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Immobilized Cell Biofilter: Results of Performance and Neural Modeling Strategies for NH_3 Vapor Removal from Waste Gases

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ABSTRACT

Artificial neural networks (ANNs) are powerful data-driven modeling tools which have the potential to approximate and interpret complex input/output relationships based on given sets of a data matrix. In this paper, a predictive computerized approach is proposed to predict the performance of an immobilized-cell biofilter treating NH_3 vapors in terms of its removal efficiency (RE) and elimination capacity (EC). The input parameters to the ANN model were inlet concentration, loading rate, flow rate and pressure drop, and the output parameters were RE and EC. The data set was divided into two parts: a training matrix consisting of 51 data points, and a test matrix with 16 data points representing each parameter considered in this study. Earlier experiments of continuous biofilter operation showed removal efficiencies from 60 to 100% at inlet loading rates (ILRs) varying between 0.5 to 5.5 g $NH_3/m^3/h$. Internal network parameters of the ANN model during simulation were selected using the 2^k factorial design, and the best network topology for the model was thus estimated. Predictions were evaluated based on their d coefficient values (R^2). The results showed that a multilayer network (4-4-2) with a back propagation algorithm was able to predict biofilter performance effectively with R^2 values of 0.9825 and 0.9982. The proposed ANN model for biofilter operation could be used as a potential alternative for knowledge-based models through proper training and testing of variables.

Keywords: Immobilization; Biofilter; Removal efficiency; Elimination capacity; Neural network; Prediction.

INTRODUCTION

Ammonia is used extensively in the semiconductor industry as the starting material for the manufacture of nitric acid and as a refrigerating fluid replacement for chlorofluorocarbons. Malodors containing NH_3 are released from pulp and paper industry, wastewater treatment plants, night soil treatment plants and aerobic composting of low C/N material. Hence, there arises a potential need to adapt suitable control techniques for the effective removal of these emissions from related process industries. Biofiltration is a cost effective technology for treatment of waste gases containing low concentrations of volatile organic compounds (VOCs) at large flow rates. The high removal efficiencies (REs) achieved along with uncomplicated flexible design, low operational and maintenance costs gives biofilters an edge over other physico-chemical treatment techniques for the removal of VOCs and other malodors, such as adsorption, absorption, incineration and condensation (Kennes and Veiga, 2001; Cheng, 2008). Biofilters have been proved to remove NH_3 emissions effectively from gas streams using a bed of biologically active material, such as compost, peat, wood bark, etc. In recent years, immobilization of microbes in support matrix, such as alginate beads or suitable polymeric materials has gained popularity in the field of biofiltration. The main advantages of adopting immobilization techniques in biofiltration is to provide high cell concentrations, improve genetic stability, protection from shear damage and to enhance favorable microenvironments for microbes (nutrient gradients and pH). Chung *et al.* (1996) evaluated the effects of operational factors, such as retention time, temperature and inlet concentration on the performance of a biofilter packed with *Thiobacillus thioparus* immobilized with Ca-alginate pellets and found an optimal S-loading of 25 $g/m^3/h$.

Traditionally, the performance of biofilters has been modeled/predicted using process-based models that are based on mass balance principles, simple reaction kinetics and a plug flow of air stream (Ottengraf and Van Den Oever, 1983; Shareefdeen *et al.,* 1993; Deshusses *et al.,* 1995; Jin *et al.,* 2006). The main advantages of these process models are that they are based on the underlying physical process and the results obtained generally provide a good understanding of the system. However, this depends on numerous model parameters and obligates information on specific growth rate of microbes, biofilm thickness and density, values of diffusivity, partition, yield and distribution coefficient, intrinsic adsorption, etc. The accurate estimation of some of these parameters requires elaborate technical facilities and expertise, the absence of which hinders the model's precision and limits its application and reliability. These parameters were not used in this study; instead, some easily measurable parameters were chosen, which are described herein.

An alternate modeling procedure consists of a data-driven approach wherein the principles of artificial intelligence are applied with the help of neural networks. It has been shown earlier that the performance of biofilters and biotrickling filters can be predicted from prior estimation of easily measurable operational parameters, such as flow rate, unit flow, inlet loading rate, pressure drop and inlet concentration (Elias *et al.,* 2006; Rene *et al.,* 2006).

THE ANN-BASED MODELING APPROACH

A multi-layer perceptron (MLP) using the back propagation algorithm (Rumelhart *et al.,* 1986) is the most widely used neural network for forecasting/prediction purposes (Maier and Dandy,

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2000). Neural networks acquire their name from the simple processing units in the brain called neurons which are interconnected by a network that transmits signals among them. These can be thought of as a black box device that accepts inputs and produces a desired output. MLP generally consists of three layers; an input layer, a hidden layer and an output layer. Each layer consists of neurons which are connected to the neurons in the previous and flowing layers by connection weights (W_{ij}). These weights are adjusted according to the mapping capability of the trained network. An additional bias term (θ_j) is provided to introduce a threshold for the activation of neurons. The input data (X_i) is presented to the network through the input layer, which is then passed to the hidden layer along with the weights. The weighted output ($X_i W_{ij}$) is then summed and added to a threshold to produce the neuron input (I_j) in the output layer. This is given by:

$$I = \sum X_i W_{ij} + \theta_j \tag{1}$$

This neuron input passes through an activation function $f(I_j)$ to produce the desired output Y_j. The most commonly used activation function is the logistic sigmoid function which takes the form:

$$f(I_j) = \frac{1}{1 + e^{-I_j}} \tag{2}$$

MATERIALS AND METHODS

Experimental details pertaining to cultivation of micro-organisms, media composition, preparation of immobilized packing media, experimental setup, biofilter operation and analytical techniques for data collection are given in our previously published work (Kim *et al.,* 2007) and is summarized in Table 1.

The inlet loading rate, removal efficiency and elimination capacity were calculated to evaluate biofilter performance, according to Eqs. (3-5),

$$\text{Inlet loading rate, ILR (g/m}^3\text{h)} = \frac{Q\,C_{go}}{V} \tag{3}$$

$$\textit{Elimination capacity, EC (g/m}^3\textit{h)} = \frac{Q(C_{go} - C_{gi})}{V} \tag{4}$$

$$\text{Removal efficiency, RE (\%)} = \frac{C_{go} - C_{gi}}{C_{go}} \times 100 \tag{5}$$

Where, C_{go} and C_{gi} are the inlet and out pollutant concentrations from the biofilter, ppm_v (or) g/m^3, V is the volume of the biofilter, m^3 and Q is the gas flow rate, m^3/h.

MODELING METHODOLOGY

Model Input-Outputs

A neural network-based predictive model was developed with flow rate, inlet loading rate, pressure drop and inlet concentration as the model inputs and elimination capacity and removal efficiency as the outputs.

Data Division

The experimental data was divided into training (N_{Tr}, 75%) and test data (N_{Te}, 25%). The test data was set aside during network training and was only used for evaluating the predictive potentiality of the trained network.

Error Evaluation

The closeness of prediction between the experimental- and model-predicted outputs was evaluated by computing the determination coefficient values computed by the following formulae (Elias *et al.,* 2006):

$$R^2 = \left[\frac{\sum_{i=1}^{N} (Y_{model_i} - \overline{Y_{model}})(Y_{observed_i} - \overline{Y_{obserbed}})}{(N-1) S_{Y_{model}} S_{Y_{observed}}} \right]^2 \tag{6}$$

Where, Y_{model_i} is the predictions made using the model, $Y_{observed_i}$ is the experimentally observed true values, Y and S_Y are the averages and standard deviations, while N is the number of sample cases analyzed.

Data Pre-Processing and Randomization

Experimental data collected from the biofilter during the 67 days of continuous operation were randomized to obtain a spatial distribution of the data, which accounts for both steady state and transient steady state operation. The data was also normalized and scaled to the range of 0 to 1 using Eq. (7), so as to suit the transfer function in the hidden (sigmoid) and output layer (linear)

$$\hat{X} = \frac{X - X_{min}}{X_{max} - X_{min}} \tag{7}$$

Where, X is the normalized value, X_{min} and X_{max} are the minimum and maximum values of X, respectively.

Table 1. Summarized preparatory details of the immobilized cell biofilter.

Method	Experimental details	Conditions
Cultivation of mixed culture	Mixed culture isolated from wastewater treatment plant was grown in batch reactors. Media composition: Kim *et al.* (2007)	Grown in a rotary shaker (150 rpm) at 30 ± 2°C.
Packing material for biofilter	Autotrophic nitrifiers were harvested by centrifugation, washed aseptically with distilled water. Concentration of autotrophic nitrifiers ~3300 mg VSS/L. Purified cells were mixed with sterilized sodium alginate solution and then mixed with 7.5 L of polyvinyl alcohol (PVA) solution. The resulting solution was injected in a mold tray, freeze gelled and by thawing, PVA cryogels (cubes) having a cell concentration of ~825 mg/L were obtained.	Cells were centrifuged at 7500 g, 15 min.
Biofilter set up	Biofilter was operated in a down flow mode. Components: Flow meters, mixing chamber, sampling ports, nutrient recycling system.	ID: 14 cm, packed bed volume: 6.5 L.

Network Parameters

The internal parameters of the back propagation network, namely epoch size, error function, learning rate (η), momentum term (μ), training count (T_c) and transfer function were appropriately selected to obtain the best network architecture that gives high predictions for the performance variables.

In this study, the number of neurons in the input layer ($N_I = 4$) and output layer ($N_0 = 2$) were chosen based on the number of input and output variables in the network. A detailed study on the effect of internal network parameters on the performance of back propagation networks and the procedure involved in selecting the best network topology has been described elsewhere (Maier and Dandy, 1998). However in most instances, literature suggests the use of a trial and error approach in which the performance goal is set by the user. In this study, the best values of the network parameters were chosen by carrying out simulations performed using the 2^k full factorial design (Montgomery, 1991). The 2^k design is of particular significance in exploring the effect of many factors on the response variable for a particular system. It provides the smallest number of runs with which 'k' factors can be studied in a complete factorial design (In this study, k = 4, hence 16 simulations were done; *data not shown*). Determination coefficient (R^2) values were taken as the response variable and the setting that yielded the maximum R^2 value in the test data was taken as the best network parameter.

Software Used

ANN-based predictive modeling was carried out using the shareware version of the neural network and multivariable statistical modeling software, NNMODEL (Version 1.4, Neural Fusion, NY) and full factorial design was carried out by the statistical software MINITAB.

RESULTS AND DISCUSSIONS

Experimental

The performance of the immobilized cell biofilter was monitored by varying the flow rate and inlet concentration. A step increase from low to high loading rates to the biofilter required a few days to adapt to the new concentration and reach a new steady state value. The results from this study are shown in Fig. 1 as a function of the operating time, loading rate, empty bed residence time (EBRT) and removal efficiency (RE). These removal profiles indicated that the immobilized cells possessed good activity with steady and consistent removal even during the beginning of the experiments. The loading rate of NH_3 was gradually increased to 2.5 g/m^3/h on the 14th day of continuous operation. The response was a sudden decline in the RE from 100% to 96% followed by a new steady state at the end of the 16th day where the RE was 98%. Hence, the loading rate was decreased to 1.7 g/m^3/h and subsequently increased in incremental time steps to a maximum of 4.5 g/m^3/h. The biofilter RE profiles displayed minor ameliorating fluctuations due to a stepwise increase in loading rate between 1 and 4.5 g NH_3/m^3/h. It was also evident that the RE was nearly 100% (> 95%) up to a loading rate of 4.5 g/m^3/h. However, after 60 days, when the inlet loading rate (ILR) to the biofilter was increased significantly by varying both the concentration and flow rate to values as high as 7.5 g NH_3/m^3/h, a noticeable decrease in the RE values from 100% to nearly 60% was observed. The critical NH_3 loading rate to the biofilter was considered as 4.5 g NH_3/m^3/h. Pressure drop values were sufficiently low during the operational time (0.1-1.7 cms of H_2O) and did not cause any significant operational problem.

ANN-Based Modeling

To model the performance of the biofilter, neural-based simulations were carried out using the standard back error propagation network. The ranges of input and output parameters for the ANN model are given in Table 2. The experimental data collected from the biofilter was suitably divided into the training and test data sets, pre-processed and randomized before carrying out simulations. The model was evaluated with the test data and the effect of network parameters on the R^2 value was used as a measure to choose the best network architecture.

Effect of Network Internal Parameters

The different values of internal network parameters used to train the network are given in Table 3. During simulations with different combinations of settings as given by the experimental design, the following interpretations were made: (i) increasing the number of neurons in the hidden layer decreases the R^2 value significantly; (ii) an increase in the training count from low to high levels displays high R^2 value for the model; (iii) the effect of learning rate did not play a major role in increasing the R^2 value, but it played a complementary role in speeding up the error Convergence, and; (iv) the momentum term increased the R^2 value when increased from lower to higher levels. The best network architecture was then selected by observing high R^2 value in the test data set (Table 4, for RE predictions, R^2 value-

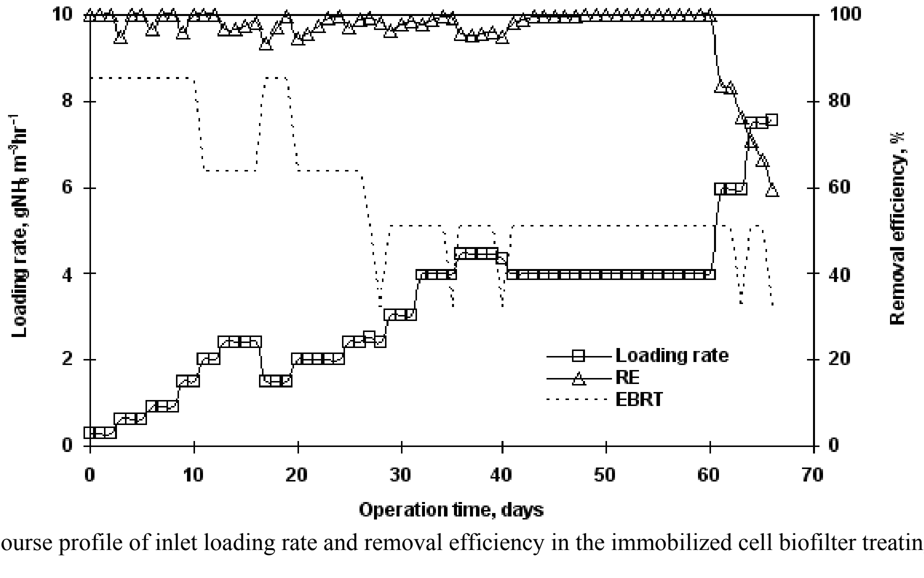

Fig. 1. Time course profile of inlet loading rate and removal efficiency in the immobilized cell biofilter treating NH_3 vapors.

Table 2. Range of input and output parameters used for training and testing ANN model developed to represent biofiltration of NH_3 vapors.

Parameter	Training data, N_{Tr}-51			Testing data, N_{Te}-16		
	Min	Max	Mean	Min	Max	Mean
Input						
Inlet concentration (ppm_v)	10	150	63.3	20	150	74.1
Flow rate (m^3/h)	6	16	9.25	6	16	9.13
Inlet loading rate (g/m^3/h)	0.3	7.5	3.08	0.6	7.5	3.54
Pressure drop (cm of H_2O)	0.1	1.5	1.1	0.2	1.5	1.16
Output						
RE (%)	60	100	97.2	66.8	100	93.2
EC (g/m^3/h)	0.3	5.3	2.93	0.5	5	3.18

Table 3. Range of values of network parameters chosen for estimating the best network architecture using full factorial design of experiments.

Parameters	Values
Neurons, N_H	4-12
Training count, Tc	1000-16000
Learning rate, η	0.1-0.9
Momentum term, μ	0.1-0.9
Best R^2	1
Error tolerance	0.0001

Table 4. Best architecture obtained with different values of network internal parameters

N_I	N_H	N_O	T_C	η	μ
4	4	2	16000	0.9	0.9

0.9825, for EC, R^2 value-0.9982).

Predictive Capability of the Model

The RE and EC values predicted by the ANN model is illustrated in Figs. 2 and 3 for the training data. It is quite apparent that, while predicting the RE and EC, the network was able to exactly map the data points. However, two or three data points were not adequately mapped by the network during training. This might have been caused by the step increase in loading rates where the microbes were reacclimatizing themselves to attain new steady states. After training, the network was provided with a separate set of data for testing the developed model. The results, presented as EC and RE, are illustrated in Figs. (4) and (5), respectively. A comparison between the EC and RE values predicted by the model with the experimental values reveals the predictive capability of the model. The model was able to adequately identify the low and high peaks in the EC and, RE values. The R^2 values obtained during training and testing were greater than 0.98 which indicated that the predictions are accurate with best network architecture of 4-4-2.

CONCLUSIONS

A laboratory-scale immobilized cell biofilter for removing NH_3 vapors showed RE higher than 90% at loading rates less than 4.5 g $NH_3/m^3/h$. This study explored the application of ANN as a performance prediction tool for a biofiltration process. The ANN model showed the ability to predict extreme operating conditions and address performance with R^2 values greater than 0.98 for training and test data sets. The best network architecture (4-4-2) during effective training of the model was determined by 2^k factorial design. The results from this study suggest that neural networks can capture and extract complex relations among easily measurable parameters in a biofiltration process and predict performance. In addition, the model may be universal and can be used for the predictions of similar systems for elimination of VOCs.

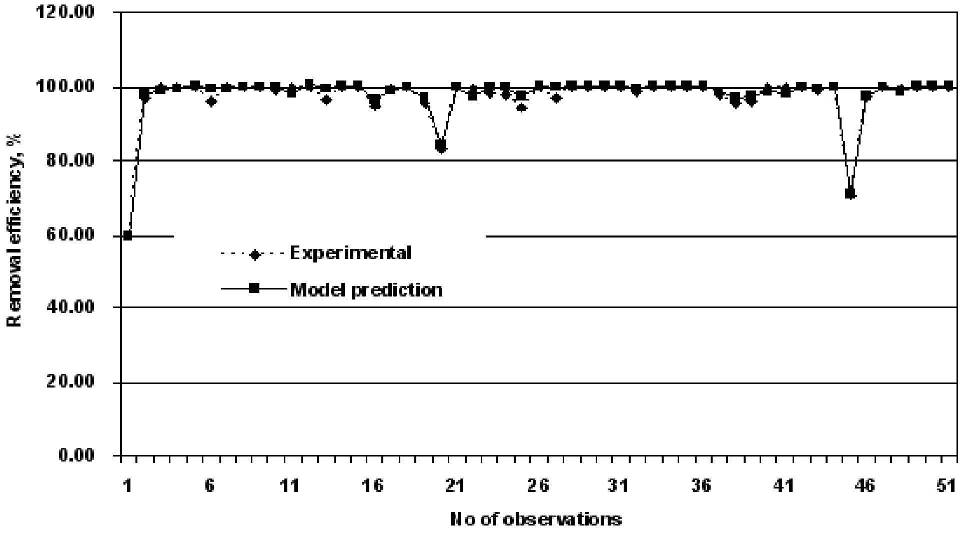

Fig. 2. Comparison of experimental and predicted values of removal efficiency during model training (N_{Tr}-51).

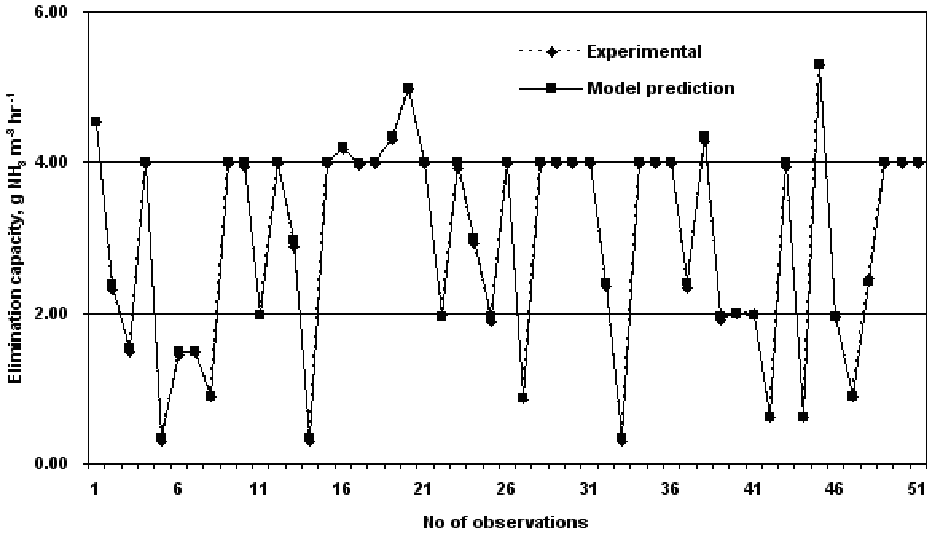

Fig. 3. Comparison of experimental and predicted values of elimination capacity during model training (N$_{Tr}$-51).

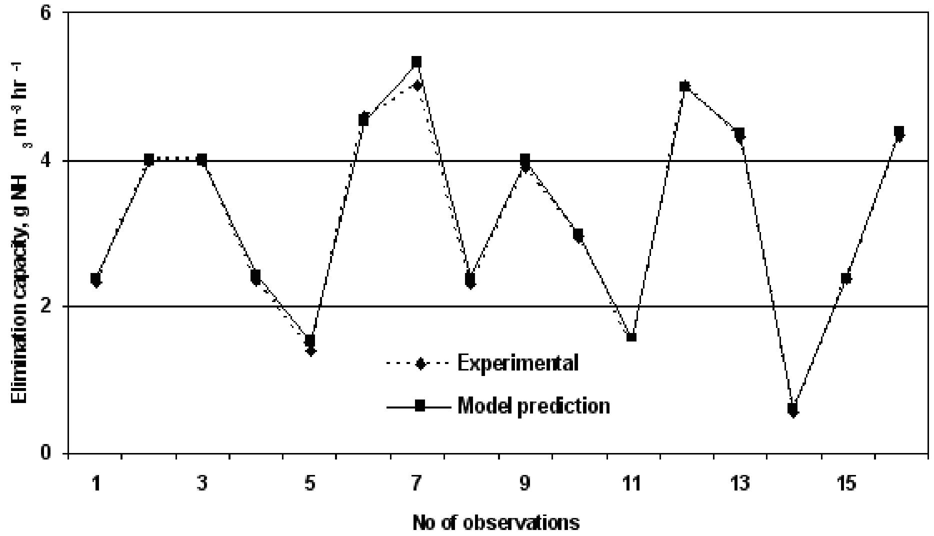

Fig. 4. Comparison of experimental and predicted values of removal efficiency during model testing (N$_{Te}$-16).

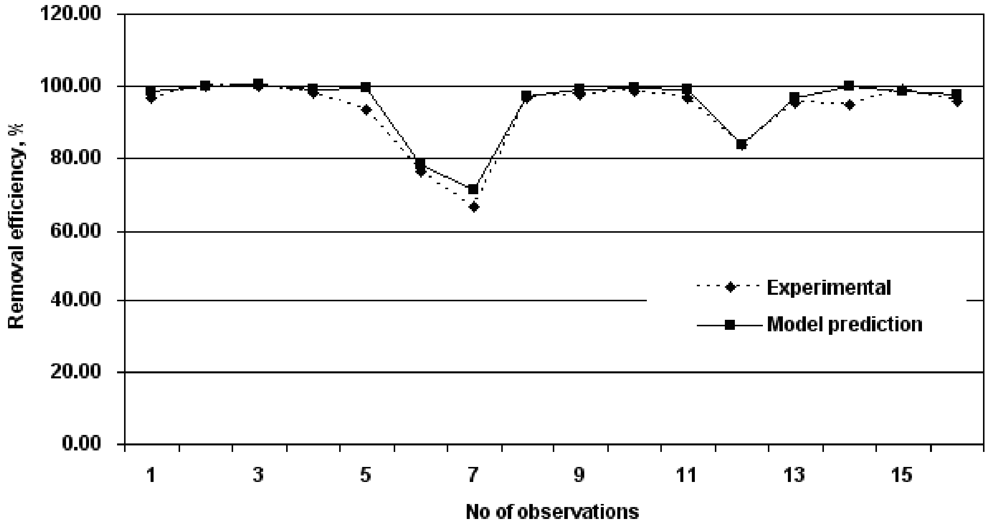

Fig. 5. Comparison of experimental and predicted values of elimination capacity during model testing (N$_{Te}$-16).

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