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Prediction of recovery of gold thiosulfate on activated carbon using artificial neural networks

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Abstract

Since a high toxicity of cyanide which use as a reagent in the gold processing plant, thiosulfate has been recognized as an environmentally friendly reagent for leaching of gold from ore.

After gold leaching process it's important for recovery of gold from solution using adsorption or extraction methods, One of these methods is activated carbon.

The loading of gold from industrial thiosulfate solution that obtained from Zarshuran gold plant-Takab-Iran, onto activated carbon have been investigated. The affecting variables of the adsorption of gold on the carbon included, temperature, concentration of gold, size of activated carbon, pH and the ratio of amount of activated carbon to the volume of solution. The results show that at low concentration of gold in solution, effective loading can be achieved at pH 10.5. The size of activated carbon has a significant effect on the loading of gold on surface of activated carbon.

In this study the recovery of gold on activated carbon has been predicted using artificial neural network. For this purpose temperature, pH, the proportion of solution volume to weight of activated carbon, gold concentration and time of adsorption were taken as input parameters, whereas, the recovery of gold on activated carbon from thiosulfate solution was considered as an output parameter. The network with LMBP algorithm with two hidden layer were used and the topology 5-4-13-1 showed the best ability for prediction. Moreover sensitive analyze indicated that pH and temperature have substantial influence on adsorption.

Keywords: *Thiosulfate, gold, activated carbon, zarshuran gold plant, artificial neural network.*

1. Introduction

Although Cyanidation is currently the main choice for the extraction of gold from ores, because the toxicity of cyanide and the failure of this complexant to extract gold from the so-called difficult to treat raw materials (e.g. carbonaceous and copper-gold ores), it is necessary that this reagent Supersede with proper solutions which are less environmentally dangerous [1]. After leaching its indispensable, the gold recover on surface of adsorbent. It is found that gold thiosulfate ions can be adsorbed on surface of strongly base exchanger with high efficiency [2]. Furthermore the zinc and copper have represented good affinity for cementation of gold from ammonical thiosulfate solutions [3-5]. In other cases alkyl phosphorus esters, alkyl amines, TOMAC, amine oxides and their mixtures with amines are used to extract gold from thiosulfate solution [6-8]. Among the different techniques of gold thiosulfate adsorption, when there is low concentration of gold in the aqueous solution, activated carbon is a rational method [9].

From the point of view in engineering, in order to design an effectual recovery process, before scaling up the technology, an empirical model of adsorption operation is needed; however, these methods in some cases may not be completely suitable and must be substituted with efficient methods [1].

Many years, it has been a goal of engineering to develop intelligent machines with a large number of simple elements. Many current researchers have acquired artificial-intelligence-based techniques such as artificial neural network (ANN), genetic algorithm (GA) and fuzzy expert systems (FES) to be effective in solving and modeling some of the excessively complicated problems. ANN technique is a relatively new branch of artificial intelligence (AI) and has been developed since the 1980s [11]. Artificial neural networks or connectionist models, as they are sometimes refereed to, have been inspired by what is known as the brain metaphor. This means that these models try to copy the capabilities of the human brain into computer hardware or software [12]. An artificial neuron, as shown in Figure 1, is the basic element of a neural network. It consists of three basic components including weights, biases, and a single activation function.

Neural networks have appeared in the last decade as a promising computing technique which enables computer systems to exhibit some of the desirable brain properties. Different kinds of networks have been employed with success in different field of science and technology. Examples are applications in industrial process modeling and control, biological and ecological modeling [13].

In this paper, an attempt has been made to model the adsorption of gold thisulfate ions on surface activated carbon using ANN technique and has been determined the best structure of network. (ANNs are classified by their architecture (number of layers), topology (connective pattern, feed forward or recurrent, etc.) and learning rule).

2. Experiment

2.1.Material and methods

The purified gold-thiosulfate solution used in this work, was obtained from Zarshuran Gold Plant-Takab- Zanjan-Iran. Also the activated charcoal carbon, obtained from Merck Company, were washed with deionized water and were dried. Table 1 shows the main properties of the activated carbon.

Table 1. Properties of activated carbon

Character	Description
Form	charcoal activated granular
Size	~ 2.5 mm
Molar mass	12.01 g/mol
Density	~ 2 g/cm³ (20 °C)
pH value	~ 6 (50 g/l 20 °C) (slurry)
Bulk density	~ 400 kg/m³
Solubility in water	(20 °C) insoluble
Substances soluble in nitric acid	<=%5
n-Hexane adsorption	>=%30
Residue on ignition	<=%8
Loss on drying	<=%10

Gold adsorption tests were done in 500 mL vessels that were placed in a water bath. They were equipped with a magnetic stirring devise. The experiments were initiated by placing the amount of activated carbon that were weighted, then adding the certain amount of solution containing gold.

The range of pH and temperature was selected between 8-11 and 15-70°C in this experiment. The cause of varied pH rang and temperature is the gold thiosulfate solutions tend to decompose in pH more than 11 and lower than 8. another reason is that ammonia were applied as pH regulator. By increasing temperature more than 70°C, the loss of ammunia by volatilization goes up; consequently, quantity of ammoniac consumption increase [2].

During adsorption process, samples were taken in different times and then were analyzed by atomic adsorption spectrometry. The efficiency of adsorption process was estimated by the percentage of gold adsorption as the following:

$$\%\,\mathrm{Au}_{ads} = 100.([Au]_0 - [Au]_t) \big/ [\mathrm{Au}]_0 \qquad (1)$$

Where $[Au]_0$ indicates the initial gold concentration while $[Au]_t$ is the final gold concentration (mg/L).
According to the classification done by dubinin, the size of activated carbon pores (X) have been divided to macropores, transition or mesopores and micropores:
Macropores: X>100-200 nm
Transition or mesopores: 1.6<X<100-200 nm
Microipores: X<1.6 nm
The size of anion of gold thiosulfate is larger than anion of gold cyanide; therefore, it is necessary that the properties of chosen activated carbon for gold recovery from ammunical thiosulfate solutions, satisfied this difference. In other word the size of activated carbon must be often macropore or transition. The researches have demonstrated that activated carbon based charcoal have more mesopores rather than other activated carbons.
The used thiosulfate solution has been prepared from Zarshuran Gold Plant and it contains 2.3 mg/l gold, 1.97 mg/l silver and 790 mg/l copper.

The important factors in the process of gold adsorption on surface of activated carbon include: the percentage of impurities, pH, temperature, the concentration of gold, the amount of activated carbon, size of activated carbon, the time of agitation and finally the efficiency of agitation.

3. Results of experiment

3.1. Effect of proportion of liquid to solid

Experiments were carried out at three different ratios of liquid volume to weight of activated carbon in solution (l/s), namely 100, 200, 400 and 800 ml/g. The variation effect of proportion of liquid to solid (l/s) on recovery of gold on activated carbon has been shown in Figure 1. Based on the line graph, it can be state that the adsorption of gold thiosulfate ions increases with increasing the quantity of the activated carbon in solution. This result may be attributed to this fact that growing the amount of carbon brings about the active sites on surface of activated carbonincreases; consequently, the gold recovery increases.

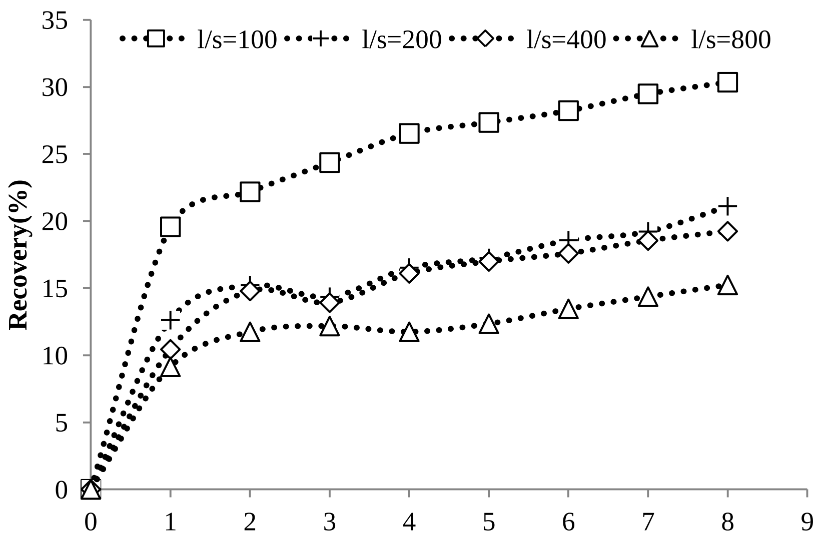

Figure 1. Effect of l/s ratio on the gold thiosulfate complexes recovery by activated carbon. Gold concentration= 2mg/l, pH=10 and T=25°C

3.2. Effect of temperature

As can be seen from Figure 2, the temperature has a significant effect on the adsorption of gold. It is clear that the higher temperature lead to the more adsorption rate. In 25°C, after 8 hours of adsorption time, the maximum recovery rate was 50.0%, while after same time period in 35°C and 45°C, the recovery percentage was 67.9% and 90.4%, respectively.

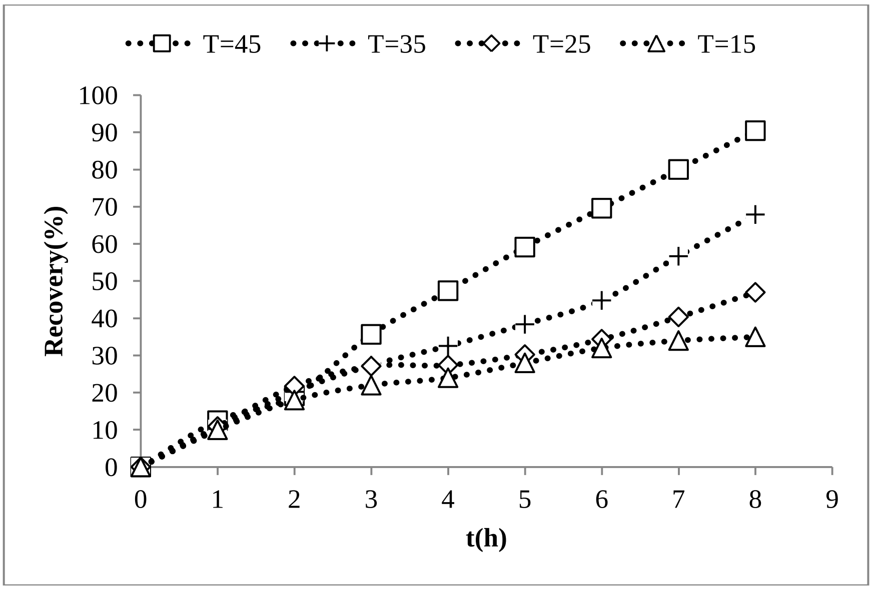

Figure 2. Temperature effect on the gold thiosulfate complex recovery by activated carbon. Gold concentration= 2 mg/l, pH=10 and l/s=100

3.3. Effect of pH

The influence of variation of pH on the adsorption of gold has been shown in Figure 3. Undoubtedly, it can be inferred that in pH higher and lower than 10.5, the gold adsorption has been severely decreased. The Optimum gold adsorption was achived at pH 10.5. This result may be ascribed to the competitive adsorption of OH with gold ion on the carbon surface. It is clear from Figure 3. Generally, the adsorption rate of gold on activated carbon during initial stage is relatively faster than other stages. According to results, the maximum recovery has been recorded in pH 10.5 which was equal to 86% while in pH 10 the recovery was 40%.

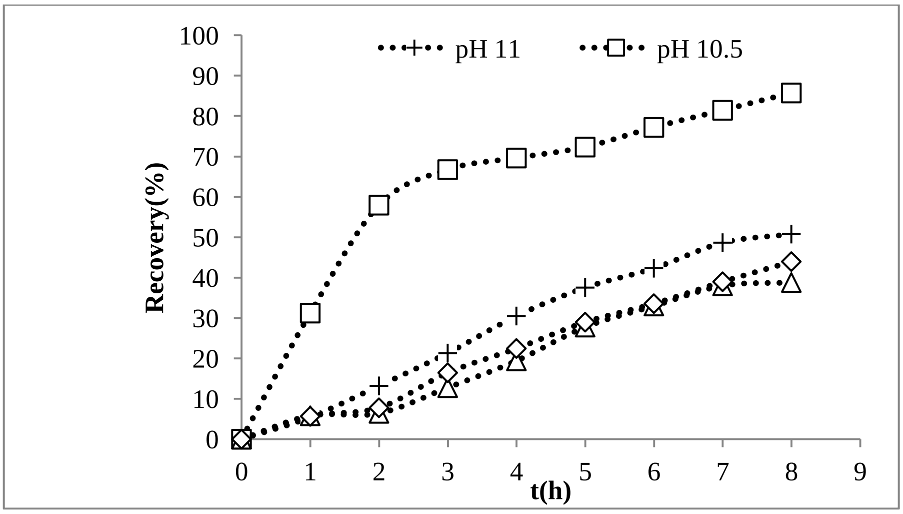

Figure 3. PH effect on the gold thiosulfate complex recovery by activated carbon. Gold concentration= 5mg/l, T=45°C and l/s=100

Figure 4 shows that at lower concentration of gold in solution and higher proportion of liquid to solid, the influence of pH variation remarkably reduced, . i.e. increasing the amount of activated carbon in solution, sensitivity of adsorption process decreases.

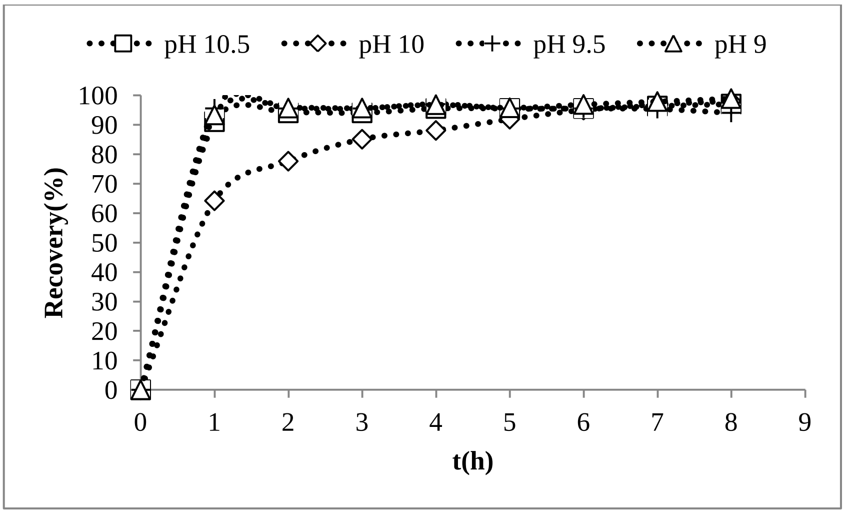

Figure 4. PH effect on the gold thiosulfate complex recovery by activated carbon. Gold concentration= 1.0 mg/l, T=25°C and l/s= 50

3.4. Effect of initial gold concentration

The effect of initial gold concentration on the adsorption by activated carbon was studied and the results are shown in Figure 5. As can be seen from the graph, the fluctuation of initial gold concentration between 2 and 5 mg/l had no remarkable effect on final recovery (after 8 h); However, the rate of adsorption, especially in initial stage, has been affected by changing initial gold concentration. It is clear that with increasing the initial gold concentration in aqueous solution, the probability of encountering gold thiosulfate ions with macropores of carbon surface increased. As a result the adsorption rate in higher initial gold concentration was faster than lower gold concentration.

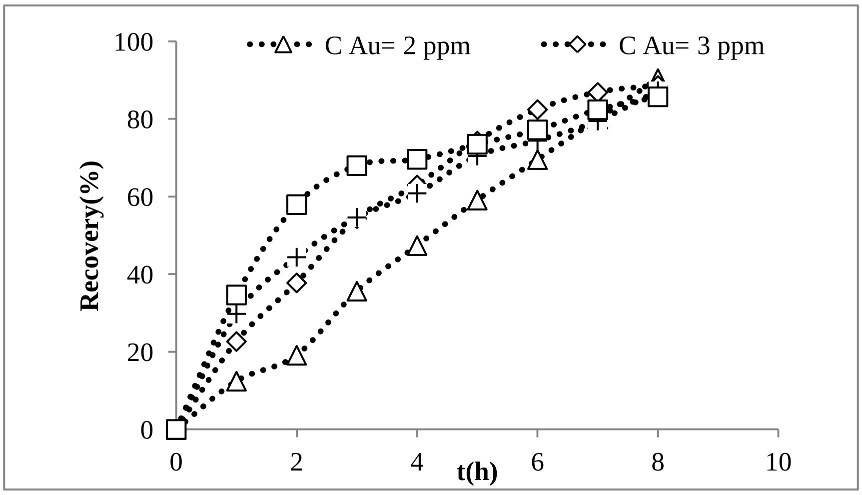

Figure 5. Effect of initial gold concentration. T=45°C, l/s=100 and pH=10.5

3.5. Effect of activated carbon sizes

The following experiment were accomplished to determine the effect of activated carbon size on the percentage of gold adsorbed and adsorption rate,. Three different sizes of carbon were used (-75 microns, +75-150 microns and +150 microns). As shown in Figure 6 in the middle sizes, the recovery of gold and the adsorption rate were higher than the two other sizes. Supposedly, This may be due to the fact that in the size of +75-150 microns, more macropores were available than other sizes. In such circumstances the probability of encountering gold thiosulfate ions with macropores increased; therefore, the adsorption rate raised.

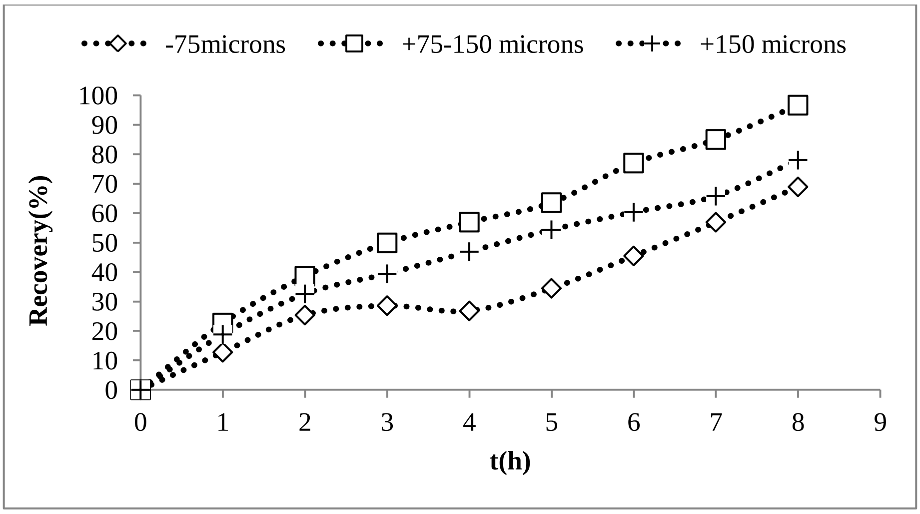

Figure 6. Effect of sizes of activated carbon. gold concentration= 2mg/l, T=45°C, l/s=100 and pH=10.5

4. Artificial neural network

Artificial neural network (ANN) has the potential to learn from the set dates and predict values at locations where no dates are present. A neural network includes interconnected neurons that may have several input layers, hidden and output is working sequentially and parallel. When an input pattern is introduced to the neural network, the synaptic weights between the neurons are stimulated and these signals propagate through layers then an output pattern is formed. The weights between layers and the neurons are modified in such a way that next time the same input pattern is introduced and then neural network will provide an output pattern which will be closer to the expected response. In order to increase or decrease the input that goes into the activation Function, the bias term is added to the summation at the neuron [14].

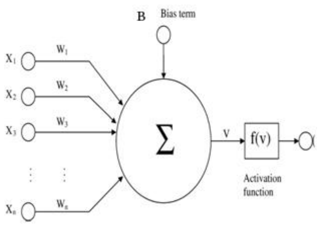

Figure 7. The basic model of neuron

4.1. Multi-layer perceptron (MLP) network

ANNs are classified by their architecture (number of layers), topology (connective pattern, feed forward or recurrent, etc.) and learning rule. Most of the engineering systems have used multi-layered feed-forward networks and use error back propagation (BP) learning. In this network Training data is passed through the system which then back propagates to assess errors and adapts by adjusting weights. Once the errors become small enough to meet specified criteria, the architecture is established and is then ready for classification or prediction depending upon the need [15, 16].

Various algorithms have been suggested for training of neural networks although the back propagation algorithm is the most versatile and robust technique, and provides the maximum effective learning procedure for MLP networks. [16].

Usually a neural network with too few hidden neurons is unable to learn adequately from the training data set whereas a neural network with too many hidden neurons will permit the network to memorize the training set instead of generalizing the acquired knowledge for unseen patterns. Each neuron in an ANN is an independent processing element, having its own inputs and output. Its function is that of a distributed parallel computation (Figure 8). The output is calculated by the equation [17].

$$O_j = f\left(\sum_{i=1}^{n} x_i w_i\right) \tag{2}$$

Where

x_i = the ith input,

w_i = the connection weight associated with the i th input,

O_j = output of the jth neuron, and

f = transfer function.

The training process consists of two steps. In the first step, the training patterns (a set of known input and output pairs) were obtained from the database is fed into the input layer of the network. These inputs are then propagated through the network until the output layer is reached. The output of each neuron is computed by the transfer function, as shown in Equation 2, which “squashes” the range of input to be between 0 and 1.0. Then, a forward preprocessing error is calculated using the following equation. In the second step, the error is minimized by back propagation through the neural network [17].

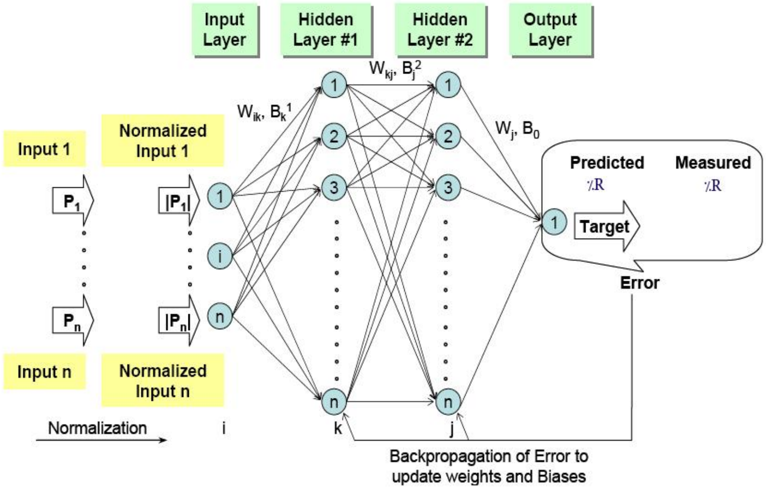

Figure 8. A neural network with feed forward back propagation algorithm

Nonlinear (LOGSIG, TANSIG) and linear (POSLIN, PURELIN) functions can be used as transfer functions (Figure 9 and Figure 10). The logarithmic sigmoid function (LOGSIG) is defined as [11].

$$f = {}^{1}\!/_{(1+e^{-e_x})} \qquad (3)$$

$$f = {}^{e^{e_x} - e^{-e_x}}\!/_{e^{e_x} - e^{-e_x}} \qquad (4)$$

Where, e_x is the weighted sum of the inputs for a processing unit.

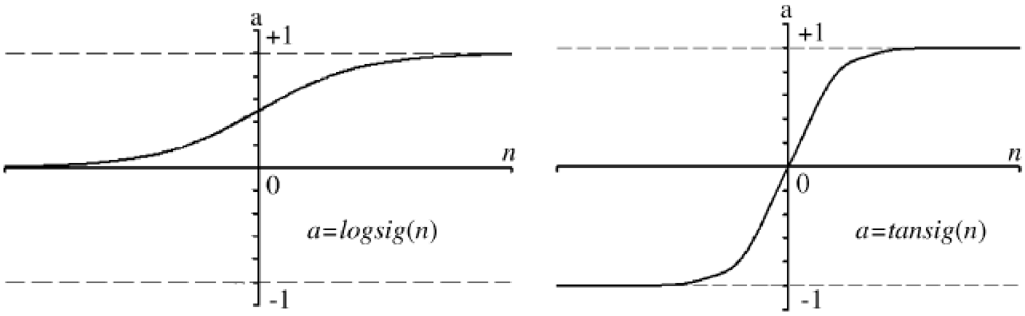

Figure 9. Sigmoid transfer functions

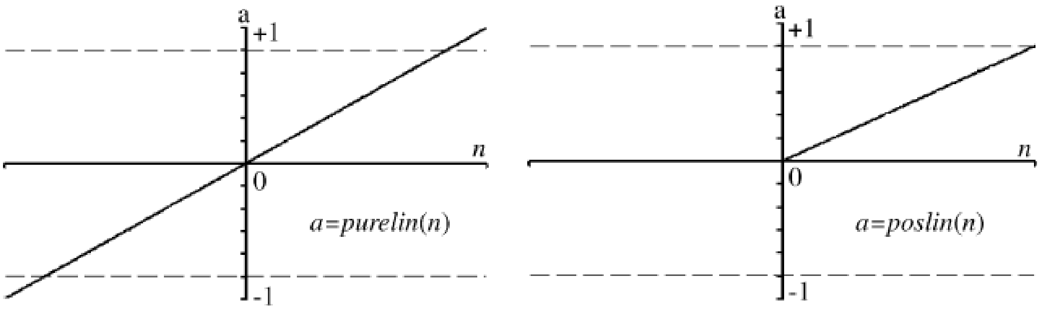

Figure 10. Liner transfer functions

4.2. Data set

A total number of 240 recoveries of gold on activated carbon were recorded at different condition of adsorption and results were used for the neural network modeling. These data sets are divided into training (90%) and testing (10%) and then the network using automated regularization methods were trained. Temperature, pH, the ratio of solution volume to weight of activated carbon, gold concentration and time of adsorption were taken as input parameters where as the recovery of gold on activated carbon from thiosulfate solution was considered as an output parameter. The minimum and maximum amount of all the parameters and their respected symbols are given in Table 2.

Table 2. Input and output parameters used for training the neural network and their range

Parameter	Symbol	Description	Min	Max
Input	A	pH	8	11
Input	B	Solution/ AC (ml/mg)	50	1600
Input	C	Temperature (°C)	15	70
Input	D	Time (h)	1	8
Input	E	Gold concentration (ppm)	0.7	6
Output	F	Recovery of gold on activated carbon (%)	2.3	98

4.3. Discussion

In this study, different combination of MLP networks with one and two hidden layer were used and the optimum network structure was determined using root mean squared error (RMSE) and mean absolute error (MAE):

$$RMSE = \sqrt{(O_i - T_i)^2 / N} \tag{5}$$

$$MAE = |\mathrm{T_i} - \mathrm{O_i}| \tag{6}$$

Where, $\mathrm{T_i}$, $\mathrm{O_i}$ and N are representative of the measured output, the predicted output and the number of input-output date sets, respectively.

Based on Table 3 it can be stated that a network with transfer functions TTPU, topology 4-13-1 with the maximum R^2, logical RMSE and MAE is the optimum model for prediction. It is also evident from Table 3 that several models are similar or near amount R^2, that in these cases the network with minimum neurons is the optimum model. In this work, performance of model based on a comparison and correlation between actual and prediction using testing set dates was evaluated. The corresponding results have been displayed in Figure 11 and Figure 12.

Table 3. Results of a comparison between some of the models based on maximum R^2

No	Transfer function	Model	RMSE	MAE	R^2
1	TANSIG-TANSIG-PURELINE (T-T-PU)	4-13-1	1.8525	1.2388	0.982
2	TANSIG-LOGSIG-PURELINE (T-L-PU)	18-14-1	0.8453	0.5523	0.981
3	TANSIG-TANSIG-POSLIN (T-T-PO)	8-19-1	1.0216	0.5717	0.982
4	TANSIG-PURELINE (T-PU)	13-1	1.6644	1.2119	0.979
5	LOGSIG-TANSIG-POSLIN(L-T-PO)	20-9-1	4.1049	2.737	0.982
6	LOGSIG-LOGSIG-PURELINE(L-L-PU)	18-12-1	1.5769	1.111	0.983

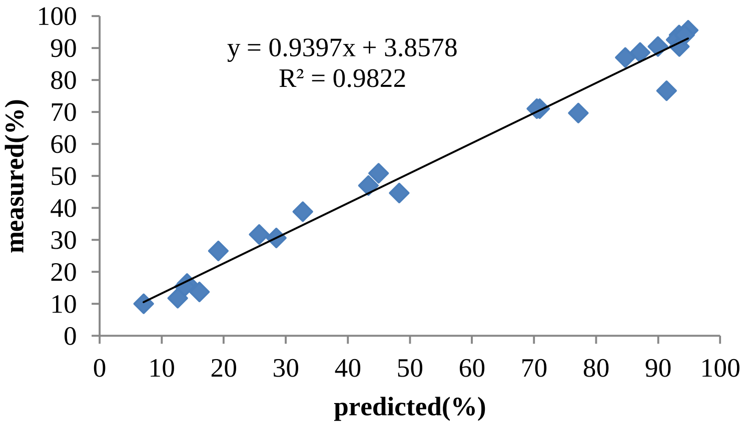

Figure 11. Correlation between Measured ANN predicted recovery

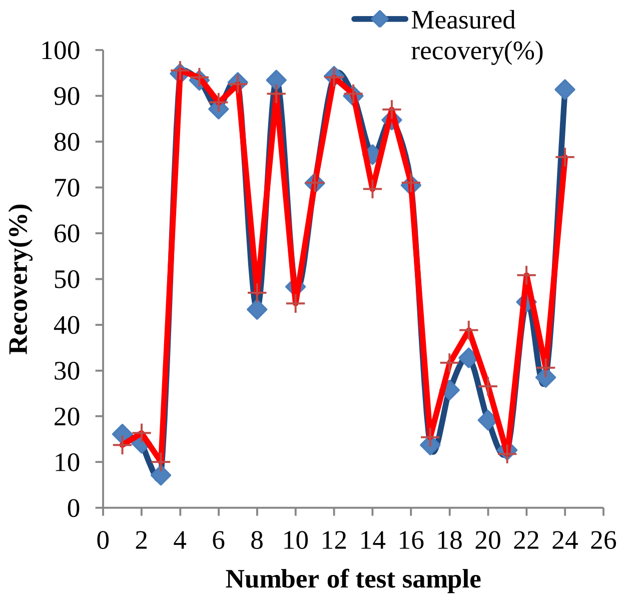

Figure 12. Comparison between measured and predicted recovery of gold on activated carbon for different type of pattern

As can be seen from Table 4, when optimum network were chosen bases on RMSE and MAE, as network performance index, the network with topology (19-20-1) showed the best prediction ability among them. In this way, the strength of estimation of network (R^2) is 0.94 Whereas R^2 is chosen for estimate of network performance, it increases to 0.98.

Table 4. Results of a comparison between some of the models based on maximum MAE and RMSE

No	Transfer function	Model	RMSE	MAE	R^2
1	TANSIG-TANSIG-PURELINE (T-T-PU)	15-18-1	0.3096	0.0534	0.89
2	TANSIG-LOGSIG-PURELINE (T-L-PU)	3-20-1	0.407	0.194	0.93
3	TANSIG-TANSIG-POSLIN (T-T-PO)	3-19-1	0.6536	0.3583	0.86
4	TANSIG-PURELINE (T-PU)	20-1	0.8885	0.5579	0.89
5	LOGSIG-TANSIG-POSLIN(L-T-PO)	19-20-1	0.539	0.3045	0.94
6	LOGSIG-LOGSIG-PURELINE(L-L-PU)	18-17-1	0.4128	0.2077	0.89

Saffarzadeh and heidaripanah [18] showed in a network with LMBP algorithm without automated regularization and with MSE as performance index, whit a specified number of neurons in the hidden layer, by decreasing training error and MSE, the prediction ability (R2) of the network increases as it reaches its maximum value. After that, by continuing training and decreasing the MSE, network over fits and the generalization ability reduces significantly [18].

In this study, the LMBP algorithm with the automated regularization with two network performance index, RMSE and MAE, has been used. As shown in Table 4, the variation of R^2 due to RMSE and MAE has no specific trend; therefore, the network should be trained in different area value of epochs and then training network must stop at the MAE and RMSE in which the R^2 has the maximum value. As can be seen from Table 4, when one network is chosen based on MAE or RMSE, may will achieved an inappropriate result. In Figure 13, R^2 is in good agreement with RMSE and MAE, respectively and the best prediction reached in 300 epochs, but basically no in the lower MAE or RMSE.

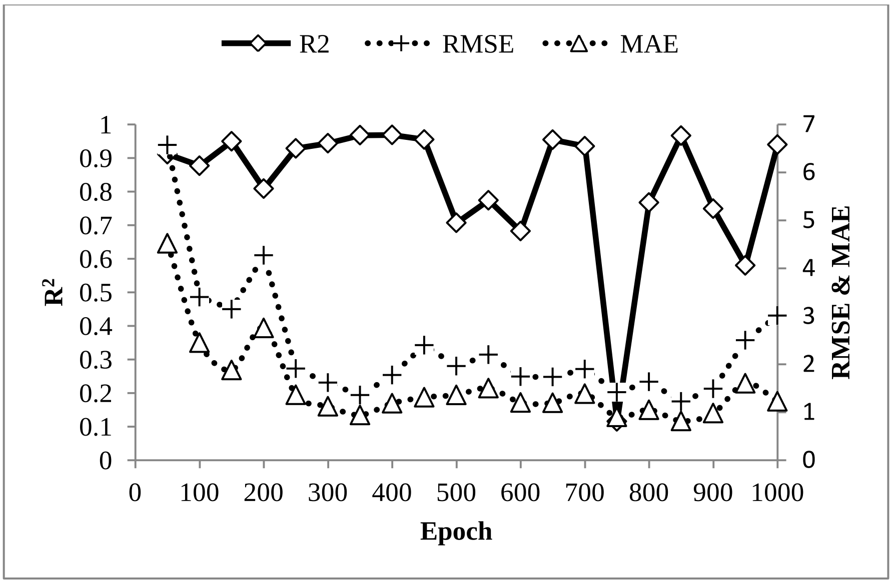

Figure 13. The variation of R2 and MAE vs. epoch

5. Statistical modeling

To evaluation efficiency of neural network, an attempt has been done for the prediction output data using regression model. The regression model can obtain correlation between output variable and input variables (equation 7). For this purpose, in this study, the datasets which were used to training of neural network are also applied for regression model. Using the following regression model, the correlation between the measured and predicted recovery is illustrated in Figure 14.

$$R = 132.6 - 45.82B \times E + 10.553A \times C + 1.507A - 0.004D + 0.763D \times D - 7.205C \times D - 3.712C \times C \quad (7)$$

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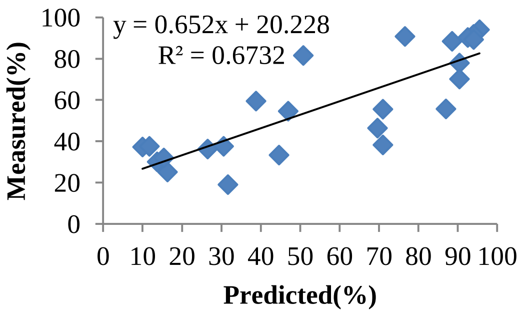

Figure 14. Correlation between measured and predicted recovery using regression model

Also the comparison between performances of two used models has been shown in Figure 15. As it can be seen, the prediction ability (R2) of neural network model is much better than regression model.

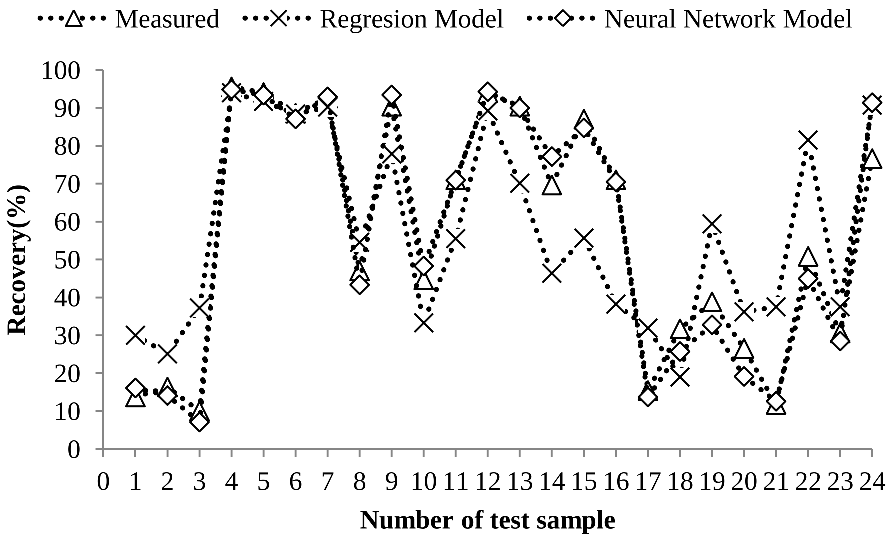

Figure 15. Comparison between measured and predicted recovery of gold on activated carbon for different type of pattern

6. Sensitivity analysis

The cosine amplitude method (CAM) was used to determine the strength of each input parameters on recovery of gold thiosulfate on activated carbon. The CAM is used to obtain the express similarity relations between the related parameters. To reach this purpose, all input-output pairs were expressed in common X-space. The data pairs used to construct a data array X is defined as:

$X = \{x_1, x_2, x_3, \ldots, x_i, \ldots, x_n\}$

Each of elements, X_i, is defined as

$X_i = \{x_{i1}, x_{i2}, x_{i3}, \ldots, x_{im}\}$

Thus, each data set can be thought of as a point in m-dimensional space, where each point requires m-coordinates for a full description. The strength of the relation between the data set, x_i and x_j, is given by the membership value expressing the strength:

$$r_{ij} = \frac{\sum_{k=1}^{m} X_{ik}X_{jk}}{\sqrt{\sum_{k=1}^{m} X_{ik}^2 \sum_{k=1}^{m} X_{jk}^2}} \quad (8)$$

The result based on r_{ij} values is shown in Figure 16. Based on the bar chart, it can be stated that pH, temperature and somewhat time of contact are the most effective parameters on the gold recovery on the surface of activated carbon; hence, it is necessary to regulate these parameters precisely prior to start the experiments.

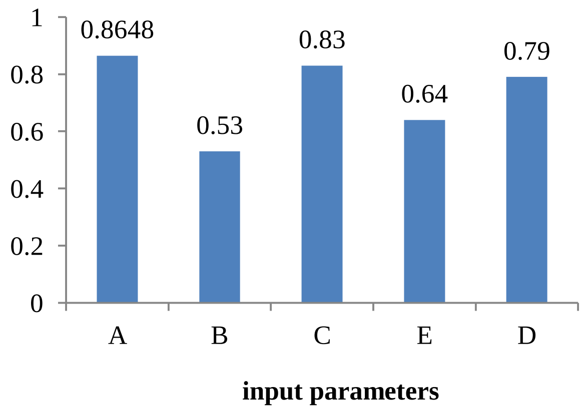

Figure 16. Sensitive analyze of input parameters

7. Conclusions

The possibility of gold adsorption from aqueous ammunical thiosulfate solutions that were obtained from Iranian Zarshuran gold plant, on to activated carbon were studied. The pH solution has a significant effect on adsorption process, which the optimum gold adsorption occurred at pH: 10.5. The obtained results revealed that by decreasing in l/s ratio, the negative effect of pH variations can be reduced; also, the results manifested the higher temperature, give rise to the higher adsorption rate three different sizes of carbon were used. Results of this section pointed that in the middle size (+75-150 microns), the recovery of gold and the adsorption rate were higher than the two other sizes.

Among the different statistical and empirical methods, the ANN was the best technique for the prediction of gold recovery on activated carbon from ammunical thiosulfate solution.

Temperature, pH, the ratio of solution volume to weight of activated carbon, gold concentration and time of adsorption were taken as input parameters whereas the recovery of gold on activated carbon was considered as an output parameter. For this purpose a database consisting of 240 datasets were used for training (90%) and testing (10%). As shown in this study, a network with LMBP algorithm and a 5-4-13-1 neuron topology were expressed the best ability for prediction, that in this way the performance index R^2 was equal to 0.98. The statistical analysis indicated that the prediction ability of neural network model is much better than regression model. According to sensitive analyze, pH, temperature and a little time of contact are the most effective parameters on the recovery of gold on the surface of activated carbon; hence, it is necessary to regulate these parameters before starting the experiments.

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