Optimization of Coagulation Parameters for Turbidity Removal Using Box-Behnken model

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ABSTRACT

This study aims to determine the efficacy of using calcium hydroxide Ca(OH)2 in coagulation process to treat urban liquid effluent from wastewater purification plant. The response surface methodology was used. The effects and interactions between three key process parameters was enhanced applying Box-Behnken design. Factors studied were: coagulant dosage (g/L), coagulation speed (rpm), and coagulation time (min). The treatment efficiency was determined by the turbidity removal rate and the final pH of the treated wastewater. The statistical soundness of the generated model was determined using analysis of variance. The optimal model for determining the relationship between the variables is a second-order quadratic model (R2>98 %). The best turbidity reduction (91.3 %) was obtained using a 0.50 g/L Ca(OH)2 coagulant and 130.1 rpm for 5 minutes. Under these conditions, the ultimate pH of the effluent reached 8.

Keywords: Process, Coagulation, Optimization, Response Surface Methodology, Box-Behnken.

1. INTRODUCTION

The optimization of water treatment processes is of great importance because it improves efficiency and reduces the cost of treatment [1, 2]. One of the more modern methodologies for process optimization is Response Surface Methodology (RSM). It involves the design of experiments, analysis, and partial regressionfit modeling of experimental parameters [3,4]. The approach may combine multiple factors at once and reveal reciprocal interactions in a process’ yield; it also minimizes the number of experimental tests necessary to achieve statistically acceptable results.

The current study adjusted the process variables previously known to affect the water coagulation process [5, 6]. RSM was used to assess the impacts of three variables (Lime dosage, coagulation speed, and coagulation time) and their effects on turbidity removal and the final pH of wastewater treated in order to eliminate the colloidal suspended matter responsible for the turbidity of the water and to achieve a final pH in the range of 6–8 to avoid a post-adjustment of pH due to the use of calcium hydroxide.

1.1. Related works and contributions

The RSM methodology is one of the more recent ways of multiple response optimizations that is being applied in industries. It is commonly used because to its low cost and low test requirement. RMS has been used to optimize different processes for wastewater treatment.

Khattaf et al. [7] used the coagulation-flocculation process to remove organic matter from surface water used for drinking water production. The variables influencing the responses are the initial pH of the water and the concentrations of coagulant and flocculant used. RSM results reveal that the optimal conditions were an initial pH of 6.9, using 0.133 g/L...
and 0.06 g/L of coagulant and flocculant, respectively. The final pH obtained was 6.78 and the abatement reached 56% and 59% in terms of COD and UV-254 respectively.

In another work by Louhichi et al. [8], similar to Khettaf et al. [7], the wastewater from the vegetable oil refinery was also treated by coagulation–flocculation. This study also used RSM but Box–Behnken (BBD) as an optimization design. The same variable factors were adopted. The optimum was acquired at an initial pH of 9.23 using 2400 mg/L and 60.05 mg/L of coagulant and flocculant, respectively. The treatment eliminated 99% of the COD and 100% of the turbidity.

Gökçek, et al. [9] optimized the coagulation process for COD, SS, and turbidity removal from slaughterhouse wastewater. The Box Behnken design was used. The various experimental factors were the alum coagulant dosage, the coagulation speed and the settling time. Optimization gave maximum removal (75, 90, and 91%), respectively using 1g/L of alum, stirring at a speed of 150 rpm and allowing the sample to settle for 10 minutes.

RSM has also been used in other studies to optimize wastewater treatment process. Table 1 presents an overview of the work carried out above.

| Table 1. Overview of water and wastewater treatment studies carried out with RSM |
|--------------------------------------|----------|-------|-----------------|--------|-----------------|-----------------|
| Type of wastewater | Process | Design | Runs | Variables | responses | Reference |
| Surface water | Coagulation | CCD | 29 | X1=coagulant concentration X2= flocculant concentration X3= initial pH | Y1= COD removal Y2= absorbance Y3= final pH | [7] |
| Vegetable oil refinery wastewater | Flocculation | Box–Behnken | 29 | X1=coagulant concentration X2= flocculant concentration X3= initial pH | Y1= turbidity removal Y2= COD removal | [8] |
| Slaughterhouse wastewater | Flocculation | Box–Behnken | 28 | X1=Coagulant concentration X2= coagulation speed X3= Settling time | Y1= COD removal Y2=SS removal Y3= Turbidity removal | [9] |
| Petroleum wastewater | | CCD | 13 | X1= initial pH X2= coagulant concentration | Y1= final pH Y2= COD removal Y3= turbidity removal Y4= TDS removal Y5= color removal | [10] |
| Palm oil mill wastewater | Electro-Coagulation | Box–Behnken | 16 | X1=time X2= Voltage X3= NaCl concentration | Y1= COD removal Y2=TSS removal Y3= TDS removal | [11] |
| Pharmaceutical wastewater | | CCD | 30 | X1= initial pH X2= Cefazolin concentration X3= Currentdensity X4= Electrolysis time | Y1= Cefazolin removal | [12] |

2. METHODOLOGY

2.1. Origin And Characteristic Of The Effluent

The studied effluent was sampled from the wastewater purification plant, located in the town of Sidi Bel Abbes (western Algeria). The effluent was collected from the clarification tank before any chemical treatment. The wastewater is characterized by a pH of 7.5, a turbidity of 489 NTU, and a dissolved salt level of 1253 g/L.

2.2. Coagulation-Flocculation Tests And Experimental Design

Tests of coagulation flocculation were realized in a flocculator device (Jar Test AO/AUTC). As a coagulant, calcium hydroxide Ca(OH)₂ was used. The volume of wastewater used is 400 mL per test.
To optimize the variables that affect the efficiency of the coagulation process, RSM was employed. The statistical software Design Expert 13.0 was utilized for the experiment design and data analysis. The Box-Behnken (BBD) design was used to optimize the turbidity removal and the final pH of the wastewater treated by coagulation method.

Three factors were varied: coagulant dosage (A), coagulation speed (B), and coagulation time (C). The coded levels and ranges of each factor are shown in Table 2. The responses of the design were: the turbidity removal (Y1) and the final pH (Y2). 15 experimental runs were performed.

**Table 2.** Experimental and coded values

| Factors              | Symbol | Coded levels |
|----------------------|--------|--------------|
| coagulant dosage (g/L) | A      | -1 0 +1     |
| Coagulation speed (rpm) | B      | -1 0 +1     |
| Coagulation time (min) | C      | -1 0 +1     |

The model’s fitness was assessed using a significance test and an analysis of variance. A quadratic equation model, expressed in Equation (1), was used to optimize the coagulation process.

\[
Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k} \sum_{j=1}^{k} \beta_{ij} X_i X_j
\]

Where Y is the response, X_i represents the input factor, \( \beta_0 \) is the intercept, the linear influence of the input factor X_i is denoted by \( \beta_i \), the interaction effect between X_i and X_j is denoted by \( \beta_{ij} \), the quadratic effect of X_i is \( \beta_{ii} \). The turbidity removal efficiency (%) is calculated according to Equation (2):

\[
\text{Turbidity removal, (\%)} = \left[1 - \left(\frac{T_i}{T_0}\right)\right] \times 100
\]

T_0 is the initial turbidity.

\[
(2)
\]

T_i is the final turbidity.

### 3. RESULTS AND DISCUSSION

The relationships between turbidity removal efficiency, the final pH of treated wastewater, and the factors (coagulant dosage, coagulant speed, and coagulant time) were evaluated. Table 3 shows the operating parameters, results of the experiments that were conducted and predicted data.

#### 3.1. Analysis Of Variances (ANOVA)

The results in Table 3 showed that the experimental data fit the model perfectly. Multiple regression analysis was used to analyze the experimental data. The second order polynomial Equations (3) and (4) were used to relate the responses values and the coded factors. The synergistic effect is indicated by the positive sign in front of the parameters, and the antagonistic effect is indicated by the negative sign.

\[
Y_1 = 98.4467 -0.45375A -1.72125B +1.6275C^2 -1.3475AB +2.22AC -0.875BC -8.06208C^2 +6.16042B^2 -8.60208C^2
\]

(3)

\[
Y_2 = 9.09333 +0.93875A +0.6975B +2.22AC -0.33AC +0.38735C^2 +0.7275AB +0.33AC +0.5375BC -0.374167A^2 +1.37333B^2 +0.845833C^2
\]

(4)

For each response, the BBD design gave statistical parameter data. The variance analysis (ANOVA) was used to generate these statistical data, which were then used to determine the optimization’s significance.

**Table 3.** Experimental and predicted data (BBD design)

| Test | A: Coagulant dosage (g/L) | B: Coagulation speed (rpm) | C: Coagulation time (min) | Y1: Turbidity removal (%) | Y2: Final pH |
|------|---------------------------|-----------------------------|---------------------------|---------------------------|--------------|
|      | Actual value | Predicted value | Actual value | Predicted value |         |
| 1    | 0.5          | 200              | 5             | 96.24         | 95.99        | 96.24        | 95.99         |
| 2    | 0.5          | 150              | 7             | 81.61         | 81.42        | 81.61        | 81.42         |
| 3    | 2            | 200              | 3             | 95.26         | 94.20        | 95.26        | 94.20         |
| 4    | 0.5          | 100              | 5             | 97.86         | 95.99        | 97.86        | 95.99         |
| 5    | 3.5          | 150              | 3             | 77.32         | 77.51        | 77.32        | 77.51         |
| 6    | 3.5          | 100              | 5             | 98.27         | 98.52        | 98.27        | 98.52         |
| 7    | 2            | 150              | 5             | 97.99         | 98.45        | 97.99        | 98.45         |
The parameters such as F-value, P-value, R² and R² adjusted were determined to assess the model's effectiveness. The Model F-value of 41.59 and 70.05 shown in Tables 4 and 5 suggest that the model is significant. P-values of the model terms less than 5.10⁻² are considered significant and confirm factor-response interactions. Based on this, the factors B and C and the interaction terms A², B², and C² had significant individual and quadratic effects on the removal of turbidity. Also, the parameters A and C had significant interactive effects between them. Analysis revealed that all three factors had significant individual (A, B, C), quadratic (A², B², C²), and interactive (AB, AC, BC) effects on the final pH value.

Table 4. Fit statistics for the response Y1 (turbidity removal)

| Source          | Sum of Squares | df | Mean Square | F-value | p-value |
|-----------------|----------------|----|-------------|---------|---------|
| Model           | 761.13         | 9  | 84.57       | 41.59   | 0.0004  |
| A-Coagulantdosage| 1.65           | 1  | 1.65        | 0.81    | 0.4094  |
| B-Coagulationspeed| 23.70          | 1  | 23.70       | 11.66   | 0.0190  |
| C-Coagulationtime| 21.19          | 1  | 21.19       | 10.42   | 0.0233  |
| AB              | 7.26           | 1  | 7.26        | 3.57    | 0.1174  |
| AC              | 19.71          | 1  | 19.71       | 9.70    | 0.0264  |
| BC              | 3.06           | 1  | 3.06        | 1.51    | 0.2743  |
| A²              | 279.44         | 1  | 279.44      | 137.43  | <0.0001 |
| B²              | 140.13         | 1  | 140.13      | 68.92   | 0.0004  |
| C²              | 239.99         | 1  | 239.99      | 118.03  | 0.0001  |
| Lack of Fit     | 9.85           | 3  | 3.28        | 20.59   | 0.0467  |
| Pure Error      | 0.3189         | 2  | 0.1594      | /       | /       |

Table 5. Fit statistics for the response Y2 (final pH)

| Source          | Sum of Squares | df | Mean Square | F-value | p-value |
|-----------------|----------------|----|-------------|---------|---------|
| Model           | 25.89          | 9  | 2.88        | 70.05   | 0.0001  |
| A-Coagulantdosage| 7.05           | 1  | 7.05        | 171.71  | <0.0001 |
| B-Coagulationspeed| 3.89           | 1  | 3.89        | 94.79   | 0.0002  |
| C-Coagulationtime| 1.18           | 1  | 1.18        | 28.69   | 0.0030  |
| AB              | 2.12           | 1  | 2.12        | 51.56   | 0.0008  |
| AC              | 0.4356         | 1  | 0.4356      | 10.61   | 0.0225  |
| BC              | 1.16           | 1  | 1.16        | 28.15   | 0.0032  |
| A²              | 0.5169         | 1  | 0.5169      | 12.59   | 0.0164  |
Figure 1 shows the relationship between experimental (actual) and predicted model values. We can observe that the predicted values of the model follow the experimental values well because they are near the diagonal axis. The values of the coefficient of determination $R^2$ of 0.98 and 0.99 (table 6) obtained for the removal of turbidity ($Y_1$) and for the final pH of wastewater ($Y_2$), respectively, revealed a good fit of the model.

Referring to table 6, the predicted $R^2$ values for turbidity removal and the final pH are in the order of 0.7948 and 0.8857, which is in agreement with the adjusted $R^2$ values of 0.9631 and 0.9780, respectively. The difference is less than 0.2, which indicates that the model adopted is significant. The correct precision value must be greater than 4. The correct precision values for the rate of turbidity removal and the final pH are 20.08 and 23.47, respectively. Therefore, both models are accepted.

### Table 6. Statistical parameter values

| Parameter            | Turbidity removal(%) | Final pH |
|----------------------|----------------------|----------|
| Standard deviation   | 1.43                 | 0.2026   |
| Mean                 | 92.79                | 10.08    |
| $R^2$                | 0.9868               | 0.9921   |
| Adjusted-$R^2$       | 0.9631               | 0.9780   |
| Predicted-$R^2$      | 0.7948               | 0.8857   |
| Adequate precision   | 20.0855              | 23.4745  |

### 3.2. Optimization Of Parameters

To obtain maximum turbidity removal under optimal conditions, an optimization analysis was performed. The objectives of this analysis and its limitations are presented in Table 7. Prior to the
analysis, aims were specified to find the optimized process conditions. The final pH of the wastewater was chosen to be in range 6 and 8. It is known that calcium hydroxide in its coagulant form induces very alkaline final pH. So this range was chosen on the basis of obtaining a final pH of the treated water close to neutral.

**Table 7. Objectives and ranges of optimization**

| Parameter               | Aim          | Minimum level | Maximum level |
|-------------------------|--------------|---------------|---------------|
| A - Coagulant dosage    | Minimum      | 0.5           | 3.5           |
| B - Coagulation speed   | In range     | 100           | 200           |
| C - Coagulation time    | In range     | 3             | 7             |
| Turbidity removal       | Maximize     | 50            | 100           |
| Final pH                | In range     | 6             | 8             |

According to the objectives of the study, the selected optimal conditions was, coagulant dosage of 0.5 g/L, coagulation speed of 130.1 rpm and coagulation time of 5 min. At these optimal conditions, the predicted removal of turbidity was 91.3% and the final pH was of 8. The desirability of these optimal conditions is 0.908. The 3D graph of this desirability is shown in Figure 2.

![Desirability surface for optimal conditions](image)

Figure 2 Desirability surface for optimal conditions Figures 3 and 4 show 3D surface diagrams for the turbidity removal and the final pH, respectively, using 0.5 g of coagulant.

The Figure 3 shows a minor interaction between the agitation speed and the reduction of turbidity; changing its value along its axis has no meaningful effect on the elimination of turbidity in the wastewater. While a considerable interaction between coagulation time and turbidity reduction can be noted in the same figure, a maximum range of elimination is reached between 4 and 6 min.
The Figure 4 shows that the interactions between time or speed of stirring and the final pH are weak; a change in the value of time or speed of stirring does not significantly affect the variation of the final pH.

4. CONCLUSION

This study aimed at optimizing the operating conditions of the coagulation process applied to the pre-treatment of urban liquid effluent in the wastewater purification plant in the town of Sidi Bel Abbes (Algeria). The Box-Behnken Design was used to assess the effects of coagulant dosage (calcium hydroxide), coagulation speed, and coagulation time on the reduction of turbidity and the final pH of the wastewater.

The results show that the coagulation speed and time individually influence the reduction of turbidity, while the final pH value is influenced by all three factors. A second-order mathematical model was found to be well-adjusted to the experimental data.

The optimal conditions obtained were a Ca(OH)₂ dosage of 0.5 g/L and a coagulation speed of 130.1 min for a time of 5 minutes. The optimum condition makes it possible to obtain maximum performance from the coagulation process while minimizing the total cost of the treatment.

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**NOMENCLATURE**

| Acronyms | Meaning                  |
|----------|-------------------------|
| ANOVA    | Analysis Of Variance    |
| UV       | Ultra Violet            |
| BBD      | Box-Behnken Design      |
| SS       | Suspended Solids        |
| RM S     | Response Surface        |
| COD      | Chemical Oxygen Demand  |