African Journal of Biotechnology Vol. 11(95), pp. 16196-16204, 27 November, 2012
Available online at http://www.academicjournals.org/AJB
DOI: 10.5897/AJB11.2047
ISSN 1684–5315 ©2012 Academic Journals

Full Length Research Paper

Modeling effluent heavy metal concentrations in a bioleaching process using an artificial neural network technique

Bulent Sari

Department of Environmental Engineering, Faculty of Engineering and Architecture, Cukurova University, 01330, Balcali, Adana/Turkey. E-mail: bsari@cu.edu.tr. Tel/Fax: +90 322 3386779.

Accepted 9 November, 2012

Artifical neural networks practices were used to predict the recovery of heavy metals (Zn, Cu, Ni, Pb, Cd and Cr) from dewatered metal plating sludge (with no sulfide or sulfate compounds) using bioleaching process involving *Acidithiobacillus ferrooxidans*. The bioleaching process was operated as a completely mixed batch (CMB) reactor. The leaching performance data of the CMB reactor in terms of heavy metals was applied to a multi-layer perceptron (MLP) neural network technique for simulation. The performance of the reactor was evaluated with this robust model using the experimental data obtained under varying heavy metal concentrations in the sludge. Agitation time, pulp density of the sludge, and pH were used as inputs for the model, whereas the heavy metals (Cd, Cr, Cu, Ni, Pb, and Zn) concentrations were the output variables. The results of the models were compared using statistical criteria such as mean square error (MSE), mean absolute error (MAE), mean absolute relative error (MARE), and determination coefficient (R^2). The results show that the MLP neural network produced highly accurate estimation of the aforementioned metals with R^2 over 97.9%.

Key words: Bioleaching, heavy metals removal, artificial neural network, multi-layer perceptron.

INTRODUCTION

The process of bioleaching offers a pollution-less low cost and low energy approach for metals extraction from low-grade ores, soil or mine tailings by chemolithotrophic bacteria, heterotrophic bacteria or fungi as previuosly reported (Mazuelos et al., 1999; Bevilaqua et al., 2002; Mandal and Banarjee, 2004; Chatain et al., 2005; Shi and Fang, 2005; Fang and Zhou, 2006; Wu and Ting, 2006; Vestola et al., 2010). Among these microorganisms, *Thiobacillus thiooxidans* and *Thiobacillus ferrooxidans* are more commonly used for the bioleaching of sulfide minerals (Donati et al., 1996; Das et al., 1999; Falco et al., 2003).

Many of exterior physical (adsorption-desorption, pulp density, particle size, agitation and temperature), biological (growth rate and cell concentration), chemical (pH and medium composition), and electrochemical (redoks potential) parameters have been determined to have an influence on the performance of the bioleaching process. The common mathematical models that are built based on these parameters and used in bioleaching applications have some advantages and disadvantages such as models of Kumar and Gandhi, Lacey and Lawson, Blancarte-Zurita and Branion, Konishi and Katoh, Sanmugasunderam and Branion, and Hanson, etc (Haddadin et al., 1995). In order to manage an optimum bioleaching process and effluent heavy metals concentrations, appropriate models that completely defines the system are required. In an effort to control the performance of the system, it is imperative to use thorough models that are dependent on the determination of specific parameters and hence predict the process performance based on these parameters.

Biological systems are non-linear, ever progressing,

Abbreviations: CMB, Completely mixed batch; **MLP,** multi-layer perceptron; **MSE,** mean square error; **MAE,** mean absolute error; **MARE,** mean absolute relative error; **R^2,** determination coefficient.

Table 1. Physicochemical characteristics of the dewatered metal plating sludge samples.

Characteristic	Value
Solids (wt.%)	4.11 - 4.95
pH	9.01 - 9.45
Particle size (mm)	$d_{90} < 0.125$
Concentration of heavy metals (mg/kg)	
Zn	22900 - 22980
Pb	828 - 850
Ni	601 - 729
Cu	625 - 691
Cd	325 - 341
Cr	210 - 212

and highly complex systems. Artificial neural network (ANN) have found vast application for biological systems and therefore its importance and favourability have increased greatly. Mathematical modeling of non-linear and complex biological systems offer a difficult task for the interested. Therefore, the regression models have been used to model most biological systems due to their wide range of application. However, regression models have proven to err in the determination of some data that are not required to be used for especially the regression equation. It has been shown by Mohanty et al. (2002) that the ANN models have better simulated the biological systems as compared to the regression models. The use of neural networks to predict the solubilization of heavy metals originating from municipal wastewater treatment sludges has already been presented by Du et al. (1994) who used *Th. thiooxidans* and *Thiobacillus thioparus* in batch systems. The authors have demonstrated that a neural network with input variables of type of sludge, initial metal concentrations and pH could satisfactorily predict heavy metal solubilization.

Since the 1990s, the studies on biological systems and bioleaching has shown that ANN-based models demonstrated better prediction performance for complex biological systems with numerous non-linearly correlated parameters as compared to conventional mathematical and statistical models (Acharya et al., 2006; Ozkaya et al., 2008; Liu et al., 2008; Jorjani et al, 2007; Nurmi et al., 2010; Laberge et al., 2000). In this study, a new ANN model was proposed for the estimation of heavy metal concentrations in a completely mixed batch reactor as an alternative to the conventional methods. The predictive ability of the proposed model was assessed using some statistical criteria - mean square error (MSE), mean absolute error (MAE), mean absolute relative error (MARE) and and determination coefficient (R^2).

MATERIALS AND METHODS

The methodology followed in this study has been previously described in details (Bayat and Sari, 2010a). Therefore, the materials and methods were concisely described herein. The dewatered metal plating sludge samples studied were collected in polyethylene bags from the Karme Metal Plating Industries in Fatsa-Ordu, Turkey. The physicochemical characteristics of the dewatered sludge samples are presented in Table 1. All analyses were carried out in duplicate (Bayat and Sari, 2010a, b). The biological leaching experiments were carried out in a completely mixed batch (CMB) reactor with a volume of 3 L and dimensions of 15 cm inner diameter (ID) and 17 cm height equipped with pH and temperature controllers, a stirrer, and an aerator system (Figure 1) (Bayat and Sari, 2010a).

Artificial neural network approach

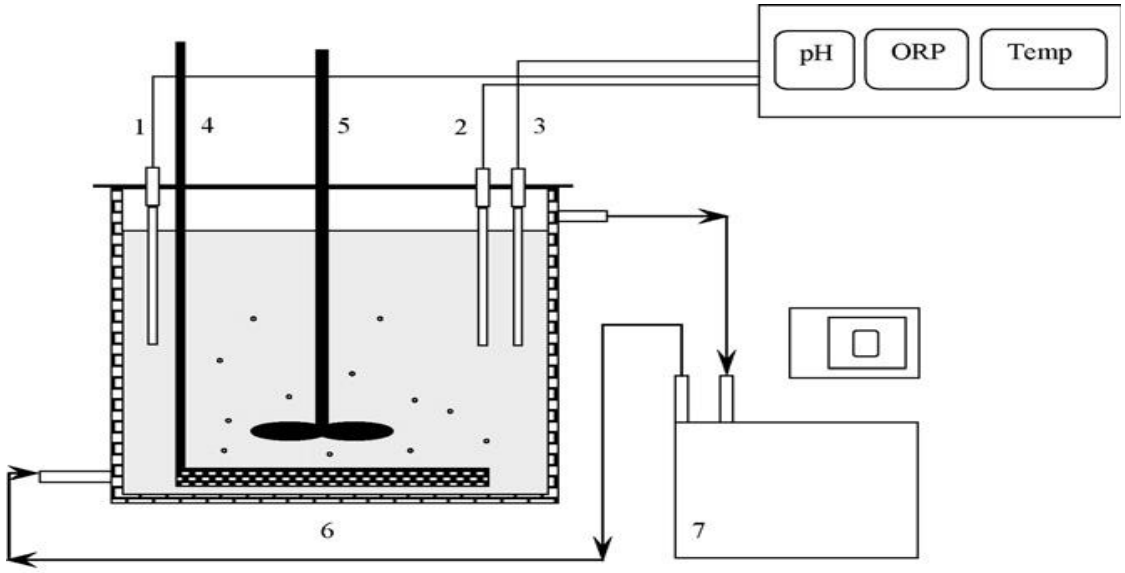
ANN has the ability to learn from examples, recognize a pattern in a group of data, adapt solutions over time, and process information rapidly. The application of ANN to issues related to wastewater treatment and water resources conservation is rapidly gaining popularity due to their immense power and potential in the mapping of nonlinear system data. In the context of hydrological forecasting, recent studies have reported that ANN technique may offer a promising alternative for bioleaching (Acharya et al., 2006; Ozkaya et al., 2008; Liu et al., 2008; Jorjani et al, 2007; Nurmi et al., 2010; Laberge et al., 2000), rainfall–runoff modeling (Lin and Chen, 2004), stream-flow prediction (Raman and Sunilkumar, 1995; Kisi, 2004a), suspension of sediments (Kisi, 2004b), water resources (Cobaner et al., 2008), reservoir inflow forecasting (Coulibaly et al., 2005) and treatment of wastewater (Elmolla et al., 2010; Pai et al., 2009; Chen and Lo, 2010). The variation in the characteristics of a bioleaching system may be non-linear and multivariate, and the variables involved may have complex inter-relationships. For most cases, ANNs provide more reliable estimates for dependent variables of concern. The processes that involve several parameters are easily amenable to neuro-computing.

Among the many ANN structures that have been studied, the most widely used network structure is the multilayer perceptron (MLP) network. An ANN consists of a number of data processing elements called neurons or nodes, which are grouped in layers. The input layer of neurons receives the input vector and transmits the information to the next layer with the help of cross connections. In the current study, a MLP modeling technique was applied.

Multi-layer perceptron (MLP) neural network

A MLP distinguishes itself by the presence of one or more hidden layers, whose computation nodes are correspondingly called “hidden neurons of hidden units”. The function of hidden neurons is to intervene between the external input and the network output in some useful manner. By adding one or more hidden layers, the network is enabled to extract higher order statistics. In a rather loose sense, the network acquires a global perspective despite its local connectivity due to the extra set of synaptic connections and the extra dimension of NN interconnections. The detailed theoretical information about MLP can be found in Haykin (1998).

The MLP network used in the current study is shown in Figure 2. Index k is referred to as the individual output layer neurons, the indices i and j refer to as the input neurons and the hidden layer neurons, respectively, while w_{ij} and w_{jk} represent the connection weights between the hidden-input layer and hidden-output layer, respectively. A hidden-layer neuron produces the following as output:



1. pH probe
2. ORP probe
3. Temprature probe
4. Air diffuser
5. Mixer
6. Completely mixed batch reactor
7. Water bath circulator

Figure 1. Schematic diagram of bioleaching experiments.

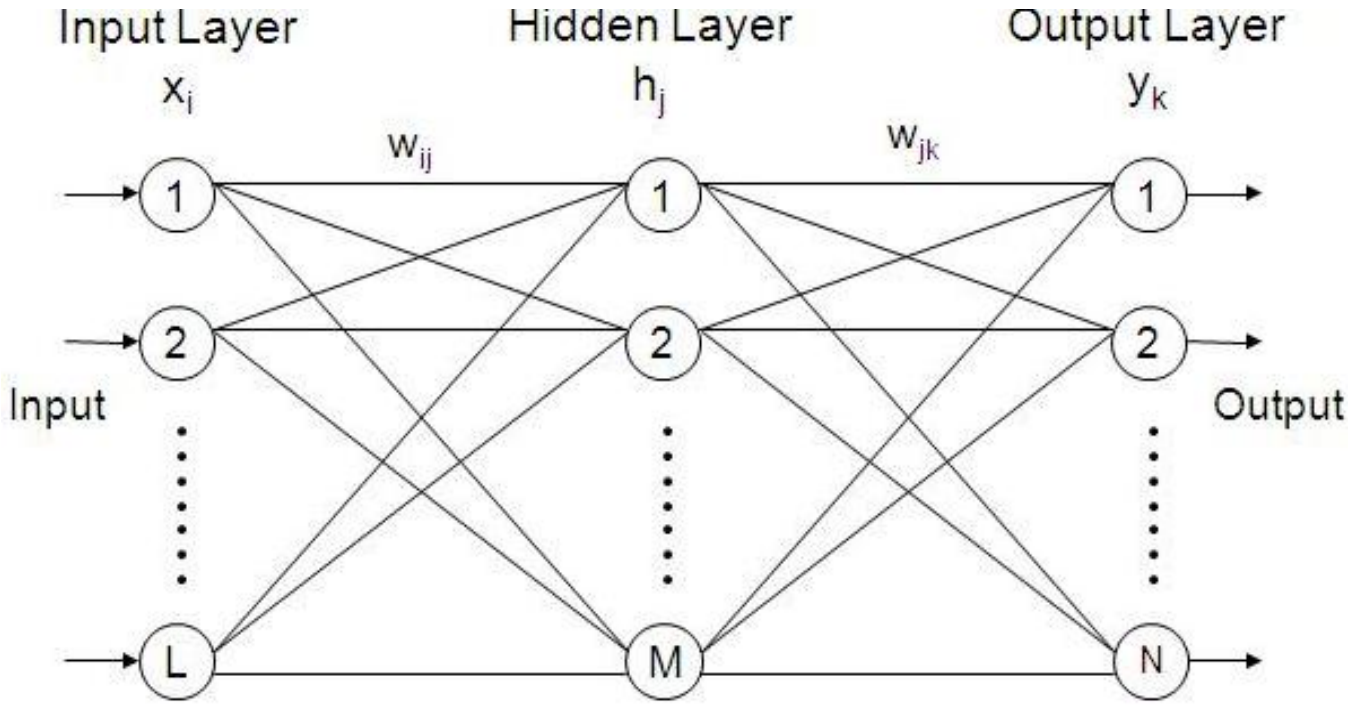


Figure 2. The MLP model network.

$$h_j = f\left(\sum_{i=1}^{n} w_{ij}x_i + b_j\right) \quad [1]$$

$$y_k = f\left(\sum_{j=1}^{n} w_{jk}h_j + b_k\right) \quad [2]$$

While an output-layer neuron produces the following as output;

Table 2. Minimum and maximum values of input and output parameters.

Model parameter	Training data set		Testing data set	
	Min	Max	Min	Max
Agitation time	1	20	2	19
Pulp density	0.02	0.24	0.08	0.18
pH	1.95	8.88	2.67	8.26
Zn	187.23	3358.42	642.32	2151.77
Cu	0.38	80.38	2.61	80.38
Cr	0.01	8.36	0.11	8.32
Cd	0.05	28.58	0.68	28.57
Ni	0.56	99.54	2.14	78.2
Pb	0.02	71.35	0.11	71.31

Where h_j is the output of the j_{th} neuron in the hidden layer; x_i is the input of the i_{th} neuron in the input layer; y_k is the output of the k_{th} neuron in the output layer; $b_j$and b_k are the threshold values, also called the bias, associated with the hidden and output nodes, respectively; and f denotes the activation function. Each neuron multiplies every input by its interconnection weight, sums the product, and then passes the sum through a transfer function to produce its result. This transfer function is usually a steadily increasing S-shaped curve, called a sigmoid function.

The MLP can have more than one hidden layer. However, theoretical works have shown that a single hidden layer is sufficient for MLP to approximate any complex nonlinear function (Maier and Dandy, 1996; Onkal at al., 2005). Therefore, in this study, one-hidden-layer MLP was used. Throughout all MLP simulations, the adaptive learning rates are used to speed up training. The numbers of hidden layer neurons are found using simple trial-and-error method in all applications. The sigmoid and linear functions are used for the activation functions of the hidden and output nodes, respectively. Some of the recent studies have reported that the performance of MLP was superior to conventional statistical and stochastic methods (Kisi, 2004a, b). Multilayer perceptions can get trapped in a local minimum when they try to find the global minimum of the error surface.

Maier and Dandy (2000) summarized the methods used in the published literature to overcome the local minima problem, such as training a number of networks starting with different initial weights, an on-line training mode to help the network escape local minima, inclusion of the addition of a random noise, and employment of second order schemes, such as Newton–Raphson and Levenberg–Marquardt algorithms, or global methods such as stochastic gradient algorithms and simulated annealing. Other ANN methods, such as conjugate gradient algorithms, the radial basis function, the cascade correlation algorithm, and recurrent neural networks, were briefly explained in the report by the ASCE Task Committee on Application of Artificial Neural Networks in Hydrology (2000a, 2000b).

Levenberg-Marquardt algorithm

In the present study, the Levenberg–Marquardt algorithm was employed because this algorithm is more powerful than the conventional gradient descent techniques (Hagan and Menhaj, 1994). The Levenberg–Marquardt algorithm is an approximation of Newton's method and is very efficient for training networks with up to a few hundred weights. Although the computational load of the Levenberg–Marquardt algorithm is greater than that of other techniques, this is compensated by the increased efficiency and much better precision in results. In many cases, the Levenberg–Marquardt algorithm was found to converge when other back-propagation techniques diverged (Hagan and Menhaj, 1994).

Determination of an appropriate ANN model

Determining an appropriate architecture of a neural network for a particular problem is an important issue as the network topology directly affects its computational complexity and its generalization capability. MLP model with one hidden layer can approximate any complex non-linear function provided sufficient amount of hidden-layer neurons are available (Hornik et al., 1989). Indeed, many experimental results seem to confirm that one hidden layer may be enough for most forecasting problems (Coulibaly et al., 1999). Therefore, in this study, one hidden-layer MLP model was used. Generally, the number of hidden layer neurons is determined by a trial-and-error method. A common strategy for finding the optimum number of hidden-layer neurons is to start with a small number of neurons and increase their number while monitoring the performance criteria until no significant improvement is observed (Goh, 1995).

RESULTS AND DISCUSSION

In this study, the concentration of each heavy metal (Cd, Cr, Cu, Ni, Pb, and Zn) was the dependent variable, while the independent variables were the agitation time, pulp density, and pH. The minimum and maximum values for the model variables are provided in Table 2. There are no acceptable rules to determine the optimum size of the training data set. The networks are not very sensitive to the number of training data, but very sensitive to the number of testing data. Attempts at reducing the training data size resulted in poor generalization capabilities in the testing phase. Therefore, the available data set was partitioned into a training set and a testing set with 75 and 25% of the available experimental measurements selected for training and testing phases, respectively. Before the training phase of the network, both input and output variables were normalized within the range of 0.1 to 0.9 as follows:

Table 3. Statistical assessment of MLP-LM models for both training and testing phases.

Parameter	Criteria	Training	Testing
		MLP-LM	MLP-LM
Cr	MAE	0.1334	0.2581
	MSE	0.0306	0.1936
	MARE	100.9855	13.7304
	R^2	0.9908	0.9812
Zn	MAE	2.6844	2.4645
	MSE	15.5973	10.5366
	MARE	22.6989	12.3220
	R^2	0.9854	0.9840
Cu	MAE	0.3549	0.9755
	MSE	0.2095	1.7330
	MARE	26.6762	11.2191
	R^2	0.9946	0.9791
Cd	MAE	76.8961	29.6200
	MSE	9007.5130	1397.3167
	MARE	9.1677	2.4401
	R^2	0.9933	0.9942
Ni	MAE	0.8476	1.0613
	MSE	1.2370	1.6854
	MARE	18.4977	4.2652
	R^2	0.9962	0.9976
Pb	MAE	0.5585	2.2865
	MSE	0.7503	10.7273
	MARE	26.4042	20.1662
	R^2	0.9971	0.9804

$$x_i = 0.8\frac{(x - x_{min})}{(x_{max} - x_{min})} + 0.1 \qquad [3]$$

Where x_i is the normalized value of a certain parameter, x is the measured value for this parameter, x_{min} and x_{max} are the minimum and maximum values in the database for this parameter, respectively.

For all created neural networks, the general structure of input, one hidden, and one output layer was used. In order to determine the optimal architecture, several neural networks were trained with different iteration number (epoch) and number of nodes in the hidden layer. For all cases, a “log sigmoid transfer function (log sig)” was used in the hidden and output layers. When the log sig was applied, the inputs and the outputs were normalized within the range of 0 to 1. The most accurate estimations of the ANNs were obtained with logarithmic sigmoid transfer function. The best MLP results were obtained from the ANN (4, 6, 1) model using the logarithmic sigmoid activation functions for both hidden and output layer neurons, respectively.

The MAE, MSE, MARE and R^2 values of ANNs for both training and testing phases are given in Table 3. The MAE, MSE and MARE are defined as follows:

$$MSE = \frac{1}{N}\sum_{i=1}^{N}(Yi_{observed} - Yi_{predicted})^2 \qquad [4]$$

$$MAE = \frac{1}{N}\sum_{i=1}^{N}\left|Yi_{observed} - Yi_{predicted}\right| \qquad [5]$$

$$MARE = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{Yi_{observed} - Yi_{predicted}}{Yi_{observed}}\right| \times 100 \qquad [6]$$

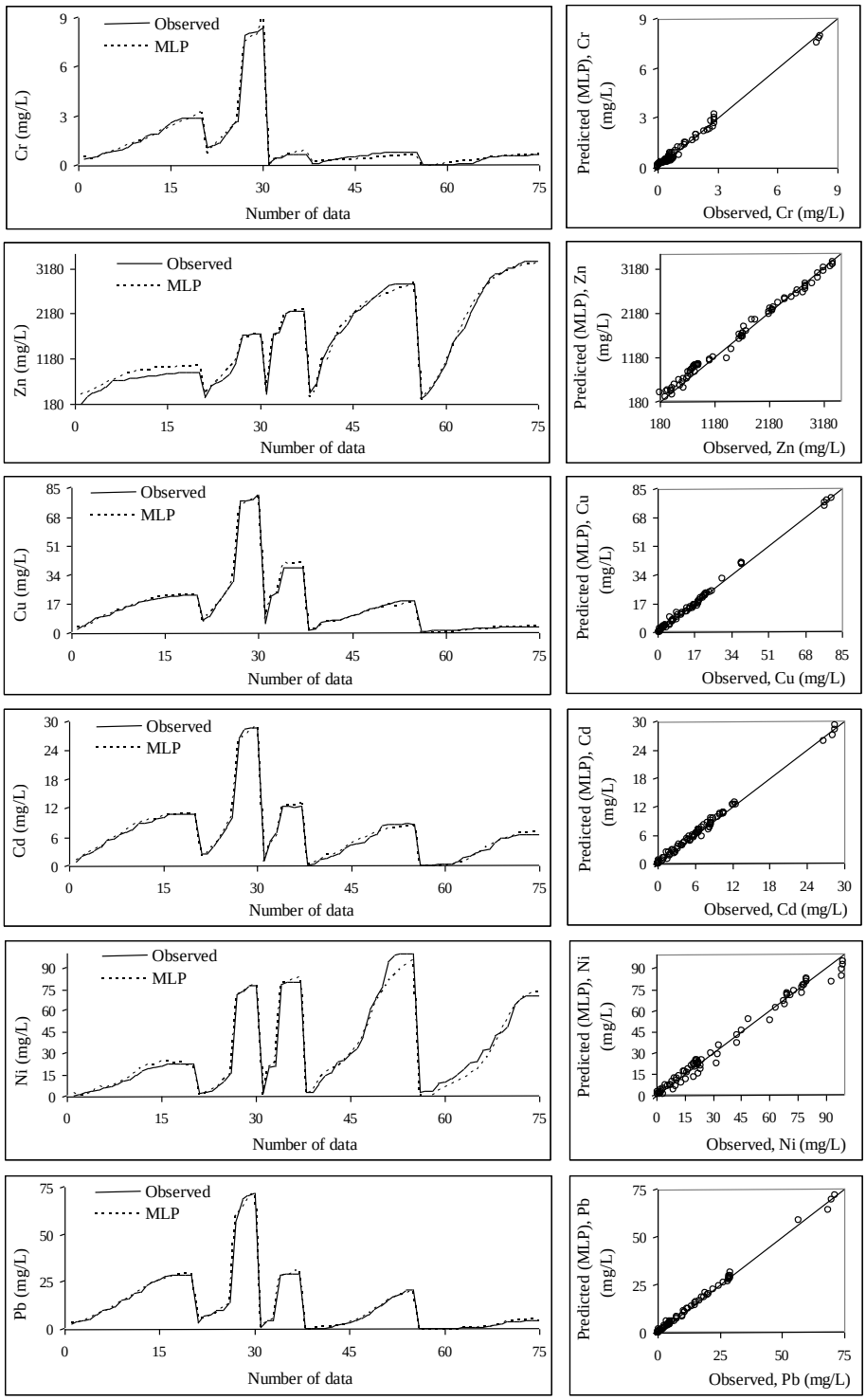


Figure 3. Comparison between observed and predicted heavy metal concentrations in training phase.

In Equations [4], [5] and [6], Y denotes the heavy metal concentrations and N is the total number of data. The MLP model was trained, tested and then the results were compared by means of MSE, MAE, MARE and R^2 statistics as shown in Table 3. As shown in Table 3, the MLP has the capability of modeling heavy metal concentrations. The performance of the MLP model analyzed herein is shown in Figures 3 and 4 for training

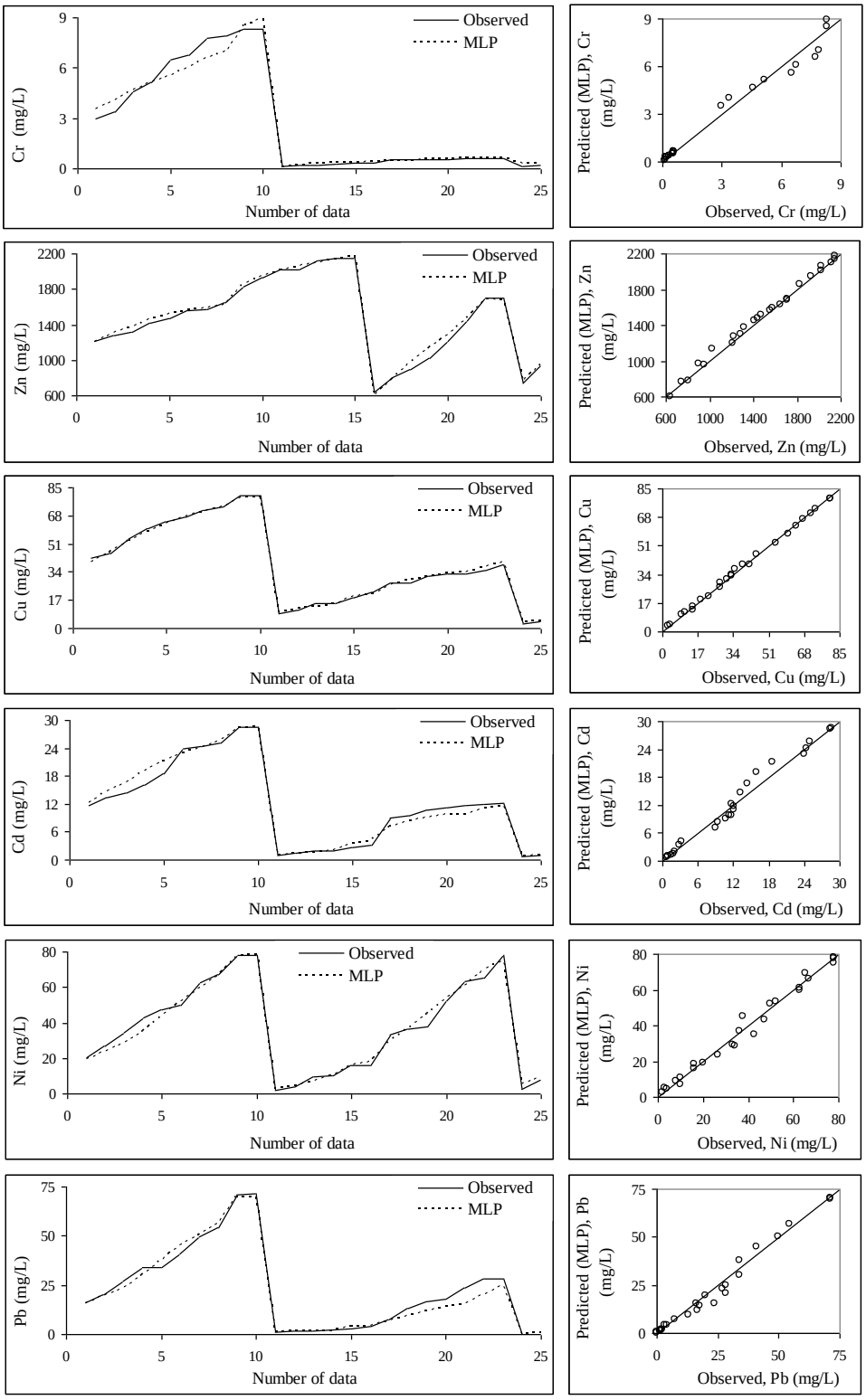


Figure 4. Comparison between observed and predicted heavy metal concentrations in testing phase.

and testing phases, respectively. As shown in these figures, the MLP produced highly accurate results in the estimation of heavy metal concentrations for both training and testing phases.

Conclusions

All biological models commonly used are developed using mass, energy, and momentum conversion. Moreover, most models have generally failed to define and predict the real system performance and behavior in advance during microbiological growth and bioleach. The development of practical mathematical models for these processes is difficult due to many affecting factors. The prediction models such as ANNs demonstrate stronger and more realistic behavior in predicting the complex non-linear and ever changing conditions of biological processes. ANN applications were previously used in some studies to predict the leaching of metals for bioleaching processes with similar or different microorganism groups and sludges from different industries and applications (Du et al., 1994; Acharya et al., 2006; Ozkaya et al., 2008; Liu et al., 2008; Jorjani et al, 2007; Nurmi et al., 2010; Laberge et al., 2000).

In this study, a three layer feed forward MLP neural network model was developed to predict the effluent heavy metal concentrations of bioleaching technique involving *A. ferrooxidans* in dewatered metal plating sludge containing no sulfide or sulfate compounds. The ability of multi-layer perceptron (MLP) neural network in the estimation of the effluent heavy metal concentrations in the CMB reactor was assessed in this paper by comparing the results with observed concentrations of heavy metals, namely Cd, Cr, Cu, Ni, Pb, and Zn. From the results obtained, the MLP technique with a Levenberg-Marquardt algorithm used in the current study appears to be a useful tool for the prediction of the effluent heavy metal concentrations in the CMB reactor. The results showed that the MLP neural network produced highly accurate estimation (R^2 over 97,9%) of the aforementioned metals. The results of this study support the findings of previous studies indicating that ANN is a strong modeling tool for the performance prediction of non-linear and time dependent biological processes.

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