

Study of decolorization variety by electrocoagulation process in the removal of Dye solution containing Remazol Yellow G: optimization of effective parameters using the Taguchi method

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ABSTRACT

The Taguchi method was applied as an experimental design to determine optimum conditions for colour removal from azo dyes solutions containing remazol yellow G by electrocoagulation (EC) using iron and steel electrodes as anode and cathode, respectively. An orthogonal array (OA9) experimental design that allows to investigation the simultaneous variations of four parameters (initial remazol yellow G concentration, initial pH of the solution, time of electrolysis and current density) which have three levels was employed to evaluate the effects of experimental parameters. Performance measure analysis was followed by performing a variance analysis, in order to determine the optimum levels and relative magnitude of the effect of parameters. The desired characteristic for response has been elected as maximum decolourization. Therefore, Taguchi's 'the larger the better' performance formula was used. The optimum conditions were found to be initial remazol yellow G concentration, 100 mg/l, initial pH of the solution 9, time of electrolysis, 25 min, and current density, 1mAcm⁻².

Keywords: Electrocoagulation; Decolorization; Remazol yellow G; Taguchi method

1. INTRODUCTION

Nowadays the colour and high chemical oxygen demand (COD) of effluents from dye house cause serious environmental contamination problems. Particular azo dyes represent about half of the dyes used in the textile industry and, as a consequence, a relevant problem of pollution related to the release of these products in the environment is taking place [1-2]. Although there were several other technologies available for the removal of colour and COD from azo dye wastewater such as biodegradation [3], sorption [4-6], electrochemical and oxidative degradation [7-11], electro-coagulation (EC) as an electrochemical method was developed to

overcome the drawbacks of conventional decolourization technologies. EC process provides a simple, reliable and cost-effective method for the treatment of wastewater without any need for additional chemicals, and thus the secondary pollution. It also reduces the amount of sludge, which needs to be disposed. In this technique, the coagulant is generated by electrolytic oxidation of an appropriate anode material that leads, at an appropriate pH, to the insoluble metal hydroxide which is able to remove a large variety of pollutants [12]. 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 [13-14]. Iron and aluminum are generally used as sacrificial anodes. A growing research interest is reported on the treatment of various wastewater types: paper industry wastewater [15-18], Landfill leachate [19-22], electroplating wastewater [23], Tannery effluent [24], laundry wastewater [25-26] and latex particles [27]. EC process has also been used in the removal of heavy metals [28-39], phosphate [40-41], fluoride [42-45], boron [46-47] and textile wastewaters [48-53]. Mean-while, EC process has been widely used to decolorize various structurally different dye containing solutions such as disperse, reactive and acidic dyes [54-60].

The technique of defining and investigating all possible conditions in an experiment involving multiple factors is known as the design of experiments [61]. In robust parameter design, the primary goal is to find factor settings that minimize response variation, while adjusting (or keeping) the process on target. After determination which factors affect variation, we can find settings for controllable factors that will either reduce the variation that makes the product insensitive to changes in uncontrollable noise factors, or both. A process designed with this goal will produce more consistent output. Robust design is an engineering methodology for low costs [61]. Taguchi's parameter design is an important tool for robust design. It offers a simple and systematic approach to optimize design for performance, quality and cost [61-62]. When a critical quality characteristic deviates from the target value, it causes a loss. Continuously pursuing variability reduction from the target value is the key to achieve high-quality and reduce cost. By applying this technique one can significantly reduce the time required

for experimental investigations. This is important in investigating the effects of multiple factors on performance as well as to study the influence of individual factors to determine which factor has more influence and which one has less [61,63]. The most important stage in the design of an experiment lies in the selection of control factors. For this purpose, many factors as possible should be included and non-significant variables must be identified at the earliest opportunity. Taguchi creates a standard orthogonal array to accommodate these requirements. Depending on the number of factors and levels needed, the choice is left to the user to select the standard orthogonal array.

The Aim of this investigation is to explore the decolourization by electrocoagulation from the solutions containing remazol yellow azo dye and to determine the influence of the variables such as initial dye concentration, current density, initial pH, and time of electrolysis on removal process. The experiments were carried out according to Taguchi orthogonal array (OA) experimental design with two replicates and three center points.

2. EXPERIMENTAL

2.1 Materials and methods

The commercial dye used in this project was purchased from Merck (Germany). The chemical structure of this dye is shown in Fig. 1. Dye solutions were prepared by dissolving dye in distilled water. The batch experimental cell is shown in Fig. 2. The conductivity of solutions was raised up and adjusted in steady value by the addition of 2g/l NaCl (Merck, Germany). The conductivity measurement was performed using a HANA conductometer (HI8733, Hungary). All pH measurements were performed with a Metrohm 691 pH-meter using a combined glass electrode. The pH of the solutions were adjusted by adding

NaOH or H₂SO₄ (Merck, Germany) solutions. Iron (ST 37-2) plates were

used as anode and steel (grade 304) plates were used as cathode.

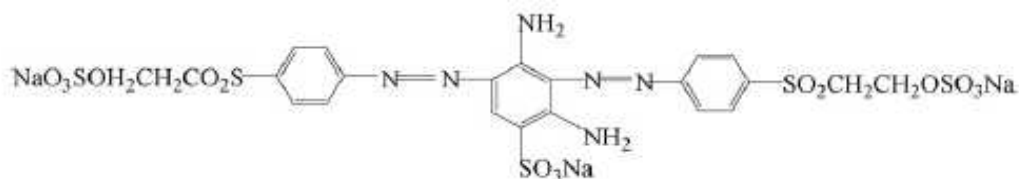


Fig. 1. Chemical structure of remazol golden yellow ($\lambda_{\max}=423\text{nm}$)

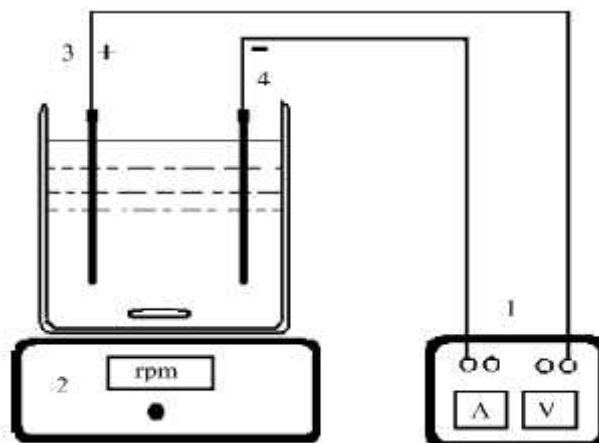


Fig. 2. An apparatus electrocoagulation cell: (1) digital DC power; (2) Magnetic bar-stirrer; (3) anode; (4) cathode.

Dimensions of electrodes were 65 mm \times 30 mm \times 3 and the distance between two electrodes in EC cell was 20 mm in all experiments. The electrodes were connected to a DC power supply (Micro, China) with galvanostatic operational options for controlling the current density. All the runs were performed at room temperature. In each run, 500 ml of the dye solution was decanted into the electrolytic cell. At the end of EC, the 4 ml of solution was centrifuged by centrifuge (Fanazma, Iran). The remanent dye concentration was determined from its absorbent characteristics in the UV-Vis range with the calibration method using Beer-Lambert's law by means of Shimadzu spectrophotometer (S 2000, Japan). The calculation of colour removal efficiency (CR%) after electrocoagulation treatment was performed using this formula:

$$\text{CR\%} = \frac{C_0 - C}{C_0} \times 100 \quad (1)$$

Where C_0 and C are concentrations of dye before and after electrocoagulation in mg/l, respectively. All experiments were repeated twice, and the experimental error was around 2%.

2.2 Statistical analysis

The collected data for colour removal efficiency was analyzed using the computer software package program (MINITAB Release 14.13) for the evaluation of the effect of each parameter on the optimization criteria. The variables chosen for this investigation were initial dye concentration, initial pH of the solution, current density and time of electrolysis. The variables were investigated and their levels were summarized in Table 1. In order to optimize the removal efficiency process, experimental parameters and their investigated levels are presented in Table 1. The experimental design, based on standard OA9 (43)

orthogonal array, was conducted to change the settings of the various

process parameters (Table 2).

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

Variables	Levels		
	1	2	3
A: initial Dye concentration (mg/l)	50	100	150
B: current density (mA/cm ²)	0.25	0.5	1
C: time of electrolysis (min)	8	15	25
D: initial of pH the solution	5	9	3

Table 2. Experimental variables, their levels and results of conducted experiments corresponding to L₉ experimental plan.

Experiment	Variables and their levels				Dye Removal efficiency (%)		
	A	B	C	D	First series	Second series	Average
1	1	1	1	1	51	49	50.00
2	1	2	2	2	96	97.12	96.56
3	1	3	3	3	92.94	91	92.00
4	2	1	2	3	80.19	81	86.00
5	2	2	3	1	99	99	99.00
6	2	3	1	2	74.95	76	75.50
7	3	1	3	2	98	99	98.50
8	3	2	1	3	57	54	55.50
9	3	3	2	1	98	95	96.50

In order to observe the effects of noise sources (uncontrollable factors) on this process, each experimental trial was repeated twice under the same conditions at different times. Also, the order of experiments was made randomly in order to avoid noise sources which was not been considered initially and which could take place during an experiment and affected the results in a negative way. Performance measure analysis reflecting the variation in the response at each setting was chosen as the optimization criteria. Its analysis determines the controllable factors and their settings, which minimize the variation in process while keeping the mean response on target. By setting those factors at their optimal levels, the process can be made robust to changes in operating and environ-

mental conditions. When the desired characteristic for the response is larger, it is better; Taguchi recommends the use of larger is better :

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

Where S/N is performance characteristics, n is the number of repetitions done for an experimental combination and Y_i is the performance value of the ith experiment. The performance value corresponding to the optimum working conditions can be predicted by utilizing the balanced characteristic of OA. For this, the following additive model may be used:

$$Y_i = m + X_i + e_i \quad (3)$$

where m is the overall mean of performance value, X_i is the fixed effect of the parameter level combination used in the ith experiment and e_i is the

random error in the i^{th} experiment. Detailed description of the Taguchi method can be found in literatures [64-67].

3. RESULTS AND DISCUSSION

The removal efficiency data were statistically analyzed to investigate the influence of each parameter. The results are given in Fig. 3. The optimal level of a process parameter is the level with the highest S/N value calculated by equation (2). Fig. 3 shows the variation of the performance characteristics with the variables. To determine the experimental conditions for the first data point, the initial pH of the solution for that point is level 1 which is 5 for this parameter. The experiments for which initial pH of the solution level is 1 are experiments 1, 4, and 7. Therefore, the performance characteristics value of the first data point is the

average of those obtained from experiments 1, 4 and 7. Thus, experimental conditions for the second data point are the conditions of the experiments for which column C is 2 (experiments 2, 4 and 9). The numerical value of the maximum point in each graph marked the best value of that particular parameter and was found as A2 (100 mg/l), D2 (pH=9), B3 (1mA/cm²) and B3 (25 minute). These parameter values provide the optimum conditions.

The optimal levels of these factors are the levels with the maximum performance measures that is, with minimum variability. From Fig. 3, the optimal levels of these factors are initial dye concentration (A2: 100 mg/l), initial pH of the solution (D2:9), current density (B3: 1 mA/cm²) and time of electrolysis (C3: 25 min).

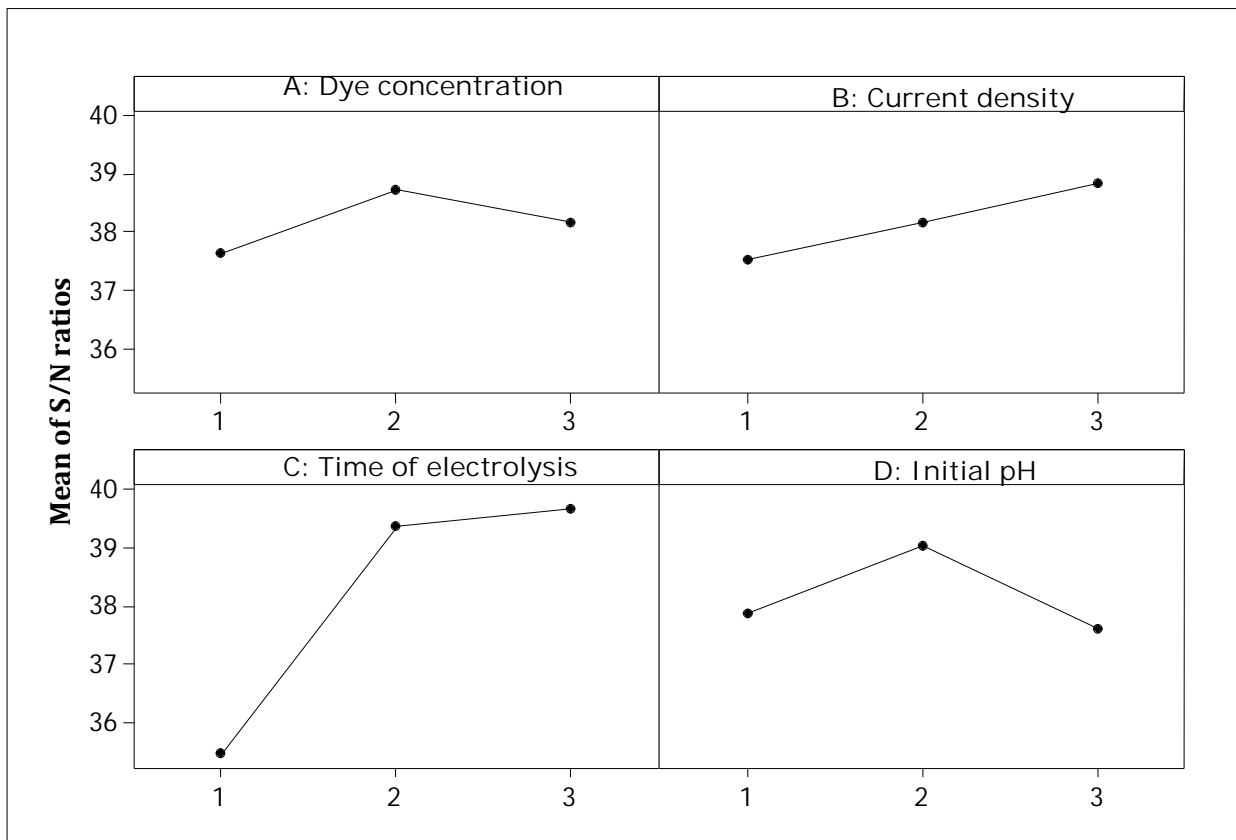


Fig. 3. Effect of parameters on optimization criteria.

3.1 Effect of initial pH

It has been established that pH is an important parameter influencing the performance of the EC process. The kinetics of Fe^{2+} conversion to Fe^{3+} are strongly affected by the pH; the surface charge of the coagulating particle also varies with pH [68]. In general, as shown in Fig. 4, at lower and higher pH Fe is increasingly soluble.

To examine its effect, the sample was adjusted to a desired pH for each experiment by using sodium hydroxide

or sulfuric acid solutions. At lower pH the protons in the solution are reduced to H_2 at the cathode and the same proportion of hydroxide ions cannot be produced. As shown in Fig. 4, at lower pH, Fe(OH)^{2+} and Fe(OH)_2^+ were produced which are disadvantageous for colorant precipitation [69]. In summary, higher colour removal efficiency was obtained in base media, as reported by several authors [68].

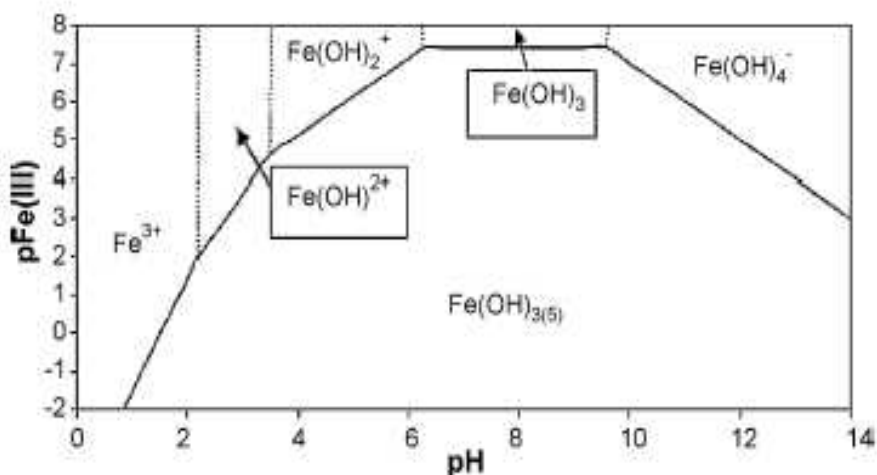
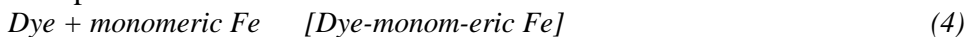


Fig. 4. Predominance-zone diagrams for Fe(III) chemical species in aqueous solution. In this case, the solid line represents the solubility equilibrium of Fe(OH)_3 and the dotted line represents the predominance limits among soluble chemical species.

The mechanism of the electrochemical process in aqueous systems is quite complex. However, the colour removal process may involve the dye molecule adsorbing by both electrostatic attraction and physical entrapment. The insoluble metal hydroxides of iron can

remove dye molecules by surface complexation or electrostatic attraction. In surface complexation, it is assumed that the dye molecule can act as a ligand to bind a hydrous iron moiety with precipitation and adsorption mechanisms [69-71]:

Precipitation



Adsorption



Also, the dye actually may be complexing with the iron hydroxide forming ionic bonds [72].

According to Fig. 5 pH of the medium has to increase during the process. As a matter of fact, as shown in Fig. 5 pH is

increased during the electrocoagulation. Another mechanism is a reductive process (i.e. hydrogen gas is produced) where some dye molecules possibly degraded by a reduction mechanism [73].

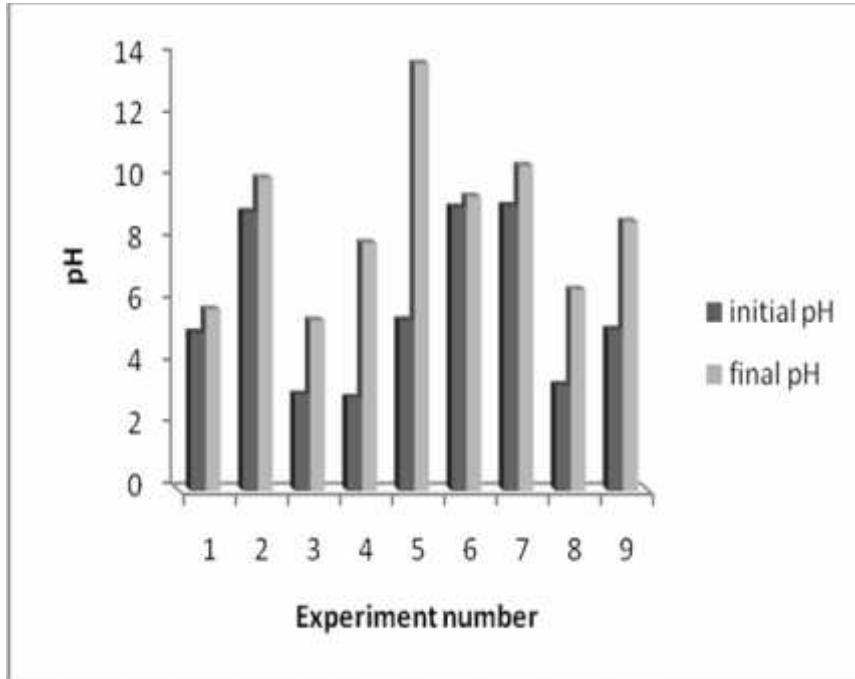


Fig. 5. The variation of pH during the electrocoagulation process.

3.2 Effect of current density

It is well known that the amount of current density determines the coagulant production rate, and adjusts the rate and size of the bubble production, and hence affects the growth of flocs [73-74]. However, it is advisable to limit the current density in order to avoid excessive oxygen evolution as well as to eliminate other adverse effect, like heat generation [75]. With

increasing current density the amount of oxidized iron and the bubble generation is increased and consequently the amount of the hydroxyl polymers available for the attraction of the dye molecule are also increased. The maximum dye removal was observed at 1 mAcm^{-2} current density for pH 9 and 25 min of electrolysis time (Fig. 6).

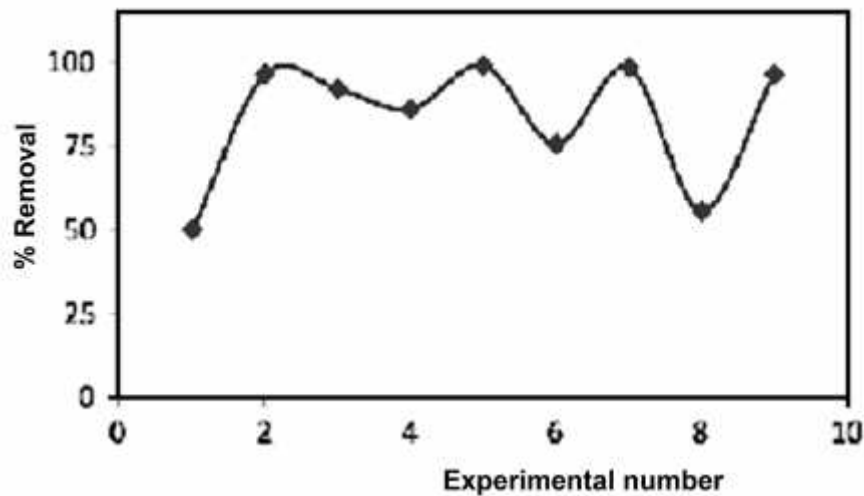


Fig. 6. The variation of color removal % during the electrocoagulation process.

3.3 Effect of electrolysis time

Time of electrolysis which increases comparable changes in the removed efficiency of remazol yellow G is observed. The colour removal efficiency depends directly on the concentration of hydroxyl and metal ions produced on the electrodes. The colour of the reactive dye was decreased as a function of elapsed time.

The rate of removal remazol yellow G can be represented by the following mth order reaction kinetics:

$$\frac{dC}{dt} = -kC^m \quad (8)$$

Where C represents the remazol yellow G concentration, m is the order of reaction, k is the reaction rate coefficient and t is the time. For a first-order reaction, the above equation becomes

$$\ln\left(\frac{C}{C_0}\right) = -kt \quad (9)$$

That C_0 is the initial remazol yellow G concentration. According to the above equation, a plot of $\ln(C_0/C)$ against t will yield a straight line with a slope of k. There was reasonably good fit of first order kinetic model to the observed data.

3.4 Effect of the initial dye concentration

The dye solutions with different initial concentrations in the range of 50–150 mg/l were treated by EC in different conditions of density, pH and time of electrolysis. According to the results, up to the concentration of 100 mg/l, the removal percentage was relatively constant. However, above this concentration, the removal percentage was decreased due to the insufficient hydroxyl and metal ions produced on the electrodes in the high dye concentrations and the constant current density (Fig. 3A).

4. CONCLUSION

In this paper, Taguchi method has been used to determine the optimum working conditions for the dye removal

from aqueous solutions by EC. The orthogonal array, OA9, technique is described for experimental design as it reduces the number of experiments required to investigate a set of parameters and to minimize time and cost while performing experiments. Experimental investigations into the parameter effects have allowed determining the optimum configuration of design parameters for dye colour removal efficiency performance. It can be said that EC is a highly effective process for dye removal from aqueous solutions, due to obtained efficiencies based on decolourization are in general satisfactory levels (Table 2).

Acknowledgments

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مطالعه انواع رنگزدایی توسط فرآیند انعقاد الکتروشیمیایی در حذف محلول رنگینه حاوی ریمازول قهوه‌ای G: بهینه‌سازی پارامترهای موثر با استفاده از روش تاگوچی

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چکیده:

روش تاگوچی به عنوان یک طرح تجربی برای تعیین شرایط بهینه برای حذف رنگ از محلول‌های رنگینه آزو محتوی ریمازول قهوه‌ای G با انعقاد الکتروشیمیایی توسط الکتروده‌های آهن و استیل به ترتیب به عنوان آند و کاتد بکار برده شد. یک طرح تجربی با آرایش اورتوگونالی که امکان بررسی تغییرات همزمان چهار پارامتر (غلظت اولیه رنگینه ریمازول قهوه‌ای G، pH اولیه محلول، زمان الکترولیز و دانسیته جریان) که سه سطح داشته، را می‌دهد، برای ارزیابی اثرات پارامترهای آزمایشگاهی بکار گرفته شد. آنالیز اندازه‌گیری عملکرد توسط اجرای یک آنالیز واریانس برای تعیین سطوح بهینه و بزرگی نسبی اثر پارامترها دنبال شد. مشخصه مطلوب برای جواب به عنوان ماکزیمم رنگ‌زدایی انتخاب شده است. بنابراین فرمول عملکرد بزرگتر-بهرتر تاگوچی استفاده گردید. شرایط بهینه بصورت غلظت اولیه رنگینه ریمازول قهوه‌ای G (100 mg/L)، pH اولیه برابر ۹، زمان الکترولیز ۲۵ دقیقه و دانسیته جریان 1mAcm^{-2} بدست آمد.

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