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Modeling of Phenol Degradation in Spouted Bed Contactor Using Artificial Neural Network (ANN)

Madhukar A. Dabhade* M. B. Saidutta†

D. V. R. Murthy‡

*National Institute of Technology Karnataka, Surathkal, dabhadema@rediffmail.com

†National Institute of Technology Karnataka, Surathkal, saidutta.mb@gmail.com

‡National Institute of Technology Karnataka, Surathkal, dvrmvzm@gmail.com

Modeling of Phenol Degradation in Spouted Bed Contactor Using Artificial Neural Network (ANN)

Madhukar A. Dabhade, M. B. Saidutta, and D. V. R. Murthy

Abstract

Presence of phenol and phenolic compounds in various wastewaters and its harmful effects has led to the use of different treatment methods. Work on biological methods shows the use of different microorganisms and different bioreactors so as to improve the removal efficiency economically. The present work deals with the use of *N. hydrocarbonoxydans* (NCIM 2386), an actinomycetes, for the degradation of phenol. *N. hydrocarbonoxydans* was immobilized on GAC and used in a spouted bed contactor for effective contact of microorganisms and the substrate. The contactor performance was studied by varying flow rates, influent concentrations and the solids loading in the contactor. The effect of these variables on phenol degradation was investigated and modeling study was carried out using the artificial neural network (ANN). A feed forward neural network with back propagation was used for the model development. The experiments were planned as per the face centered cube design (FCCD) and used for training of the model, whereas data from four other experimental runs were used for testing and validation of the model. The network was optimized for the number of neurons based on the mean square error. The ANN model with three layers with three input neurons, eight neurons in hidden layers and one output neuron was found to predict effectively the effluent concentration for the given operating conditions in the spouted bed contactor. The mean square error was found to be 9.318e-12 for this ANN model. Also the experimental data was used to develop second order nonlinear empirical model obtained using multiple regression (MR) and the results compared with ANN using correlation coefficient (R^2), average absolute error (AAE) and root mean square error (RMSE). Results show that R^2, AAE and RMSE values of MR model were 0.9363, 2.085 % and 2.338 % respectively, while in case of ANN model these values were 0.9995, 0.59 % and 1.263 % respectively. This shows that ANN model prediction is better than multiple regression model prediction.

KEYWORDS: spouted bed contactor, artificial neural network, phenol biodegradation, multiple regression

1. INTRODUCTION

The agricultural and industrial activities introduced different organic pollutants into the environment which are toxic and carcinogenic. The harmful effects of these pollutants on human and aquatic life and its strict discharge norm have led to the development of treatment technologies like biodegradation which emphasizes the destruction of pollutants rather than conventional disposal. Phenol and its derivatives is the basic structural unit in a wide variety of synthetic organics including pesticides. Phenol is listed as a priority pollutant by USEPA. Phenolic waste is generated from pesticide manufacturing, dye manufacturing, synthetic resin, refinery, coke and ammoniacal liquor (Autenrieth et al., 1991). The ingestion of phenol contaminated water in the human body causes protein degeneration, tissue erosion, paralysis of central nervous system, damage to kidney, liver and pancreas, vomiting and lung failure (Kirk and Othmer, 1982). EPA has set a limit of 0.1 mg/l of phenol in waste water. According to Bureau of Indian Standard, the permissible limit for the discharge of phenolic effluent into inland surface water is 1 mg/l and 5 mg/l in public sewer and marine coastal areas (Gupta et al., 2006).

Different biofilm reactors like packed bed, fluidized bed, RBC and membrane bioreactor etc are used for phenol degradation due to their various advantages (Sheeja et al., 2002; Israni et al., 2002; Livingston and Chase, 1989; Gonzalaz et al., 2001; Kai-Chee et al., 2000). The drawbacks associated with packed column, trickle bed and fluidized beds are discussed by Shetty et al. (2007). The problems like clogging, channeling and increased pressure drop etc restrict their uses. In spouted bed, there are three different flow regions. The recirculation of solids and liquids between the central up flow region and annular down flow region helps to keep a thin biofilm on solids and avoid any buildup of biomass. In the present study, spouted bed contactor was used for phenol biodegradation. Granular activated carbon was used for immobilization of the cells of *N. hydrocarbonoxydans* (NCIM 2386). Activated carbon was selected due to its better adsorption characteristics, better spouting behavior, light weight and easy removal of biomass from its surface. *N. hydrocarbonoxydans* is an actinomycetes capable of degrading phenol (Vidyavathi N, 1998; Shetty et al., 2007). Literature on spouted bed technology shows its wide applications in drying, coating, granulation, solids blending, combustion, pyrolysis and charcoal activation (Mathur and Epstein, 1974). Silva and Yang (1998) used gas-solid spouted bed bioreactor for production of amylases from rice by solid state fermentation. Recently, Osmary et al. (2007) used three phase spouted bed for solid catalyst alkylation. Literature on use of three phase spouted bed as a bioreactor is rare.

The objective of this work was to study the performance of spouted bed contactor for phenol biodegradation. The artificial neural network for the modeling of spouted bed contactor would be attempted. To predict the effluent concentration the multiple regression model would also be developed and its efficiency compared with ANN model using correlation coefficient, average absolute error (%) and root mean square error (%).

2. MATERIALS AND METHODS

Activated carbon (specific surface area (BET) = 700 m^2/g, bulk density = 0.53±0.05 g/cm^3, average pore radius = 10 x 10^{-10} m and micro pore volume = 0.38 cm^3/g) and all the chemicals (except 4-aminoantipyrene) were obtained from NICE Chemicals, Cochin, India. 4-aminoantipyrene was obtained from Central Drug House, New Delhi. A strain of microorganism *N. hydrocarbonoxydans* (NCIM 2386), was obtained from National Chemical Laboratory, Pune, India. It uses phenol as a sole source of carbon and energy (Vidyavathi, 1998; Shetty et al., 2007). The strains were sub cultured once in fifteen days on agar slants and were stored at 4 ºC.

The growth culture was prepared using nutrient medium consisting of media-A (ammonium nitrate-1 g/l, ammonium sulphate-0.50 g/l, sodium chloride-0.50 g/l, di-potassium hydrogen orthophosphate-1.50 g/l, potassium di-hydrogen orthophosphate-0.50 g/l, ferrous sulphate-0.002 g/l) and media-B (calcium chloride 0.01 g/l and magnesium sulphate-0.50 g/l). To avoid the precipitation of calcium chloride and magnesium sulphate, media A and media B were prepared and separately steam sterilized at 1.5 kg/cm^2, 120 ^oC for 20 minutes before addition to the growth media in a suitable volume. The solution was adjusted to pH 7.0 by using 0.1 N NaOH. A loop full of test organism was taken from a freshly sub cultured slant and inoculated into 100 ml sterilized media containing 100 mg/l phenol and the nutrients. This culture was incubated at a 30 ^oC for 3 days in a shaker operated at 150 rpm. This formed the primary culture. Second, third and fourth acclimatization was also prepared. The acclimatization was followed according to procedure mentioned by Shetty et al. (2007). While inoculating secondary acclimatization 1 ml of primary culture was used instead of the subculture in 250 ml flask and incubated for 48 h. The fourth acclimatized culture was then gradually acclimatized to higher concentrations of phenol.

Granular Activated Carbon (GAC) was sieved for required size (average size 2.4 mm), thoroughly washed with distilled water to remove all dirt and fines, dried at 105 ^oC for one day and then it was used for immobilization. The immobilization on GAC was done by adsorption. The use of activated carbon for immobilization of phenol degrading microorganism has been reported in the literature, as activated carbon provides a high adsorption capacity for phenol and

also acts as a support material for microorganisms (Ehrhardt and Rehm, 1985). For every gram of GAC, 7.5 ml of cell suspension from fourth acclimatized culture was added and refrigerated at 4 ^{o}C for two days with occasional stirring.

After immobilization, solids were filtered, washed and loaded into the contactor. At regular interval contactor effluent samples were collected, filtered using Whattman filter paper (no.1) and analyzed for phenol concentration. Phenol concentration was measured according to standard method, (APHA, 1975) of using 4-aminoantipyrene which in presence of potassium ferricyanide at pH of 10 reacts with phenol and produces amber like color. The absorbance was measured at 510 nm using UV-VIS spectrophotometer (HITACHI-2000). The dry biomass weight was measured at the end of the experiment by weight difference before and after washing GAC in 0.25 M NaOH and drying at 105 ^{o}C for 2 days. The washing and drying was repeated till the constant value was obtained (Livingston and Chase, 1989).

3. EXPERIMENTAL

3.1 Experimental Set Up

Experimental set up is as shown in Fig.1. Spouted bed contactor is made of Perspex column of 25 mm inside diameter and 26 cm length. The working volume of the contactor is 136.4 ml. It consists of lower conical section and upper cylindrical section. Phenolic influent and air enter the contactor through a small orifice. Care was taken to avoid any photo degradation of phenol by blackening the phenolic solution tank and the tubes. The column was loaded with different quantities of GAC (0.010 to 0.020 kg) immobilized with *N. hydrocarbonoxydans* and operated at three different flow rates (0.100, 0.200 and 0.300 l/h) and influent concentrations of 0.100, 0.200 and 0.300 kg/m^3. Immobilization was done under batch mode according to procedure given in section 2 prior to its use in the contactor.

Effluent concentration was measured at regular intervals of time. Experiment was continued till steady state was reached, which was indicated by the constant effluent concentration. As GAC loading was varied from 0.010 kg to 0.020 kg, air flow was varied from 1.50 lpm to1.75 lpm so as to have spouting of bed giving systematic recirculation of solids inside the contactor. D.O. in the contactor was measured using pre-calibrated HACH probe and it was above 5 mg/l.

3.2 Design of Experiments

The experiments were planned based on face centered cube design. This design has been found to be adequate to model multifactorial response (James and Muth, 1999; Montgomery, 2004). Three factors and three levels were selected. Twenty experimental runs as per this design were conducted which includes, one central point, eight points on the corners of the hyper cube, six points on the face of the hyper cube plus five replicates. Data from these runs were used for training the ANN. Data from four other experimental runs randomly selected were used for testing the model.

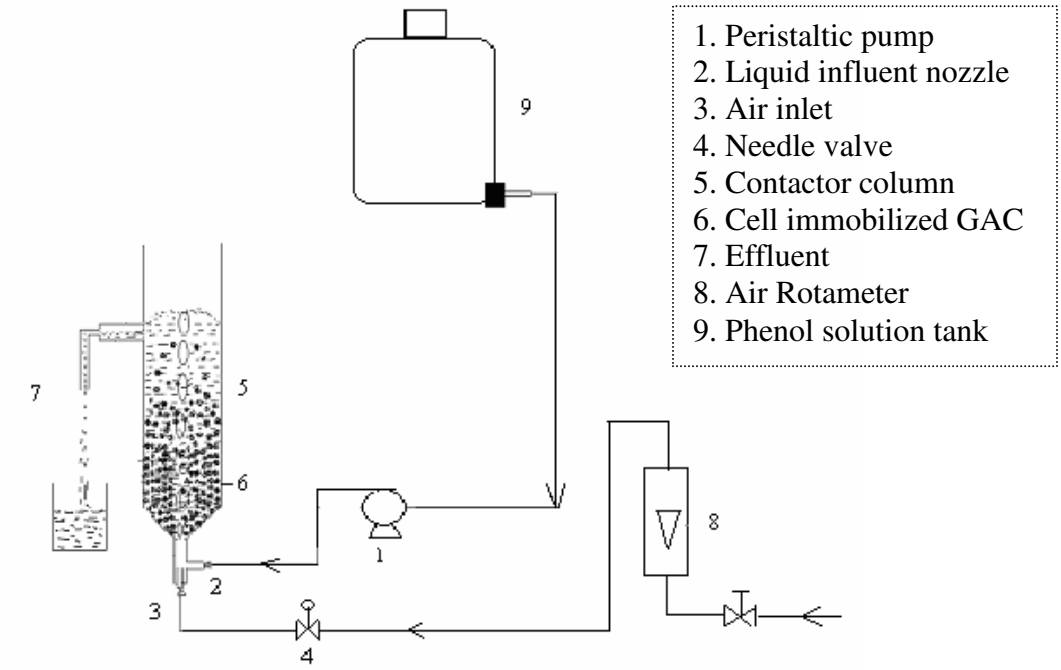

Fig.1: Schematic diagram of three phase spouted bed contactor

4. BIOREACTOR MODELING

4.1 Artificial Neural Network (ANN)

ANN is direct inspiration from our biological neural network. The ANN consists of interconnected processing units which consist of a summing part followed by an output part. The summing part receives N input values, weights each value and computes a weighted sum. The weighted sum is called the activation value. The output part produces a signal from the activation. The processing units are interconnected according to some topology. The arrangement of the processing

units, connections and pattern input/output is referred to as topology. ANN networks are normally organized into layers of processing units. The connections across the layers and among the units within a layer can be in a feed forward manner or in a feedback manner. The processing unit (neurons) process information by their dynamic response to external inputs. In case of multiple layer feed forward with back propagation, generalized delta rule is used as learning law which is supervised learning. Here the weights are adjusted based on the error between the desired and actual output value for a given input (Yenanarayana, 2005). The minimization of the mean square error is the main technique used in the back propagation algorithm.

In the present work, the inputs to ANN model were phenol concentration, flow rate and solids loading in the contactor. The output parameter was percentage degradation. Three layered feed forward neural network with back propagation having topology as 3:8:1 was used. The 'logsig' transfer function was used in the input and hidden layer and 'purelin' function between hidden layer and output layer. As per universal approximation theory, network with single hidden layer and sufficiently large number of neurons can interpret any input output structure (Daneshvar et al., 2006). For training 'trainlm' function was used. ANN model was developed under a MATLAB (7.20R2006A) environment (Math Work, Natick, Mass, U.S.A.).

4.2 Multiple Regression Model

For three factors and three levels of experiment, following second order empirical model was considered. (Annadurai et al., 2000)

$$Y = \beta_0 + \Sigma\beta_i X_i + \Sigma\beta_{ii} X_i^{\,2} + \Sigma\Sigma\beta_{ij} X_i X_j \qquad (1)$$

Where,

Y is the predicted response (steady state percentage degradation), X_i is independent variable (factor), β_0 is intercept term, β_i linear coefficient. β_{ii} is quadratic coefficient, β_{ij} is interaction (cross product) coefficients. Three factors affecting the response are phenol concentration (X_1), flow rate (X_2) and GAC loading (X_3). Table1 shows the coded factors at three levels (-1, 0, +1), calculated using the equation (2),

$$X_i = \frac{2x_i - \left(x_{ihi} + x_{ilo}\right)}{x_{ihi} - x_{ilo}} \qquad (2)$$

X_i (i = 1, 2, 3) are the respective coded factors, x_{ihi} and x_{ilo} is higher and lower value of x_i, respectively.

Table 1: Coded factor at three level

Concentration, kg/m^3 (X_1)	Flow rate, l/h (X_2)	Weight of GAC, kg (X_3)
0.100 (-1)	0.100 (-1)	0.010 (-1)
0.200 (0)	0.200 (0)	0.015 (0)
0.300 (+1)	0.300 (+1)	0.020 (+1)

5 RESULTS AND DISCUSSIONS

5.1 Adsorption and Biodegradation

To differentiate the phenol removal by biodegradation from adsorption, a separate experiment of adsorption and biodegradation was conducted. Fig.2 shows the adsorption and degradation plot for 0.050 kg/m^3 phenol concentration and 0.010 kg of GAC. In case of adsorption, the effluent concentration reduced to minimum within first 3 to 5 h.

Activated carbon contains a highly developed pore structure. The major adsorption takes place in the micro pores. The adsorption of phenol from water is by physical adsorption and is reversible. This helps the microorganisms immobilized on GAC to consume phenol easily. Initial decrease in effluent concentration is due to adsorption. When the microorganisms were immobilized on GAC, and used in a contactor, initially effluent phenol concentration decreased suddenly as observed in adsorption studies up to 3 to 5 h, afterwards very slow decrease in concentration was observed. During this period the growth of microorganism takes place and when the biodegradation becomes predominant, the effluent phenol concentration decreases reaching a steady value. The adsorption dominates at the initial stage and biodegradation dominates after reaching the adsorption equilibrium (Mondal and Balmomajumder, 2007).

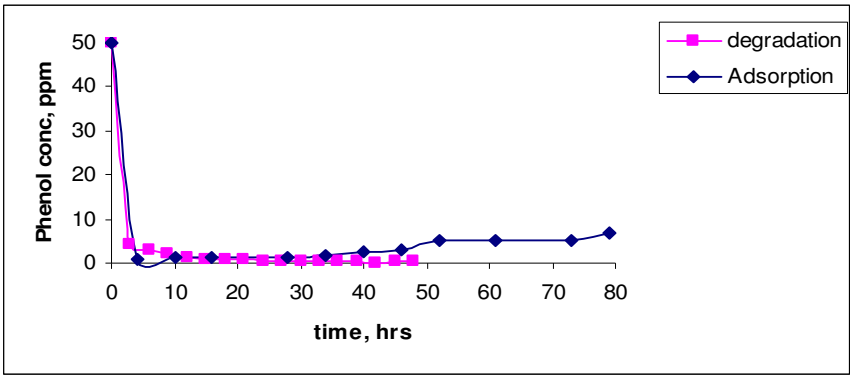

Fig.2: Adsorption and degradation plot for 0.050 kg/m^3 phenol conc., 0.100 l/h flow rate and GAC loading 0.010 kg.

5.2 Artificial Neural Network

An attempt was made to model the performance of spouted bed contactor for phenol degradation, using artificial neural network. Input variables for the method were scaled in between 0 to 1. Table 2 shows the variables with their range and new scale. Initially the data was trained by varying the number of neurons in the hidden layer from three to eight. After repeated training, for 8 neurons and 245 epochs, mean square error observed was 9.318e-12. Fig. 3 shows the plot of number of neurons against mean square error. Fig 4 shows the architecture of the given neural network

Table 2: Model variable and their range

Layer	Variables	Range	Scale
Input layer	Phenol concentration	0.100-0.300 kg/m^3	0.333-1.00
	Flow rate	0.100-0.300 l/h	0.333-1.00
	GAC loading	0.010-0.020 kg	0.500-1.00
Output layer	% degradation	0-100%	0-100%

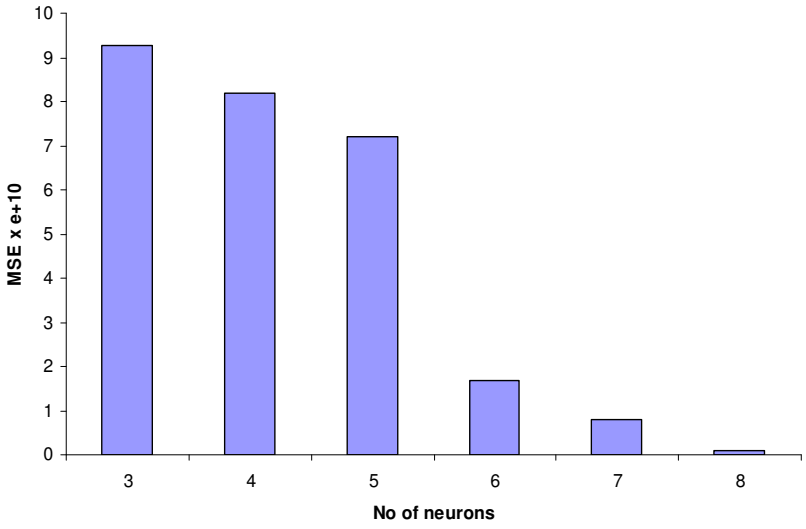

Fig. 3: Variation of mean square error with neurons

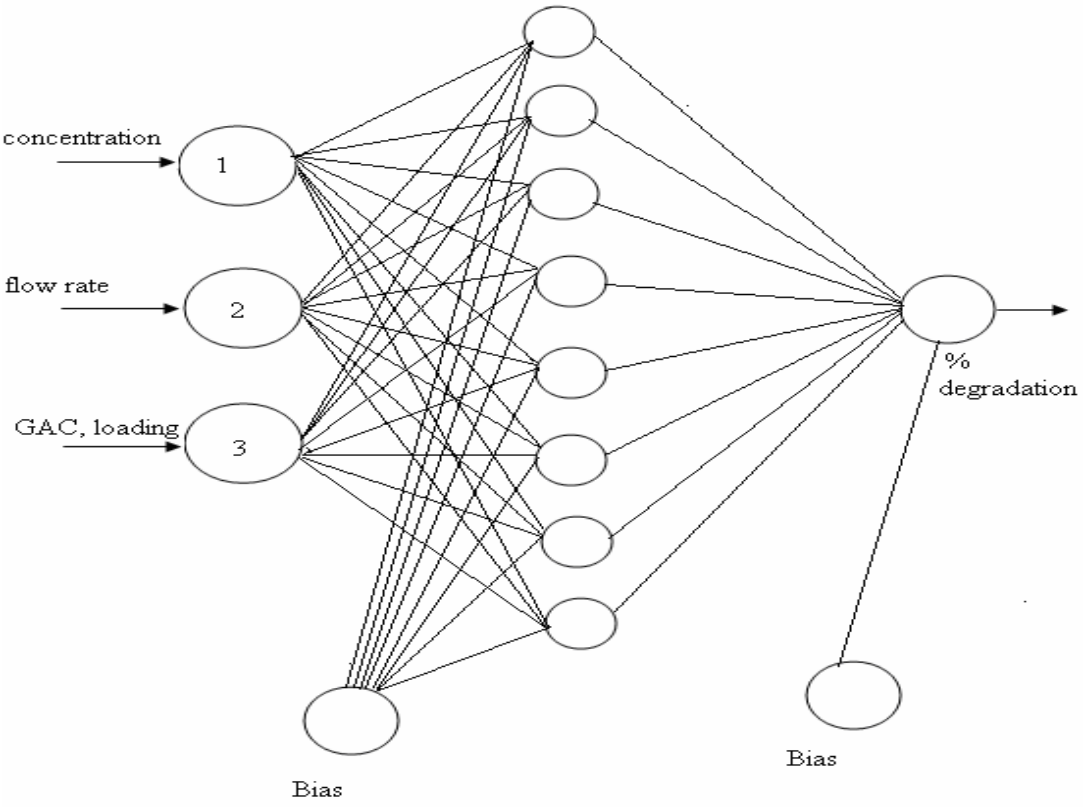

Fig. 4 ANN Architecture for phenol degradation system

The experimental and predicted values of percentage degradation are shown in Fig.5 and Table 3. The correlation coefficient of 0.9995 for training the model and 0.9953 for testing the model was obtained. The output of ANN for testing and validation of model is shown in Table 4.

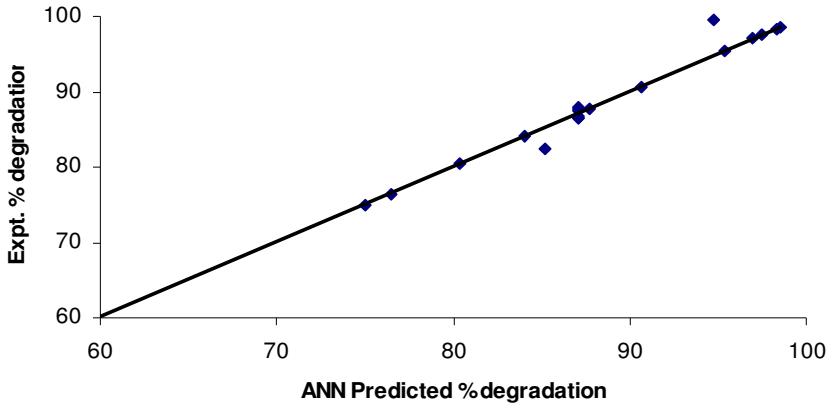

Fig. 5 Comparison of experimental and ANN predicted values

Table 3: Output of ANN and Multiple Regression Model for Training data set

Sr. no	Phenol conc. Kg/m^3	Influent flow rate l/h	GAC loading kg	% phenol degradation Experimental	% phenol degradation ANN output	% Phenol degradation MR output
1	0.300	0.200	0.015	76.50	76.50	80.05
2	0.100	0.200	0.015	98.50	98.50	92.46
3	0.200	0.300	0.015	84.00	84.00	79.57
4	0.200	0.100	0.015	97.00	97.00	98.94
5	0.200	0.200	0.020	90.62	90.62	89.69
6	0.200	0.200	0.010	82.50	85.20	83.64
7	0.300	0.300	0.020	75.00	75.00	73.69
8	0.300	0.300	0.010	58.85	58.85	59.96
9	0.300	0.100	0.020	95.35	95.35	94.71
10	0.300	0.100	0.010	99.47	94.70	91.96
11	0.100	0.300	0.020	87.70	87.70	91.05
12	0.100	0.300	0.010	80.40	80.40	81.68
13	0.100	0.100	0.020	98.30	98.30	97.80
14	0.100	0.100	0.010	97.50	97.50	99.42
15	0.200	0.200	0.015	86.50	87.12	87.95
16	0.200	0.200	0.015	87.85	87.12	87.95
17	0.200	0.200	0.015	87.50	87.12	87.95
18	0.200	0.200	0.015	86.50	87.12	87.95
19	0.200	0.200	0.015	86.75	87.12	87.95
20	0.200	0.200	0.015	87.65	87.12	87.95
Correlation Coefficient					0.9995	0.9363

5.3 Multiple Regression Model

The experimental data was analyzed statistically using LABFIT V 7.2.34 (trial version) software (Silva et al., 2007).The following empirical model was obtained.

$$Y = 87.951 - 6.203X_1 - 9.687X_2 + 3.029X_3 - 1.691X_1^2 + 1.309X_2^2 - 1.281X_3^2 - 3.566X_1X_2 + 2.746X_2X_3 + 1.091X_1X_3 \quad (3)$$

The percentage degradation obtained in each experiment was compared with that calculated using empirical model equation (3) and the results are given in Table 3. Fig.6 shows the experimental and predicted percentage degradation. This gives correlation coefficient of 0.9363.

The comparison of both the models was made using correlation coefficient, average absolute error (%) and root mean square error (%). Balan et al. (1999) used similar method to check the performance of models and optimized the nutrient media composition for phenol degradation. The correlation coefficient, average absolute error (%) and root mean square error (%) for ANN model were 0.9995, 0.59 % and 1.263 % ; while for multiple regression model these values were 0.9363, 2.085 % and 2.338 % respectively. Both the models predict well the effluent phenol concentration, but ANN model shows higher correlation coefficient, low average absolute error and also low root mean square error compared to multiple regression model. In testing of models using blind data sets similar results were obtained as shown in Table 4 and hence ANN model is better compared to multiple regression model. Table 5 gives the comparison of both the models for training and testing data set.

Table 4: Output of ANN and Multiple Regression Model for Testing of the model

Sr. no.	Phenol conc. Kg/m^3	Influent Flow rate, l/h	GAC loading, kg	% phenol degradation, Experimental	% phenol degradation ANN output	% phenol degradation, MR output
1	0.100	0.100	0.015	99.73	97.66	99.89
2	0.200	0.100	0.010	96.63	95.97	97.38
3	0.300	0.200	0.010	76.67	75.85	74.66
4	0.100	0.300	0.010	82.34	83.28	87.65
Correlation Coefficient					0.9953	0.9275

Table 5: Comparison between ANN and Multiple Regression Model

	Correlation Coefficient		Average Absolute Error (%)		RMS Error (%)	
	ANN	Regression	ANN	Regression	ANN	Regression
Training data	0.9995	0.9363	0.590	2.085	1.263	2.338
Testing data	0.9953	0.9275	1.254	2.864	1.25	2.864

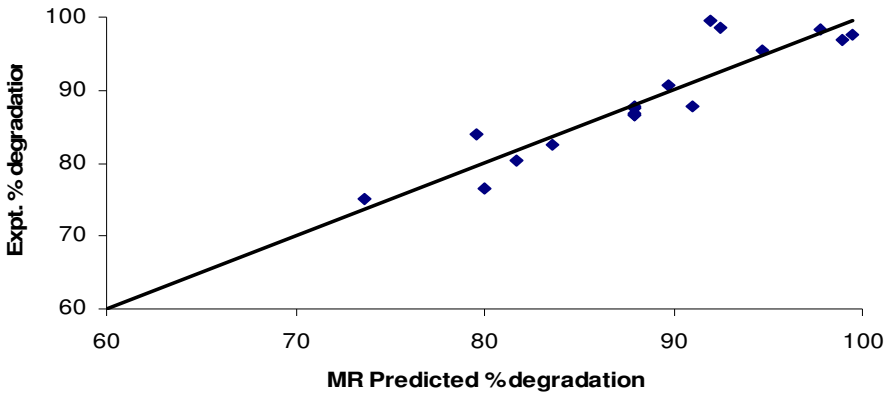

Fig.6: Experimental and predicted percentage degradation using Multiple Regression model

5.4 Effect of Influent Concentration, Flow rate and Solids loading on % degradation

Influent substrate concentration is an important parameter affecting the percentage degradation, as the microbial kinetics gets affected. It has been observed that at all the flow rates, there is a decrease in percentage degradation with increase in influent concentration. At higher flow rates and higher influent concentration, percentage degradation drops below 75 % where as at lower flow rates and higher concentration it is above 95 % (Table 3). Typical bar chart for 0.010 kg GAC and flow rate of 0.200 l/h is shown in Fig.7. There is a good agreement between the experimental and model predicted results.

The residence time of substrate and hence the contact time of substrate with microorganisms is dependant on the influent flow rate. Higher residence time gives rise to higher percentage degradation for low substrate concentration whereas for higher substrate concentration and higher flow rates, degradation showed a decline. Wang et al. (2003) reported that higher liquid flow rate increases shear between the spout and the annulus and enhance turbulence. This results in small and uniform bubble formation and increase in gas hold up. But, in case of biofilm reactors, at higher flow rate, shear action will detach the biomass and results in poor degradation. The solids loading in a contactor determine the surface area available for microorganisms for immobilization and hence the concentration of microorganism is higher for higher solids loading resulting in higher percentage degradation. GAC used in this study was found to adsorb and biodegrade phenol simultaneously due to its inherent adsorption capacity as well as the presence of immobilized microorganisms, leading to increased capacity to remove phenol.

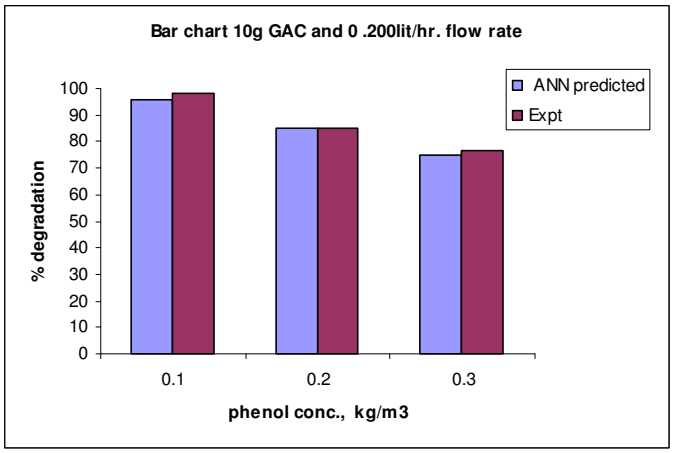

Fig.7: Comparison of ANN predicted percentage degradation with Experimental result (GAC = 0.010 kg and Influent flow rate = 0.200 l/h)

6. CONCLUSIONS

A phenol degrading microorganism *N. hydrocarbonoxydans* was immobilized on GAC and used in spouted bed contactor. No backwashing was required in spouted bed due to its systematic flow pattern. Both the ANN model and multiple regression model can predict the effluent concentration.

The correlation coefficient of 0.9995, low average absolute error (0.59 %) and low root mean square error (1.263 %) of ANN shows good prediction using ANN model as compared to multiple regression model. This model can be used to predict the effluent concentration for the given range of parameters under study. Artificial neural networking is a simple and a good method for modeling phenol biodegradation.

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