A$  – factor A degrees of freedom in eq. (12)
- $\sigma_A$  – variance for the factor A in eq. (13)
- $R^2$  – regression coefficient

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