Optimization of adsorption of cationic dye from aqueous solution by biochar from artichoke waste using response surface methodology

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Abstract: The use of experimental design and in particular the response surface methodology (RSM) allowed the determination of the influence of the simultaneous effects and the interaction of the operating parameters on the methylene blue removal efficiency. The parameters studied were the initial concentration of the adsorbate, the stirring speed, mass and particle size of the adsorbent. The results show that the application of RSM allows describing the influence of these four experimental parameters on the treatment effectiveness. The second-order model obtained, for the Methylene Blue (MB) removal efficiency was validated by using different statistical approaches. The use of the ANOVA showed that the model is significant and in functional adequacy with the experimental results.

Keywords: Adsorption, Experimental design, response surface methodology RSM, ANOVA.

1. Introduction

The discharges of the effluent colored in nature are not only unpleasant for the environment but also affects a lot of biological cycles. These discharges present a real danger to humankind and the environment by cause of their poor biodegradability [1–9]. Several treatments have been used to reduce the harmful effects of effluent discharge [10]. One of the most used techniques in water purification discharges of these industries is adsorption, which is a technique of low cost compared to other methods used in the bleaching process [11–18].

The design of experiments methodology (DEP) is a strategy for planning scientific and industrial experiments to remove information corresponding to the objective that has been previously set. The analysis of the information collected uses statistical methods [19–21].

It can be applied to many disciplines and industries as long as we search for the correlation between a quantity of interest (Y) and variables (X_i) [22]. In this method, the response can be written as follows:

$$Y = f(X_1, X_2, X_3, \ldots\ldots\ldots\ldots, X_k) + \varepsilon \qquad (1)$$

Where f is the response function that depends on random variables X_i; ε is a term that represents other sources of variability.

This work consists of optimizing the removal of MB dye by adsorption. The influence of the pollutant concentration, the stirring speed, mass and particle size of the adsorbent was also studied using the response surface methodology. This method allows, through empirical mathematical models to determine an approximate relationship between the output response and the input variables to optimize the process parameters in order to achieve a desirable response [23–28].

The advantages of this method are:

a decrease in the number of tests,
a higher number of factors studied,
a determination of the interactions between factors,
a better precision on the results [29].

2. Materials and methods

2.1. Adsorbent
The adsorbent used in this work is artichoke waste, which was washed, then dried in an oven at 100°C for 24 hours. It was crushed, then carbonized at 650°C for one and a half hours.
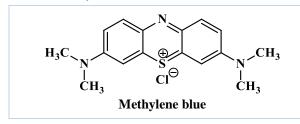
The material used as an adsorbent in this study is the biochar, which can be obtained from the artichoke waste [30].

2.2. Adsorbate
The dye we used is methylene blue (Table 1). The concentration

of the dye solution MB was determined using a UV-vis spectrophotometer (Shimadzu UV-mini spectrometry 1240) in maximal wavelength of 664 nm. The percentage removal of the contaminant was calculated by the following equation [31]:

$$R(\%) = \frac{C_0 - C_e}{C_0} *100 \qquad (2)$$

Where C_0 and C_e are respectively, the initial and equilibrium concentrations of dye (mg l^{-1}).

Table 1. Methylene blue characteristics.

	Chemical formula	$C_{16}H_{18}N_3S^+$ Cl$^-$
	Charge	Positive
	Molar mass (g mol^{-1})	373,90
	Solubility (g l^{-1})	50
Methylene blue	Wave length (nm)	664

2.3. Mathematical model

The behavior of the response surface was explained by a full second-order polynomial model, as shown in the following equation [28,32–35]:

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i X_i \sum_{i=1}^{n} \beta_{ii} X_i^2 + \sum \sum_{i=1 \ j=1}^{n} \beta_{ij} X_i X_j \qquad (3)$$

This type of model can estimate a response surface to study the linear effects, quadratic effects and interaction effects:

- Y is the response function.
- β_0 is the constant polynomial that expresses the general average effect.
- β_i, β_{ii}, β_{ij} are the coefficients of the linear effect, quadratic and interaction, respectively.
- X_i and X_j represent the independent coded variables.

The NEMRODW [36] software was used to determine the polynomial coefficients for the response. The degree of significance of the coefficients was determined using the test student. Verification of the fitted model was performed by the regression coefficient (R^2).

2.4. Matrix experience and experimental field

We chose a central composite design in a cubic field allowing 3 levels of factor which are (-1), (0) and (1) as shown in Table 2. The experience of the number of central plans is determined using the following equation (4). According to this equation [33], the total number of tests to be performed for k = 4 and $n_0 = 2$, is of 27 tests. The results are summarized in Table 3.

$$N = 2^K + 2K + n_0 \qquad (4)$$

Where k is the number of input factors. And n_0 the number of field tests in the center.

Table 2. Experimental field.

Factors	Level (-1)	Level (0)	Level (1)
Concentration of dye (mg l^{-1}) (X_1)	10	20	30
Mass (mg) (X_2)	90	107.5	125
particle size (mm) (X_3)	0.08	0.115	0.15
stirring speed (rpm) (X_4)	800	1200	1600

Table 3. Matrix of tests and experimental response.

N°Exp	X_1 (mg l^{-1})	X_2(mg)	X_3 (mm)	X_4(rpm)	Y(%)
1	-1	-1	-1	-1	72.76
2	1	-1	-1	-1	54.50
3	-1	1	-1	-1	79.32
4	1	1	-1	-1	54.22
5	-1	-1	1	-1	36.23
6	1	-1	1	-1	51.61
7	-1	1	1	-1	42.77
8	1	1	1	-1	51.96

9	-1	-1	-1	1	62.90
10	1	-1	-1	1	53.63
11	-1	1	-1	1	75.31
12	1	1	-1	1	49.32
13	-1	-1	1	1	30.90
14	1	-1	1	1	52.31
15	-1	1	1	1	49.82
16	1	1	1	1	53.32
17	-1	0	0	0	56.38
18	1	0	0	0	53.54
19	0	-1	0	0	40.74
20	0	1	0	0	47.03
21	0	0	-1	0	49.19
22	0	0	1	0	38.50
23	0	0	0	-1	44.41
24	0	0	0	1	44.27
25	0	0	0	0	41.13
26	0	0	0	0	37.31
27	0	0	0	0	39.64

3. Results and Discussion

3.1. Statistical analysis of the results

In order to determine the quality description of the model and to know if the model is correct, it is necessary to realize the statistical analysis of the model in its totality. A variance analysis Table 4 (ANOVA) is constructed using the statistical test of Fisher [37,38]. The analysis of variance is to explain the total variation of the response which decomposes into a sum of squares of the variation due to the regression and a sum of squares differences or residuals [31,39].

The quality of the adjustment of the model was verified by the coefficient of determination (R^2) [34]. In this case, the value of the coefficient of determination ($R^2 = 0.9697$) indicates that only 2.06 % of the total variations are not explained by the model. The value of the adjusted coefficient of determination ($R^2_{adj} = 0.9344$) is also very high, indicating a high significance of the model, using the statistical test of Fisher.

Table 4. ANOVA Results.

Source of variation	Sum of squares	degree of freedom	mean square	Fc	Fcritique
Régression	3467.9866	14	247,713	Fc=27.493	2.64
Résidus	108.12224	12	9.010		
Total	3576.373	26			

3.2. Statistical analysis of the coefficients of the model

The purpose of this statistical test is to know if there are coefficients that are not influential, that is to say, which do not affect each response. The estimated values of the coefficients of the model and the significance are given in Table 4.

Analysis of the results shows that the concentration (X_1), particle size (X_3) and the stirring speed (X_4) have a negative influence. Indeed, the minimum value of these factors results in a higher response. X_2 factor (the mass of adsorbent) has a positive sign indicating that the maximum value of this factor results in a higher response.

Table 5. Analysis table coefficients

Nam	Coefficien	Standard Deviatio	t.exp.	Signif. %

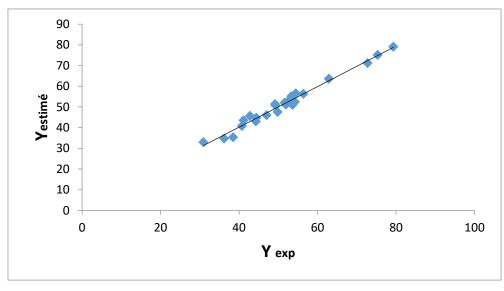|									
b0	42.228	1.106	38.17	< 0.01 ***		b44	0.678	1.872	0.36	72.3
b1	-1.777	0.708	-2.51	2.63 *		b12	-2.979	0.750	-3.97	0.192 **
b2	2.638	0.708	3.73	0.291 **		b13	8.006	0.750	10.67	< 0.01 ***
b3	-7.985	0.708	-11.2	< 0.01 ***		b23	0.778	0.750	1.04	32.2
b4	-0.889	0.708	-1.26	23.2		b14	0.528	0.750	0.70	50.2
b11	11.298	1.872	6.04	< 0.01 ***		b24	0.929	0.750	1.24	23.8
b22	0.223	1.872	0.12	90.3		b34	1.464	0.750	1.95	7.2
b33	0.183	1.872	0.10	92.1						

Figure 1. Estimated Response in the experimental response

The Figure 1 shows that there is a good correlation between the experimental values and the estimated values because they are very close to the straight line.

3.3. Graphical analysis of the results
The experimental field being defined from the variation of two factors. The graphic analysis and more particularly, the response surfaces for studying the behavior responses depending on input parameters.

Figures 2 to 6 represent the response surface of three-dimensional, which was constructed to show the effects of the parameters on the removal efficiency of methylene blue.
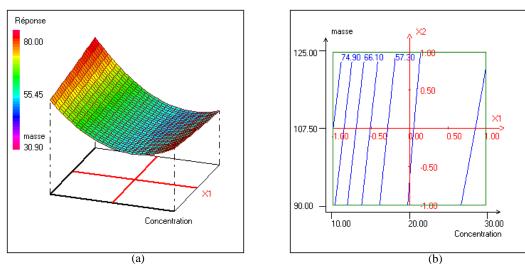

(a)   (b)

Figure 2. Representation of the response in the plan: Concentration vs masse (a:3D et b:2D)

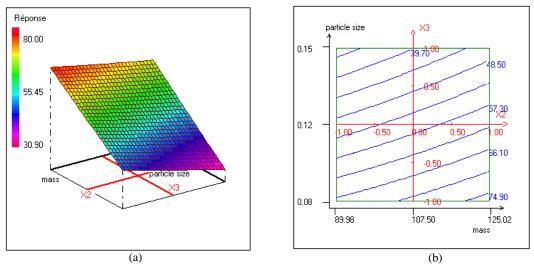(a)   (b)
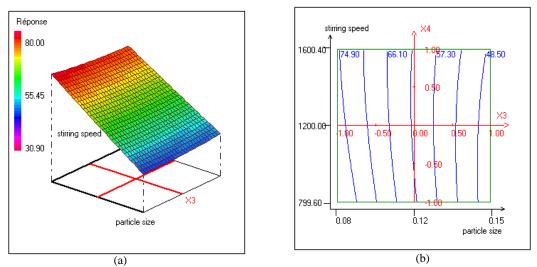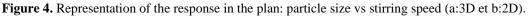
Figure 3. Representation of the response in the plan: Masse vs particle size (a:3D et b:2D)
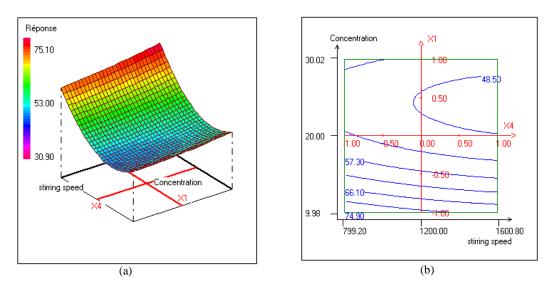

(a)   (b)

Figure 4. Representation of the response in the plan: particle size vs stirring speed (a:3D et b:2D).

(a)   (b)

Figure 5. Representation of the response in the plan: stirring speed vs Concentration

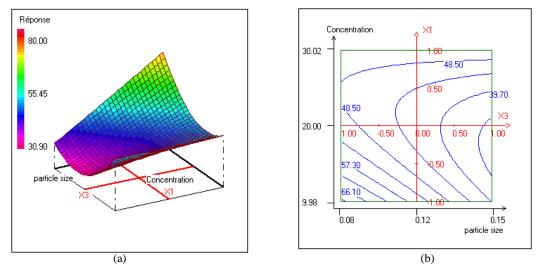(a)   (b)

Figure 6. Representation of the response in the plan: Concentration vs particle size (a:3D et b:2D)

Figures 2-6 represent the response surface of three dimensions which was constructed to show the effects of the parameters on the removal efficiency of methylene blue. Figure 2 shows the effect of mass and concentration of adsorbate on the decolorization efficiency, the influence of these variables on the dye removal efficiency was significant for low values of dye concentration and higher values of the amount of adsorbent. Figure 3 shows the effect of particle size and mass of adsorbent. A decrease in particle size leads to a significant increase in the elimination of the dye, while an increase in the mass of adsorbent gives the same result. The effect of the stirring rate and particle size on the response is shown in Figure 4. The yield was increased with decreasing particle size and increases slightly with low stirring speed. The results illustrated in Figure 5 shows that the concentration has a significant effect on the elimination of methylene blue. The yield increases slightly with decreasing the stirring speed. Figure 6 shows the effect of particle size and concentration of adsorbate. The decolorization efficiency increased with a decrease in the two variables studied.

4. Conclusion

To study the effect of the independent variables: the concentration of dye, stirring speed, mass and particle size of adsorbent on the elimination of methylene blue, we modeled the response as a polynomial function of these parameters. The results show that the application of response surface methodology and the central composite design are used to describe and model a correct manner the influence of these four experimental parameters on the efficacy of treatment. The reliability of the model-based second order was tested by the analysis method (ANOVA). This analysis showed that the model is highly significant and in good adequacy with experimental results [40].

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