Optimization of RB5 Effluent Decolorization Efficiency Using KCT Composites - Statistical Approach with ANN Modeling

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ABSTRACT: *Calcined Kaolin / Calcium carbonate / TiO_2 (KCT) composites were prepared using 1:1:1 and 1:1:2 proportions at 1000 °C calcination temperature. The efficiency of KCT composites on the decolorization of Reactive Black 5 (RB5) effluent was investigated based on the composite concentration, pH, and time of treatment. Box and Behnken experimental design was used to optimize the decolorization efficiency of RB5 using KCT composites, followed by modeling of the treatment using an artificial neural network. Optimized parameters based on the Box and Behnken design for 1:1:1 KCT composites resulted in a decolorization efficiency of 94.5 %, using 20 g/L composites treated at pH 3 for a treatment duration of 3 hours 48 minutes. Similarly, for 1:1:2 KCT composites, 94 % decolorization efficiency of RB5 effluent has been achieved using 18.9 g/L composite treated at pH 3.5 for a treatment duration of 3 hours 56 minutes. Experimental results and the predicted results show close conformity in decolorizing RB5 effluents using KCT composites. Physico-chemical treatment of dyes using KCT composites was found to be efficient due to the formation of calcium silicate and calcium titanate, resulting in a strong photocatalytic adsorbent, leading to physical sorption and photochemical oxidation.*

KEYWORDS: *Composite, Kaolin, Decolorization, Optimization, Modeling Physico-Chemical Oxidation.*

INTRODUCTION

The rising use of textiles globally has never put a rest on these industries [1] and the global textile market is expected to grow by US$ 549.9 billion from 2021 to 2025. According to the claims made by the World Health Organization, these industries contribute around 17-20 % of the total pollution. Estimations show ~ 80% of dyes used for the applications of textiles are azo dyes. Though most of the dyes have good affinity towards the textile materials around 15 – 20 % of dyes are expected to wash out of textile on usage adding to pollution load [2]. Dyes have also found to release toxic

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amines putting water bodies at high risk. Thus treatment of dye effluents has been made as one of the important effluent treatment processes over years [3]. There have been physical, chemical, physico-chemical, biological and electrochemical methods of treating dye effluents [4].

Physical treatment methods include membrane filtration and adsorbents for physical entrapment of effluents [1], however membrane and adsorbents are costlier and disposal of the same after usage seems to be the most challenging task. Chemical treatment methods utilize various chemical interactions using coagulants, flocculants, Fenton's reagent and oxidizing agents. These chemical reagents generate huge amount of useless sludge [5]. Decolorization of dyes by using aerobic and anaerobic microorganisms breaks the azo groups in dye, however, it results in aromatic amine byproduct which could be a threat to the environment [1]. Electrochemical methods of dye removal efficiently decolourises dyes by electro coagulation and anode interactions. Compared to other methods discussed above, electrochemical interactions with dye effluents produce less sludge, however end up in the formation of unavoidable toxic compounds [5,6]. Recent studies on dye effluents using Advanced Oxidation Processes (AOPs) have provided certain promising outcomes in dye effluent treatments [7]. AOPs have capabilities of oxidizing organic and inorganic compounds by generation of strong oxidizing agents and hydroxyl radical (•OH). UV-Photolysis AOPs using Titanium dioxide (TiO_2) produces hydroxyl radical (•OH) by UV irradiation and is found to be popular among AOPs due to its cost, non-toxic nature and commercial availability. Studies show that the quantum yield of TiO_2 responsible for oxidation and reduction of contaminants are low and thus simple modifications can aid in improving its performance [8].

TiO_2 in its natural and modified nano forms have been interacted with clay minerals by electrostatic interaction, complexation of clay surfaces and coordination of ionic species on the clay surfaces [9]. Synthesis of TiO_2 – Zeolite photocatalyst composite prepared using solvothermal method has effetively decolourised Methylene Blue with 99% efficiency [10]. Similarly synthesis of TiO_2-Fe_3O_4-Bentonite clay prepared by sol gel method have been found to be effective in Decolorization of Basic Blue 41 [11]. Though most of the recent study is emphasized on the use of nano TiO_2 for the formation of composites, its use has been highly criticized for its carcinogenic, genotoxic and photosensitization behavior by researchers [12]. On the other hand TiO_2 in its natural form is found to be non-toxic and safe to the environment.

Usage of statistical tools in determining the efficiency of Decolorization percentage of dyes has been a key to understand various influencing factors [13]. Design of experiments like Taguchi, Box Behnken, central composite designs have been used for such optimization processes [14,15]. ANN modeling is known for its optimization and precise predictions of the experimental data with better reproducibility [16]. ANN has the capability of learning from a set of training data and effectively provides its corresponding outputs [17–19].

The major objective of the present study is to investigate the physico-chemical oxidation efficiency of the prepared KCT composites of proportions 1:1:1 and 1:1:2 on Decolorization of RB5 effluents by UV-Photolysis AOP. Box and Behnken design of experiments is used to optimize the variable parameters (composite concentration, pH and time of treatment) along with Response Surface Modeling (RSM). ANN model, is used to predict Decolorization the efficiency using four input parameter namely, composite concentration, pH, treatment time and composite proportion by varying the network parameters namely, learning rate, momentum constant, number of hidden layers, number of nodes in each hidden layers and number of cycles.

EXPERIMENTAL SECTION

Reactive dye

Jackzol Black BN 150 with the C.I name Reactive Black 5 and C.I Number 20505 was supplied by Jay Chemical Industries Limited, Gujarat, India and was used for the study without any further modifications [20].

Chemicals

Laboratory grade chemicals were used in this study. Kaolin was purchased from Loba Chemie Pvt Ltd, India, Calcium Carbonate ($CaCO_3$) was purchased from S D Fine Chem Limited, India and Titanium Dioxide (TiO_2) was purchased from Merck, India.

Preparation of dye effluent

Dyeing of cotton was carried with Reactive Black 5 using the recipe shown in Table 1. The dye effluent after dyeing were collected for further studies [20] .

Table 1: Dye Recipe.

Reactive Black 5	2% (owf*)
Sodium chloride	40 g/L
Sodium carbonate	15 g/L
Temperature	70°C
Time	60 minutes
M:L ratio**	1:40

**owf – on weight of fabric;*
***M:L ratio – Material-to-liquor ratio*

KCT preparation

Calcination of Kaolin ($2Al_2Si_2O_5(OH)_4$)/Calcium carbonate ($CaCO_3$)/Titanium dioxide(TiO_2) (KCT) composite at proportions 1:1:1 and 1:1:2 respectively was carried out using a muffle furnace. The samples were calcined at 1000 °C with a heating rate of 10 °C per minute and were held at final temperature for a period of 1 hour in an air environment. These proportions and calcinations temperatures were chosen based on the conditions required for the formation of calcium silicate (Ca_2O_4Si) and calcium titanate (CaO_3Ti) [21,22] that contributes physical sorption and photochemical oxidation respectively [23] and its thermal transition is shown in equation [1 - 6].

$$CaCO_3 \xrightarrow{\Delta} CaO + CO_2 \uparrow \quad (1)$$

$$Al_2SiO_5(OH)_4 \xrightarrow{\Delta} Al_2SiO_7 + 4H_2O \uparrow \quad (2)$$

$$TiO_2 \xrightarrow{\Delta} TiO_2 \quad (3)$$

$$2Al_2SiO_7 \xrightarrow{\Delta\,1000°C} Si_3Al_4O_{12} + SiO_2 \quad (4)$$

$$3Si_3Al_4O_{12} \xrightarrow{\Delta\,1000°C} 2Si_3Al_6O_{13} + 5SiO_2 \quad (5)$$

$$3CaO + SiO_2 + TiO_2 \xrightarrow{\Delta\,1000°C} Ca_2O_4Si + CaO_3Ti \quad (6)$$

Decolorization analysis

The prepared composites were cooled and stored until further usage. Dye effluents were treated using the composites in a UV chamber (Figure 1) and the absorbance was measured using UV-Vis Spectrophotometer (Shimadzu UV 1700), capable of scanning the samples in wavelength range 190 - 700 nm. Decolorization (%) of the sample was expressed as the ratio of difference between initial absorbance [I_0] and final absorbance at time t [F_t] to the initial absorbance (*David, Arivazhagan, et al*., 2015)

Table 2: Coded and actual values of experimental design.

Coded Variable	Description of the variable	Coded and Actual Values		
		-1	0	+1
X_1	Concentration of Composite (g/l)	10	15	20
X_2	pH	3	5	7
X_3	Time (Hours)	2	3	4

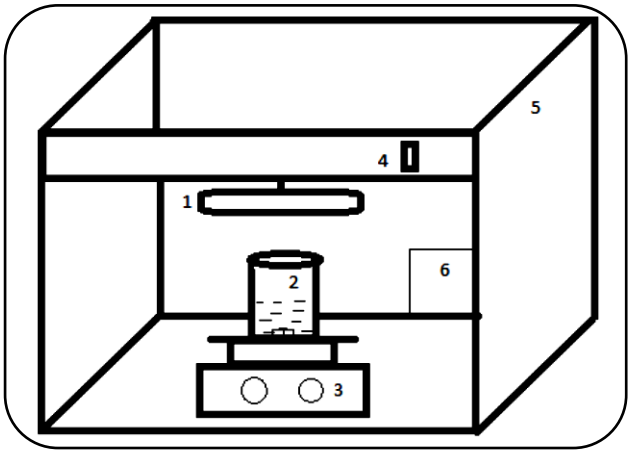

Fig. 1: UV Chamber for Effluent Treatment (1) UV Light, (2) Beaker (3) Magnetic stirrer (4) Switch controls (5) wooden box (6) Exhaust arrangement.

As shown equation [7]. The initial absorbance of dye before Decolorization treatment and the final absorbance of dye after treatment were measured at λ_{max} of 597 nm [24].

$$Decolourisation\ (\%) = \frac{I_0 - F_t}{I_0} \times 100 \quad [7]$$

FT-IR analysis

Degradation of dye with respect to pH was studied using Fourier-transforms infrared analysis. FT-IR was studied between 650 – 4000 cm^{-1} with 32 scans per minute and resolution of 1 cm^{-1} [25].

Response surface method

Box and Behnken design of experimental design meant for three factors and three levels was used to analyze the effect of composite concentration, pH and time on Decolorization efficiency. The coded and actual values of experiment are shown in Table 2.

Generic form of second order polynomial equation for 3 factor design is given by equation [8] (*Tayeb, Tony, et al*., 2018).

$$\gamma = \beta_0 + \sum \beta_i X_i + \sum \beta_{ij} X_i^2 + \sum\sum \beta_{ij} X_i X_j \quad [8]$$

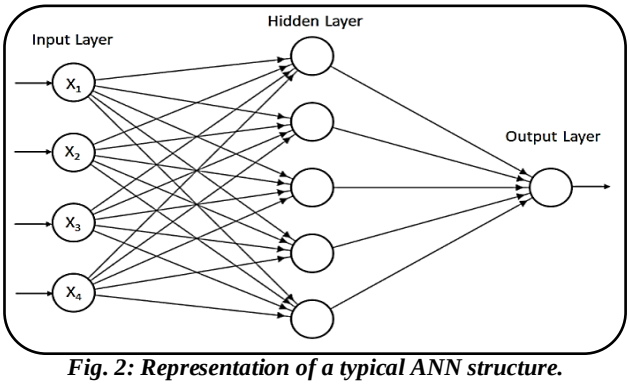

Fig. 2: Representation of a typical ANN structure.

Where γ being the predicted response (Decolorization %), $\beta_0, \beta_i, \beta_{ii}$ and β_{ij} are the model regression coefficient parameter and X_i is the input controlling code variable. Design Expert software (Version 7.0.0) was used to analyze the data [26,27].

Artificial Neural Network (ANN)

ANN develops a model that aids in understanding the relation between input and output parameters. It consists of Input layer, output layer and hidden layer(s). These layers consist of nodes that act as computational elements. Experimental design parameters decide the number of nodes in input layer and output layer, whereas the nodes in hidden layer vary based on network optimization and performance. Signals received from previous layers by each node are multiplied by weights known as synaptic weight. These inputs of synaptic weights are summed up and fed as the input to the successive layer using transfer function. ANN models were developed and studied using EASYNN Software. Mostly Physico-chemical process modeling uses back propagation method with feed forward neural network [28]. Thus, determination of weights is done by training neural network using back propagation algorithm. A typical network structure is shown in Figure 2, which shows input layers (X_1, X_2, X_3 and X_4), hidden layers and output layer. Input vector with the help of random weights is used by the network to produce output vector, compares the predicted output with the desired output vector and thus calculates the error vector.

The error vector is calculated using equation [9] where, E denotes Error vector, T is the target output vector, Out_i is predicted output vector and n denotes training patterns [29].

$$E = \frac{1}{2}\sum_{i=1}^{n}(T_i - Out_i)^2 \qquad [9]$$

Backward propagation algorithm propagates the error signal and synaptic weights are adjusted to achieve minimum possible error for the particular network parameter combinations. Changing of weights for input-output pair after the presentation is given by equation [10].

$$\Delta W_{ij} = \eta\left[\frac{\delta E}{\delta W_{ij}}\right] \qquad [10]$$

Where ΔW_{ij} denotes the correction applied to synaptic weights W_{ij} that connects the ith node present in the hidden layer and jth output node. Learning rate is denoted by η and is a constant. Sigmoid transfer functions has been used in this network [29].

Optimization of ANN parameters

Prediction performance of network is highly influenced by the selection and optimization of parameters used for the study. The present study uses four input parameters namely, composite concentration (X_1), pH (X_2), treatment time (X_3) and composite proportions (X_4). The independent network was trained to predict the Decolorization percentage of RB5 effluents. In order to predict the Decolorization percentage of RB5, one hidden layer with five nodes were used, as one hidden layer itself was capable of handling nonlinearity. Adaptive learning and momentum constant are used to predict the performance, as necessary parameter changes are controlled by the program itself to train the network. Learning rate and momentum constant were optimized at 1 and 0.8 respectively. Training was stopped at 769 iterations for Decolorization efficiency percentage. The dataset used in ANN training were divided into two subsets in which 86.66 % of data was used to train the network and 13.33% of data was used for validation. The mean square error (MSE) and mean absolute error percentage (MAEP) used in ANN prediction is given by equation [11] and [12] respectively.

$$MSE = \frac{1}{n}\sum_{i=1}^{n}(Y_e - Y_p)^2 \qquad [11]$$

$$MAEP = \frac{1}{n}\sum_{i=1}^{n}\frac{|(Y_e - Y_p)|}{Y_e} \times 100 \qquad [12]$$

Where, Y_e is the experimental values, Y_p is the predicted values obtained from ANN and n is the number of experiences. MSE determines the relation between the hidden layer neurons and network error or ANN model [28].

Table 3: Experimental and predicted values of composite on RB5.

S.No	Composite concentration (g/L)	pH	Time (Hour)	Composite Proportion	Decolorization Efficiency (%)	
					Experimental	Predicted by ANN
1	10	3	3	1	72.35 (0.57)	72.24
2	20	3	3	1	93.54 (0.68)	92.70
3	10	7	3	1	64.43 (1.53)	64.55
4	20	7	3	1	87.10 (1.03)	87.15
5	10	5	2	1	58.52 (1.12)	59.73
6	20	5	2	1	76.34 (0.69)	76.66
7	10	5	4	1	73.39 (0.68)	73.78
8	20	5	4	1	93.87 (0.19)	93.35
9	15	3	2	1	69.67 (0.62)	69.85
10	15	7	2	1	63.66 (0.60)	61.24
11	15	3	4	1	86.28 (1.43)	86.51
12	15	7	4	1	81.80 (1.74)	81.89
13	15	5	3	1	69.67 (0.47)	69.73
14	15	5	3	1	70.82 (0.84)	69.73
15	15	5	3	1	68.47 (0.69)	69.73
16	10	3	3	2	71.09 (1.27)	70.66
17	20	3	3	2	88.85 (1.13)	89.24
18	10	7	3	2	67.07 (0.45)	66.99
19	20	7	3	2	85.68 (0.40)	85.50
20	10	5	2	2	58.91 (1.58)	60.55
21	20	5	2	2	76.83 (1.52)	76.76
22	10	5	4	2	74.48 (1.50)	74.36
23	20	5	4	2	93.26 (0.57)	92.83
24	15	3	2	2	69.84 (1.02)	68.65
25	15	7	2	2	63.82 (1.04)	63.87
26	15	3	4	2	83.17 (0.63)	83.03
27	15	7	4	2	79.29 (0.64)	79.66
28	15	5	3	2	74.75 (1.10)	74.09
29	15	5	3	2	73.82 (1.33)	74.09
30	15	5	3	2	73.71 (0.72)	74.09

*Values in parentheses indicate CV%

Table 4: Regression equation of KCT composites on RB5.

Composite	Equation	R^2
1:1:1 KCT	$y = +69.18 + 10.27\,X_1 - 3.11\,X_2 + 8.39\,X_3 + 5.18\,X_1^2 + 5.0\,X_2^2 + 1.17\,X_3^2 + 0.37\,X_1X_2 + 0.67\,X_1X_3 + 0.38\,X_2X_3$	0.993
1:1:2 KCT	$y = +73.06 + 9.13\,X_1 - 2.14\,X_2 + 7.60\,X_3 + 3.48\,X_1^2 + 1.64\,X_2^2 - 0.67\,X_3^2 + 0.21\,X_1X_2 + 0.21\,X_1X_3 + 0.53\,X_2X_3$	0.990

RESULTS AND DISCUSSION

Statistical analysis - Response surface methodology

Decolorization efficiency using 1:1:1 and 1:1:2 KCT composite on RB5 obtained by experimental results and predicted results by ANN are given in Table 3. Table 4 shows the regression equation and coefficient of determination for Decolorization efficiency of effluents. The p value of 1:1:1 and 1:1:2 KCT composite is less than 0.0001 and thus the result is significant. The p value of 1:1:1 KCT composite for the factors composite concentration,

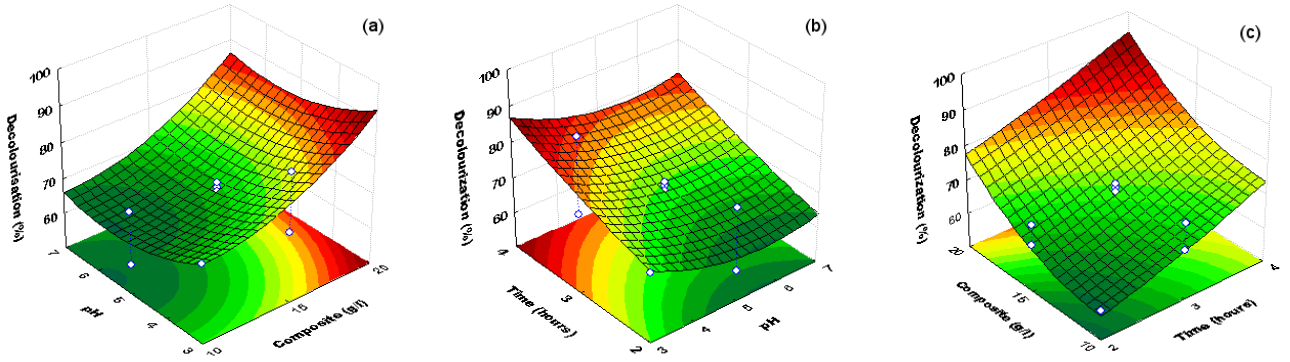

Fig. 3: Response surface of UV-photolysis of 1:1:1 KCT composite Decolorization efficiency.

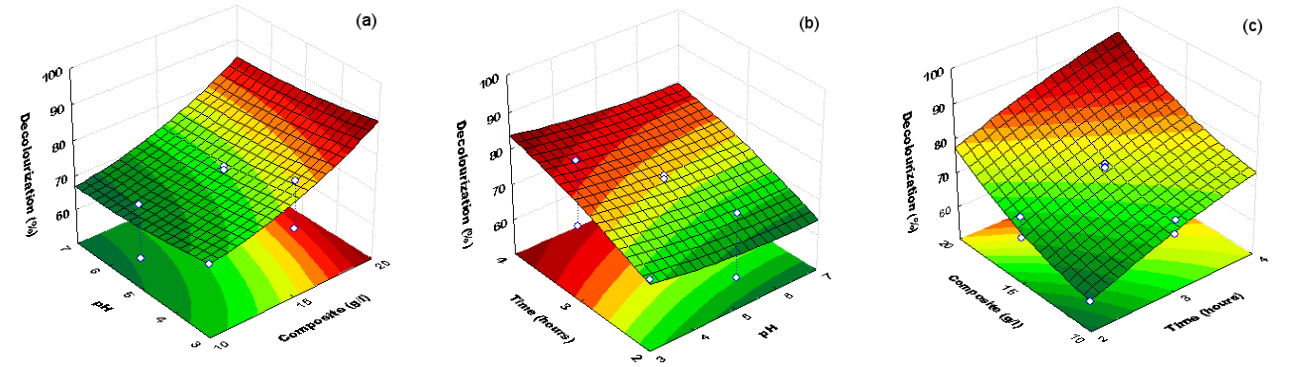

Fig. 4: Response surface of UV-photolysis of 1:1:2 KCT composite Decolorization efficiency.

*p*H and time of treatment is less than 0.0001, 0.0002 and 0.0001 respectively.

Similarly, the p value of KCT composite for the factors composite concentration, *p*H and time of treatment is less than 0.0001, 0.0021 and 0.0001 respectively. Both the composite p value is less than 0.05 thus the result is significant. The high values for the coefficient of determination of 1:1:1 KCT composite (R^2 = 0.993) and 1:1:2 KCT composite (R^2 = 0.990) suggests a good fit of the model with the experimental data. The 3D response surface of the performance variables composite concentration, pH and time for 1:1:1 and 1:1:2 KCT composite on Decolorization of RB5 effluents is presented in Figure 3 (a) - (c) and Figure 4 (a) - (c,) respectively.

Response surface models confirm significant interaction of performance variables on Decolorization of RB5. It can be seen from Figure 3 and 4, that both the composite follows similar trend irrespective of proportion due to formation of similar phases. Increase in composite concentration shows an increasing trend and increase in pH shows a decreasing trend on Decolorization percentage of RB5 as shown in Figures 3 and 4 (a). Time of treatment responds linearly with respect to pH and composite as shown in Fig 3 and 4 (b-c). The maximum composite efficiency is found at pH 3 with maximum time of 4 hours of treatment.

These interactive studies were used to determine the optimum condition with the help of Design Expert software and the optimum conditions for 1:1:1 and 1:1:2 KCT composite is presented in Table 5. The dye effluent treated using 1:1:1 and 1:1:2 KCT composite under optimized condition results in 94.42 % and 93.93 % Decolorization efficiency respectively and is shown in Fig. 5. The statistically predicted Decolorization efficiency is well in agreement with the experimental results performed using optimized conditions.

Dye degradation – FT-IR analysis

FT-IR spectra of Reactive Black 5, its residues after treatment of effluents using 1:1:1 KCT composites treated at pH 3, 5 and 7 are shown in Figure 6. FT-IR spectra of original dye sample exhibited the peaks at 3450 cm^{-1}, 2968 – 2888 cm^{-1}, 1640 cm^{-1}, 1400 – 1580 cm^{-1} and 1078 – 1256 cm^{-1} accountable for the presence of –NH, CH2, C=N,

Table 5: Optimum treatment conditions for KCT composite with predicted and achieved Decolorization efficiency.

Response variables	Optimum conditions	
	1:1:1 KCT Composite	1:1:2 KCT Composite
Composite Concentration (g/L)	18.4	19.8
pH	3.3	3.6
Time (Hours)	4	4
Decolorization efficiency (%) – Predicted	94.20	93.65
Decolorization efficiency (%) – Achieved	94.42	93.93

Table 6: Prediction performance summary

Statistical Performance	Decolorization Efficiency			
	Prediction by Statistical Model		Prediction by ANN Model	
	Equation 1	Equation 2	Prediction by Training data-set	Prediction by Testing data-set
Correlation coefficient	0.997	0.997	0.99	0.97
MAEP (%)	0.0094	0.0069	0.0074	0.0062
MSE	0.657	0.405	0.59	0.4

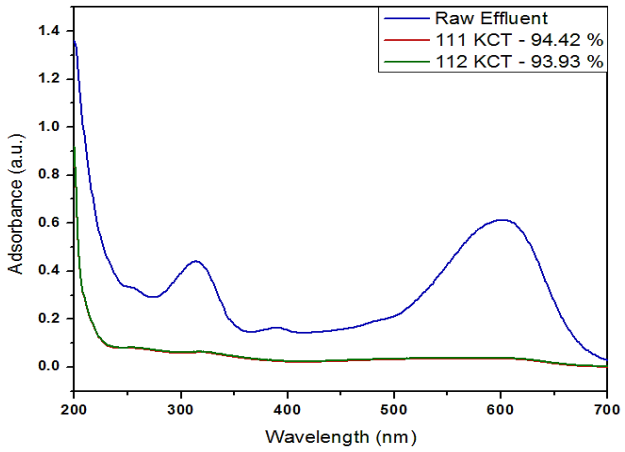

Fig. 5: Decolorization efficiency of RB5 under optimized condition

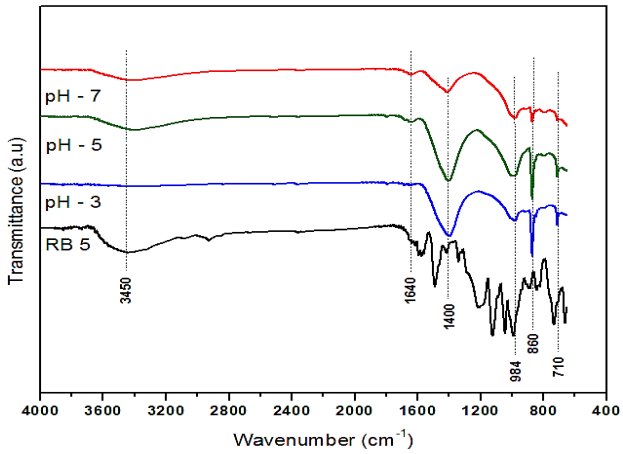

Fig. 6: FT-IR spectra of Reactive Black 5 and its residues after treatment.

N=N and O-C stretches [30]. After the treatment of effluents with KCT composites, significant changes in the peaks were observed – (i) absence of peaks around 1400 – 1580 cm^{-1}, indicating removal of N=N group, responsible for colour of the effluent (chromophore of the dye molecule), (ii) reduction / completely disappearance of –NH stretch (3000 – 3718 cm^{-1}) and (iii) reduction of peaks in 650 – 2000 cm^{-1} region and formation of new peaks indicating C=O stretch at 1640 cm^{-1} , -CH2 stretch at 1400 cm^{-1} and C-H stretch at 710 - 984 cm^{-1} [31].

Artificial Neural Network

Experimental results of Decolorization percentage using various combinations were used to train ANN (Table 2). Among 30 data-sets, 26 data-sets were used for training the network, and 4 data-sets were used for validation of the model. ANN training were stopped and optimized based on the minimum possible MSE achieved. The difference between the experimental values and the predicted values were used to calculate the absolute error percentage. Table 6 shows the prediction performance summary of statistical and ANN models.

The correlation coefficient is more than 0.99 for training data set and 0.97 for testing data. The correlation coefficient of statistical model is 0.997 for both the equations.

CONCLUSIONS

The composite concentration, pH and time of treatment were found to have significant effect on

Decolorization of RB5. Increase in composite concentration and time increases the Decolorization efficiency, while lower pH values increase the Decolorization efficiency. Based on the optimization conditions, about 94.42% and 93.93 % of Decolorization efficiency was achieved for 1:1:1 and 1:1:2 KCT composite, respectively. In ANN model number of hidden layers, number of hidden nodes in each hidden layer, number of iterations, momentum constant and learning rate were optimized to predict Decolorization efficiency. Experimental values and ANN predicted values were compared and the prediction performance was assessed by mean square error, mean absolute percentage error and correlation coefficient, which showed marginally better prediction performance in the case of ANN model than statistical model in terms of mean square error and correlation coefficient.

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