

# Optimization of Methylene Blue Adsorption onto Activated Carbon from *Bos Indicus* Gudali Bones Using a Box Behnken Experimental Design

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**Abstract** A phosphoric acid activated adsorbent produced from *Bos Gudali Indicus bones* (BGIB) was used for Methylene Blue adsorption in aqueous solution. Adsorption performances were assessed using a Box Behnken Experimental Design (BBED). Three fundamental parameters were investigated: adsorbent mass, initial dye concentration and temperature while a pH at 5 was considered from passed studied as convenient. The amount of adsorbed Methylene Blue regards the responses was used for modeling and optimization. From obtained result, it appeared that Methylene blue adsorption was significantly affected by singular and quadratic interaction effects of temperature, adsorbent mass and initial dye concentration. Optimal adsorbed dye amount of 15.08 mg.g<sup>-1</sup> was achieved with an initial concentration of 45 mg.L<sup>-1</sup>, a temperature of 45°C, and adsorbent mass of 0.2 g. Statistical analysis were performed using analysis of variances (ANOVA) and R<sup>2</sup> = 98.99 lack of fit adequacy which gave good interpretation of different interactions of experimental parameters, with highly significant with confidential level > 99%. It was then observed that, when adsorbent mass increases, temperature and initial dye concentration decrease considerably. The quadratic effect of adsorbent mass is 5 times greater than adsorbent mass while decreasing when the first one is increasing.

**Keywords** Box-Behnken Experimental Design, Methylene Blue, Adsorption Modeling and Optimization

## 1. Introduction

Many interesting organic molecules were developed and produced by chemical industries all over the 20<sup>th</sup> century. These molecules are used in many fields as medicine, agriculture, chemistry and chemical formulation (ref). Some of these molecules are called "micropollutants" because they present known or suspected harmful effects even at low concentrations (Halling-Sorensen *et al.*, 1998).

However, in recent years, recently, scientific community paid particular attention to micropollutants mainly on their occurrence and their fate in the environment. These molecules include formulation additives, hormones, metabolites, pharmaceutical residues and organic dyes (Deblonde *et al.*, 2011; Murray *et al.*, 2010). Organic dyes

are commonly used in many industrial activities such as dyeing leather, paper, textiles, wood, silk, plastics, as well as in the cosmetic, food and pharmaceutical industries. Among these organic dyes is methylene blue (Domga *et al.*, 2015).

Methylene blue is one of the most widely used dyes for dyeing many substrates, including silk, cotton, and wood, and for temporarily coloring paper ref. It is used as a colored redox indicator, histological dye, antiseptic in the treatment of superficial wounds (Atef, 2009). Maurya *et al* (2008), and Vanessa *et al.* (2017) reported that organic dyes damages human health; some dyes are carcinogenic, mutagenic and teratogenic and can cause several conditions such as nausea, hemorrhoids, ulcers of the skin and mucous membranes, severe irritation of the respiratory system. Madhura *et al* (2014) indicated that wastewater from processes using organic dyes has high BOD and COD. Their elimination calls for appropriate methods (Zidane *et al.*, 2011).

Removal of organic dyes as well as organic or inorganic pollutants from wastewater is necessary. Many water treatment processes have been developed for the reduction

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and elimination of pollutant load from organic dyes in wastewater, including biological, chemical and physical methods. These methods include, among others, flocculation, precipitation, membrane filtration, ion exchange, ozonation, irradiation and adsorption (Kifuani *et al.*, 2004; Hameed *et al.*, 2007; Li *et al.*, 2009; Muswema *et al.*, 2010; Sun *et al.*, 2012; Balogoun *et al.*, 2015; Kassale *et al.*, 2015; Berradi *et al.*, 2016; Ousmaila *et al.*, 2016; Oliveira *et al.*, 2017). Most of these methods are expensive, require a lot of energy, produce huge quantities of sludge or generate derivatives that sometimes are more toxic than the initial products (Masson, 2015; Tchuifon, 2015). Among water polluted (organic dyes) treatment processes, adsorption is found to be the most efficient (Ertaş *et al.*, 2010; Yao and Kouassi, 2015). Activated carbon is one of the most widely used adsorbents (Sark *et al.*, 2015), but the high cost production remains a challenge. However, Several works are being carried for the preparation of low-cost adsorbent using inexpensive precursor, available locally from renewable natural sources, such as agro-industrial waste (Arivoli *et al.*, 2009; Kazlauskienė *et al.*, 2009; Li *et al.*, 2009; Laximi and Ahmazaman, 2010).

To study the adsorption process, several parameters can be followed and the experimentation should be repeated for each parameter. To solve this problem, it is necessary to reduce the number of experiments to be carried out without losing the quality of the results sought. The use of experimental design method gives a strategy in the choice of experimental methods. Their success in research and industry is linked to need of competitiveness, improvement in quality and a low-costs (Domga *et al.*, 2015; Domga *et al.*, 2016; Domga *et al.*, 2018).

There are two approaches for this: The first is a classic intuitive application that consists of setting the level of all but one factor, then repeating the procedure until the last factor (Dejaegher *et al.*, 2011). This method does not allow to study the evolution of the system compared to the variation of a single factor: mathematical model is impossible to adjust.

The second is a multivariate approach which consists in varying several factors simultaneously. This approach is subdivided into sequential and simultaneous approaches (Massart *et al.*, 1997. Vander Heyden *et al.*, 2000). The sequential approach consists of carrying out some initial experiments and uses their results to define experiments (Dejaegher and Vander Heyden, 2009). This approach can be applied when the experimental domain contains an unknown optimum. The simultaneous approach gives a representation of the predefined number of experiments, according to a precise experimental organization as an experimental design (Montgomery, 1997; Dejaegher and Vander Heyden, 2009; Dejaegher *et al.*, 2010).

Experimental design method is introduced mainly to evaluate several factors simultaneously and to predict interactions between factors, hence the choice of modeling designs or response surfaces mainly on the Box-Behnken model. The latter is from the three large families of design

of experiments with the mixing and screening designs (Dejaegher *et al.*, 2011; Cobas *et al.*, 2014). Experimental technical design is a very tool for this purpose, as it provides statistical models which help in understanding the interactions among the parameters that have been optimized. Another important advantage of the use of statistical models in optimization processes is the requirement of very small number of experiments and therefore minimizes time required to carry out the experiments, also minimizes the amount of chemicals, glassware, and manpower needed (Cobas *et al.*, 2011; Dejaegher *et al.*, 2011). In contrast to the classical optimization processes, which did not account the effect of different combination of parameters, response surface methodology (RSM) provides elaborative vision over various combinations of parameters. RSM is essentially a particular set of mathematical and statistical methods for designing experiments, building models, evaluating the effects of variables, and searching optimum conditions of variables to predict targeted responses (Myers and Montgomery, 2002). RSM is an important tool to develop novel processes, optimizing their performance and improving their design and formulation of new products. RSM shows promising results in industrial research, especially where large number of variables influences the system (Myers and Montgomery, 2002). The adsorption capacity of the adsorbent was assessed using a Box-Behnken Experimental Design (BBED).

The Box-Behnken (BB) model therefore has the advantage of being less costly in terms of time (reduced number of tests) and resources to invest in experimentation. The BB model does not allow to test treatments for which all the factors are placed simultaneously at their high or low level. Box-Behnken matrix therefore minimizes the combinations located at the ends of the variation domain of the factors, where generally an unsatisfactory response (high or low response) is observed. This can be advantageous when points on the corners of the cube represent combinations of factor levels that are expensive or impossible to test due to physical constraints of the process (Nosrati *et al.*, 2011; Cobas *et al.*, 2011; Yazici and Deveci, 2013; Yazici and Deveci, 2013; Cobas *et al.*, 2014).

The present work interest is to develop a potential adsorbent for the adsorption of organic dyes using waste cattle's (*Bos Indicus* Gudali) bones from the slaughterhouse of Dang (Ngaoundéré 3 sub-division) in Adamawa region. Almost 40% of these bones are released as waste into environment. Converting these bones into a potential adsorbent thus constitutes a way of recovering waste and cleaning up the environment.

The adsorption performances of *Gudali* species (*Bos Indicus*) activated carbon bones was evaluated using an aqueous solution of methylene blue (BM). Several parameters influence this adsorption process, hence the need an experimental design. Hence, the aim of the present study was to optimize the adsorption of Methylene Blue using Response Surface Method: Box Behnken experimental design.

## 2. Material and Methods

### 2.1. Preparation of Activated Carbon

Zebu cattle bones are degreased *Gudali* collected and thoroughly washed with distilled water and dried in an oven at 110°C for 24h, 200g *Gudali* bone were dried, milled and sieved (0.2-5mm) for 18 h and impregnated with a solution of phosphoric acid H<sub>3</sub>PO<sub>4</sub> 85%. Impregnated bones were washed with hot distilled water until neutral pH and dried at 110°C for 24h. Samples were then pyrolyzed during 2 h at 500°C. Pyrolysis was conducted in a muffle furnace Heraeus, MR model 260E with a heating rate of 5°C. min<sup>-1</sup>.

### 2.2. Preparation of Methylene Blue Solutions

Methylene blue (MB) is a cationic dye with a molecular formula C<sub>16</sub>H<sub>18</sub>N<sub>3</sub>SCl (Figure 1) was purchased without any treatment from Merck. The dye stock solution was prepared by dissolving accurately weighted MB in distilled water. Experimental solutions were obtained by diluting MB in accurate proportions to required initial concentrations.

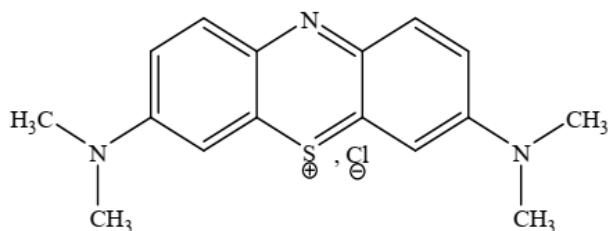


Figure 1. Methylene blue molecule

### 2.3. Determination of MB Concentration

UV-visible spectrum of MB absorption was obtained by a spectral scan from 300 to 1100 nm, of a 10 ppm solution (Figure 2). This spectrum shows that the peak of absorption of is 664 nm. The concentrations of MB solution are obtained by measuring the absorbance at 664.5 nm by a spectrophotometer “Pharo 100”.

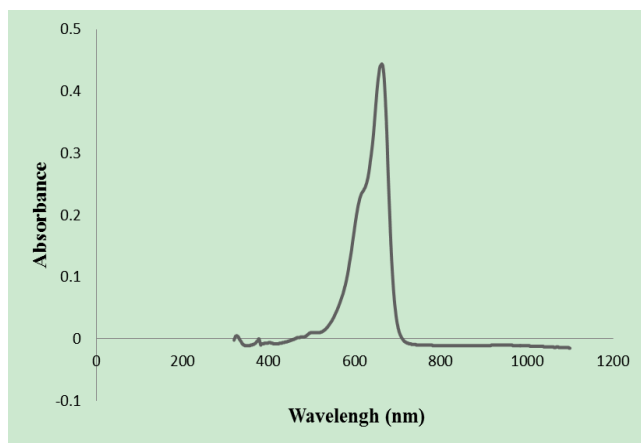


Figure 2. Absorption spectrum of methylene blue

### 2.4. Batch Equilibrium Adsorption Studies

Adsorption studies were carried out at room temperature

in a 250 mL reactor. For each experiment, dose of 0.2-0.6 g of adsorbent was weighed and put in reactors containing 50 mL of concentration ranging from 10-70 ppm, pH increasing from 3 to 11 (Domga *et al.*, 2015).

The mixture was subjected to magnetic stirring over the interval of time between 10 to 60 minutes, using a magnetic stirrer. The suspension was filtered and the residual concentration was determined by UV-visible spectrophotometry. Concentration of remaining MB in the solution was calculated by the difference of the initial and the final BM concentrations. The research work of Domga *et al* (2015) allows us to fix the most influential factors in this work.

Dye removal efficiency was expressed as:

$$q_t = (C_o - C_f) \frac{V}{m} \quad (1)$$

C<sub>o</sub> and C<sub>f</sub> are respectively initial and final concentration of dye (mg·L<sup>-1</sup>), m is adsorbent mass and V de volume of the solution.

### 2.5. Response Surface Methodology (RSM)

A 3-factor 3-level factorial Box–Behnken experimental design (BBED) matrix was used to investigate the effects of selected parameters. BB designs are response surface methods (RSM), specially made to require only 3 levels, coded as -1, 0, and +1. BB designs are formed by combining two-level factorial designs with incomplete block designs that the geometry of this design suggests a sphere within the process space such that the surface of the sphere protrudes through each face with the surface of the sphere tangential to the midpoint of each edge of the space (Myers and Montgomery, 2002). This procedure creates designs with desirable statistical properties but, most importantly, with only a fraction of the experiments required for a three-level factorial (3<sup>k-p</sup>) designs. Since there are only three levels, the quadratic model is appropriate. Observations were fitted to a second order polynomial model as given below:

$$Y = a_0 + \sum a_i X_i + \sum a_{ij} X_i X_j + \sum a_{ii} X_i^2 \quad (2)$$

Table 1. Factors and levels of the Box-Behnken design

Variables	Factors	Level		
		Low	Middle	High
X <sub>1</sub>	Mass (g)	0.20	0.40	0.60
X <sub>2</sub>	Concentration (mg/L)	20.00	45.00	70.00
X <sub>3</sub>	Temperature (°C)	30.00	45.00	60.00
Response				
Y	Adsorbed Amount (mg/g)			

Where Y is the predicted response associated with each factor level combination, a<sub>0</sub> is the intercept term, a<sub>i</sub> is linear effect, a<sub>ii</sub> is the quadratic effect, and a<sub>ij</sub> is the 2-way linear by linear interaction effect. Additionally, X<sub>i</sub> and X<sub>j</sub> represent the real values of independent variables. Important chosen variables for the BBED are temperature,

mass and initial concentration of methylene blue designated as  $X_1$ ,  $X_2$  and  $X_3$ , respectively. The dependent variable ( $Y_i$ ) is the amount of adsorbed dye per gram of adsorbent ( $q$ ). The range of experimental design matrix is shown in Table 1.

## 2.6. Statistical Analysis

Regression analysis, statistical significance and response surfaces were obtained to find the most suitable combination of factors resulting in optimal Methylene Blue adsorption capacity of *BGI*. Statistical significance of variables was evaluated using Analysis of Variance (ANOVA). Constructed model adequacy was investigated via lack of fit, coefficient of determination ( $R^2$ ) and F-values. Statgraphics and sigma plot software packages were used for the regression, residual and graphical analyses.

## 3. Results and Discussion

### 3.1. Textural Characteristics of the Adsorbent

Table 2 shows the iodine value of 666.251 mg/g. This high value of iodine index makes it possible to conclude that adsorbent has micropores on its surface available to fix polluting micromolecules (Domga *et al.*, 2016).

Adsorbent specific surface (Table 2) shows a significant exchange surfaces reaching 1045 m<sup>2</sup>/g. This high value of the specific surface makes it possible to assume that this adsorbent complies with standards of adsorbents specific surface and it justifies good adsorbent capacity (Domga *et al.*, 2018).

**Table 2.** Adsorbent characteristics

Parameters	Values
Sieved (mm)	0.2-5.0
pH <sub>ZPC</sub>	6.84
Residual moisture (%)	1.50
Ash content (%)	17.63
Iodine Index (mg/g)	666.25
$S_{BM}$ (m <sup>2</sup> /g)	1045.00

### 3.2. Development of Regression Model Equation for Methylene Blue Removal

The complete design matrix and values of both responses obtained from the experimental works are given in Table 3. BBED is a modeling technique used to evaluate correlation between experimental variables for MB adsorption and corresponding response of adsorbed dye quantity from aqueous solution. The second order polynomial equation fitted between responses represents adsorbed quantities ( $Y$ ) and input variable of mass ( $X_1$ ), initial concentration of dye ( $X_2$ ) and temperature ( $X_3$ ). Empirical model for adsorption MB is given in Eq 3:

$$Y = 5.43472 - 3.31491X_1 + 3.2113X_2 + 0.298774X_3 + 1.68547X_1X_1 - 1.47902X_1X_2 - 0.0590403X_1X_3 - 0.357626X_2X_2 + 0.0722022X_2X_3 - 0.219239X_3X_3(3)$$

Equation 3 has been used to observe the effects of experimental factors on removal efficiency. Effect of mass of adsorbent is negative and greater than the effect of temperature and initial dye concentration. However, quadratic effect of mass is 5 times greater than mass and is positive. Experimental and predicted values of adsorbed amount of MB are given in Table 3. Experimental values are measured for a particular run, predicted values are evaluated from the model and are generated by using the approximating functions. Predicted  $R^2$  of 98.99% is reasonable.

**Table 3.** Experimental and predicted values of the BBED matrix for methylene blue uptake

Run	$X_1$	$X_2$	$X_3$	Adsorbed amount (mg/g)		
				Experiment	Predicted	Residue
1	0.2	45	30	10.07	9.86	-0.21
2	0.6	45	30	3.275	3.35	0.07
3	0.4	20	30	1.80	1.42	-0.43
4	0.4	70	30	7.178	7.70	-0.52
5	0.2	70	45	15.08	14.77	-0.301
6	0.2	20	45	4.79	5.39	0.59
7	0.6	20	45	1.406	1.72	0.31
8	0.6	70	45	5.77	5.18	-0.59
9	0.4	45	45	5.61	5.43	-0.17
10	0.4	45	45	5.17	5.43	0.27
11	0.4	45	45	5.53	5.43	0.09
12	0.4	70	60	8.06	8.44	0.38
13	0.4	20	60	2.40	1.87	-0.52
14	0.2	45	60	10.65	10.57	-0.07
15	0.6	45	60	3.61	3.83	0.21

### 3.3. Analysis of Variance (ANOVA) BBED for Methylene Blue Uptake

ANOVA of MB removal obtained in the study is presented in the Table 4. In general terms, ANOVA explains any variation in the statistically derived model and significance of the model parameters. Degrees of Freedom (DF), Sum of Squares (SS), quadratic mean standard deviation probability (P) and F ratio are calculated and tabulated in the ANOVA table. Normally, a lower P value of a model term in ANOVA indicates good significance of the term over others. In this study, the models obtained for MB dye removal were able to explain experimental results on color removal. Regression model, linear model and square model well fitted with 100% confidential level. The interaction of the parameter temperature, adsorbent mass and initial concentration of MB adsorption is also highly significant with confidential level > 95%. Figure 3 shows predicted versus actual plots for adsorbed amount,

demonstrating that the values are distributed relatively near to the straight line. This indicates that models are adequate for predicting color removal efficiency within the range of studied variables. Test of lack of adequacy is used to determine if selected model can be used to describe experimental data or if another complicated one should be chosen. If probability for lack of fit is bigger than 0.05 there is evidence that quadratic linear regression model is more appropriate to explain parameters relationship on Methylene Blue adsorption. In the present study, probability of lack of fit is lower than that of pure errors sum of square and quadratic mean.

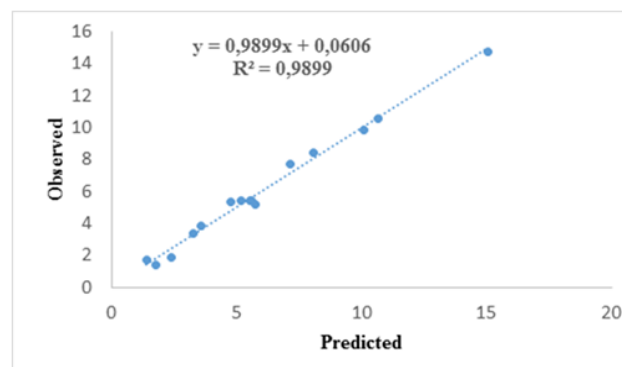
**Table 4.** ANOVA for MB removal

Run	Sum of Square	Quadratic Mean	Degree of Freedom	Proba.	F ratio
X <sub>1</sub>	87.91	87.91	1.00	0.00	225.64
X <sub>2</sub>	82.50	82.50	1.00	0.00	211.75
X <sub>3</sub>	0.71	0.71	1.00	0.24	1.83
X <sub>1</sub> X <sub>1</sub>	10.50	10.50	1.00	0.00	26.92
X <sub>1</sub> X <sub>2</sub>	8.75	8.75	1.00	0.01	22.46
X <sub>1</sub> X <sub>3</sub>	0.01	0.014	1.00	0.86	0.04
X <sub>2</sub> X <sub>2</sub>	0.47	0.47	1.00	0.32	1.21
X <sub>2</sub> X <sub>3</sub>	0.02	0.02	1.00	0.83	0.05
X <sub>3</sub> X <sub>3</sub>	0.18	0.18	1.00	0.53	0.46
<b>Pure Error</b>	1.95	0.34	5.00		
<b>Total (corr.)</b>	193.60		14.00		

The ANOVA table breaks down the variability in adsorbed amount into separate rows for each effect. It then tests the statistical significance of each of the effects by comparing the root mean square against an estimate experimental error. In this case, 4 effects have probabilities less than 0.05, indicating that they are significantly different from zero at the 95.00% confidence level.

The R<sup>2</sup> statistic indicates that fitted model explains 98.99% of the variability in adsorbed amount. The adjusted R<sup>2</sup> statistic, which is preferable for comparing models with

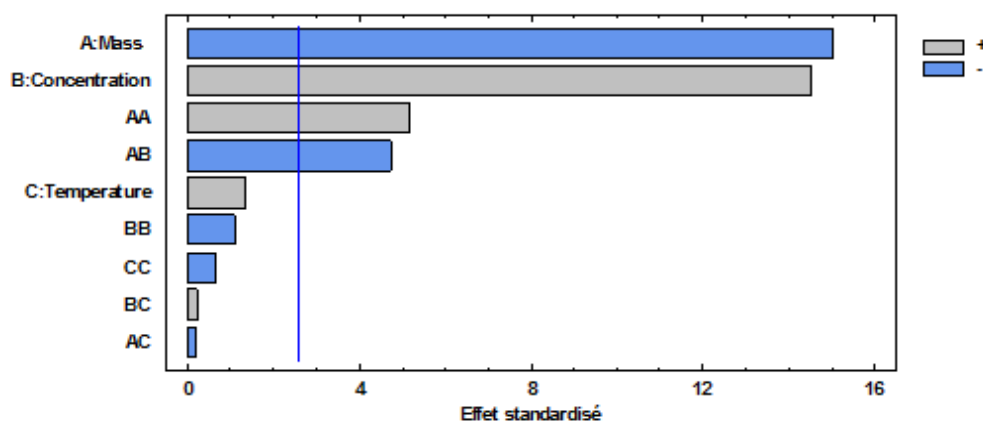
different numbers of explanatory variables, is 97.18%. The standard error of the estimate indicates that standard deviation of residuals is 0.624181. The mean absolute error (MAE) of 0.32 is the mean value of the residuals. The Durbin-Watson (DW) statistic tests the residuals to determine if there is a significant correlation based on the order in which they appear in data file. Since the probability value is greater than 5.00%, this does not indicate the serial autocorrelation of the residuals at the 5.00% significance level.



**Figure 3.** Experimental vs. Predicted of Methylene Blue adsorbed

### 3.4. Graphic Representation of Effects a. Pareto Chart

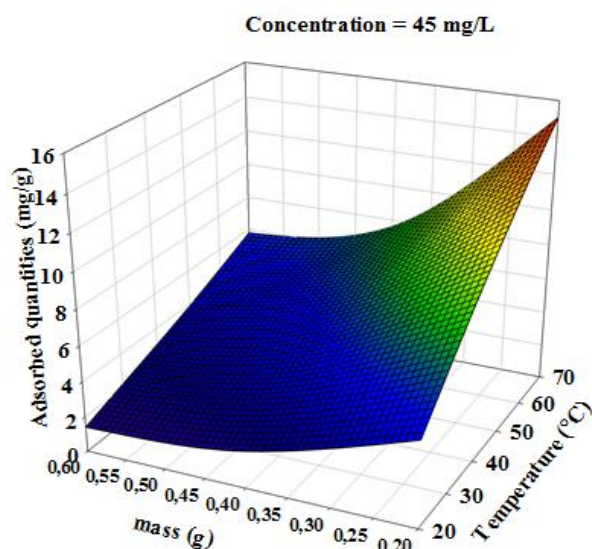
This diagram (figure 4) makes it possible to extract the most important parameters. Among all the factors studied and at chosen confidence level ( $\alpha = 0.05$ ), concentration (B) appears to be the very influential factor. The temperature (C) can according to this same diagram have an effect on the answer. Mass (A) has no significant effect on response. The positive sign of each of the factors (B, C) indicates that the maximum value of these factors results in a higher response than with the minimum value. The factor (A) has a negative sign indicating that the minimum value of this factor results in a higher response. The quadratic factors have respectively a significant (AA) and insignificant (B, C) effect on the response, while the quadratic effects (AB) influence the response less. The quadratic effects (BB, CC, AC) influence the response less and less.



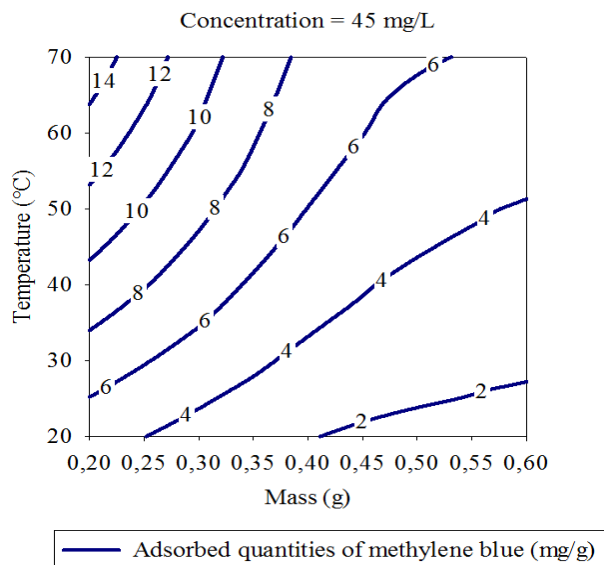
**Figure 4.** Pareto plot of normalized effects

Equation model has been simplified, and the ranking of factors is done according to the Pareto chart (Figure 4). Furthermore, examination of responses diagram estimated as a function of measured responses (Figure 3) shows a good coefficient of determination  $R^2 = 98.99\%$ . This means that there is a good correlation between theoretical and measured responses. This correlation is confirmed by the value of adjusted R squared ( $R^2_{aju} = 97.18$ ).

### 3.5. Estimation of Quantitative Effects of the Factors



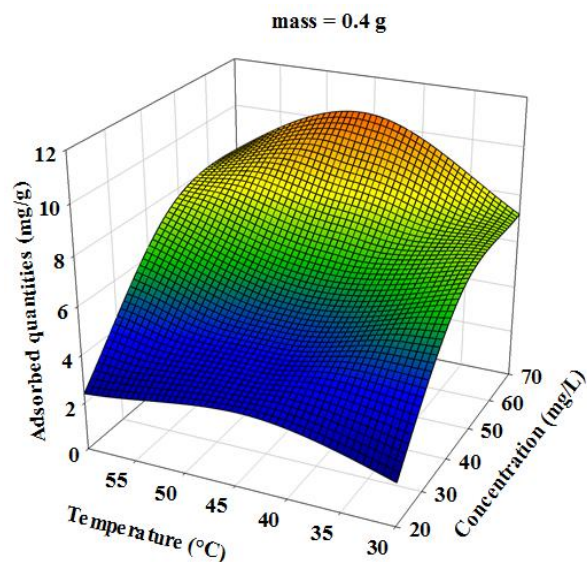
**Figure 5.** Response surfaces of the effect of temperature and adsorbent dosage, on the adsorption of MB at constant initial concentration of dye ( $45 \text{ mg/L}^{-1}$ ) and pH (5)



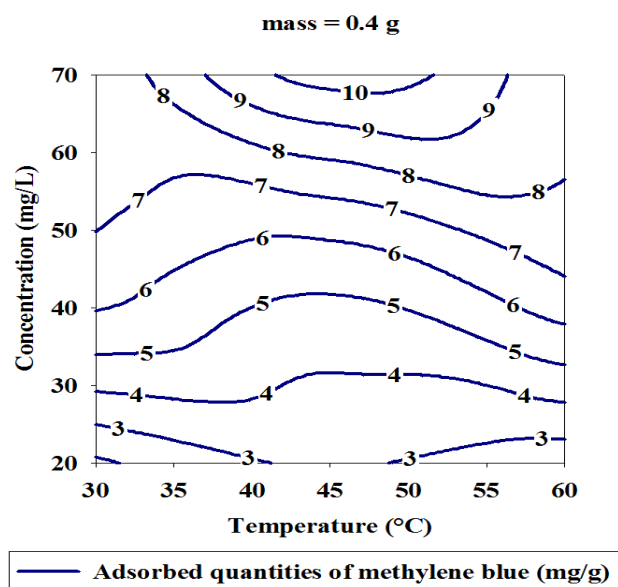
**Figure 6.** Iso-response curves of the effect of temperature and adsorbent dosage, on the adsorption of MB at constant initial concentration of dye ( $45 \text{ mg/L}^{-1}$ ) and pH (5)

Three dimensional surface responses obtained from second polynomial equations are shown in figures ranging from 5 to 10. Optimum values of variables were obtained as maximized response using RSM. The best response range is

obtained by analyzing response surface plots and iso-responses curves. Combined effect of temperature, adsorbent mass, at constant initial dye concentration ( $45 \text{ mg/L}^{-1}$ ) and pH (5), for MB adsorption from aqueous solution are made. Adsorbed quantity increased with increasing temperature and initial dye concentration. This is due to the increase in number of binding sites on adsorbent surface. It is clear from figures 5-10 that adsorbed amount decreases with increase of adsorbent mass from 0.2 to 0.6 g. It is also noted, that maximum adsorption at all temperature takes place at 0.20g. Maximum adsorbed amount ( $15.08 \text{ mg/g}^{-1}$ ) was observed at constant temperature ( $45^\circ\text{C}$ ), initial dye concentration ( $45 \text{ mg/L}^{-1}$ ) and pH (5).

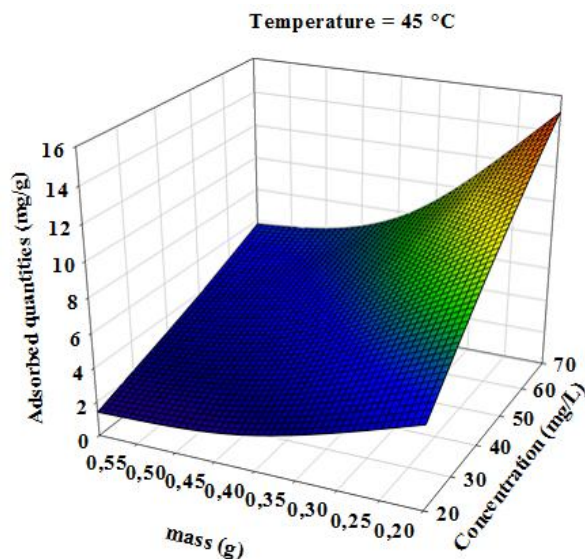


**Figure 7.** Response area of the effect of temperature and initial concentration, on the adsorption of Methylene Blue at constant adsorbent dosage (0.4 g) and pH (5)

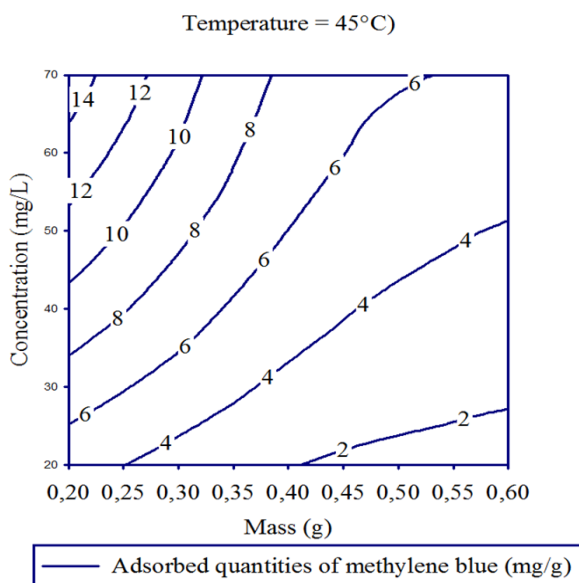


**Figure 8.** Iso-response curves of the effect of temperature and initial concentration, on the adsorption of Methylene Blue at constant adsorbent dosage (0.4 g) and pH (5)





**Figure 9.** Response surfaces of the effect of adsorbent dosage and initial concentration, on the adsorption of Methylene Blue at constant temperature (45.0°C) and pH (5)



**Figure 10.** Iso-response curves for the effect of adsorbent dosage and initial concentration, on the adsorption of Methylene Blue at constant adsorbent dosage (0.4 g) and pH (5)

Figure 7 shows three-dimensional response surface for the interaction effect of temperature and initial dye concentration on adsorption of MB at constant adsorbent dosage (0.4 g) and pH (5). Increasing temperature from 30 to 60°C facilitated the removal efficiency of MB, probably due to the increase in the mobility of large dye ions. Increase in amount of dye sorption increases with temperature also due to either higher affinity of sites for dye or an increase in number of binding sites on adsorbent. Enhancement of adsorption capacity of MB at higher temperature may be the possibility of an increase in the porosity and in total pore volume of the adsorbent (Dogan and Alkan, 2003). Interactive effect of adsorbent mass and concentration of the solution on the removal of MB at constant pH (5) and

temperature (45°C) is shown in Figure 9. It is clear from figure 9 that the adsorption of MB is efficient at initial dye concentration of 45 mg·L<sup>-1</sup> for 0.2g of adsorbent mass.

## 4. Conclusions

The adsorption efficiency of *Bos Gudali Inducus* was studied using Methylene Blue dye solution. Adsorption experimental process parameters namely temperature, initial concentration and adsorbent mass were considered to optimize the removal of Methylene Blue using response surface methodology. Experimental design approach consists of organizing the experimental approach and decision analysis by applying several tools and statistical tests. The use of experimental designs leads to establish a design with maximum precision with minimum experiments. In this work, we first used as preliminary step Domga *et al* (2015) work, in order to select most influencing factors. Among studied factors, temperature, initial dye concentration and adsorbent mass appeared to be the most influential parameters on the amount of methylene blue adsorbed on *Gudali* bones activated carbon. These factors were then examined by the response surface methodology using the Box-Behnken design. Adsorption experimental process parameters like temperature, initial concentration and adsorbent mass were optimized using RSM for removal of Methylene Blue. Furthermore, interactions among all variables were investigated. Regression analysis, statistical significance and response surface well predicted responses in experimental regions. Multiple correlation coefficient of determination  $R^2$  was 98.99%, showing that experimental data fitted well with the predicted data. The lack of adequacy test showed that this model fit very well experimental data. To study the effect of independent variables, we modeled the response as a polynomial based on these parameters. Obtained model is significant  $P = 0.02$  ( $<0.05$ ). There is a satisfactory correlation between measured values and adjusted values,  $R^2 = 98.99\%$  and  $R^2_{\text{ajus}} = 97.18$ . Optimal adsorption of Methylene Blue using *Bos Gudali Indicus* activated carbon at temperature, initial concentration and adsorbent mass were found to be 45°C, 45 mg·L<sup>-1</sup> and 0.2 g respectively with 15.08 mg·g<sup>-1</sup> as dye adsorbed amount. This results shows *Bos Gudali Inducus* is better adsorbent for Methylene Blue adsorption from aqueous solution than many others adsorbents including some activated carbons and biosorbents.

## REFERENCES

- [1] Arivoli S, Nandhakumar V, Saravanan S, Sulochana N. 2009. Adsorption dynamics of copper ion by low cost activated carbon. *The Arabian Journal for Science and Engineering*, 34(1A): 112. PMID: 40064143.
- [2] Atef SA. 2009. Adsorption of methylene blue from aqueous

- solution onto a low-cost natural Jordan Tripoli. *Amer. J. Environ. Sci.*, 5(3): 197-208. DOI: <http://www.doi.org/10.3844/ajessp.2>.
- [3] Balogoun C. K, Bawa ML, Osseni S, Aina M. 2015. Préparation des charbons actifs par voie chimique à l'aide de l'acide phosphorique à base de coque de noix de coco. *Int. J. Biol. Chem. Sci.*, 9(1): 563580. DOI: <http://dx.doi.org/10.4314/ijbcs.v9i1.48>.
  - [4] Berradi M, Essamri A, Al Harfi A. 2016. Discoloration of water loaded with vat dyes by membrane process of ultrafiltration. *J. Mater. Sci.*, 7(4): 10981106.
  - [5] Cobas M., Sanromán M. A., Pazos M., *Bioresource Technol.* 2014. 160, 166–174.
  - [6] Deblonde, T., Cossu-Leguille, C., Hartemann, P. 2011. Emerging pollutants in wastewater: A review of the literature. *Int. J. Hyg. Environ. Health, The second European PhD students workshop: Water and health.* Cannes 2010 214, 442–448. <https://doi.org/10.1016/j.ijheh.2011.08.002>.
  - [7] Dejaegher B., Vander Heyden Y., titre de l'article *Phar J. Biome. Anal.* 2011. 56, 141–158.
  - [8] Dejaegher B., Vander Heyden Y., titre de l'article *Comprehensive Chemometrics.* 2009. (Chapitre 17), 547–575.
  - [9] Dejaegher B., Vander Heyden Y., titre de l'article *Acta Chromatogr.* 2009. 21, 161–201.
  - [10] Dejaegher B., Durand A., Y. Vander Heyden Y., *Chemometric Methods in Capillary Electrophoresis.* 2010. (Chapitre 2), Editeur 11–74.
  - [11] Dogan, M. And M. Alkan. 2003. Adsorption kinetics of methyl violet onto perlite. *Chemosphere*, Editeur 50(4), 517-528.
  - [12] Domga R., Tcheka C., Anombogo G. A. M., Kobbe Dama N., Domga, Tchatchueng J. B., Tchigo A., Tsafam A. 2016. Batch equilibrium adsorption of methyl orange from aqueous solution using animal activated carbon from gudali bones. *International Journal of Innovation Sciences and Research*, vol 5, iss7, pp.798-805.
  - [13] Domga R., DomgaTchatchueng, J. B., Nyounai F., Gaineunbo S. F., F. Fabane F., Béné M., Assokeng T., Tsafam A., NÔN B. E. 2018. Synthesis of animal activated carbon from gudali (bos indicus) bones and its adsorption for chromium (VI) in aqueous solution. *International Journal of Engineering Research and Science and Technology*, vol 7, iss 1, pp. 1-15.
  - [14] Ertaş M, Acemioğlu B, Hakki Alma M, Usta M. 2010. Removal of methylene blue from aqueous solution using cotton stalk, cotton waste and cotton dust. *J. Hazard. Mater.*, 183(1-3): 421-427. DOI: <http://www.doi.org/10.1016/j.jhazmat.2010.07.041>.
  - [15] Goupy J. 2016. Modélisation par les plans d'expériences, *Techniques de l'ingénieur, mesures et contrôles.* R 275, 1-23
  - [16] Halling-Sorensen, B., Nielsen, S., Lanzky, P., Ingerslev, F., Lutzhoft, H., Jorgensen, S. 1998. Occurrence, fate and effects of pharmaceutical substances in the environment - A review. *Chemosphere* 36, 357–394. [https://doi.org/10.1016/S0045-6535\(97\)00354-8](https://doi.org/10.1016/S0045-6535(97)00354-8).
  - [17] Kassale A, Barouni K, Bazzouai M, Albourine A. 2015. Kinetics and Modeling of the adsorption of methylene blue by the grafted cotton. *J. Chem. Bio. Phy. Sci.*, 5(2): 1205-1216.
  - [18] Kifuani KMA, Noki VP, Ndelo DPJ, Mukana WMD, Ekoko BG, Ilinga LB, Mukinayi MJ. 2012. Adsorption de la quinine bichlorhydrate sur un charbon actif peu coûteux à base de la Bagasse de canne à sucre imprégnée de l'acide phosphorique. *Int. J. Biol. Chem. Sci.*, 6(3): 1337-1359. DOI: <http://www.doi.org/10.4314/ijbcs.v6i3.36>.
  - [19] Kifuani KMA. 2013. Adsorption des composés organiques aromatiques sur charbon actif à base des déchets agroindustriels. *Thèse de Doctorat, Université de Kinshasa*, 265 p.
  - [20] Laximi GS, Ahmazman MD. 2010. Adsorption technique for the removal of phenolic compounds from wastewater using low-cost natural adsorbents. *Assam University Journal of Science and Technology*, 5(2): 156-166.
  - [21] Li K, Zheng Z, Huang X, Zhao G, Feng J, Zhang J. 2009. Equilibrium, kinetic and thermodynamic studies on the adsorption of 2-nitroaniline onto activated carbon prepared from cotton stalk fiber. *J. Hazard. Mater.*, 166(1): 213-220. DOI: <http://www.doi.org/10.1016/j.jhazmat.2008.11.007>.
  - [22] Masson S, Reinert, Guittonneau S, Duclaux L. 2015. Cinétiques et isothermes d'adsorption de micropolluants sur un tissu et un feutre de carbone activé. *Revue des Sciences de l'Eau*, 28(3): 207-213. DOI: <http://www.doi.org/10.7202/1034009ar>.
  - [23] Madhura C, Priyanka S, Charmi N, Sunil K, Sonali D. 2014. Removal of dye by adsorption on various adsorbents: A review. *Inter. J. Sci. Eng. And Technol. Res.*, 3(4):835-840.
  - [24] Masson S, Reinert L, Guittonneau S, Duclaux L. 2015. Cinétique et isothermes d'adsorption de micropolluants sur un tissu et un feutre de carbone activé, *Rev. Sci. de l'Eau*, 28(3): 179-247. DOI: <http://www.doi.org/10.7202/1034009ar>.
  - [25] Maurya MS, Mittal AK, Correl P. 2008. Evaluation of adsorption potential of adsorbents: A case of uptake of cationic dyes. *J. Envir. Biol.*, 29(1): 31-36.
  - [26] Massart D. L., Vandeginste B. G. M., Buydens L. M. C., De Jong S., P. J. Lewi P. J., Smeyers-Verbeke J., *Handbook of Chemometrics and Qualimetrics, Part A*, Elsevier, Amsterdam, 1997.
  - [27] Montgomery D. C., *Design and Analysis of Experiments*, 4<sup>th</sup> edition, John Wiley, New York, 1997.
  - [28] Myers R.H. Montgomery D.C. 2002. Response Surface Methodology: Process and Product Optimization Using Designed Experiments, 2<sup>nd</sup> ed., John Wiley and Sons, USA.
  - [29] Muswema JL, Ekoko BG, Lobo KK, Ilinga L, Monama O, Mvele OM, Kifuani KM. 2010. Photodégradation de l'Acide Rouge G et l'Acide Orange 7 en milieu basique en présence d'une suspension de TiO2 seule ou couplée à Ag<sup>+</sup>. *Rev. Cong. Sci. Nucl.*, 24: 1-9.
  - [30] Murray, K.E., Thomas, S.M., Bodour, A.A. 2010. Prioritizing research for trace pollutants and emerging contaminants in the freshwater environment. *Environ. Pollut.* 158, 3462–3471. <https://doi.org/10.1016/j.envpol.2010.08.009>.



- [31] Nosrati S., N. S. Jayakumar N. S., Hashim M. A., *Desalination*. 2011, 266, 286–290.
- [32] Oliveira P, Coufort-Saudejaud C, Alliet M, Frances C. 2017. Procédé de traitement des eaux usées par boues activées: lien entre les propriétés morphologiques des floes et l'âge de boue. *Revue des Sciences de l'Eau*, 30(1): 29-32. DOI: <http://www.doi.10.7202/1040060ar>.
- [33] Sark F, Sennaoui A, Elouardi M, Tamimi M, Assabbane A. 2015. Etude de l'adsorption du bleu de méthylène sur un biomatériau à base de Cactus. *J. Mater. Environ. Sci.*, 6(2): 397-406.
- [34] Tchuifon TDR, Nche G, Tchouanyo DH J, Ngakou SC, Djoumbissie AL, Anagho SG, Kamgaing T, Ketcha JM. 2015. Adsorption studies of methylene blue on methanol modified and unmodified corn stalks and egussi peeling. *Inter. J. Engin. and Techn. Res. (IJETR)*, 3: 8-14.
- [35] Vander Heyden Y., C. Perrin C., Massart D.L., *Handbook of Analytical Separations, K. Edition Valkó*, 2000, 163–212.
- [36] Vanessa P, Andrin A, Le Behec M, Lacombe S, Frayret J, Pigot T. 2017. Couplage photocatalyse-oxydation par le ferrate (VI) pour le traitement du colorant rhodamine 6G. *Revue des Sciences de l'Eau*, 30(1): 35–39. DOI: <http://www.doi.10.7202/1040061ar>.
- [37] Yao MK, Kouassi NL-K. 2015. Etude des propriétés d'adsorption et de désorption du Pb et Cd par les sédiments d'une lagune tropicale en présence d'Allythiourée. *Int. J. Biol. Chem. Sci.*, 9(1): 483-491. DOI: <http://www.doi.10.4314/ijbcs.v9i1.41>.
- [38] Yazici E.Y., Deveci H., *Hydrometallurgy*. 2013, 139, 30–38.
- [39] Zidane F, Ohazzar A, Blais JF, Ayoubi K, Bensaid J, Basri SE, Kaba N, Fakhreddine Q, Lekheif B. 2011. Contribution à la dépollution des eaux usées de textile par électrocoagulation et par adsorption sur des composés à base du fer et d'aluminium. *Int. J. Biol. Chem. Sci.*, 9(1): 1727-1745. DOI: <http://www.doi.10.4314/ijbcs.v5i4.35>.