World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:6, No:8, 2012

A Critics Study of Neural Networks Applied to ion-Exchange Process

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***Abstract*—**This paper presents a critical study about the application of Neural Networks to ion-exchange process. Ion-exchange is a complex non-linear process involving many factors influencing the ions uptake mechanisms from the pregnant solution. The following step includes the elution. Published data presents empirical isotherm equations with definite shortcomings resulting in unreliable predictions. Although Neural Network simulation technique encounters a number of disadvantages including its "black box", and a limited ability to explicitly identify possible causal relationships, it has the advantage to implicitly handle complex nonlinear relationships between dependent and independent variables. In the present paper, the Neural Network model based on the back-propagation algorithm Levenberg-Marquardt was developed using a three layer approach with a tangent sigmoid transfer function (*tansig*) at hidden layer with 11 neurons and linear transfer function (*purelin*) at out layer. The above mentioned approach has been used to test the effectiveness in simulating ion exchange processes. The modeling results showed that there is an excellent agreement between the experimental data and the predicted values of copper ions removed from aqueous solutions.

***Keywords*—**Copper, ion-exchange process, neural networks, simulation

I. Introduction

IN recent years, ion-exchange techniques have been widely investigated for the removal of heavy metals from wastewaters. Acquiring sufficient understanding of the system to develop an appropriate conceptual, logical and then simulation model is one of the most difficult tasks in simulation analysis. In many previous studies done with the aim of predicting ion exchange profiles, the focus was on using empirical and/or fundamental equilibrium isotherms and rate equations.

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This involved resistance modeling where it is assumed that equilibrium exists at the solid liquid interface and that film or intraparticle diffusion could be rate limiting. Artificial Intelligence is a branch of science which deals helping machines finds solutions to complex problems in a more human-like fashion. This generally involves borrowing characteristics from human intelligence, and applying them as algorithms in a computer based manner. Intelligent systems adapt themselves using example situations and their correct decisions, which after the learning phase, can make decisions automatically for future situations [1]. The aim of this paper was to investigate the effectiveness in simulating ion-exchange processes by removing heavy metals such as Cu (II) from this synthetic solution.

II. Materials and Methods

A.Preparation of the adsorbent and synthetic solution

The clinoptilolite used in this study was sourced from the Vulture Creek, KwaZulu-Natal Province of South Africa. The clinoptilolite was crushed and milled into powder with average particle sizes of approximately 75µm [2]. The powder was then examined using an X-ray powder diffractometer (XRD) Phillips X'pert Model 0993 to determine its composition. Its elemental composition was determined using X-ray fluorescence spectroscopy (XRF, Phillips Magix Pro) while the surface area was analyzed using BET (Tristar 3000) [2]. The measurements were done under a nitrogen atmosphere. Prior to porosity and surface area analysis, 2g of sample was first degassed and nitrogen gas was flushed through for 4h at 120^{o}C [2]. Clinoptilolite grains of sizes in the range of 2.8 mm to 5.6 mm were used for adsorption studies. A fraction of these grains was treated in HCl at concentrations of 0.02M and 0.04M at room temperature over a period of 8h. The clinoptilolite was then washed in deionised water to remove the fine fractions and thereafter dried in the oven at 50^{o}C for 24h [2]. The solution of Cu was prepared by dissolving $CuSO_4.5H_2O$ in deionised water at pH 6.5. These solutions were assayed using atomic absorption spectroscopy (AAS), (model Varian Spectra (20/20)) [2].

B.Batch adsorption studies on synthetic solution

The Cu ion-exchange process on the clinoptilolite was conducted at room temperature. Glass columns of 2cm diameter and 30 cm of length were pre-loaded with 25g of either natural clinoptilolite (as received) or HCl-activated clinoptilolite. Aliquots of 25ml of the prepared Cu-bearing solution of desired concentrations were passed through each of the two types of zeolites.

There were afforded the same solution-zeolite contact time. After passing through the zeolite-packed column the resultant solution was assayed using atomic absorption spectroscopy (AAS) in order to ascertain the zeolite's removal efficiency. The flame type used was air-acetylene and the adsorption wavelengths for the two metals then prepared and a calibration curve was drawn using these standards. Dilution was applied stoichiometrically where the concentrations of the unknown solution of copper exceeded the standards concentration range of the standards [2].

The metal uptake percentage from the solution and distribution ration was calculated as in (1).

$$\% \text{ Uptake} = (C_o - C_f)\, x100/C_o \quad (1)$$

Where: C_o and C_f are the initial and final metal concentration respectively.

The percentage of Cu (II) ions removal as the output parameter of the Neural Network model was considered as a measure of the uptake efficiency.

C. Definition of the Neural Network model

Neural Networks (NN) are known for their superior ability to learn and classify data. The inspiration of NN came from studies on the structure and function of the brain and nerve systems as well as the mechanism of learning and responding. The potential applications include prediction, classification, data conceptualization, data filtering and optimization [3].

In this study, Neural Network Toolbox of MATLAB mathematical software was used to predict the adsorption (removal) efficiency. Hundred experimental sets were used to develop the NN model. The uptake of Cu (II) mechanisms from the pregnant solution is strongly dependent on pH, temperature and initial concentration as variable parameters. The data gathered from batch experiments was divided into input matrix and desired matrix. The procedures for obtaining reliable Neural Network models are summarized in Fig. 1. Training and validation data sets are obtained by varying inputs of the columns covering several scenarios in experiments. The data sets need to be scaled in order to overcome the significant minimum and maximum values used in the training process [4].

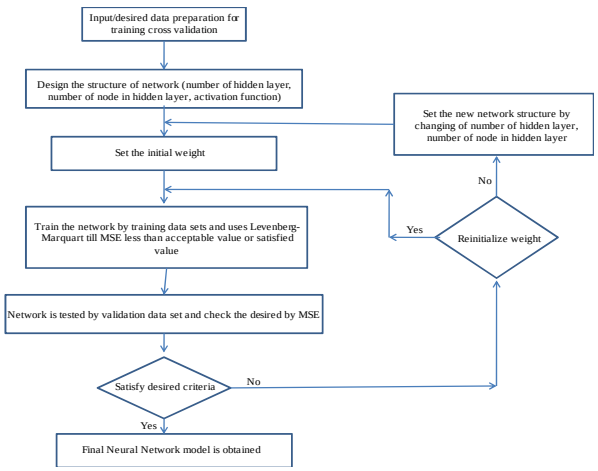

Fig. 1 Procedure for obtaining Neural Networks [4]

III. Results and Discussion

NN models could describe adsorption systems better than general rate models [5]. Even the adsorption isotherms can be represented by neural networks [6]. So, it is preferable to use a non-parametric technique such as a back-propagation NN to represent such an equilibrium relationship [7] and this have been confirmed in [8,9].

A. Optimization of NN structure

The optimal architecture of the NN model and its parameter variation were determined based on the minimum value of the (Minimum Mean Squared Error) MSE of the training and prediction set. In optimization of the network, one neuron was used in the hidden layer as an initial guess. With an increase in the number of neurons, the network gave several local minimum values and different MSE values were obtained for the training set. With 11 neurons at hidden layer, the MSE reached its minimum value. Hence, the NN containing 11 neurons at hidden layer was proposed as the best case.

Pursuing benchmark comparisons of back-propagation algorithms, a study was conducted to determine the optimal network structure. Finally, the optimal NN is shown in Fig. 1 consists of a three-layer NN, with tangent sigmoid transfer function (*tansig*) at hidden layer with 11 neurons and a linear transfer function (*purelin*) at out layer. A training set of hundred experimental data sets was selected to develop the model. NN model based on Levenberg-Marquardt Algorithm (LMA) for the experimental data was applied to train the NN. During training, the output matrix is computed by a forward pass in which the input matrix is propagated forward through the network to compute the output value of each unit. The output matrix is then compared with the desired matrix which results into error signal for each output unit. In order to minimize the error, appropriate adjustment were made for each of the weights of the network. The training was stopped after 30 iterations (TRAINLM, Epoch 30/100) for the LMA to give the desired output for a given input matrix because the differences between training error and validation error started to increase. Fig. 3 illustrates training, validation and test mean squared errors for the LMA. A regression analysis of the network response between NN outputs and the corresponding targets was performed. The graphical output of the network outputs plotted versus the targets as open circles is illustrated in Fig. 4. Taking into account the non-linear dependence of the data, linear regression shows a good agreement between NN outputs (predicted data) and the corresponding targets (experimental data).

The input vectors and target vectors was randomly divided into three sets as shown in Fig. 3. as follows: 70% was used for training, 15% was used to validate that the network is generalizing and to stop training before overfitting and the last 15% was used as a completely independent test of network generalization. The following regression plots in Fig. 4 display the network outputs with respect to targets for training, validation and test sets.

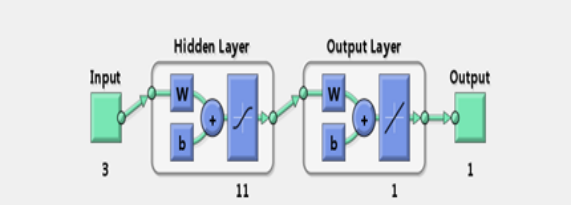

Fig. 2 Proposed NN structure

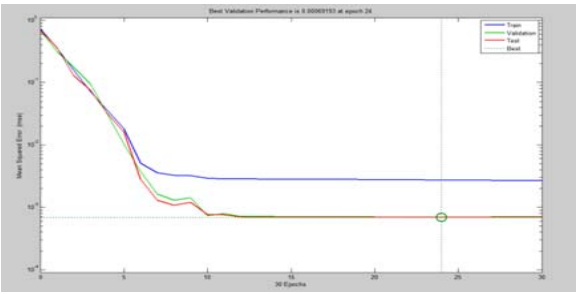
Fig. 3 Training, validation and test mean squared errors for the LMA

The fit is reasonably good for all data sets and the output tracks the targets very well for training, testing and validation, and the R-value is over 0.99 for total response.

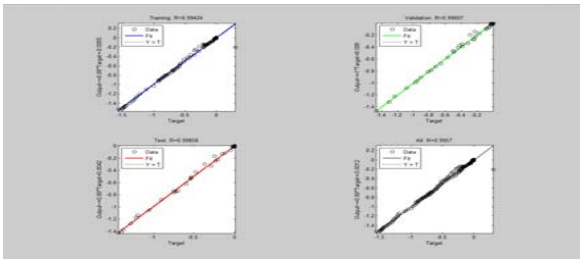
Fig. 4 Plot regression for the NN training

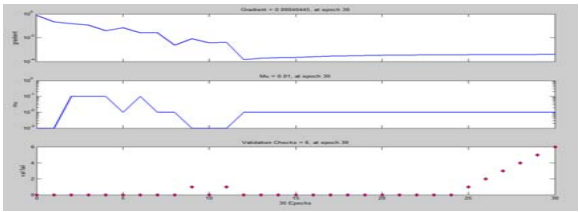
Fig. 5 Plot training state for the NN training

B.Sensitivity analysis

Sensitivity analysis was conducted to determine the degree of effectiveness of variables. Performance of the group of input vectors includes initial concentration, pH and temperature. Series of experiment resulted into the evaluation of the performance based on 70% data for training, 15% for testing and 15% for testing at 30 epochs.

During training, the process is constantly updated in the training window. Of most interest are the performance, the magnitude of the gradient of performance and the number of validation checks (see Fig. 5). The magnitude of the gradient and the number of validation checks were used to terminate the training. The gradient was become very small as the training reached a minimum of the performance.

The training was stopped when the magnitude of the gradient was 0.000254. The number of validation checks Mu in the Figure 5 represents the number of successive iterations that the validation performance failed to decrease. The training was stopped when the number of validation checks reached 6.

IV. Conclusions

A critical study of Neural Network (NN) model was developed using the Clinoptilolite as an adsorption agent to predict the efficiency of Cu uptake mechanisms from the pregnant solution. The effect of operational parameters such as initial concentration of Cu ions, solution pH and operating temperature were studied to optimize the conditions for maximum removal of Cu ions. After back-propagation (BP) training, the NN model was able to predict adsorption efficiency with a tangent sigmoid transfer function (*tansig*) at hidden layer with 11 neurons and a linear transfer function (*purelin*) at out layer. The Levenberg- Marquardt algorithm (LMA) was found as the best of 11 BP algorithms with a minimum mean squared error (MSE) of 0.00069193 at epoch 24.

Acknowledgment

Research project is financed by University of Johannesburg/ New Generation Scholarship.

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