



RESEARCH PAPER

Modeling and Optimization by RSM for the Removal of the Dye "Palanil blue R" by Coagulation-Flocculation Process

Malika Chenna^{1,2} · Maya Kebaili^{2,3} · Nadia Lardjane⁴ · Nadjib Drouiche⁵ · Hakim Lounici²

Received: 13 February 2022 / Revised: 2 May 2022 / Accepted: 4 May 2022
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Abstract

This study aims to optimize the factors influencing the removal of textile dyes by a physicochemical treatment, coagulation-flocculation, using an experimental design. By carrying out the tests and analysing the data, the screening of the factors made it possible to identify the optimum conditions necessary to obtain better elimination. These operating conditions are pH, coagulant dose, the concentration of initial solutions, and stirring speed. Our study demonstrated the importance of applying the design of experiments methodology, particularly the response surface methodology (RSM). A full factorial design allowed the optimization of operational parameters affecting flocculation coagulation.

Article Highlights

- The factors influencing the removal of textile dyes by a physicochemical treatment have been studied.
- A full factorial design allowed the optimization of operational parameters affecting flocculation coagulation.
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Keywords Optimization · Response surface methodology · Textile dye · Coagulation-flocculation

Introduction

Water is the most important raw material on our planet, whether for humans, animals, plants, or microorganisms. Practically all the vital phenomena of the biosphere are linked to the availability of water. Therefore, water is not only a living space, an energy carrier, or a means of transport but also an essential element for all kinds of production.

Nowadays, the protection of our water wealth is a major problem for humanity. Important efforts are being made to preserve water resources, which are threatened by diffuse human pollution. Textile industry waste presents a threat to the environment because of its stability and biodegradability. The elimination of dyes from dye-laden waters is based on the use of several physical–chemical processes.

The novelty of this work is to contribute to this field; thus, the improvement of the elimination efficiency becomes a paramount necessity. Applying statistical techniques such as the response surface design makes this improvement increasingly possible (Gullon et al. 2021). These methods, which allow experimentation in a minimal number of experiments equal to 4, also allow factor screening.

The response surface methodology (RSM) has been proven to be a powerful tool to determine the effects of each factor and the interactions between them from the most influential to the least influential (Mahallati 2020) which allows extracting the maximum information with a minimum of experimental trials, more economical for the collection of research results than that of a classical variable at a time

✉ Nadjib Drouiche
nadjibdrouiche@yahoo.fr

¹ University of M'sila, Algiers, Algeria

² MDD Lab, University of Bouira, Bouira, Algeria

³ Centre de Recherche Mécanique, Chaab Erssas, Route de Aïn ElBey, BP 73B Constantine, Algeria

⁴ Faculty of Biological Sciences and Agronomic Sciences, University Mouloud Mammeri of Tizi Ouzou, Algiers, Algeria

⁵ CRTSE-Division CCPM- N°2, Bd Dr. Frantz FANON, P.O.Box 140, 16038 Alger Sept Merveilles, Algeria

or full factor experimentation, which can save time, money and lives (Yim et al. 2019). The experiment is expensive in time and means to characterize a process as accurately as possible.

In this context, this study investigates the efficiency and applicability of a physicochemical process called coagulation-flocculation for dye treatments using response surface designs. The experiments were designed according to the Factorial design with four factors, each at two different levels (Jahan et al. 2016; Ani et al. 2019) because it is simpler to implement and quickly highlights the existence of interaction between the factors.

The influence of the pollutant concentration, the quantity of catalyst ($\text{Al}_2(\text{SO}_4)_3$, FeCl_3), and the solution's pH on the quality of the process, were investigated. Using the complete factorial design methodology, a modeling of the effects of these different parameters on the kinetic constant will be performed. In this study, we optimized these influential factors and tested the quality of the coagulants used. The choice of a wastewater treatment process depends on several factors: the composition of the affluent, the type of reuse, the quality of the requirements, and the size of the plant.

Materials and Methods

Materials

The solutions were prepared from demineralized water and *palanil blue R*. The different concentrations are obtained simply by adding adequate amounts of palanil. Table 1 summarises the chemical characteristics.

- Aluminum sulfate powder 18 times hydrated ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$) with a molar mass of 666 g mol^{-1} and a purity between 95 and 98% of a PANREAC brand prepared by dissolution in distilled water;
- Iron chloride also called ferric chloride or iron perchloride is an iron salt with the chemical formula FeCl_3 . It is a very hygroscopic compound, which emits vapors in humid air under the effect of hydrolysis. The dissolution reaction in water is very exothermic and forms a brown acid solution. A stock solution of 10 mg L^{-1} was

prepared periodically by dissolving these powders in distilled water.

Methods

This study describes the treatment of a dye using the physico-chemical process, which consists of coagulation-flocculation at different concentrations of the coagulant ($\text{Al}_2(\text{SO}_4)_3$), at different dye concentrations, and different pH values to determine the optimal conditions for coagulation-flocculation. The Jar test of the Wise Stir® type was carried out on a flocculator with six one-liter beakers, each with an arm equipped with a paddle capable of generating a maximum rotation speed of 300 rpm and equipped with a lamp at the base that allows us to visualize the flocs formed. The experiments were performed at room temperature.

Six one-liter beakers were prepared by pouring in 1 L of the solution to be treated prepared from distilled water and dye; the different concentrations were obtained by dilution. The acidic or basic pH values were obtained by adjusting the solution with a 1 N NaOH and 1 N H_2SO_4 solution.

The beakers were placed on the flocculation ramp at $\frac{3}{4}$ of the depth. Coagulant was added simultaneously at different concentrations; the addition of coagulant was followed by rapid mixing in a beaker at about 200 rpm for 30 s. Stirring was continued for 60 min to obtain larger, more settleable flocs, with 40–150 rpm blade speeds. Samples were taken 30 s after adding the coagulant (after the high agitation phase), then every 1 min for a 10-min interval, and then every 5 min for a 60-min interval. Finally, the recovered samples were filtered through a Whatman N° 5 filter paper, and the filtered solution was analyzed by spectrophotometer, and the recovered flocs were characterized.

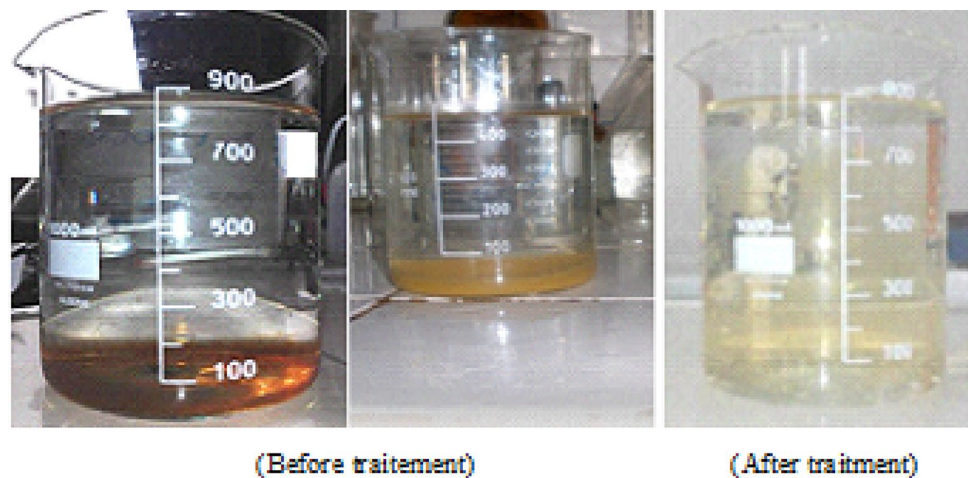
Identification of the Flocs Formed

The addition of coagulants associated with the coagulation-flocculation steps produces flocs that may contain some of the coagulants used in laboratory effluent treatment (Fig. 1).

Table 1 Characteristics of the dye used (palanil blue R)

Coded experimental variables (X_i)	Factors unity (U_i)	Lower level	Higher level
X_1	U_1 C_{dye} mg/l	30	70
X_2	U_2 Coagulant mg/l	3	7
X_3	U_3 stirring speed rpm	30	90
X_4	U_4 pH of Solution	4	10

Fig. 1 The dye before and after treatment with FeCl_3 coagulant



Results and Discussion

Application of Experimental Design Methods for Response Surfaces

We studied the influence of four factors: dye concentration (U1), coagulant concentration (U2), stirring speed (U3), and pH (U4) of the solution. We adopted the experimental research methodology, the ordered sequence of trials in an experiment following the experimental design approach. Each of these provides new knowledge that allows the effects of various factors on a response to be quantified and optimized in well-determined experimental areas (Goupy 2013). This is, in effect, the translation of the experiment matrix into real values. These matrices are meshes of a network of points uniformly distributed in the space of coded variables (X_i), allowing the estimation of the coefficients of a polynomial mathematical model of the second degree, whose essential interest is to be able to predict, at any point of the experimental domain, the values of the response (Myers et Montgomery 2002). The predictions thus calculated must be as close as possible to the values obtained by experimentation. In other words, the model's overall quality is important here and not the individual influence of the factors. Table 2 lists the experimental levels chosen for the four factors studied.

Definition of Objectives and Responses

The response is a grade that is measured at each experimental trial. It is the quantity of interest. In our case, it is the removal rate of the treated solutions and to estimate the effect of the factors identified as potentially influencing the observed response. The interactions will also be observed to complete the estimation of the average effects of the factors:

- U1: Dye concentration;

- U2: Concentration of coagulants (FeCl_3 or $\text{Al}_2(\text{SO}_4)_3$) added;
- U3: Stirring speed;
- U4: pH of the solution.

Experimental Area Chosen

The experimental domain consists of all the combinations of factors that can be realized (Goupy 2013). In our case, we study four factors with two levels. Table 2 summarises the different levels of the factors.

The experimental design used in this work is based on a mathematical model with four factors and eleven interactions. The response studied is the elimination rate, which translates into the following mathematical model:

$$Y = a_0 - a_1X_1 - a_2X_2 - a_3X_3 - a_4X_4 + a_{12}X_1X_2 + a_{13}X_1X_3 + a_{23}X_2X_3 - a_{14}X_1X_4 - a_{24}X_2X_4 - a_{34}X_3X_4 + a_{11}X_1X_1 + a_{22}X_2X_2 + a_{33}X_3X_3 + a_{44}X_4X_4 + a_{123}X_1X_2X_3 + a_{124}X_1X_2X_4 + a_{134}X_1X_3X_4 + a_{234}X_2X_3X_4 + a_{1234}X_1X_2X_3X_4.$$

With:

Y_1 : experimental response (FeCl_3);

Y_2 : experimental response ($\text{Al}_2(\text{SO}_4)_3$);

X_i : coded variables (-1 or $+1$);

a_i : estimated main effect of factor i for response Y ;

a_{ij} : estimated interaction effect between factor i and factor j for response Y .

The response studied in our work is the rate of removal ($R\%$) of the coagulation-flocculation reaction of the dye by the coagulants iron chloride (FeCl_3) and aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$). In the presence of four factors X_1, X_2, X_3, X_4 , consists of 24 experiments. The application of a model with interaction gives 24 parameters to determine. As a result, we have a saturated design, requiring additional tests in the

Table 2 Experimental range for palanil blue R mineralization

Essay N°	Experience matrix				Experimental design					
	X1	X2	X3	X4	U1	U2	U3	U4	Y1	Y2
1	0	− 1	− 1	0	50	3	30	7	64	8
2	1	1	0	0	70	7	90	10	56	55
3	0	− 1	0	− 1	50	3	60	4	65	45
4	0	1	− 1	0	50	7	30	7	45	88
5	0	0	0	0	50	5	60	7	70	84
6	0	1	1	0	50	7	90	7	30,2	75
7	1	0	− 1	0	70	5	30	7	36	85
8	0	0	1	− 1	50	5	90	4	78	69
9	0	1	0	− 1	50	7	60	4	85	69
10	0	0	1	1	50	5	90	10	65	78
11	0	0	0	0	50	5	60	7	63	74
12	0	1	0	1	50	7	60	10	62	74
13	1	0	0	− 1	70	5	60	4	74	78
14	− 1	0	0	1	30	5	60	10	85,2	96
15	− 1	0	0	− 1	30	5	60	4	74	85
16	1	0	0	1	70	5	60	10	69	68
17	0	0	− 1	− 1	50	5	30	4	65	70
18	− 1	− 1	0	0	30	3	60	7	66	69
19	− 1	0	1	0	30	5	90	7	47	84
20	− 1	1	0	0	30	7	60	7	85	75
21	1	− 1	0	0	70	3	60	7	74	85
22	0	− 1	0	1	50	3	60	10	72	69
23	0	− 1	1	0	50	3	90	7	71	69
24	1	0	1	0	70	5	90	7	76	78
25	0	0	0	0	50	5	60	7	78	74
26	0	0	− 1	1	50	5	30	10	75	74
27	− 1	0	− 1	0	30	5	30	7	77	78

Table 3 Experimental matrix and experimental design of dye removal rates with responses recorded for each trial Y1 (FeCl₃), Y2 (Al₂(SO₄)₃)

Coefficient	Value	Standard deviation	t statistic	T _{Ratio}
Constant	63	4.138941	15.22	< .0001
X ₁	8.4	2.069471	4.06	0.0016
X ₂	− 3.566667	2.069471	− 1.72	0.1104
X ₃	− 2.95	2.069471	− 1.43	0.1795
X ₄	− 4.55	2.069471	− 2.20	0.0483
X ₁ *X ₂	11.5	3.584428	3.21	0.0075
X ₁ *X ₃	12	3.584428	3.35	0.0058
X ₂ *X ₃	2.15	3.584428	0.60	0.5598
X ₁ *X ₄	12.7	3.584428	3.54	0.0040
X ₂ *X ₄	− 15.55	3.584428	− 4.34	0.0010
X ₃ *X ₄	3	3.584428	0.84	0.4190
X ₁ *X ₁	− 0.133333	3.104206	− 0.04	0.9664
X ₂ *X ₂	3.1666667	3.104206	1.02	0.3278
X ₃ *X ₃	5.1416667	3.104206	1.66	0.1235
X ₄ *X ₄	− 0.108333	3.104206	− 0.03	0.9727

center of the domain that allows for statistical analysis. The tests were carried out after randomization. Table 3 provides the matrix of experiments and the experimental design. It is necessary to make sure that the experimental design can achieve the objectives of the experiment with the desired precision (Baraka 2014; Torrades et Garcia-Montaña 2014).

Overall Analysis of Test Results

The statistical analysis is an aid to the interpretation of the results. It will allow us to calculate the coefficients of the models we are looking for and, above all, identify the factors that do not significantly influence the measured response and, consequently, can be removed from our study.

Mathematical Analysis of Test Results

The individual, quadratic, and interaction effects of the different factors were estimated (Table 4). A coefficient with a (+) sign means that the factor has a synergistic effect. On the other hand, a sign (−) indicates an antagonistic effect of the factor.

Table 4 Coefficient of abatement rate models (FeCl₃)

Coefficient	Value	Standard deviation	<i>t</i> statistic	<i>T</i> _{Ration}
Constante	70.555556	0.6348	111.15	< 0.0001
<i>X</i> ₁	− 0.4375	0.673307	− 0.65	0.5825
<i>X</i> ₂	− 4.0625	0.673307	− 6.03	0.0264
<i>X</i> ₃	− 7.3125	0.673307	− 10.86	0.0084
<i>X</i> ₄	− 10.6875	0.673307	− 15.87	0.0039
<i>X</i> ₁ * <i>X</i> ₂	4.8125	0.673307	7.15	0.0190
<i>X</i> ₁ * <i>X</i> ₃	3.0625	0.673307	4.55	0.0451
<i>X</i> ₂ * <i>X</i> ₃	− 5.0625	0.673307	− 7.52	0.0172
<i>X</i> ₁ * <i>X</i> ₄	0.1875	0.673307	0.28	0.8068
<i>X</i> ₂ * <i>X</i> ₄	− 1.6875	0.673307	− 2.51	0.1291
<i>X</i> ₃ * <i>X</i> ₄	− 7.9375	0.673307	− 11.79	0.0071
<i>X</i> ₁ * <i>X</i> ₂ * <i>X</i> ₃	8.5625	0.673307	12.72	0.0061
<i>X</i> ₁ * <i>X</i> ₂ * <i>X</i> ₄	3.4375	0.673307	5.11	0.0363
<i>X</i> ₁ * <i>X</i> ₃ * <i>X</i> ₄	2.6875	0.673307	3.99	0.0574
<i>X</i> ₂ * <i>X</i> ₃ * <i>X</i> ₄	− 2.1875	0.673307	− 3.25	0.0831
<i>X</i> ₁ * <i>X</i> ₂ * <i>X</i> ₃ * <i>X</i> ₄	1.1875	0.673307	1.76	0.2198

Table 5 Coefficients of abatement rate models with (Al₂(SO₄)₃)

Coefficient	Value	<i>t</i> _{crit} = 2.64
Constant	63	Significant
<i>X</i> ₁	8.4	Significant
<i>X</i> ₂	− 3.566667	Significant
<i>X</i> ₃	− 2.95	Significant
<i>X</i> ₄	− 4.55	Significant
<i>X</i> ₁ * <i>X</i> ₂	11.5	Significant
<i>X</i> ₁ * <i>X</i> ₃	12	Significant
<i>X</i> ₂ * <i>X</i> ₃	2.15	Significant
<i>X</i> ₁ * <i>X</i> ₄	− 12.7	Significant
<i>X</i> ₂ * <i>X</i> ₄	− 15.55	Significant
<i>X</i> ₃ * <i>X</i> ₄	3	Significant
<i>X</i> ₁ * <i>X</i> ₁	− 0.133333	Not Significant
<i>X</i> ₂ * <i>X</i> ₂	3.1.666667	Significant
<i>X</i> ₃ * <i>X</i> ₃	5.1.416667	Significant
<i>X</i> ₄ * <i>X</i> ₄	− 0.108333	Not Significant

Table 5 groups the effects of all the factors studied together with the statistical values of the *t* student and the observed probability (*p* value). The mathematical analysis consists of identifying the *p* coefficients of the models from the results of the *N* experiments carried out. The analysis of these results will make it possible to identify the factors that do not significantly influence the measured responses (Evrard et al. 2009).

Statistical Analysis of the Model

The statistical analysis involves implementing statistical tests, which refer to hypotheses and require knowledge

Table 6 Analysis of model coefficients

Coefficient	Value	<i>t</i> _{crit} = 3.68
Constant	70.555556	Significant
<i>X</i> ₁	− 0.4375	Not Significant
<i>X</i> ₂	− 4.0625	Significant
<i>X</i> ₃	− 7.3125	Significant
<i>X</i> ₄	− 10.6875	Significant
<i>X</i> ₁ * <i>X</i> ₂	4.8125	Significant
<i>X</i> ₁ * <i>X</i> ₃	3.0625	Not Significant
<i>X</i> ₂ * <i>X</i> ₃	− 5.0625	Significant
<i>X</i> ₁ * <i>X</i> ₄	0.1875	Not Significant
<i>X</i> ₂ * <i>X</i> ₄	− 1.6875	Not Significant
<i>X</i> ₃ * <i>X</i> ₄	− 7.9375	Significant
<i>X</i> ₁ * <i>X</i> ₂ * <i>X</i> ₃	8.5625	Significant
<i>X</i> ₁ * <i>X</i> ₂ * <i>X</i> ₄	3.4375	Not Significant
<i>X</i> ₁ * <i>X</i> ₃ * <i>X</i> ₄	2.6875	Not Significant
<i>X</i> ₂ * <i>X</i> ₃ * <i>X</i> ₄	− 2.1875	Not Significant
<i>X</i> ₁ * <i>X</i> ₂ * <i>X</i> ₃ * <i>X</i> ₄	1.1875	Not Significant

Table 7 Coefficient analysis for the coagulant Al₂(SO₄)₃

Source	Sum of squares	DDL	Mean square	<i>F</i> _{observed}	<i>F</i> _{critic}
(a) (FeCl ₃)					
Bond	4329.2841	14	309.235	61.47	4.10
Residues	616.7100	12	51.393		
Total	4945.9941	26			
(b) Al ₂ (SO ₄) ₃					
Bond	6511.93	15	434.12	59.85	3.68
Residues	14.50	2	7.253		
Total	6526.44	17			

of variability. Table 6 presents the model coefficients, standard error values, *t* value, and *p* value of (Prob > |*t*|) for each factor and interaction. Student's *t* student test (parametric test): The *t* student values are used to determine the significance of the coefficients of each parameter as it compares the observed mean value of a statistical sample to a fixed value. If *t_i* > *t_c*, then the coefficient is significant; otherwise, it is rejected from the model. Recall that a factor is significant at 5% when it is experimentally observed student value (*t*_{obs}) is greater than or equal to the student value (*t_c*) at a 95% confidence level.

For a significance, level α equal to 5%:

Case of FeCl₃ : $t_{\text{critical}} = t(\alpha/2, N - p)t_{\text{critical}} = 2.64$,

N: number of tests performed;

P: number of coefficients of the model;

Table 8 Regression analysis (a) (FeCl_3) , (b) $\text{Al}_2(\text{SO}_4)_3$

Source	Sum of square	DDL	Mean square	F_{observed}	F_{critic}
a) (FeCl_3)					
Bond	4329.1	14	309.235	61.47	4.10
Residues	616.71	12	51.393		
Total	4945.9941	26			
(b) $\text{Al}_2(\text{SO}_4)_3$					
Bond	6511.93	15	434.12		
Residues	14.50	2	7.253		
Total	6526.44	17			

The results of the coefficient analysis are shown in Table 6 (FeCl_3) and Table 7 ($\text{Al}_2(\text{SO}_4)_3$).

It can be seen that the higher the absolute value of the coefficient, the more significant the single factor or interaction will be on the response studied.

Analysis of Variance

The analysis of variance is a means of validating a mathematical model using the Fisher criterion, mainly comparing two residual and experimental dispersions. The results indicate that the main effect of the regression is significant since the probability of significance of the p value risk is less than 0.05 (Table 8).

F_{critical} readings from the Fisher-Snedecor table with (p-1) and (n-p) degrees of freedom and a 95% confidence level are 4.10 for FeCl_3 and 3.68 for $\text{Al}_2(\text{SO}_4)_3$. The F_{obs} value adequately explains the variation of data from their mean value. In addition, it attests to whether the main effect is significant (Tinsson 2010; Sado and Sado 2001).

We notice that $F_{\text{obs}} \gg F_{\text{theoretical}}$. Thus, we accept hypothesis H: “the mean square of the regression is significantly greater than the mean square of the residuals”. The regression is globally significant (Evrard et al. 2009). It can be seen that the value of the probability F_c is much lower than 1%, so we can say that the model allows describing correctly the variation of the test results for the response. The regression is, therefore, significant at a confidence level of about 99.99% (Montgomery 2001). The observed differences in the influential variables at different operating points suggest interactions between the parameters.

The coefficient of determination R^2 and adjusted R^2 , which are indices of the quality of the regression being equal to 1.088, the value of R^2 and adjusted R^2 are very close to 1. The coefficient of determination gives several indicators of the quality of the model, thus good compatibility between the experimental and predicted values of the fitted model. Therefore, we can deduce that the mathematical model is

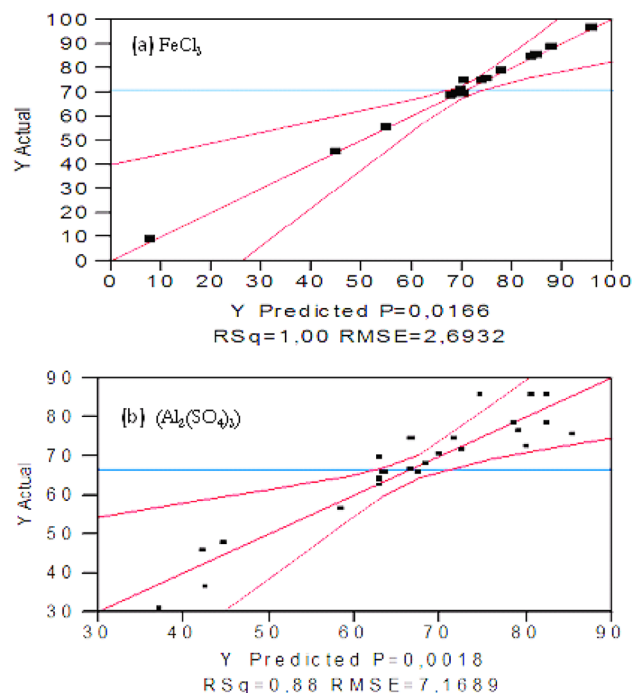
satisfactory and accepted because there is a good agreement between the data and the predicted removal rate from the model.

Model Fit Graph

Figure 2 shows that the descriptive quality can be observed through a model fit graph. It allows comparing the answers calculated by the model with the answers measured by this one. The bisector represents the line of correspondence. The two graphs confirm that the curve of the observed values according to the predicted values has perfectly the aspect in the bisector of a straight line. Indeed, the cloud of points is on the straight line for iron chloride and between the red dotted lines that delimit the confidence zones for alumina sulfate. The descriptive quality is well represented in a plane. The proposed models strongly approach the phenomenon of elimination. Inevitably some points are poorly represented for alumina sulfate.

Circle of Correlations

The objective is to study the relationship between k quantitative variables. The results are given in graphs: a graph of variables and a graph of individuals. The graph of variables is given by the circle of correlations, with radius $R = 1$. It allows us to see which variables are explained by the factorial axes (a variable close to 1 and parallel to an axis is well

**Fig. 2** Fuitability graph of the model abatement rates interaction

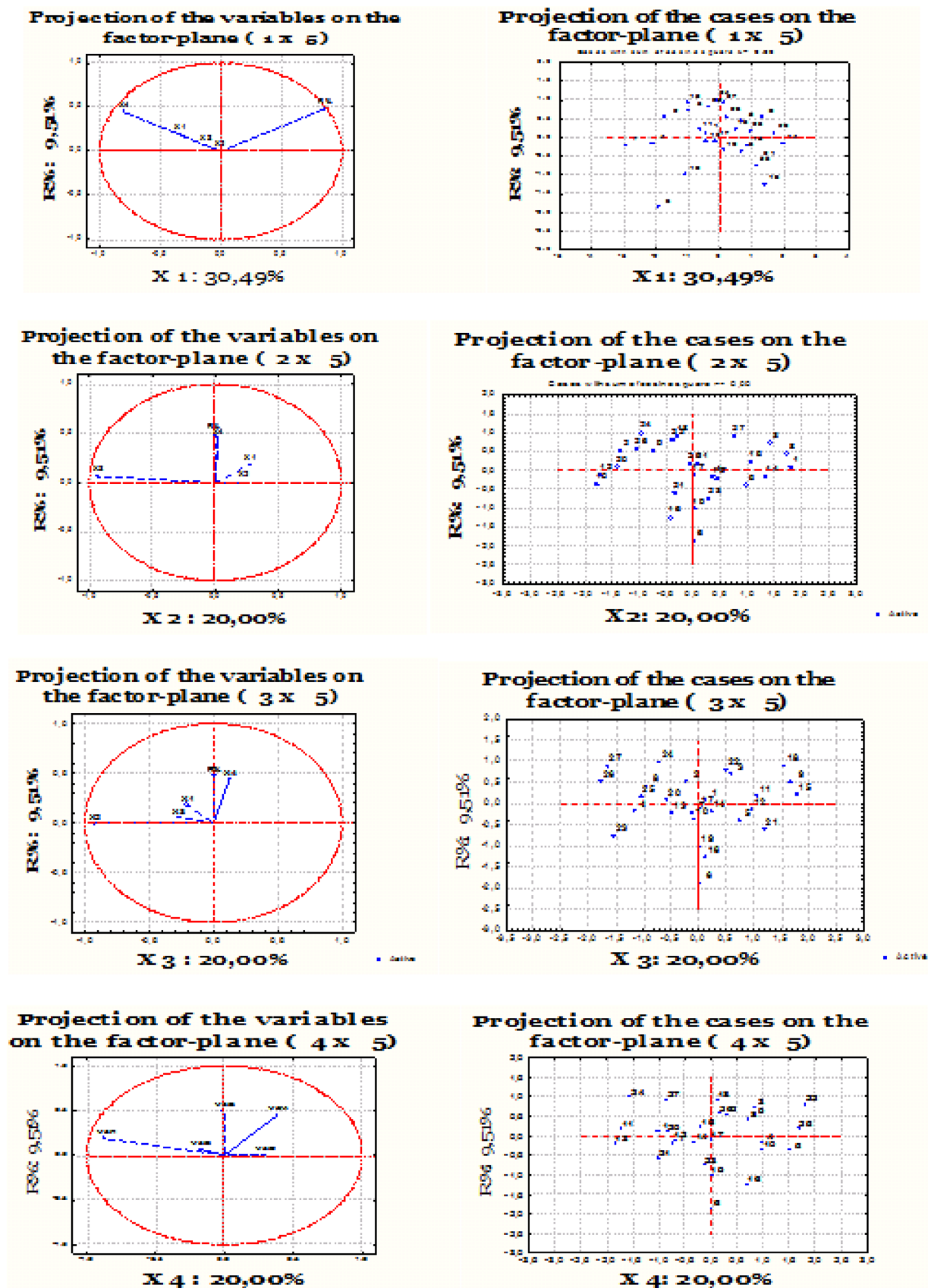
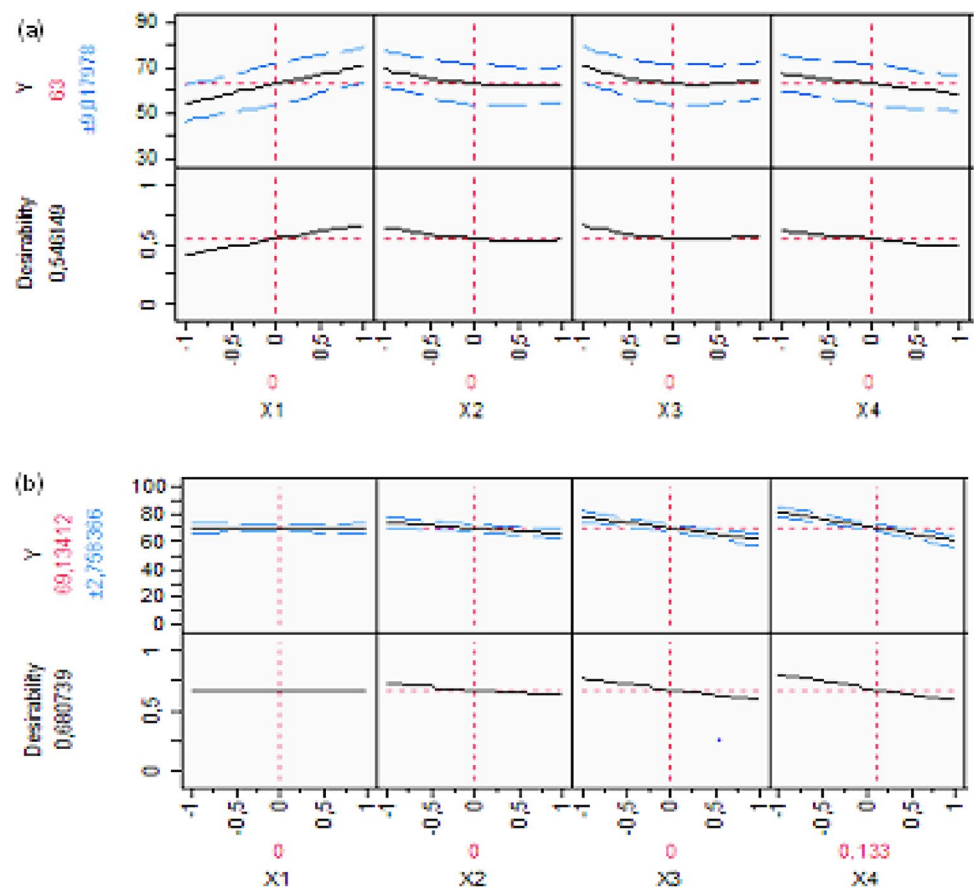


Fig. 3 Projection of variables on the factorial plane (2×5) and projection of individuals on the factorial plane (3×5), (FeCl₃)

Fig. 4 Effects of **a** FeCl_3 , **b** $\text{Al}_2(\text{SO}_4)_3$



represented) and which are correlated with each other (those close on the factorial axis). When two perpendicular variables are uncorrelated, two opposite variables are negatively correlated (Taylor 2017).

The graph of the individuals is read simultaneously with that of the variables. It allows deducing the individual who gives the best (or the lowest) value of the variable considered in the reading. Before analyzing the correlation circle, it is important to check if the variables are well represented on the graph.

It is often agreed that:

- If the share of information exceeds 70%, the variable is very well represented;
- If the share of information is less than 70%, the variables can be, for example, 60% or poorly represented.

In our case, the variables most related to axis1 (Z_1) are X_1 and X_3 . The vectors corresponding to these two variables will have the same direction but opposite directions because the correlation coefficients do not have the same sign. This means that when one increases, the other decreases. The variables X_5 and X_2 are the most related to axis 2 (Z_2). Their direction of variation is also opposite. In general, the

relationship between the different variables is defined by the value of the angle:

- X_1 and X_2 are positively correlated;
- X_1 and X_3 are inversely correlated;
- X_4 is inversely correlated with X_3 and X_2 ;
- X_3 and X_2 are independent.

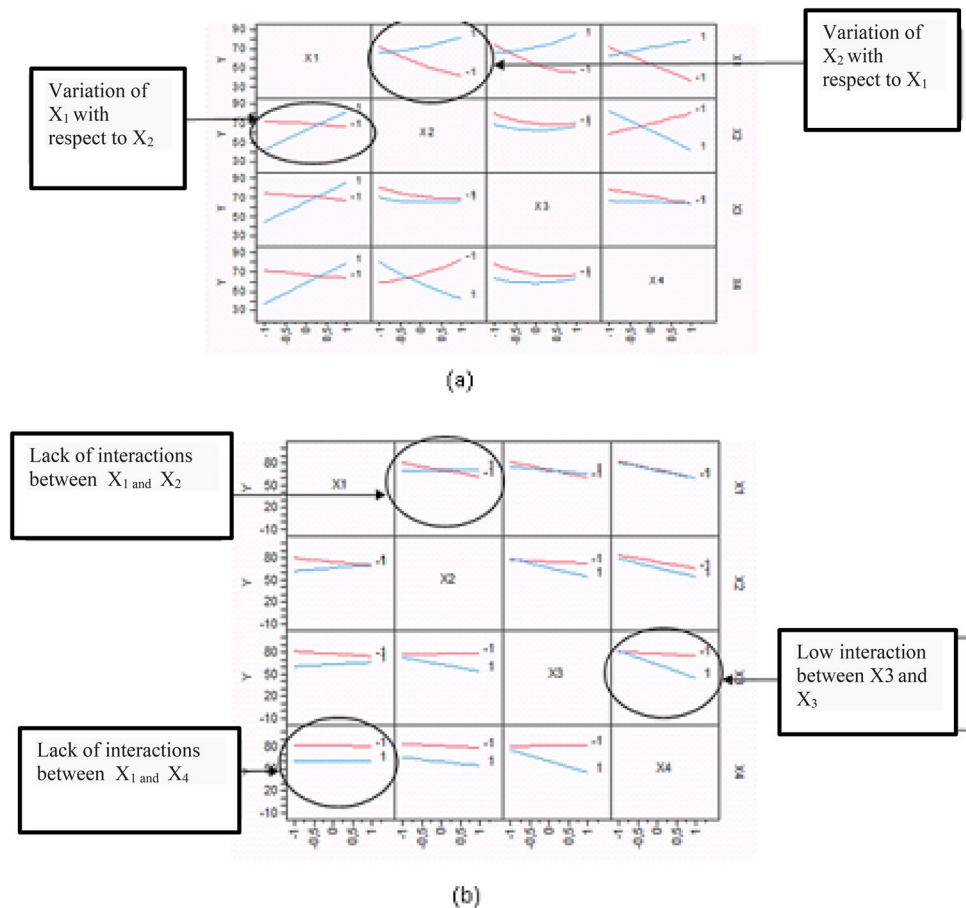
When the absolute value of the angle between the factors between 0° and 90° : The variables are correlated in the same direction. For example, when the value of X_1 increases, the value of X_4 increases, the value of X_2 also increases.

When the absolute value of the angle between the factors is between 90° and 180° : The variables are inversely correlated. When X_1 increases, X_3 decreases. In the same way, when X_2 increases, X_3 decreases.

When the angle is close to 90° : the variables are independent, i.e., uncorrelated. For example, X_1 and X_2 vary independently. There is no relationship between these two variables.

Figure 3 shows that an excellent correlation exists between the experimentally obtained removal rate and that which the mathematical relationship could predict.

Fig. 5 Interaction plot: **a** FeCl_3 ,
b $\text{Al}_2(\text{SO}_4)_3$



Plotting of Factor Effects Graphs

The effect of the bills is shown graphically in Fig. 4: (a) iron chloride and (b) aluminum sulfate. To interpret the results suggested by this type of graph, it is sufficient to compare the slopes of each of the segments of the straight lines whose ends correspond to the average effects calculated at the -1 and $+1$ levels. The greater the line slope characterizing the effect, the greater the factor's weight (Kitous et al. 2014).

Interaction Plot

In a complex system, the parameters are often coupled. Knowing the effects of each parameter on the response is not possible because when you change the level of one factor, you change the effects of all other factors. Interpretation based solely on the main effects of the factors would be error-prone. Therefore, information is needed on the influence of the variation of each factor on the effect of the other factors. This notion, called interaction (Erper et al. 2011), is represented graphically by Fig. 5. When observing the graphs, there are two interactions whose effect is statistically significant. These are coagulant concentration and dye concentration, coagulant concentration, and pH, but the

interaction between time and pH and stirring speed and pH are weak interactions. The effect of coagulation time on the response is not the same when changing the temperature factor. Such interactions should not be neglected in future investigations and projects.

Graphical Representation of Response Surfaces

The response surface is defined as the dimensional space associating all factors with a response. Response surfaces can show the variation of responses as a function of only two factors at a time, with the other factors set to a fixed value (Goupy 2013). The response surfaces and contours are shown in Figs. 6, 7, and 8 for iron chloride and aluminum sulfate. The graphs can be used to visualize and determine the optimal conditions in the defined study area. By fixing the level of an independent variable at the center of the experimental range, it is possible to follow the evolution of the other two variables and their influence on the percentage of water discoloration.

Based on Figs. 6, 7, and 8, we can question the relevance of the chosen model. The response surfaces seem to be parabolic (Kitous et al. 2014; Tinsson 2010). We can

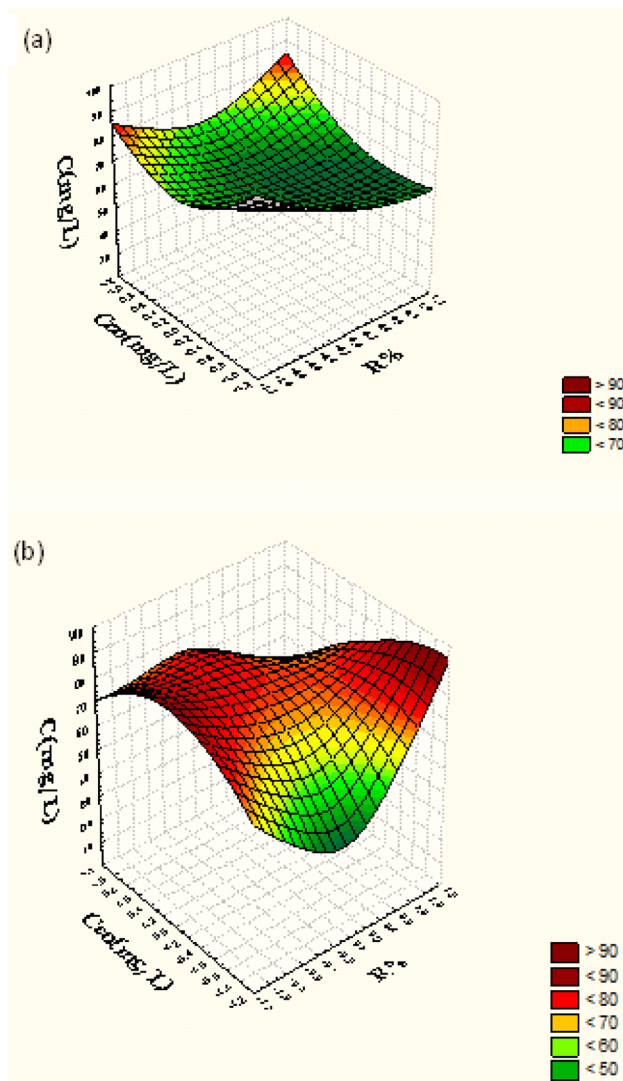


Fig. 6 The response surface as a function of dye concentration X_1 and coagulant concentration X_2 (iron chloride) in a three-dimensional space **a** FeCl_3 , **b** $\text{Al}_2(\text{SO}_4)_3$

deduce that the chosen model is very close to the elimination phenomenon.

Conclusion

By applying the design of experiments methodology, we reduce the number of necessary tests by half, which allows a considerable saving of time and energy, the use of charts and graphs is a convenient way to exploit the results. These results can be a basis for extracting knowledge about the system and studying the extrapolation of experimental conditions. The analysis of the data allowed us to find on the one hand the existing effects between the different parameters,

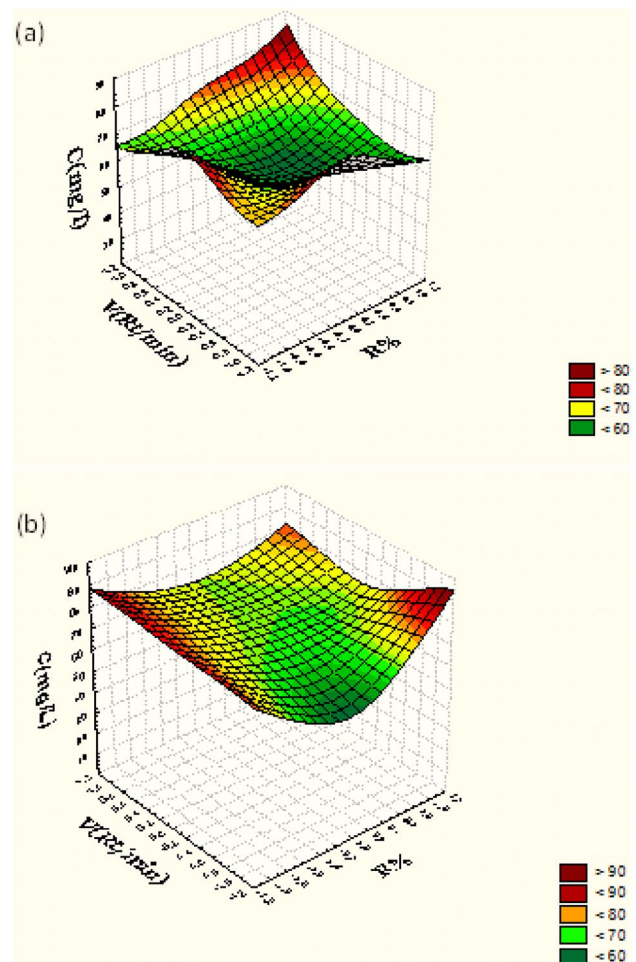


Fig. 7 The response surface as a function of dye concentration X_1 and stirring speed X_3 in three-dimensional space

establishment of a mathematical model, and correctly identify their influences on the response.

A screening plan allowed us initially to classify the factors in order of their influence on the elimination of dyes by coagulation-flocculation, this study highlighted the complementarity of the sequence of these methods, the description of the mathematical analyzes and statistical analyzes allowed us to identify the most influential parameters and to optimize them, to find the correlation (positive and negative) existing between the different factors.

The obtained results made it possible to draw the following conclusions:

- The models are most often linear sometimes with interactions (X_1 , X_2), rarely with square terms; since the chemical laws which underlie chemical phenomena are not necessarily so, it is desirable to limit the gap between the limits of the domain. The results presented in this

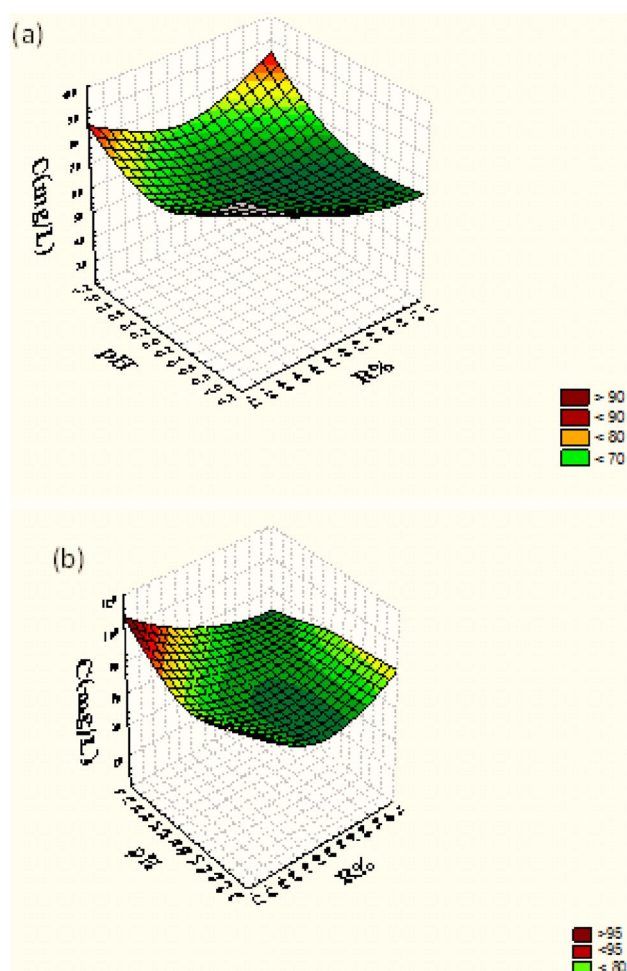


Fig. 8 The response surface as a function of dye concentration X_1 and pH in three-dimensional space **a** FeCl_3 , **b** $\text{Al}_2(\text{SO}_4)_3$

report provide information on the efficiency of the coagulation-flocculation process with iron chloride. However, the setting up of pilot projects using this technology is essential to assess with more certainty its profitability and effectiveness.

- The results show that the type of coagulant, the coagulant concentration, and the pH influence the percentage of elimination, given the high values of their effects observed compared to the effects of other factors. The type of coagulant and its concentration have a negative influence on the elimination as it decreases from ferric chloride (FeCl_3) to aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$) and from a concentration of 1 ppm to a concentration of 3 ppm. In contrast, the acidic pH has a positive effect. The existence of interactions between coagulant-pH and coagulant concentration and dye concentration means that the effects of the main factors vary according to the hydrogen potential. This leads us to study the effects of interactions between the parameters.

Funding This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Availability of data and materials The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

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Chapter 15 - Supercritical Extraction of Value-Added Compounds From Empty Fruit Bunch: An Optimization Study by Response Surface Methodology