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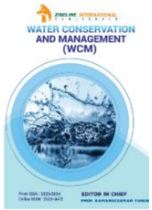

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*RESEARCH ARTICLE*

# OPTIMIZATION OF THE COLOR CORRESPONDING TO THE DIFFERENT ABSORBENCIES OF TANNERY WASTEWATER BY RESPONSE SURFACE DESIGNS

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**ABSTRACT**

The control of pollutant discharges, during the treatment of wastewater from tanneries, is the main task to restore the ecological environment and minimize the time and cost. In this work, we performed experiments concerning the treatment of industrial discharges from a tannery located in the city of Mohammedia-Morocco. An optimization study will be presented to model and optimize the corresponding absorbances at the following wavelengths: 436 nm, 525nm and 620 nm, using the most influential factors such as coagulant, pH and flocculant. In order to minimize the number of experiments performed, the Box-Behnken response surface design was applied. Quadratic models characterizing the three absorbances studied during the treatment will be obtained. The statistical, graphical and algebraic characteristics will be presented. A perfect agreement between the theoretical and experimental results has been established, with a very good performance in the depollution of treated wastewater.

**KEYWORDS**

Industrial Tannery Discharges, Wastewater, Absorbance, Response Surface, Optimization.

## 1. INTRODUCTION

Pre-treatment taking into account the treatment of color variations corresponding to the three different absorbances 436 nm, 525nm and 620 nm, is essential for the elimination of pollution from tannery wastewater. According to a previous study based on the treatment of tannery wastewater by coagulation-flocculation, the removal efficiency and color generated depend on the coagulant and flocculant used (Bouazza et al., 2019; Assou et al., 2017a). In this study, the nature of the coagulant, the flocculant and the pH value are the factors that were taken into consideration, in order to obtain an optimization of the color of tannery wastewater, exploiting a quadratic model. These factors have significant effects on the treatment process and their selection was made from a screening by experimental designs (Assou et al., 2017b). Optimization by response surface design was adopted in order to obtain good information from the experiments performed, in the shortest time and at the lowest cost.

The high load of oxidizable materials is mainly due to biogenic materials in the hides and the organic chemicals used in the hide processing. The treatment of color is related to the elimination of pollution, and for this reason its optimization is very important for the depollution of treated water from tanneries. The principle of the optimization plan used, consists in exploring a surface of the color, in order to locate a possible extremum in the considered study domain (Karam, 2004). We have chosen for our approach a Box-Behnken response surface design, to reduce the number of experiments to be performed. Theoretical quadratic models of the studied absorbances were derived and exploited, studying response surfaces, statistical characteristics and graphical methods. The performance of the tannery wastewater treatment was evaluated by quantifying the color (pollution removal efficiency between 78% and 80%).

## 2. MATERIALS AND METHODS

### 2.1 Experimental Procedure

The aesthetic aspect of the reclaimed water, i.e., its color, smell or taste, also plays a decisive role in the acceptance of water respecting the Moroccan standards of rejection. The total effluent discharged into the main sewer where all the plant's discharges end up, showed that the latter are complex, highly charged with organic and mineral matter, have variable characteristics over time and are not easily biodegradable. In fact, color have been collected (Ghariba, 2020). Laboratory-scale performance evaluation of effluent treatment by coagulation-flocculation was conducted using the Jar-Test technique. To improve the efficiency of pollution removal, coagulants such as: ferric chloride $FeCl_3$ and aluminum sulfate $Al_2(SO_4)_3$, are the reagents used. Concerning the flocculants, the agents Chimec 5161 and Chimec 5264 were selected for this study. The process goes through three phases: first an initial high-speed stirring (about 160 rpm) during the addition of the coagulants, flocculants or both for 10 min, followed by a slow stirring of the mixture at 30 rpm for 20 min. The process is completed by a decantation during one hour. After the predetermined settling, the color removal of tannery wastewater was studied by measuring the three absorbances at 436 nm, 525 nm and 620 nm. The responses observed in this study are the absorbances that correspond to the wavelengths 436 nm, 525 nm and 620 nm.

### 2.2 Design of Experiments Method

An additive model was obtained from a screening study, which allowed us

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to identify the elements having a significant influence on the objective functions (the color), among a list of factors (Assou et al., 2017; Taylor et al., 2010; Assou et al., 2017; Assou et al., 2014). The translation of response variations in an experimental domain was represented from a response surface study by deriving a polynomial model. Screening designs and response surface designs have the same steps to follow during the conduct of the tests, except that the model established with the response surface performs better than the one obtained by the screening technique. The response surface of the color materializes the regression surface from a graph in a three-dimensional space. This response surface design allows for better understanding and optimization of the objective function. We adopted Box-Behnken response surface designs that incorporate information from a properly designed factorial experiment. They fit a more detailed full quadratic model.

### 2.3 Factors and Field of Study

The variations of the three factors determined from the screening method as influential (coagulant, flocculant and pH), define the experimental domain. A Box-Behnken response surface design was adopted to construct the experiment matrix (Assou et al., 2017). These designs are economical to perform the experiments to optimize the explicit response function. In our case, we performed fifteen tests for three influential factors. The experimental response is the color. The factors used are summarized in Table 1:

**Table 1:** Factors Used to Materialize The Response Surface

| Factors | Name of the factors | Number of levels | Level 1 | Level 2 |
|---|---|---|---|---|
| A | Coagulant | 2 | $Al_2(SO_4)_3$ | $FeCl_3$ |
| B | pH | 2 | Chimec 5161 | Chimec 5264 |
| C | Floculant | 2 | 5 | 6 |

We used the factors in coded values during the construction of the experiment matrix

### 2.4 Quadratic Model and Its Coefficients

Second degree polynomials translate the desired models from the experiments performed. The effects of each factor and their interactions are calculated to obtain the following models:

$$Y_1 = 0{,}4473 - 0{,}0426\ A_1 - 0{,}0564\ B_1 - 0{,}0060\ C_1 + 0{,}0032\ A_1{*}A_1 + 0{,}1012\ B_1{*}B_1 + 0{,}1210\ C_1{*}C_1 + 0{,}0052\ A_1{*}B_1 + 0{,}1230\ A_1{*}C_1 - 0{,}0660\ B_1{*}C_1$$

Where $Y_1$ represents the color that corresponds to the absorbance of wavelength 436 nm.

$$Y_2 = 0{,}2620 - 0{,}0347\ A_2 - 0{,}0258\ B_2 - 0{,}0190\ C_2 + 0{,}0121\ A_2{*}A_2 + 0{,}0731\ B_2{*}B_2 + 0{,}0656\ C_2{*}C_2 - 0{,}0107\ A_2{*}B_2 + 0{,}0677\ A_2{*}C_2 - 0{,}0437\ B_2{*}C_2$$

Where $Y_2$ represents the color that corresponds to the absorbance of wavelength 525 nm.

$$Y_3 = 0{,}2823 - 0{,}0446\ A_3 - 0{,}0116\ B_3 - 0{,}0325\ C_3 + 0{,}0247\ A_3{*}A_3 + 0{,}0727\ B_3{*}B_3 + 0{,}0530\ C_3{*}C_3 - 0{,}0163\ A_3{*}B_3 + 0{,}0640\ A_3{*}C_3 - 0{,}0420\ B_3{*}C_3$$

Where $Y_3$ represents the color that corresponds to the absorbance of wavelength 620 nm.

### 2.5 Pareto Diagram

The influence of each of the three factors analyzed and their interactions was demonstrated on the Pareto charts (Figure 1a, Figure 1b, Figure 1c) for the absorbance of wavelength 436 nm, the absorbance of wavelength 525 nm and the absorbance of wavelength 620 nm.

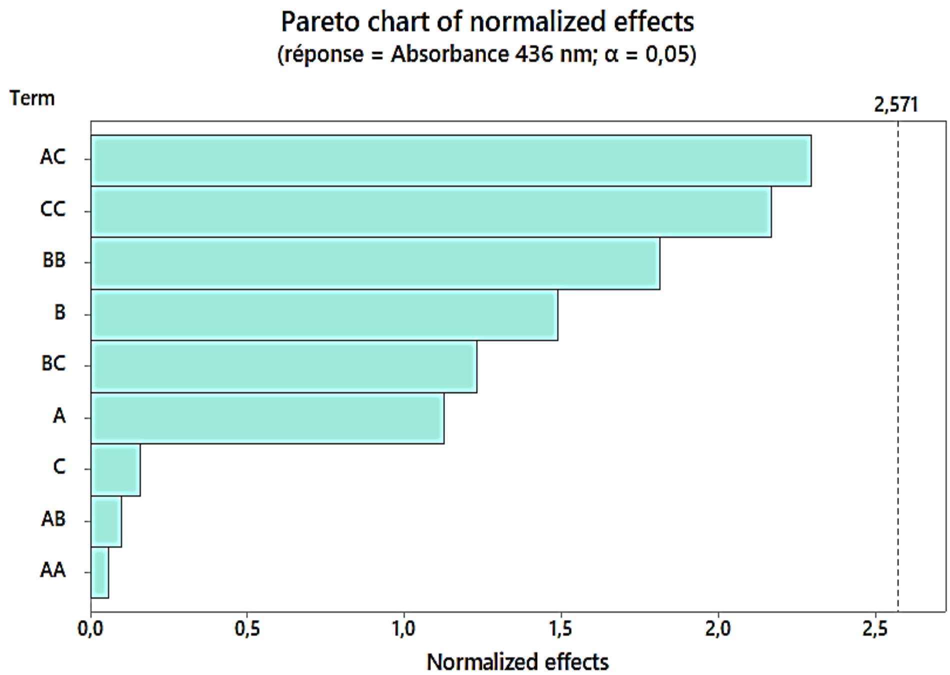


**Figure 1-a:** Pareto Diagram of the absorbance of wavelength 436 nm

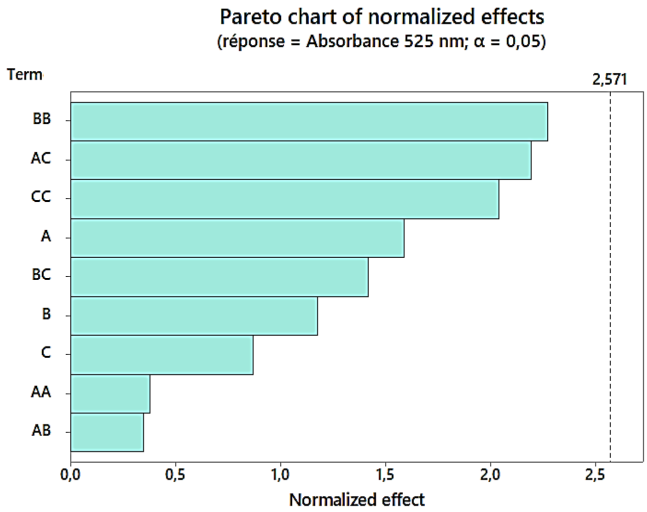


**Figure 1-b:** Pareto Diagram of the absorbance of wavelength 525 nm

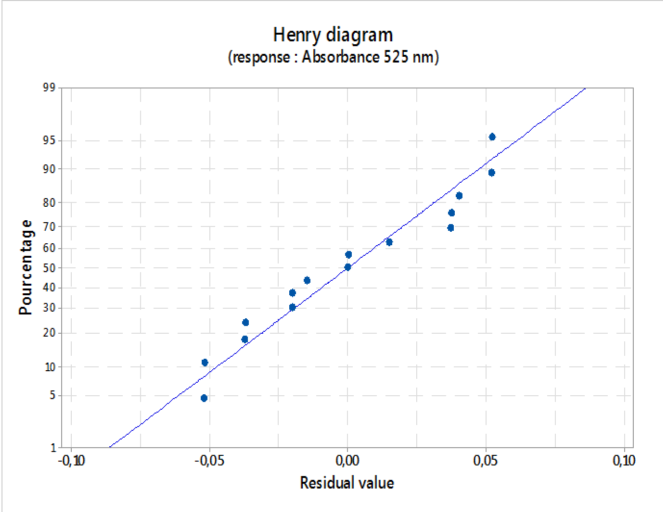


**Figure 2-b:** Henry Diagram of the absorbance of wavelength 525 nm

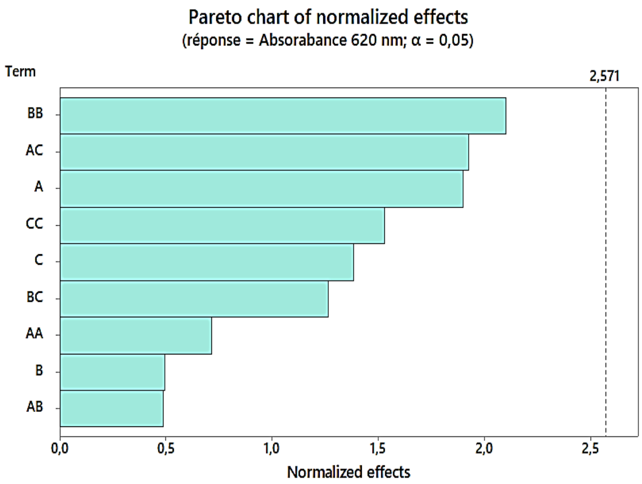


**Figure 1-c:** Pareto Diagram of the absorbance of wavelength 620 nm

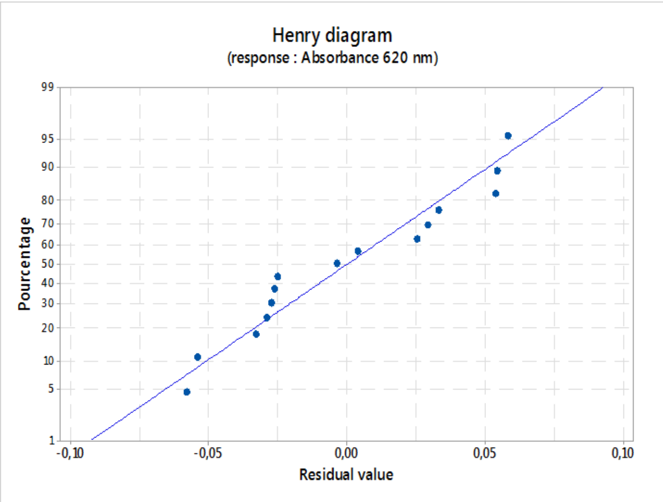


**Figure 2-c:** Henry Diagram of the absorbance of wavelength 620 nm

The quadratic terms seem to be perfectly important during the analysis of the results, hence the quadratic models perform much better than the linear models obtained by previous works (Assou et al., 2017b).

### 2.6 Henry Diagram

The following figures allow us to evaluate the normality of the color distribution, i.e., the percentage of acceptances for each reference value.

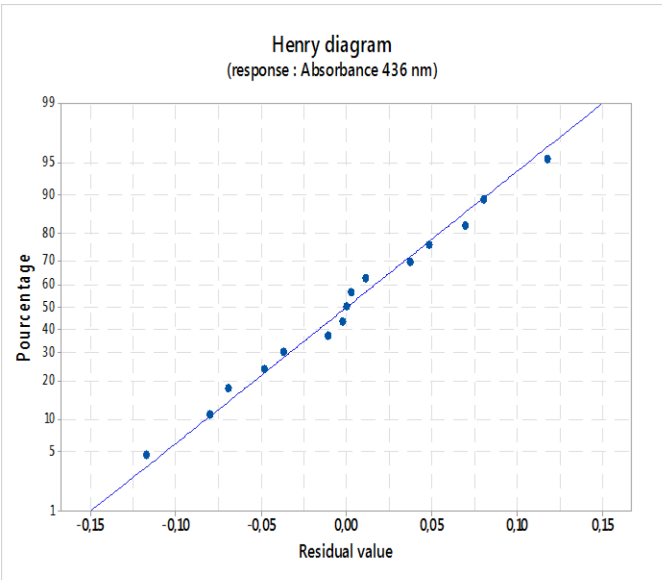


**Figure 2-a:** Henry Diagram of the absorbance of wavelength 436 nm

These lines fit a Gaussian distribution to the series of observations of the three absorbances studied. They allowed us to quickly read the mean and standard deviation of the distribution in question, which confirms that the models obtained, are more accurate by exploiting the interactions between the factors, in order to eliminate the color of the tannery wastewater.

## 3. Results and Discussions

### 3.1 Statistical Analysis of The Coefficients

Quadratic terms have nontrivial effects on color. The analyses that have been performed confirm this result.

**Table 2-A:** Statistical Analysis of Coefficients (The Absorbance of Wavelength 436 Nm)

| Factors | Weight of the factors | Standard Error | t value | P value |
|---|---|---|---|---|
| Constante | 0,4473 | 0,0619 | 7,22 | 0,001 |
| A | -0,0426 | 0,0379 | -1,12 | 0,0312 |
| B | -0,0564 | 0,0379 | -1,49 | 0,0197 |
| C | -0,0060 | 0,0379 | -0,16 | 0,0880 |
| A*A | 0,0032 | 0,0558 | 0,06 | 0,0956 |
| B*B | 0,1012 | 0,0558 | 1,81 | 0,0130 < 0.05 |
| C*C | 0,1210 | 0,0558 | 2,17 | 0,0082 < 0.05 |
| A*B | 0,0052 | 0,0536 | 0,10 | 0,00926 < 0.05 |
| A*C | 0,1230 | 0,0536 | 2,29 | 0,0070 < 0.05 |
| B*C | -0,0660 | 0,0536 | -1,23 | 0,0273 < 0.05 |

| **Table 2-b:** Statistical Analysis of Coefficients (The Absorbance of Wavelength 525 nm) | | | | |
|---|---|---|---|---|
| Factors | Weight of the factors | Standard Error | t value | P value |
| Constante | 0,2620 | 0,0357 | 7,34 | 0,001 |
| A | -0,0347 | 0,0219 | -1,59 | 0,0173 |
| B | -0,0258 | 0,0219 | -1,18 | 0,0292 |
| C | -0,0190 | 0,0219 | -0,87 | 0,0424 |
| A*A | 0,0121 | 0,0322 | 0,38 | 0,0722 |
| B*B | 0,0731 | 0,0322 | 2,27 | 0,0072 < 0.05 |
| C*C | 0,0656 | 0,0322 | 2,04 | 0,0097 < 0.05 |
| A*B | -0,0107 | 0,0309 | -0,35 | 0,00742 < 0.05 |
| A*C | 0,0677 | 0,0309 | 2,19 | 0,0080 < 0.05 |
| B*C | -0,0437 | 0,0309 | -1,42 | 0,0216 < 0.05 |

| **Table 2-c:** Statistical Analysis of Coefficients (The absorbance of Wavelength 620 nm) | | | | |
|---|---|---|---|---|
| Factors | Weight of the factors | Standard Error | t value | P value |
| Constante | 0,2823 | 0,0384 | 7,35 | 0,001 |
| A | -0,0446 | 0,0235 | -1,90 | 0,0116 |
| B | -0,0116 | 0,0235 | -0,49 | 0,0642 |
| C | -0,0325 | 0,0235 | -1,38 | 0,0226 |
| A*A | 0,0247 | 0,0346 | 0,71 | 0,0507 ≤ 0.05 |
| B*B | 0,0727 | 0,0346 | 2,10 | 0,0090 < 0.05 |
| C*C | 0,0530 | 0,0346 | 1,53 | 0,0187< 0.05 |
| A*B | -0,0163 | 0,0333 | -0,49 | 0,0646 |
| A*C | 0,0640 | 0,0333 | 1,92 | 0,0112< 0.05 |
| B*C | -0,0420 | 0,0333 | -1,26 | 0,0262< 0.05 |

The obtained models contain quadratic terms as well as interactions between factors, which have significant effects. These models are more adequate than the linear models. Statistical analyses confirm these results (p-value is below the threshold value 0.05).

### 3.2 ANOVA For The Models

| **Table 3-a:** Statistical Analysis of First Model (The Absorbance of Wavelength 436 nm) | | | | | |
|---|---|---|---|---|---|
| Source of variation | Degrees of Freedom | Sum of squares | Mean squares | F Ratio | Pr > F |
| Model | 9 | 0,204291 | 0,022699 | 1,97 | 0,00235 |
| Residual | 5 | 0,057535 | 0,019174 | | |
| Total | 14 | 0,261826 | | | |

| **Table 3-b:** Statistical Analysis of Second Model (The Absorbance of Wavelength 525nm) | | | | | |
|---|---|---|---|---|---|
| Source of Variation | Degrees of Freedom | Sum of Squares | Mean Squares | F Ratio | Pr > F |
| Model | 9 | 0,077465 | 0,008607 | 2,25 | 0,00192 |
| Residual | 5 | 0,057535 | 0,005571 | | |
| Total | 14 | 0,096578 | | | |

| **Table 3-c:** Statistical Analysis of Third Model (The Absorbance of Wavelength 620 nm) | | | | | |
|---|---|---|---|---|---|
| Source of Variation | Degrees of Freedom | Sum of Squares | Mean Squares | F Ratio | Pr > F |
| Model | 9 | 0,078735 | 0,008748 | 1,98 | 0,00234 |
| Residual | 5 | 0,022131 | 0,004426 | | |
| Total | 14 | 0,100866 | | | |

Very low probabilities of the studied responses showed that the absorbance models are well fitted and appropriate.

### 3.3 Graphical Representations of Response Surfaces

The experiments carried out during our research have enabled us to present the response surfaces of the objective functions, by showing the variations in the elimination of the color which correspond each time to the absorbances of different wavelengths. Furthermore, the optima of objective responses are more adequately indicated by iso-response projections. The following figures (3, 4 and 5) represent the variations in the color of wastewater from tanneries for different wavelength absorbances (436 nm, 525 nm and 620 nm), thus, the projection of each surface, in function of the coagulant (factor A), of the pH (factor B) and of the chimec 5161 type flocculant. The curves are represented in the form of three-dimensional curves. Each time the third factor is fixed, at a suitable value, while respecting the experimental range (maintenance value), to show the variations of the responses as a function of the other two, so that a significant elimination of the color is obtained.

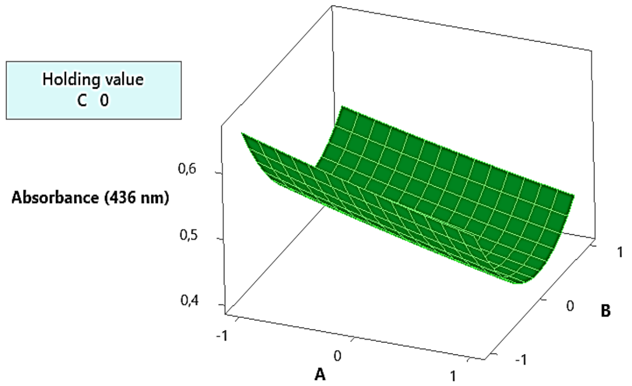


**Figure 3-a:** Surface of the absorbance of wavelength 436 nm as a function of A and B

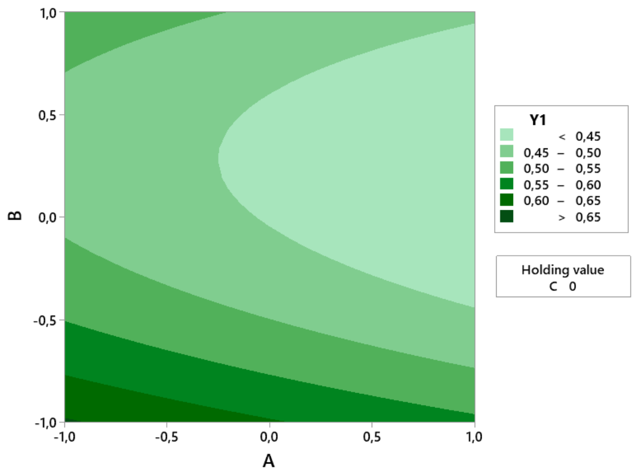


**Figure 3-aa:** Contour plot of the absorbance of wavelength 436 nm as a function of A and B

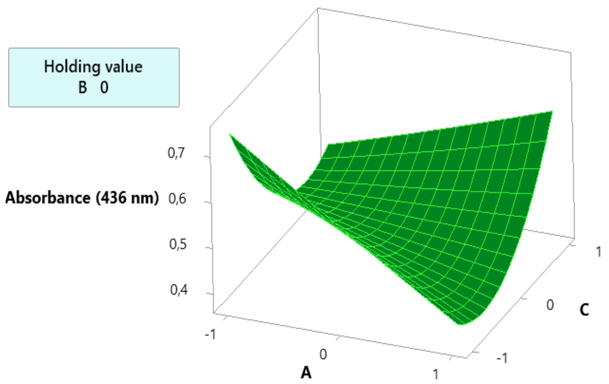


**Figure. 3-b:** Surface of the absorbance of wavelength 436 nm as a function of A and C

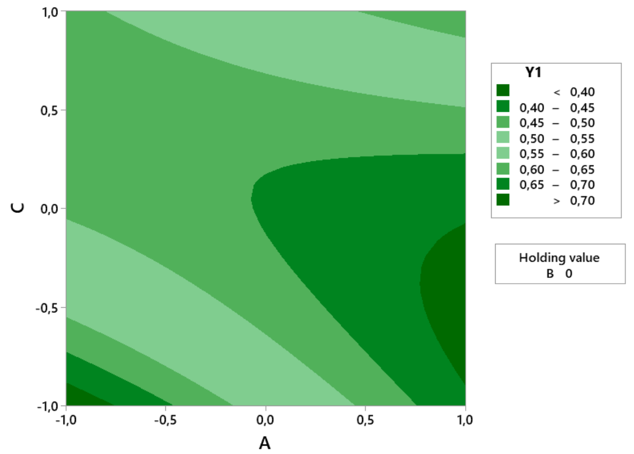


**Figure 3-bb:** Contour plot of the absorbance of wavelength 436 nm as a function of A and C

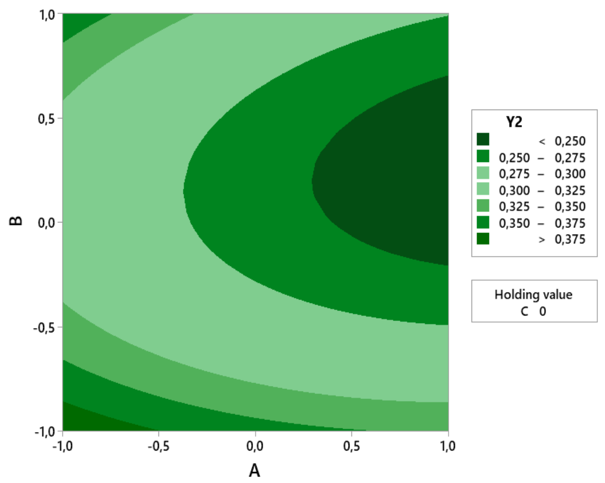


**Figure 4-aa:** Contour plot of the absorbance of wavelength 525 nm as a function of A and B

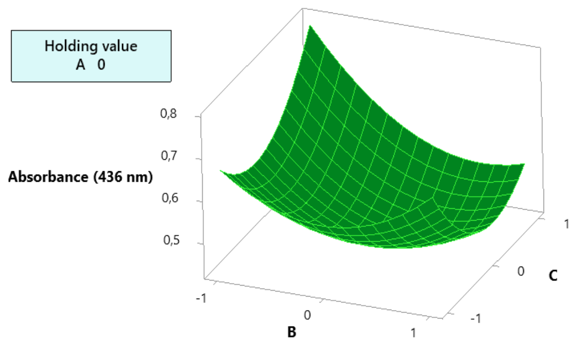


**Figure 3-c:** Surface of the absorbance of wavelength 436 nm as a function of B and C

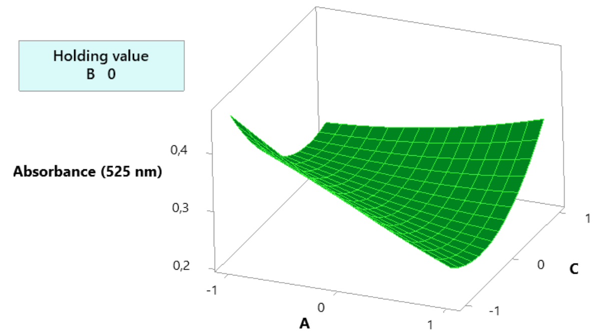


**Figure 4-b:** Surface of the absorbance of wavelength 525 nm as a function of A and C

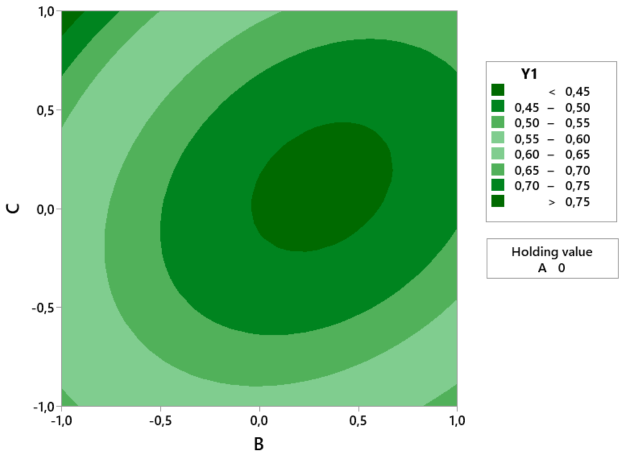


**Figure 3-cc:** Contour plot of the absorbance of wavelength 436 nm as a function of B and C

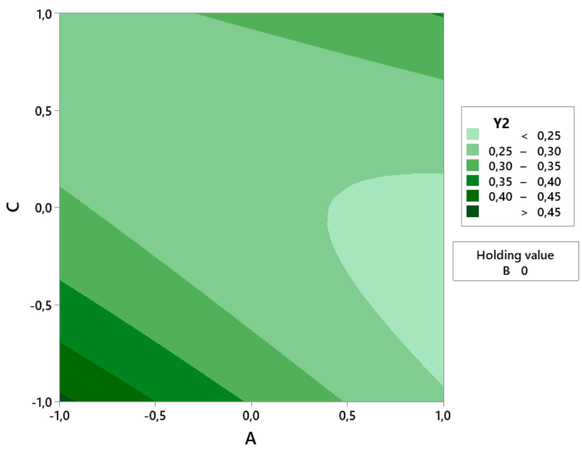


**Figure 4-bb:** Contour plot of the absorbance of wavelength 525 nm as a function of A and C

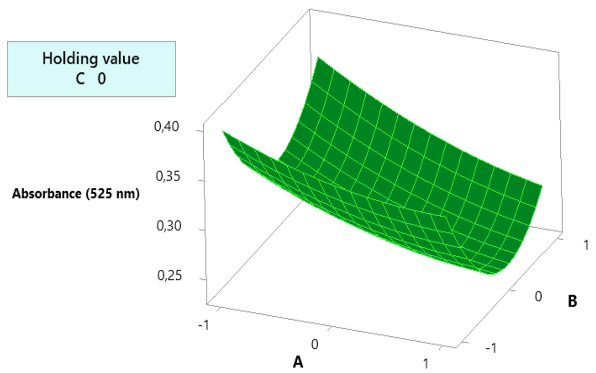


**Figure 4-a :** Surface of the absorbance of wavelength 525 nm as a function of A and B

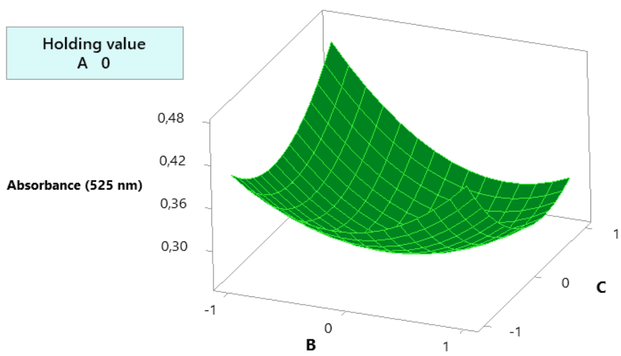


**Figure 4-c:** Surface of the absorbance of wavelength 525 nm as a function of B and C

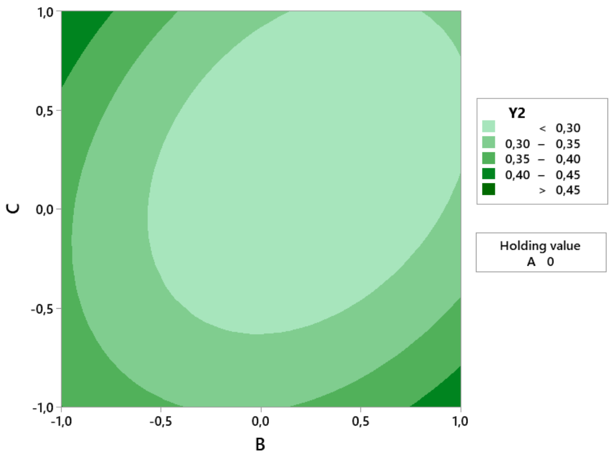


**Figure 4-cc:** Contour plot of the absorbance of wavelength 525 nm as a function of B and C

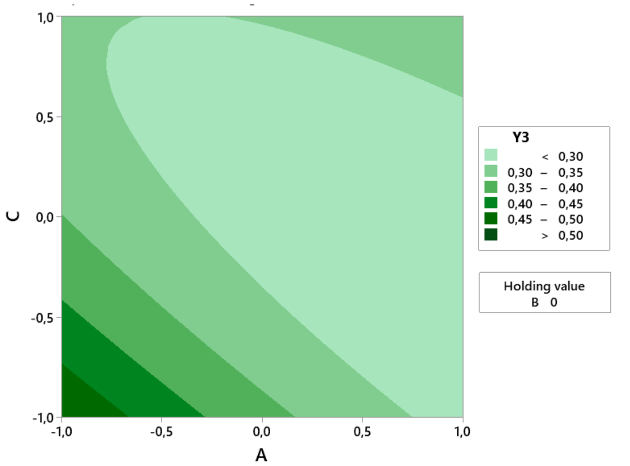


**Figure 5-bb:** Contour plot of the absorbance of wavelength 620 nm as a function of A and C

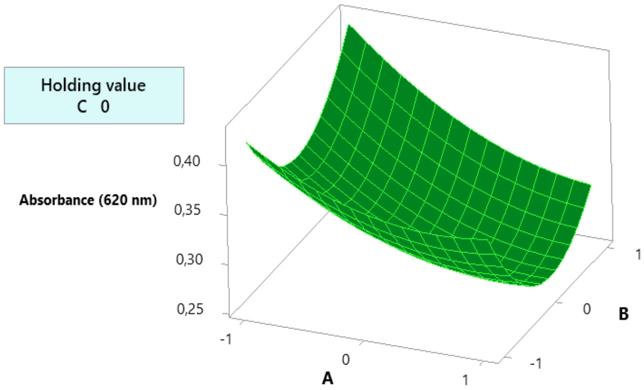


**Figure 5-a:** Surface of the absorbance of wavelength 620 nm as a function of A and B

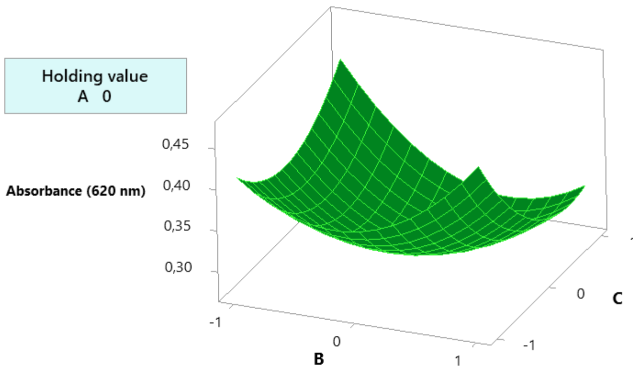


**Figure 5-c:** Surface of the absorbance of wavelength 620 nm as a function of B and C

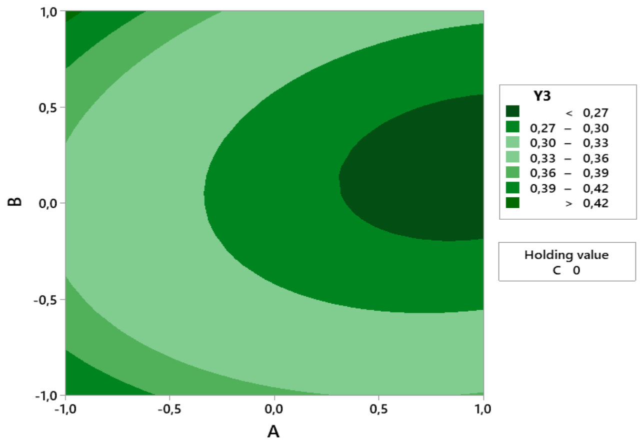


**Figure 5-aa:** Contour plot of the absorbance of wavelength 620 nm as a function of A and B

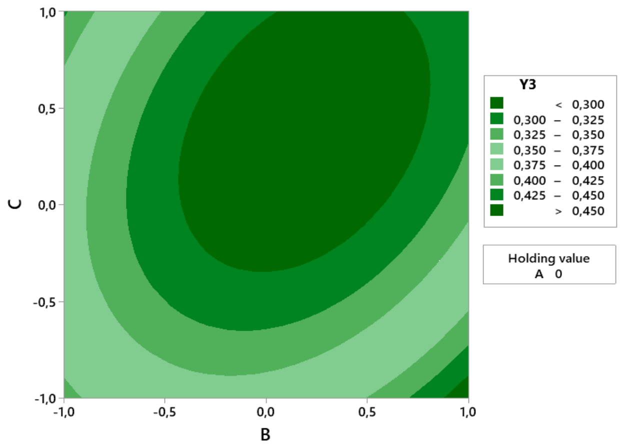


**Figure 5-cc:** Contour plot of the absorbance of wavelength 620 nm as a function of B and C

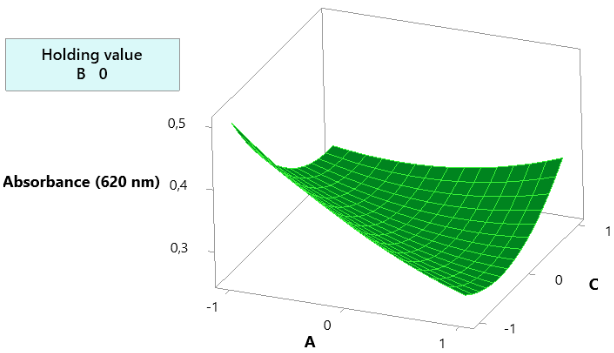


**Figure 5-b:** Surface of the absorbance of wavelength 620 nm as a function of A and C

As the absorbance measurement indicates the elimination of color, this optimization study is carried out on three responses (absorbances at 436 nm, 525 nm and 620 nm), in order to determine the optima of each of the most influential factors on the removal of color from tannery wastewater. Analyzing the response surface and iso-response figures, the maximum color removal efficiency (for the different wavelengths), was obtained using the aluminum sulfate reagent with the high pH value (6) in parallel with the use of anionic flocculant (chemec 5161). This confirms the results of previous work that $Al_2(SO_4)_3$ achieves higher removal efficiencies and generates higher color contents (Assou et al., 2017; Aboulhassan, 2008; Ellouzi and Halouali, 2015). As well as anionic flocculant (Chimec 5161), whose main role is to improve the quality of the treated water and further reduce the residual chromium content.

### 3.4 Validation of The Model

The adequacy graphs of the models are well represented in figures 6-a, 6-b and 6-c, in the form of a cloud of points which materializes the variations of the measured responses on the abscissa and the variations of the responses on the ordinate calculated from the quadratic models obtained (paragraph 2.3).

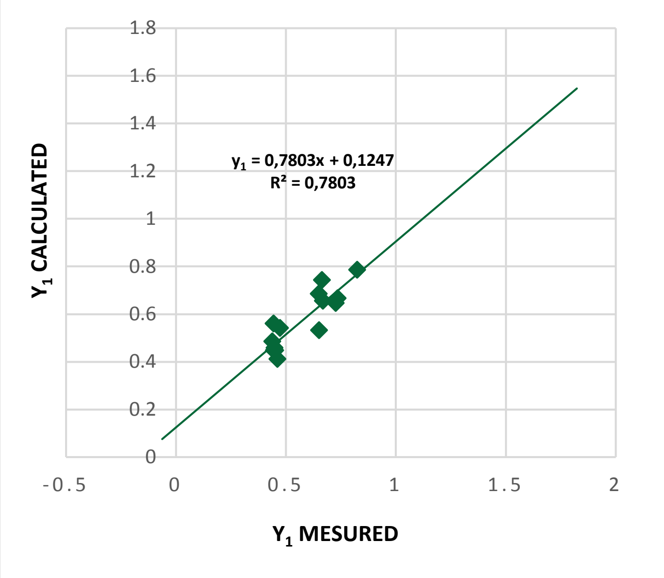


**Figure 6-a:** Correlation between theoretical and estimated yield (The absorbance of wavelength 436 nm)

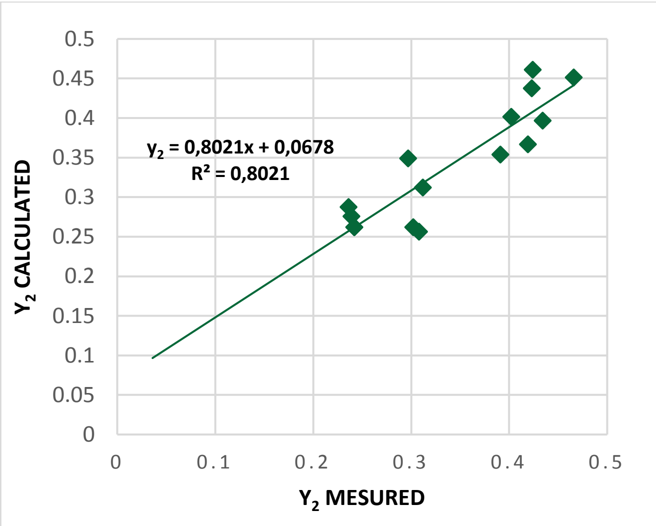


**Figure 6-b:** Correlation between theoretical and estimated yield (The absorbance of wavelength 525 nm)

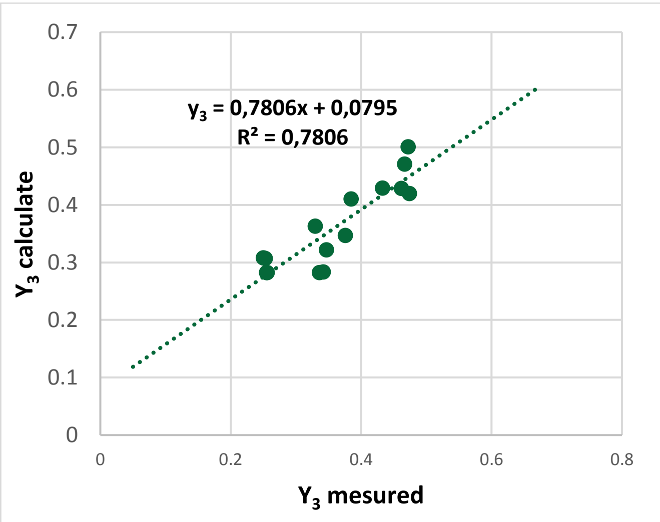


**Figure 6-c:** Correlation between theoretical and estimated yield (The absorbance of wavelength 620 nm)

The values of the coefficients of determination for the three responses are very close to unity, so they confirm the perfect agreement between the experimental values and the theoretical values of the absorbances at different wavelengths, hence the theoretical models obtained are very efficient (Pichet, 2020; Bosnic et al., 2013; Assou et al., 2017c).

## 4. Conclusion

The tannery's wastewater discharges have a devastating impact on the environment. For this reason, the fight against water pollution becomes a necessity, if not, we can at least reduce the harmful effects of these toxic discharges on the environment by techniques simple to implement and whose operation is possible. Coagulation-flocculation, in combination with experimental designs, is considered one of the most widely used techniques to clean up these discharges. In this study, the treatment of wastewater from an industrial tannery in Mohammedia, Morocco, was evaluated using the coagulation-flocculation process. The evaluation of the pollution of tannery wastewater is done by determining a number of physicochemical parameters that characterize them.

The work presented in this article, aims to better understand the methodology of experimental design in the research conducted on one of these parameters. An application of the concepts of this methodology was exploited, in order to bring answers to the problems of wastewater treatment. The response surface approach applied, has implemented the estimation of the respective roles of the most influential factors during the treatment of tannery wastewater: the coagulant, the flocculant and the pH on the color of the tannery wastewater which corresponds to the absorbances of different wavelengths. Optimal operating conditions are determined to obtain a significant reduction in pollution of treated water (elimination of discharges heavily loaded with organic matter). We have optimized the color of tannery wastewater by focusing on absorbances at different wavelengths, in order to create interesting quadratic models of objective functions. We measured the absorbance of wavelength 436 nm, 525 nm and 620 nm.

The optimization is carried out on these three responses which indicate the elimination of color, in order to determine the optima of each of the most influential factors on the discoloration of the waste water coming from the tannery. The analysis of the results obtained, based on the response surfaces, highlights the importance of using the aluminum sulphate reagent $Al_2(SO_4)_3$ in combination with the anionic flocculant Chemic 5161, and with an equal pH value to 6, in order to obtain a very high yield of color removal from tannery wastewater, for the three absorbances studied during the treatment (78.03%; 80.21%; 78.06%).

In view of the results obtained, aluminum sulphate makes it possible to generate higher color contents, in the presence of anionic flocculant (chemec 5161), while increasing the pH value in the study area considered. From the experiments carried out, we found that the chemec 5161 flocculant improves the quality of the treated water while showing the effectiveness of improving the quality of the treated water and further reducing the residual chromium content. Finally, the quadratic models seem perfect to describe the responses studied according to the significant factors mentioned during the treatment of tannery wastewater.

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