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Application of the Taguchi Method for Optimizing the Process Parameters for the Removal of Sulfur Black dye

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ABSTRACT

In this study, the parameters for the adsorption of sulfur black dye onto ZnO and ZnO-ME nanoparticles were optimized in a batch system (ZnO-nanomaterials synthesized using bacterial extracts and ZnO-ME-nanomaterials synthesized using bacterial cell mass). For estimating the significant and interaction effects of several investigated factors (initial concentration of sulphur black dye, adsorbent dose and contact time) on the adsorption process, the Taguchi optimization approach (an L9 array design) was used. It was discovered that 15 mg of adsorbent, 55 mg/l of initial dye concentration and 2 h of contact time at pH 7 and room temperature were the optimum conditions. Using the analysis of variance (ANOVA) method, the percentage contribution of each process parameter to the removal of sulphur black dye was estimated. It was discovered that Taguchi predicted well results. It was discovered that the percentage removal of the sulphur black dye obtained in the confirmatory experiments conducted under optimized conditions was higher than that optimized in each of the Taguchi design's test runs, supporting the accuracy of the process parameter optimized under the specified experimental conditions.

Key words: ZnO nanomaterials, sulfur black dye, Taguchi, analysis of variance, optimization

INTRODUCTION

In recent decades, industrialization has a substantial impact on the global economy, but it has also had a number of negative environmental repercussions (Patnaik, 2018). The release of dangerous chemicals into the environment through industrial wastewaters is a major source of pollution (Shindhal *et al.*, 2021; Yadav *et al.*, 2022). One of the main causes of water pollution is the dumping of coloured chemicals. Since these dyes are typically released in natural streams that people use for daily activities like drinking, washing and taking showers, this type of pollution may result in several severe issues for people. Most importantly, these wastewaters pose serious health risks to people since they are poisonous and cancer-causing. Additionally, the introduction of synthetic dyes into natural streams has detrimental impacts on the photochemical processes that occur in aquatic environments by obstructing the passage of light (Zolgharnein and Rastgordani, 2018). As a result, in order to prevent the biological ecosystem from being contaminated, it is imperative that dyes be removed from all discharges due to increased awareness of their toxicity and strict environmental safety standards (Pundir *et al.*, 2018). A class of dyes known as sulphur dyes have been linked to considerable amounts of water pollution because they are resistant to biodegradation. These cancer-causing chemicals drastically alter the natural food chain in the environment and have several detrimental impacts on aquatic species. The design and development of new industrial techniques with high efficacy and reusability to treat these wastewaters before releasing them into the environment is a crucial requirement despite the development of various chemical, physical, and biological methods such as coagulation/ flocculation, ozonation, electrochemical methods, biological treatment, photocatalyst and adsorption for the treatment of wastewaters containing sulphur dyes in the last decade (Haghgir *et al.*, 2022).

Adsorption, one of these techniques, has demonstrated highly promising effectiveness in removing various pollutants from wastewater streams with high efficacy (Abbasi, 2020). A range of synthetic and natural materials can be used as adsorbents in the simple, effective and affordable adsorption method (Mosoarca *et al.*, 2022b, c). Activated carbon, lignin, zeolite, metal-organic framework, biomass residue, nanomaterials, etc. are examples of commonly used adsorbents. The use of nanomaterials in the realm of the environment has increased recently due to the rapid growth of nanotechnology (Sultan *et al.*, 2022; Wang *et al.*, 2022). Compared to their commercial analogs, nano-sized materials are destructive sorbents with several advantages, such as strong surface reactivity and simplicity of synthesis from available natural materials. They have also been demonstrated to be superior materials for adsorbents (Janani *et al.*, 2022; Mansour *et al.*, 2022). Time, initial dye concentration, pH, adsorbent dose, temperature and ionic strength influence the adsorption process. These parameters should be optimized to get the best dye removal efficiency, if possible. For adsorption, experiment condition optimization is crucial. The Taguchi approach is one of the most effective techniques for identifying an optimum design configuration for multifactor situations (Babji *et al.*, 2022; Mosoarca *et al.*, 2022a). The literature has many past studies that optimize dye removal using various optimization strategies.

Nevertheless, a few papers suggest using Taguchi designs to improve dye removal. Abbas *et al.* (2022) and Mahapatra and Kumar (2022), also found the Taguchi method was appropriate for the optimization process. This study used zinc oxide nanoparticles as an adsorbent to remove the sulfur black dye from the synthetic water through adsorption. One of the main aims was to identify the optimum adsorption parameters using the Taguchi technique after examining the effects of several controllable elements on the effectiveness of the process and the signal-to-noise ratio (S/N). The ANOVA analysis was used to calculate the percentage contribution of each parameter to the dye removal process. Additionally, it was determined that Taguchi model forecasts were accurate.

MATERIALS AND METHODS

UV-visible spectroscopy was used to calculate the initial and final concentrations of sulfur black dye in the solutions, and 600 nm was used as the analytical wavelength. 1000 mg/l was used to generate the standard stock solution for sulphur black dye with the working ranges of 10, 20, 40, 60, 80 and 100 mg/l. Samples absorbance was measured, and the concentration was calculated using the calibration curve generated by the produced standard solutions of sulfur black dye. The percentage of sulfur black dye adsorption by zinc oxide nano-adsorbents was expressed as follows:

$$\% \text{ b dye removal} = \frac{Co - Ce}{Co} \quad ...(1)$$

Where Co was the initial concentration and Ce (mg/l) was the final concentration in the solution (Ghosh and Sinha, 2015).

As independent (input) variable, the initial sulfur black dye concentration, contact time and adsorbent dosage were utilized to examine the effects of each on the removal of sulfur black dye from the aqueous matrix. Based on the literature, these parameters and their range were chosen (Palai *et al.,* 2021) and according to some preliminary experiments. The pH and temperature conditions were set at 7 and 28°C, respectively, to make the adsorption process more cost- and environmentally friendly.

The initial sulfur black dye solution range was chosen to be 10-100 mg/l because most traditional technologies were ineffective for treating dye pollution at low concentrations due to high running costs (Ghosh and Sinha, 2015). Thus, the range of initial sulfur black dye concentration, contact time, and adsorbent dosage used are shown in Table 1. Using Minitab software version 21, all statistical analyses for optimizing experimental variables were examined.

Table 1. Range of the investigated parameters

Parameters	Range	
	Low	High
Dose (mg)	5	15
Conc. (mg/l)	10	100
Contact time (min)	60	180

The variance analysis (ANOVA) was used to determine how well these models fitted the experimental data. This method was used to determine the statistical significance of the models and the interaction effects of each factor on the responses using the Fischer (F-test) test with a 95% confidence interval. The decision was made under the assumption that a model was a suitable approach to the actual data, the error resulting from lack of fit was insignificant, and the variation brought on by the regression was substantial at a 95% confidence level. Additionally, the model's appropriateness was evaluated using the regression coefficient (R^2), corrected regression coefficient (ADJ-R^2), predicted multiple determination coefficient (Pre-R^2), and the sum of squares of predicted residual errors (PRESS).

The engineer and statistician Genichi Taguchi created the Taguchi technique, a fractional factorial design based on an orthogonal design array. With the use of this design, it was possible to do a limited number of tests to look into how different elements affect reactions at multiple levels. Organization of the variables and the levels at which they should be altered was made easier by the orthogonal array architecture. Additionally, the Taguchi technique only examined pairs of combinations, whereas the factorial design evaluated all potential combinations, saving time, money and resources. These arrays were used to identify key factors in an experiment and forecast how variables and operational circumstances interact (Sy Mohamad *et al.*, 2020).

Additionally, analyzing the signal-to-noise ratio (S/N) was crucial for evaluating the experimental outcomes. Generally speaking, there were three alternative signal-to-noise ratios (S/N) that can be used: lower was better, nominal was best and larger was better. Signal-to-noise ratio was yielded with following equation:

$$S/N = -10\log_{10}[1/2å(1/PRE_i) \qquad ...(2)$$

Where, n was the number of experiments conducted under similar experimental conditions, and PRE denoted the outcomes of the measurements (Ghosh and Mondal, 2019).

RESULTS AND DISCUSSION

The optimum conditions for sulfur black dye adsorption were determined using the Taguchi method. The terms "signal" and "noise" in the Taguchi technique denote the desired and unwanted values for the output characteristic, respectively. The signal-to-noise (S/N) ratio is used by the Taguchi technique to quantify quality characteristics that deviate from the target value. Depending on the type of characteristic, the S/N ratios vary. The initial dye concentration, contact time and adsorbent dose were the three factors used in an orthogonal array of Taguchi experimental designs with two levels. The present experiment concentrates on the "larger the better" choice and, according to the Taguchi technique, evaluating experimental outcomes necessitated an analysis of the signal-to-noise ratio. The optimal levels of the parameters for getting the maximal response variable (dye adsorption %) were identified and are displayed in Fig. 1 according to the S/N ratio as the primary effect graph for the SN ratio. The main effect graphs showed the findings of the regression analysis. Only the significant

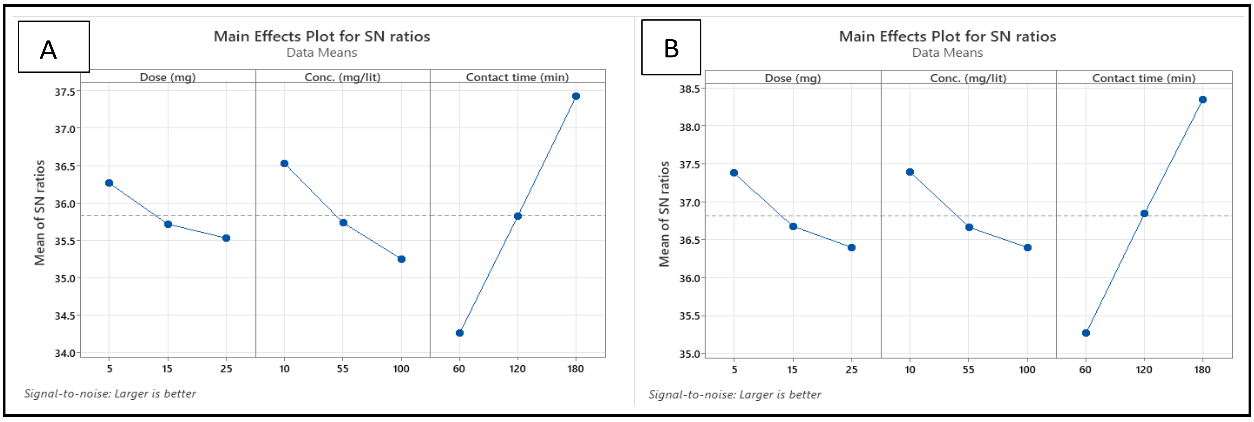

Fig. 1. Main effect plot for SN ratio (A) ZnO and (B) ZnO-ME nanoparticles.

variables at the 95% confidence level were displayed.

The adsorption rate of sulfur black dye was determined by regression analysis (Eqs. 3 and 4). Analysis of variance (Table 2) and model statistic summary (Table 3), as well as model graphs, including 3D graphs and predicted vs actual value plots, were used to express the mathematical model fitting and statistical analysis of observed experimental data of both nanoparticles for sulfur black dye adsorption (%). A Student's t-test with a 95% level of confidence was used to assess the significance of the regression coefficients. Additionally, the model showed an adjusted square correlation coefficient R^2 (adj) for ZnO and ZnO-ME nanoparticles of 97.73 and 97.82%, respectively, which well fitted the statistical model.

Dye adsorption (ZnO-ME) = 55.67 - 0.3667 Dose (mg) - 0.0778 Conc. (mg/lit) + 0.2028 Contact time (min) ...(3)

Dye adsorption (ZnO-ME) = 49.04 - 0.2333 Dose (mg) - 0.0926 Conc. (mg/lit) + 0.1861 Contact time (min) ...(4)

The accuracy of the anticipated adsorption % data using the Taguchi approach is shown in Fig. 2. Although Taguchi experimental design is based on a small number of studies, it can be seen that it reasonably predicted the adsorption of sulfur black dye using ZnO and ZnO-ME nanoparticles. Whether these outcomes were "real" or "random" was unclear. The "actual" impacts were determined using a normal probability plot.

On the Pareto chart, the relative significance of the main effects and their interactions were also visible (Fig. 3). These values for each effect are shown in the Pareto chart by horizontal columns. A student's t-test was used to assess whether the calculated effects differed statistically from zero. Significant values were those that were above a reference line or those that fell inside the 95% confidence interval. Fig. 3 shows the three main components that go beyond the reference line (A, B and C) were significant at the 0.05 level.

Table 2. Analysis of variance (ANOVA) for the adsorption of sulfur black dye using ZnO and ZnO-ME nanoparticles

Source	ZnO					ZnO-ME					
	d. f.	Adj	Adj	F-Value	P-Value	Source	d. f.	Adj	Adj	F-Value	P-Value
Regression	3	885	295	71.76	0	Regression	3	1042.33	347.444	74.81	0
Dose (mg)	1	32.67	32.667	7.95	0.037	Dose (mg)	1	80.67	80.667	17.37	0.009
Conc. (mg/lit)	1	104.17	104.167	25.34	0.004	Conc. (mg/lit)	1	73.5	73.5	15.83	0.011
Contact time (min)	1	748.17	748.167	181.99	0	Contact time (min)	1	888.17	888.167	191.23	0
Error	5	20.56	4.111			Error	5	23.22	4.644		
Total	8	905.56				Total	8	1065.56			

Table 3. Model summary statistics

Model	R^2	Adjusted R^2	Predicted R^2	Std. Dev	Mean
Taguchi (ZnO)	0.9773	0.9637	0.9118	2.0275	62.78
Taguchi (ZnO-ME)	0.9782	0.9651	0.9132	2.1551	70.22

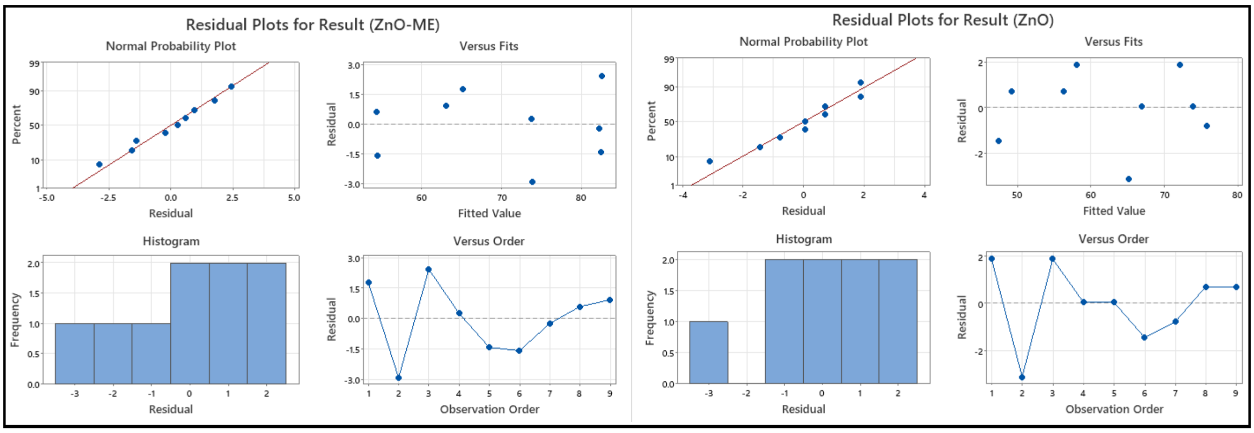

Fig. 2. Residual plots for PBD for ZnO and ZnO-ME nanoparticles.

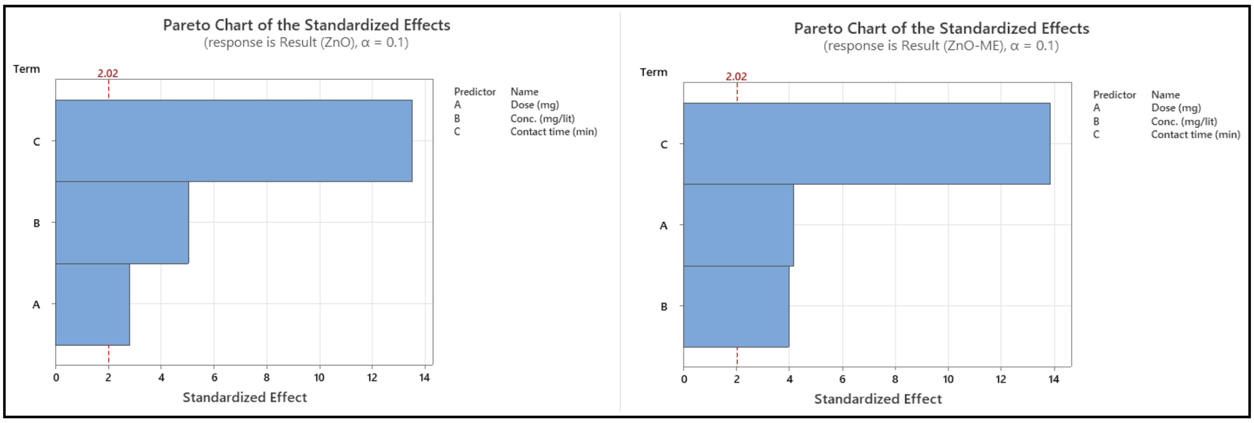

Fig. 3. Pareto plots of ZnO and ZnO-ME nanoparticles.

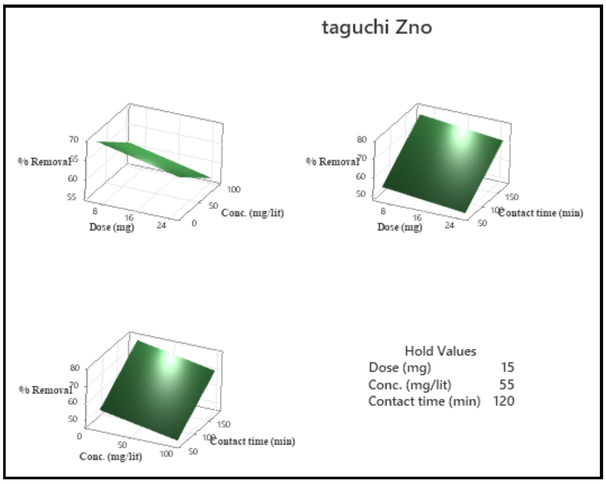

Fig. 4. Three-dimensional response surfaces for sulfur black dye adsorption using ZnO nanoparticle.

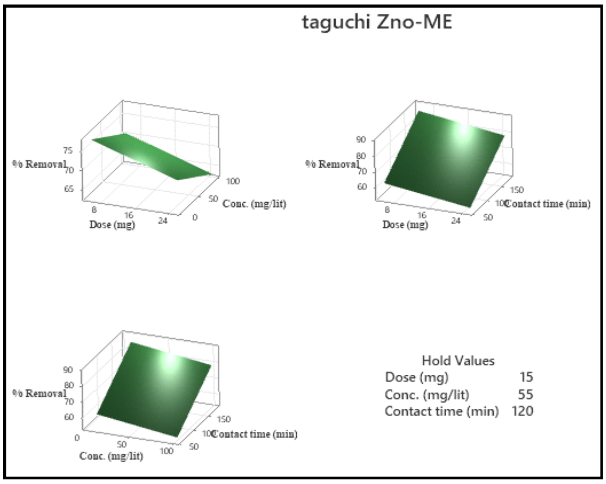

Fig. 5. Three-dimensional response surfaces for sulfur black dye adsorption using ZnO-ME nanoparticle.

The simultaneous impacts of the experimental variable on the dye adsorption process are better understood by looking at surface plots of the adsorption % versus various pairs of effective parameters. Figs. 4 and 5 depict the combined effects of initial dye concentration, contact time and adsorbent dosage.

CONCLUSION

The current study concentrated on utilizing ZnO and ZnO-ME nanoparticles as adsorbents for sulfur black dye removal from synthetic aqueous solution. Using batch mode experiments, the process variables for the adsorption of the sulfur black dye were optimized by the Taguchi method. The process parameters were optimized for the maximum per cent removal of sulfur black dye using the Taguchi experimental design with an L9 orthogonal array. The following was the order in which each parameter contributed to the per cent removal of sulphur black dye: contact time > initial dye conc. > adsorbent dose for ZnO and contact time > adsorbent dose > initial dye conc. for ZnO-ME. The confirmation experiment's dye removal percentage was higher than all test runs. To increase the percentage of sulphur black dye removal, process parameters were optimized accurately. Additionally, the model showed an adjusted square correlation coefficient R^2 (adj) for ZnO and ZnO-ME nanoparticles of 97.73 and 97.82%, respectively, which fitted the statistical model well. In this study, at optimized conditions, the per cent dye removal for ZnO and ZnO-ME was 71 and 80, respectively. This indicated that ZnO-ME was more significant than ZnO for dye adsorption. These findings imply that ZnO nanomaterials can remove sulfur black dye from synthetic aqueous medium and can be used to treat industrial wastewater.

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