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REMOVAL OF METHYLENE BLUE FROM AQUEOUS SOLUTIONS USING ALUM SLUDGE: SORPTION OPTIMIZATION BY RESPONSE SURFACE METHODOLOGY

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ABSTRACT

A sorption process on low cost sorbents is one the promising methods for removing of these pollutants from aqueous solutions. This report describes use of drinking water treatment sludge which is abundantly available from drinking water treatment plants, to remove methylene blue (MB) dye from aqueous solutions. The main objective of this work is to determine the optimum conditions for removal of MB dye, a common compound that is used as a model for organic chemicals. Response surface methodology (RSM) is used in the optimization of the sorption process. Six independent important factors which are temperature of treatment, pH of solution, dosage of sorbent, initial dye concentration, contact period and temperature of sorption were investigated using batch sorption technique with a 26 half factorial faced centered central composite design.

Keywords: drinking water sludge; methylene blue; response surface methodology.

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1. INTRODUCTION

Industrial effluents which contain various organic and inorganic pollutants contribute mainly to environmental pollution problem [1]. Synthetic dye is one of the organic chemicals which is widely used in various industries such as textile, cosmetics, pharmaceutical [2], paper, printing and food industries [3]. Consequently, the discharge of the synthetic dyes into

hydrosphere cause significant pollution [4]. Consuming water contaminated by synthetic dye such methylene blue (MB) will seriously affect human cardiovascular, gastrointestinal, central nervous system [5-6] hematologic, dermatologic and genitourinary. Hence, the treatment of dye effluents before discharged to the water bodies is highly important [5, 7].

Adsorption process is the most widely method used to remove dyes [8-9] due to its ease of application and cost effectiveness [10]. In addition, the adsorption process has no side product and the adsorbent can be regenerated and reused as well [6]. The cost-effective waste materials that have been employed as dye adsorbents including Ephedra *strobilacea* sawdust, wastewater sludge [3] and sewage sludge [11]. The removal of MB dye onto various adsorbents were due to the formation of physical and chemical bonds including van der Waals forces, hydrogen bonding, and hydrophobic interactions [10] between adsorbent and MB molecules.

This report describes the results of our study on the potential of drinking water treatment sludge which is abundantly available from drinking water treatment plants, to remove MB from aqueous solutions. Six independent important factors which are temperature of treatment, pH of solution, dosage of sorbent, initial dye concentration, contact period and temperature of sorption were investigated using batch sorption technique with a 2^6 half factorial faced centred central composite design. A 2^6 half factorial faced centred central composite design was employed to investigate the effects of temperature of treatment, pH of solution, dosage of sorbent, initial dye concentration, contact period and temperature of sorption at laboratory scale. In order to determine the optimum conditions of the sorption process, response surface methodology (RSM) was applied.

2. METHODOLOGY

2.1. Preparation of Sorbent

The sorbents were prepared from a fresh drinking water treatment sludge cake which was obtained from a local waste water treatment plant. The dried alum sludge (labelled as T100) was obtained by drying the collected sludge in an oven at 105^{o}C for an overnight. The thermally treated sludge sorbents were prepared by heating the dried sludge in a furnace at a heating temperature of 450 (labelled as T450) or 800 (labelled as T800) ^{o}C for 7 hr. The heated sludge samples were cooled to room temperature, sieved to obtain particle sizes of 100 to 150 µm and stored in polyethylene bottles with screwed caps.

2.2. Preparation of Methylene Blue Solutions (Sorbates)

Methylene blue (MB) dye supplied by Sigma Aldrich (United Kingdom) was used as a sorbate. A stock solution of MB dye solution (1000 mg/L) was prepared by dissolving 0.5 g of the dye powder into distilled water in a 500 mL of volumetric flask. Then, the stock solution was diluted with distilled water to obtain the desired MB dye concentrations (Table 1) to prepare various MB working solutions. The initial pHs of the solutions were adjusted to the desired values by adding 0.1 M NaOH or 0.2 M HCl solutions.

2.3. Experimental Design

Six process parameters, i.e. temperature of treatment, pH of solution, dosage of sorbent, initial MB dye concentration, contact period and temperature of sorption were used as the independent factors and the percentage removal of MB dye as the dependent response. Table 1 shows the independent factors, experimental range and levels used in the batch sorption experiments of MB dye removal with the total of 49 experiments conducted at α = 1 using a 2^6 half factorial faced centered central composite design with five replicates at the center point and twelve experiments at axial point.

2.4. Batch Sorption Experiments

The sorption experiments were conducted by shaking the required amounts of sorbents into 20 mL of MB aqueous working solutions in 250 mL Erlenmeyer flasks and agitated at 180 rpm in a water bath at chosen temperature and contact period. The solutions were filtered using filter paper (Whatman, grade 3) and their final concentrations were analysed using UV-vis Spectrophotometer (Thermo Scientific, Genesys 6) at the wavelength of 665 nm. The percentage removal of MB dye was taken as a response (R) in this study and calculated as:

$$R = \left(\frac{C_i - C_f}{C_i}\right) * 100 \qquad (1)$$

where, C_i and C_f are the initial and final MB dye concentrations (mg/L) of the solutions respectively.

2.5. Statistical Analysis

The experimental data were subjected to a second order polynomial regression analysis using MINITAB 16 software to analyse the experimental data by estimating the response as the function of independent variables. The general form of the second order polynomial regression model used to explain MB dye removal (Equation (2)).

$$\hat{R} = \beta_0 + \sum \beta_i X_i + \sum \beta_{ij} X_i X_J + \sum \beta_{ii} X_i^2 \qquad (2)$$

where $\hat{R}$ is the predicted response (MB dye removal), β_0 is the intercept, β_i is effects of

the linear terms, β_{ii} is effects of the quadratic terms, β_{ij} is effects of the interaction terms and X_i are coded value of the corresponding i^{th} factors. The analysis of variance (ANOVA) with F-value, the lack of fit test (*LOF*) and the coefficient of determination, R^2 were used to determine the appropriate model. The optimum conditions for the removal was predicted using 'response optimizer'. The response contour plot was used to present the interaction effects of the significant variables.

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3. RESULTS AND DISCUSSION

3.1. Development of Regression Model Equation for MB Removal

Table 1 and 2 show the experimental design matrix, the actual and predicted dye removal efficiencies respectively. The predicted values were calculated from RSM while the actual values are measured from the experiment. The estimated optimum settings of each variable for removal of MB dye (%) and the interaction effects among the variables were analysed using the FCCCD method. The maximum removal efficiency of dye was found to be 100%. The second order polynomial equation fitted between the responses represent colour removal efficiency (R) and the input variable of temperature of treatment (X_1), pH of solution (X_2), dosage of sorbent (X_3), contact period (X_4), initial dye concentration (X_5) and temperature of sorption (X_6) is expressed in Equation (3) after the insignificant terms (P-value > 0.05) were eliminated.

$$\hat{R} = 99.923 + 17.8933X_3 + 0.0777X_4 - 0.2138X_5 - 0.036X_6 - 7.1018X_3X_3 - 0.028X_3X_4 + 0.093X_3X_5 + 0.067X_3X_6 - 0.001X_5X_6 \quad (3)$$

where $\hat{R}$ represents the predicted response (MB dye removal) while X_1, X_2, X_3, X_4, X_5 and X_6 are the coded values of the six independent variables.

Table 1. Central composite design matrix

Independent factor	Symbol	Unit	Low (-1)	Centre (0)	High (+1)
Temperature of treatment, X_1	Temp. Heat.	℃	100	450	800
pH of solution, X_2	pH	pH	2	6	10
Dosage of sorbent, X_3	W	g	0.1	1.55	3.0
Contact period, X_4	Time	min	60	120	180
Initial dye concentration, X_5	Conc.	mg/L	50	150	250
Temperature of sorption, X_6	Temp. Sorp.	℃	25	52.5	80

Table 2. Experimental and predicted values

Run	Temp. Heat	pH	Weight	Time	Conc.	Temp. Sorp.	R	$\widehat{R}$
1	100	10	3	180	50	80	99.5	100
2	800	10	0.1	180	250	25	73.3	57.0
3	800	2	3	60	50	25	99.8	95.4
4	450	6	0.1	120	150	52.5	73.3	70.5
5	100	2	3	180	250	80	99.9	98.0
6	450	6	1.55	120	150	52.5	100	99.9
7	100	10	3	60	250	80	99.9	98.8
8	100	10	0.1	180	250	80	43.2	41.7
9	100	2	3	60	50	80	100	100
10	450	10	1.55	120	150	52.5	99.9	99.9
11	100	2	3	180	50	25	99.8	94.5
12	450	6	1.55	120	150	52.5	100	99.9
13	100	2	0.1	180	250	25	55.0	57.0
14	100	2	0.1	180	50	80	99.0	98.5
15	100	10	0.1	180	50	25	99.3	100
16	800	10	0.1	60	250	80	39.2	32.7
17	450	6	1.55	120	250	52.5	99.9	87.7
18	100	2	0.1	60	250	80	13.8	32.7
19	800	2	0.1	60	50	80	90.3	89.5
20	800	2	0.1	180	50	25	92.9	100
21	450	6	1.55	120	150	52.5	100	99.9
22	450	2	1.55	120	150	52.5	99.9	99.9
23	450	6	1.55	120	150	25	99.9	100
24	800	10	3	180	50	25	94.3	94.5
25	100	10	3	180	250	25	99.9	100
26	450	6	1.55	120	150	80	100	97.7
27	450	6	1.55	120	150	52.5	99.9	99.9
28	800	2	3	180	50	80	99.8	100
29	100	2	3	60	250	25	100	100
30	450	6	1.55	60	150	52.5	100	97.9
31	100	10	0.1	60	50	80	99.8	89.5

32	450	6	3	120	150	52.5	99.9	99.4
33	800	10	0.1	180	50	8	98.8	98.5
34	800	2	3	180	250	25	99.9	100
35	450	6	1.55	180	150	52.5	100	100
36	800	10	3	180	250	80	99.9	98.0
37	100	6	1.55	120	150	52.5	100	99.9
38	450	6	1.55	120	150	52.5	99.9	99.9
39	800	10	0.1	60	50	25	99.5	93.9
40	100	10	0.1	60	250	25	48.9	48.0
41	800	2	3	60	250	80	99.9	98.8
42	800	10	3	60	50	80	98.5	100
43	800	2	0.1	180	250	80	38.5	41.7
44	100	10	3	60	50	25	99.5	95.4
45	800	10	3	60	250	25	99.9	100
46	100	2	0.1	60	50	25	95.0	93.9
47	450	6	1.55	120	50	52.5	99.7	100
48	800	6	1.55	120	150	52.5	99.8	99.9
49	800	2	0.1	60	250	25	39.6	48.0

Results displayed in Table 3 showed that main effects of Weight, Time, Conc. and Temp. Sorp., squared effect of Weight*Weight, interaction effects of Weight*Time, Weight*Conc., Weight*Temp. Sorp. and Conc.*Temp. Sorp. were significant. The coefficient of determination (R^2) value of the model is 0.9358 indicating that the experimental data can be predicted well by the model.

Table 3. Statistical regression coefficients forMBremovel efficiency (%) in coded units

Term	Coef	SE Coef	T-Value	P-Value
Constant	99.923	6.0371	16.552	0
Weight	17.8933	3.72058	4.809	0
Time	0.0777	0.02602	2.988	0.005
Conc.	-0.2138	0.02588	-8.26	0
Temp. Sorp.	-0.036	0.08186	-0.439	0.663
Weight*Weight	-7.1018	0.90184	-7.875	0
Weight*Time	-0.0282	0.01243	-2.268	0.029
Weight*Conc.	0.093	0.00746	12.468	0

Weight*Temp. Sorp.	0.067	0.02712	2.47	0.018
Conc.*Temp. Sorp.	-0.001	0.00039	-2.535	0.015

Table 4. ANOVA results for the quadratic model

Source	DF	Seq SS	Adj MS	F	P
Regression	9	21262.6	2362.51	63.13	0
Linear	4	12464	1175.88	31.42	0
Square	1	2320.5	2320.54	62.01	0
Interaction	4	6478.1	1619.52	43.28	0
Residual Error	39	1459.4	37.42		
Lack-of-Fit	15	824.4	54.96	2.08	0.053
Pure Error	24	634.9	26.46		
Total	48	22722			

R^2=0.9358; $R^2_{(pred)}$=0.8884; $R^2_{(adj)}$=0.9209

The results of analysis of variance (ANOVA) is presented in Table 4. From the regression, low P-value (<0.05) and large F-value (63.13) implied that the model was accurate. The non-significant value of lack of fit (P > 0.05) also supports the accuracy of the model. In addition, high coefficient of determination R^2 (0.9358) is adequate to describe the relationship between the response and variables. As it can have been observed, the values of R^2 (0.9358) and $R^2_{(adj)}$ (0.9209) of the model are close to each other which confirm the accuracy of the model. This denotes that 93.58 % of the sample variation can be described by the independent variables.

3.2. Main Effects

Fig. 1 presents the main effect plot of all independent variables on the removal of MB dye molecules, which is the response (R) of this study with the grand mean of 89.58 %. The effect of changing treatment temperature on sorption, while keeping the other parameters constant is illustrated in Fig. 1(a). The removal increased from 87.74 % to 98.16 % using T450 and then decreased to 84.97 % when T800 was applied, showing that T450 sludge has the highest removal efficiency. Adsorption of methylene blue by kaolinite has been documented by [12]. Based on our previous report [13], kaolinite was found in both raw and thermally treated alum sludge, but kaolinite in the sludge decreased remarkably when thermal treatment of sludge was conducted at 800^{o}C. Thus, removal efficiency of MB decreased when T800 was used.

To investigate the effect of pH on MB uptake, sorption experiments were performed at different pHs in the range of 2.0-10.0, as shown in Fig. 1(b). The pH of MB solution was

adjusted using 0.2 M of HCl or 0.1 M of NaOH solutions which introduced H^+ and OH^- ions into the solutionsrespectively. The MB removal efficiency remarkably increased from 86.36 % at pH 2 to 98.17 % at pH 6.0 and then decreased to 86.36 % at pH 10. Similar result was documented by [14]. Lower removal efficiency at pH 2 might due to the presence of higher amount of H^+ ions in the medium compared to that at pH 6, which caused higher competition between H^+ ions with the S^+ from MB dye molecules for the sorption sites. Similar explanation was also documented by numerous researcher [10, 15-17]. However, the reason for lower removal efficiency at higher pH i.e. pH 10 is different from the above explanation. Introduction of OH^- ions into the MB solution, caused competitions between OH^- ions and Cl^- (from MB molecules) for the sorption sites, thus leading to decrease in the sorption sites for MB molecules, consequently lower removal efficiency at pH 10. Similar explanation was reported by [16].

The removal of MB as a function of sorbent dosage was evaluated at dosage between 0.1 and 3.0 g. as shown in Fig. 1(c). As it can be seen, the percentage removal increased drastically with the increase of sorbent dosage from 0.1 to 1.55 g could be attributed to higher surface area of sludge and a greater number binding sites are available. However, when highest sorbent dosage (3.0 g) was used, there was a little change in percentage removal. Similar result has been documented by [18] using montmorillonite clay to remove MB. According to [19], the sorbents may overlap each other which might prevent some of the active sites reached by the MB molecules. Thus, this may explain the observation at the highest dosage.

The percentage of MB dye removal varied with the varying contact period of sorption as depicted in Fig. 1(d). It can be seen that the percentage of MB dye removal increases rapidly with contact period and thereafter, the percentage removal decreased gradually. For example, the percentage removal increased from 86.38 to 98.16 % when contact period increased from 60 to 120 min. But, the percentage removal decreased from 98.16 to 86.33 % when the contact period increased from 120 to 180 min. Similar trend was found by [20]. According to [19], MB molecules have sufficient time for the adsorption when long contact period is provided. However, aggregation of dye molecules might occur when longer contact period is applied.This might cause difficulties in diffusion of MB molecules to the adsorbent. As a result, percentage removal decreased when longer contact period (180 min) was used. In contrast, lower percentage removal at contact period of 60 min might due to less sufficient time for MB dye molecules to adsorb onto the active sites.

Besides that, there was significantly change of percentage removal in the effect of initial

concentration of MB dye, thus, the initial concentration of MB dye is one of the important factor in sorption of MB dye onto sludge sorbent. As concentration of MB dye increases from 50 to 150 mg/L, the percentage removal increases slightly from 97.20 to 98.18 % but decreases drastically (75.50 %) as the initial concentration increased to 250 mg/L (Fig. 1(e)). Increase in MB removal with initial concentration can be caused by increase of MB which are available for more sorption but lower sorption at 250 mg/L, as the sorption sites was saturated with MB molecules. The initial concentration provides the necessary driving force which this force overcome the resistance to the mass transfer of MB dye between solid and aqueous phase, therefore the interaction between dye molecules and sorbent enhanced and the sorption uptake increased [20]. Although driving force existed, high initial MB dye concentration may cause overload of the dye molecules. At 250 mg/L, the active sites on the sorbent could be occupied completely and the higher amount of dye molecule remained in the solution which caused lowest percentage removal of MB molecules.

To study the effect of temperature of sorption on MB dye sorption by drinking water treatment sludge, the experiments were performed at temperatures of 25, 52.5 and 80 ^{o}C. It is evident that the percentage removal increased rapidly as sorption temperature increased from 25 to 52.5 ^{o}C, but the percentage removal decreased remarkably as sorption temperature increased from 52.5 to 80 ^{o}C (Fig. 1(f)). The increase of percentage removal implies not only the temperature has a significant effect on the sorption, but also the sorption of MB is an endothermic process. Some researcher has reported similar findings [10, 21]. Higher percentage removal at 65 ^{o}C compared to 25 ^{o}C might due to the mobility of the molecule dye increases [21]. However, in [10] documented that the rate of diffusion of the dye molecules across the external boundary layer and the internal pores of the sorbents particle increases when temperature increased due to the decrease in the viscosity of the solution. This might explain lower removal when temperature increased from 60 to 80 ^{o}C as more MB molecules diffuse from the sorbent.

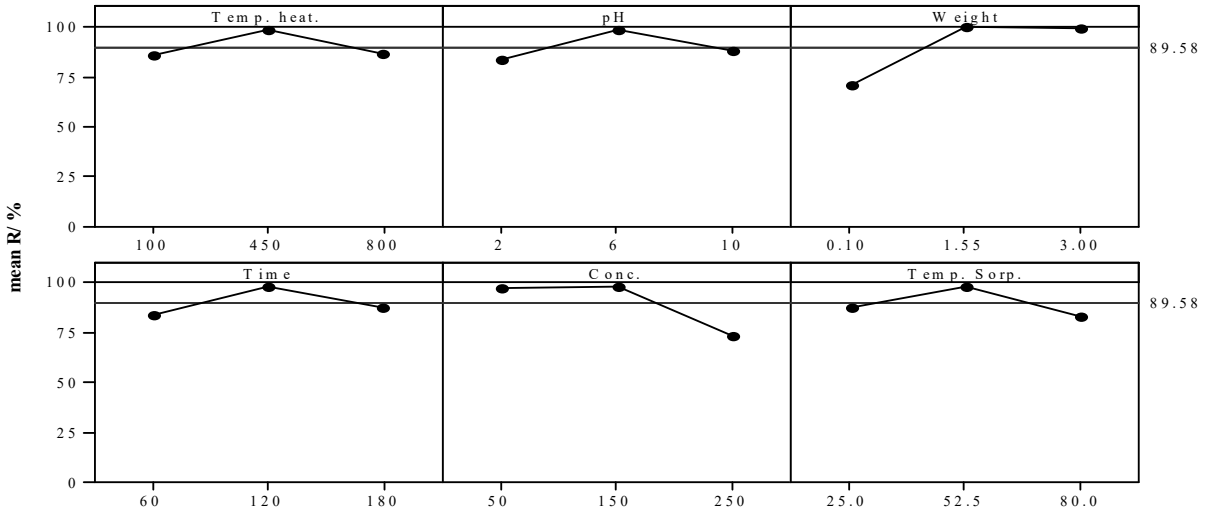

Fig.1. Effect of important parameters on the removal of MB dye molecules

3.3. Response Contour Plot

Fig. 2 illustrates the three-dimensional relationship between sorbent dosage and initial concentration was constructed for MB removal at constants pH (6), contact period (120 min), sorption temperature (52.5 ^{o}C) and treatment temperature (450 ^{o}C). The interaction between both parameters were highly significant compared to other significant interaction effect as the MB removal increased from 95.00 to 110 % at initial concentration of 50 mg/L when dosage of sorbent increased from 0.1 to 3.0 g. The percentage removal for MB decreased from 95.00 to 45.00 % dosage of sorbent at 0.1 gas initial concentration increased from 50 to 250 mg/L. Consequently, with appropriate sorbent dosage (1.55 g) and initial concentration (150 mg/L), the optimum removal of 100 % can be obtained as indicated by the peak of the plot. A good agreement was found when comparing the optimum value with that obtained by optimizing the regression model (Equation (3)).

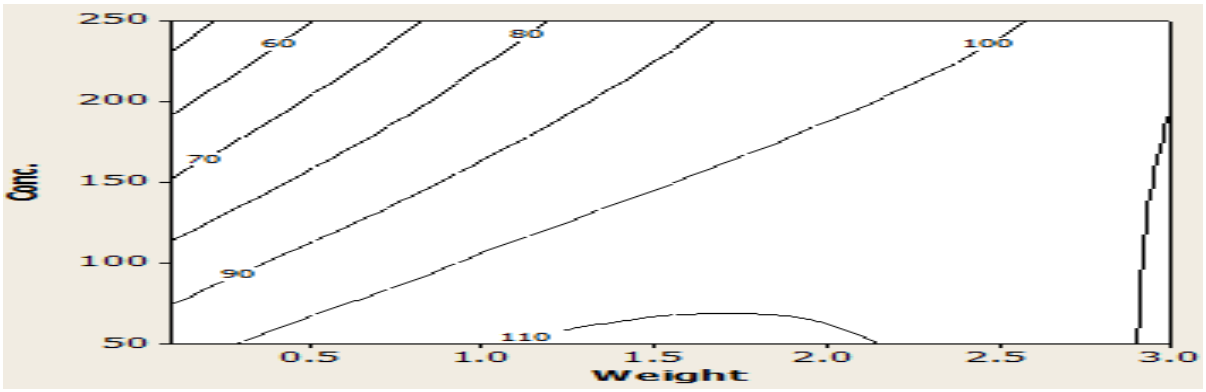

Fig.2. Response contour plot of percentage removal versus the effect of variables of initial concentration and dosage of sorbent

3.4. Optimization by Response Surface Optimizer

The optimum conditions to achieve 99.94 % removal of MB dye with 0.999 of composite desirability were predicted using response surface optimizer analysis and the results are depicted in Fig. 3. The results showed that the optimum conditions were suggested as: 1.55 g of sorbent dosage, 150 mg/L of initial dye concentration, treatment temperature of 450 ^{o}C, pH 6 of MB solution, 120 min of contact and 52.5 ^{o}C of sorption temperature. Sorption experiments were conducted in 4 replications at these suggested optimum conditions and the removal percentages were 100, 99.99, 100 and 99.99 % with the mean removal percentage of 100 % which was close to the prediction removal of 99.94 %. Thus, the experimental results were in good agreement with the predicted values. It can be said that the model can predict the percentage removal accurately.

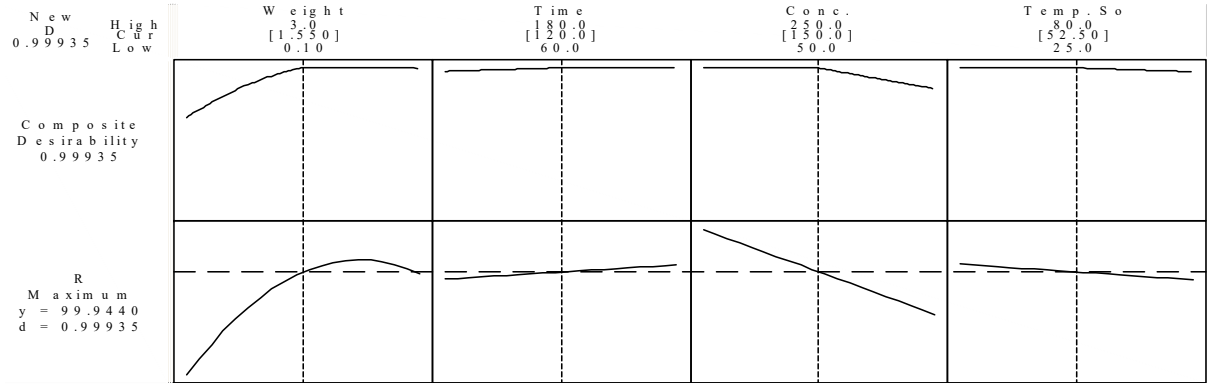

Fig.3. Response surface optimizer of MB dye removal using drinking water sludge

4. CONCLUSION

In this study, the sorption efficiency of drinking water treatment sludge was examined using MB dye solution. The effects of temperature of treatment, pH of solution, dosage of sorbent, initial dye concentration, contact period and temperature of sorption were investigated using ANOVA and the sorption conditions were optimized using RSM. The significance main effects were Weight, Time, Conc. and Temp. Sorp., squared effect of Weight*Weight, interaction effects of Weight*Time, Weight*Conc., Weight*Temp. Sorp., and Conc.*Temp. Sorp.. The optimum temperature of treatment, pH of solution, sorbent dosage, initial dye concentration, contact period and temperature of sorption were 450 ^{o}C, 6, 1.55 g, 150 mg/L, 120 min and 52.5 ^{o}C respectively for removal of 100 % of MB removal using drinking water treatment sludge. It can be concluded that drinking water treatment sludge is effective in removing MB dye from the aqueous solution. Model prediction was in good agreement with the experimental data as indicated by high coefficient of determination (R^2) of 0.9358.

5. ACKNOWLEDGMENTS

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