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Estimation of Reaction Parameters for Phenol Biodegradation Using Trainable Artificial Neural Networks

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Biological systems typically respond non-linearly to the external stimuli such as food availability or toxic exposure. Analytical models based on empirical and semi-empirical representations only simulate a narrow range of conditions. Simulation of the developed kinetic laws on wider scale normally fails. A black box approach is normally applied to simulate responses outside of the studied range. One such system that could be used without worrying about the internal mechanisms is the Trainable Artificial Neural Network (TANN). This system offers the capability to predict the next steps in the behaviour of the system using data from the past. In this study, a multi-layer feed forward neural network was capable of simulating phenol biodegradation without a kinetic model. The network consisted of an input layer with three neurons, a single hidden layer with ten neurons and an output layer with one neuron. Network testing achieved a Mean Squared Error (MSE) of 0.001 with the regression coefficient (R^2) of 0.984. The predicted trend was validated by substrate inhibited Monod-type kinetic using actual experimental data from a batch system using a laboratory enrichment of an environmental sample containing Pseudomonas aeruginosa. Both the model and the TANN was further tested against literature data from Chirwa and Wang (2000), where the researchers achieved simultaneous Cr(VI) reduction and phenol degradation using an anaerobic consortium of bacteria containing *Escherichia coli* ATCC 33456. In all cases tested, using the TANN algorithm ahead of the kinetic model generated a dataset that reduced convergence time during parameter search in the highly non-linear reaction kinetics.

1. Introduction

Phenol is a commonly found pollutant in industrial waste effluents from factories of iron-steel, coke, petroleum, pesticide, paint solvent, pharmaceutical, wood processing chemicals, pulp and paper (Kavuri, 2011). The Environmental Protection Agency (EPA) has proposed a water purification standard for phenol in surface water due to the documented effect of phenol on aquatic life. Aromatic compounds and halogenated aromatics are water-soluble and highly mobile, resulting in resistance to degradation (Collins and Daugulis, 1997). For more complex phenolic compounds, analytical solutions for the complex kinetic models arising from complex metabolic pathways are usually impossible to solve. The degradation of the compounds is typically highly inhibited and sometimes interlinked with electron transport in the conversion of inorganic substances such as metals (Chirwa and Smit, 2010). An example of a more simplified version is presented by Chirwa and Wang (2000, 2001) and later articles by the same researchers. I the previous studies, a Monod-type kinetic for phenol degradation was suggested in which phenol degradation transitioned from first-order kinetics at low concentration to zero-order kinetics at high concentrations with substrate level inhibited Monod kinetics at very high phenol concentrations as shown in Eq(1) below:

$$-\frac{dS}{dt} = \frac{k_{ms} \cdot S \cdot}{K_s + S + S^2 / K_I} X \tag{1}$$

where k_{ms} = maximum substrate (phenol) utilisation rate coefficient (h^{-1}), K_s = half velocity concentration ($mg.L^{-1}$) and K_I = inhibition coefficient ($mg.L^{-1}$). Simulation of the developed kinetic laws on a wider scale, normally fails when the system is operated under conditions outside the range in which the model was

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tested and optimised. Neural network and fuzzy logic systems came into being in an effort to answer the limitation of the mechanistic simulations (Hajmeer and Basheer, 2002).

1.1 Application of Trainable Artificial Neural Networks (TANN)

TANNs are nonlinear computational systems used in approximating the behaviour of systems associated with explicit modelling difficulties without assuming the type of relationship and degree of nonlinearity between various independent and dependent variables. TANNs have a wide range of application and the ability to handle problems in noisy and highly complex nonlinear data (Balan et al., 1999).

Conventional kinetic modelling entails certain assumptions about kinetic equations and requires experimental estimation of kinetic constants. The assumptions made usually increase calculation errors and model feasibility is limited to reaction conditions in which the model and associated constants were determined.

Parallel, real time processing is achieved with TANNs whereas traditional modelling is sequential, logical and restricted by rules/algorithms. TANNs are known as universal function approximators, capable of adaptive learning and self-organization (Anjum et al., 1997). Their current limitation is that they are most interpolative in nature, thus they cannot be relayed upon to estimate points outside the current training dataset.

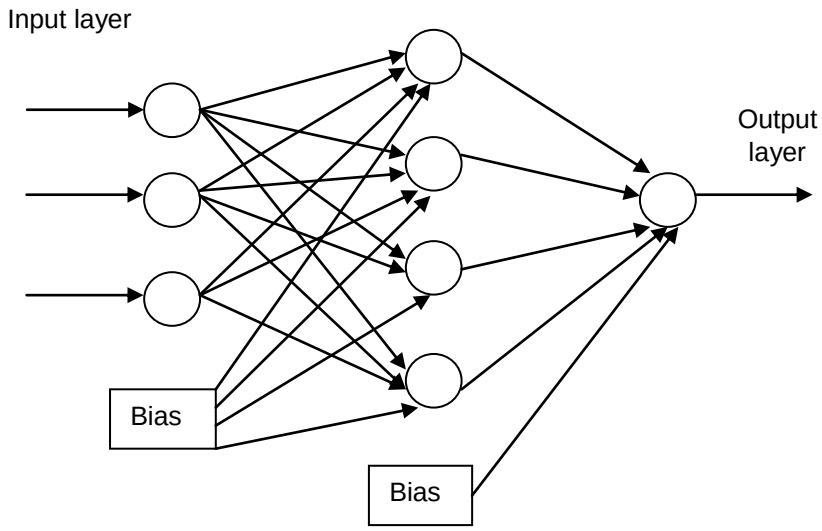

Figure 1: Schematics of a multi-layer artificial neural network.

Artificial neurons accept different signals from neighbouring neurons and process them in a predefined simple way. The neuron either fires an output signal or not, depending on the outcome of this processing. The weights in the artificial neuron decide what proportion of the incoming signal is transmitted into the neuron body. A negative weight reflects an inhibitory connection, while positive values designate excitatory connections.

The output signal can be either zero or one, or have any real value between zero and one, for binary or real valued artificial neurons. The input signals are normalised between zero and one, and are regarded as non-active layers of neurons serving only to distribute the signals to the first layer of active neurons.

The function which calculates the output from the input vector is composed of two parts. The first part evaluates the net input and the first function is a linear combination of the input variables multiplied with the weight coefficients. The second part transfers the net input in a non-linear manner to the output value. The net input is calculated using Eq(2) and the output value is determined using Eq(3).

$$Net_j = \sum_{i=1}^{m+1} w_{ji} x_i \tag{2}$$

$$y_i = \frac{1}{1 + e^{-(Net_j)}} \tag{3}$$

where N_{etj} = net input, w_{ji} = weight coefficient, x_i = input variable, and y_i = output variable. The subscripts and superscripts m, i, and j are iteration numbers.

2. Error Back-Propagation Training

Artificial neurons try to mimic the adaption of synapse strength by iterative adaption of weights in neurons, according to the difference between the actual obtained outputs and the targets. Neurons in error back-propagation learning try to yield quantitatively an answer as close as possible to the target, with the order of weight correction enabling accurate determination of the error in each output node. Weight corrections are done according to Eq(4), after each input-target pair has produced an output vector.

$$\Delta w_{ji}^{i} = \eta \delta_{j}^{i} y_{i}^{i-1} + \mu \Delta w_{ji}^{i(previous)} \tag{4}$$

The assumption is made that the errors have been evenly distributed when the signals were passing from the last hidden layer to the outer layer. The δ_j^l term, calculated using Eq(5) represents the error that occurs on a specific hidden layer. The output layer error, δ_j^{last}, is calculated according to Eq(6).

$$\delta_{j}^{i} = \left(\sum_{k=1}^{k} \delta_{j}^{i+1} w_{j}^{i+1} \right) y_{j}^{i} \left(1 - y_{j}^{i} \right) \quad \text{for l = 1,..., last – 1} \tag{5}$$

$$\delta_{j}^{last} = \left(t_{j} - y_{j}^{last} \right) y_{j}^{last} \left(1 - y_{j}^{last} \right) \tag{6}$$

where k = number of output units, t_j = target value, η = learning rate, μ = momentum, and δ_j = error term. Learning by error back-propagation is carried out in epochs, where one epoch is a period in which all input-target pairs are presented once to the network. After each epoch, the MSE is reported according to Eq(7) (Engelbrecht, 2007).

$$Mean\ Square\ Error = \frac{\sum_{p=1}^{p_T} \sum_{k=1}^{k} \left(t_{k_p} - y_{kp} \right)^2}{p_T k} \tag{7}$$

in which p = individual patterns, p_T = total number of patterns. The accuracy of the predictions of the neural network is quantified by the MSE difference between the measured and the predicted phenol biodegradation (Arce-Medina and Paz-Paredes, 2009). The MSE is used as the objective function and supervised training aims to reach the smallest possible MSE in the shortest possible time.

3. Artificial Neural Network Setup

A multi-layer feed-forward neural network with error back-propagation was used to develop the model. The network was designed with the Neural Network Toolbox, MATLAB (Beale et al., 2013). Published data from earlier studies (Chirwa and Wang, 2000) and later (Chirwa and Wang, 2001) was used in the evaluation of degradation parameters. The inputs of the neural network were time, chromium (VI) (Cr(VI)) concentration and initial phenol loading concentration, and the network output was the percentile of phenol biodegraded. The network's predicted phenol biodegradation was compared to the actual phenol biodegradation achieved during the laboratory study.

It is important to note that the neural network feasibility greatly depended on the range of data used during the design process. The test set data was utilized to evaluate the prediction accuracy of the network using data outside the range covered in the training and control data sets.

A log-sigmoid transfer function was used in the hidden layer of the multi-layer network and a linear transfer function was used as function approximator in the final layer of the network. During the learning procedure the experimental data was divided into three sets namely a training set, a control set and a final test set. The training set contained 70 % of the data and the control and final test sets each contained 15 % of the data.

In this study, training was performed to minimize the MSE by adjusting the weights and biases. The control set was used to find the best neural network configuration and training parameters.

4. Results and Discussion

The neural network was design as a multi-layer feed forward neural network, with an input layer, a single hidden layer and an output layer (Figure 2). Good correlation was achieved during training, shown in Figure 3, with a minimized MSE of 0.001. The neural network design was optimized using the control data set and the optimum network configuration is depicted in Figure 3. The input layer consisted of three neurons, with ten neurons in the hidden layer and one neuron in the output layer.

The control data set achieved high correlation, shown in Figure 4, for the optimized network configuration. The initial phenol loading concentrations utilized during testing exceeded to maximum training and control set data range by 20 %. The correlation between the estimated and experimental biodegradation for the test set data is shown in Figure 5. The neural network exhibited high correlation between experimental and estimated biodegradation for input values exceeding the initial data range. Excellent prediction accuracy with a high regression coefficient of determination (R^2 = 0.984) was obtained, demonstrating the reliability, accuracy and feasibility of the network for future phenol biodegradation estimations.

The optimum parameters determined in this study were in agreement within the acceptable range of biological parameters for phenol degradation in the presence of Cr(VI) with less than 5 % error difference in the maximum phenol degradation rate coefficient, k_{ms} = 0.033 $L.h^{-1}$ and K_s = 833 $mg.L^{-1}$ this study versus k_{ms} 0.035 $L.h^{-1}$ and K_s = 940 $mg.L^{-1}$ from Nkhalambayausi-Chirwa and Wang (2005).

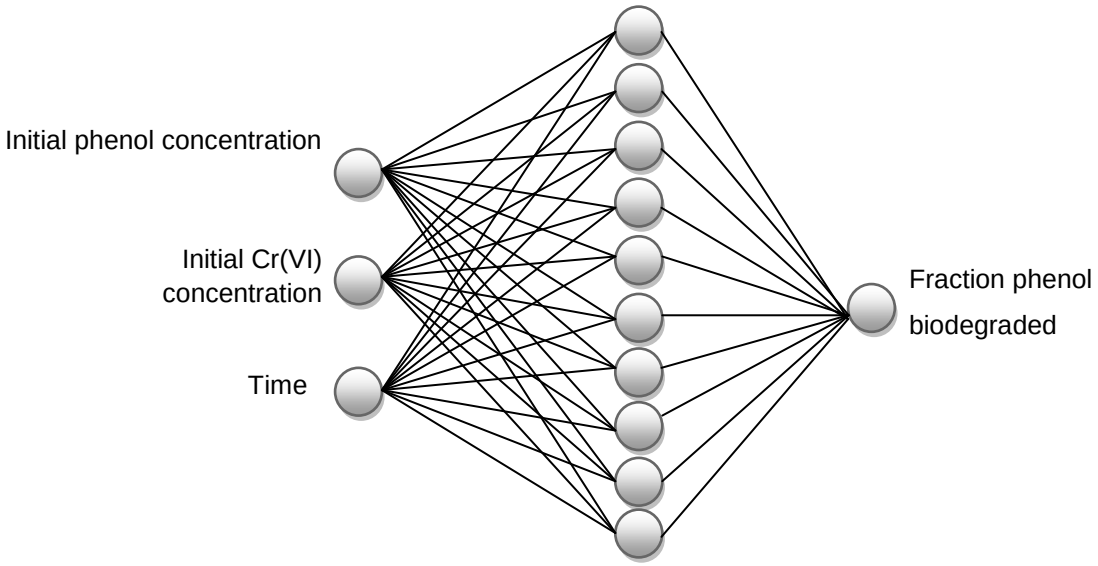

Figure 2: Neural network design architecture.

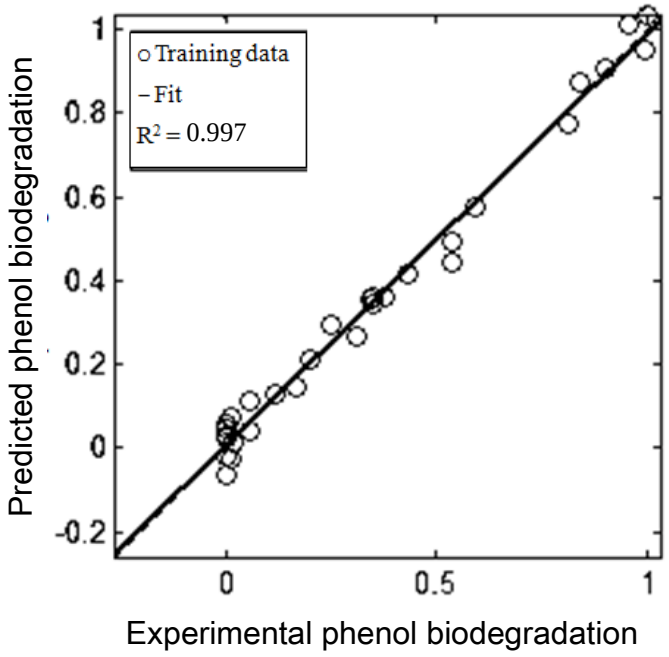

Figure 3: Correlation between estimated and experimental biodegradation for the training set data.

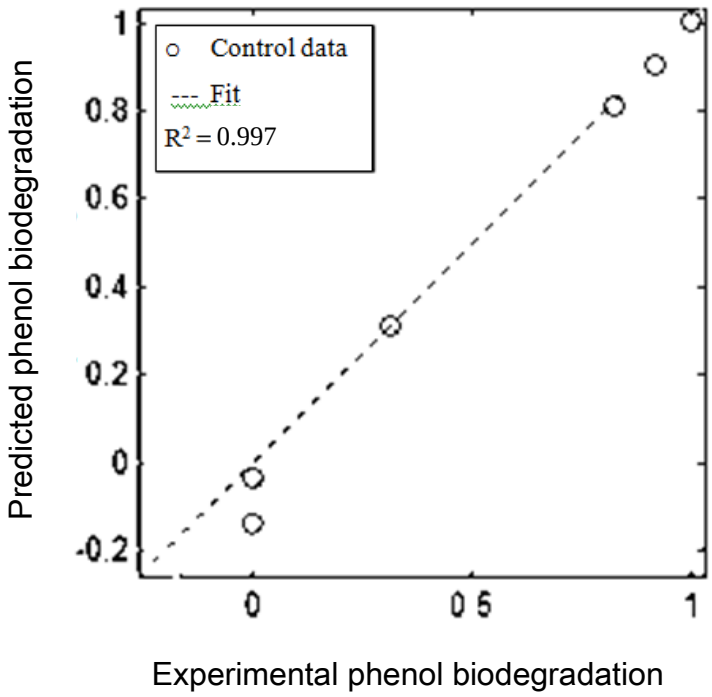

Figure 4: Correlation between estimated and experimental biodegradation for the control set data.

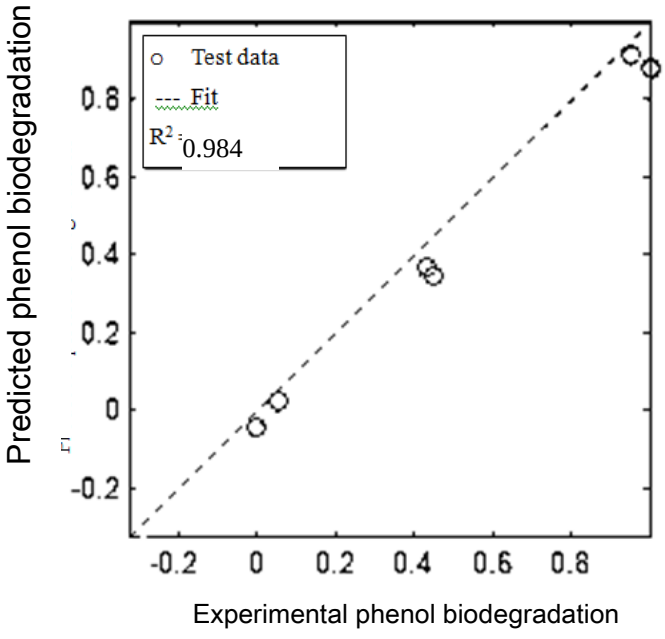

Figure 5: Correlation between estimated and experimental biodegradation for the test set data.

5. Conclusions

A multi-layer feed forward neural network consisting of an input layer with three neurons, a single hidden layer with ten neurons and an output layer with one neuron, provided good correlations between experimental and estimated biodegradation for the training and control data sets used. The model accuracy extended towards data up to 20 % outside of the training range while maintaining a MSE of 0.001. The regression coefficient of determination (R^2 = 0,984) obtained demonstrated the reliability, accuracy and feasibility of the network for future phenol biodegradation predictions. The neural network only considers the effect of initial phenol loading and inhibitor concentrations on phenol biodegradation, but previous studies have shown that temperature, pH, aeration and agitation also influence phenol biodegradation. It is recommended that the neural network be expanded to incorporate these parameters, allowing computation of optimal control inputs.

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