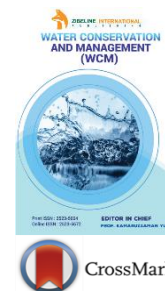


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## RESEARCH ARTICLE

# USE OF RESPONSE SURFACE METHODOLOGY AND ARTIFICIAL NEURAL NETWORK APPROACH FOR METHYLENE BLUE REMOVAL BY ADSORPTION ONTO WATER HYACINTH

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## ABSTRACT

The release of coloured effluents from various dyeing industries are of great concern due to the challenge involved in the treatment process. In present work, response surface methodology (RSM) and artificial neural network (ANN) were used to predict the color removal using adsorption process. Water hyacinth (WH) was used as an economical adsorbent for color removal from aqueous solution in a batch system. The individual effect of influential parameter viz. initial pH, MB (dye) concentration, and the adsorbent dose were studied using the central composite design of RSM. The RSM result was used as an input data along with final pH (non-controllable parameter) after adsorption to train the ANN model. Color removal of 96.649% was obtained experimentally at the optimized condition. A comparison between the experimental data and model results shows a high correlation coefficient ( $R^2_{RSM} = 0.99$  and  $R^2_{ANN} = 0.98$ ) and showed that the two models predicted MB removal indicating WH can be used as an adsorbent for color removal from dye wastewater.

## KEYWORDS

Water hyacinth, Adsorption, Methylene blue, RSM, ANN.

## 1. INTRODUCTION

Dyes are one of the important materials used in the industries like textile, plastic, leather, food and paper and effluents are characterised by high content of colour and organic content. The wide use of synthetic dye in textile industry has resulted into a major water pollutant. It is estimated that about 15% of dye material is lost during manufacturing and processing operation (Anwar et al., 2015) from industries like paper, plastic, rubber, leather, food, and textile (Chowdhury et al., 2011). Dyes are toxic, carcinogenic, non biodegradable and stay in the environment for a longer period. Its disposal into the water bodies causes undesirable change in water colour which hinders the penetration of sunlight and limit the photosynthetic activities of the aquatic life. Among different dyes classification cationic dyes are of highest toxicity. Methylene blue (MB), a cationic dye is used in the application of biology, chemistry, and dyeing industries. Its long term exposure leads to vomiting, nausea, eye burn, increase in heart rate, anemia, and hypertension (Foo & Hameed, 2012). Hence, the removal of MB from the effluents is need of the time and there is a need of an efficient method for removal before being discharged into the environment.

Various physicochemical and biological methods has been reported to be used by various researchers for treating coloured wastewater (Blanco et al., 2014; Monsef Khoshhesab et al., 2015; Mouni et al., 2018). Physicochemical methods involve process like coagulation, filtration, photodegradation, adsorption, and chemical oxidation. Recently, several

technologies have been utilized for removal of dye effluents and its intermediates from contaminated wastewater like adsorption (Yagub et al., 2014), biodegradation, membrane separation (Zhu et al., 2017) and photocatalyst (Blanco et al., 2014). Chemical coagulation is a very common method for treatment but generates huge amount of sludge which further needs to be treated, making it a very expensive method (Sala and Gutiérrez-Bouzán, 2012). Adsorption is extensively utilised for colour removal from dye wastewater due to its ease in operation and maintenance, highly effectiveness and low cost (Konicki et al., 2018).

Water hyacinth (WH) is an aquatic floating plant with prolific growth rate having capacity to double its biomass in 7-15 days. It is an invasive plant and abundantly available. Its widespread and rapid growth rate are of serious concern for its control. The floating mat of WH blocks the water surface interfering with the fishing, navigation, recreation and prevents the penetration of light into the water leading problems to the marine life. The present method of control of WH include extraction using mechanical extractor and dumping into the land. Much research has been conducted on for biogas production (Lu et al., 2009; Rai et al., 2011; Rodrigues et al., 2014; Álvarez et al., 2016). The disposal option of the water hyacinth is still a challenging task and need for a viable method. This has led to the usage of water hyacinth for various purposes. In this study, WH was selected for adsorption process as an adsorbent for removal of MB from wastewater. Previously, researchers have used materials such as fly ash and red mud (Wang et al., 2005), agricultural residue (Ibrahim et al., 2010).

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The traditional methods of modelling are costly, tedious, and do not represent effects of all variable process parameter. It does not take into the effect of all the interactions that are taking place between the process variables. The most common methods used to define the kinetics and equilibrium study are Langmuir and Freundlich isotherm, pseudo first and second order models (Aljeboree et al., 2017; Ji et al., 2019; Mohan et al., 2008; Tharaneedhar et al., 2017). These are insufficient to describe the connections between the operating parameters of the adsorption process. It includes varying one parameter at a time, making it time consuming, non-effective and costly. Hence, the results obtained cannot be validated accurately (Shojaeimehr et al., 2014). To overcome this problem, RSM has been employed by many researchers to optimize the conditions for colour removal from wastewater (Mourabet et al., 2017; Owolabi et al., 2018; Sadhukhan et al., 2016). It offers advantages in reduction of the number of experiments to be conducted, prediction of responses and is time-saving. It utilizes the polynomial equation for the independent variable parameter and analyses to obtain the optimum values to get a suitable response (Şahan et al., 2010).

The use of ANN has been widely used for prediction of responses of environmental problems due to its non-linear relationship between variables in the complex systems (Turan et al., 2011). This process is used where complexity is involved in the application. Some researchers have tried both RSM and ANN modelling to predict and evaluate the removal process such as disperse dye removal from water treatment residue (Gadekar and Ahammed, 2019), copper from simulated wastewater, dissolution of alumina from Azaraegbelu clay (Bhatti et al., 2011), biosorption process (Witek-Krowiak et al., 2014), light expanded clay aggregate for  $\text{Cu}^{2+}$  adsorption optimization (Shojaeimehr et al., 2014). Both RSM and ANN are used to predict, generalise and optimize the effectiveness of adsorbent in color removal from effluent. Therefore, the aim of this study was to study the possibility WH for adsorption of MB from coloured wastewater. Based on the results of adsorption process, mathematical modelling CCD and RSM was develop to find the correlations between the the initial pH, WH dose, and dye concentration. The result of RSM was further used for ANN modelling.

## 2. MATERIALS AND METHODS

### 2.1 Adsorbate

MB known as basic blue 9 ( $\text{C}_{16}\text{H}_{18}\text{N}_3\text{ClS}$ , CI = 52,015, MW:319.85 g/mol,  $\lambda_{\text{max}} = 665 \text{ nm}$ , structure shown in Figure S1) was used in this study for preparation of synthetic color wastewater without any additional purification. Deionized water was used for stock solution preparation using dissolving precise quantity of dye. Synthetic colour wastewater of different concentration for adsorption experiments were prepared in double-distilled water.

### 2.2 Preparation of Adsorbents

WH, a free-floating aquatic macrophytes, was collected from Tapi River, Surat, India. It was washed thoroughly under running tap water to remove undesirable components followed by deionized water wash. The adsorbent was initially sun-dried for 5-6 days (to remove excess water) thereafter hot air oven-dried at  $80 \pm 2 \text{ }^\circ\text{C}$  for 24 hours (till constant weight). Grinding of dried WH to fine powder followed by sieving to a uniform particle size of  $216 \mu\text{m}$  and stored in desiccator and used without any further modifications.

### 2.3 Characterization of Sorbent

Characteristics of WH (physical and chemical) was done using Scanning Electron Microscopy (SEM) (Hitachi-S3400N) and Fourier Transform Infrared (FTIR) (Shimadzu- FTIR-8400S) spectra to understand the surface properties. SEM were conducted to understand surface structure and characterization on the mechanism of adsorption of MB onto WH with an electron acceleration voltage of 15kV. Gold coating on the adsorbent was done with the help of sputter to make WH powder electrically conductive and prevent the electrostatic charge accumulation onto the adsorbent surface. Colour was measured using spectrophotometer

(Systronics-169) at maximum wavelength of 665 nm. Zero potential charge ( $\text{pH}_{\text{ZPC}}$ ) of water hyacinth was determined as per (Shirzad-Siboni et al., 2014): NaCl (0.01 M) solution of different pH (2-12) was prepared by adding HCl/NaCl. Then 50 ml of samples of 0.01M NaCl was prepared of different pH (2-12) followed by addition of 0.20 g of adsorbent in the reagent bottles. After 48 hours, samples were centrifuged and pH of solution was measured. Figure S2 shows the graph of  $\text{pH}_{\text{ZPC}}$  of water hyacinth.

### 2.4 Adsorption Experiments

The adsorption experiment was performed in 250 ml Erlenmeyer flask with different dose of adsorbent and 100 ml dye solution of known concentration. A predetermined quantity of adsorbent was added to the Erlenmeyer flask. The initial pH of dye solution was adjusted with 0.1 M HCl / NaOH. From equilibrium study, the equilibrium time of 90 min was calculated. The flask was held in orbital shaker (Eltek OD-4) at 200 rpm followed by 10 min of settling. The effect of influential parameters viz. initial pH (2-12), dye concentration (100-300 mg/L), and adsorbent dose (0.5-3 g/L) was studied. The final pH of the sample after adsorption was also measured. Filtrate thus obtained was measured at maximum absorbing wavelength 665 nm. The removal (%) was calculated using:

$$\text{Removal (\%)} = \frac{C_i - C_o}{C_i} \times 100 \quad (1)$$

Where  $C_i$  and  $C_o$  are MB dye concentration (mg/L) initial and after adsorption, respectively.

Experiments of adsorption were executed in triplicate and average values of the results were reported. Control experiments were also carried out in Erlenmeyer flask without adding adsorbent which established that adsorption was negligible on the wall of the flask.

### 2.5 Response Surface Methodology (RSM)

Design of experiments is mostly used to optimize the influencing process variables by improving the characteristics performance and reducing the errors of the experiments (Roosta et al., 2014). Several factors and their interaction between responses is evaluated by RSM. It consists of three steps namely designing the experiment, modelling of response surface and optimization of process variables. Three-level CCD was performed to evaluate the influences of initial variable parameters on the removal process (color) with total of 20 runs for the optimization process. The outcomes were fitted in the second-order quadratic equations. A total of 30 experiments were calculated using  $N = 2^n + 2n + n_c$  where  $N$  and  $n$  signifies number of experiment and factors respectively (Hameed et al., 2008).

The points used in experimental design comprise of  $2^n$  factorial points;  $2n$  axial points, and  $n_c$  central points and including their test results for the response variables. From centre points the experimental errors and reproducibility of data can be determined. Coding of independent variables were (+1) high and (-1) low levels. A  $\alpha$  represents the distance at which axial points are located to make design rotatable with fixed value of 1.682 in this study.  $(\pm \alpha, 0, 0)$ ,  $(0, \pm \alpha, 0)$  and  $(0, 0, \pm \alpha)$  are location of axial points.  $\alpha$  represents the axial point distance from the centre making the design rotatable. The independent values are shown in Table S1. The experimental data of three independent variables were analyzed and fitted into second-order polynomial model given by following equation (Hameed et al., 2008).

$$Y = b_0 + \sum_{i=1}^n b_i x_i + \left( \sum_{i=1}^n b_{ii} x_i \right)^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^n b_{ij} x_i x_j \quad (2)$$

where  $Y$  represents predicted response,  $b_0$  as constant coefficient,  $b_i$  is linear coefficients,  $b_{ij}$  is interaction coefficients,  $b_{ii}$  as quadratic coefficients and  $x_i, x_j$  are the coded values. In the present study, RSM was applied for modelling the process parameter for color removal efficiency by WH using three independent variables (initial pH, dye concentration, and WH dose). The results obtained after the analysis of RSM along with

final pH was utilized as input parameter for modelling the process using ANN.

## 2.6 Artificial Neural Network (ANN)

The ANN modelling was done with RSM-CCD data with additional data of final pH of solution after adsorption as shown in Table S2. A neural network tool (nntool) in MATLAB (R2017a), the MathWorks, Inc. was used in this study. It is used for nonlinear processes. It comprised of an input layer, a hidden layer and one or more than one hidden layer. The weighted sum of inputs arriving at each neuron is passed through activation function to get an output (Bhatti et al., 2011). The ANN structures are feed forward network trained from input using error back propagation algorithm. Input node takes independent variables and in return output nodes variables are dependent one. Hidden layers are used to perform nonlinear transformations function on the input which are further utilised for computation purpose. The model complexity increases with increase in the no of hidden layer. In this study neural network consisted of one input layer with four inputs, one hidden layer with 10 neurons and one output layer as shown in Figure S3. In this, 52 data sets of the experimental results were for modelling ANN network. The total data sets were divided as training (70%), validation (15%), and testing (15%). The performance was measured using mean square error (MSE).

Neuron transfer are used to transfer the input values to the next layers and strength of these connections are determined by weights. Modification on weights were performed to achieve minimum error between observed and predicted values for color removal efficiency. The validation of data was carried out till the error between the observed and predicted values reached to minimum. In present study, feed forward back propagation algorithm was selected with log sigmoidal (logsig) or tan sigmoid (tansig) function in hidden layer and output layer with linear transfer function (purelin) to achieve the best back propagation algorithm having minimum MSE and minimum relative error (MRE). The Levenberg–Marquardt (LM) and Gradient decent (GDA) back propagation algorithms were applied for training the model. The generalization ability of the observed values was assessed of the ANN model during the testing process. Further analysis of the trained network was carried out by linear regression between the observed and the predicted one.

## 3. RESULTS AND DISCUSSION

Removal of color from synthetic dye wastewater was optimized using variable parameter *viz.* initial pH, dye concentration and adsorbent dose. All the combinations of the variable parameters were used for modelling the ANN. Experiments were conducted as per the design of RSM and results were reported as observed results. It was then analysed for removal prediction from RSM software.

### 3.1 WH Characterization

FTIR analysis was used to determine functional groups characteristics present on WH before and after dye adsorption as shown in Figure 1. The broad spectrum at peak  $3330\text{ cm}^{-1}$  corresponds to O-H stretching vibrations of alcohols and carboxylic acid as in cellulose and lignin showing presence of free hydroxyl group on the adsorbent surface. Other peaks at 2918.28, 1619, 1414, 1321, 1025, and  $512\text{ cm}^{-1}$  corresponds to N-H stretching, C-N, C-C, C-O,  $>C=O$ , and S-S respectively. After the dye adsorption onto WH, the peak is shifted to 3332, 2914, 1640, 1428, 1319, 1035,  $541\text{ cm}^{-1}$  suggesting interaction of dye molecule with the functional groups. The presence of irregular and porous surface on water hyacinth indicates the possibility of adsorption as can be seen in Figure 2 (a) and therefore confirms its suitability for adsorption onto WH. It can be said that adequate surface is present on the WH for MB adsorption. After adsorption of MB dye (Figure 2 (b)), the surface of adsorbent became smooth showing the adsorption of dye onto the rough surface of the adsorbent.

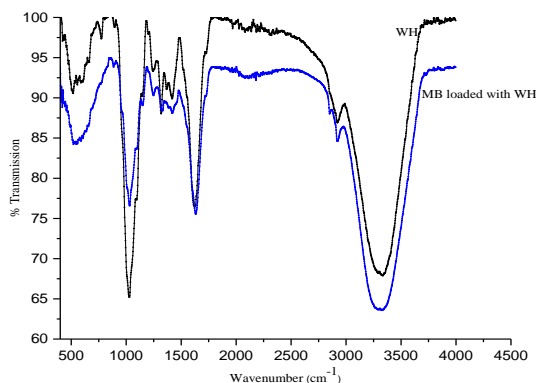


Figure 1: FTIR spectrum of water hyacinth

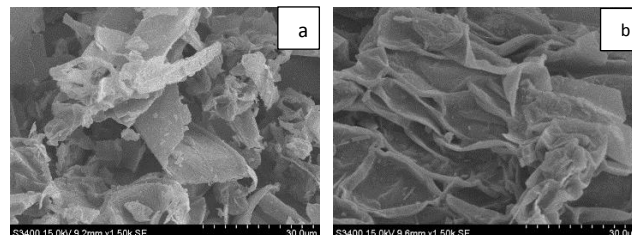


Figure 2: SEM image of WH before (a) and after adsorption (b) of MB dye

### 3.2 Effect of Operating Parameter

In this study, the effect of three parameters (initial pH, WH dose and dye concentration) was studied on the removal capacity of WH for MB removal through adsorption process. Experiments were conducted at different factors levels as per the design of Design Expert 11. The maximum color removal was obtained at higher dose of water hyacinth. Two-factor interaction (2FI) showed that colour removal was affected by two operating parameters. The presence of functional group -OH plays an important role in adsorption at higher dose of WH.

Figure 3 shows effect of initial pH and adsorbent dose on the color removal. It can be observed from the plot that with increase in pH and adsorbate dose, the color removal (%) increases. The maximum color removal was observed at pH of 10-12, adsorbent dose 3 g/L and dye concentration 300 mg/L as constant. Under acidic medium condition, removal was observed due to the fact of repulsion between the  $H^+$  ions present in the solution and in the water hyacinth. The point of zero charge for the adsorbent was 6.65. The maximum adsorption was observed at  $pH > pH_{ZPC}$ , because charge on the adsorbent surface changed to negative and there are probabilities of attraction between the cationic and anionic group present in the adsorbate. Similar results were reported by Roosta et al. (2014) for methylene blue removal by gold nanoparticle loaded on activated carbon.

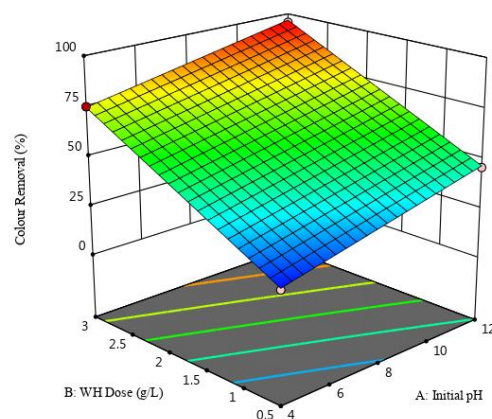


Figure 3: Interaction between water hyacinth dose and initial pH on the dye removal (at dye concentration of 300 mg/L)



Adsorbent dose plays a vital role in influencing the adsorption process by determining the adsorption capacity of the process under a given variable (Saha et al., 2010). Figure 4 shows the relation between the adsorbent dose and dye concentration of MB for adsorbent dose of 0.5-3 g/L. It can be seen that as dose of adsorbent increases, the removal also increases which can be attributed to the increase in the sorption surface area and more active adsorption sites available for adsorption of the dye molecule (Chowdhury and Saha, 2010)(Chowdhury et al., 2013). While color removal (%) increases, the adsorption capacity of the adsorbent decreases with increase in the adsorbent dose. This was attributed to the reduction in the total adsorption surface area and hence causes aggregations of adsorption sites (Chowdhury and Saha, 2010). Similar trend was observed by (Saeed et al., 2010) for adsorption of CV from grape fruit peel (Pathania et al., 2017) and for adsorption of MB onto activated Ficus carica bast.

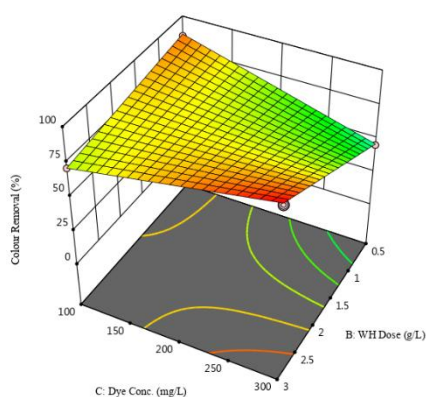


Figure 4: Interaction between water hyacinth dose and dye concentration on the dye removal (at initial pH of 12)

The rate of adsorption was also affected by the concentration of dye (Figure 5). The removal percentage decreases with increase in the dye concentration. This can be due to the saturation of adsorbent sites after a limit of number of active sites (Tsai and Chen, 2010). The dye concentration has contrary effect on the adsorption frequency (Marungrueng and Pavasant, 2007).

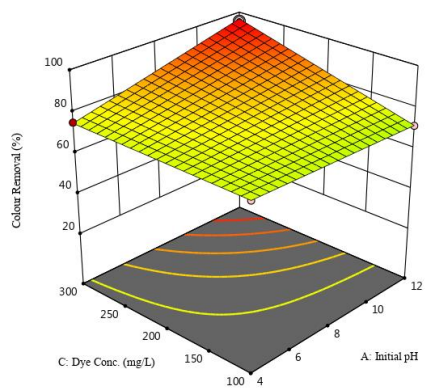


Figure 5: Interaction between water dye concentration and initial pH the dye removal (at adsorbent dose of 300 mg/L)

3.3 Modelling and Optimization using RSM

Different variables were selected to know the effect on colour removal using central composite design (CCD) were performed. Table S2 shows design matrix, experimental and predicted results. The second-order polynomial model was developed and analyzed from the observed data in terms of coded factor employing RSM. The two factor-interaction was suggested by the model as shown in Table S3. The two-factor interaction model (2FI) in terms of coded factors is expressed below to predict the response given as:

$$\text{Colour Removal (\%)} = 69.98 + 5.59 \times A + 9.28 \times B - 9.81 \times C - 0.6837 \times AB + 5.99 \times AC + 16.31 \times BC$$

Analysis of variance shows model to be significant as shown in Table 1

(ANOVA). The significance of the model can be seen from p-value (0.99) which shows that the model was significant and can be used for prediction of color removal from aqueous medium.

**Table 1: Analysis of variance (ANOVA) test results**

Source	Sum of Squares	Degree of freedom	Mean Square	F-value	p-value
Model	5154.50	6	859.08	506.45	< 0.0001 (significant)
A-Initial pH	381.16	1	381.16	224.70	< 0.0001
B-WH Dose	1038.54	1	1038.54	612.24	< 0.0001
C-Dye Conc.	1314.39	1	1314.39	774.86	< 0.0001
AB	3.74	1	3.74	2.20	0.1614
AC	287.40	1	287.40	169.43	< 0.0001
BC	2127.80	1	2127.80	1254.39	< 0.0001
Residual	22.05	13	1.70		
Lack of Fit	10.94	8	1.37	0.6152	0.7424 (not significant)
Pure Error	11.11	5	2.22		

R<sup>2</sup>: 0.99; Adjusted R<sup>2</sup>: 0.99; Predicted R<sup>2</sup>: 0.98; AP: 96.47

**Table 2: Design matrix with experimental and predicted values**

Sr. No.	A: pH	B: WH dose g/L	C: Dye conc. mg/L	Color removal (%)	
				Experimental	Predicted
1	1.2 (-1.682)	1.75 (0)	200 (0)	59.77	60.58
2	8 (0)	1.75 (0)	200 (0)	73.43	69.98
3	4 (-1)	3 (+1)	300 (+1)	75.22	74.86
4	8 (0)	0.35 (-1.118)	200 (0)	60.64	59.60
5	8 (0)	1.75 (0)	200 (0)	69.58	69.98
6	8 (0)	1.75 (0)	368.17 (1.682)	53.52	53.48
7	8 (0)	3.85(1.682)	200 (0)	85.32	85.58
8	8 (0)	1.75 (0)	31.82 (-1.682)	85.86	86.47
9	12 (+1)	0.5 (-1)	100 (-1)	85.77	87.09
10	12 (+1)	0.5 (-1)	300 (1)	45.98	46.84
11	8 (0)	1.75 (0)	200 (0)	69.61	69.98
12	4 (-1)	0.5 (-1)	300 (1)	21.54	22.31
13	8 (0)	1.75 (0)	200 (0)	69.66	69.98
14	12.7 (1.182)	1.75 (0)	200 (0)	78.22	76.58
15	12 (+1)	3 (+1)	300 (1)	95.45	96.65
16	4 (-1)	0.5 (-1)	100 (-1)	86.78	86.54
17	12 (+1)	3 (+1)	100 (-1)	71.48	71.66
18	8 (0)	1.75 (0)	200 (0)	70.20	69.98
19	8 (0)	1.75 (0)	200 (0)	70.16	69.98
20	4 (-1)	3 (+1)	100 (-1)	73.75	73.85

3.4 Optimization by RSM

Optimization of color removal was done using variables viz. WH dose, initial pH, and initial dye concentration. The favorable condition and its responses selected from the options provided to obtain optimum conditions. Desired goal was set to “maximise” and other parameters were set as “in range” as shown in Table S4. All other factors were given equal importance. A color removal of 96.649% was obtained with WH dose of 3 g/L, initial pH 12 and dye concentration 300 mg/L. To validate the predicted optimum condition, experiments were conducted at above optimize conditions and 93.86 % color removal efficiency was achieved as shown in Table S5. This shows that this model can be efficiently utilized for colour removal process. A statically significant model (P-value < 0.05) was developed using ANOVA, F-value (506.45) of the model implies the model was significant for the adsorption process.

The  $R^2$  Pred. (0.987) and  $R^2$  Adj. (0.993) values for MB biosorption capacity of water hyacinth biomass well satisfied the model. "Adequate precision" ratio of 96.474 showed an adequate signal for the model to be used for navigation in the design space. The  $p < 0.050$  indicate model terms are significant, whereas  $p > 0.100$  are not significant. In our study, all terms were significant except the interaction between the term AB i.e. initial pH and WH dose which had p-value of 0.161. Two-factor interaction was suggested by the model after using sequential model of square. Experimental responses are the responses measured for the specific run whereas predicted values are software assessed values using approximate functions of the model (Körbahti and Rauf, 2008). The adequacy of the model was confirmed by the responses obtained from experimental and predicted values.

### 3.5 ANN modelling

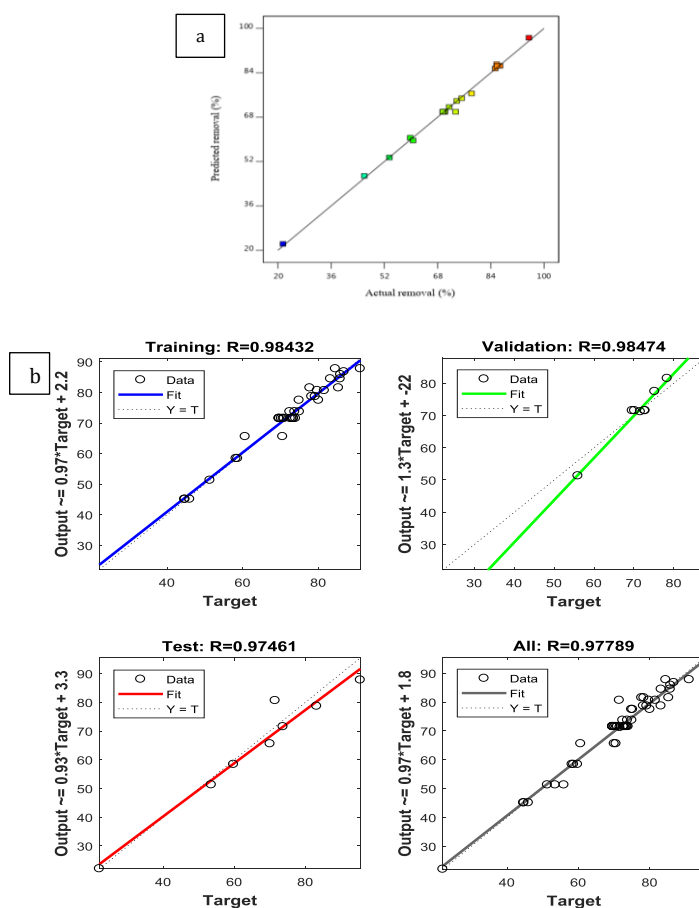
Artificial Neural Network was used to develop a mathematical model from the results obtained in RSM design. Artificial network was designed to train the neural network and to obtain the optimal number of neurons in hidden layer. Initial input data and results of central composite design were used to obtain the neural network model as show in Table 2. The original data sets with 50 data were divided into three sets viz. training, testing and validation of 70%, 15% and 15% respectively of the ANN model. The outliers were excluded from the data used for modelling in ANN. A feed forward network with three layers and back-propagation algorithm with log sigmoidal (logsig) or tan sigmoid (tansig) function in hidden layer including output layer with a linear transfer function (purelin) were developed. The no of hidden layer selected were 8 neurons (Dil et al., 2016). The Levenberg-Marquardt (LM) training function were

used for ANN model training. The modelling was found to be satisfactory with mean square error of 5.705 during the analysis phase. This signifies the accuracy of adsorption process prediction. The value of  $R^2$  (0.98) indicates excellent agreement between the experimental and predicted values of ANN.

### 3.6 RSM and ANN modelling

RSM and ANN are among the most widely used mathematical technique for adsorption study (Shojaimehr et al., 2014;Ohale et al., 2017). Both methods provide a good relationship between the influencing input parameters involved in the process and the result outcome due to its nonlinear relations between responses of the system and independent variables (Witek-Krowiak et al., 2014). It allows output prediction on the basis of inputs without defining the interrelationship between them. The validity of both models was assessed using same data points but in ANN the number of data points used were 52 excluding the outliers. The comparisons of results are shown in Figure 6, where experimental and model predicted results are compared. The importance of operating parameter as per ANOVA analysis were as shown in Table 1.

Hence, both models (RSM and ANN) provided a good prediction for dye removal in this study. RSM allowed prediction by giving regression equation for result predicted and the interactions between the operating parameters. It needs a standard experimental design for prediction of results whereas, ANN does not require any standard results for its predictions. Hence, both models provide a reliable way to interpret MB dye removal from aqueous solution onto water hyacinth.



**Figure 6:** Comparison of the experimental (line) with predicted data (symbols) using (a) RSM and (b) ANN model

## 4. CONCLUSION

Two modelling approach, ANN and RSM were used to model the colour removal using water hyacinth. RSM provides an aid in visualization of the result obtained in interactions between the parameters with 99% variance ( $R^2=0.99$ ). Both models provided good quality prediction for three

independent variables. However, ANN gave good results due to its non-linear relations between the input parameters. The optimal conditions for 95.45% colour removal were initial pH of 12, dye concentration of 300 mg/L, and WH dose of 3 g/L. The results of both RSM and ANN methodologies showed that RSM ( $R^2=0.99$ ) and ANN ( $R^2=0.977$ ) are good method of prediction of colour removal process by adsorption. In this

study RSM provided better removal efficiency and were in agreements with the experimental data. Feed forward BP with the Levenberg–Marquardt training algorithm was found best with highest R<sup>2</sup>. The present adsorbent study showed WH as an economical and effective adsorbent for removal of MB dye.

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