



Open Access: ISSN 1847-9286

<https://pub.iapchem.org/ojs/index.php/JESE>

Original scientific paper

Optimization of parameters for dye removal by electro-oxidation using Taguchi Design

Mani Nandhini, Balasubramanian Suchithra,

Ramanujam Saravanathamizhan✉ and Dhakshinamoorthy Gnana Prakash

Department of Chemical Engineering, SSN College of Engineering, Kalavakkam, Chennai 603110, India

✉Corresponding author: E-mail: thamizhan79@rediffmail.com; Tel. +91 44 27469700; Fax +91 44 27469772

Received: April 18, 2014; Revised: May 9, 2014; Published: December 6, 2014

Abstract

The aim of the present investigation is to treat the dye house effluent using electro-oxidation and to analyse the result using Taguchi method. L16 orthogonal array was applied as an experimental design to analyse the results and to determine optimum conditions for acid fast red dye removal from aqueous solution. Various operating parameters were selected to study the electro-oxidation for the colour removal of the effluent. The operating parameter such as dye concentration, reaction time, solution pH and current density were studied and the significance of the variables was analysed using Taguchi method. Taguchi method is suitable for the experimental design and for the optimization of process variables for the dye removal.

Keywords

Electro-oxidation; Taguchi design; colour removal; acid fast red; optimization

Introduction

Effluents discharged from textile industries have high intensity of colour, which leads to pollution. Highly coloured wastewater can be treated by different methods such as biological treatment, chemical coagulation, activated carbon adsorption, ultrafiltration, ozonation, wet oxidation, photocatalysis, electrochemical methods etc. [1-5]. Among these methods the electrochemical treatment has been receiving greater attention in recent years due to its unique features such as versatility, energy efficiency, automation and cost effectiveness [6]. The electrochemical technique offers high removal efficiencies and the main reagent is the electron called 'clean reagent' which degrades all the organics present in the effluent without generating any secondary pollutant or by-product/sludge.

Industrial wastewaters have been treated by electro-oxidation techniques and the operating parameters have been optimized by different techniques. The widely used technique is response surface methodology for the experimental design and process optimization. Taguchi method is another method in the experimental design, proposed by Genichi Taguchi, contains system design, parameter design, and tolerance design. Taguchi method was effectively used to improve the product or process effectiveness by using a loss function to attain the product quality in terms of the parameter design [7]. Taguchi is the preferable technique among statistical experimental design methods since it uses a special design of an orthogonal array to study the effective parameters with a minimum number of experiments. 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 parameter design using Taguchi method minimizes the time and experimental runs. In the design, 'signal' and 'noise' (S/N) represent the desirable and undesirable values for the output characteristics, respectively and the ratio is a measure of the quality characteristic deviating from the desired value. Further an analysis of variance (ANOVA) is used to determine the significant parameters.

Recently Taguchi's designs have been applied to various chemical and environmental engineering studies for the experimental design and for the optimization of the process variables. Asghari *et al.* [8] used Taguchi method to determine the optimum conditions for methylene blue dye removal from aqueous solutions using electrocoagulation. The authors found that the amount of electrolyte was the most significant parameter for the colour removal. Srivastava *et al.* [9] used Taguchi method to determine the optimum conditions for the orange-G dye removal from aqueous solution by electrocoagulation using iron plate electrodes. Author also applied this methodology to optimize the process variables for the multi-component adsorption of metal ions onto bagasse fly ash and rice husk ash [10]. Kaminari *et al.* [11] used Taguchi method to determine the optimum condition of process variables and find the influencing parameters for the recovery of heavy metals from acidified aqueous solutions using electrochemical reactor. Kim *et al.* [12] optimized the experimental process variable using Taguchi technique for the nano-sized silver particles production by chemical reduction method. In another study Mohammadi *et al.* [13] used Taguchi method to determine the optimal experimental conditions for the separation of copper ions from a solution. Moghaddam *et al.* [14] used Taguchi method to design the experimental runs and optimize the parameters for the ammonium carbonate leaching of nonsulphide zinc ores. Maria *et al.* [15] designed the experimental runs and optimized the process parameters using Taguchi method for the adsorption of acid orange 7 dyes on guava seed. In that way Taguchi method was used to study the electro oxidation of dye removal.

The objective of the present study is to treat the acid fast red dye using electro-oxidation. The effect of experimental parameters such as initial dye concentration, reaction time, solution pH and current density on colour removal was investigated using an L16 orthogonal array. The Taguchi experimental design has been used to determine the optimum conditions for the maximum colour removal of the dye from aqueous solutions.

Materials and Methods

All the chemicals used in this study were AR grade (Merck). Acid fast red was prepared by dissolving definite quantity of dye in the distilled water. To increase the conductivity of the solution of 1 mg L⁻¹ NaCl was used as supporting electrolyte for all experimental runs. The initial

solution pH was adjusted by 0.1 M HCl or 0.1 M sodium hydroxide solution. Experiments were repeated twice to minimize the experimental error.

Experimental setup

The experimental setup of the batch reactor is shown in the Figure 1. The volume of the reactor was 250 ml and electrodes used were fixed inside the reactor with 1 cm space between them. Stainless steel sheet cathode and mesh type Ruthenium oxide coated Titanium anode were used. The void fraction of the mesh type anode accounts 20 % by area, which resulted in an effective anode area of 28 cm² (7×5 cm). The electrodes were connected to a 5 A, 10 V DC regulated power supply, through an ammeter and a voltmeter. The solution was constantly stirred at 200 rpm using a magnetic stirrer in order to maintain a uniform concentration. DC power supply was given to the electrodes according to the required current density and the experiments were carried out under constant current conditions. The samples were analysed for the colour removal using UV-Vis spectrophotometer (Jasco, V-570). The percentage colour removal was calculated by:

$$\text{Colour removal, \%} = \frac{Abs_i - Abs_t}{Abs_i} \times 100 \quad (1)$$

where Abs_i and Abs_t are absorbance of initial and at time t at the corresponding wavelength λ_{max} .

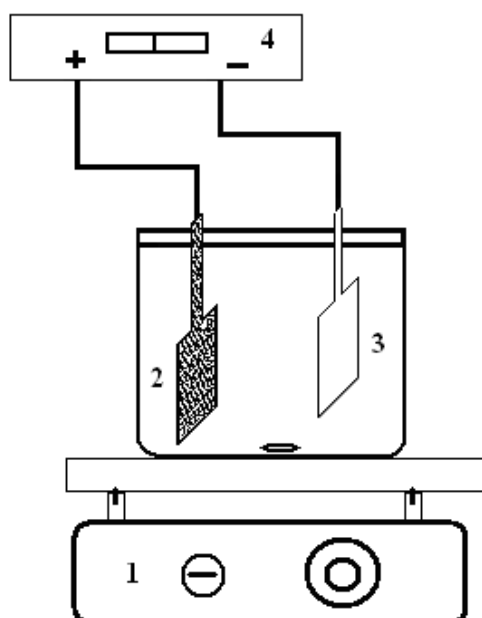


Figure 1. Schematic diagram of the experimental setup
1. Magnetic stirrer 2. Anode 3. Cathode 4. DC Power supply

Taguchi design

The following procedure was adopted for the parameter design.

1. Planning of experiment

- i. Determine the experimental responses of the process.
- ii. Determine the levels of each variable.
- iii. Select a suitable orthogonal array table. The selection based on the number of variables and number of levels.
- iv. Transform the data from the experiments into a proper S/N ratio.

2. Implementing the experiment, based on design table.

3. Analyzing and examining the result

- i. ANOVA analysis to determine the significant parameters in the process.
- ii. Draw the main effect plot, S/N ratio plot, mean plot to analysis the optimal level of the control variables.

The factors and levels chosen for the present experiment are shown in the Table 1. L16 orthogonal array design was selected for the four variables with four different levels for the each factor. Table 2 shows the Taguchi design for the electro oxidation of acid fast red dye. Each row represents one experimental run. Based on Taguchi design the experiments were carried out and the percentage colour removal was observed as response. The proposed design was an orthogonal array, for which each pair of the columns had all the possible combinations of levels. The S/N ratio characteristics can be divided into three categories when the characteristic is continuous:

(i) Nominal is the best characteristic

$$\frac{S}{N} = 10 \log \frac{\bar{y}}{S_y^2} \tag{2}$$

Table 1. Variables and their values corresponding to their levels investigated in the experiments

Variables		Level			
		1	2	3	4
A	Dye concentration, mg l ⁻¹	25	50	75	100
B	Time, min	15	30	45	60
C	pH	2	5	7	10
D	Current density, mA cm ⁻²	5	7.5	10	12.5

Table 2. Experimental variables, their levels and results of conducted experiments corresponding to L16 experimental plan

S. No	Levels				Run 1	Run2
	A	B	C	D	Colour removal, %	
1	1	1	1	1	78.50	78.40
2	1	2	2	2	81.82	81.20
3	1	3	3	3	86.42	86.40
4	1	4	4	4	89.75	90.00
5	2	1	2	3	70.08	69.98
6	2	2	1	4	84.41	84.15
7	2	3	4	1	77.20	78.01
8	2	4	3	2	91.53	91.50
9	3	1	3	4	71.55	71.54
10	3	2	4	3	68.21	68.12
11	3	3	1	2	86.78	86.78
12	3	4	2	1	83.44	83.58
13	4	1	4	2	52.05	52.19
14	4	2	3	1	56.70	57.25
15	4	3	2	4	91.66	91.89
16	4	4	1	3	96.32	96.80

(ii) *Smaller the better characteristics*

$$\frac{S}{N} = -10 \log \frac{1}{n} \sum y^2 \quad (3)$$

(iii) *Larger the better characteristics*

$$\frac{S}{N} = -10 \log \frac{1}{n} \sum \frac{1}{y^2} \quad (4)$$

Where, \bar{y} is the average of observed data, s_y^2 the variance of y , n is the number of observations, and y the observed data. For each type of the characteristics, with the above S/N ratio transformation, the higher the S/N ratio the better is the result. The experimental data were analysed using MINITAB 14 (PA, USA) [16].

Result and discussion

Main effect plot

Main effect plot for the percentage colour removal using electro oxidation of acid fast red dye is shown in the Figure 2. The plot is used to visualize the relationship between the variables and output response. The effect of initial dye concentration on colour removal is shown by the factor 'A'. The percentage colour removal of the dye increases with decrease in dye concentration. This is due to the fact that at higher initial dye concentrations, the intermediate products formed due to the degradation of dyes increase the resistance of current flow by blocking the electrode active sites, and thus, decrease colour removal. The effect of electrolysis time on colour removal is shown in the figure by a factor 'B'. The percentage colour removal depends on the electrolysis time. When time increases the generation of OCl^- radical increases due to electro-oxidation which results increase the percentage colour removal. The effect of pH on the mean colour removal is represented by the factor 'C'. As it is observed from the figure, colour removal increases with the decrease of pH. This is due the fact that the hydroxyl radical generation is high at acidic pH which results to increase the rate of colour removal. The effect of current density on the removal of dye is shown by the factor 'D'. The increase of current density increases the OCl^- generation hence the percentage colour removal increases. High concentrations of chloride ions and salts in water can improve the performance and effectiveness of the electro-oxidation process. Various levels (1, 2, 3, 4) of the operating parameter (A, B, C, D) and their mean colour removal is shown in the Table 3. It is observed form the table, Level '1' shows highest colour removal of 84.06 %, 84.37 % for the initial dye concentration and pH, respectively. Level '4' shows highest colour removal of 90.37 %, 86.52 % for electrolysis time and current, respectively.

Table 3. Mean colour removal for electro oxidation of acid fast red

Level	Mean colour removal, %			
	A	B	C	D
1	84.06	68.04	86.52	74.14
2	80.86	72.74	81.71	77.98
3	77.50	85.64	76.62	80.29
4	74.36	90.37	71.94	84.37

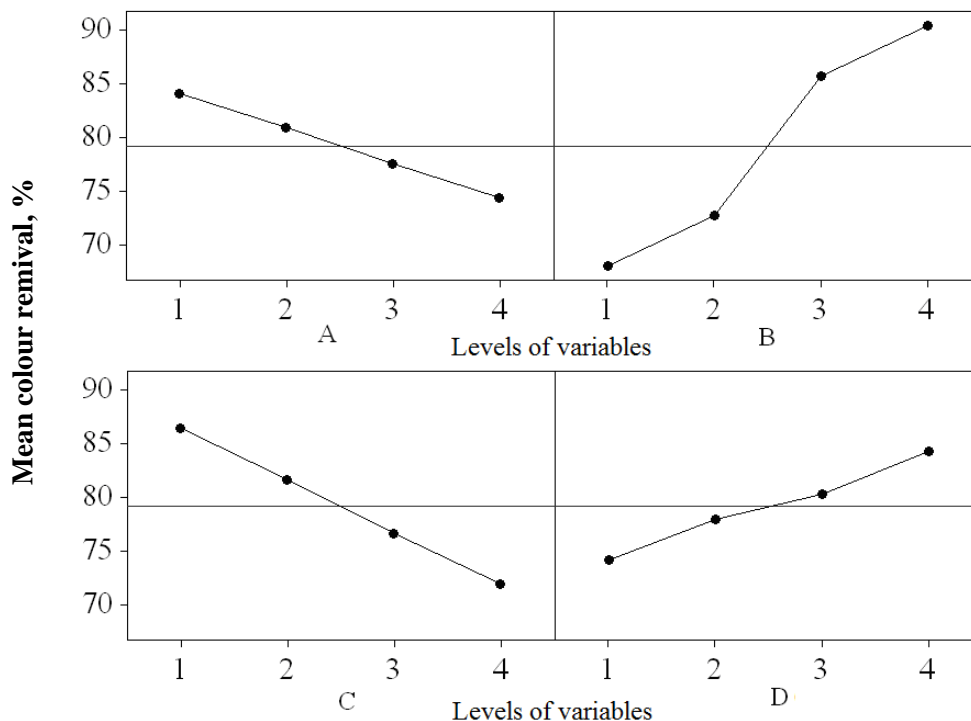


Figure 2. Main effect plot for the percentage colour removal of acid fast red
 A: Dye concentration parameter; B: Time parameter; C: pH parameter; D: Current density parameter

Signal to noise (S/N) ratio

Taguchi method was used to identify the optimal conditions and most influencing parameters on colour removal. In the Taguchi method, the terms ‘signal’ to ‘noise’ ratio represent the desirable and undesirable values for the output response, respectively. The *S/N* ratios are different according to the type of output response. In the present case, larger *S/N* ratio is better for high colour removal. Figure 3 shows the *S/N* ratio of dye removal using electro-oxidation.

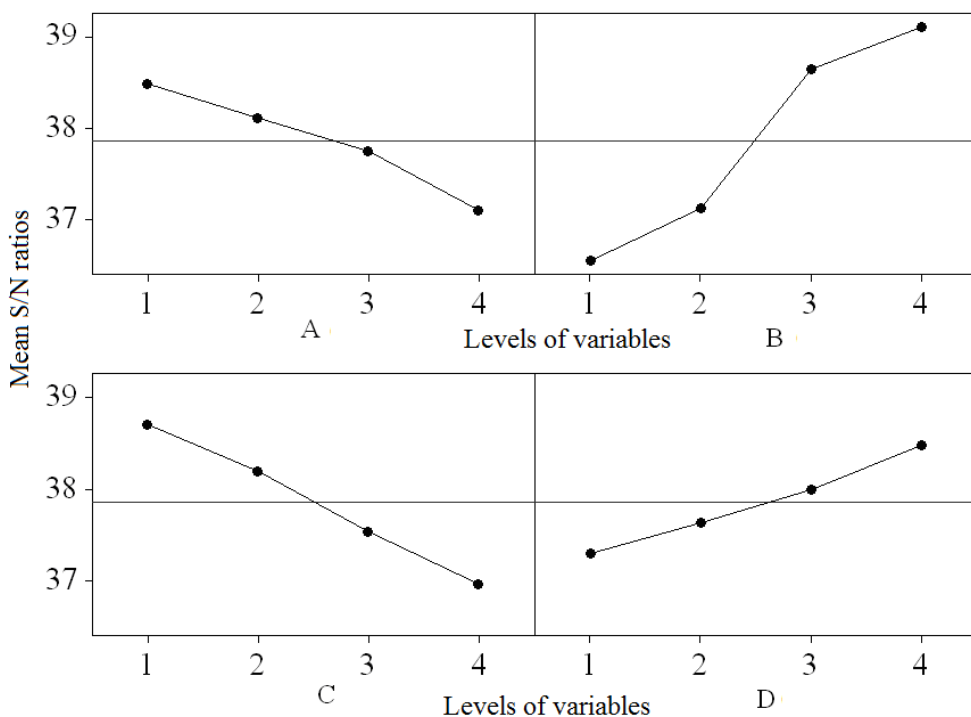


Figure 3. *S/N* ratio plot for the percentage colour removal of acid fast red .
 A: Dye concentration parameter; B: Time parameter; C: pH parameter; D: Current density parameter

It can be noticed that at higher *S/N* ratio better level for colour removal was achieved. It is observed that factor 'A', initial dye concentration, and factor 'C', pH, are required in a lower level and electrolysis time 'B' and applied current density 'D' are required at higher level for the maximum colour removal.

Optimization of process parameters based on *S/N* ratio

The process parameters were optimized based on *S/N* ratio. Larger the *S/N* ratio higher the percentage colour removal and vice versa. The values of the *S/N* ratios for the operating parameters are shown in the Table 4. It is observed from the Table that the *S/N* ratio of level '1' 38.48, 38.72 shows the higher value for the factor 'A' and 'C', respectively. It shows that level '1' gives the higher colour removal. The larger *S/N* ratios of factor 'B' and 'D' for the level '4' are 39.11, 38.48, respectively. It shows that level '4' gives the higher colour removal.

Table 4. *S/N* ratio for electro oxidation of acid fast red

Level	<i>S/N</i> ratio of electro oxidation			
	A	B	C	D
1	38.48	36.56	38.72	37.31
2	38.11	37.13	38.20	37.64
3	37.74	38.64	37.54	38.00
4	37.10	39.11	36.97	38.48

Analysis of variance (ANOVA)

Analysis of variance (ANOVA) was performed to see whether the process parameters were statistically significant or not. The F-test is a tool to check which process parameters have a significant effect on the colour removal. The P value less than 0.05 shows that the parameter is significant. ANOVA table for the colour removal of the dye is shown in the Table 5. It is observed from the table the most influential factor was electrolysis time because the P value is 0.049 with the corresponding sum of the square is higher compared to other variable in the Table. Then the less significant variables are pH, current density and initial dye concentration.

Table 5. ANOVA table for the electro oxidation of acid fast red

Source	Degree of freedom	Sum of the squares	Mean of squares	F	P
A	3	210.9	70.29	1.50	0.374
B	3	1330.5	443.48	9.46	0.049
C	3	476.8	158.94	3.39	0.171
D	3	220.2	73.40	1.56	0.361
Residual Error	3	140.7	46.90		
Total	15	2379			

Conclusion

Taguchi experimental design was used to determine the optimum operating conditions of the dye removal from aqueous solutions using electro oxidation. The significant variables were identified for the colour removal process. Optimum levels for operating parameters can be simultaneously identified with the Taguchi method. The advantage of the Taguchi method is the reduction in time and minimization of the number of experimental runs. For the present study

level '1' is best for initial dye concentration (25 g L^{-1}) and initial pH (2), level '4' is best for electrolysis time (60 min) and current density (12.5 mA cm^{-2}) for the highest colour removal. It can be concluded that Taguchi method is suitable for the experimental design and to optimize the process variable for the colour removal of the dye effluent.

Acknowledgement: The authors are grateful to the SSN Trust for the financial support of this work.

References

- [1] M. C. Gutierrez, M. Crespi, *Journal of the Society of Dyers and Colorists* **115** (1999) 342–345.
- [2] M. Panizza, G. Cerisola, *Chemical Reviews* **109** (2009) 6541–6569.
- [3] C. A. Martinez-Huitle, E. Brillas, *Applied Catalysis B: Environmental* **87** (2009) 105-145.
- [4] L. C. Davies, I. S. Pedro, J. M. Novais, S. Martins-Dias, *Water Research* **40** (2006) 2055 - 2063.
- [5] G. Chen, *Separation & Purification Technology* **38** (2004) 11–41.
- [6] F. C. Walsh, *Pure and Applied Chemistry* **73** (2001) 1819–1837.
- [7] J. A. Ghani, I. A. Choudhury, H. H. Hassan, *Journal of Materials Processing Technology* **145** (2004) 84–92.
- [8] A. Asghari, M. Kamalabadi, H. Farzinia, *Chemical and Biochemical Engineering Quarterly* **26** (2012) 145–154.
- [9] V. C. Srivastava, I. D. Mall, I. M. Mishra, *Chemical Engineering Journal* **140** (2008) 136–144
- [10] V. C. Srivastava, I. D. Mall, I. M. Mishra, *Industrial & Engineering Chemistry Research* **46** (2007) 5697-5706.
- [11] N. M. S. Kaminari, D. R. Schultz, M. J. J. S. Ponte, H. A. Ponte, C. E. B. Marino, A. C. Neto, *Chemical Engineering Journal* **126** (2007)139-146.
- [12] K. D. Kim, D. N. Han, H. T. Kim, *Chemical Engineering Journal* **104** (2004) 55-61.
- [13] T. Mohammadi, A. Moheb, M. Sadrzadeh, A. Razmi, *Desalination* **169** (2004) 21-31.
- [14] J. Moghaddam, R. Sarraf-Mamoory, M. Abdollahy, Y. Yamini, *Separation & Purification Technology* **51** (2006)157-164.
- [15] M. P. Elizalde-Gonzalez, V. Hernandez-Montoya, *Journal of Hazardous Materials* **168** (2009) 515–522.
- [16] K. Ravikumar, S. Ramalingam, S. Krishnan, K. Balu, *Dyes and Pigments* **70** (2006) 18-26.