The use of Neural Network for modeling of copper removal from aqueous solution by the ion-exchange process

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Abstract—In this study, Neural Network (NN) was applied for the prediction of ion-exchange process efficiency for the removal of Cu (II) from aqueous solutions by clinoptilolite. The effect of operational parameters such as pH, temperature and initial concentration were investigated to optimize the conditions for maximum removal of Cu (II). The NN model, based on the backpropagation algorithm Levenberg-Marquardt algorithm (LMA) has been selected to train the model. This model was able to predict removal efficiency with a tangent sigmoid transfer function at hidden layer with 11 neurons and a linear transfer function at out layer. The performance of the network for predicting the adsorption efficiency for copper was found to be very impressive.

Keywords: Neural network, copper removal, aqueous solution, ion-exchange process.

I. INTRODUCTION

A NUMBERS of purification methods are currently employed in the water purification industry and these include ultrafiltration, phytoextraction, electrodialysis, reverse osmosis and ion-exchange [1], [2]. Among the water purification technologies, ion-exchange is most commonly employed in industrial water treatment [3]. The main advantage of this process is the possibility of replacing expensive synthetic ion-exchangers by low-cost sorbents such as natural zeolites [4]. The removal of Cu (II) from aqueous solution by the ion-exchange process is quite complex, since the process is influenced by several factors [5]. Due to complexity of the process, it is difficult to be modeled and simulated using conventional mathematical modeling. Application of NN has been considered as a promising tool because of their simplicity towards simulation, prediction and modeling. The advantages of NN are that the mathematical description of the phenomena involved in the process is not required, less time is required for model development than the traditional mathematical models and prediction ability with limited numbers of experiments [6].

The use of NN on the removal of heavy metals has been reported in literature. NN were applied in biological wastewater [7], [8] and physic-chemical wastewater treatment [5]. However, few studies on applications of NN on ion-exchange process have been reported [5]. The aim of this study is to describe the ion-exchange process potential in removal of Cu (II) ions from aqueous solutions. The NN results were compared with those obtained through experiments.

II. NEURAL NETWORK

A. Basic concepts

Neural network (NN) is an information processing system that is inspired by the way such as biological nervous systems e.g. brain. The objective of a NN is to compute output values from input values by some internal calculations [9]. The basic component of a NN is the neuron, also called “node”. Fig. 1 illustrates a single node of a NN.

Inputs are represented by \( a_1, a_2, \ldots, a_n \), and the output by \( O_j \). There can be many input signals to a node. The node manipulates these inputs to give a single output signal [10]. The values \( w_{ij}, w_{ij}, \ldots w_{nj} \), are weight factors associated with the inputs to the node. Weights are adaptive coefficients within the network that determine the intensity of the input signal. Every input \( (a_1, a_2, \ldots, a_n) \) is multiplied by its corresponding weight factor and the node uses summation of these weighted inputs \( (w_{1j}a_1, w_{2j}a_2, \ldots, w_{nj}) \) to estimate an output signal using a transfer function. The other input to the node, \( b_j \) is the node’s internal threshold, also called bias. This is a randomly chosen value that governs the node’s net input through (1):

\[
    u_j = \sum_{i=1}^{n} (w_{ij}a_i) - b_j
\]

Node’s output is determined using a mathematical operation on the node’s net input. This operation is called a transfer function.
Zeolite was then washed in deionized water to remove the fine fractions and thereafter dried in the oven at 50°C for 24h. The synthetic wastewater (solution of Cu) was prepared by dissolving CuSO$_4$.5H$_2$O in deionized water at pH 6.5. This solution was assayed using atomic adsorption spectroscopy (AAS), (Model Varian Spectra (20/20)).

**B. Batch uptake studies**

The Cu ion-exchange process on the zeolite was conducted at room temperature. Glass columns of 2cm diameter and 30cm of length were pre-loaded with 25g of either natural zeolite (as received) or HCl-activated zeolite. Aliquots of 25 ml of the prepared Cu-bearing solutions of desired concentrations were passed through each of the two types of zeolites. These were afforded the same solution-zeolite contact time. After passing through the zeolite-packed column the resultant solutions were 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 metal were Cu (324.7nm). Standards of 1000 mg/l, 2000 mg/l and 3000 mg/l were 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 [13].

The percentage of ions removal as the output parameter of the NN model was considered as a measure of the uptake percentage. The uptake efficiency (%) was calculated as follows:

$$\%\text{Uptake} = \frac{C_o - C_f}{C_o} \times 100$$

Where $C_o$ and $C_f$ are the initial and final ions concentrations of the solution (mg/l), respectively.

**C. Application of Neural Network (NN)**

In this study, NN Toolbox V4.0 of MATLAB mathematical software was used to predict the removal efficiency. Hundred experimental sets were used to develop the NN model. The removal of Cu (II) is strongly dependent on pH, temperature, contact time and initial concentration as variable parameters. A three-layer NN with tangent sigmoid transfer function (tansig) at hidden layer was used. The data gather from batch experiments was divided into matrix [p] and target matrix [t].

**IV. RESULTS AND DISCUSSION**

**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 MSE of the training and prediction set. In optimization of the network structure, two neurons were 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 hidden neurons, the MSE reached its
minimum value of 0.000227875. Hence, the neural network containing 11 hidden neurons was chosen as the best BP algorithm. When the number of neurons exceeded 11, the MSE showed a slight increase. The increment can be attributed to the characteristics of the MSE performance index and the input vector [p] used in this study.

The best trained network was selected as a trained network for that structure. Fig. 3 illustrates training, validation and test mean squared errors for the LMA.

![Fig. 3 Training, validation and test mean squared errors for the LMA](image)

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).

![Fig. 4 Plot regression for the NN training](image)

**B. Sensitivity analysis**

A sensitivity analysis was conducted to determine the degree of effectiveness of a variable using the proposed NN model. In the analysis, performance evaluations of various possible combinations of variables were investigated. Therefore, performance of the groups of one, two, three, four, and five variables were tested by the optimal NN structure using the LMA with 11 hidden neurons. The loading (uptake) of heavy metals ions by clinoptilolite is affected by certain variables such as pH, temperature, initial metal concentration and contact time.

**C. Effect of pH on loading efficiency**

To examine the effect of pH on loading, the solution containing Cu (II) concentration of 30 mg/L was mixing 1.0 g of adsorbent with 200 ml of solution at various pH values ranging from 2.5 to 6. Figs. 5-7 show a comparison between the predicted and experimental values of metals loaded. Findings of batch experiments showed that the pH of the solution was found to be an important parameter affecting the loading performance. The variation in the removal of metal ion with respect to pH can be explained by considering the surface charge of the adsorbent material and the metal chemistry in water. The difference of the adsorption capacity of zeolite for heavy metal ions may be due to a number of factors which include hydration diameter and solubility of cations. It can be seen in Figs 5-7 that the obtained results from the proposed NN model are in good agreement with the experimental data at pH 4.

![Fig. 5 NN outputs and experimental data for pH 2.5](image)

![Fig. 6 NN outputs and experimental data for pH 4](image)
D. Effect of temperature

The effect of temperature on the adsorption of Cu (II) onto clinoptilolite was studied at various temperatures 30, 45 and 60°C. It was observed that the initial uptake rate for two metals was very high, as a large number of adsorption sites are available for adsorption at the onset of the process. As the sites are gradually filled up, adsorption proceeds slower and the kinetics becomes more dependent on the rate at which the adsorptive is transported from the bulk phase to the actual adsorption sites. Adsorption of metal ion on zeolite adsorbent shows similar behaviour with respect to increase in temperature. The equilibrium adsorption capacity of Cu (II) onto zeolite was favoured at higher temperatures. The increase on the uptake of heavy metal ions with increase in temperature may be attributed to the increase in the average kinetic energy of the metal ions. The attractive forces between metal ions and zeolite surface sites will become insufficient to retain the metal ion at the binding site of zeolite. Figs. 8-10 show a comparison between the experimental data and the predicted on the loading efficiency with a better fit at 60°C.

E. Effect of initial concentration of Cu (II) on loading efficiency

Effect of initial concentration of Cu (II) on adsorption was determined by mixing 1.0 g of adsorbent with 200 ml of solution containing various concentrations ranging from 50 to 200 mg/L at an initial pH of 4. Batch experiments apparently showed that the percentage of Cu (II) removal increased when the initial concentration of Cu (II) per 200 ml of solution was increased from 50 to 100 mg/L for each agitation period. However, a small decrease was observed in the percentage of Cu (II) removal for the solution containing 100 mg/L of Cu (II). This was basically due to the saturation of adsorbent above an initial Cu (II) concentration of 50 mg/L. The amount of metal ions adsorbed per unit mass \((q_e)\) of Clinoptilolite increases gradually with an increase in metal concentration, whereas the extent of radiation (%) decreases with increasing metal ion loading. At low concentration of the metal ions, a unit mass of the zeolite is exposed to small number of metal ions and consequently the adsorption is independent of the initial metal ion concentration. The extent of adsorption comes down for a fixed adsorbent content at high metal ions concentration due to decreased number of available adsorption sites on zeolite surface. Figs. 11-13 show that the predicted values are in good agreement with the experimental data with best fit for initial concentration of 200 mg/L.
V. CONCLUSION

Neural Network modeling has been used to investigate relation between the effects in the ion-exchange studies of Cu$^{2+}$ ions. The NN model could describe the behavior of the ion-exchange process with the adopted experimental conditions. The configuration of the backpropagation NN giving the smallest MSE was three-layer NN with tangent sigmoid transfer function at out layer and Levenberg-Marquardt backpropagation training algorithm (LMA). NN predicted results are very close to the experimental results with correlation coefficient ($R^2$) of 0.9967 and MSE 0.000227875. The sensitivity analysis showed that the variables studied have strong effect on the copper removal during the process. NN results showed that NN modeling could effectively predict the behavior of the process.

REFERENCES