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Artificial Neural Network Based Prediction of Removal of Basic Dye from Its Aqueous Solution Using Activated Carbon

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Abstract - Management of solid wastes and liquid effluents are a great menace and has become a formidable social problem. Many countries including India find it difficult to have a control on it. Disposal of highly coloured water and municipal solid waste is a problem for the municipal planners. In the present study an attempt has been made to convert putrescible vegetable waste into phosphoric acid activated carbon (PAAC) for the removal of methyl violet from its aqueous solution. Characterization of the activated carbon was done using various physico chemical methods and characterized with FT-IR, SEM EDAX and BET surface area analysis. Batch mode adsorption study was employed for studying the efficiency of PAAC to remove methyl violet from its aqueous solution. To optimize the study, a high quality representative model called artificial neural network (ANN) was employed using feed forward back propagation algorithm. A comparison of experimental and ANN data was done for the effect of pH, adsorbent dosage, initial dye concentration and time. It was found that the developed ANN model predicted the efficient adsorption of methyl violet onto PAAC.
Keywords - Artificial neural network, Adsorption, Activated carbon, Surface area, Acidic dye, Basic dye.

I. INTRODUCTION

Due to rapid urbanization the amount of solid waste problems are also increasing. Solid waste generated from houses increased significantly in quantity and complexity with the advent of the throwaway society and the growth of the packaged and processed food industry. In spite of incurring huge expenditure, the services that are provided to the solid waste management are not fulfilling the requirement, causing public health hazards and nuisance. Hence, there is a strong need to develop appropriate technology for the proper management of urban as well as rural solid wastes. Due to massive Industrialization and urbanization the development in printing, dyeing and textile industries has increased multifold. The effluent released proves to be highly hazardous [1]. The untreated effluents cause drastic reduction in the quality of water in the receiving body since they possess strong colors, high pH and large amount of suspended solids, TDS, BOD and COD. In order to develop a solution to solid waste disposal problem as well as treatment of dye loaded effluent water a novel method has been generated for the synthesis of activated carbon using putrescible vegetable waste collected from market places, kitchens of home, hostels, hotels and also food processing units. Data analyzed were optimized using ANN modeling technique. ANNs are intelligent, thinking machines which work in the similar way as the human brain. They learn from experience in a way that no conventional computer can and they can rapidly solve hard computational problems. With the spread of computers, these models were simulated and later research was also directed at exploring the possibilities of using and improving them for performing specific tasks. In the last decade, research into ANNs has shown explosive growth. ANN modeling technique is employed in waste water treatment by several researches. Dynamic adsorption of crystal violet from aqueous solution using citric-acid-modified rice straw as adsorbent [2], estimation of lead concentration in grasses [3] prediction of the bulking phenomenon in wastewater treatment plants [4], prediction of membrane fouling during nano filtration of ground and surface water [5], assessing of ecosystem quality and community variations [6], prediction of optimum body diameter of air cyclones[7] and predicting single droplet collection efficiency of counter current spray towers [8] were analyzed using ANN modeling technique. Based on various studies, the ANN model proves to be the most appropriate modeling technique for describing the dynamic dye adsorption process. The present work describes the adsorption potential of methyl violet by phosphoric acid activated carbon (PAC) prepared from putrescible vegetable waste. The effects of various operational parameters such as adsorbent dosage, initial dye concentration, initial pH and contact time were also investigated. On the basis of batch adsorption experiments, the researchers proposed a three-layer ANN model using a back propagation (BP) algorithm to predict the methyl violet removal by

PAC as an adsorbent material. An optimization study was conducted to determine the optimal network structure and finally the outputs obtained from the ANN modeling were compared with the experimental data.

II. MATERIALS AND METHODS

A. Preparation Of The Adsorbent

Putrescible vegetable waste (PVW) has been collected from market places, kitchens of college hostels and marriage halls in and around Coimbatore city, Tamilnadu, India throughout the year 2010. During the one year period all types of vegetable wastes and their peel off could be collected. PVW was dried in sunlight to remove the moisture content. It was then dried in a hot air oven at 110^0C for 4-6 hours. The dried PVW to be carbonized is impregnated with a boiling solution of 30 % H_3PO_4 for 2 hours and soaked in the same solution for 24 hours. At the end of 24 hours, the excess solution decanted off and air dried and was carbonized in muffle furnace at 400°C. The dried material was powdered and activated in a muffle furnace at 800°C for a period of 10 minutes. Finally the activated carbon (PAC) was washed with plenty of water to remove residual acid, dried and powdered.

B. Adsorbate

The methyl violet (MV) was selected as the model compound having chemical formula $C_{23}H_{26}N_3Cl$ and λ_{max} 578nm. MV dye stock solution of 1000mg/L was prepared by dissolving it in double distilled water.

C. Batch Mode Adsorption Studies

The batch technique was selected because of its simplicity. The experiments were carried out in a mechanical shaker (KHAN shaker - KEMI make) working at a speed of 150rpm. MV solution of 50ml with desired concentrations and initial pH values were used. The adsorbent of 200mg were added to each flask and then the flasks were sealed to prevent any change in volume during the experiments. It was agitated for predetermined time intervals at room temperature in the mechanical shaker. After shaking, the adsorbent were separated by centrifugation and the supernatant solutions were estimated by measuring absorbance at maximum wavelengths using UV-Visible spectrophotometer (Model: JASCO V-570) at the desired wave length. The effect of each parameter like adsorbent dose, adsorbent particle size, different dye concentrations and agitation time were studied by fixing the values of other parameters.

The percentage of dye adsorbed by the adsorbent was calculated using the following equation.

$$\% \text{ removal} = \frac{C_0 - C_e}{C_0} \text{ X } 100$$

where C_o and C_e (mg/L) are the initial and equilibrium liquid-phase dye concentration.

D. ANN Model

For the study, Neural Network Toolbox V4.0 of MATLAB® mathematical software was used to predict the adsorption efficiency. Eighty five experimental sets were used to develop the ANN model. A three-layer ANN with tangent sigmoid transfer function (tansig) at hidden layer and a linear transfer function (purelin) at output layer was used. The data gathered from batch experiments was divided into input matrix [p] and target matrix [t]. Variables like adsorbent dosage, initial dye concentration, initial pH and contact time at a particular temperature were taken in the input layer (p). Adsorption efficiency (%) was taken in the output layer. The analysis was followed by the division of the original data into training, validation and test subsets. One fourth of the data was taken for the validation set, one fourth for the test set and one half for the training set. The experimental data was loaded into the workspace at random for each subset [9]. An optimization was carried out as an important task between the neuron number and mean square error (MSE) for the best back propagation (BP) algorithm. Finally some analysis of the network response was carried out. The entire data set was put through the network and a linear regression between the network outputs and the corresponding targets was performed.

III. RESULTS AND DISCUSSION

A. Characterization Of Activated Carbon

Physico chemical property of PAC is presented in Table 1. The data shows that PAC is having a greater BET surface area of 603.7m^2/g and Langmuir surface area of 2469m^2/g respectively. Total pore volume and average pore diameter for PAC were 0.7053cm^3/g and 2396 A^0 respectively. SEM EDX analysis of the activated carbon particles revealed the percentage of all the elements present (Table 1). SEM images of PAC are presented in Fig.1 and it shows that PAC surface is porous in nature. An FTIR spectrum of PAC is given in Fig.2. PAC displayed a broad band at 3429cm^{-1} was attributed to O-H stretching in hydroxyl functional

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groups [10]. The band at 2930cm^{-1} and its shoulder at 2859cm^{-1} were ascribed to C-H stretching and C-H bending absorption bands. A sharp peak at 1609 cm^{-1} resulting from C=C stretching vibration in aromatic ring enhanced by polar functional groups [11]. The broad peak at 1109 cm^{-1} in PAC can be assigned to the stretching vibrations of P=O groups and 669cm^{-1} due to C-P bonding. From the physico chemical analysis, surface area, SEM analysis and IR spectroscopy it is undoubtedly proved that the phosphoric acid treated carbon (PAC) is highly porous in nature with various functional groups and could be used for adsorption studies.

TABLE I
PHYSICO-CHEMICAL CHARACTERISTICS OF PAC

Parameters	Values	Parameters	Values
Moisture content (%)	4.5	pH	6.67
Ash content (%)	8.37	pH_{ZPC}	6.87
Water soluble content (%)	19.26	Electrical conductivity	0.151
Acid soluble content (%)	1.63	Yield (%)	76.48
Volatile matter (%)	20.5	Bulk density (g/ml)	0.388
Iodine number (mg/g)	1101.9	Specific gravity	0.98
Methylene blue number	525	Porosity	60.4
BET Surface area (m^2/g)	603.7	Carbon (%)	49.76
Total pore volume (cm^3/g)	0.7053	Oxygen (%)	37
Average pore diameter (A^0)	46.74	Phosphorus (%)	13.09
Langmuir surface area (m^2/g)	2469	Sulphur (%)	0.15

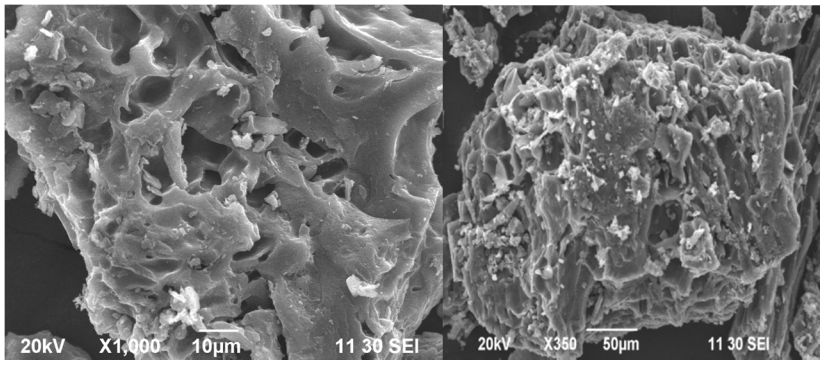

Fig. 1 SEM images of PAC

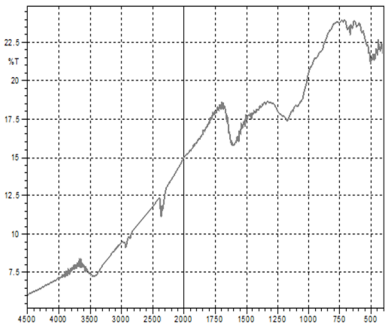
Fig. 2 FTIR spectrum of PAC

B. Optimization Of The ANN Structure

A typical architecture of a three-layered feed forward back propagation hierarchical network is shown in Fig.3. The network was trained automatically with the MATLAB function 'train' with the 'weights' and 'biases' initialized to random values. Before training, the data set was divided randomly into training and test data set. Data sets of 87 were used as training set, while 10 data sets were used for testing the network. In order to determine the optimum network generalization, Levenberg–Marquard algorithm

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was used. During training, the 'weights' and 'biases' of the network are adjusted so as to minimize the mean square error between the expected and the predicted values. The training can be terminated when the mean square error is less than 0.001 or when the number of iterations is equal to 1000 [12].

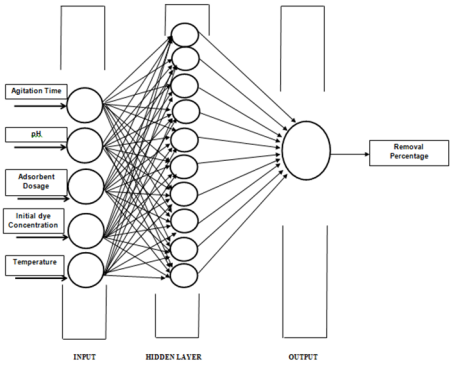

Fig.3 Network structure (5-10-1) used for prediction

The correlation coefficient between the predicted and experimental values using the entire data set for adsorption efficiency is in the range 0.9656 to 0.9895. It is a good sign for the model to be accurate. A perfect prediction would see all the plotted points sitting on the central line (the solid line), the accuracy of the model can be easily compared by the closeness of the data clusters to this line [13]. The best linear fit is indicated by a dashed line. It can be inferred from Fig. 3 that most of the values fall closer to the central line which proved the model to be accurate. A plot of the training errors, validation errors, and test errors for percentage removal is shown in Fig. 4. The results observed were reasonable because of the following considerations: the final mean square error was small. The test set error and the validations set error had similar characteristics. No significant over fitting occurred by iteration (where the best validation performance occurs).

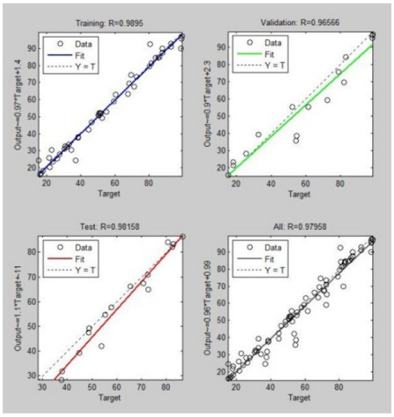

Fig. 4 Correlation between predicted and experimental values for adsorption efficiency

C. Comparison Of ANN And Experimental Values

On comparing the experimental values with ANN values, it was found that the neural network predictions were in good agreement with the experimental data. Figure 5 show the agreement between ANN outputs and experimental data as a function of adsorbate concentration, contact time at 100 mg/L, pH and adsorbent dosage. It was thus concluded that by increasing the concentration of methyl violet the removal percentage decreased [9] and as the contact time increased the removal percentage increased [14]. At pH 5 the removal of methyl violet by PAC was higher and as the adsorbent dosage increased the removal of methyl violet also increased. It was observed that the predicted (ANNs) and experimental values were fairly following the similar trend for the removal of methyl violet by PAC. Figure 6 show the mean square plot for measuring the adsorption efficiency of methyl violet onto PAC.

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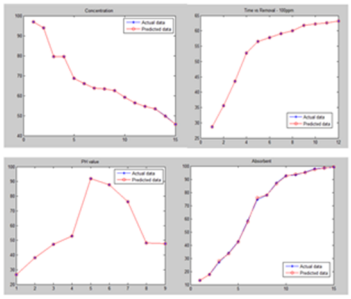

Fig.5 Comparison of ANN and experimental data for the adsorption of methyl violet onto PAC

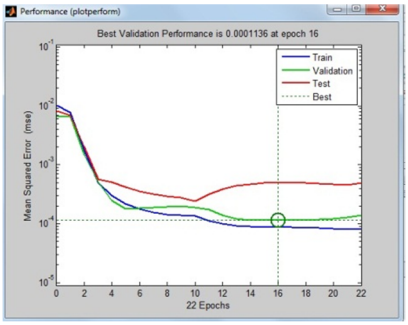

Fig.6 Mean square error plot for adsorption efficiency of methyl violet onto PAC

IV. CONCLUSIONS

The present study utilized a low-cost adsorbent, an activated carbon prepared from putrescible vegetable waste treated with phosphoric acid.

Physico-chemical properties of the activated carbon were analyzed and it showed that it has good adsorption properties.

BET analysis concluded a better surface area of 603.7m^2/g.

High porosity was observed by SEM analysis and its pore characteristics.

FT-IR proved the functional groups and XRD decided it to be amorphous in nature.

Artificial Neural Network optimization was done and various parameters like effect of pH, adsorbent dosage, concentration and time were studied. It has proved that the experimental and ANN data were in good agreement with each other.

The correlation coefficient between the predicted and experimental values using the entire data set for adsorption efficiency is in the range 0.9656 to 0.9895.

Mean Square Error value of 0.0001166 at Epoch 16 showed the best validation performance.

From all the above data it could be concluded that a simulation based on the ANN model can provide a further contribution to develop a better understanding of the dynamic behavior of process where still phenomena cannot be explained in all detail.

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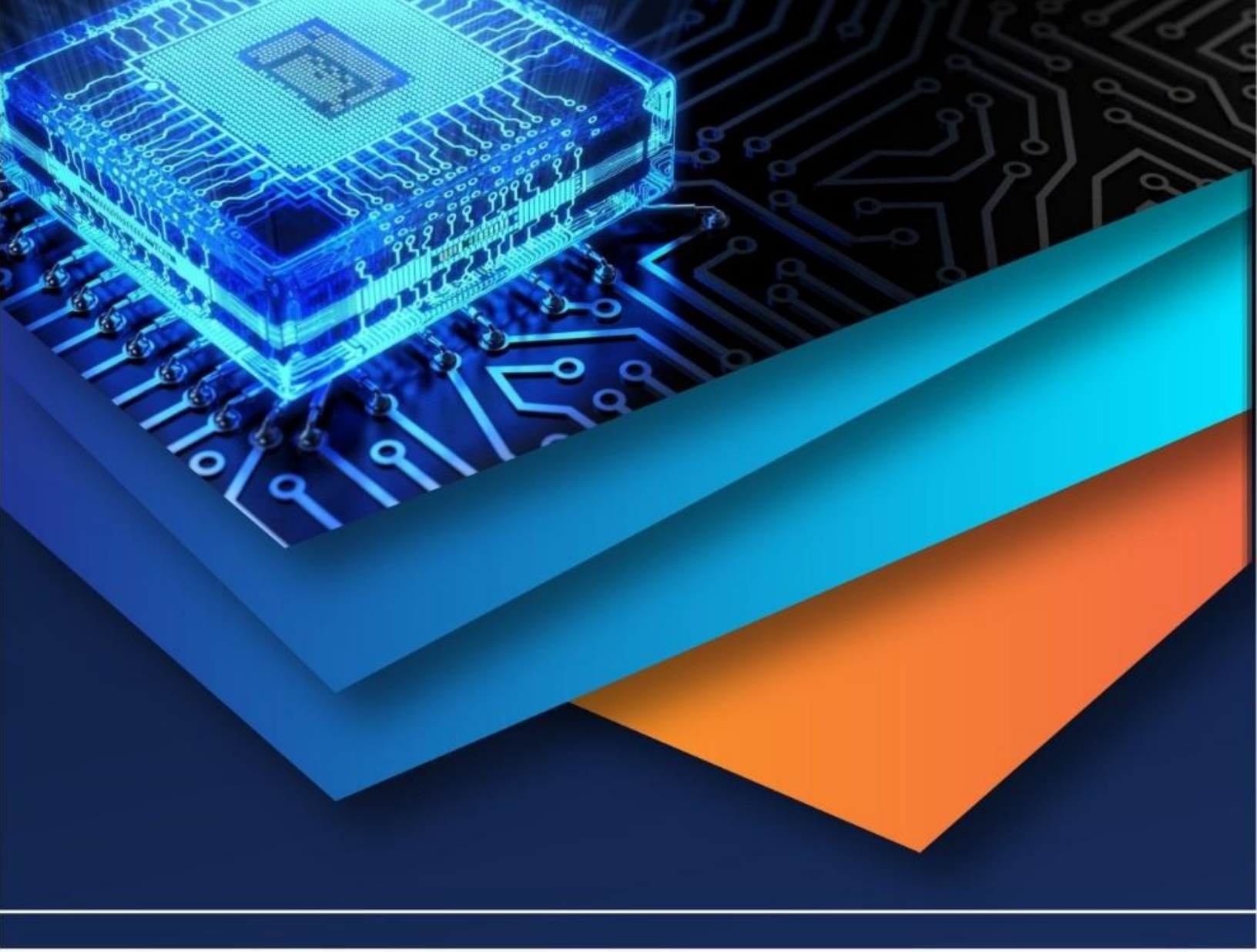

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