Optimization of Methylene Blue Dye Removal by Electroflocculation Using Response Surface Methodology

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**Abstract.** This study investigates the decolorization of methylene blue (MB) dye using an electroflocculation process in a batch reactor. The objective is to develop a mathematical model for optimizing and evaluating the effects and interactions of three primary operating parameters: the number of plates, applied voltage, and electroflocculation time, on MB color removal. Initially, a preliminary experimental investigation was conducted to examine the individual effects of these parameters on MB removal efficiency using a one-factor-at-a-time approach. Subsequently, the response surface method incorporating the Box-Behnken design was employed to determine the optimal values for the three primary operating parameters. This experimental design consisted of seventeen runs. The experimental data were analyzed using analysis of variance (ANOVA) with conventional quadratic and Two-factor Interaction (2FI) model regression. The high R2 value of 0.9988 (with adjusted and predicted R2 values of 0.9981 and 0.9959, respectively) from the ANOVA for the 2FI model indicates a significant fit between the proposed mathematical equation and the experimental data. This model can be used to predict removal efficiency and optimize operating parameters for MB decolorization. According to the model, the optimal operating conditions for maximum color removal in the shortest time are a reaction time of 20 minutes, an applied voltage of 19.29 V, and six aluminium electrode plates. Under these conditions, the experimental and predicted color removal efficiencies were 93.50 % and 93.72 %, respectively.

# Introduction

Dyes, commonly used as colorants for various materials, are prevalent in several industries such as textiles, printing, paper, plastic, and leather. The textile industry consumes a significant portion of the world's dye production. With a growing global population and increasing demand for clothing, the utilization of dyes in textile dyeing is expected to rise. Globally, it is estimated that 10-15% of the dyes used in textile production, amounting to millions of liters of wastewater annually, are released into the environment, underscoring the urgent need for effective treatment solutions.[1] Dye-containing wastewater is characterized by its intense color, high chemical and biological oxygen demand, pH, temperature, and turbidity. In some cases, it may also contain harmful compounds that can produce hazardous by-products during hydrolysis, oxidation, or other reactions in the waste solution.[1]

The direct discharge of untreated dye wastewater into water bodies can significantly threaten surrounding ecosystems, causing biological alterations, dissolved oxygen (DO) depletion, and aquatic life extinction. As a result, proper treatment of dye effluents is critical before discharging them into water bodies. Numerous methods, including chemical coagulation, adsorption, photocatalytic degradation, aerobic and anaerobic digestion, chemical degradation, ozonation, and electrocoagulation/electroflocculation, have been extensively researched for removing dye pollutants.[1]–[7] While a variety of methods have been explored for dye wastewater treatment, each comes with notable limitations. Chemical coagulation often generates large volumes of sludge that require further treatment [2], while adsorption can become cost-prohibitive due to the need for regenerating or replacing adsorbents [3]. Photocatalytic degradation typically requires high energy input [4], and biological methods may be limited by the toxicity of certain dyes and the time required for microbial adaptation [5]. These constraints highlight the need for alternative methods that are both cost-effective and environmentally sustainable, such as electroflocculation (EF). EF addresses many of these limitations by requiring no chemical additives, producing minimal sludge, and being operable within a compact system with low energy requirements, making it a promising solution for industrial applications [6].

In wastewater treatment, electroflocculation (EF) technology is garnering interest due to its simplicity, efficiency, and cost-effectiveness compared to other methods. EF has been successfully applied to treat various organic effluents, including those from tanneries,[8] refineries,[9] food industries,[10] heavy metals,[11] and dairy wastewater.[12] The technology offers numerous advantages, such as no need for chemicals during treatment, no secondary pollutants, reduced sludge disposal, and minimal space requirements.[13]

EF is an electrochemical method involving the release of metal ions from sacrificial electrodes when a direct current is applied. The electrochemical dissolution of these electrodes generates coagulants in water, enhancing the coagulation and precipitation of soluble or colloidal pollutants.[14] Aluminum (Al) or iron (Fe) are common electrode materials due to their affordability and availability. In this study, aluminum plates were used as electrodes for their exceptional coagulation efficiency with trivalent Al³⁺ ions.[13] The reactions that occur in the reactor cell include:

1. Anode: Al ⇌ Al³⁺ + 3e⁻
2. Cathode: 3H₂O + 3e⁻ ⇌ 3OH⁻ + 3/2 H₂

The Al³⁺ and OH⁻ ions formed through electrolysis reactions (1) and (2) create various monomeric species[15] such as Al(OH)₄⁻, Al(OH)₂⁺, Al(OH)₂²⁺, and Al₂(OH)₂⁴⁺, over a wide pH range. These species eventually convert into Al(OH)₃ via complex precipitation kinetics:

1. In solution: Al³⁺ + 3OH⁻ → Al(OH)₃

Aluminum hydroxide formation in the solution acts as an active coagulant, which binds to colloidal particles and counter ions, creating agglomerates or flocs. These flocs settle at the reactor's bottom and can be filtered through gravity filtration. Hydrogen gas bubbles evolved at the cathode and oxygen at the anode may also trap pollutants, causing flotation of the flocculated pollutants to the solution's surface.[16] The electroflocculation method combines three key pollutant removal mechanisms: anode dissolution, gas bubble formation, and floc sedimentation or flotation. Pollutant removal is a result of these processes and their interactions within the EF cell. However, performance is influenced by factors such as applied voltage, reaction time, initial conductivity, initial concentration, and electrode number. To improve EF efficiency, operating parameters must be optimized. Traditional multifactor experiments often use the one-factor-at-a-time approach, which adjusts one factor while keeping others constant. This method is time-consuming and overlooks interactions between EF process factors.[17] Response surface methodology (RSM) is an alternative technique for successful EF process optimization.

RSM is a mathematical tool for designing experiments and developing models to assess the impact of operating parameters and achieve optimal working conditions with minimal experimental trials.[18] Among the various RSM experimental designs, the Box-Behnken design is the preferred choice for this study.

Preliminary experiments utilized the one-factor-at-a-time method to evaluate the influence of operating parameters (applied voltage, electroflocculation time, concentration of NaCl, initial dye concentration, and electrode number) on the color removal efficiency (CRE %) of methylene blue using EF technique. The one-factor-at-a-time results indicated that applied voltage, electroflocculation time, and the number of plates significantly affected EF performance. Therefore, the latter part of this study focused on applying RSM to design and optimize the CRE % of methylene blue dye using the EF technique as a function of the number of plates (A), applied voltage (B), and electroflocculation time (C). By using RSM, particularly the Box-Behnken design, the study aimed to minimize the number of experimental trials while maximizing the CRE % of methylene blue dye in the EF process. This approach enables researchers to identify the optimal operating conditions for the EF process, taking into account the interactions between various factors, such as the number of plates, applied voltage, and electroflocculation time. Ultimately, RSM allows for a more efficient and effective optimization of the EF process, leading to better wastewater treatment outcomes and reduced environmental impact.

In summary, the electroflocculation (EF) process relies on the electrochemical dissolution of sacrificial electrodes, such as aluminum, to generate coagulant species that facilitate pollutant removal through coagulation, flocculation, and flotation mechanisms. Aluminum hydroxide acts as an active coagulant that binds pollutants into flocs, which either settle or float due to gas bubble formation at the electrodes. The efficiency of EF is influenced by operating parameters like applied voltage, electroflocculation time, and electrode number. While traditional one-factor-at-a-time experiments provide initial insights, they fail to account for parameter interactions. Response surface methodology (RSM), specifically the Box-Behnken design, allows for a more systematic optimization of EF performance. By minimizing experimental trials and identifying optimal conditions, RSM enhances pollutant removal efficiency and contributes to more sustainable wastewater treatment solutions. Compared to conventional methods such as chemical coagulation or adsorption, the RSM-optimized electroflocculation process demonstrated in this study offers enhanced efficiency in dye removal, reduced environmental impact due to minimal chemical usage, and improved sustainability through lower sludge production and energy consumption.

# Materials and methods

## Dye wastewater

In this study, methylene blue was dissolved in water to create a synthetic dye wastewater solution. The dye's chemical structure and physical properties are illustrated in Fig. 1 and detailed in Table 1, respectively.

|  |
| --- |
| A chemical structure with letters and numbers  Description automatically generated |
|  |

**FIGURE 1.** Chemical structure of methylene blue dye.

**TABLE 1.** Physical properties of methylene blue dye.

|  |  |
| --- | --- |
| Property | Value |
| Color Index (C.I.) | 52030 |
| Chemical class | Basic Blue 9 |
| Maximum absorbance wavelength ( / nm | 663 |
| Color | Dark green powder |
| Chemical formula |  |
| Molecular mass / g mol⁻¹ | 319.85 |

## Standard calibration method

To quantify residual dyes in the effluent, the standard calibration technique was employed. Standard solutions of 0, 5, 10, 20, and 30 ppm were prepared from the 1000 ppm stock solution. The absorbance of each standard was measured at its corresponding wavelength of 663 nm using a UV-Vis Spectrophotometer (Dynamica HALO XB-10).

## Experimental set-up

Figure 2 presents a schematic representation of the experimental setup employed in this study. The batch experiment was conducted in a 0.7 L reactor with a working capacity of 0.5 L. The EF reactor used was a rectangular plastic container, approximately 12.5 cm × 7.0 cm × 8.0 cm in size. For this study, both anodes and cathodes were composed of aluminum, and the Al plates were arranged in monopolar-parallel connections. The electrodes had dimensions of 5.0 cm × 10.0 cm × 0.04 cm and an exposed surface area of 35 cm2 per plate. The sacrificial electrodes were directly connected to a digital DC power supply (WANPTEK DC Power Supply GPS305D). During the treatment process, a magnetic stirrer (Thermo Scientific Cimarec Hot Plate Magnetic Stirrer SP131320-33) was employed to thoroughly agitate the wastewater solution at a constant stirring speed and room temperature.

A diagram of a magnetic stirrer

Description automatically generated

**FIGURE 2**. Schematic diagram of the EF experimental setup: (1) DC power supply, (2) cables/wires, (3) electrodes, (4) EF reactor cell and (5) magnetic stirrer.

## Experimental procedure

A measured volume (0.5 L) of methylene blue solution, adjusted to the required initial dye and electrolyte concentrations, was placed in the reactor cell. Solutions with various sodium chloride (NaCl) concentrations were prepared to achieve the desired electrolyte concentration. In each run, 0.5 L of the dye solution was added to the EF reactor cell. After each run, the treated dye sample was allowed to settle for 20 minutes and subsequently filtered using a 125 mm diameter filter paper (Sartorius – Grade 389). The operating parameters examined using the one-factor-at-a-time approach are provided in Table 2.

**TABLE 2**. Variables studied in the electroflocculation of methylene blue solutions.

|  |  |
| --- | --- |
| Operating parameters | Values |
| Applied voltage / V | 12, 14, 18 and 22 |
| Electrolysis time / minutes | 10, 15, 30, 45 and 60 |
| Electrolyte concentration / ppm | 200, 300 and 400 |
| Initial dye concentration / ppm | 10, 20, 30 and 40 |
| Number of plates | 2, 4 and 6 |

## Analysis

A UV-Vis spectrophotometer (Dynamica HALO XB-10) was utilized to analyze the remaining dye concentration (λ\_max = 663 nm) in the filtered sample by measuring the absorbance of each sample three times and calculating the average reading. The color removal efficiency (CRE %) of synthetic MB dye wastewater was determined using Equation (4), which is expressed in terms of methylene blue concentration:

CRE (%) = (1)

Here, and represent the initial and final concentrations of the methylene blue wastewater solution, respectively.

## Response Surface Methodology using Box-Behnken Design

The Response Surface Method (RSM) is a statistical tool used to optimize various processes. In this research, the primary objectives of employing RSM are to determine the optimal operating parameters for the decolorization of MB dye and to identify the relationship between factors and responses. In this study, the Box-Behnken design (BBD), an experimental design for RSM, was employed to create a set of design experiments using Design Expert software (version 12). As depicted in Table 3the Box-Behnken design assigns three levels (-1, 0, and 1) for each factor, which is analogous to coding, with -1 representing the lowest value and +1 representing the highest value. The three factors considered to have a significant influence on MB removal are: number of plates (A), applied voltage (B), and electroflocculation time (C). These parameters and values were chosen based on the findings of the preliminary studies conducted at the beginning of this research. The other factors, such as initial MB concentration and electrolyte concentration, were held constant at 20 ppm and 350 mg/L, respectively.

**TABLE 3.** Original and coded factors for the Box-Behnken design.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Factor | Original  factor (X) | Coded factor | | |
| **-1** | **0** | **1** |
| Number of plates | A | 2 | 4 | 6 |
| Applied voltage (V) | B | 10 | 15 | 20 |
| EF time (min) | C | 20 | 40 | 60 |

Table 4presents the model terms included in the Box-Behnken design. These consist of the direct effects of the three independent factors (A, B, and C), their interactions (AB, AC, and BC), and quadratic effects (A2, B2, and C2). The analysis of model terms produced standard errors with values in close proximity, indicating a balanced design. Factor independence and non-collinearity are demonstrated by a variance inflation factor (VIF) of 1.0. VIFs larger than 1.0 suggest multi-collinearity; the higher the VIF, the more severe the factor correlation. Additionally, R2 values of 0 indicate that these terms are not correlated with one another.

**TABLE 4.** Model terms included in the Box-Behnken design.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Term** | **Standard Error\*** | **VIF** | **Rᵢ²** | **Power** |
| A | 0.3536 | 1 | 0.0000 | 68.1 % |
| B | 0.3536 | 1 | 0.0000 | 68.1 % |
| C | 0.3536 | 1 | 0.0000 | 68.1 % |
| AB | 0.5000 | 1 | 0.0000 | 40.8 % |
| AC | 0.5000 | 1 | 0.0000 | 40.8 % |
| BC | 0.5000 | 1 | 0.0000 | 40.8 % |
| A² | 0.4873 | 1.00588 | 0.0058 | 93.8 % |
| B² | 0.4873 | 1.00588 | 0.0058 | 93.8 % |
| C² | 0.4873 | 1.00588 | 0.0058 | 93.8 % |

# Results and discussion

## Effects of operating parameters

*Applied Voltage.*

Applied voltage is a crucial variable for regulating the reaction rate within the EF reactor cell in all electrochemical processes. This parameter is well-known for determining the coagulant production rate and adjusting gas bubble formation rate, thereby influencing floc formation. To evaluate the impact of voltage on MB removal efficiency, an electroflocculation procedure was conducted at a fixed initial MB dye concentration (20 mg/L) and salt concentration of 400 ppm for a 45-minute operating time. In this study, the applied voltage range was 12 – 22 V. Figure 3shows the MB decolorization efficiency in relation to the applied voltage. The graph reveals that color removal significantly increases with increasing applied voltage. It can be observed that, for a given reaction time of 45 minutes, the removal increases from 75.6% to 81.0%, 93.7%, and up to 98.0% for applied voltages of 12, 14, 18, and 22 V, respectively.

The increase in voltage enhances color removal efficiencies for MB dye, and this finding aligns with existing results reported by other researchers.[19] The improved removal effectiveness of MB dye with increased applied voltage can be attributed to an increase in the number of ions generated by the dissolution of electrode materials, which then produced aluminum hydroxide. This process promotes the destabilization of pollutants and eventually leads to the agglomeration of induced flocs while enhancing hydrogen evolution.

**FIGURE 3**. Effect of voltage on color removal efficiency. (Operating parameters: time = 30 min, initial MB concentration = 20 ppm and NaCl concentration = 400 ppm)

*Electroflocculation time.*

As depicted in Fig. 4, increasing the reaction time leads to an increase in the color removal of the MB dye solution. In addition to applied voltage, electrolysis time is a crucial parameter that significantly influences the treatment efficiency of the electroflocculation process, as it is directly proportional to the metal hydroxides produced by the dissolution of electrodes.[20] To investigate this parameter, a series of tests were conducted at a constant voltage of 22 V.

As shown in Fig. 4, the color removal increases from 57.1% to 99.6% when the reaction time is extended from 10 to 45 minutes. Beyond 45 minutes, treatment efficiency remains nearly constant and exhibits no substantial increase. This finding aligns with previous research.[21] A longer reaction time results in increased floc production, leading to a higher MB percentage removal. When the time is further extended beyond the optimal reaction time, MB removal does not show any significant increase, which could be due to the sufficient metal hydroxide already generated to adsorb the dye pollutant. Consequently, 45 minutes was determined to be the ideal electroflocculation time, and this value was used in subsequent experiments.

**FIGURE 4.** MB color removal efficiency as a function of electroflocculation time (Operating parameters: voltage = 22 V, initial MB concentration = 20 ppm and NaCl concentration = 400 ppm)

*Electrolyte concentration.*

Previous research has identified a significant relationship between electrocoagulation efficacy and electrolyte concentration.[22] Various electrolytes, such as NaCl, KCl, MgCl₂, and MgSO₂, can be used; however, in this study, NaCl was chosen due to its low cost and easy availability. Experiments were conducted to determine the effect of electrolyte concentration on the EF removal efficiency by varying the doses of NaCl from 200 to 400 mg/L. Figure 5shows the influence of NaCl concentration on the percentage color removal of MB. The percentage color removal can be observed to increase from 91.6% to as high as 99.1% when the NaCl concentration is raised from 200 to 400 mg/L.

The increase in percentage color removal can be attributed to a rise in chloride ions in the solution. Indeed, the concentration of sodium chloride is directly proportional to the solution's conductivity. As the solution's conductivity increased, so did the current flow across the sacrificial electrodes, enhancing the effectiveness of MB dye removal. Moreover, due to their catalytic action, chloride ions not only improve the solution's conductivity but also reduce electrode passivity by eliminating the passivating oxide layer that forms on the surface of the electrodes.[23] This process makes more aluminum hydroxide available in the solution. Consequently, it is expected that the percentage removal of MB dye will be relatively high in the presence of abundant chloride ions.[24]

**FIGURE 5**. Effect of NaCl concentration on color removal efficiency of MB dye (Operating parameters: voltage = 22 V, time = 45 min and initial MB concentration = 20 ppm.

*Initial MB dye concentration.*

Utilizing the optimal parameters previously determined for applied voltage, electroflocculation time, and salt concentration, Fig. 6 shows the trend of color removal efficiency as a function of the initial MB dye concentration. To investigate the effect of initial dye concentration on EF process performance, dye solutions with different initial concentrations in the range of 20 to 40 mg/L were prepared by diluting the appropriate amount of a 1000 ppm stock solution. As anticipated, the results showed that as the starting concentration of dye increased, the dye removal efficiency declined. For instance, after 45 minutes of operating time, the percentage color removal decreased from 96.8% to 84.5% when the dye concentration increased from 20 to 40 mg/L.

This can be explained by the fact that with a constant applied voltage, the same quantity of aluminum ions is generated in the solution at varied MB dye concentrations. As a result, the amount of aluminum hydroxide produced was insufficient to destabilize the dye pollutants and facilitate the growth and formation of flocs.[25] Consequently, the reduction in removal efficiency with increasing dye concentration may be related to the need for additional coagulant as dye pollutant levels rise. These findings are consistent with those previously published by Daneshvar’s group,[26] where they studied the removal efficiency of Orange II dye using the electrocoagulation method and investigated the color removal efficiency with varying initial concentrations. They discovered that the percentage removal decreased to less than 28.15% when the starting concentration of Orange II increased from 50 to 400 ppm.

**FIGURE 6**. Effect of initial MB concentration on color removal efficiency (Operating parameters: voltage = 22 V, time = 45 min and NaCl concentration = 400 ppm.

*Number of electrodes.*

Figure 7shows the relationship between the number of electrode plates and the percentage removal of dye wastewater following the electroflocculation process. It can be observed that an increased number of electrode plates enhance the clarity of MB dye wastewater. The color removal result from treatment with two plates was 88.1%. The removal efficiency after using four electrodes reached 93.8%. Meanwhile, when employing six plates of electrodes, a maximum removal efficiency of 98.1% was attained.

The exposed surface area for each plate of electrode used in this study is 35 cm2. Utilizing a greater number of electrodes means the total area of electrodes that are immersed or exposed to the solution also increases. According to the literature, increasing the total area improves the dispersion of the active coagulant's density by generating more aluminum ions. This can lead to better coagulation of particles and, consequently, enhance the elimination of dye pollutants.[27]

**FIGURE 7**. Effect of the number of plates on color removal efficiency. (Operating parameters: voltage = 22 V, time = 45 min, initial dye concentration of 20 ppm and NaCl concentration = 400 ppm.

## Response surface methodology using Box-Behnken design (BBD)

In this section of the study, a Box-Behnken response surface design with three factors and three levels was applied to determine the optimal operating variables for decolorization of methylene blue dye. Table 5 presents the experimental design matrix, which consists of seventeen trials of electroflocculation experiments for the three analyzed factors, along with the values of the experimental and predicted response.

The most commonly used empirical model in the response surface method, which describes the relationship between design factors and response, can be expressed by the following second-order polynomial equation: [28]

+ (2)

Where represents the predicted response, which in this case corresponds to the color removal, denote the regression coefficients for the intercept, linear, quadratic, and interaction between input factors, respectively. and are the levels of independent factors in coded units, while ε represents the model's error.

**TABLE 5**. Box-Behnken design for three factors together with the experimental and predicted response.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Run** | **Coded factors** | | | **Response (%)** | |
| **A** | **B (V)** | **C (min)** | **Y (experimental)** | **Ŷ (predicted)** |
| 1 | -1 | 1 | 0 | 89.1 | 88.9 |
| 2 | -1 | 0 | -1 | 65.0 | 64.0 |
| 3 | 0 | 0 | 0 | 81.1 | 80.5 |
| 4 | 0 | 0 | 0 | 80.0 | 80.5 |
| 5 | -1 | -1 | 0 | 53.8 | 53.6 |
| 6 | 0 | -1 | 1 | 76.9 | 76.5 |
| 7 | 1 | 1 | 0 | 96.3 | 96.1 |
| 8 | 1 | 0 | -1 | 86.7 | 86.2 |
| 9 | 0 | -1 | -1 | 60.3 | 60.7 |
| 10 | 0 | 1 | -1 | 89.1 | 89.5 |
| 11 | -1 | 0 | 1 | 78.1 | 78.5 |
| 12 | 1 | 0 | 1 | 93.1 | 93.4 |
| 13 | 0 | 0 | 0 | 80.2 | 80.5 |
| 14 | 0 | 1 | 1 | 95.8 | 95.5 |
| 15 | 0 | 0 | 0 | 79.9 | 80.5 |
| 16 | 0 | 0 | 0 | 80.3 | 81.0 |
| 17 | 1 | -1 | 0 | 83.7 | 83.5 |

*ANOVA analysis: Quadratic versus Two-factor Interaction (2FI).*

The DOE software tested various models, and in this paper, ANOVA results for both quadratic and 2FI models are included. Analysis of variance (ANOVA) was used to evaluate the adequacy of each model and assess the correlation between factors and response before determining the most suitable model. Table 6 and Table 7 present the ANOVA results for quadratic and 2FI models, respectively. With p-values of <0.0001 and model F-values of 774.15 for quadratic and 1403.78 for 2FI (large value due to noise), both regression models are statistically significant. Although both models are acceptable, the software recommends using the 2FI model to achieve the 'goodness of fit.' This rationale can be explained by examining the significance p-values for each linear term (A, B, and C), interaction term (AB, AC, and BC), and quadratic term (A², B², and C²) on the response. The p-values indicate the importance of each coefficient and the effectiveness of the interaction between each independent factor. P-values less than 0.0500 suggest that the model terms are significant at the 95% confidence level.

The key difference observed between the ANOVA of both models is that the quadratic model contains three additional model terms not included in the 2FI model. These additional model terms are the squared interactions of each factor. ANOVA for the quadratic model reveals that the linear effects of A, B, and C, as well as the interaction effects of AB, AC, and BC, are significant model terms with p-values less than 0.001. Values larger than 0.1000 indicate that the model terms are not significant. In this case, the remaining model terms (quadratic effects of A², B², and C²) with p-values of 0.3839, 0.5973, and 0.902, respectively, can be considered statistically insignificant. Further improvement of the BBD design can be achieved by excluding these insignificant squared interactions, making the 2FI model a better option.

**TABLE 6.** ANOVA results for Quadratic model.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Source** | **Sum of Squares** | **df** | **Mean Square** | **F-value** | **p-value** |  |
| **Model** | 2232.35 | 9 | 248.04 | 774.15 | < 0.0001 | significant |
| A-Number of plates | 689.72 | 1 | 689.72 | 2152.70 | < 0.0001 |  |
| B-Voltage | 1141.94 | 1 | 1141.94 | 3564.13 | < 0.0001 |  |
| C-Time | 234.82 | 1 | 234.82 | 732.90 | < 0.0001 |  |
| AB | 128.28 | 1 | 128.28 | 400.37 | < 0.0001 |  |
| AC | 13.00 | 1 | 13.00 | 40.56 | 0.0004 |  |
| BC | 24.18 | 1 | 24.18 | 75.46 | < 0.0001 |  |
| A² | 0.2764 | 1 | 0.2764 | 0.8627 | 0.3839 |  |
| B² | 0.0981 | 1 | 0.0981 | 0.3061 | 0.5973 |  |
| C² | 0.0052 | 1 | 0.0052 | 0.0163 | 0.9020 |  |
| **Residual** | 2.24 | 7 | 0.3204 |  |  |  |
| Lack of Fit | 1.35 | 3 | 0.4508 | 2.03 | 0.2529 | not significant |
| Pure Error | 0.8904 | 4 | 0.2226 |  |  |  |
| **Cor Total** | 2234.59 | 16 |  |  |  |  |
| Adjusted Predicted | | | | | | |

**TABLE 7.** ANOVA results for Two-factor Interaction (2FI) model.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Source** | **Sum of Squares** | **df** | **Mean Square** | **F-value** | **p-value** |  |
| **Model** | 2231.94 | 6 | 371.99 | 1403.78 | < 0.0001 | significant |
| A-Number of plates | 689.72 | 1 | 689.72 | 2602.81 | < 0.0001 |  |
| B-Voltage | 1141.94 | 1 | 1141.94 | 4309.36 | < 0.0001 |  |
| C-Time | 234.82 | 1 | 234.82 | 886.15 | < 0.0001 |  |
| AB | 128.28 | 1 | 128.28 | 484.08 | < 0.0001 |  |
| AC | 13.00 | 1 | 13.00 | 49.04 | < 0.0001 |  |
| BC | 24.18 | 1 | 24.18 | 91.24 | < 0.0001 |  |
| **Residual** | 2.65 | 10 | 0.2650 |  |  |  |
| Lack of Fit | 1.76 | 6 | 0.2933 | 1.32 | 0.4122 | not significant |
| Pure Error | 0.8904 | 4 | 0.2226 |  |  |  |
| **Cor Total** | 2234.59 | 16 |  |  |  |  |
| Adjusted Predicted | | | | | | |

To assess the accuracy of the 2FI model, we can evaluate the determination coefficient. According to the data given in Table 7, the predicted R² of 0.9959 and adjusted R² of 0.9981 are closely related to one another with only a small difference, indicating that the 2FI model is in excellent agreement and can strongly predict the experimental data. The R² value of 0.9988 indicates that the suggested mathematical model can describe 99.88% of the total variation for MB color removal, with only about 0.12% of the total variation unexplained by this model. Based on the ANOVA results (Table 7), the Lack of Fit (F-value of 1.32) is not statistically significant compared to the pure error. A Lack of Fit F-value of this magnitude has a 41.22% chance of occurring due to noise. A non-significant Lack of Fit is desired because we want the model to fit.

Furthermore, the validity of the 2FI model was also ascertained using the diagnostic plot of predicted against experimental values, as shown in Fig. 8. The experimental values obtained and the predicted values calculated by the chosen model showed a linear relationship with very small differences between the two, signifying that both data generation methods were efficient and accurate. Based on these findings, we can conclude that the suggested 2FI model was adequate and effective for the study and optimization of MB color removal by electroflocculation.

A black background with colorful dots

Description automatically generated

**FIGURE 8.** The relationship between the experimental and predicted values of the response, Ŷ (CRE %).

*Model equation.*

Based on the ANOVA analysis, the 2FI model demonstrated a good fit with the data.Table 8 presents the estimated regression coefficients for all the linear and interaction effects of the factors on the percentage removal of MB. The predicted response of percent removal can be expressed by the following equation in terms of coded factors:

80.54 + 9.29A + 11.95B + 5.42C – 5.66AB – 1.80AC – 2.46BC (3)

**TABLE 8.** Coefficient estimates in terms of coded factors.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
| **Factor** | **Coefficient Estimate** | **df** | **Standard Error** | **95% CI Low** | **95% CI High** | **VIF** |
| Intercept | 80.54 | 1 | 0.1249 | 80.26 | 80.82 |  |
| A-Number of plates | 9.29 | 1 | 0.1820 | 8.88 | 9.69 | 1.0000 |
| B-Voltage | 11.95 | 1 | 0.1820 | 11.54 | 12.35 | 1.0000 |
| C-Time | 5.42 | 1 | 0.1820 | 5.01 | 5.82 | 1.0000 |
| AB | -5.66 | 1 | 0.2574 | -6.24 | -5.09 | 1.0000 |
| AC | -1.80 | 1 | 0.2574 | -2.38 | -1.23 | 1.0000 |
| BC | -2.46 | 1 | 0.2574 | -3.03 | -1.89 | 1.0000 |

*Model graphs.*

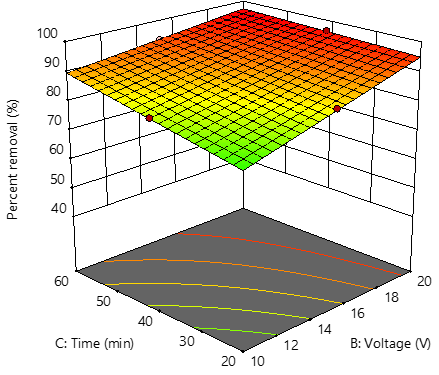
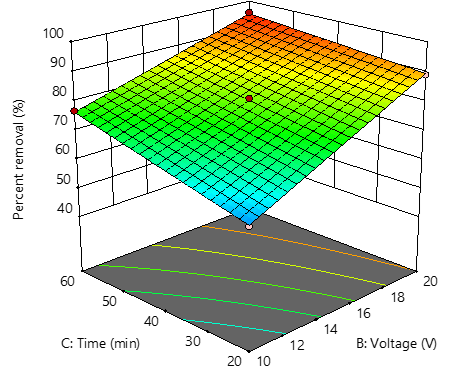
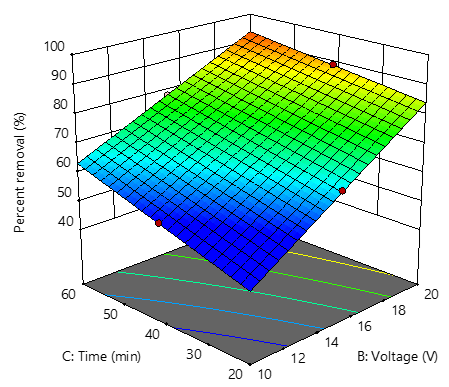
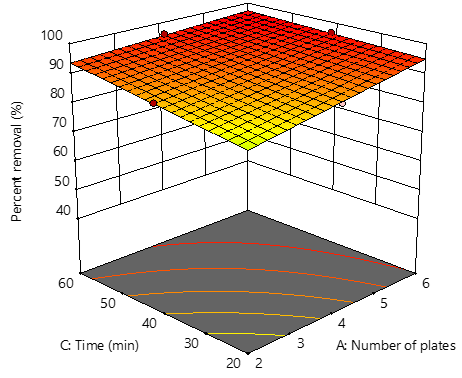
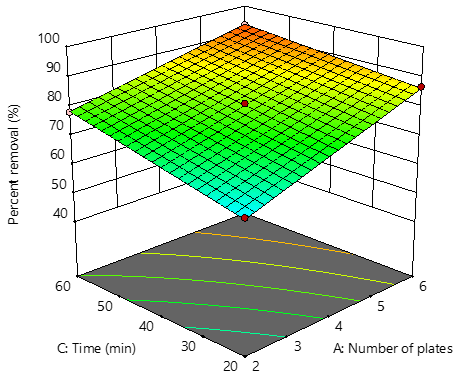
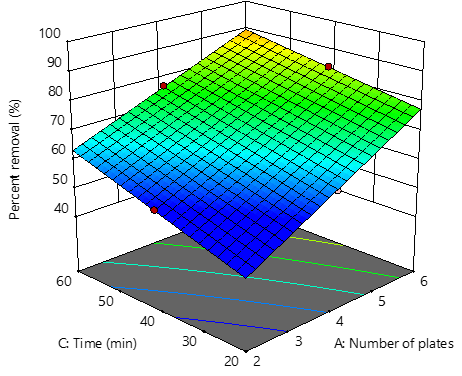
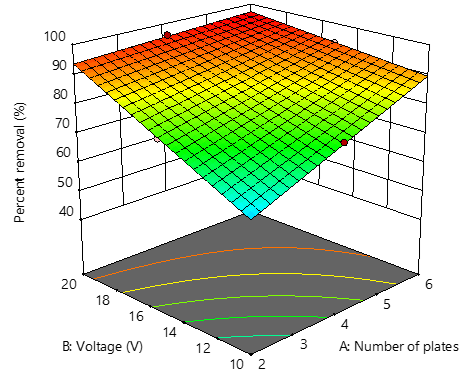
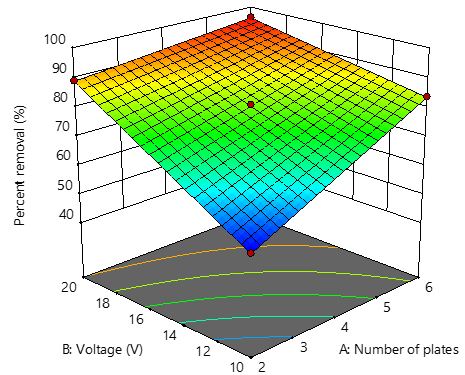
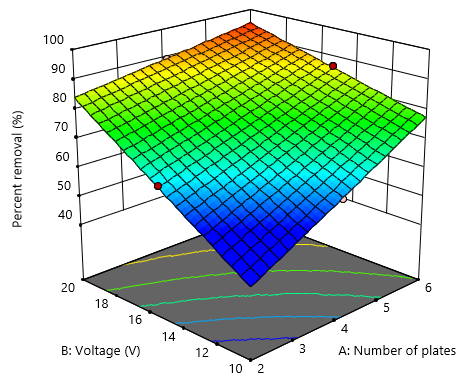
The model graphs in Fig. 9 depict the effects of percent removal resulting from the interaction of the three operating variables, as predicted by the model equation. In each case, two operational factors were analyzed while the other factor remained constant. The interaction between voltage (B) and the number of plates (A) is referred to as the AB interaction when the electroflocculation time is set to 20, 40, and 60 minutes (***a1, a2, and a3***). The interaction between electroflocculation time (C) and plate number (A) is specified as an AC interaction while the voltage remains at 10, 15, and 20 volts (***b1, b2, and b3***). Lastly, the interaction between electroflocculation time (C) and voltage (B) is referred to as the BC interaction, with the number of plates kept constant at 2, 4, and 6 (***c1, c2, and c3***).

*Model optimisation and confirmation*.

The software's numerical optimization was employed to determine the specific point that maximized the desirability function. In this study, the aim was to achieve a removal efficiency of 95.0%, with an acceptable range of ± 5.0%, while minimizing electroflocculation time and voltage and maintaining the number of plates at six. The desirable point prediction function in the experimental design generated four solutions that could be applied to reach the target removal efficiency of MB dye within the criteria set for both factors and response. Experiments were conducted under the suggested operating condition and three other predicted conditions to validate the model's validity. Table 9summarizes the outcomes for the removal efficiency of MB dye. As observed, the predicted results closely align with the experimental results, indicating that RSM can be effectively utilized to optimize operating parameters using statistical design of experiments.

**TABLE 9.**  Confirmation experiments using the solutions suggested by the model.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Number** | **Number of plates** | **Voltage** | **Time** | **Percent removal (%)** | | **Desirability** |  |
| **Experimental** | **Predicted** |
| 1 | 6.000 | 19.292 | 20.000 | 93.503 | 93.715 | 0.675 | Selected |
| 2 | 6.000 | 19.332 | 20.000 | 93.779 | 93.785 | 0.675 |  |
| 3 | 6.000 | 19.137 | 20.000 | 93.006 | 93.444 | 0.673 |  |
| 4 | 6.000 | 19.004 | 20.000 | 92.812 | 93.212 | 0.668 |  |



a1

b1

c1

a2

a3

b2

c2

b3

c3

**FIGURE 9.** 3D surface plots for interactive effect between two variables (voltage and number of plates, time and number of plates, time and voltage) at constant: (a1) V = 10V; (a2) V = 15V; (a3) V = 20V; (b1) EF time = 20 mins; (b2) EF time = 40 mins; (b3) EF time = 60 mins; (c1) 2 plates; (c2) 4 plates; (c3) 6 plates on color removal efficiency.

# Conclusion

This study successfully developed a mathematical model to optimize and evaluate the effects and interactions of three primary operating parameters—number of plates, applied voltage, and electroflocculation time—on the decolorization of methylene blue (MB) dye using electroflocculation in a batch reactor. The response surface methodology with the Box-Behnken design led to a highly accurate Two-factor Interaction (2FI) model, as evidenced by the high R² value of 0.9988. This model effectively predicts removal efficiency and optimizes operating parameters for MB decolorization.

Beyond its technical achievements, this research offers significant contributions to the treatment of dye-contaminated wastewater in the textile industry. By identifying optimal conditions for electroflocculation, including a reaction time of 20 minutes, an applied voltage of 19.29 V, and six aluminum electrode plates, the study demonstrates a practical and efficient approach to achieving high color removal efficiency (93.50% experimentally and 93.72% predicted). These results highlight the potential for the optimized EF process to improve wastewater treatment outcomes, reducing environmental impact while offering a cost-effective and sustainable solution.

Furthermore, this work aligns with broader industry goals of regulatory compliance and sustainable practice adoption. The reduced reliance on chemicals, minimal sludge production, and lower space requirements of the EF process make it an attractive alternative for industrial-scale implementation. As such, this research not only advances knowledge in electroflocculation optimization but also provides actionable insights for the textile industry to address its environmental challenges effectively.

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