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DEVELOPMENT OF MODELS FOR DYE REMOVAL PROCESS USING RESPONSE SURFACE METHODOLOGY AND ARTIFICIAL NEURAL NETWORKS

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ABSTRACT

In this study, simple, green and economic bio sorbent was examined for dye removal process. The influence temperature, initial pH, contact time, adsorbent dosage and initial dye concentration of the solution on dye removal capacity of Ficus religiosa seeds was studied by using statistical designs. Response surface methodology was used to develop a model to predict the effects of all the above parameters on dye removal. Model analysis revealed that the adsorption capacity was affected by the changes in temperature, initial pH, contact time, adsorbent dosage and initial dye concentration of the solution. The results obtained were also modeled by using Artificial Neural Networks (ANN). High values of correlation coefficients indicated the best fits of experimental results with that of values obtained from both the modeling methods. From these studies, it may be concluded that green carbon adsorbent prepared is efficient and economical for crystal violet removal from aqueous solutions.

KEYWORDS: ANN, Dye, Model, RSM

INTRODUCTION

Wastewater discharged from dye, food, textile, leather, cosmetic, paper and printing industries contains significant amount of color coupled with poor biodegradable complex components, high pH, temperature and COD concentration (Alinsafi et al., 2006). Presence of color even in small quantities makes it unusable and ineffective. Most of the dye materials are toxic and potentially carcinogenic, they may cause skin allergy, eye irritation and other carcinogenic problems (Moller and Wallin 2000; Stolz, 2001; Mechichi et al., 2006). Untreated or partially treated dye effluents cause aesthetic and environmental problems, thus they need to be treated before their discharge into water streams. Recently, interest of research on dye removal has been increasing, as government is imposing stringent legislations concerning the release of contaminated effluent (R. Rajeshkannan et al., 2009).

Industrially dye containing waste water is usually treated with conventional methods such as membrane separation, dialysis, electro dialysis, coagulation, flocculation, adsorption etc. Among these methods, adsorption is by far the most versatile and widely used method because of its wide ranging availability, low cost and ease of operation. Different adsorbents such as shrimp waste, crab shells, aspergillus niger, apricot stone shells, bamboo, plum kernels etc., have been studied to assess their capacity to remove dyes from wastewater (Khalil and Girgis, 1998; Chan et al., 2008; Wu et al., 1999a; Wu et al., 1999b), but most of them found to be uneconomical and ineffective. Hence, still there is a need to develop new and efficient adsorbent for the removal of dyes from industrial effluents. Keeping this in mind, present study is aimed to develop an efficient adsorbent for the dye removal.

Designing of experiment and standardization of variables affecting the system is very critical in optimization process. Generally this optimization is carried out by using traditional one factor at a time method, which is simple, time and chemicals are consumed in large quantities. Moreover this method neglects the interaction effects of process variables. [Duff et al., 1973; S. Kara et al., 2007; V. Ponnusami et al., 2008; Yujie Shi et al., 2009; M. Mohammadian Fazli et al., 2010; Narayana Saibaba et al., 2011a; Narayana Saibaba et al., 2011b; Narayana Saibaba et al., 2011c; Narayana Saibaba et al., 2012].

Numerous researchers have applied neural networks in the modeling of various systems in which no explicit scientific solutions were available (Zhang et al., 1992; Sayeed et al., 1995; Fang et al., 1998; Ni and Gunasekaran, 1980; Liao et al., 1993; Muller H. G.,1988; Ruan R, 1995). Hence, in the present study statistical approach such as Response Surface Methodology (RSM) was adopted to study the correlation among the process variables affecting the process. The objective of this work was to explore the use of different methods such as Central Composite Design and Artificial Neural Networks to predict the color removal capacity of biosorbent.

MATERIALS AND METHODS

Materials

Crystal Violet was chosen as model molecule to evidence the potentiality of adsorption for the treatment of waste water containing dye. Crystal violet dye, HCl and NaOH were purchased from lotus enterprises, visakhapatnam, India. Deionized water was used to prepare the dye solutions. All the glassware was washed thoroughly with deionized water before use.

Preparation of Sorbent

The Ficus religiosa seeds used in this study were obtained from Dupputuru Village in Visakhapatnam District. The collected biomaterial was extensively washed with tap water to remove soil and dust. The bio material was then dried by exposure to the sunlight for 3 days and subsequently at 80°C for 3 h in a hot air oven. The dried material was milled in to a powder using a domestic mixer and 200 mesh size materials was separated and sealed in plastic bags, and stored in desiccators for use.

Adsorption Studies

Stock solutions of Crystal Violet dye was prepared by dissolving the calculated amount of the dye in 1 L volumetric flask of de-ionized water and kept in dark colored glass bottles for further use. Test solutions of this dye were prepared from stock solutions by dissolving appropriate amounts of de ionized water. Dye solutions of 50 ml were taken in a 100 ml Erlenmeyer flasks and required pH's were maintained by using 0.1N HCl or 0.1N NaOH. The pH of the initial solutions was adjusted using dilute HCl or NaOH. Dye solutions were contacted with the appropriate amounts of sorbent and kept at 120 rpm in an orbital shaker at the predetermined temperatures. The concentration of residual dye in the test solution was determined after the completion of predetermined time intervals as per experimental design. UV spectrophotometer with wavelength corresponding to the maximum absorbance of 590 nm was used for measuring dye concentration. All the experiments were carried out triplicate.

RESULTS AND DISCUSSIONS

The effect of Temperature (x_1), pH (x_2), time (x_3), adsorbent dosage (x_4) and initial dye concentration (x_5) on percentage dye removal efficiency(Y) was studied by using a Central Composite Design. A total number of 32 experiments were employed for the optimization of parameters for removal of CV dye from aqueous solution. The experimental results show that the dye removal percentage was a function of temperature, pH, time, adsorbent dosage and initial dye concentration. Using the experimental results, the following second order polynomial modeling equation was fitted to the decolonization results and obtained in terms of factors:

$$Y_1 = 3.488 + 1.485X_1 + 3.374X_2 - 0.448X_3 + 134.227X_4 + 0.286X_5 - 0.037X_1^2 - 0.198X_2^2 -0.001 X_3^2 - 38.065 X_4^2 + 0.0001X_5^2 + 0.044 X_1X_2 + 0.001 X_1X_3 - 0.350 X_1X_4 - 0.002 X_1X_5 + 0.043 X_2X_3 -1.142 X_2X_4 - 0.014 X_2X_5 - 0.082 X_3X_4 + 0.003 X_3X_5 - 0.559 X_4X_5 \text{ -------- } (1)$$

The experimental results along with predicted values of percentage removal using the above equation were given in Table -1. The modeling equation efficiency in predicting the dye removal was evaluated by regression coefficients, standard error, t-values, p-values, correlation coefficient (R) and the determination coefficient, R^2, (Table-2).

It was observed from table-2 that dye removal is highly influenced by temperature, pH and time where as the effect of adsorbent dosage and dye concentration were had insignificant effect. Figure-1 compares the experimental dye removal efficiency with the predicted values obtained from the model. The figure indicated good agreements between the experimental and predicted values of dye removal efficiency.

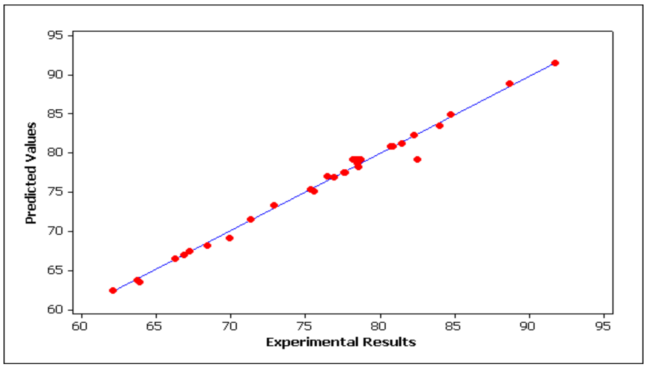

Figure 1: Comparison of Predicted Results with Experimental Results

Table 1: CCD Plan Matrix in Coded Values and Responses

Run Order	X_1	X_2	X_3	X_4	X_5	% Removal of dye	
						Observed	Predicted
1	0	0	0	0	0	82.500	79.157
2	-1	-1	1	-1	-1	66.250	66.517
3	-1	1	-1	-1	-1	78.436	78.738
4	-1	-1	-1	-1	1	72.879	73.353
5	0	0	0	0	0	78.145	79.157
6	0	0	0	-2	0	69.876	69.117
7	1	1	-1	1	-1	81.456	81.238
8	1	-1	-1	1	1	63.756	63.710
9	-1	-1	1	1	1	75.365	75.343
10	1	1	1	-1	-1	77.552	77.504
11	2	0	0	0	0	67.214	67.496
12	0	0	0	2	0	76.444	77.017
13	0	0	2	0	0	80.665	80.898
14	0	0	0	0	0	78.356	79.157
15	0	0	0	0	-2	80.865	80.860
16	-1	1	1	-1	1	88.688	88.872
17	-1	1	1	1	-1	91.689	91.495

Table 1: CCD Plan Matrix in Coded Values and Responses – Contd.

Run Order	X_1	X_2	X_3	X_4	X_5	% Removal of dye	
						Observed	Predicted
18	-1	1	-1	1	1	76.899	76.913
19	0	0	0	0	2	77.660	77.478
20	0	0	0	0	0	78.666	79.157
21	1	1	1	1	1	78.521	78.184
22	-2	0	0	0	0	83.943	83.475
23	1	-1	-1	-1	-1	62.112	62.355
24	1	-1	1	1	-1	68.443	68.190
25	0	-2	0	0	0	63.854	63.505
26	-1	-1	-1	1	-1	82.236	82.333
27	1	1	-1	-1	1	71.326	71.485
28	0	0	0	0	0	78.546	79.157
29	1	-1	1	-1	1	66.858	66.982
30	0	0	0	0	0	78.544	79.157
31	0	2	0	0	0	84.755	84.917
32	0	0	-2	0	0	75.576	75.157

Table 2: Response Surface Regression of Percentage Removal Efficiency of CV Dye

Term	Constant	SE	t	p
b0	3.488	17.1237	0.204	0.842
b_1	1.485	0.6653	2.232	0.047
b_2	3.374	1.1309	2.984	0.012
b_3	-0.448	0.1508	-2.968	0.013
b_4	134.227	14.2477	9.421	0.000
b_5	0.286	0.1192	2.396	0.035
$b_1 * b_1$	-0.037	0.0090	-4.091	0.002
$b_2 * b_2$	-0.198	0.0359	-5.511	0.000
$b_3 * b_3$	-0.001	0.0006	-1.259	0.234
$b_4 * b_4$	-38.065	5.6091	-6.786	0.000
$b_5 * b_5$	0.000	0.0004	0.013	0.989
$b_1 * b_2$	0.044	0.0243	1.791	0.101
$b_1 * b_3$	0.001	0.0032	0.243	0.812
$b_1 * b_4$	-0.350	0.3038	-1.154	0.273
$b_1 * b_5$	-0.002	0.0024	-0.889	0.393
$b_2 * b_3$	0.043	0.0065	6.666	0.000
$b_2 * b_4$	-1.142	0.6076	-1.880	0.087
$b_2 * b_5$	-0.014	0.0049	-2.781	0.018
$b_3 * b_4$	-0.082	0.0810	-1.013	0.333
$b_3 * b_5$	0.003	0.0006	5.118	0.000
$b_4 * b_5$	-0.559	0.0608	-9.193	0.000

Table 3: Analysis of Variance for Removal of CV Dye using CCD

Source	DF	SS	MS(Adj)	F	p
Linear	5	1230.90	165.907	33.1814	22.47
Square	5	121.32	121.322	24.2643	16.43
Interaction	10	255.19	255.191	25.5191	17.28
Residual Error	11	16.24	16.243	1.4766	.
Lack of Fit	6	2.42	2.417	0.4029	0.15
Pure Error	5	13.83	13.826	2.7652	
Total	31	1623.66			
S = 1.215		R-Sq = 99.0%		R-Sq(adj) = 97.2%	

The developed model was further analyzed by ANOVA. The results of ANOVA indicated that model can predict the results well (Table-3). Here, P values are used as a tool to check the significance of each of the coefficients, which in turn are necessary to understand the pattern of the mutual interactions between the test variables. The value of correlation coefficient (R= 0.998) indicates a high agreement between the experimental and predicted values. The value of determination coefficient ($R^2 = 0.997$) indicates that the response model can explain 99% of the total variations.

The experimental results were also trained and validated with artificial neural Network (ANN). The Multilayer Perceptron Neural Network (MLP 5-8-1) was designed and trained. The ANN was trained using standard back propagation architecture with BFGS training algorithm and this architecture used was comprised of two layers. Fig-2 shows the architecture of the network used. The tansigmod function was used as the transfer function in the hidden layer due to its suitable application for the data set of this kind. The output layer was made up of exponential transfer function. The optimal hidden layer was determined by varying the total number of neurons from 3 to 20.

The stop criteria were based on Sum of Square error (SOS) on the validation set for model generalization. The optimum hidden layer comprised of 8 neurons. The developed ANN with MLP 5-8-1 design gave 0.99870 and 0.99552 performances during training and testing stages which resulted the errors of 0.0001 and 0.00011 respectively at training and testing. Incremental training style is used where the weights and biases of the network are updated each time an input is presented to the network.

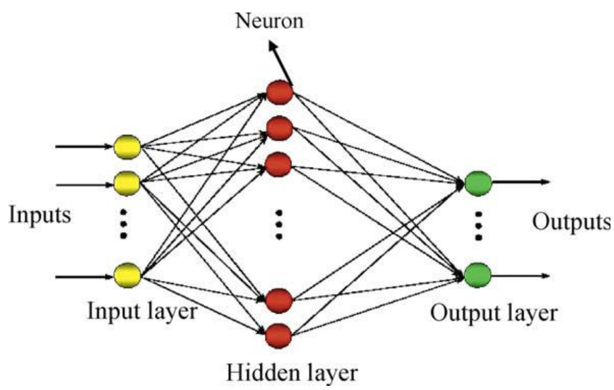

Figure 2: Multilayer Perceptron Neural Network

CONCLUSIONS

Effect of various parameters on dye removal process could be predicted well by both response surface method and ANN. Prediction ability of both the methods were very high (>99%) however, ANN gave more accurate results. The accuracy of prediction was significantly improved compared to statistical model. The neural network model developed could better predict the properties than the previously regression model. The network model had R = 0.999 which showed that the neural network model was capable of learning the relationships among the input and output variables for given data set.

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