Journal of the Nigerian Association of Mathematical Physics
***Volume* 19 (November, 2011), pp 395 – 398**

Predicting the Biodegradation of Polycyclic Aromatic Hydrocarbons (PAHs) in Continuous Stirred Tank Reactor by Artificial Neural Network

****Owabor, C.N.and Ayodele,B.V.***
Department of Chemical Engineering,
University of Benin, Benin City, Nigeria.

Abstract

This study investigates the prediction of biodegradation of polycyclic aromatic hydrocarbons using a mixture of naphthalene; anthracene and pyrene in a continuously stirred tank reactor by an artificial neural network. Artificial neural networks are relatively crude electronic networks of "neurons" whose operations are based on the neural structure of the brain. They process records one at a time, and "learn" by comparing their prediction of the record (which, at the onset, is largely arbitrary) with the known actual record. Experimental data were employed in the design of the feed forward neural networks for modeling the prediction of biodegradation process. Comparatively, results showed that predictions from the feedforward neural network closely fitted the measured values. The degradation pattern was characterized by an exponential decline in the concentrations of the polycyclic aromatic hydrocarbons, and this was followed by a 'plateau' concentration signifying the attainment of endpoint of the degradation process.

Keywords: Model, Neuron, Feed forward, Training, Input, Hidden and Output layers.

1.0 Introduction

In recent years, there has been growing interest on appropriate bioremediation technology for cleaning up environment contaminated by polycyclic aromatics hydrocarbons (PAHs). This is due to the potential deleterious effect of PAHs on human health. The success of the technology depends on the biodegradation process, including the rate, extent, pathways and related mechanisms. While extensive reports have been made on PAH degradation by pure cultures and enrichment in natural sediments [1; 2; 3; 4; 5; 6; 7; 8] there is surprisingly, lack of accurate data on biodegradation rate especially across various types of aquatic/sediment systems. Studies on the prediction of PAH biodegradation are also limited. One of the ways of predicting PAHs biodegradation is through development of mathematical models. Examples of such models are mass transfer and biodegradation models which were developed to estimate the treatment period needed for a complete removal of PAHs from contaminated soils under batch conditions [9; 10].The extent at which the overall biodegradation rates were affected by the accumulation of the intermediate during phenanthrene degradation has been predicted by the mathematical calculation of permeability based on the membrane transport model [11]. The fate and degradation of chemical pollutants in soil/water microcosm systems has also been evaluated using a mathematical model based on mass balance equations, which integrated Fick's law, a linear isotherm, pseudo-first order kinetics and mono kinetics [12;13].

Though the methods highlighted above accurately predicted PAHs biodegradation in the systems specified, limited information on degradation kinetics, unknown biodegradation mechanisms and inherent biological variations of the biodegradation process restricted their applications. Artificial neural networks (ANNs) have been shown to provide a simple and flexible approach to a non-linear regression problem in environmental sciences. Some recent applications include statistical downscaling [14], water level-discharge modeling [15], river stage forecasting [16] and air quality forecasting [17]. However, the application of ANN as a tool for the continuing discourse on PAHs biodegradation is yet to gain public knowledge. The present study aims at investigating the feasibility of ANN to predict the biodegradation of tests PAHs (naphthalene, anthracene and pyrene in aqueous-sediment matrix.

2.0 Material and Methods

2.1 Data Collection

A large number of data sets are important in order to have valid net training since an artificial neural network learns by examples. A representative training data set should include the cause-effect relationship between the input and output variables. In this case, the sets of training and testing data were taken from experiments performed in a continuous stirred tank reactor (CSTR) by [8]

2.2 Feed forward Neural Network Architecture

Initial PAH concentration, biodegradation time and biomass concentration were the input variables making the number of

*Corresponding author: Owabor, C.N., E-mail: Owabor4you@yahoo.com, *Tel*.: +2348034101218.

neurons in the input layer to be equal to 3. The study takes into account two hidden layer. The output layer is considered to be the percentage biodegradation of the PAHs which contain only one neuron. The three layer network architecture is summarized in Figure 1. Data were flown in a forward direction from the input layer to the output in this architecture. Each neuron was connected to all the neurons in subsequent layer. This makes it a fully connected structure. A weight coefficient was attached to each connection between two neurons.

2.3 Model formulation

A universal algorithm for the feed forward neural network was formulated by [18] as follows:
For an input layer neuron, the output (X_o) is equal to its input (X):

$$X_{oi}^{L} = X_{i}^{L} \ \ (i = 1,2 \dots n; L = 1,2 \dots S) \quad (1)$$

The subscript i indicates the ith input layer neutron and L indicates the Lth trained sample. N is the number of input layer neurons, and S is the number of trained samples.
The outputs from the input layer were weighted and fed in the hidden layer. The net input of the jth hidden layer neuron is the Lth trained sample was:

$$net_{j}^{L} = \sum_{n=1}^{n} X_{oi}^{L} W_{ij}^{L} \quad (j = 1,2 \dots n; L = 1,2 \dots S) \quad (2)$$

Where W_{ij}^{L} indicates the weight of the connection between the ith input layer neuron and the jth hidden layer neuron in the Lth trained sample, and m is the number of hidden layer neuron.

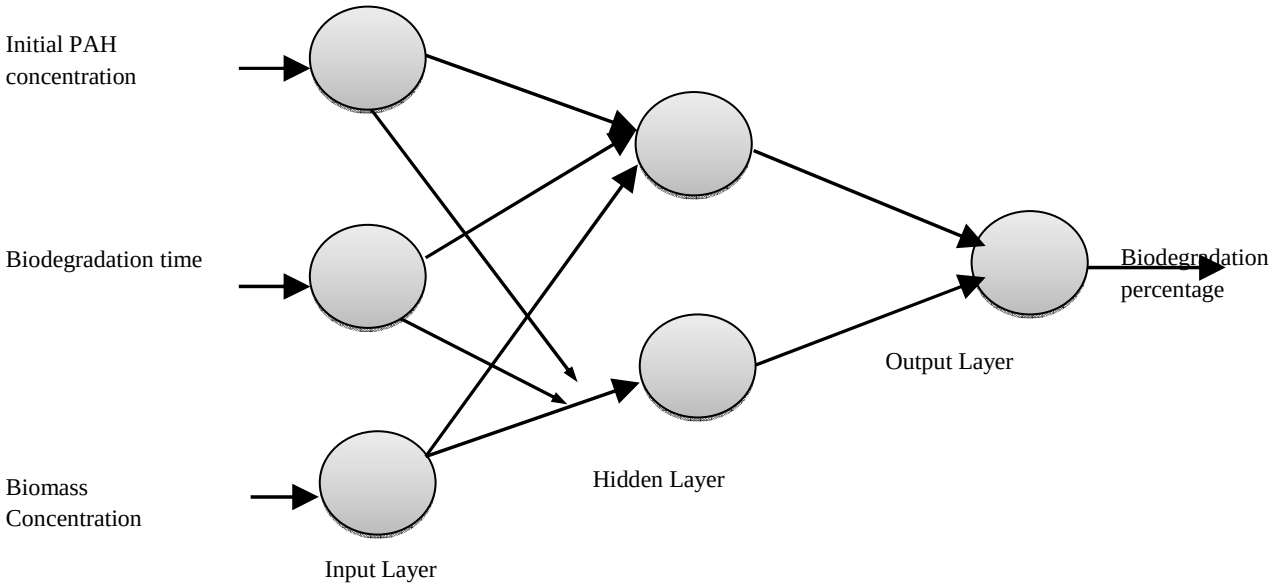

Figure 1: Three-layer feed forward neural network showing input, hidden and output layers.

The outputs of the hidden layer neuron (Y_{ok}^{L}) were given after transformation by the sigmoid function:

$$Y_{ok}^{L} = f(net_{k}^{L}) \qquad (k = 1,2 \dots n) \quad (3)$$

Here $f(x)$ is the sigmoid function:

$$f(x) = \frac{1}{1+e^{-x}} \quad (4)$$

The output of the output layer neuron was the summation of the weighted output from hidden layer neuron and was transformed:

$$O_{i}^{L} = f\left(\sum_{j=1}^{m} Y_{oj}^{L} V_{jl}^{L}\right) \qquad (l = 1,2 \dots p; L = 1,2 \dots S) \quad (5)$$

Here V_{jl}^{L} is the weight connecting jth hidden layer neuron to the lth output layer neuron.
After carrying out the procedures mentioned above for all combinations of input signals, the root-mean-square-error (RMSE) was calculated by

$$RMSE = \sqrt{\sum_{\alpha=1}^{p} \sum_{\beta}^{S} \frac{E_{\alpha\beta}^{2}}{pS}}, \ E_{\alpha\beta} = \frac{Y_{\alpha}^{\beta} - O_{\alpha}^{\beta}}{Y_{\alpha}^{\beta}} \quad (6)$$

Here p is the number of output layer neuron (p=1 in the present study because the biodegradation percentage was the only one neuron in the output layer). And is the number of trained sample, Y_{α}^{β} is the target value of βth trained sample.
Every weight with its initial value randomly generated by the computer was changed automatically. Training was repeated until the RMSE values fall into an acceptable region:

$$\Delta W_{ij}^{L}(t) = \eta\delta_j^L X_{ij}^L + \mu\Delta W_{ij}^{L}(t-1) \;\; (i = 1{,}2 \ldots n; j = 1{,}2 \ldots m; L = 1{,}2 \ldots S) \quad (7)$$

$$\Delta V_{ij}^{L}(t) = \eta\xi_i^L Y_{ok}^L + \mu\Delta V_{kl}^{L}(t-1) \;\; (k = 1{,}2 \ldots n; l = 1{,}2 \ldots m; L = 1{,}2 \ldots S) \quad (8)$$

Here t is the number of iteration of the neural computation; η is the learning rate and determines how fast the change is. $\Delta W_{ij}^{L}(t)$and $\Delta V_{ij}^{L}(t)$ were calculated in the iteration cycles. Where the momentum constant which prevents sudden change in direction in which corrections were made is **μ**. The errors ξ_i^L for the hidden layer ith neuron and δ_j^L for the output layer jth neurons, respectively, were expressed as:

$$\xi_i^L = (Y_l^L - O_l^L) f'(O_l^L) \qquad i = (1.2 \ldots\ldots p) \quad (9)$$

and

$$\delta_j^L = \sum_{\alpha=1}^{p} \xi_\alpha^L V_{\alpha j}^L f'\left(y_j^L\right) \qquad j = (1.2 \ldots\ldots m) \quad (10)$$

Here f' is the derivative with respect to x of the transformation function, and

$$f'(x) = (1 - f(x) \times f(x) \quad (11)$$

3.0 Results and Discussion

One of the important tools of ANN is the selection of the number of neuron in the hidden layer. In order to obtain the best neural network structure, different numbers of hidden layer neuron from one to six were tested in this study. The number of neuron in the hidden layers for percentage PAH biodegradation was determined according to the values of RMSE and this is shown in Figure 2. The RMSE decreased with neuron number of hidden layer. The four neurons had the lowest RMSE, and the value then increased slightly with subsequent addition of the hidden layer neurons. The final selected architecture consists of two hidden layers as shown previously in Figure 1.

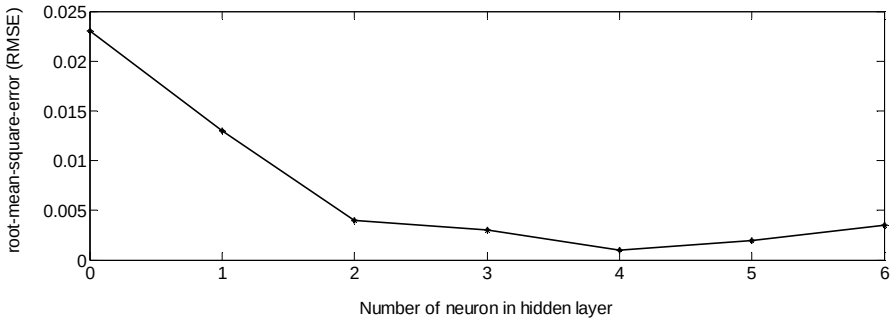

Figure 2: Number of neurons selection in the hidden layer for the anthracene biodegradation

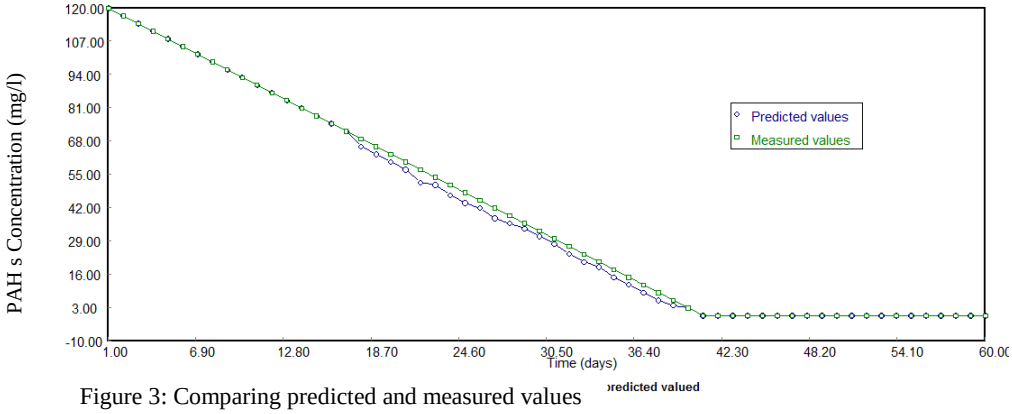

Figure 3: Comparing predicted and measured values

A parity plot was used to compare the experimental values with the predicted values for PAH biodegradation using the feedforward neural network. The prediction of the biodegradation of PAHs is shown in Figure 3. Most of the data points were well predicted. The result indicates a close prediction by the neural network. The observed deviation in the result could be attributed to factors such as temperature of the soil, pH of the soil etc which were not taken into account in the work and thus not used as input layer neurons during the training of the feedforward neural network. On the one hand, omission of certain factors in the input layer which likely may affect the output layer could be the primary reasons for failure of the developed Artificial Neural Network in accurately predicting biodegradation. The observed variation is consistent with the literature report of [19]. On the other hand, the deviation could also be as a result of unknown human error while conducting the biodegradation experiment.

Three neurons were found to be most appropriate for the input layer of the feedforward neural network in this present study. The introduction of time as one of the input layer neurons ensures that all of the data set was independent and that the percentage PAH biodegradation was predicted at different time. This is significant because for bioremediation of PAHs in contaminated environments, biodegradation rate and percentage are important indicators. Since time was included in the input layer neuron, the biodegradation rate could not be used in the output layer neuron and only the biodegradation percentage was determined as a single output layer neuron. However, for high molecular PAHs such pyrene and benzo[a]pyrene it is difficult to employ just a single biodegradation constant to describe their biodegradation rate. It would be simpler to use biodegradation percentages at different time to indicate the trend of the biodegradation rate.

Conclusion

It can be established from this study that the feed forward neural network, a common model of ANN is a very useful tool for predicting the biodegradation of PAHs in a continuously stirred tank reactor. The architecture of two-layer model was determined using three neurons in the input layers, two neurons in the hidden layer based on the lowest RMSE (0.0015) for the percentage biodegradation and one neuron in the output layer. The predicted values from the feed forward neural network are almost the same with experimental values.

References

[1] Bauer, J.E., and Capone, D.G. (1985) "Degradation and mineralization polycyclic aromatic hydrocarbons anthracene and naphthalene in intertidal marine-sediments." Applied and environmental microbiology, 50: 81-90.

[2] Shearis, M.P., (1989). "Seasonal biotranformation of anthracene, phenathrene and benzo[a]pyrene in surficial estuarine sediment." Applied and environmental microbiology, 55: 1391-1399.

[3] Wilson, S.C. and Jones, K.C., (1993). "Bioremediation of soil contaminated with Polynclear Aromatic Hydrocarbon (PAHs) - A Review." Environmental Pollution, 81: 229 – 249.

[4] Guerin, T.F.(1999). "Bioremediation of phenols and polycyclic aromatic hydrocarbons creosote contaminated soil using ex-situ land treatment." Journal of Hazardous Material. 65: 305-315.

[5] Hwang, S.C. and Cutright, T.J., (2002). "Biodegradability of aged pyrene and phenathrene in a natural soil." Chemosphere, 47: 891 – 899.

[6] Lei, L., Khodadoust, A.P., Suidan, M.T., and Tabak, H.H. (2005). "Biodegradation of sediment-bond PAHs in field contaminated soil." Water Research, 39: 2997 – 2404.

[7] Kim, H.M., and Lee, C.G. (2007). "Microbial degradation of polycyclic aromatic hydrocarbons in soils by bacterium-fungus co-cultures. Biotechnology and Bioprocess Engineering, 12: 410-416.

[8] Owabor, C.N. (2007). "Substrate bioavailability and biodegradation of naphthalene, anthracene and pyrene on contaminated aqueous-soil matrix", Ph.D Thesis, University of Lagos, Nigeria.

[9] Mulder, H., Breure, A.M., and Rilkens, W.H., (2001). "Prediction of complete bioremediation period for PAH soil pollutants in different physical physical state by mechanistics models." Chemosphere, 43: 1085-1095.

[10] Owabor, C.N., Ogbeide, S.E. and Susu, A.A. (2010). "Degradation of polycyclic aromatic hydrocarbons: Model simulation for bioavailability and biodegradation". Can. J. Chem. Eng. Vol. 88, Issue 2, 268-276.

[11] Ahn, C.K., Woo, S.H., Lee, D.S., and Park, J.M. (2006). "Mathematical evaluation of intermediate accumulation during microbial phenanthrene degradation." Korean Journal of Chemical Engineering, 23: 415-418.

[12] Liu, L., Tindal, J.A., Friedel, M.J., and Zhang, W.X., (2007). "Biodegradation of organic chemicals in soil/sediment microcosm system: model development." Water Air and Soil Pollution. 178: 131-143.

[13] Chang, J.S., Lu, S.C., and Chiu,Y.L., (2007). "Dynamic modeling of batch polymerization reactors via the hybrid neural-network rate-function approach." Chemical Engineering Journal , 130: 19-28.

[14] Harpham, C and Wilby, R.L., (2005). "Multi-site downscaling of heavy daily precipitation occurrence and amounts." Journal of Hydrology, 312: 235-255.

[15] Battacharya, B and Solomitine, D.P. (2005). "Neural network and M5 model trees in modeling water level-discharge relationship." Neurocomputing, 63 : 381-396.

[16] Dawson, C.W., See, L.M.,Abrahart, R.J., Wilby, R.L., Shamseldin, A.Y., Anctil, F., BelBachir, A.N., Bowden, G., Dandy, G., Lauzon, N. and Maier, H. (2005). "A comparative study of artificial neural network techniques for river stage forecasting." In proceedings of the IEEE international joint conference on neural networks: Vol 4, 2666-2670.

[17] Schlink,U., Dorling, S., Pelikan, E., Nunnari, G., Junninen, H., greig, A., Foxall, R., Eben, K, Chatterton, T., Vondraceh, J., Richter, M., Dustal,M., Bertucco,L, Kolehmainen,M., and Doyle,M.,(2003). "A rigorous inter-comparison of ground-level ozone predictions." Atmospheric Environmental Microbiology, 51: 1028-1035.

[18] Zhang,W.Q., Tray, J.H, Maszenen, A.M., Krumholz, L.R and Tray, S.L.T.(2003). "Importance of Grain-positive naphthalene degrading bacteria in oil-contaminated tropical marine sediments." Letters in Applied Microbiology, 36: 251-257.

[19] Kovesdi, I., Domiuguez-Rodiguez, M.F., Orfi, L., Narcy-Szabo, G., Varro, A., Papp, J.G. and Matyus, P., (1999). "Application of neural networks in structure –activities relationships. Medicinal Research Reviews , 19: 249-269.