

Design and implementation of heavy metal prediction in acid mine drainage using multi-output adaptive neuro-fuzzy inference systems (ANFIS) - a case study

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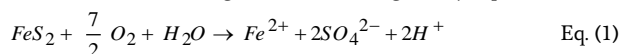
ABSTRACT

This paper reports an attempt to show how acid mine drainage (AMD), as well as other heavy metals, pollute the environment and how this problem can be resolved. AMD is considered to be the main source of environmental pollution in areas where mining operations are undertaken. Since AMD and the factors that control it are of prime importance regarding the environmental preservation activities, this study investigates the presence of heavy metal pollutants in AMD. To achieve this goal, we implemented the ANFIS method to predict the presence of heavy metals (Zn, Mn, Fe, and Cu), taking into account pH, as well as SO_4 and Mg concentrations. Having used the ANFIS method, the comparison of predicted concentration with calculated data resulted in correlation coefficients of 0.999, 0.999, 0.999, and 0.999 for Cu, Fe, Mn, and Zn, respectively. The employed procedure proved to be easy to use and cost-effective to foresee the presence of heavy metals in AMD.

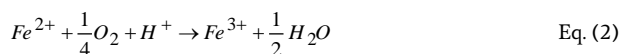
Keywords : ANFIS, Heavy Metals, Acid Mine Drainages, Shur River, Sarcheshmeh Copper Mine

1. Introduction

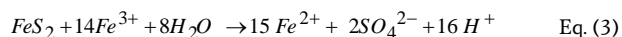
Acid mine drainage (AMD) is a severe environmental issue in the mining industry and is responsible for water pollution surrounding mining areas. [1,2]. AMD typically consists of a high concentration of iron, sulfates, and various concentrations of trace elements having low pH [3,4,5]. Therefore, AMD is considered among the main threats to the environment near mining operations, and where sulfide-rich dumps are stored [6]. Not many studies in the literature have tried to tackle this problem. The prediction and control of this issue requires comprehension of chemical reactions playing active roles in pyrite oxidation. The overall stoichiometric reaction that can describe pyrite oxidation, as well as AMD generation, can be given by Eq. (1) [7]:



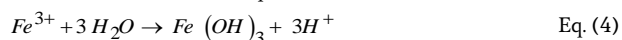
As Equation (1) shows, 1 mole of Fe^{2+} and 2 moles of SO_4^{2-} and H^+ are produced if 3.5 moles of oxygen reacts with 1 mole of pyrite.



As Eq. (1) shows, Fe^{2+} may be further oxidized by O_2 and then transformed into Fe^{3+} [8].



As Eq. (3) shows, Fe^{3+} can also oxidize pyrite to generate additional Fe^{2+} and SO_4^{2-} according to Eq. (3). It should be noted that reaction 2 seems to be rather slow at low pH values [8]:



Based on Eq. (3), pyrite is hydrolyzed into amorphous iron hydroxide. In addition, at pH values higher than 3.5 (Eq. 4), the H^+ ions are released in the environment. Indeed, factors such as moisture, acidophilic Fe microorganisms, pH, and temperature control the rate of pyrite oxidation. In addition, the existence of oxygen inside the trailing is mainly controlled by the gaseous diffusion process [9]. AMD with low pH and high SO_4 and iron contents can potentially move toxic metals, such as Cu, Pb, Ni, Mo, Co, Zn, Cd, and other dissolved materials from the trailing sites, in the environment.

The Sarcheshmeh Copper Complex (SCC) located in the southeast of Iran is considered as the third-largest copper mine in the world containing around 1 billion tons of ore with an average grade of 0.03% molybdenum and 0.9% copper [12]. Mining activities and mineral processing operations have produced enormous amounts of low-grade waste and tailing materials that are causing serious environmental problems [12]. The tailing materials in the SCC contain reactive sulfide minerals, particularly pyrite [13].

Many studies have focused on the environmental issues related to the oxidation of sulfide materials and AMD in the SCC and their effects on the Shur river located in the area [14, 15, 16, 17, 18, 19, 20]. Modeling systems play a significant role in this regard since they help researchers understand how the system is functioning.

In recent years, water treatment scholars have focused on the design and construction of facilities to control heavy metals instead of predicting their dispersion patterns. It is due to the fact that the management and operation of AMD play a rather significant role in the efficiency of water quality control systems. The Shur river, which is located near the SCC, is already contaminated with acidic waters (pH values of 2 to 4.5) and with large amounts of heavy metals resulting from AMD.

To develop appropriate monitoring as well as remediation

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procedures, the prediction of heavy metals content in water using fast and cost-effective methods, such as the Adaptive neuro-fuzzy inference system (ANFIS), is of high importance. Artificial neural networks (ANNs) are among the prevalent techniques gaining attention in different areas of engineering in recent years. Neural networks are considered robust in environmental studies since they incorporate a non-linear model-free estimator. Several such studies have been conducted reporting successful implementation of these techniques to forecast the presence of heavy metals in water systems. For instance, the general regression neural network (GRNN) has been implemented to predict the presence of Rare Earth Elements (REEs) in neutral alkaline mine drainage samples obtained from the Razi coal mine (RCM) in the northeast of Iran [21].

In another similar study, ANN was implemented to predict the presence of oxidation for pyrite in a coal washing refuse pile [22]. Other researchers have implemented Applied Artificial Neural Networks (AANN) to predict the presence of heavy metals in AMD from the Shur river near the SCC [19]. Another group of researchers predicted the presence of toxic metals using Artificial Intelligence Techniques (AIT) [23]. Two researchers applied Modular Neural Networks (MNN) and estimated the distribution of nitrate in surface waters using on-surface nitrogen recharge and loading data [24]. Two Japanese researchers implemented neural networks to determine principal metal contents in the Hokuroku district located in northern Japan [25]. They also implemented the same procedure to evaluate the content of impurities in a limestone mine, namely, SiO_2 , Fe_2O_3 , MnO , and P_2O_5 .

The performance of neural networks has also been compared through geostatistical approaches to estimate bauxite and gold grades. Two research studies estimated the presence of heavy metals in soil samples derived from reflectance spectroscopy by implementing a backpropagation network coupled with the multiple linear regression method [26]. In addition to these techniques, fuzzy logic has been increasingly implemented in the mining industry. For instance, [27] investigated the application of fuzzy logic to predict the rate of roof fall in coal mines. Another group of researchers implemented support vector machines to assess heavy metal pollution in the Shur river near the SCC [14]. Some others used fuzzy logic to estimate rock fragmentation coming from blasting in the Gol Gohar mine in Iran.

This study, therefore, was devoted to the combination of fuzzy logic (FL) and Artificial Neural Networks (ANNs) to produce a powerful tool to estimate the presence of heavy metals. Combining ANN with ANFIS, some researchers used these two to predict the existence of pyrite oxidation in a pile located in a coal washing site [29]. Two researchers implemented an adaptive neuro-fuzzy inference system to estimate the grade of waste materials in the SCC [30]. Moreover, in recent decades, several studies have been conducted in environmental engineering using ANFIS [13, 17, 31, 32, 33, 34, 35, 36, 37, 38, 39, and 40].

Having reviewed the literature in the field, we came to the conclusion that although ANFIS has been implemented in the mining industry and its related environmental problems, no applications have been reported for predicting the presence of heavy metals in AMD [41]. In this study, we focused on heavy metal prediction in the Shur river originating from AMD. The obtained results from ANFIS estimations were compared with actual samples derived from the river. The authors hoped this would encourage the implication of this fast and cost-effective method to predict the presence of heavy metals. The data used in this study are taken from Aryafar et al. (2012).

2. Case study

The Sarcheshmeh Copper Complex (SCC) is located in the Kerman province in central Iran [7]. The complex is situated at an average elevation of 1,600 m and an average annual precipitation value of 300 to 550 mm [14]. The temperature in the area fluctuates between +35C in summer and -20C in winter. Fig. 1 depicts the geographical location of the mine. The mineral processing activities in the mine has resulted in the creation of more than 24 Mt of tailings measuring approximately 13 meters in height and covering an area of about 2 to 4 squared kilometers.

The catchment area occupied by the Shur river is around 220 km² and the discharge rate is measured to be 0.52 m³/s [29].

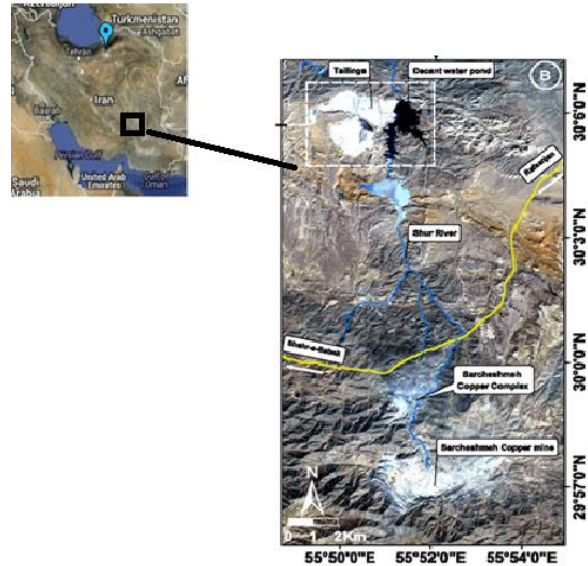


Fig. 1. Geographical location of the complex and the study area [42].

3. Materials and Methods

3.1. Neuro-fuzzy system

A neural-fuzzy model is considered to be an efficient method to model linear systems. It has been derived from artificial intelligence techniques combining the merits of ANN with FIS [43]. The basic idea behind this combination is to develop an architecture implementing fuzzy logic to display knowledge in an interpretable way and to provide a platform for optimizing various parameters involving in the process. The resulting combination constitutes an interpretable model that can learn from the data and implement prior knowledge [41].

An adaptive network is considered to be a network consisting of nodes as well as directional links connected to an ANFIS, which was proposed for the first time by Jang [44]. In addition, nodes are adaptive, meaning each output from such nodes depends on the parameters related to the relevant nodes, and the learning rules decide how and when these parameters should be changed to reduce a prescribed error measure [45, 46]. Since ANFIS can be easily adapted and implemented for a given input/output task, it can be an attractive choice to be implemented successfully for various applications. On the other hand, ANFIS combines ANN and FIS into a *compound*, meaning there are no boundaries differentiating the respective features of both ANN and FIS [47, 48].

Neuro-fuzzy is considered to be a common framework to solve complicated problems [49]. If information is expressed in terms of linguistic rules, one can easily build a fuzzy system generating inferences. If it is given the data or can be trained from a simulation, artificial neural networks can be implemented. In order to build a FIS, the fuzzy sets, fuzzy operators, and the databases have to be denoted; similarly, in order to create an ANN, the architecture, as well as the algorithm, have to be specified. Therefore, as far as FIS is concerned, the learning ability can be considered an advantage, and as far as ANN is concerned, the formulation of a linguistic rule base will be considered as the merit [50]. Fig. 2 depicts a six-layer architecture proposed for multiple output ANFIS, the functionality of which is given below:

The form of the two-rule Sugeno ANFIS:

$$\text{If } x \text{ is } A_1 \text{ and } y \text{ is } B_1 \text{ THEN } f_1 = p_1x + q_1y + r_1 \quad \text{Eq. (5)}$$

$$\text{If } x \text{ is } A_2 \text{ and } y \text{ is } B_2 \text{ THEN } f_2 = p_2x + q_2y + r_2 \quad \text{Eq. (6)}$$

In order to achieve network training, there exist two passes, namely, forward and backward. Each layer within the forward pass will be taken

into consideration. Layer by layer, this forward pass broadcasts the input vector via the network. The error in the backward pass is returned back via the network in a manner compatible with backpropagation.

Layer 1: The output for each node is:

$$O_{1,i} = \mu_{A_i}(x) \quad \text{for } i = 1, 2 \tag{Eq. 7}$$

$$O_{1,i} = \mu_{B_{i-2}}(y) \quad \text{for } i = 3, 4$$

So, the $O_{1,i}(x)$ is the membership grade for x and y .

The membership functions are given only for illustrative purposes; therefore, we will implement the function which seems to be bell-shaped by:

$$\mu_{A_i}(x) = \frac{1}{1 + \left| \frac{x - c_i}{a_i} \right|^{2b_i}} \tag{Eq. 8}$$

where a_i, b_i, c_i are parameters that have to be learned. These can be considered as the premise parameters.

Layer 2: Every node in the layer is permanent. This is where the t-norm is implemented to 'AND' the membership grades - for instance, the product:

$$O_{2,i} = w_i = \mu_{A_i}(x) \mu_{B_i}(y), \quad i = 1, 2 \tag{Eq. 9}$$

Layer 3: This layer containing fixed nodes, calculates the ratio for the firing strengths for the rules:

$$O_{3,i} = \bar{w}_i = \frac{w_i}{w_1 + w_2} \tag{Eq. 10}$$

Layer 4: The nodes in this layer are presumed to be adaptive and accomplish the consequent of the rules:

$$O_{4,i} = \bar{w}_i f_i = \bar{w}_i (p_i x + q_i y + r_i) \tag{Eq. 11}$$

The parameters in this layer (p_i, q_i, r_i) have to be specified and are therefore named as the consequent parameters.

Layer 5: In here, we only have a single node computing the overall output:

$$O_{5,i} = \sum_i \bar{w}_i f_i = \frac{\sum_i w_i f_i}{\sum_i w_i} \tag{Eq. 12}$$

This is how the input vector is passed via the network in a layer-wise fashion. It is now time to consider how ANFIS learns the consequent and premise parameters for the rules and membership functions. There exist a number of various approaches; however, we only discuss the hybrid-learning algorithm proposed by Jang, implementing a combination of Least Square Estimation (LSE) and Steepest Descent (SD).

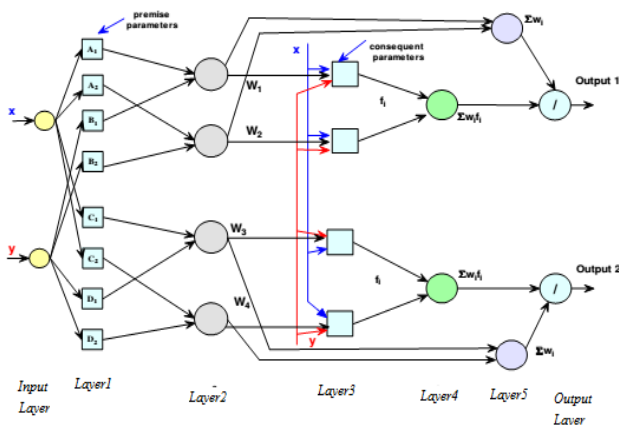


Fig. 2. The architecture of ANFIS with multiple outputs (two input and output).

4. Results and Discussion

In this study, we reported the building of an efficient ANFIS to predicate the concentration of heavy metals. In our study, ANFIS was applied for four outputs to model the predicted values using the MATLAB program. Based on the given matrix given in Table 1, pH, SO₄, and Mg, which are mostly affected by heavy metals (Zn, Mn, Fe, and Cu), were given as input parameters to ANFIS models, and Cu, Fe, Mn, and Zn were selected as the output, respectively. The generated models were trained with 48 datasets. Table 1 displays the correlation matrix among heavy metal concentrations, and the variables were considered independent.

Table 1. Correlation matrix between independent variables and the concentration of heavy metals.

EC (IS/cm)	TDS	Zn	Mn	Fe	Cu	Mg ²⁺	Ca ²⁺	HCO ₃ ⁻	Cl ⁻	SO ₄ ⁻	pH	
870	446	0	0.04	0	0	13	92	0	0	27	3.3	Min
2260	2080.68	31.48	52	23	158	123	460	628	230	1526	7.20	Max
1306.52	1009.90	6.33	16.05	4.60	20.29	56.70	182.78	34.01	23.56	778.45	5.27	Mean

Based on the requirements of neuro-fuzzy, the data for both input and output variables had to be normalized to an interval by a transformation process. We had 58 datasets available, and the concentration of most of the chemical parameters in running waters was below the standard levels. Data normalization was implemented among the range of [-1, 1] using Eq. (12) and the number of test (13) and train data (48) were randomly appointed:

$$p_n = 2 \frac{p - p_{\min}}{p_{\max} - p_{\min}} - 1 \tag{Eq. 13}$$

Where, p_n is the normalized parameter, p indicates the actual parameter, p_{\min} denotes a minimum of the actual parameters, and p_{\max} refers to a maximum of the actual parameters. The characterizations of the MANFIS models are given in Table 2.

Table 2. The characterizations of the MANFIS models.

Zn	Mn	Fe	Cu	ANFIS parameter
Gbellmf	Gbellmf	Gbellmf	Gbellmf	Type of membership function
Linear	Linear	Linear	Linear	Output membership function
286	286	286	286	Number of nodes
500	500	500	500	Number of linear parameters
45	45	45	45	Number of nonlinear parameters
545	545	545	545	Total number of Parameters 63
				70 Data pairs
40	40	40	40	Number of training data
18	18	18	18	Number of checking data pairs
125	125	125	125	Number of fuzzy rules

In order to investigate the efficiency of the ANFIS models, the variance account for (VAF) (Eq. (14)) mean square error (MSE) (Eq. (17)), the root mean square error (RMSE) (Eq. (16)) with 12 datasets were used and R2 Eq. (17).

$$VAF = \left(1 - \frac{\text{var}(y - y')}{\text{var}(y)} \right) \tag{Eq. 14}$$

$$MSE = \frac{1}{n} \sum_{i=1}^n (y - y')^2 \tag{Eq. 15}$$

$$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^n (y_i - y')^2} \tag{Eq. 16}$$

$$R^2 = 1 - \frac{\sum_{i=1}^n (y_i - y')^2}{\sum_{i=1}^n y_i^2 - \frac{(\sum_{i=1}^n y_i')^2}{N}} \tag{Eq. 17}$$

Where, y and y' are the predicted and measured values, and N refers to the number of samples.

An MSE of zero means that the estimator has predicted the values of the parameter with perfect accuracy, and therefore it is ideal, but never practically possible. The more the VAF, the better the model performance. For example, a VAF of 100% denotes that the measured output has been predicted perfectly (exact model). If VAF is equal to zero, it means that the model performs very poorly and cannot be used

as a predictor employing simply the mean value generated by the data. In addition, a lower RMSE shows that the model enjoys a better performance. The determination coefficient (R^2) was calculated, as well. Fig. 3 depicts the correlation between the predicted and measured values of the deformation modulus for ANFIS models.

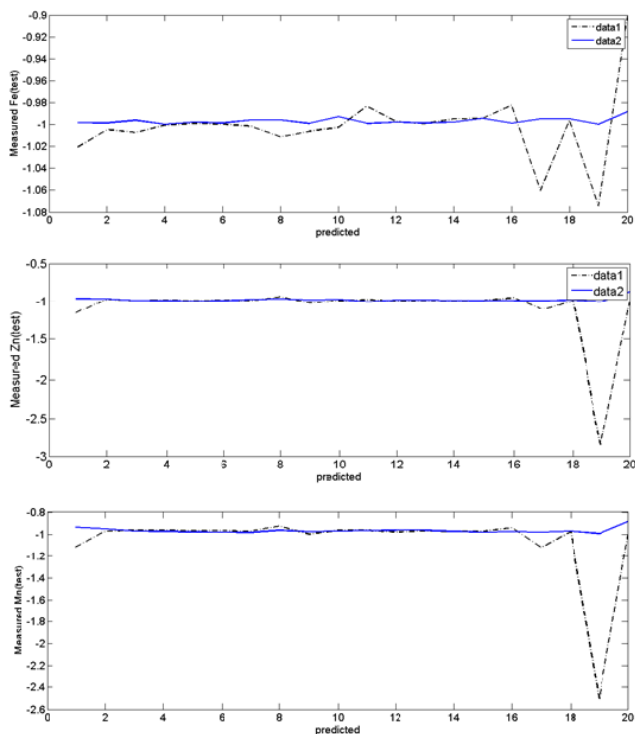


Fig. 3. Correlation between predicted values and measured ones related to the deformation modulus ANFIS.

Fig. 3 depicts the resulting work of ANFIS in the test dataset. As Fig. 3 indicates, there is a reasonable agreement between the measured and predicted values. A comparison between the results of ANFIS models is shown in Table 3.

Table 3. Comparison between the results of the models.

MSE	VAF	RMSE	ANFIS model
0.3079	99.9983	0.0055	ANFIS(CU)
0.1020	99.9999	0.0010	ANFIS(Fe)
0.4031	99.9975	0.0063	ANFIS(Mn)
0.1639	99.9990	0.0040	ANFIS(Zn)

As it can be deduced from Table 3, the ANFIS model with VAF=99.9983, MSE = 0.3079 and RMSE= 0.0055 for Cu and VAF=99.9999, MSE = 0.1020 and RMSE =0.0010 for Fe and VAF=99.9975, MSE = 0.4031 and RMSE = 0.0063 for Mn and VAF=99.9990, MSE = 0.1639 and RMSE = 0.0040 for Cu outperforms other models for modeling the concentration of heavy metals.

5. Conclusion

In order to predict the presence of heavy metals in the Shur river originating from acid mine drainage (AMD), a new ANFIS method was proposed in this study. ANFIS is believed to be a valuable tool for predicting the presence of heavy metals. In addition, when the relation function is unknown or complicated, this model proves to be a powerful tool to connect input variables to output ones to produce random values for each variable. In this study, we investigated the presence of polluting heavy metals in the waste dumps of the Sarcheshmeh Copper Complex (SCC). The SCC is characterized with high Mg and sulfate concentrations as well as a neutral pH concentration. We implemented the ANFIS method to predict the Cu, Zn, Fe, and Mn concentrations from the SCC resulting from AMD using four outputs. The pH, Mg, and

SO_4^{2-} concentrations were considered as the input layer. The obtained results indicated that because of the high ability of ANFIS, it was implemented as a useful method against expensive and time-consuming laboratory-based methods. It could quickly and efficiently forecast Cu, Zn, Fe, and Mn concentrations by testing for pH, Mg, and SO_4^{2-} in the mine drainages, where the concentration of metals (Cu, Zn, Fe, and Mn) was low. The results of the current study provide important implications for environmental scientists since it can help deal with the existence of pollutant heavy metals in the environment, particularly running waters.

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