Deterministic and Neural Network Study of Cobalt and Copper Removal from Contaminated Water

John Kabuba

***Abstract*—**In this study, a neural network (NN) was proposed to predict the removal of copper-cobalt ions from contaminated water. The effect of operating parameters such as initial metal ion concentration, pH, temperature and adsorbent dosage were studied to optimize the conditions for removal efficiency. The optimal operating conditions were determined to be a pH of 4.0, an adsorbent dosage of 1.2g, a temperature of 30^{o}C, and an initial concentration of 150 mg/l (copper) and 120 mg/l (cobalt). The experimental data were used to train a multilayer perceptron network (MLP) having three-layer (4-10-2) architecture giving the smallest error. The model used tangent sigmoid transfer function at input to hidden layer whereas a logistic sigmoid transfer function was used at output layer. Levenberg-Marquardt backpropagation training algorithm (LMA) was employed for training the neural network. It was found that the NN yielded values are within the acceptable limits.

***Keywords*—**Cobalt and copper removal, contaminated water, deterministic, neural network.

I. Introduction

In the recent years, the applicability of Neural Network (NN) technique to remove heavy metals from contaminated water has gained significant success. This technique has been employed in the different processes such as coagulation, flotation, chemical precipitation, membrane processes, adsorption and ion-exchange [1-3]. Ion-exchange is one of the popular water treatment process used [2]. This process is complex, time consuming and costly, especially for studies which use many parameters. Due to complexity of the process, it makes difficult to model using conventional mathematical modeling. Neural network is considered as promising tool because of their simplicity toward simulation, prediction and modeling [4].

The aim of this work is the introduction of a new approach based on NNs to predict the removal of Co (II) and Cu (II) from contaminated water. The effects of various operating parameters such as initial concentration, pH, temperature and adsorbent dosage were investigated using clinoptilolite for the removal efficiency of metals.

II. Neural Network

The progress of neurobiology has allowed researchers to build mathematical models of neurons to simulate neural behavior. Neural Network approach has been one of the well-known type of evolutionary computation algorithm [5]. NNs are a good alternative to conventional empirical modeling based on polynomial and linear regressions. NNs have also been used in analysis and predicting ion-exchange process [1,2,6]

Neural Network is composed of large numbers of highly interconnected processing elements known as neurons. The basic elements of an artificial neuron are shown in Fig. 1. Artificial neuron consists of weight, bias and activation function mainly.

Each neurons receives inputs x_1, x_2, x_3,…, x_n attached with a weight w_1 which shows the connections strength for a particular input for each connection. Every input is then multiplied by the corresponding weight of the neuron connection.

A bias, b_i can be defined as a type of connection weight with a constant nonzero value added to the summation of inputs and corresponding weights u, given as follows:

$$u_i = \sum_{j=1}^{n} w_{ij} x_j + b_j \quad (1)$$

The summation u_i is transferred using a scalar-to-scalar function called an "activation or transfer function", $f(u_i)$, to yield a value called the unit's "activation", given as:

$$y_i = f(u_i) \quad (2)$$

Activation function serves to introduce non-linearity into Neural Network which makes it more powerful than linear transformation. Backpropagation algorithm (BPA) which is the most widely used training algorithm for the multilayer perception, is a gradient descent technique to minimize the error for a particular training pattern. Accordingly, for a given input pattern, a flow of activation is forwarded from the input layer to the output layer via hidden layer(s). Then the errors in the output are initiated. BPA is used to adjust the weights, a small amount at a time, in a way that reduces the error. The training of the network is accomplished by adjusting the weights and it carried out through a large number of training sets and training cycles (eporches). The goal of the learning procedures is to find the optimal set of weights, which in the ideal case would produce the right output for any input. The output of the network is compared with a desired response to produce an error. Once the NN is adequately trained, it can have generalized to similar cases, which it has never seen.

Manuscript received September 4, 2017.

J. Kabuba is with the Department of Chemical Engineering, Vaal University of Technology, Vanderbijlpark, Private Bag X021.

https://doi.org/10.17758/EARES.EAP1117025

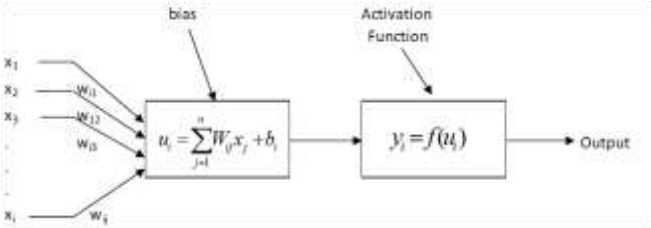

Fig. 1. Basic elements of an NN neuron

Neural Network are computing systems composed of neurons and are used to solve complex functions, which attempt to simulate the structure and function of biological neurons. A NN system has three layers, namely the input, the hidden layer and the out layer. The input layer consists of all the input factors, information from the input layer is then processed in the course of one hidden layer, and following output vector is computed in the output layer. The estimation problem using neural network models has three successive steps: model building or NN architecture; the learning or training procedure; and the testing procedure. An important stage when accommodating a NN is the training step, in which an input is introduced to the network together with the desired outputs, the weights and bias values are initially chosen randomly and the weights are adjusted to that the network attempts to produce the desired output. The weight after training contain meaningful information whereas before training, they are random and have no meaning. When a satisfactory level of performance is reached, the training stops. And the network uses these weights to make decisions. NN has been applied successfully in various fields of mathematics, engineering, medicine, economics, meteorology, psychology and neurology.

Many alternative training processes, such as back-propagation are available. The goal of any training algorithm is to minimize the global error level, such as the mean % error, root-mean-squared (RMS) and absolute fraction of variance (R^2) [7]. An important characteristic of this function is differentiable throughout its domain. The errors for the hidden layers are determined by propagating back the error determined for the output layer.

III. Materials and Methods

The clinoptilolite and synthetic solutions were prepared as described by Abdulkareem et al. [8] and Kabuba [9]. Batch experiment were conducted to determine the effect of pH, initial concentration, temperature and adsorbent dosage. The effect of pH was carried out in pH range of 2-10; the effect of various operating temperature ranging from 30 to 90^{o}C and the adsorbent dosage of 1-5g was investigated. The initial concentration of synthetic solution 100-200 mg/L for both Co (II) and Cu (II) were analyzed using atomic adsorption spectroscopy. The samples were taken at predetermined time intervals (0-120 min) on the removal efficiency. Each experiment was performed in duplicate t observe the reproducibility and the mean value used for each set of values. The removal efficiency for the metals (Co(II) and Cu (II)) as the output parameters of NN model.

The removal efficiency was calculated as follows:

$$\% = \frac{(C_a - C_b)}{C_a} x100 \quad (3)$$

The main focus of this study is the prediction of Cu and Co removal from contaminated water using NN based on experimental results. pH, adsorbent dosage, temperature and initial concentration have been used as input while Co and Cu removal efficiency were used as output.

In the NN model, 92 experimental data were divided into input matrix and output matrix for training the network. The back-propagation learning algorithm has been used with one hidden layer. The hidden layer neurons process the incoming information and extract useful features to reconstruct the mapping from the input space. The architecture of the network was obtained by trying different number of hidden layers and neurons. The performance of each network was checked by correlation coefficient (R). The goal is to maximize correlation coefficient to obtain a network with the best generalization. The proposed NN model architecture is presented in Fig. 2. The learning algorithm is Levenberg-Marquardt (LM), activation function is logistic sigmoid (logsig) transfer function.

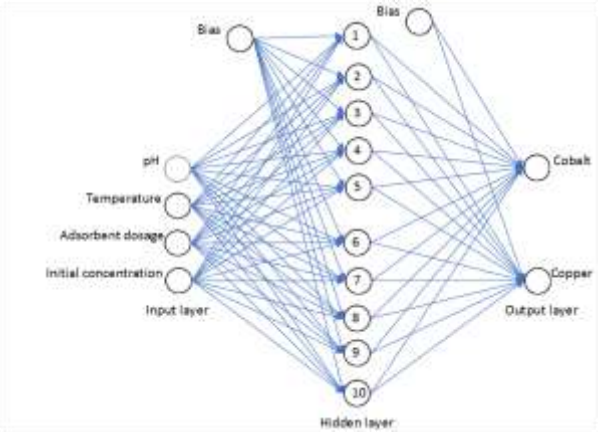

Fig. 2. The proposed NN model architecture

IV. Results and Discussion

A. Determination of Data and the Network Model Employed

For Neural Network, two data sets are needed (training-testing). The usual approach is to prepare a single data set, and differentiate it by a random selection. In this study, experimental results were used to train and test an NN. Five different NNs (Batch gradient descent, Scaled conjugate gradient, One step secant, Variable learning rate and Levenberg Marquardt) are studied, all the networks, the learning algorithm called back-propagation was applied for the single hidden layer, Levenberg-Marquardt (LM) has been used for the variants of the algorithm. These normalized both for the inputs and outputs are realized between the values of 0 and 1. Neural Network was trained and tested by means of the MATLAB software. In order to identify the output precisely for training stage, increased number of neurons in the hidden layer was tried. Firstly, the network was trained successfully, and then the test data were used to test the network. By means of the results deduced by the network, a comparison was corrected out using the statical methods. Errors that happed at the learning and testing, stages are described the RMS and R^2, mean error percentage values,

which are defined by Eqs. (4) and (5), respectively.

$$RMS = \left[\left(\frac{1}{p}\right)\sum_j \left|t_j - o_j\right|^2\right]^{\frac{1}{2}} \quad (4)$$

$$R^2 = 1 - \left[\frac{\sum_j \left(t_j - o_j\right)^2}{\sum_j \left(o_j\right)^2}\right] \quad (5)$$

where t is the target value o is the output value, and p is the pattern [7].

The RMS, R^2 and the mean error % values were used for comparing all of them.

B. Analysis Results

The NN model developed is used to predict the removal efficiency of Cu (II) and Co (II) from contaminated water. In order to check the generality of network prediction and to prevent the data overfitting, the data sets were divided into training, validation and test subsets. The performance of the proposed NN model was plotted in Figs. 3-4 for both Cu and Co, respectively. Taking into account the nonlinear dependence of the experimental data, linear regression shows a good agreement between NN out predicted data and the corresponding targets (actual values). The correlation coefficient (R^2) were 0.9987 and 0.9997 for Cu and Co, respectively.

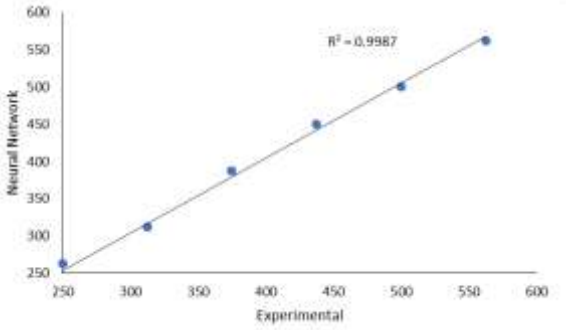

Fig. 3. Prediction of NN and actual values for training-validation-testing (Cu)

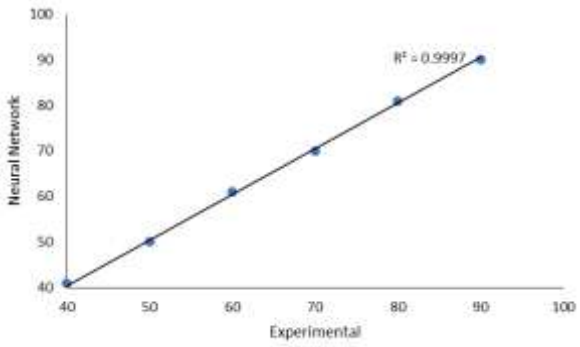

Fig. 4. Prediction of NN and actual values for training-validation-testing (Co)

The RMS of the Cu (II) prediction was about 0.2801, 0.314 and 1.63 for the training, testing and validation set, respectively. Similarly, RMS of the Co prediction was about 1.017, 1.214 and 2.647 for training, testing and validation set, respectively. As it can be seen, these RMS are fairly reasonable. Figs. 3-4 demonstrated that NN was quite successful in learning the relationship between the different input parameters and the outputs. The result was showed that the NN was capable of generalizing between input variables and output reasonably well. NN applications are treated as black-box applications in literature [10]. However, this study opens this blackbox and introduces the NN application in a closed form solution. This study aims to present the closed form solution of Cu and Co removal based on the trained NN parameters (weights and biases) as a function pH, temperature, absorbent dosage and initial concentration. Using weights and biases of trained NN model, Co removal efficiency can be given as presented in Eq. (6).

$$Co = \frac{400}{1+e^{-\left(\frac{0.0315}{1-e^{-U1}} - \frac{0.0475}{1-e^{-U2}} - \frac{0.0700}{1-e^{-U3}} + \frac{0.1229}{1-e^{-U4}} + \frac{0.5190}{1-e^{-U5}} - \frac{0.1521}{1-e^{-U6}} + \frac{0.0712}{1-e^{-U7}} + \frac{0.0259}{1-e^{-U8}} + \frac{0.0511}{1-e^{-U8}} + \frac{0.1022}{1-e^{-U10}} - 0.7010\right)}} \quad (6)$$

where the transfer function used for this approach is given as in Eq. (7).

$$F_1 = \frac{1}{1+e^{-ui}} \quad (7)$$

where U_i are given in Eq. (7) for the Co (II).

$$U_i = C_{1i}\,x\,pH + C_{2i}\,x\,temperature + C_{3i}\,x\,adsorbent\ dosage + C_{4i}\,x\,initial\ concentration \quad (8)$$

Similarly, Cu can be found as in Eq. (9).

$$C_u = \frac{450}{1+e^{-\left(\frac{0.0306}{1-e^{-U1}} - \frac{0.0486}{1-e^{-U2}} - \frac{0.0720}{1-e^{-U3}} + \frac{0.1335}{1-e^{-U4}} + \frac{0.6160}{1-e^{-U5}} - \frac{0.1523}{1-e^{-U6}} + \frac{0.0723}{1-e^{-U7}} + \frac{0.0278}{1-e^{-U8}} + \frac{0.0548}{1-e^{-U9}} + \frac{0.1134}{1-e^{-U10}} - 0.7001\right)}} \quad (9)$$

where U_i are given in Eq. (10) for Cu (II).

$$U_i = C_{5i}\,x\,pH + C_{6i}\,x\,temperature + C_{7i}\,x\,adsorbent\ dosage + C_{8i}\,x\,initial\ concentration \quad (10)$$

where the constants (C_{ij}) are given in Table I for LM algorithm with 10 neurons. It should be noted that the proposed explicit formulation of the NN model presented is valid for the ranges of training set.

TABLE I: THE WEIGHTS AND BIASES BETWEEN INPUT LAYER AND HIDDEN LAYER FOR EQS. (6) AND (8)

i	Co C_{1i}	C_{2i}	C_{3i}	C_{4i}
1	-1.2725	0.1925	- 0.0034	11.789
2	-1.6310	- 0.1166	0.0006	26.542
3	0.0061	- 0.0831	0.0023	3.458
4	1.5900	- 0.0879	0.0010	- 4.963
5	- 0.2590	0.1259	0.0020	15.789
6	0.0533	- 0.1256	0.0020	- 0.359
7	- 0.8738	0.4050	- 0.0038	- 5.653
8	2.3690	- 0.1951	0.0039	5.362
9	- 0.6472	0.2504	0.0039	-28.693
10	- 0.0355	- 0.2315	- 0.0032	30.016

TABLE II: THE WEIGHTS AND BIASES BETWEEN INPUT LAYER AND HIDDEN LAYER FOR EQS. (9) AND (10)

i	Co C_{5i}	C_{6i}	C_{7i}	C_{8i}
1	-1.3856	0.1936	- 0.0035	11.799
2	-1.7420	- 0.1172	0.0008	26.545
3	0.0067	- 0.0841	0.0033	3.473
4	1.8600	- 0.0898	0.0010	- 4.964
5	- 0.2620	0.1286	0.0020	15.799
6	0.0559	- 0.1261	0.0020	- 0.362
7	- 0.8758	0.4056	- 0.0042	- 5.668
8	2.3720	- 0.1956	0.0040	5.369
9	- 0.6478	0.2512	0.0039	-28.699
10	- 0.0365	- 0.2318	- 0.0038	30.018

C. *Effect of pH*

The experiments were conducted by varying the pH values and keeping another parameters constant. The removal efficiency was 78% and 77% at pH 4 for Cu and Co, respectively. This low pH can be attributed to the fact that a high concentration of H^+ ions may have affected metals ions removal via ion-exchange by direct competition effects between H^+ and metals ions for the exchange site on the clinoptilolite particle. In terms of the relation between the experimental data and the NN models, Figs. 5 and 6 show that predicted values (NN) are in good agreement with the experimental data.

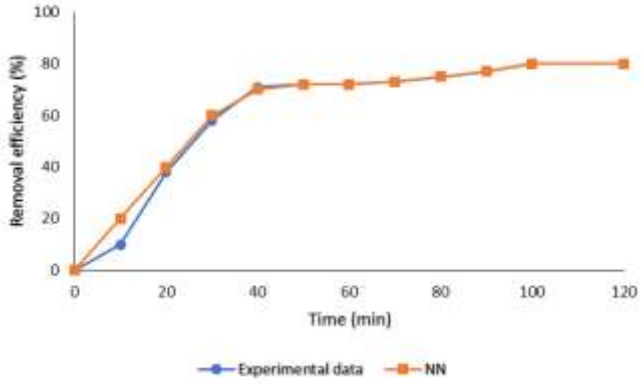

Fig. 5. Comparison between NN outputs and experimental data at pH 4 for Cu (II)

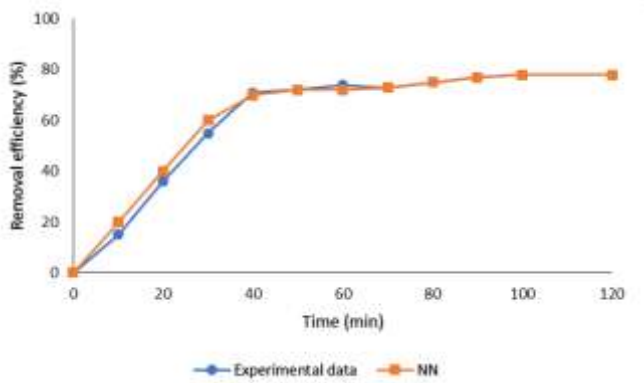

Fig. 6. Comparison between NN outputs and experimental data at pH 4 for Co (II)

D. *Effect of Temperature*

Experimental results show that the removal of Cu (II) and Co (II) from contaminated water by clinoptilolite was dependent on the temperature. The removal efficiency (%) was increased to 30°C for both metals. This can be attributed to the fact that the attraction forces between material surface and metal ions are weakened and the removal decreases above a certain temperature threshold.

Kabuba et al. [9] found that the thickness of the boundary layer decreases at relatively high temperature. This is due to the increased tendency of the metal ion to escape from the clinoptilolite materials surface to the solution, which results in a decrease in the removal. In terms of the relation between the experimental data and the predicted values, Fig. 7 and 8 show that predicted values are in good agreement with the experimental data.

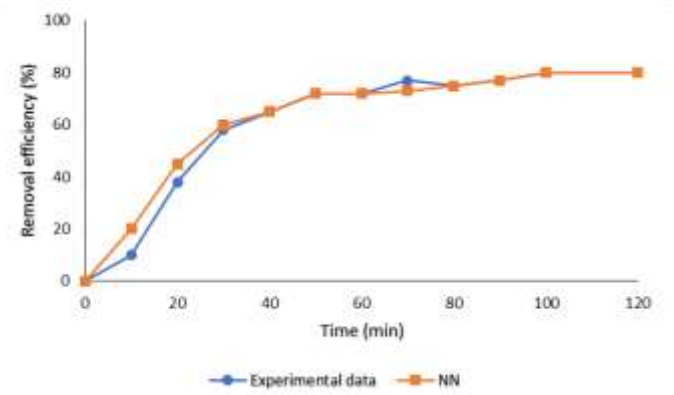

Fig. 7. Comparison between NN outputs and experimental data for temperature 30°C for Cu (II)

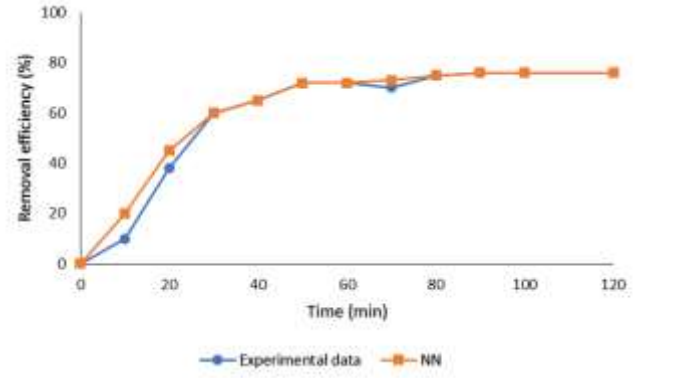

Fig. 8. Comparison between NN outputs and experimental data for temperature 30°C for Co (II)

E. *Effect of ion Concentration*

The effect of initial concentration was determined at pH 4, temperature 30°C and adsorbent dosage 1.2 g for Cu (II) and Co (II). At high concentration, metals need to diffuse to the clinoptilolite surface by intraparticle diffusion and greatly hydrolyzed ion will diffuse at a slower rate because of the saturation of the active site available on the clinoptilolite for interaction with metal ions. The removal efficiency for metals decreases with increasing metal concentration in aqueous solutions. In terms of the relation between the experimental results and the predicted values by the model, Figs. 9 and 10 show that the predicted values with NN are in good agreement with the experimental data.

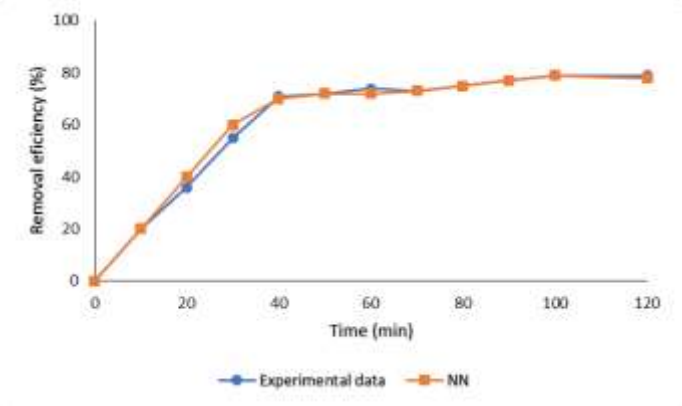

Fig. 9. Comparison between NN outputs and experimental data for Initial concentration 150 mg/L for Cu (II)

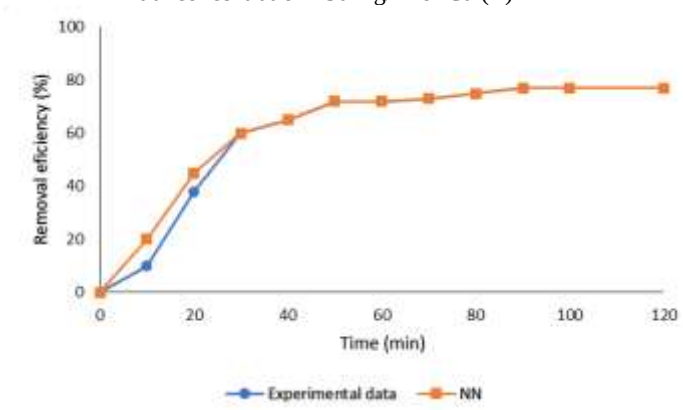

Fig. 10. Comparison between NN outputs and experimental data for Initial concentration 120 mg/L for Co (II)

F. Effect of Adsorbent Dosage

Adsorbent dosage is an important parameter because is influencing the capacity of clinoptilolite for s given initial concentration. The removal efficiency increases with the increase of clinoptilolite dosage due to the active site on the clinoptilolite making the penetration easy. Figs. 11 and 12 show that the predicted values with NN are in good agreement with the experimental data.

IV. Conclusion

This study presents a new and efficient approach for the prediction of Co (II) and Cu (II) using NN. The database used for NN training is based on experimental results. To train the network; pH, temperature, adsorbent dosage and initial concentration were used as the input while the Co (II) and Cu (II) removal efficiency are used as the output. BPNN with LM algorithm was used for the training process. The explicit formulation of Cu and Co based on the proposed NN model has strong potential as feasible tools for prediction of Cu (II) and Co (II). The usage of NN was found to be working well. This found that the network yielded values are within the acceptable limits. It has been shown that the values produced with NN are parallel to the experimental results and the network method reproduces experimental data within 99.87 and 99.97% confidence. This confirms that the proposed Neural Network model agreed perfectly with the experimental results.

Acknowledgment

The authors gratefully acknowledge the support received from the Vaal University of Technology.

References

[1] A. Amankwah, J. Kabuba, A.F. Mulaba-Bafubiandi. "Modeling of Ion-exchange process using time delayed neural networks". Proc. Int. Conf. Mech. Electr. Technol., vol. 1, pp. 447-451, 2011. https://doi.org/10.1115/1.859810.paper73

[2] J. Kabuba and A.F. Mulaba-Bafubiandi, "Comparison of equilibrium study of binary system Co-Cu ions using adsorption isotherm models and Neural Network" Proc. Int. Conf. Min. Proc. Met. Eng., vol. 1, pp. 126-130, 2013.

[3] N.G. Zaki, I.A. Khattab, N.M.A. El-Morem, "Removal of some heavy metals by CKD leachate" J. Hazard. Mater., vol. 147, pp. 21-27, 2007. https://doi.org/10.1016/j.jhazmat.2006.12.057

[4] E.S. Elmolla, M. Chaudhuri, M.M. Eltoukhy, "The use of artificial neural network (ANN) for modeling of COD removal from antibiotic aqueous solution by the Fenton process". J. Hazard. Mate., vol. 179, pp. 127-134, 2010. https://doi.org/10.1016/j.jhazmat.2010.02.068

[5] A. Sozen, E. Arcakloglu and M. Ozkaymak. "Turkey's net energy consumption" Applied Energy, vol. 81, pp. 209-221, 2005. https://doi.org/10.1016/j.apenergy.2004.07.001

[6] A. Amankwah, J. Kabuba. "Comparison of Neural Networks and Kalman Filter for the Modelling of Ion-Exchange Process", Life Sci J., vol. 10, pp. 1012-1015, 2013.

[7] M. Canakci, A. N. Ozsezen, E. Arcaklioglu, A. Erdil. "Prediction of performance and exhaust emissions of a diesel engine fueled with biodiesel produced from waste frying palm oil". Expert Systems with Applications, vol. 36, pp. 9268-9280, 2009. https://doi.org/10.1016/j.eswa.2008.12.005

[8] S.A. Abdulkareen, E. Muzenda, A.S. Afolabi, J. Kabuba, "Treatment of clinoptilolite as an adsorbent for the Removal of copper ion from synthetic wastewater solution", Arab. J. Sci. Eng., vol. 38, pp. 2263-2272, 2012. https://doi.org/10.1007/s13369-012-0505-x

[9] J. Kabuba, A. Mulaba-Bafubiandi, K. Battle. "Neural Network Technique for Modeling of Cu (II) Removal from Aqueous Solution by Clinoptilolite". Arab. J. Sci. Eng., vol. 39, pp. 6793-6803, 2014. https://doi.org/10.1007/s13369-014-1277-2

[10] N. K. Togun and S. Baysec." Prediction of torque and specific fuel consumption of a gasoline engine by using artificial neural networks" Applied Energy, vol. 87, pp. 349-355, 2010. https://doi.org/10.1016/j.apenergy.2009.08.016 https://doi.org/10.1016/j.apenergy.2010.04.027