

Single objective optimization of surface water coagulation process using inorganic/organic aid formulation by taguchi method

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ABSTRACT

The combined inorganic/organic coagulant has received much attention in turbidity removal for its positive influence on the environment and economy when introduced to the classic coagulation-flocculation clarification process. The present study investigates the effect of different parameters on turbidity removal. These parameters are initial turbidity, coagulant dosage, organic load, and pH. The Taguchi method was utilized in designing the experiment. The Analysis of the variable indicates that the organic load, initial turbidity, coagulant dose, and pH contributions to the turbidity removal was 58.92%, 20.03%, 11.67%, and 6.73% respectively. Results show that the predicted quadratic model can represent the turbidity removal with an R^2 value of 99.24%. Optimum operation conditions were: an organic load of 25% of the total dose, total coagulants dose of 10 ppm, pH of 6 with an estimation of high turbidity value of 100 NTU. Confirmation experiments were conducted and the experimental results have a slight deviation from theoretical values representing model strength. The results confirm the role of an organic aid to provide superior turbidity removal at reasonable economic and operational benefits.

Keywords: Taguchi, Coagulation, Organic Coagulan

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1. Introduction

Water is not found in nature in pure form because it tends to dissolve organics, gases, and impurities. These soluble caused physical change (turbidity, temperature, color, and electrical properties), chemical change (pH, alkalinity, and total organic carbon) or biological change (biological demand for oxygen) depending on its concentration and compositions [1].The turbidity of water is one significant water characteristic in plants that treat water from natural resources such as lakes and rivers. To obtain clarified water, turbidity must be removed [2].The most common techniques used for the turbidity removal of freshwater are by addition of coagulant and flocculation chemicals where common inorganic coagulants such as Alum, poly-aluminum chloride, and Ferric Chloride are used in addition to synthetic organic polymers such as polyacrylamide derivatives and polyethylene amine. These technologies followed by filtration are the key steps in conventional water treatment and are approved for the removal of suspended solids, color, bacteria, viruses, and other micro-organisms. The coagulation and flocculation mechanisms involve electrostatic attraction, sorption, and bridging [3], [4].

zeta-potential measurement and jar tests are methods used to determine the best coagulant and flocculent dosage. In a typical jar test practice, the operator determines the optimum dosage and pH by changing these factors with time and holds the other factors constant. The value of the factor that gives in the best response

with low turbidity is then selected and used in the following tests which continue in the same procedure with other factors [5]. The main challenges of surface water treatment are the seasonal changes in surface water turbidity [6]. With a range of commercial coagulants and different water character in various water sources, the difficulty arises in the proper choice of coagulants for particular surface water. In the conventional water treatment process, the correct coagulants selection with an economic dose to the enhancement of turbidity removal [7]. Most conventional inorganic coagulants used in water treatment contains aluminum chloride and ferric chloride [8], [9-10]. There are various drawbacks of using metal salts such in water treatment including high operational costs, health effects from residual metals, the influence on pH of clarified water, and huge sludge volumes [11-12], [13]. The search for the best alternative coagulants to match the increasing requirement for water quality has become a critical area of study. It is important to decrease the dosage of conventional coagulants to reduce unfavorable effects. The best familiar method to fix the above issues is by applying coagulant aids. Organic Coagulant enhanced the total cost of coagulation and speed up the later flocculation process, reduce the production of sludge, and make flock easy to capture by a filter. Synthetic polymers are the best-accepted coagulant aids. while inorganic and naturally extracted polymers can promote the floc formation by acting as sites of nucleation [14]. The combined organic coagulant aid was widely investigated with polymeric aluminum, ferric chloride, and aluminum chloride and showed that it can improve both the turbidity removal and organic matter removal [11]. The use of organic as a coagulant aid for treating wastewater and oily produced water has been confirmed in recent researches [15], [16-17]. Novel covalently bound organic hybrid coagulants tested against PAC and applied to treat turbid water and coking wastewater achieved about 82.05% turbidity removal than PAC [18]. Recently, statistical methods such as Taguchi, factorial design analysis, response surface methodology, and multiple linear regression (MLR) have been employed to optimize the water coagulation process and improve the yield [19], [20-21]. Taguchi design is an efficient quality control method for engineers that affirm the roles of research and development (R&D). The special orthogonal array design comprising controllable variables and their estimated levels that significantly reduce the overall testing time and cost [22-23], [24]. The previous researches used Taguchi analysis in wastewater treatment, less focused on surface water [25], [26]. The primary objective of the current study to investigate the efficiency of using inorganic (Ferric Chloride) /organic (SoliSep MPT150) aid coagulants formulation on the turbidity removal percentage. These coagulants were chosen because of their frequent use in conventional water treatment plants around the Tigris River in Baghdad. This research also aims to study the signs and optimized the process variables (initial turbidity, organic load, coagulant dose, and pH) on the turbidity removal percentage by using the Taguchi design of experiment method and analysis of variance (ANOVA). The study identified the process condition of achieving the highest turbidity removal of surface water which could be applied practically.

2. Materials and methods

Surface water samples 20 L were collected from the Tigris River area in Baghdad in the experimental period (January /February 2020). The water sample was collected and characterized within 90 minutes as explained in Table 1. Two different coagulants were used in this study. Ferric Chloride anhydrous (CDH) was used as an inorganic coagulant and industrial product SoliSep MPT150 (SUEZ Water Co.) was used as an organic coagulant. Kaolin powder from Sigma- Aldrich Co. was used to simulate different levels of turbidities. The pH of the samples was adjusted by HCl (CDH) or NaOH (Alfa Chemika) as needed. The turbidity of samples was measured using HACH 2100Q turbidimeter. pH was measured using Pocket Pro+ Multy 2 by HACH. The water clarification experiments performed using the jar test method. All jar testing experiments were done using the FP4 Flocculator tester by Velp Scientifica. Ten grams of Kaolin were dehydrated over 105 °C for three hours and dissolved in 50 ml deionized water then additional water was added to reach 1 liter volume. The suspension then stored to be used as a stock turbidity solution.

A weight measurement of 10 gm of the coagulants mix at different ratios was dissolved in 250 ml of deionized water to prepare the stock coagulant solution. The solutions are then agitated for better mixing and homogenization. The term “organic load” was used to describe the strength of organic coagulants in total

coagulants mixture. Four coagulant samples were prepared based on the organic load at 0, 0.25, 0.5, and 0.75 mg organic coagulant/mg of the total dose. For example, to prepare a 0.5 organic load coagulant stock, 5 mg of Ferric Chloride, and 5 gm of SoliSep MPT150 were mixed in a 250 ml demineralized water solution. Turbidity and pH were adjusted to the required measures, coagulant was injected to the required dosage, and the mixture was agitated for 1, 1.5, and 5 minutes at speed of 200, 40, and 20 rpm respectively. The sample then allowed to rest for 30 minutes, about 10 mL was collected for turbidity measurement from a point of 2 centimeters below the surface. All the experiments were conducted at the laboratory temperature (18 -20 C).

Table 1. Tap water characteristics

Parameter	Unit	Value
pH		7.34
Connectivity	μS/cm ²	1178
Total Dissolved Solids	PPM	889
Turbidity	NTU	
Chloride	PPM	220
Total Hardness	PPM as CaCO ₃	468
Ca Hardness	PPM as CaCO ₃	272
Total Alkalinity	PPM as CaCO ₃	172

The performance of jar test runs was described by turbidity removal percentage, calculated from Eq. (1) [26]:

$$\text{Turbidity removal \%} = \left[1 - \frac{T_{ur_f}}{T_{ur_i}} \right] 100 \quad (1)$$

Where Tur_i, Tur_f initial, and final turbidities NTU units respectively.

3 Taguchi method for optimizing the coagulation Process

The Taguchi method with L16 orthogonal array was employed for the robust design of the experiments by using software Minitab 19. The parameters presumed are initial turbidity, pH, organic load, and coagulant dose. Table 2 explains the studied experimental parameters and their levels.

The objective function employed to analyze the response of the experimental results of the present study is the signal to noise ratio (S/N) ratio larger-is-better, which relates to the maximum turbidity removal percentage.

The signal to noise ratio (S/N) larger-is-better can be realized by applying the Eqn. (2) [27].

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

Where n is repeat number and y_i is measured variable value.

Table 2. Experimental parameters and their levels

Parameter	Code	Unit	Levels			
			1	2	3	4
Initial Turbidity	X ₁	NTU	25	50	75	100
Organic load %	X ₂	mg/m	0	25	50	75
Coagulant Dose	X ₃	g ppm	10	20	30	40
pH	X ₄		5	6	7	8

3. Results and discussions

3.1. Analysis of response and S/N ratio

The observed values of the experimental response (turbidity removal percentage) and the S/N ratios higher is better explained in Table 3. The values of the S/N ratio for all levels of the studied parameters and their effect on turbidity removal percentage in rank wise are explained in Table 4. The main effect plots of studied

variables (initial turbidity, organic load, coagulant dose, and pH) on the S/N ratio and mean of response are illustrated in Figures 1 and 2. It's clear from Fig.1 and Fig.2 that the S/N ratio and mean of response increased with increasing initial turbidity from 25 to 100 NTU. The S/N ratio reaching the best value of 39.60 at level 4 with initial turbidity of 100 NTU as explained in Table 4. Increasing the initial turbidity will increase the turbidity removal percentage this may be attributed to sweeping flocculation mechanism. The large density of suspended solids and collides corresponding to high turbidity enhanced the size and weight of flocs, making turbidity removal more efficient. On the other hand, the studied coagulant mixture has good potential to treat a wide range of turbid water [28]. The effect of organic load on S/N ratio and mean of turbidity removal percentage illustrated that the best S/N ratio attributed to organic load was 39.78 at level two as described in Table 4. It can be observed from Figures 1 and 2 that the S/N ratio and turbidity removal percentage increased with increasing organic load to level %25 and then decrease with a more organic load. The addition of organic coagulants may reduce the dose of inorganic coagulant by %25 and results in a turbidity removal of 97.4%. The organic coagulant found to be effective and economics to enhance the coagulation efficiency and to the formation of denser flocs. The use of organic coagulants will also reduce the risk of iron carryover to the downstream equipment. S/N ratio was constant at a coagulants dose range from 10 to 20 ppm. Further increasing of coagulant dose caused a decrease in S/N ratio and turbidity removal percentage from 93.7 % at 20 ppm dose to 86.8 % at 40 ppm dose. This drop may be attributed to the charge reversal phenomenon and destabilization of colloidal particles because of excess coagulants dose [29]. pH found to have a slight effect on the S/N ratio and turbidity removal percentage compared with the other parameters. The best turbidity removal achieved at pH 6 is about 93.8%. It is clear from Figures 1 and 2 that the studied coagulants act efficiently in a wide range of initial sample pH. The turbidity removal percentage range between 89% to 93.8% at a pH range from 5 to 8.

The contour and surface plots represented in Fig.3-8 describe the combined effects and influences of different variables on turbidity removal. of and the importance of each variable to the coagulation process. Fig.3 and Fig.6 represent the combined effect of organic load and initial turbidity on the turbidity removal in both contour and surface plots respectively. The turbidity removal achieved a very high value of > 98% at initial turbidity ranges from 60 to 100 NTU and an organic load of 15% to 50%. It can be also observed that high organic dosage with low initial turbidity values reduces turbidity removal significantly. The organic coagulants SoliSep MPT150 used in the present study appear efficient with a wide range of turbid water even with a low dose.

Table 3. Taguchi design with an observed and predictive response and S/N Ratio higher is better

Run	X1	X2	X3	X4	turbidity removal %	S/N Ratio
1	25	0	10	5	93.16547	39.3851
2	25	25	20	6	97.08475	39.74302
3	25	50	30	7	87.54967	38.84509
4	25	75	40	8	67.72334	36.61477
5	50	0	20	7	95.85616	39.6324
6	50	25	10	8	96.18307	39.66197
7	50	50	40	5	81.8662	38.26209
8	50	75	30	6	83.07018	38.3889
9	75	0	30	8	95.26174	39.57837
10	75	25	40	7	99.03388	39.91568
11	75	50	10	6	96.46907	39.68776
12	75	75	20	5	85.33686	38.62273
13	100	0	40	6	98.94286	39.90769
14	100	25	30	5	97.53	39.78276
15	100	50	20	8	96.61739	39.70111
16	100	75	10	7	89.0991	38.99747

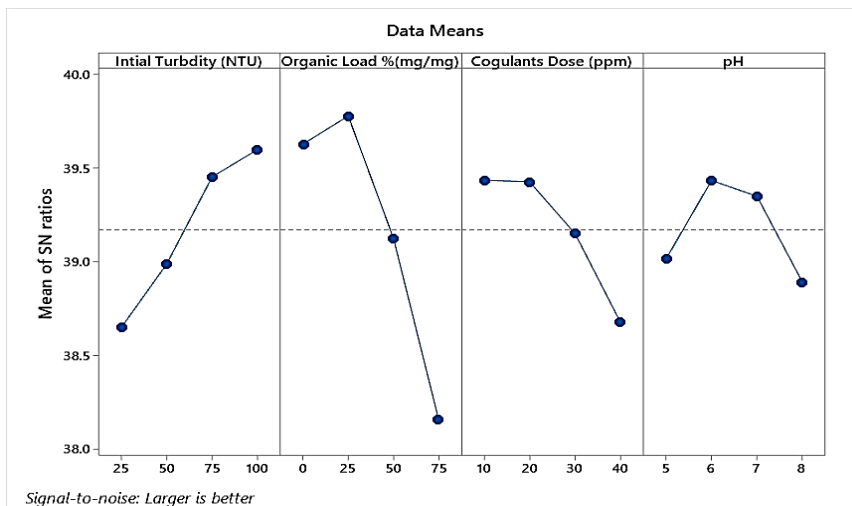


Figure1. Main effect plot of studied parameters on S/N Ratio

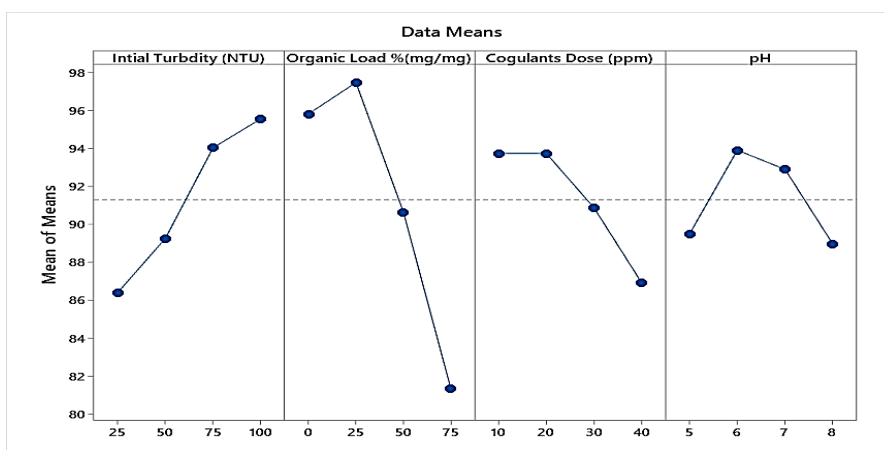


Figure 2. Main effect plot of studied parameters on the mean of response turbidity removal percentage

Fig.4 and Fig.7 show the combined effect of organic load and pH on turbidity removal. At pH value between 6.5 to 7.5 and the organic load of 20% to 25%, the turbidity removal reaches as high as >98 %. It appears from the second zone in the graphs that the target turbidity removal percentage > 98% can be achieved by reducing the organic load to 0% (increasing ferric chloride dose) at a pH range of 5.3 to 6 which required chemical addition for stabilization and corrosion control [30]. The addition of organic coagulants enhances the pH range of the coagulation process of surface water. The combined effect of organic load and total dose explained in Fig.5 and Fig.8. The addition of organic load to the inorganic coagulants reduces the total dose required to achieve >90% of turbidity removal. on the other hand, the zero organic loads require high inorganic coagulants dose 35 to 40 ppm to reach maximum turbidity removal percentage of >98 %. The used inorganic and organic aid coagulant mixture appears efficient in the dose between 10 to 25 ppm with broad initial turbidity range 25 to 100 NTU and can provide an efficient turbidity removal.

Table 4 .The Response for signal to noise ratios larger is better

Level	Initial Turbidity	Organic Load	Dosage	pH
1	38.65	39.63	39.43	39.01
2	38.99	39.78	39.42	39.43
3	39.45	39.12	39.15	39.35
4	39.60	38.16	38.68	38.89
Delta	0.95	1.62	0.76	0.54
Rank	2	1	3	4

3.2. Statistical Analysis and Building Model

In this section, the relationship between the four studied variables and their influence on single response turbidity removal percentage (Y) was studied. Minitab 19 statistical software used to analyze the data and develop a predictive model. Analysis of variance (ANOVA) was utilized to generate the relationships between turbidity removal and the investigated parameters. The result of ANOVA analysis is represented in Table 5. Multiple regression analysis was performed to represent the relationship between the expected variable and response (Y). the quadratic and interaction model is represented in Eqn. (3). The predicted model has an R² value of 99.24% this shows that the fit model demonstrated 99.24% of the percentage of turbidity removal, reflecting the model's effectiveness.

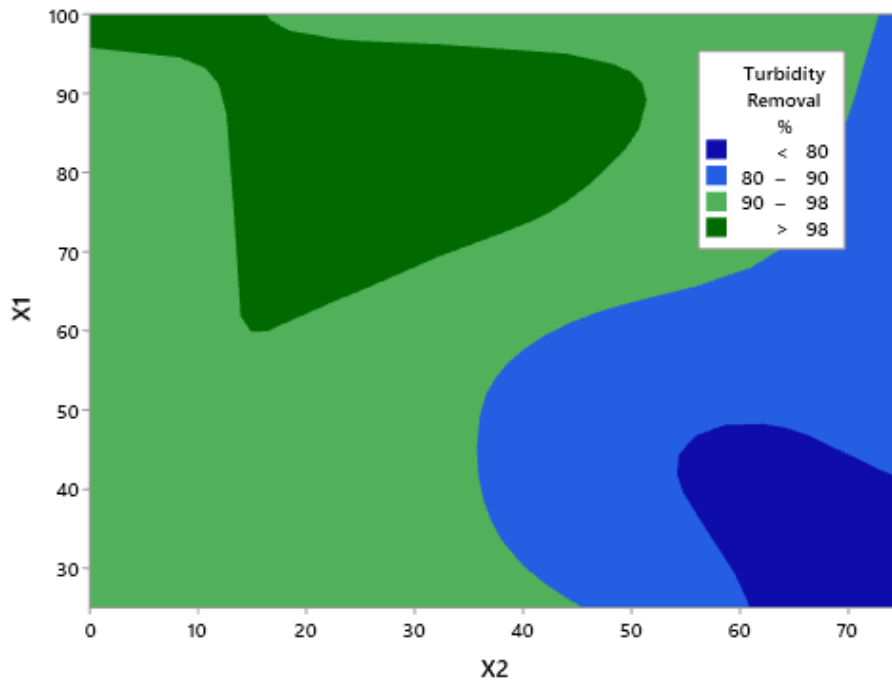


Figure 2. Contour plots for turbidity removal percentage, hold values: X₃=25 ppm, X₄=6.5

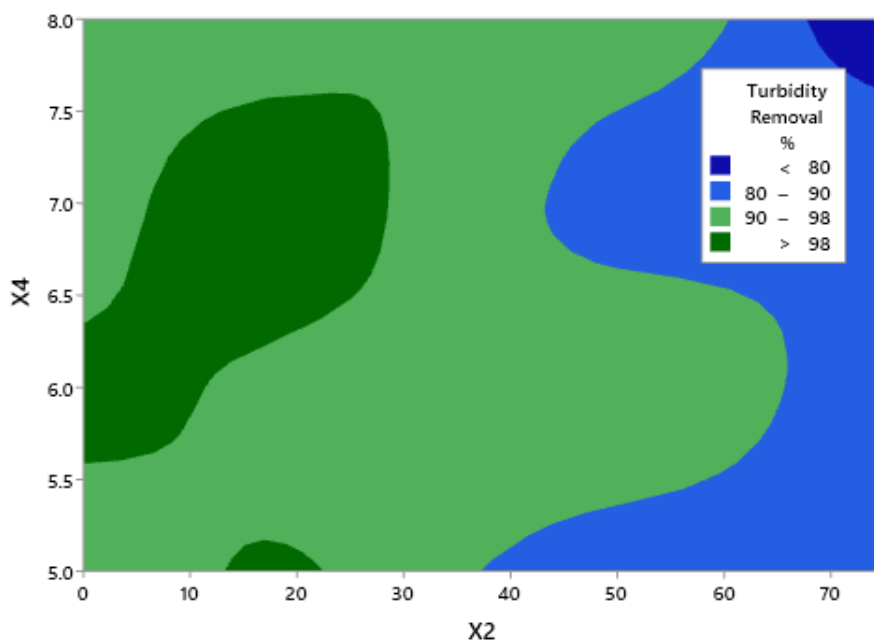


Figure 3. Contour plots for turbidity removal percentage, hold values: X₁=62.5 NTU, X₃=25 ppm

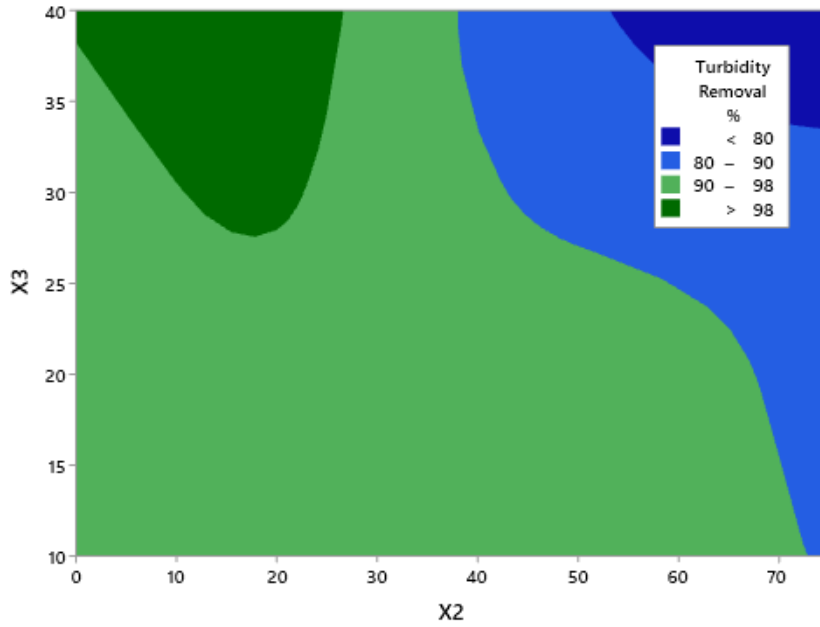


Figure 5. Contour plots for turbidity removal percentage, hold values: $X_1= 62.5$ NTU, $X_4= 6.5$

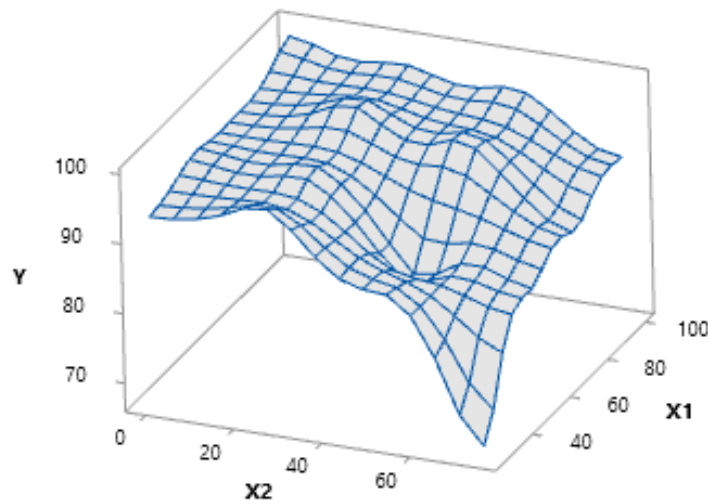


Figure 6. Surface plots for turbidity removal percentage, hold values: $X_3= 25$ ppm, $X_4= 6.5$

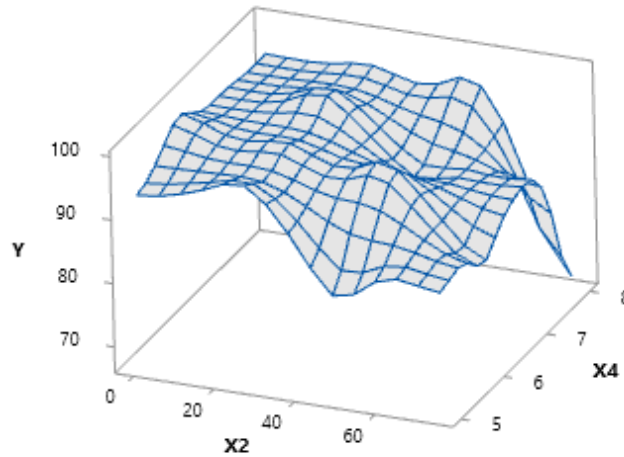


Figure 7. Surface plots for turbidity removal percentage, hold values: $X_1= 62.5$ NTU, $X_3= 25$ ppm

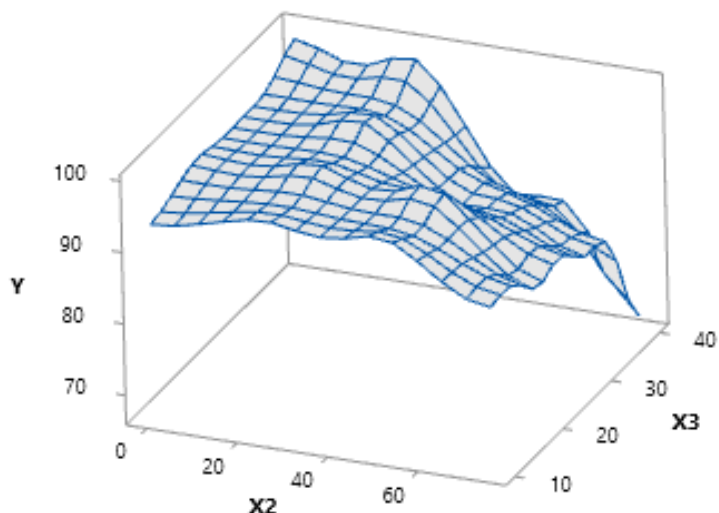


Figure 8. Surface plots for turbidity removal percentage, hold values: $X_1= 62.5\text{NTU}$, $X_3= 6.5$

The model adjusted R^2 is about (98.10%), this indicated that the total change of 98.10% of the model variables covered by this model and cannot describe only (1.9%) of the total variability. The predicted R^2 (94.51%) indicated the model well to predict the response (turbidity removal percentage) for new observations

$$Y\% = 11.1 - 0.1859 X_1 + 0.1250 X_2 - 0.298 X_3 + 28.12 X_4 - 0.004388 X_2^2 - 0.00989 X_3^2 - 2.089 X_4^2 + 0.001528 X_1X_2 + 0.01031 X_1X_3 \quad (3)$$

3.3. Analysis of variance (ANOVA)

The most important coagulation variables have been determined by calculating the percentage of each variable's contribution to the turbidity removal model. Table 5 and Fig.12 represents the analysis of variance (ANOVA). It shows that the organic load (X2) has a 58.92% contribution to the response turbidity removal. The organic load was noticed to be the most substantial variable for the coagulation process followed by initial turbidity (X1) with a 20.03% contribution and the coagulant dose (X3) contribute with 11.67%. Finally, the pH variable (X4) is less effective with a 6.73% contribution.

Table 5. Analysis of variance for turbidity removal %

Source	Degree of freedom	Sum of squares	Mean square	Percentage contribution (%)
X_1	3	215.57	71.858	20.03
X_2	3	635.15	211.382	58.92
X_3	3	125.64	41.880	11.67
X_4	3	72.40	24.133	6.73
Error	3	28.57	9.523	2.65
Total	15	1076.33		100

3.4. Prediction of optimum turbidity removal and its validation

The theoretical turbidity removal percentage was tested by applying Taguchi optimum variables levels to the predicted model represented in Eqn. (3). The optimum variables level for surface water coagulation by using inorganic/organic aid coagulants was obtained as follows: initial turbidity of 100 NTU, an organic load of 25%, total coagulants dose of 10, and pH of 6. However, these conditions should be experimentally proved. Two experiments were conducted within the range of initial turbidity of 25 NTU and 100 NTU to test the

validity of the predicted model under different initial turbidity range. The experiment test results explained in Table 6. The predicted turbidity removal in both experiments was found to be significant. There is a slight difference between the experimental and the predicted values.

Table 6. Observation vs. theoretical values

Run	X ₁	X ₂	X ₃	X ₄	Observation Value	Theoretical Value
Run 1	100	25	10	6	95.33	96.563
Run 2	25	25	10	6	99.94	99.907

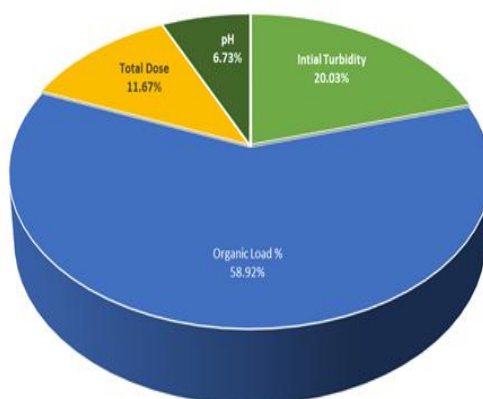


Figure 9. Variables contribution percentage on response turbidity removal percentage

4. Conclusion

The Taguchi method serves as a powerful tool for the significant design of experiments and system representation. The use of the Taguchi method can significantly reduce the number of experiments and time required to evaluate the effect of different variables on the process response. The effect of studied variables (initial turbidity, organic load, coagulant dose, and pH) on the response the turbidity removal and SN/ratio was observed. The analysis of variance (ANOVA) assumes that the most significant variable relates to the turbidity removal to be the organic load at a contribution of 58.92%. Optimum condition established was an organic load of 25% of the total dose, total coagulants dose of 10 ppm, pH of 6 with an estimation of high turbidity value of 100 NTU. The performance of water turbidity removal by using inorganic/organic aid coagulants is feasible for treating surface water. The results confirm the role of an organic aid to provide superior turbidity removal at reasonable economic and operational benefits. This can be achieved with a total coagulant dosage of 10 ppm, 25% organic load, and a high initial turbidity value of 100 NTU. The optimum inorganic/organic aid performance was at pH around 6. A second-order mathematical model developed using regression and the predicted model was well fitted to the obtained experimental data. The validation of the predicted model with further experiments was achieved and represents model strength and integrity. These results give a better understanding of the possibility of applying the inorganic/organic aid coagulant formulation in water treatment. The optimum levels achieved from this study could be scaled up and applied to practical surface water treatment

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