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Optimizing decolorization of methylene blue dye by electrocoagulation using Taguchi approach

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ABSTRACT

In this study, the removal of methylene blue (MB) using electrocoagulation (EC) process has been investigated. Taguchi method was applied to determine the optimum MB removal from aqueous solution by EC using iron electrodes. An orthogonal array, L_9 (3^3), experimental design that allows to investigate the simultaneous variations of three controlling factors, namely: current density (A), initial pH (B), and reaction time (C), having three levels was employed to evaluate the effects of experimental parameters. A statistical analysis of variance (ANOVA) was performed to see whether the process parameters were statistically significant or not. According to the *F*-test results, it can be concluded that the degrees of the influences of parameters on the decolorization efficiency are reaction time, initial pH of the solution, and current density. The optimum conditions were found as the third level of current density (150 mA/cm^2), second level of pH (6.5), and third level of treatment time (20 min). Under these conditions, DE was predicted as 96.3% which was within the range of confidence limit of the observed value of 95.5%.

Keywords: Methylene blue; Decolorization; Electrocoagulation; Optimization Taguchi method

1. Introduction

The textile industry is an important sector that consumes extensive volumes of water and chemicals for dyeing processes. Diverse synthetic dyes, widely used in the textile, printing, paper, and leather industries [1], are toxic as well as carcinogenic, mutagenic, and teratogenic [2]. Large amounts of highly colored effluents are esthetically displeasing and affect the nature of the water, impeding light penetration and reducing photosynthetic activity within the stream [3]. The dyes-associated environmental problem comes from their high visibility, undesirability, and recalcitrance. Therefore, their removal from industrial effluents as the most urgent task makes possible to a safe and clean environment [4].

Methylene blue (MB) as a thiazine cationic dye is commonly used for coloring paper, as a temporary hair colorant, for dyeing cottons, wools, and as a coating for paper stock. MB is not strongly toxic, but it can cause some harmful effects [5,6].

Due to stringent environmental regulation, this wastewater has to be treated before disposal into the

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environment. Many technologies have been developed for treating dye wastewater including adsorption [7,8], oxidation [9,10], biodegradation [11,12], membrane separation [13,14], and coagulation–flocculation [15,16] showed better performance, but each technique are costly and cannot be used by small industries to treat the wide range of dyeing wastewaters. The design of novel procedure based on nontoxic and low cost are the best choice for wastewater treatment.

Electrochemical techniques are considered as an attractive methodology for the treatment of dyeing wastewater and have significant advantages namely: wide application, simple equipment, easy operation, lower temperature requirements, and no sludge formation [17]. Several researchers have investigated the feasibility of electrochemical degradation of dye with various electrode materials for wastewater treatment, such as titanium-based dimensionally stable anode, platinum, conductive-diamond, metal alloy, and boron-doped diamond [18].

Electrocoagulation (EC) is a simple and cost-effective technique for the treatment of dye wastewater. During EC, hydroxide ions (OH^-) are produced at the cathode, thus increasing the solution pH, which affects the formation of metallic hydroxide flocs. The important factors affecting dye removal by EC include nature of dye, electrode material and surface area, applied current, solution pH, and mixing speed [19,20].

In order to attain the maximum removal efficiency of dyes, an optimization strategy is required to find the best experimental conditions. Recently, statistical experimental designs described as design of experiments (DOE) can be used to investigate the effect of all the possible combinations of the conditions. In this context, the Taguchi method as a statistical technique is used to determine the optimum conditions. Moreover, the use of Taguchi method is to minimize the experimental runs by fractional factorial designs based on robust orthogonal arrays (OA) of factors.

In this study, the electrochemical decolorization of MB was investigated and a statistical experimental design, Taguchi Method, is used to identify the best optimum conditions from the key experimental parameters such as current density (A), initial pH of the solution (B), and reaction time (C) on the decolorization of MB from aqueous solution by EC process using iron as an electrode material.

2. Materials and methods

2.1. Materials

The cationic dye, MB (purity > 98%, MW 373.90) was purchased from the Duksan Pharmaceutical Co. Ltd., and its properties are given in Table 1. MB solutions were prepared by dissolving desired amounts of MB in 1 L of distilled water. The UV–vis spectrophotometer (UV, model 1240/Shimadzu) was used to measure the concentration of MB at $\lambda_{max} = 658$ nm. The decolorization efficiency (DE) of MB was calculated using the following relationship:

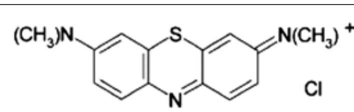
$$DE\ (\%) = \left(1 - \frac{C_e}{C_0}\right) \times 100 \tag{1}$$

where C_0 is the initial MB concentration (mg/l) and C_e is the final MB concentration (mg/L).

2.2. Experimental method

The EC experimental apparatus consists of an electrolytic cell which was a Plexiglas reactor containing two iron electrode plates that were connected to a digital DC power supply (0–30 V, 2.5 A) in monopolar mode. An ampermeter and voltmeter were used to measure the current passing through the circuit and the applied potential, respectively. The electrodes were situated vertically in the cell and the gap between the electrodes is 10 mm. The volume of MB solution treated was 400 mL and the total effective electrode area was 35 cm^2. Experiments were conducted at initial MB concentration of 50 mg/L. The pH and conductivity were adjusted to a desirable value using NaOH or H_2SO_4 and Na_2SO_4, respectively.

Table 1
Characteristics of MB

Dye	Structure	Molecular weight	λ max (nm)	Color index (CI)
Methylene blue [3,7bis(dimethylamino) phenazathionium chloride trihydrate]		373.9	658	52,015

2.3. Taguchi design of experiments

Taguchi method is a multi-parameter optimization procedure, which is very useful in identifying and optimizing dominant process parameters with a minimum number of experiments [21]. This technique is based on an OA of experiments and includes data transformation into an accurate and desirable signal-to-noise (S/N) ratio [22]. An OA is a minimal set of experiments with various combinations of parameter levels. The selection of a suitable OA depends on the number of control factors and their levels [23]. The optimization of the observed values is determined by using the signal-to-noise (S/N) ratios and analysis of variance (ANOVA). S/N values may be optimized using three types of performance characteristics: the larger–the better or maximum response, the smaller–the better, or minimum response, and the nominal–the better, or a goal of reducing the standard deviation. The use of ANOVA (analysis of variance) is to find out the percentage contributions of individual parameters.

In this work, the variables chosen for this investigation are current density (A), initial pH of the solution (B), and reaction time (C). The variables investigated and their levels were summarized in Table 2.

To analyze the significance of three factors at three different levels, a full factorial design requires 3^3 (=27) experiments to find the influencing parameter while Taguchi design involves only nine experiments using an OA $L_9(3^3)$. Experimental plan table according to L_9 (3^3) is shown in Table 3.

In Taguchi method, the optimum conditions should be determined using the S/N ratio of the results obtained from experiments designed by OA technique. For optimization of DE, larger-the-better type of objective function has been used. In this case, the exact relation between S/N ratio and the signal is given by Eq. (2)

$$S/\mathrm{NL} = -10\,\log\left(\frac{1}{n}\sum_{\mathrm{i}=1}^{n}\frac{1}{Y_i^2}\right) \tag{2}$$

where n is the number of repetitions performed for an experimental combination and Y_i is the DE of the ith experiment.

Table 2
Parameters and their values corresponding to their levels studied in experiments

Factors	Designation	Levels		
		1	2	3
Current density (mA/cm^2)	A	20	85	150
Initial pH	B	3	6.5	10
Time (min)	C	2	11	20

Table 3
Experimental variables, their levels, and results of conducted experiments corresponding to L_9 experimental plan

Experiment No.	Variables and their levels			DE of MB (%)	S/N ratio
	A	B	C		
1	1	1	1	10.65	20.54
2	1	2	2	51.65	34.26
3	1	3	3	90.95	39.17
4	2	1	2	36.22	31.18
5	2	2	3	97.76	39.8
6	2	3	1	50.55	34.07
7	3	1	3	75.32	37.54
8	3	2	1	46.53	33.35
9	3	3	2	85.52	38.64

The experimental variables, their levels, and results of conducted experiments are given in Table 3.

3. Results and discussion

3.1. Taguchi design analysis

3.1.1. Taguchi results

In this study, Taguchi method was used to identify the optimal conditions and to select the parameters having the most principle influence on the MB dye removal. Standard L_9 OA table with three levels and three factors is shown in Table 3. Each row in the table represents a trial condition with the level of factors.

Table 3 shows the DE and S/N ratio for decolorization of the solution containing MB calculated using equation (1) and (2), respectively.

Fig. 1 shows the S/N response graph for decolorization of MB solution by EC process. Therefore, the current density (factor A), pH (factor B), and reaction time (factor C) exhibit much variation which indicates that these are the factors having a major affect on the removal efficiency of decolorization of MB.

Fig. 1 illustrates the S/N ratio averages for each factor at the three different levels and the corresponding response variable. As seen in Fig. 1, the effect of three parameters on MB DE increased in the following order: A (current density) < B (initial pH) < C (reaction time). The variation is smaller in the cases of the current density, making this the parameter with less influence in the EC process. According to the analysis, while the reaction time is the most effective parameter, the current density is the least effective in the decolorization of MB by EC process.

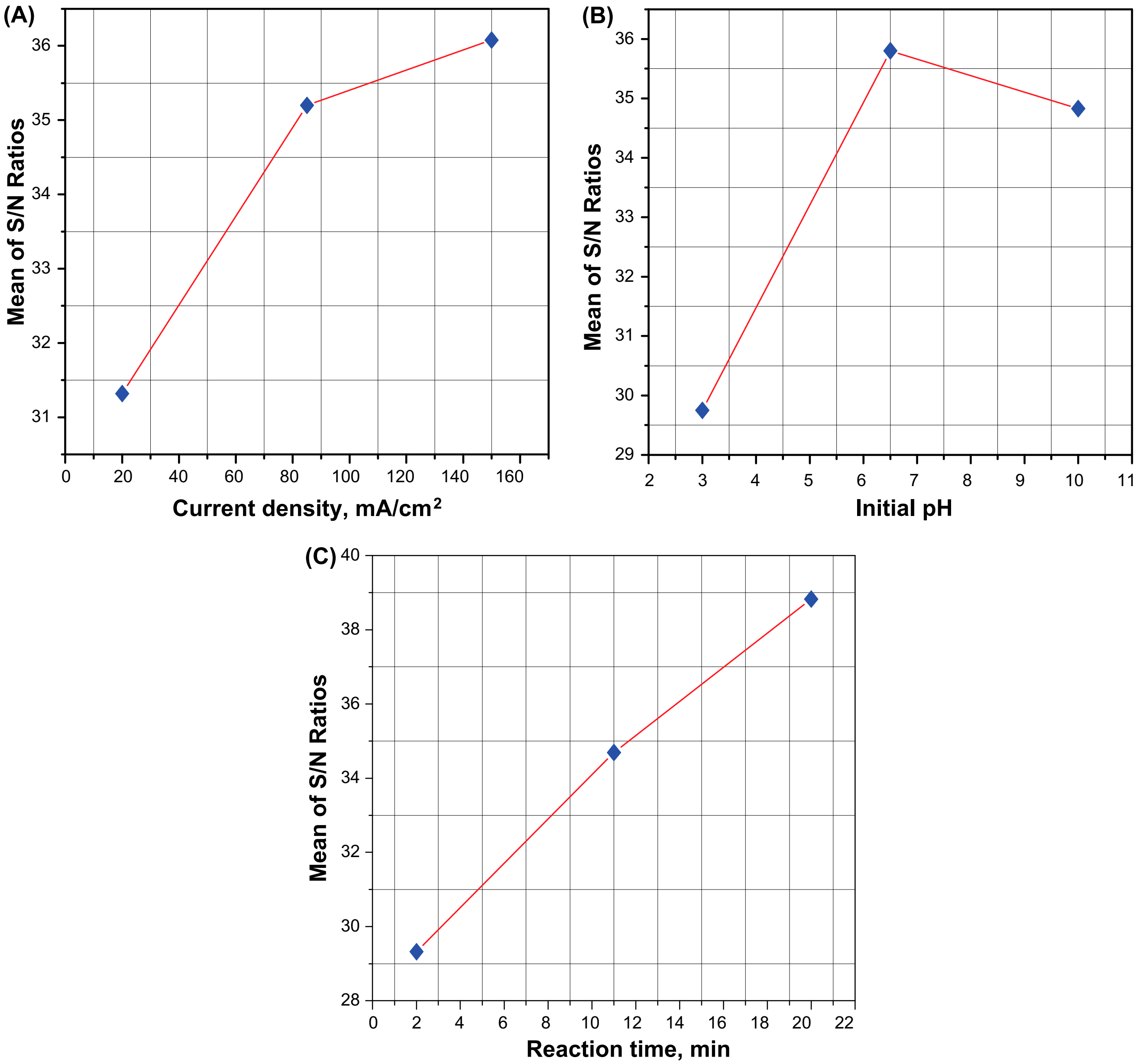


Fig. 1. Main effects plots for S/N ratios of the EC process. (A) current density, (B) initial pH, and (C) reaction time.

Considering the highest value of the S/N ratio as the optimum, the optimum combination of conditions was determined to be A3, B2, and C3. In other words, based on the S/N ratio, the optimal factors for MB dye removal are A (current density) at level 3 (150 mA/cm^2), B (initial pH) at level 2 (6.5), and C (reaction time) at level 3 (20 min). Under these conditions of process variables optimized by Taguchi method, the predicted DE was estimated to be 96.3% which is in good agreement with the value observed as 95.5%.

3.1.2. ANOVA results

In order to conduct an analysis of the relative importance of each factor more systematically, an ANOVA was applied to the data.

The main objective of ANOVA is to extract from the results how much variations each factor causes relative to the total variation observed in the result [24]. The results of ANOVA are listed in Table 4, the reaction time had the largest variance and the initial pH indicated the second, respectively.

Table 4
Results of ANOVA for the removal efficiency of MB by EC

Factor	Degree of freedom	Sum of squares	Mean squares	F-Ratio	Percent (%)
A	2	492.12	246.06	6.39	6.28
B	2	1,932.70	966.35	25.09	28.08
C	2	4,106.41	2,053.205	53.31	60.97
Error	2	77.03	38.515		4.67
Total	8	6,608.26	3,304.13		100

$R^2 = 0.967$, $R^2_{adj} = 0.948$, Fcr (Table) = 19.

F-ratio is a tool to indicate which parameter has a significant effect on the DE of MB by EC process. In this study, degree of freedom for the error is 2. Fcr value of the parameters for degrees of freedom of 2 and 2 at a confidence level of 95% is 19. According to the results, the *F*-ratio (6.39) for current density is smaller than the Fcr-value and the F-Ratio for initial pH (25.09) and reaction time (53.31) are higher than the Fcr. This means that the variance in current density is insignificant compared with the variance in initial pH and reaction time that has a significant effect on the responses.

Table 4 shows that the error is 4.67% (<50%), which is far enough from the limit. It means that the error of the experiment is not significant.

The use of the *F*-ratios in an ANOVA is only helpful for the qualitative evaluation of factorial effects. Quantitative evaluation can be achieved using percentage contribution (*P*%) [25]. It is calculated by dividing the source's net variation by SS_T, which is given by Eq. 3.

$$P\ (\%) = \frac{SS_A - (DOF_A \times MS_e)}{SS_T} \times 100 \tag{3}$$

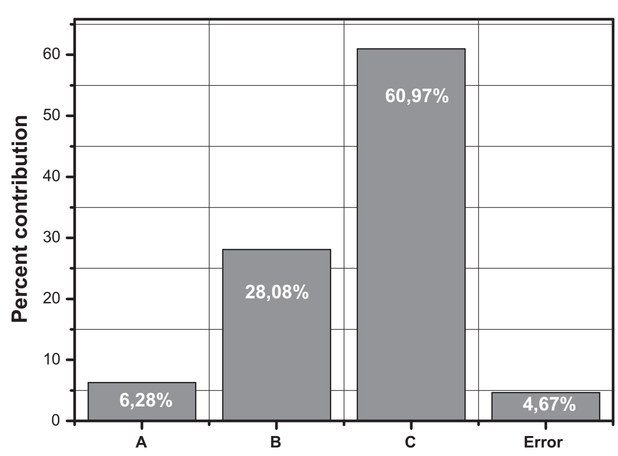


Fig. 2. Contribution of each factor on the performance statistics.

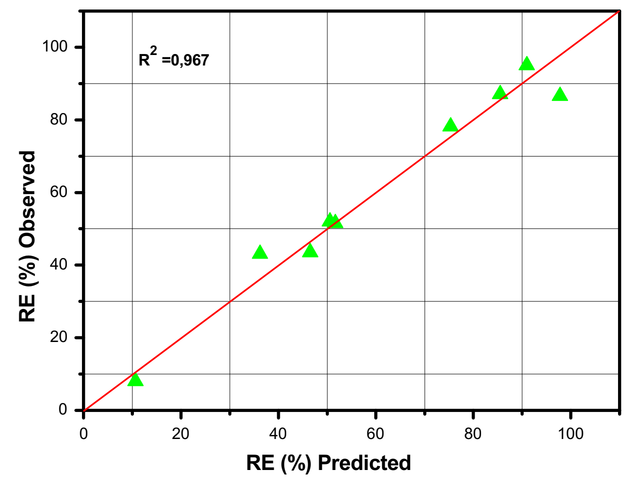


Fig. 3. Observed response vs. predicted response for the EC of MB dye.

Percent contributions of all factors are presented in Fig. 2. The order of importance of factors is as follows: reaction time > initial pH > current density. Reaction time is the most important parameter for decolorization of MB dye by EC process. Moreover, the predicted values were compared with the experimental values and are shown in Fig. 3. The predicted values are closer with experimental values for all runs. Also, the R^2 and adjusted R^2 values are 0.967 and 0.948, respectively. This confirms a good agreement between the predicted and the observed values in all cases.

4. Conclusion

In this study, Taguchi DOE (L_9) has been used to determine the optimum conditions for the MB dye removal from aqueous solutions by EC. Effect of current density, initial pH of the solution, and reaction time on the EC of MB has been investigated and effects of these parameters on the system performance have been evaluated based on DE. The larger-the-better S/N ratio was used to analyze the results of experiments. According to the obtained results, the following conclusions were made: the reaction time is the most significant factor on the DE, and it contributes 60.97%; the second most significant factor is the initial pH, and its contribution percent to the DE is 28.08%; and finally, the least effective process parameter is the current density.

The optimum conditions within the selected parameter values were found as the third level of current density (150 mA/cm^2), second level of pH (6.5), and third level of reaction time (20 min). The confirmation experiment was carried out at optimum conditions. Under the optimized conditions, 95.5% of DE was obtained.

The predicted and observed DE values are close to each other with a high coefficient of determination value (R^2) of 0.967. This confirms a good agreement between the predicted and the observed values.

The study shows that the Taguchi method is suitable to optimize the experiments for MB dye removal from aqueous solution by EC process.

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