

ISSN 1996-3343

Asian Journal of  
**Applied**  
Sciences

## **Prediction of Adsorption Capacity of Microwave Assisted Activated Carbon for the Decolorization of Direct Blue 86 by using Artificial Neural Network**

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### **ABSTRACT**

A three-layer feed forward neural network was used to predict the rate of adsorption of textile dye direct blue 86 using microwave assisted activated carbon. In the present study, the Levenberg-Marquardt back-propagation algorithm was used to train the neural network at various experimental conditions. The pH, contact time, initial dye concentration, adsorbent dose and temperature were chosen as the input variables whereas, the dye uptake capacity was considered as the output variable. The tan sigmoid and linear transfer functions had been selected to train the hidden and output layer of the network, respectively. According to Levenberg-Marquardt back-propagation algorithm the optimum number of neurons was found to be eight. The predicted and experimental values of the desired output variables were in good agreement with each other with a correlation coefficient value of 0.982.

**Key words:** Adsorption, neurons, transfer function, back-propagation, Levenberg-Marquardt algorithm

### **INTRODUCTION**

Artificial Neural Networks (ANNs) have a wide application in the prediction of the rate of adsorption of an adsorbent in solid-liquid system. The ANN is an advanced mathematical tool which is used to determine the network output based on the available experimental information. The neural networks are made up of simple elements i.e., input layer, hidden layer and output layer, operating in parallel which are inspired by biological nervous systems. A typical neural network can be trained by adjusting the values of weights and bias between elements (Bhattacharya *et al.*, 2008; Sato *et al.*, 1999). The main purpose of the training was to make the experimental output reach as close as to the desired target output for a particular input. Till now various researchers used the ANN for predicting the performance of various adsorption systems successfully (Du *et al.*, 2007). Among all these adsorption process the adsorption of dye stuffs from different industrial effluents has become a major environmental concern (Dekhil *et al.*, 2011; Rajeswari *et al.*, 2011). Most of the dyes are non biodegradable in nature and produce toxic amines over incomplete degradation which has an adverse effect on the aquatic life (Rajendran *et al.*, 2011). These colored substances also retard the rate of photosynthesis by preventing the sunlight penetration into the water bodies (Wanchanthuek and Thapol, 2011; Arunachalam and Annadurai, 2011). Therefore, removal of these substances from the industrial wastewater is utmost

important (Wanchanthuek and Nunrung, 2011; Dutta and Basu, 2012). In most of the cases, the adsorption of dyes is carried out by using commercial activated carbon (Eckenfelder, 1989; Dutta and Basu, 2011). However, its higher cost limits its widespread uses. So far limited applications has been found on the removal of direct dyes from industrial waste water by using microwave assisted activated carbon.

Therefore, in the present investigation the adsorption capacity of microwave assisted activated carbon had been tested against a typical textile dye direct blue-86 (DB 86) through artificial neural network. For this purpose, a multilayer feed-forward neural network model was constructed which was found to predict the decolorization efficiency of DB 86 successfully through a non linear regression analysis.

## **MATERIALS AND METHODS**

**Materials:** The scrap wood of *Acacia auriculiformis* was collected from local saw mill of Kharagpur. Direct blue-86 (DB-86), hydrochloric acid, sulphuric acid and ammonia were procured from Merck Specialities Private Limited, Mumbai, India.

**Preparation of the activated carbon:** The wood scraps were first cut into small pieces of 2 mm width and 40 mm of length, cleaned with distilled water and was sun dried for 24 h prior to the carbonization process. The wood pieces were kept on a ceramic boat which was placed at the center of a 40 mm i.d tubular furnace. The material was then heated from ambient temperature to the carbonization temperature of 750°C at the rate of 4°C min<sup>-1</sup> in a continuous flow of N<sub>2</sub> (300 mL min<sup>-1</sup>) and then it was kept at this temperature for 1 h for subsequent activation. The product was then allowed to cool to ambient temperature in presence of N<sub>2</sub> flow and sieved to obtain the desired size fractions. It was further stored in a desiccator over silica gel. The char prepared in this way is termed as C750N.

The C750N was further activated in a domestic microwave oven (IF20PG3S) for 5 min at a constant input power of 800 W and a frequency of 2450 MHz. After treating in the microwave oven C750N was termed as AC750NMW5.

**Characterization of activated carbon:** The surface area and the total pore volume of the prepared char and the activated carbons were determined by using N<sub>2</sub> adsorption-desorption method by using Brunauer Emmett Teller (BET) apparatus (Autosorb-1, Quantacrome). The surface properties and the microporous structure were investigated by using Scanning Electron Microscope (SEM) (Hitachi, model SU-70).

**Batch adsorption study:** The effect of contact time and initial concentration on the amount of dye adsorbed were investigated at an initial dye concentration of 40-100 mg L<sup>-1</sup> whereas, the contact time was varied from 10-140 min. The pH effect was studied over a pH range of 1.5-9.4. The adsorbent dosage was varied from 1-3 g L<sup>-1</sup> in order to investigate the effect of adsorbent dosage on the instantaneous uptake of DB 86. The kinetic study was carried out by analyzing adsorptive uptake of the dye from aqueous solution at different time intervals. The samples were centrifuged and the concentrations were analyzed in a UV-Vis spectrophotometer.

**Equilibrium and kinetic study of DB-86:** The monolayer adsorption capacity of the prepared activated carbon was determined against direct blue-86 (DB-86) by the equilibrium study.

The equilibrium study was carried out by adding 0.1 g of adsorbent into a series of 250 mL conical flask containing 100 mL solution of direct blue 86 and was shaken for 36 h at room temperature. After this the samples were centrifuged and the concentrations were analyzed in a UV-Vis spectrophotometer. The equilibrium adsorption capacity was calculated from the relationship:

$$q_e = \frac{(C_0 - C_e) V}{w} \quad (1)$$

where,  $q_e$  ( $\text{mg g}^{-1}$ ) is the equilibrium adsorption capacity,  $C_e$  is the dye concentration at equilibrium ( $\text{mg L}^{-1}$ ),  $V$  is the volume of solution (l) and  $w$  is the weight of adsorbent (g). In order to establish an appropriate relationship between the adsorption capacity ( $q_e$ ) and dye concentration ( $C_e$ ) at equilibrium by using AC750NMW5, the data were fitted to Freundlich, Langmuir and Temkin isotherms. The Langmuir isotherm represents the uni-molecular adsorption of the adsorbate molecule on the adsorbent surface (Song *et al.*, 2010). The model can be expressed as:

$$\frac{1}{q_e} = \frac{1}{K_1 Q_0} + \frac{1}{Q_0} \quad (2)$$

where,  $K_1$  is the Langmuir constant related to the energy of adsorption ( $\text{L mg}^{-1}$ ) and  $Q_0$  is the maximum amount of adsorption corresponding to complete the monolayer coverage on surface ( $\text{mg g}^{-1}$ ).

Similarly, the Freundlich isotherm can be used for non-ideal sorption that involves heterogeneous surface energy systems (Ozturk and Kavak, 2005) and is expressed by the following equation:

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (3)$$

where,  $K_F$  is a rough indicator of the adsorption capacity and  $1/n$  is the adsorption intensity. In general, with the increase of the adsorption capacity  $K_F$  value increases. The Temkin isotherm describes the heat of adsorption and interaction between adsorbent-adsorbate molecules (Ergene *et al.*, 2009). The Temkin isotherm can be expressed as:

$$q_e = B_1 \ln k_T + B_1 \ln C_e \quad (4)$$

where,  $k_T$  and  $B_1$  are Temkin isotherm constant. The equilibrium plot of direct blue 86 is shown in Fig. 1 and values of different adsorption isotherm constants are also shown in Table 1. It was seen from Fig. 1 that the equilibrium behavior of Db 86 was successfully predicted by Langmuir adsorption isotherm. Besides, it was observed from Table 1 that the Langmuir adsorption isotherm gave the most satisfactory result for DB 86 adsorption with a higher correlation coefficient value ( $R^2$ ) of 0.98. The monolayer adsorption capacity was found to be  $500.16 \text{ mg g}^{-1}$  for an initial dye concentration of  $500 \text{ mg L}^{-1}$ . The chi-square value ( $\chi^2$ ) for different isotherms are also shown in Table 1. The chi-square value is a test statistic that is calculated as the sum of the squares of

observed values minus expected values divided by the expected values. Lower value of chi-square signifies more accurate system. It was observed from Table 1 that Langmuir isotherm had the lowest value of chi-square which signified a better fit.

**The artificial neural networks:** ANN is an advanced mathematical modeling procedure inspired by biological neuron system. In the present investigation, Neural Network Toolbox V5.0 of MATLAB R2006a mathematical software was used. A three-layer (one input layer, one hidden layer and one output layer) feed forward ANN network with tangent sigmoid transfer in the hidden layer linear function in the output layer was constructed for predicting the adsorption capacity of the prepared adsorbent. A typical ANN is shown in Fig. 2 where various input variables namely initial solution pH, initial adsorbate concentration, adsorbent concentration, reaction time and reaction temperature were fed to the input layer whereas, the dye adsorption capacity  $q_t$  ( $\text{mg g}^{-1}$ ) was considered as the output of the respective network. The ranges of different input variables are given in Table 2. Altogether 166 experimental data points were fed to the ANN network. The data points were divided for training, test and validation purpose in a ratio of 3:1:1. The training of the

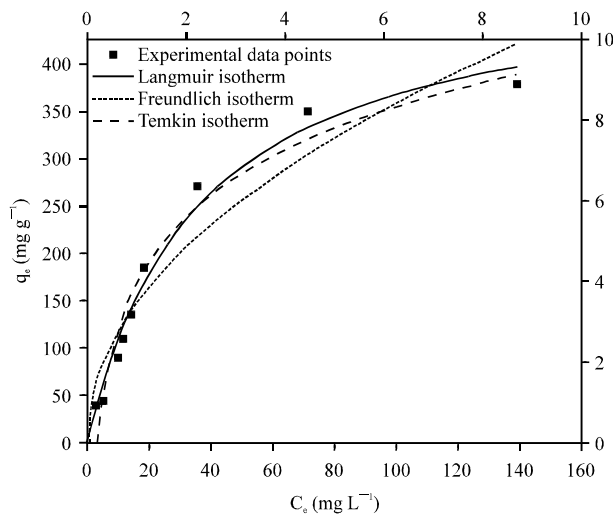


Fig. 1: The equilibrium curve for DB 86

Table 1: Equilibrium model parameters

Isotherm model	$R^2$	$\chi^2$	Kinetic parameters
Freundlich $K_F = 38.36$ ; $n = 2.06$	0.93	1372.66	$q_e = K_F C_e^{1/n}$
Langmuir $K_L = 0.03$ ; $Q_0 = 500.16$	0.98	295.00	$q_e = K_L q_m C_e / (1 + K_L C_e)$
Temkin $KM = 0.32$ ; $B = 102.59$	0.96	677.67	$q_e = K_m \log(hC_e)$

Table 2: The ranges of different input variables used for experimental run

Parameters	Range
pH	1.5-9.4
Contact time (min)	10-120
Initial concentration ( $\text{mg L}^{-1}$ )	30-100
Adsorbent dose ( $\text{g L}^{-1}$ )	1-3
Temperature ( $^{\circ}\text{C}$ )	10-50

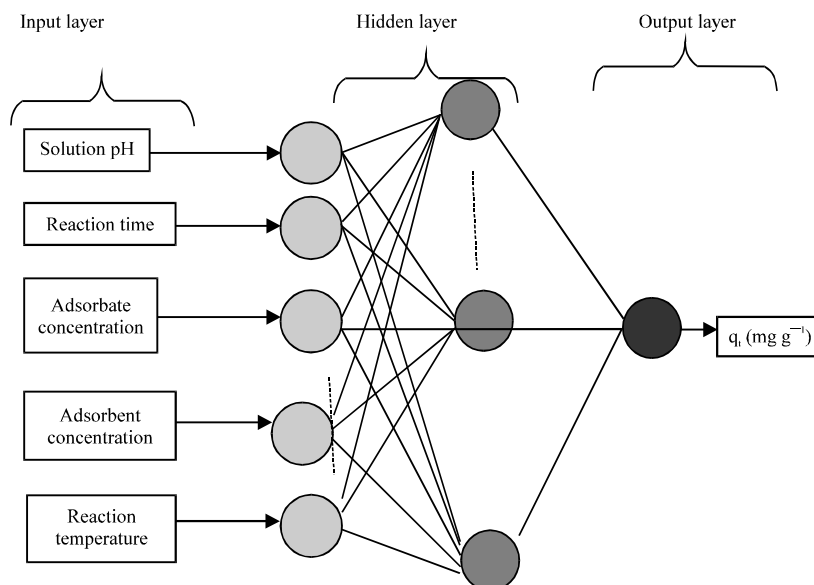


Fig. 2: Schematic diagram of the three layers ANN network with three input and one output layer

ANN network depends on the number of neurons in the hidden layer. The performance of the network can be developed by increasing the number of neurons in the hidden layer but too many neurons may affect the performance conversely in the over fitting (Kumar and Porkodi, 2009).

## RESULTS AND DISCUSSION

**Comparison of surface properties of activated carbons:** The BET surface area ( $S_{\text{BET}}$ ), total pore volume ( $V_{\text{tot}}$ ) and average pore size of C750N and AC750NMW5 were determined from the physical adsorption data of  $\text{N}_2$  at 77 K and the values are shown in Table 3.

It can be seen from Table 3 that AC750NMW5 has the highest surface area and total pore volume. Inside the microwave oven a high temperature could be reached in comparatively shorter period of time resulting dissipation of huge amount of energy at a molecular level. Consequently, the roughness of the pore walls may also be increased due to rapid heating with the formation of additional active sites (Huang *et al.*, 2011). Besides, rapid heating could accelerate the release of tar or volatile matter from the pore interior which results into higher pore volume (Liu *et al.*, 2010).

**Surface morphology of activated carbons:** The surface morphology of activated carbons was investigated through Scanning Electron Microscope (SEM) analysis. The SEM images of various activated carbons are shown in Fig. 3a and b. It was observed from Fig. 3a that a typical honeycomb structure with pores of different size was formed on the surface of char (C750N) when it was treated at optimum condition. The similar surface morphology could also be observed when activated carbon was prepared from corncob by chemical activation (Tseng, 2006). The roughness of the interior pores of activated carbon was increased when it was further treated in a domestic microwave oven (Fig. 3b).

**Optimization by using artificial neural network (ANN):** In the present work Levenberg-Marquardt back-propagation was implied to predict the rate of DB 86 adsorption. Trial and error

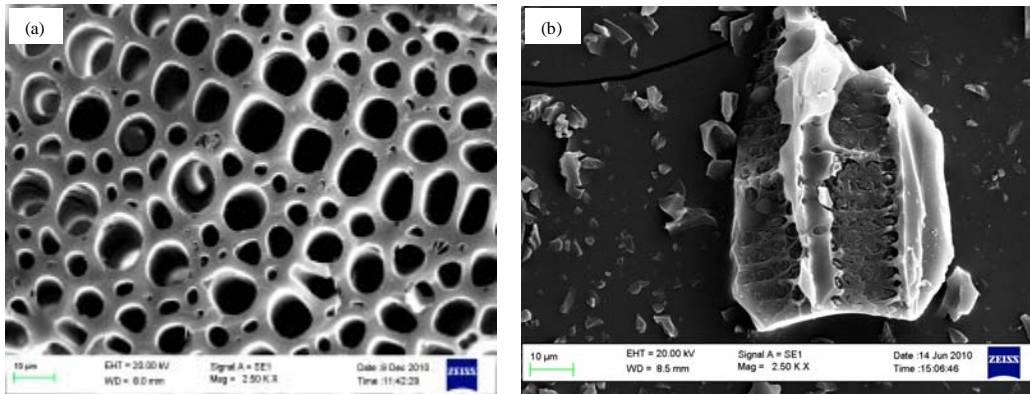


Fig. 3(a-b): The SEM image of (a) C750N and (b) AC750NMW5

Table 3: Comparison of surface properties of different adsorbents

Adsorbent	Surface area ( $\text{m}^2 \text{g}^{-1}$ )	Total pore volume ( $\text{cc/g}$ )	Average pore diameter ( $\text{\AA}$ )
C750N	514.2	0.36	27.99
AC750NMW5	695	0.50	28.55

Table 4: The Levenberg-Marquardt backpropagation of various algorithm

Algorithm	Function	NHL	$R^2_{\text{training}}$	$R^2_{\text{Val}}$	$R^2_{\text{test}}$	$R^2_{\text{All}}$	Iteration	LR	MSE
Levenberg-Marquardt backpropagation	Trainlm	5	0.979	0.990	0.985	0.982	28	0.03	0.0091
		8	0.981	0.976	0.993	0.982	11	0.02	0.0087
		10	0.991	0.987	0.936	0.992	51	0.03	0.0088

NHL: Neurons in the hidden layer, LR: Learning rate

method was followed to find out the optimum number of neurons in the hidden layer. It had been found that 8-12 neurons produce minimum Mean Square Error (MSE).

Therefore, 8-12 neurons were considered for this study. The Levenberg-Marquardt back-propagation algorithm along with its corresponding correlation coefficient values ( $R^2$ ) is shown in Table 4. The number of iterations required to find out the optimum number of neurons was also mentioned. The simulation result of “Levenberg-Marquardt back-propagation” algorithm is shown Fig. 4. It was seen from Fig. 4 that the network was successfully trained by using “Levenberg-Marquardt back-propagation” algorithm. The similar transfer function was also used by Oguz and Ersoy (2010) where removal of  $\text{Cu}^{2+}$  was carried out in a fixed bed column.

**Prediction of the adsorption capacity through ANN:** The actual and ANN model predicted values for adsorption capacity were compared through “Levenberg-Marquardt back-propagation algorithm” (Fig. 5). It was found that both of these values were in reasonable agreement with each other which is in commensurate with the result obtained by Aber *et al.* (2007). The values were compared for training, test and validation purpose. The values of corresponding correlation coefficient were also provided in the figures.

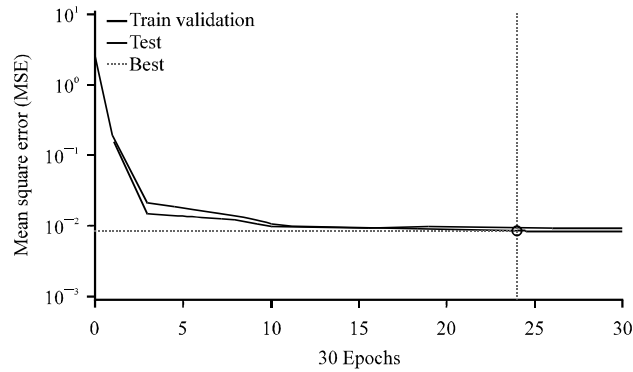


Fig. 4: Simulation curve for Levenberg-Marquardt backpropagation algorithm (best validation performance is 0.0085922 at epoch 24)

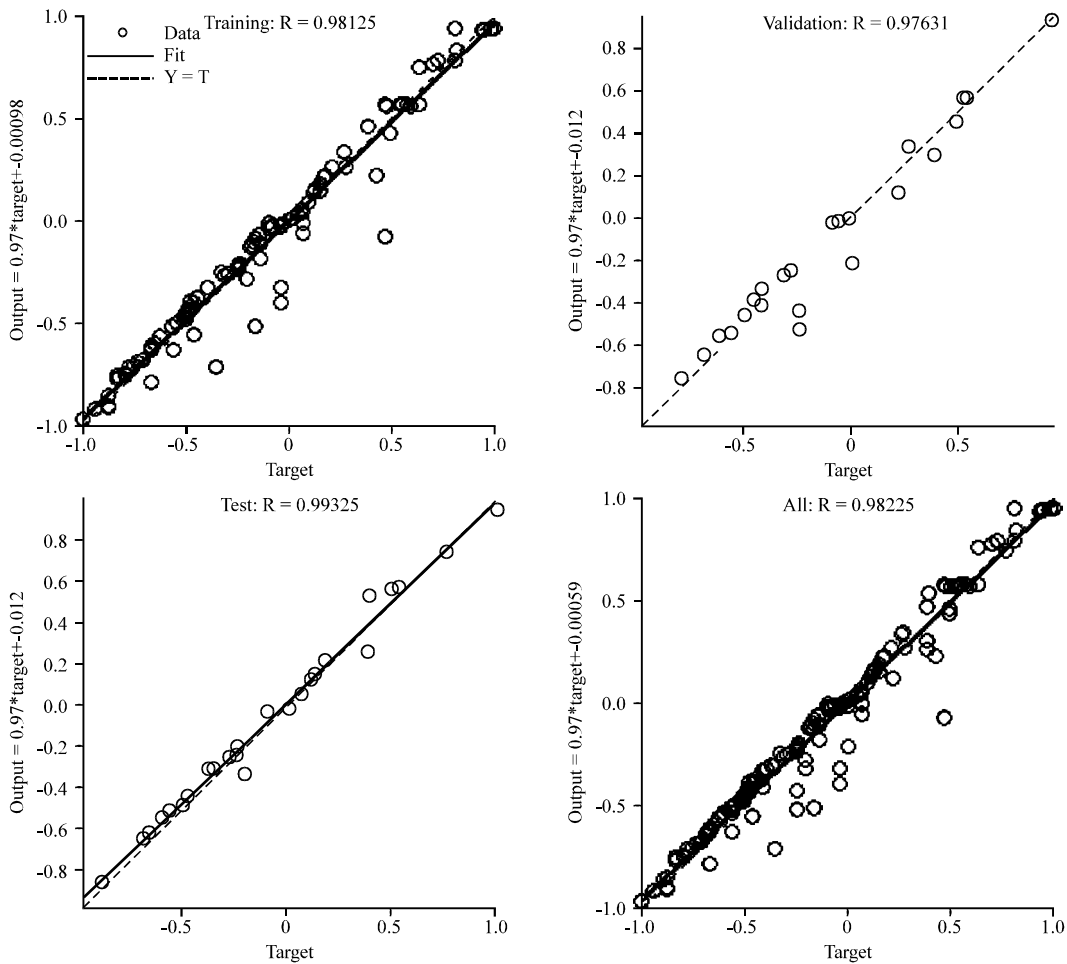


Fig. 5: Comparison of experimental and model predicted values of adsorption capacity by Levenberg-Marquardt back-propagation algorithm

**CONCLUSIONS**

In the present scenario ANN was successfully implemented for predicting the adsorption capacity in a solid-liquid adsorption system. The value of MSE was also calculated for



Levenberg-Marquardt algorithm. It was observed that 8 numbers of neurons in the hidden layer was required for the above algorithm. A reasonable agreement between the simulated results and experimental results was found with a correlation coefficient value of 0.982. Therefore, it can be said that ANN model could be used efficiently to run an automated water treatment process.

Pinaki Ghosh and Monal Dutta has conducted the experiments and Jayanta K Basu has developed the idea.

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