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Article

Process Optimization of Chitosan Adsorption of Methylene Blue Dye from Aqueous Solution Using a Central Composite Design

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Abstract

This study investigates the potential of commercial chitosan, a natural and eco-friendly biosorbent, for the removal of methylene blue (MB) dye from synthetic wastewater. Experiments were designed and optimized using central composite design (CCD) to evaluate the interactive effects of initial dye concentration (6.39–12.79 mg/L) and contact time (10–120 min) on removal efficiency. Statistical analysis (ANOVA) revealed that the quadratic model was highly significant (F-value = 128.88, p-value = 0.0002) with an excellent coefficient of determination (R^2 = 0.9938) and adjusted R^2 = 0.9861, indicating strong agreement between experimental and predicted values. The derived quadratic equation accurately described the relationship between the variables. Optimization results showed that the maximum MB removal efficiency of 71.74% was achieved at an initial MB concentration of approximately 7.17 mg/L and a contact time of approximately 103.81 min. These findings confirm that chitosan is a promising, low-cost, biodegradable adsorbent for cationic dye removal, offering a sustainable solution for industrial wastewater treatment.

1. Introduction

The rapid expansion of textile, printing, dyeing, and related industries has led to the discharge of large volumes of wastewater containing synthetic organic dyes, which pose severe threats to aquatic ecosystems and human health[1, 2, 3]. These effluents are characterized by high chemical oxygen demand (COD), intense colour, low biodegradability, and the presence of toxic, carcinogenic, and mutagenic compounds. Even at very low concentrations, dyes can reduce light penetration in water bodies, disrupt photosynthesis in aquatic plants, and alter the ecological balance of receiving waters. Among the various classes of dyes, cationic dyes such as methylene blue (MB), are among the most common organic pollutants discharged in wastewater from textile, printing, and dyeing industries[4]. Due to its high chemical stability, potential toxicity, and visibility even at low concentrations, MB poses serious environmental and health risks to aquatic ecosystems. Although conventional treatment methods (e.g., chemical coagulation, advanced oxidation processes) can be effective, they are often expensive, energy-intensive, and may generate secondary sludge or toxic by-products[1, 5].

Methylene blue, a thiazine dye commonly employed in textile processing, paper manufacturing, and biological staining, exhibits high solubility in water and resistance to conventional biological treatment processes[6]. Its discharge into natural water systems not only causes aesthetic pollution but also leads to adverse effects on aquatic organisms, including reduced growth rates, respiratory distress, and bioaccumulation in the food chain. Moreover, prolonged exposure to MB in humans has been linked to methemoglobinemia, skin irritation, and potential carcinogenic risks. Given these environmental and health concerns, effective, economical, and sustainable methods for MB removal from industrial effluents have become a priority in wastewater treatment research[7].

Chitosan, a linear polysaccharide derived from the deacetylation of chitin (primarily sourced from crustacean shells such as shrimp), stands out as an excellent natural biosorbent for cationic dyes like methylene blue. Its chemical structure features abundant amino ($-NH_2$) and hydroxyl (-OH) functional groups, which become protonated in acidic to neutral media, enabling strong electrostatic interactions with positively charged dye molecules[8, 9].

The present study focuses on the adsorptive removal of MB using commercial chitosan under controlled batch conditions. central composite design (CCD) was applied to systematically investigate and optimize the effects of the two most influential operational parameters initial dye concentration and contact time while developing a reliable quadratic mathematical model to predict removal efficiency. This work aims to identify the optimal conditions for maximum MB removal, elucidate key adsorption trends, and contribute to the development of practical, low-cost, and environmentally benign technologies for treating dye-containing industrial wastewater.

2.Materials and Methods

2.1. Materials

Chitosan was obtained from Aldrich (Aldrich Co. Steinheim, Ger-many), those being chitosan (powder, shrimp shells, product number 50494). Methylene blue ($C_{16}H_{18}ClN_3S$, 82%) was acquired from Biochem Chemophara.

2.2. MB dye removal

Utilizing the CCD design and the statistical program Design Expert ver.13, the test was designed. Table 1 displays the factors used in this research. By adding a defined amount of chitosan to 0.5 g flasks that held 30 mL of dye solution at various concentrations (6.39–12.79 mg L^{-1}) for different times (10-120 min). Equilibrium tests were carried out). A temperature of 25°C was maintained while the mixture was shaken in an orbital shaker at 150 rpm. Centrifugation through the liquid at a speed of 6000 rpm was then used to separate the solid phase. The UV/VIS spectrophotometer was used to compare the MB dye concentration before and after the adsorption operations at a maximum wavelength of 664 nm. MB removal efficiency (%) was calculated using the formula below [10] .

$$Dye\ removal(\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (1)$$

where C_o and C_e, respectively, represent the starting and equilibrium dye concentrations (mg L^{-1}).

Table 1: levels of experiment design for diverse factors.

Factors	Codes	Level		
		-1.0	0.0	1.0
MB concentration (mg L^{-1})	A	6.39	9.59	12.79
Time (min)	B	10.00	65.00	120.00

The second-order quadratic polynomial equation for a Central Composite Design (CCD) with two independent factors (k = 2) can be expressed as follows[11]:

$$Y = \beta_0 + \sum_{j=1}^{k} \beta_j x_j + \sum_{j=1}^{k} \beta_{jj} {x_j}^2 + \sum \sum_{i<j=2}^{k} \beta_{ij} x_i x_j + \varepsilon \quad (2)$$

Where ε represents the error, x_i and x_j represent variables. The linear, binominal, and combination effects are represented by the coefficients β_i, β_{ii} , and β_{ij}, respectively. β_0 is a constant coefficient, and while k denotes the total number of studied factors (k=2)[12].

2.3. Analytical and Laboratory Equipment

Dye absorption spectra were recorded by ultraviolet spectroscopy (UV-2450, Shimadzu, Germany). Phase separation of the solid-liquid mixture after experiments was performed using a high-speed benchtop centrifuge (Model Hermle Z326, Hermle Labortechnik GmbH, Wehingen, Germany) operated at 6000 rpm. All weight measurements were carried out on a high-precision analytical balance.

3. Results and discussion

3.1. Experimental Design Matrix and Responses

The experimental findings for the methylene blue removal parameters are displayed in Table 2. The numbers obtained from the anticipated model are also shown in the table, illuminating that the proposed model and the actual results are not considerably different.

Table 2: Design matrix for various factor values and experimental responses.

Experiment order	Experimental factors		Response (MB Removal)	
	MB concentration (mg L^{-1})	Time (min)	Actual	Predicted
1	6.39	65	56.3	56.51
2	9.59	65	61.45	63.29
3	9.59	120	85.6	82.17
4	6.39	120	75.45	75.39

5	6.39	10	36.8	37.62
6	9.59	10	46.6	44.41
7	9.59	65	61.44	63.29
8	12.79	120	87.44	88.95
9	12.79	65	70	70.07
10	12.79	10	51.8	51.19

3.2. Fitting of model

Table 3 presents the ANOVA results for the quadratic model describing methylene blue (MB) dye removal using chitosan as the adsorbent. The coefficient of determination $R^2 = 0.9938$ is very close to 1, indicating excellent agreement between the experimental data and the predicted values from the model meaning there is nearly perfect correspondence between observed and model-predicted responses. Similarly, the adjusted $R^2 = 0.9861$ is also very high and close to 1, confirming that the model fits the data exceptionally well after accounting for the number of terms, with minimal unnecessary complexity.

The overall model is highly significant, as evidenced by the model F-value of 128.88 and a corresponding p-value of 0.0002 (well below the typical threshold of 0.05). This strongly rejects the null hypothesis that the model has no explanatory power, demonstrating that the quadratic model reliably captures the relationship between the independent variables (MB concentration and time) and the response (MB removal percentage).

Furthermore, the relationship between the F-value and p-value for individual terms provides insight into their effects: an inverse correlation exists, where higher F-values correspond to lower p-values and thus greater statistical significance of a term's influence on MB removal. In this case, the linear terms for MB concentration (A) and time (B) exhibit highly significant effects ($p < 0.05$), underscoring their dominant roles in the adsorption process, while interaction and quadratic terms remain non-significant.

Despite these strong fit indicators (high R^2 values and significant model overall), the significant Lack of Fit ($p = 0.0023$) warrants careful consideration. This indicates that the quadratic model does not fully capture all systematic variations in the data beyond pure experimental error a situation that can arise when pure error is extremely low (excellent replication at center points), yet subtle deviations persist at other design points. While the exceptionally high R^2, adjusted R^2, and adequate precision (34.145 > 4) suggest the model remains useful for navigation within the design space and for identifying key trends/trends, the significant Lack of Fit advises caution in precise predictions or optimization. Model refinement (e.g., exploring higher-order terms if feasible, outlier checks, or confirmation experiments at predicted optima) is recommended to enhance reliability before practical application. Overall, the model demonstrates robust explanatory power and statistical significance, supporting its utility in understanding and guiding the chitosan-based MB removal process.

Table. 3 Removal using ANOVA variance analysis.

Source	**Sum of Squares**	**df**	**Mean Square**	**F-value**	**p-value**
Model	2426.75	5	485.35	128.88	0.0002
A-MB concentration	275.95	1	275.95	73.27	0.0010
B-Time	2139.10	1	2139.10	568.02	< 0.0001
AB	2.27	1	2.27	0.6015	0.4813
A^2	2.89	1	2.89	0.7683	0.4302
B^2	7.87	1	7.87	2.09	0.2218
Residual	15.06	4	3.77		
Lack of Fit	15.06	3	5.02	1.004E+05	0.0023
Pure Error	0.0001	1	0.0001		
Cor Total	2441.81	9			
R^2	0.9938				
Adjusted R^2	0.9861				
Predicted R^2	0.9566				
S.D	1.94				
CV	3.07				

The analysis has shown that a quadratic polynomial model's equation, which expresses the responses in coded factors, links the responses with the factors under study. This model is given below:

$$\text{MB Removal (\%)} = +62.85 + 6.78\ \text{A} + 18.88\ \text{B} - 1.11\text{A}^2 + 1.84\ \text{B}^2 - 0.7525\ \text{AB} \quad (3)$$

A is MB concentration (mg L^{-1}), and B is the Time (min).

The mathematical signs and magnitude of the coefficients in Equation 3 provide crucial insights into the individual and interactive effects of the operational parameters on Methylene Blue (MB) removal efficiency. The positive coefficients for the linear terms +6.78 A (MB concentration) and +18.88 B (Contact time) clearly demonstrate that increasing both initial dye concentration and contact time individually enhances the absolute removal efficiency within the evaluated experimental design range. Conversely, the negative coefficient for the interaction term (-0.7525 AB) suggests a slight antagonistic interaction, meaning that the time required to achieve a specific removal percentage shift at higher initial concentrations. However, according to the ANOVA results (Table 3), this interaction term exhibits a high p-value of 0.4813, which confirms that there is no statistically significant interactive effect between MB concentration and contact time on the adsorption efficiency. The adsorption process is largely dominated by the independent influences of the linear factors [18]. Plots of the expected vs. actual values for the MB dye removal reaction are shown in Figure 1 demonstrates that the response-predicted values are very near to the experimental values, demonstrating the accuracy of the prediction made by the model.

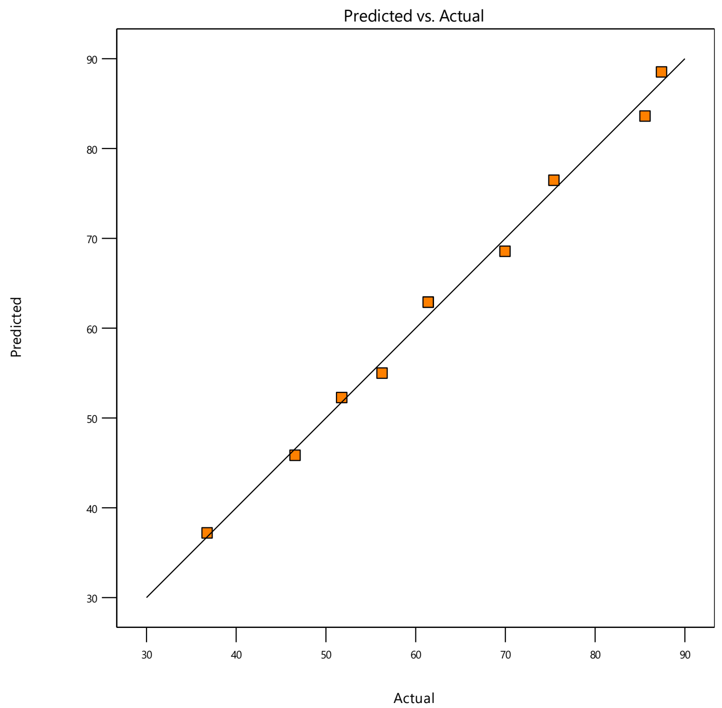

Fig. 1 Predicted vs. experimental values for removal of MB.

3.3 Internal vs. Normal

Figure 2 normal probability plot, which indicates whether the data are normally distributed, is particularly useful. If the data is regularly distributed, then a linear pattern indicates this and vice versa. The data, in this case, is roughly distributed in a straight line, indicating that it is regularly distributed[13].

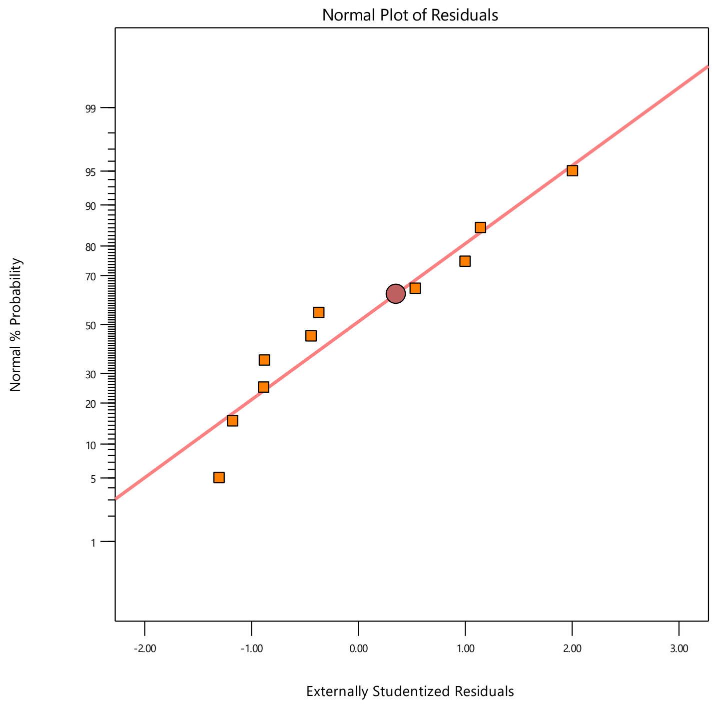

Fig .2 The normal % probability residuals and studentized residuals for MB dye removal.

3.4. Impact of process factors on MB removal

Figure 3 illustrates 3D drawings of the response surfaces, and how each of the three independent parameters affects (MB concentration, and time) affected the removal of MB. Understanding the individual and combined impacts of the input factors on the replies is made possible by the response surface analysis. The maximum removal efficiency of MB Removal was 71. 74% at MB concentration of 7.17 mg L^{-1}, and the contact time of 103.81 min.

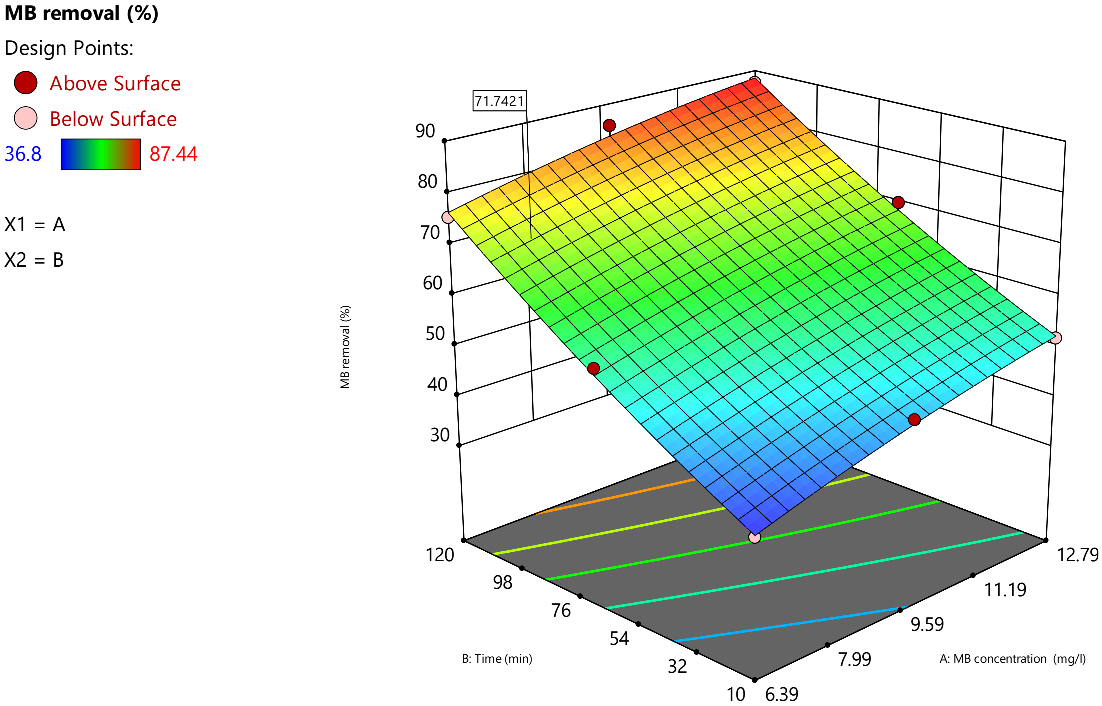

Fig .3 3-D Surface contour plot for the effects of MB concentration & time on % adsorption of MB.

3.5.Optimization Using the Desirability Functions

The optimization of methylene blue dye removal was carried out utilizing the desirability functions. The Design Expert 13 program enables the setting of optimization objectives to minimize, maximize, achieve the desired value or find a solution within a certain response range. In this study, all of the factor ranges were placed between their minimum (1) and maximum (+1) values with the optimization aim of maximizing MB elimination. The outcomes of the mathematical model in Figure 4 showed a maximum desirability of 1.00 utilizing the prior optimization criteria.

Using these conditions, the maximum MB Removal (%)= 71.74 %, experimental) obtained corresponds to the MB concentration (A) of ~ 7.17 mg L^{-1} and the reaction time (C) of ~103.81 min.

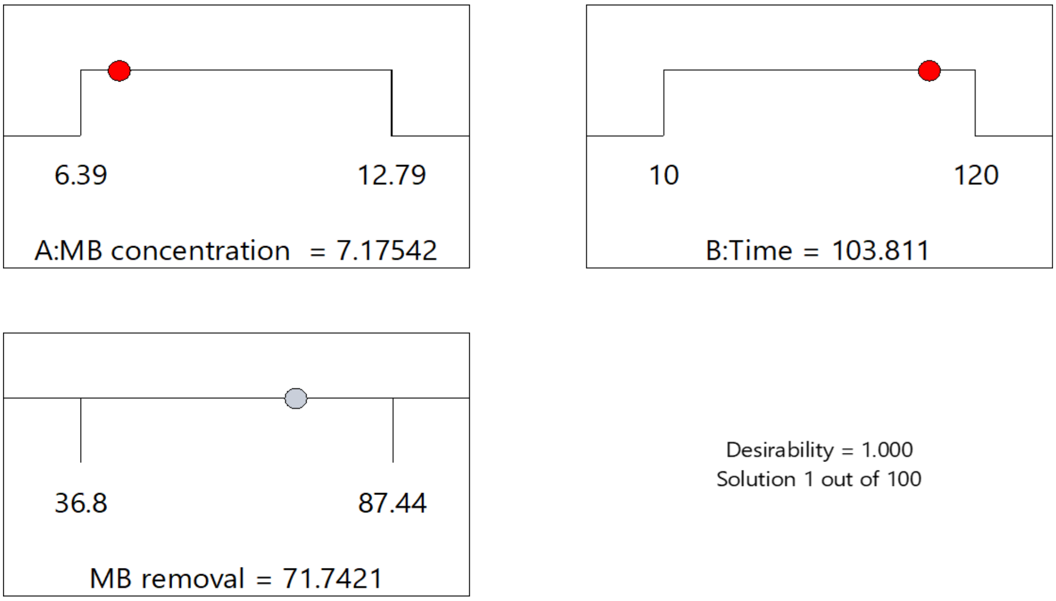

Fig .4 Desirability ramp for optimization.

4.Conclusion

This study demonstrates that chitosan is an effective, environmentally friendly, and low-cost biosorbent for the removal of methylene blue from aqueous solutions. Through the application of a CCD a highly significant quadratic model (R^2 = 0.9938) was developed, accurately capturing the influence of initial dye concentration and contact time on removal efficiency. Optimization results revealed that the highest removal percentage of 71.74% could be achieved at an initial MB concentration of ~7.17 mg/L and a contact time of ~103.81 min. These findings highlight the strong potential of chitosan as a viable alternative to conventional adsorbents, particularly in regions with abundant natural raw materials.

Future research should focus on evaluating chitosan performance in real industrial effluents (containing salts, organic matter, and multiple pollutants), investigating regeneration and reusability to improve economic feasibility, and exploring surface modification strategies to further enhance adsorption capacity and performance at higher dye concentrations.

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Ethical Statement

This research does not contain any studies with human or animal subjects performed by any of the authors

Data Availability Statement

Not Applicable

Conflicts of Interest

The authors declare no conflicts of interest

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