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APPLICATION OF RESPONSE SURFACE METHODOLOGY FOR OPTIMIZATION OF CADMIUM ADSORPTION IN AN AQUEOUS SOLUTION BY ACTIVATED CARBON PREPARED FROM *Bauhinia Purpurea* LEAVES

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ABSTRACT

An empirical model was developed and validated applying ANOVA analysis incorporating interaction effects of all parameters and optimized using response surface methodology (RSM) for the adsorption of cadmium onto activated carbon prepared from *Bauhinia Purpurea* leaves.These analyses were performed by means of Fisher's *F*- test, Student *t*-test and Probability '*P*' (< 0.05). For the effect of process parameters, the square (F = 806.731 and P = 0.000061) and linear (F = 8035.63 and P = 0.000035) terms are having a significant effect than interactive (F = 101.578 and P = 1.829998) terms on % removal of cadmium. The process variables of X_1 and X_4 showed the greatest significant positive effect and the other variables of X_2 and X_3 are having an insignificant effect on the cadmium adsorption process. All the squared terms, X_1, X_2, X_3 and X_4 shows a significant negative influence on the adsorption of cadmium. The interaction effect between process variables of X_1X_3 (P = 0.000808, t = 4.5669), X_2X_3 (P = 0.000018, t = 7.1926), were found to be statistically significant and having a positive effect, whereas the combinations of X_1X_2 (t =1.0919, P =0.298219), X_1X_4 (P = 0.185458, t = -1.4125), X_2X_4 (P = 0.832451, t = -0.2166), X_3X_4 (P= 0.513044, t = -0.6759) are having an insignificant and a negative effect on the % removal of cadmium. The optimal values from RSM for cadmium was pH (X_1) = 6.45, C_i (X_2) = 3.54 mg/L, w (X_3) = 0.14 g, $T(X_4)$ = 313.56K and maximum removal efficiency is 87.63%.

Keywords: Bauhinia *purpurea* leaves, Central Composite Design, optimization.

INTRODUCTION

The presence of heavy metals in the aquatic ecosystem results in a serious threat because of their toxicity, bioaccumulation, and bio-magnifications in the food chain.[1-3] Industries produce much wastewater and sludge containing heavy metal (Zinc, Copper, Nickel, Lead, Chromium, Cadmium, Mercury, etc.) ions with various concentrations, which are very toxic and carcinogenic to the human and other living organisms[4,5].The scope of this research work is the optimize the process parameters by using Response Surface Methodology. The main reason for implementing RSM is to evaluate the optimum process conditions for the adsorption or to determine a region that satisfies the operating specifications[6]. Response Surface Methodology is an efficient statistical method to assess the optimum operating conditions of the process or to resolve the range of process parameters that meets the process specifications with a minimum number of experiments as well as to inspect the relationship between one or more response variables and a set of quantitative experimental variables or factors. To provide efficient conditions for the process, RSM consists of design and experiments, response surface modeling through regression and

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optimization[7]. The application of statistical experimental design technique was adopted in the adsorption process to attaining a high degree of metal removal, closer confirmation of the output response to nominal and target requirements and reduced development time and overall costs[8].The central composite design was used to optimize cadmium removal by activated carbons prepared from *Bauhinia Purpurea* leaves in a batch system. It is used to examine the relationship between one or more response variables and a set of quantitative experimental variables or factors. The process optimization of the Cadmium recovery from ACBPL as the adsorbent using adsorption process has not been reported in the literature. Hence the present work intends to assess the effects of variables such as initial cadmium concentration, the weight of the adsorbent, temperature and pH of the solution to identify the optimum conditions using a central composite design (CCD). Designing of experiment and consistency of variables affecting the system is critical in the optimization process. The conventional optimization is carried out by varying a parameter whilst maintaining all other parameters involved at constant levels. Such methods are simple but do not describe the interaction effects of process variables. Moreover, this method is time consuming and gives low efficiency in optimizing the variables. In addition, the conventional classical method requires a large number of experimental runs to determine the optimum values. RSM is a collection of the statistical and mathematical technique used to develop, improve and optimize different processes. RSM is used to understand the crucial role of the experimental conditions (process parameters), as well as their interactions in a adsorption process[9,10]. It also gives the information about the complete interaction effects of all the process parameters influencing the adsorption process. In the present research work, RSM was adopted to study the influence of various process variables in the adsorption process and to determine the accurate model which in turn is used to get more accurate optimized values.

EXPERIMENTAL

Materials and Methods

RSM is one of the relevant techniques of experimental design which is frequently utilized after the essential controllable factors are distinguished and to discover the variable settings that optimize the response. RSM is preferred as it is convenient in making various projections, providing graphical illustrations. Consequently, this technique provides visual elucidation of the practical relations between the response and experimental variables.[11,12] In numerous chemical, biochemical and bio-environmental processes, RSM has been adapted to analyze, optimize and assess the interactive effects of independent factors. As per the research works conducted, Box and Wilson inspired the use of orthogonal design for evaluating the first-order model[13]. Eminent scientists and engineers have gained proficiency in applying central composite designs (CCDs) and three-level designs by the Box and Behnken for the second-order models[14]. In spite of many available designs available for fitting a second-order model, CCD suggested by Box and Wilson is the most preferred one among all the designs. It consists of factorial points (from a 2^m design), central points, and axial points. Based on these points, the total number of experiments designed by CCD will be:

$$N = 2^m + 2m + k_o \qquad (1)$$

Where, N is the total no. of experiments, m is the no. of the factors studied, and k_o is the no. of replicates. Design expert software or Minitab is generally preferred for using CCD under RSM. In this study, statistical software STATISTICA 6.0 (Stat Soft Inc.) is used as an optimization software. The value of α is important to calculate in CCD, as the location of axial points in the experimental domain could be easily determined using the value. The α value determines the design to be applied, whether spherical, orthogonal, rotatable, or face-centered. More accurately, the CCD is made by choosing a value of :

$$\alpha = \left[2^{\frac{m}{4}} \right] \qquad (2)$$

To study the influence of pH of the solution (2-8) , initial lead concentration in the solution (C_i= 2-10 mg/L), Dosage of adsorbent (w = 0.025-0.15g) and temperature of the solution (303-323K) on the % removal, a Central Composite Design (CCD) is used to optimize the levels of these variables. Analysis of variance (ANOVA) was utilized to test the significance of each term in the equation and the goodness of fit of the regression model.[15-17]

This RSM is applied to gauge the result by contour plots in order to examine the individual and cumulative effects of the variables and the mutual interactions between the variables on the dependent variable.Using the response surface regression procedure of statistical analysis system experimental data obtained was analyzed. The interaction between responses and independent factors was obtained by fitting them into second-order polynomial equation:

$$Y = \alpha_o + \sum_{i=1}^{k} \alpha_i x_i + \sum_{i=1}^{k} \alpha_{ii} x^2{}_i + \sum\sum_{i<j}^{k} \alpha_{ij} x_i x_j + \in \tag{3}$$

Where Y is the response, α_0 is the constant, α_i is the incline or straight impact of the data element x_i, α_{ii} is the quadratic effect of input factor x_i, α_{ij} is the linear by linear interaction effect between the input factor x_i and ϵ is the residual term.

RESULTS AND DISCUSSION

Activated carbon prepared from *Bauhinia Purpurea* leaves (ACBPL) was used as low-cost adsorbent for removal of cadmium from aqueous solution. Optimization of adsorption process parameters using response surface methodology was also reported in this section.

Table -1: Range of Adsorption Process Parameters covered in the Present Study

Process parameter	Activated Carbon prepared from *Bauhinia Purpurea* leaves	
	Cadmium	
	Min	Max
Contact time, *t* (min)	2	80
Initial metal ion concentration of the solution, C_i *(mg/L)*	2	10
Solution pH	2	10
The average particle size of the adsorbent, *d* (µm)	74	177
The dosage of the adsorbent, *w* (g)	0.025	0.15
Temperature, *T* (K)	303	323

Table -2: Experimental Variables and Levels investigated by Central Composite Design

Variable	Process parameter	Level of Process parameters				
		-2	-1	0	1	2
X_1	Solution pH	3	4	5	6	7
X_2	Initial metal concentration ,C_i *(mg/L)*	2	4	6	8	10
X_3	Dosage of adsorbent, *w (g)*	0.05	0.075	0.1	0.125	0.15
X_4	Temperature (K)	303	308	313	318	323

Optimization of Adsorption Process Parameters using Factorial Experimental Design (CCD)

The response surface methodology is a collection of useful mathematical and statistical technique for analyzing the effects of several independent parameters on the response of interest. Preliminary experimental results indicated that the pH (X_1),initial metal concentration (X_2) , adsorbent dosage (X_3) and temperature of metal solution (X_4) were all the five essential parameters of adsorption studies, significantly affected the percentage of adsorption and metal deposition on activated carbon of *Bauhinia Purpurea* leaves. Therefore, these parameters were chosen to achieve the optimized conditions for the maximum percentage of adsorption of Cadmium onto activated carbon of *Bauhinia Purpurea* leaves using response surface methodology. The response was expressed as the percentage of adsorption of Cadmium onto the adsorbent. A CCD with 26 experiments, which includes a 8 cubic point runs, 6 center point runs and 2 axial point runs, was utilized for the optimization of process parameters. For statistical calculations, all independent variables were coded to five levels as X_i according to the following equation,

$$X_i = \frac{x_i - x_{oi}}{\Delta x_i} \quad (i=1,2,3,\ldots\ldots\ldots k) \tag{4}$$

Where, X_i is the dimensionless value of an independent variable, X_i is the real value of an independent variable, x_{oi} is the real value of the independent variable at the center point, and ΔX_i is the step change. On the basis of preliminary experimental results, it is noted that all the four parameters pH, initial metal ion concentration, the dosage of the adsorbent and temperature significantly affected the adsorption efficiency of Cadmium and metal deposition on the adsorbent. For statistical calculations, all independent variables were coded using Equation (3) as discussed in section (2). Based on the analysis of preliminary experimental results the levels of independent process variables used in a CCD are shown in Table-1. The application of RSM yielded the following regression models and the following equation was found to represent the % removal of cadmium (Y) using activated carbon of *Bauhinia Purpurea* leaves as an adsorbent.

$$(Y\%) = -12587.5 + 83.9X_1 - 0.5X_2 + 813.1X_3 + 78.7X_4 - 5.2X_1^2 - 0.7X_2^2 - 2775.3X_3^2 - 0.1X_4^2 + 0.2X_1X_2 + 52.7X_1X_3 - 0.1X_1X_4 + 41.5X_2X_3 - 1.6X_3X_4 \tag{5}$$

In order to study the combined effect of the factors, experiments were performed for different combinations of the process parameters using statistically designed experiments (Table-2). Apart from the linear effect of the parameters on the adsorption efficiency of cadmium, the RSM also gives insight into the squared and interaction effects of the parameters. These analyses were performed by means of Fisher's *F*- test and Student *t*-test[18].The Student *t*-test was used to determine the significance of the regression coefficients of the parameters. In general, the larger the magnitude of *t* and smaller the value of *P*, the more significant is the corresponding coefficient term. The influence of linear, square and interaction effects of process variables on the adsorption efficiency of cadmium is shown in Table-3. These results were demonstrated from the cadmium adsorption process by means of Fisher's *F*-test and Student *p*-value. For all the parameters, the square model (F = 806.731 and P = 0.000061) and linear (F = 8035.63 and P = 0.000035) model terms are having significant effect than interactive (F = 101.578 and P = 1.829998) model terms of cadmium adsorption process using activated carbon of *Bauhinia Purpurea* leaves as an adsorbent. Based on *t*-test and *p*–values of the process variables of X_1 and X_4 showed the greatest significant positive effect and the other variables of X_2 and X_3 (Table-4) are having an insignificant effect on the cadmium adsorption process. All the squared terms, X_1, X_2, X_3 and X_4 shows a significant negative influence on the adsorption of cadmium using activated carbon of *Bauhinia Purpurea* leaves as an adsorbent. The interaction effect between process variables of X_1X_3 (p = 0.000808, t = 4.5669), X_2X_3 (p = 0.000018, t = 7.1926), were found to be statistically significant and having positive effect on adsorption efficiency (%) of cadmium using activated carbon of *Bauhinia Purpurea* leaves as an adsorbent, whereas the combinations of X_1X_2 (t =1.0919, p =0.298219), X_1X_4 (p = 0.185458, t = -1.4125), X_2X_4 (p = 0.832451, t = -0.2166), X_3X_4 (p = 0.513044, t = -0.6759) are having an insignificant effect on the adsorption efficiency of cadmium. The shapes of the surface plots indicate an interaction between the variables. The elliptical shape of the response surface curve indicates good interaction between two variables. It represents the interactive effect of any two variables on the response variable when the remaining variables kept constant. The surface and contour plots (Figs.-2a to 2f) are used to show the pictorial representation of the influence of independent variables and their interaction on the dependent variable. The response surface plots had a clear peak, which indicated that the optimum conditions fall inside the design boundary. The maximum adsorption efficiency of Cadmium is indicated by the surface confined to the smallest curve of the plot with the other variable maintained at hold value. The projection of the surface and contour plots (Figs.-2a and 2b) indicate that the adsorption efficiency increased with increasing temperature and attained maximum efficiency of 60 % at pH: 6.5 and T: 314K. Similarly, Figs.-2d and 2f indicate that the adsorption efficiency is highly influenced by the initial concentration (80 % at 2 mg/L) and moderately with the dosage of the adsorbent (60 % at 0.14 g of *ACBPL*) with increasing temperature. Figs.-2d and 2e show that the adsorption efficiency increased with an increase in pH from 4 to 6.5; the less removal observed at low pH may be due to the more availability of H^+ ion activity comparable to Cd^{+2} in the solution. Besides, it was explained that at lower pH value, the surface of the adsorbent is surrounded by H^+ ions, thereby preventing the metal ions from approaching the binding sites of the adsorbent. This means that at the higher H^+ concentration, the repulsion is taking place between the metal and adsorbent and decreased adsorption efficiency. In contrast, as the pH increases, the adsorbent surface converted as more negatively charged surface and capture the lead ions to increase the adsorption efficiency. However, when the pH was greater than 6.5, there was a decrease in the adsorption capacity. This may be due to the occurrence of Cadmium precipitation. At this stage there are three species present in solution, Cd^{2+} in very small quantities, $Cd(OH)^+$ and $Cd(OH)_2$ in large quantities.[19-22] The % removal of Cadmium increased when the initial concentration was increased from 1 to 3 mg/L (Figs.-2b, 2d and 2f). Besides, higher initial concentrations of Cadmium to an increase in the affinity of the lead ions towards the active sites. However, a further increase in the initial concentration (>3 mg/L) resulted in a decrease in the adsorption efficiency of Cadmium. This indicates that the initial Cadmium concentration is an effective parameter to maximize adsorption efficiency. The % removal of Cadmium increased when the dosage of the adsorbent was increased from 0.05 to 0.15 g (Figs.-2c and 2f). This was due to the limited availability of the number of adsorbing species for a relatively larger number of surface sites on the adsorbent at a higher weight. However, a further increase in adsorbent dosage (> 0.14 g) resulted in a decrease in % removal of Cadmium. These results may be due to the overlapping of the adsorption sites as a result of overcrowding of adsorbent particles. In order to maximize the adsorption efficiency of metals, regression model equations (5) developed by using response surface methodology for the prediction of the effect of process variables on the % removal of Cadmium was optimized separately. The optimal values from RSM for cadmium was as follows: pH = 6.45, initial metal ion concentration = 3.54 mg/L, dosage of the adsorbent = 0.14 g, temperature = 313.56 and maximum removal efficiencies of 87.63%. The optimum values for process variables obtained by using RSM were validated by conducting experiments at these optimum conditions and obtained the maximum % removal of Cadmium is 85.57%. The above results proved that the statistical experimental design using RSM could be effectively used to optimize various process parameters for improving the adsorption of cadmium onto activated carbon of *Bauhinia Purpurea* leaves as an adsorbent.

Table - 3: Analysis of Variance (ANOVA) for a response surface quadratic model for removal of Cadmium

Source	SS	DF	MS	F	P (Prob>F)
Linear	8035.63	4	8035.63	6034.433	0.000035
X_1	4022.27	1	4022.27	3020.561	0.000000
X_2	2758.89	1	2758.89	2071.821	0.000000
X_3	1195.11	1	1195.11	897.484	0.000000
X_4	59.34	1	59.34	44.567	0.000035
Square	806.731	4	806.731	605.822	0.000061
X_1^2	465.338	1	465.338	349.450	0.000000
X_2^2	120.235	1	120.235	90.291	0.000001
X_3^2	52.517	1	52.517	39.438	0.000060
X_4^2	168.641	1	168.641	126.643	0.000000
Interaction	101.578	6	101.578	76.281	1.829998
X_1X_2	1.588	1	1.588	1.192	0.298219
X_1X_3	27.773	1	27.773	20.856	0.000808
X_1X_4	2.657	1	2.657	1.995	0.185458
X_2X_3	68.890	1	68.890	51.734	0.000018
X_2X_4	0.062	1	0.062	0.047	0.832451
X_3X_4	0.608	1	0.608	0.457	0.513044
Error	14.648	11	1.332		
Total SS	8649.094	25	R^2 =.9983	R^2 (Adj) = .9961	

DF: degree of freedom; SS: the sum of squares; F: factor F; P: probability.

Table-4: Estimated Regression Coefficients and Corresponding *t*- and *P*- Values for the Adsorption of Cadmium.

Adsorption parameter (Mean value)	Regression Coefficient	Standard Error	t-Value	p-Value
Constant	-12587.5	1095.705	-11.4881	0.000000
X_1	83.9	18.328	4.5763	0.000795
X_2	-0.5	9.115	-0.0514	0.959945**
X_3	813.1	730.933	1.1124	0.289673**
X_4	78.7	6.928	11.3659	0.000000
X_1^2	-5.2	0.276	-18.6936	0.000000
X_2^2	-0.7	0.069	-9.5022	0.000001
X_3^2	-2775.3	441.934	-6.2800	0.000060
X_4^2	-0.1	0.011	-11.2536	0.000000
X_1X_2	0.2	0.144	1.0919	0.298219**
X_1X_3	52.7	11.540	4.5669	0.000808
X_1X_4	-0.1	0.058	-1.4125	0.185458**
X_2X_3	41.5	5.770	7.1926	0.000018
X_2X_4	-0.0	0.029	-0.2166	0.832451**
X_3X_4	-1.6	2.308	-0.6759	0.513044**

**insignificant ($P \geq 0.05$)

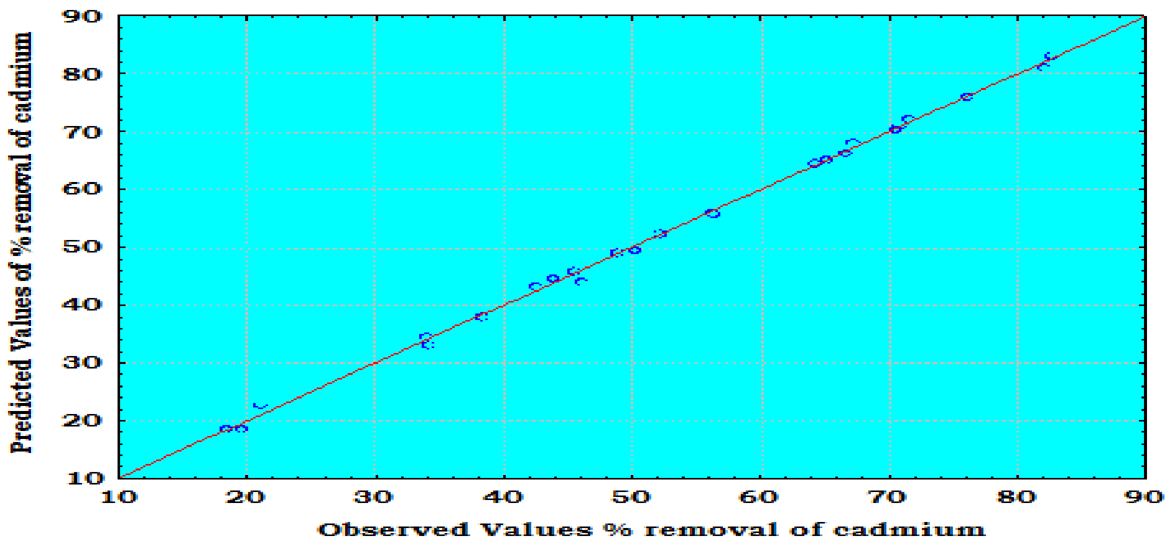
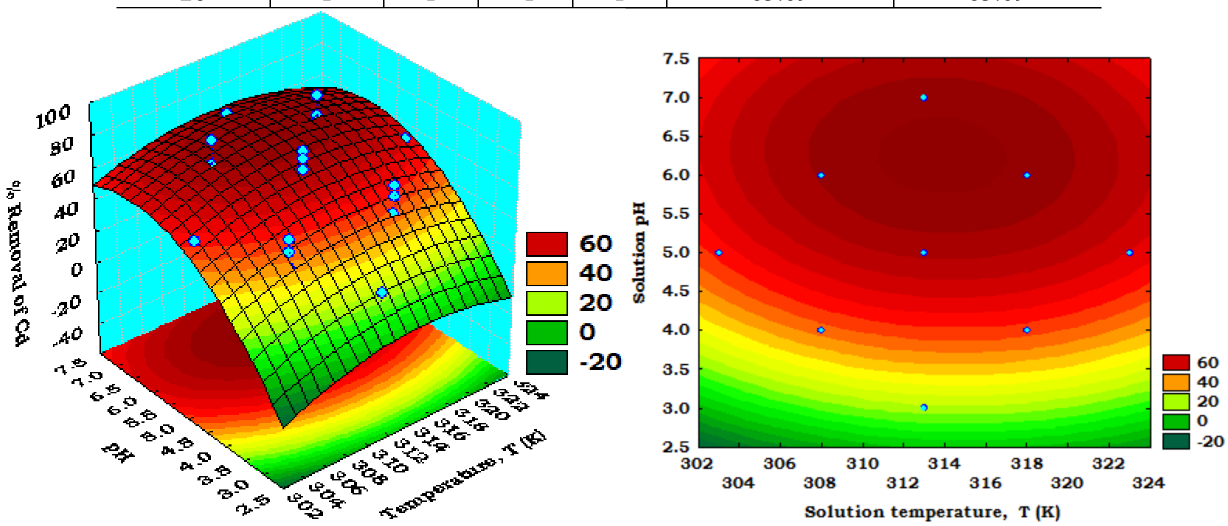


Fig.-1: Correlation Plot of Experimental Values Vs Predicted Values

Table-5: Experimental Design Matrix and Results in the Adsorption of Cadmium

Run	X_1	X_2	X_3	X_4	% Removal of Cadmium	
					Observed	Predicted
1	-1	-1	-1	1	43.77	44.64
2	-1	1	1	1	48.91	49.12
3	-1	-1	1	1	52.17	52.36
4	0	2	0	0	56.21	56.06
5	-1	1	1	-1	18.42	18.55
6	0	0	0	2	21.03	22.77
7	2	0	0	0	34.04	34.56
8	0	0	0	-2	38.35	38.01
9	0	0	2	0	67.24	68.09
10	-1	1	-1	-1	70.78	70.93

11	0	0	0	0	82.14	81.07
12	1	-1	-1	-1	82.76	83.14
13	1	-1	1	1	42.42	43.25
14	1	1	-1	-1	45.53	45.84
15	0	-2	0	0	64.24	64.54
16	-1	-1	-1	-1	66.55	66.35
17	-2	0	0	0	19.55	18.54
18	-1	-1	1	-1	70.52	70.32
19	1	-1	-1	1	76.17	76.03
20	1	-1	1	-1	34.21	33.14
21	0	0	-2	0	46	44.03
22	1	1	1	-1	71.5	72.26
23	1	1	1	1	50.24	49.51
24	0	0	0	0	56.27	55.80
25	-1	1	-1	1	65.09	65.09
26	1	1	-1	1	65.09	65.09



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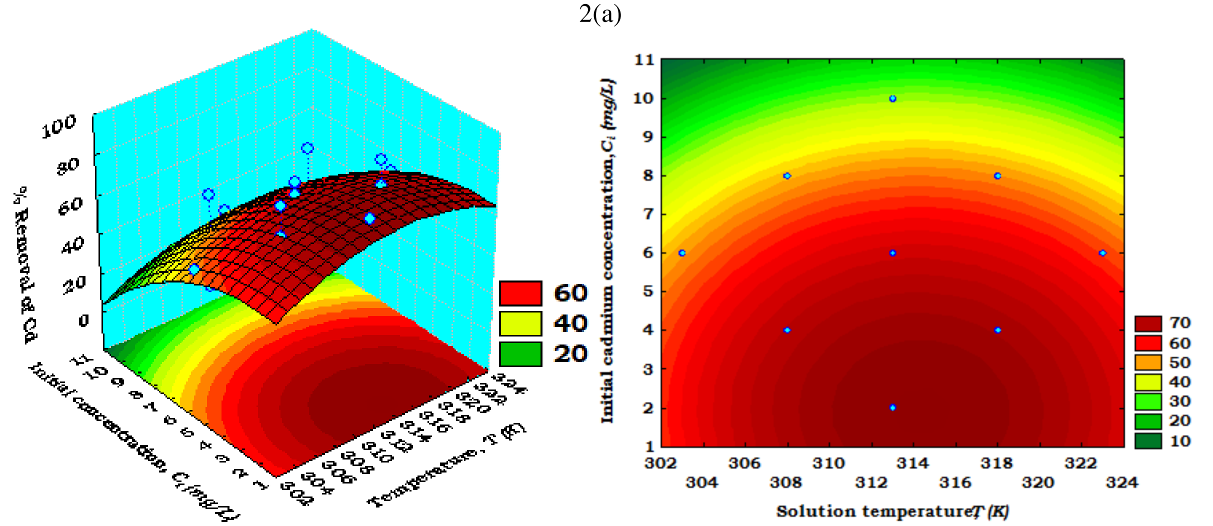
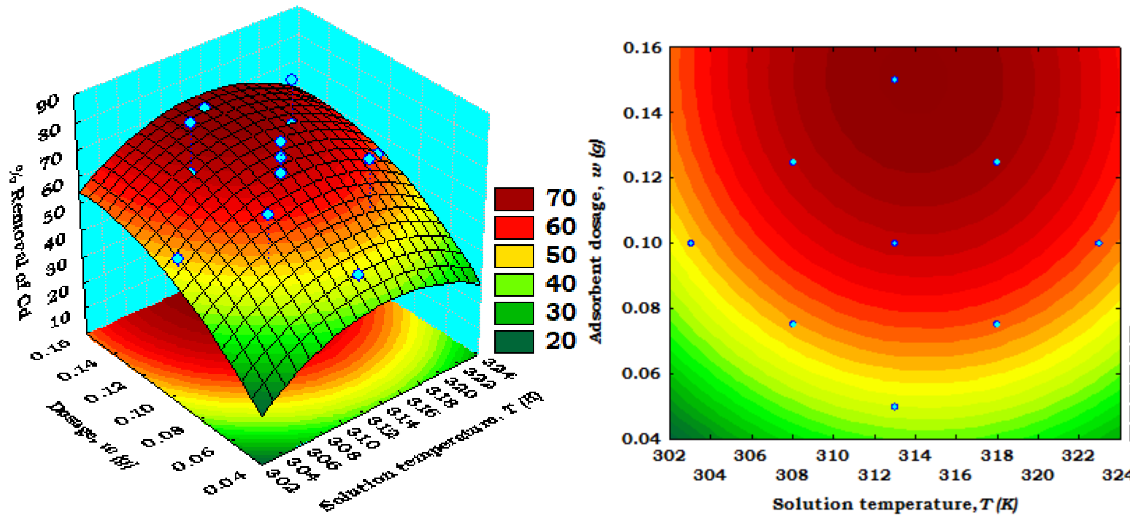


2 (b)

Fig.-2: Response surface and contour plots showing (a) the effect of temperature (X_4), Solution pH(X_1) and their mutual interaction on % removal of Cadmium, with constant adsorbent dose (X_3) and initial Cadmium concentration (X_2); (b) the effect of X_4, X_2 and their mutual interaction on % removal of Cadmium, with constant level of X_1 and X_3.



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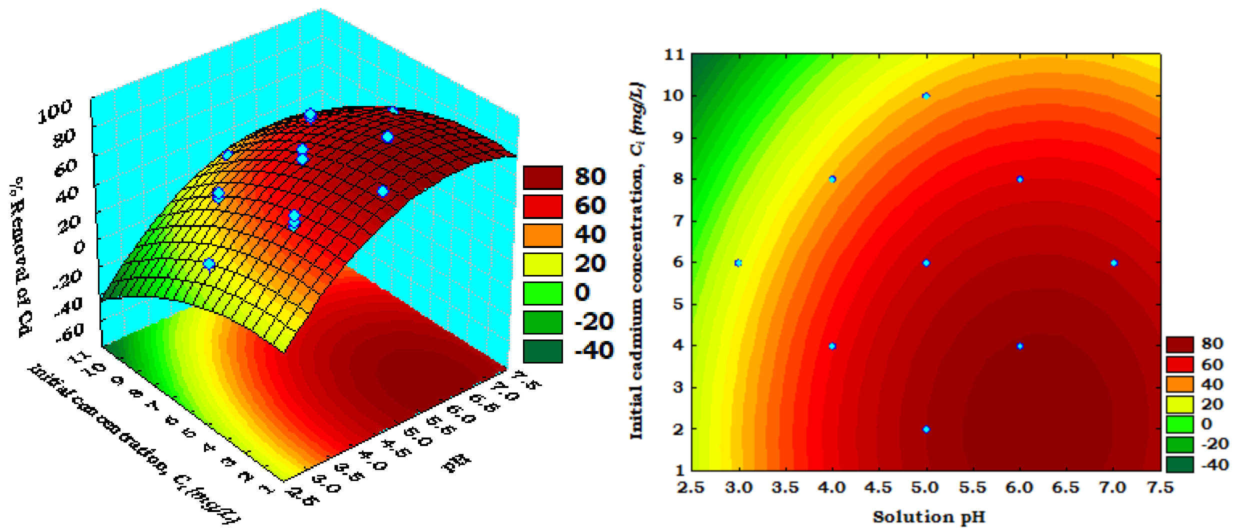
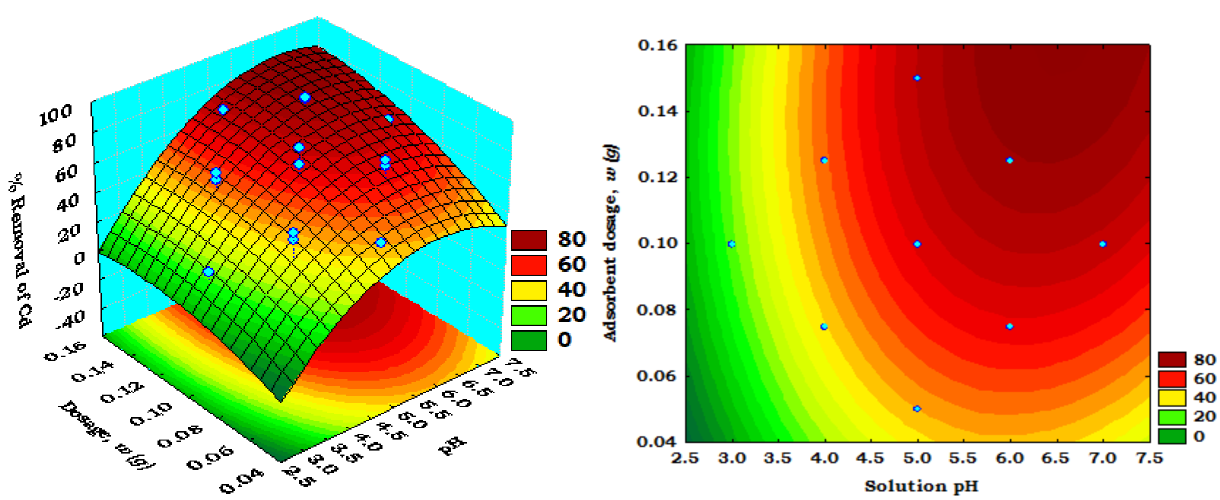
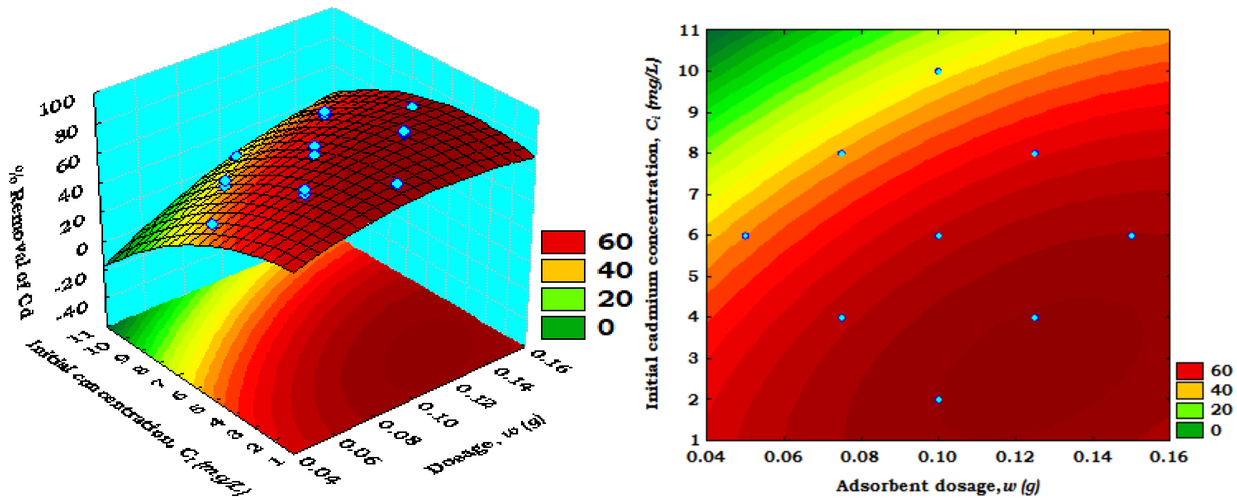


2(d)

Fig.-2: Response surface and contour plots showing (c) the effect of temperature (X_4), Dosage (X_3) and their mutual interaction on % removal of Cadmium, with constant pH (X_1) and initial Cadmium concentration (X_2); (d) the effect of X_1, X_2 and their mutual interaction on % removal of Cadmium, with constant level of X_3 and X_4.



2(e)



2(f)

Fig.-2: Response surface and contour plots showing (e) the effect of pH (X_1), Dosage (X_3) and their mutual interaction on % removal of Cadmium, with constant temperature (X_4) and initial Cadmium concentration (X_2); (f) the effect of X_2, X_3 and their mutual interaction on % removal of Cadmium, with a constant level of X_1 and X_4.

Table-6: The Optimal Values of the Process Variables and Responses (% Removal) of Cadmium.

	Activated carbon prepared from *Bauhinia Purpurea* leaves	
Process parameter	Cadmium	
	Opt.(Using CCD)	Opt.(Experimentaion)
Solution pH (X_1)	6.45	6.5
Initial metal concentration (X_2), (mg/L)	3.54	4
Dosage of the Adsorbent (X_3), g	0.14	0.15
Solution temperature (X_4), K	313.56	313
Maximum% removal	87.63	88.76

CONCLUSION

The effects of various process parameters on % removal of cadmium and metal uptake of activated carbon of *Bauhinia Purpurea* leaves as adsorbents were determined using adsorption technique. The following conclusions could be drawn from the present study on the removal cadmium from aqueous solutions using adsorption technique:

- The optimized process variables obtained from the RSM for the adsorption of cadmium onto activated carbon prepared from Bauhinia Purpurea leaves adsorbent was in close agreement with the experimental data.
- The optimal values from RSM for cadmium was as follows: pH = 6.45, initial metal ion concentration = 3.54 mg/L, dosage of the adsorbent = 0.14 g, temperature = 313.56 and maximum removal efficiencies of 87.63%.
- The optimum values for process variables obtained by using RSM were validated by conducting experiments at these optimum conditions and obtained maximum % removal of Cadmium is 85.57%.
- Second order polynomial equation to represent the adsorption of cadmium onto activated carbon of Bauhinia Purpurea leaves:

$$(Y_4\%) = -12587.5 + 83.9X_1 - 0.5X_2 + 813.1X_3 + 78.7X_4 - 5.2X_1^2 - 0.7X_2^2 - 2775.3X_3^2 - 0.1X_4^2 + 0.2X_1X_2 + 52.7X_1X_3 - 0.1X_1X_4 + 41.5X_2X_3 - 1.6X_3X_4.$$

- Surface and contour plots were successfully used for the study of evaluation of the effects of two combined process variables on % removal of metal ions.

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