

## Estimation of grade and recovery in the concentration of barite tailings by the flotation using the MLR and ANN analyses

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**Abstract:** This study aimed to find optimal models in a comparative framework to estimate the recovery and grade of barite concentrate obtained from the rougher flotation of the barite tailings. Therefore, firstly, the effect of four operating parameters (flotation time, pH, collector dosage, and depressant dosage) on the rougher flotation of the barite tailings containing 37.23% BaSO<sub>4</sub> was experimentally investigated. Secondly, two models called the multivariable linear regression (*MLR*) and the artificial neural network (*ANN*) were used for the estimation of the recovery and grade of the barite concentrate for the rougher flotation optimization. The *R*<sup>2</sup> values found from the *MLR* and *ANN* models were 0.828 and 0.995 for the concentrate recovery, and 0.977 and 0.960 for the barite concentrate grade, respectively. In the comparison of the models determined, it was found that the *ANN* model expressed quite well than the *MLR* models, especially for the recovery of the rougher concentrate.

**Keywords:** barite, tailing, flotation, recovery, grade, *MLR* model, *ANN* model

### 1. Introduction

Barite (BaSO<sub>4</sub>) is the mineral with the highest density other than metal minerals, and for this reason, it finds wide use as drilling mud in oil drilling. On the other hand, it is used as a filler in paint, glass, and paper industries due to its white color. Moreover, barite minerals are used to view cancerous cells in colonoscopy due to their adsorption properties of X-rays and gamma rays (Deniz, 2012; Deniz & Guler, 2018).

Barite ores are generally evaluated in less than 10 gravity-based concentration plants (facilities established as jigs and shaking tables) in different regions of Turkey. During the barite concentration in these concentrator plants, an important amount of very fine barite minerals escapes in the tailing fraction.

In recent years, the reserves of high-quality barite deposits are decreasing and grinding energy costs are increasing; therefore, such gravity concentration plant tailings have started to gain economic importance due to their significant barite content and fine size. Additionally, this situation not only causes resource losses but also important environmental problems. For this reason, more researchers will need to focus on the concentration of barite minerals from gravity concentration plant tailings in terms of environmental protection and economic point.

Due to its low cost and high selectivity in the concentration process, gravity methods such as jigs for coarse barite ores and shaking tables for the concentration of fine barite ores are generally used. However, the very fine barite minerals (under 75 μm) can not be concentrated efficiently using these gravity methods. For this reason, the flotation method comes to the fore in the efficient evaluation of very fine barite slimes.

While the industrial scale flotation method has been used in the concentration of barite ores in the world for years, it has not been applied in Turkey until now. Also, since Turkey already has reserves of high-quality barite ore, barite tailings have been evaluated in only one barite plant so far, and that is the gravity-based multi-gravity separator (*MGS*) method (Deniz, 2015; 2021). Therefore, barite tailings have been generally stored in heaps in other concentration plants in Turkey. However, the barite particles

again escape to the tailing due to the low recovery and selectivity of the MGS method. In the coming years, it will be necessary to evaluate the barite tailings in terms of both the decrease in the quality of barite deposits in Turkey and environmental problems. Therefore, this study will make an important contribution to guiding the use of the flotation method for barite ores in the coming years.

Barite is evaluated in the group of non-metallic minerals by flotation. The flotation foundations of non-metallic minerals were first introduced by Aplan & Fuerstenau (1962) and then studied by the flotation of oxides by Hanna & Somasundaran (1976) and Smith & Akhtar (1976).

Barite is often found together with gangue minerals such as quartz and calcite. Although due to the differences in composition, structure, and properties between barite and quartz, it is relatively easy to separate them by flotation with fatty acids and their salts, whereas calcite and barite minerals show similar surface properties, selective flotation is difficult (Marinakakis & Shergold, 1985; Ren et al., 2017). Therefore, barite flotation with fatty acids is almost impossible without a depressant from a barite ore containing calcite minerals (Feng et al., 2015). In addition to them, Bulatovic (2015) showed that  $\text{Ca}(\text{OH})_2$  gave better results than  $\text{NaOH}$  as a pH regulator on barite recovery.

In barite flotation, sodium oleate is widely used as a collector in a highly alkaline pulp, but it is susceptible to slime and hard water ions (Gurpinar et al., 2004; Chen et al., 2019; Lu et al., 2020). Also, Bhatti et al. (2017) reported that a concentrate grade of over 91% could be obtained from a barite ore using sodium oleate with sodium silicate.

The mechanism of adsorbing alkali sulfate and sulfonates to the surface of barite was investigated by Andrew & Collings (1989) and showed that the collector adsorbed specific (chemically) to the cation in the barite crystal in the stern layer.  $\text{Ba}^{2+}$  cations are the potential determining ion (*pdi*) for barite and  $\text{BaCl}_2$  increases the efficiency of barite flotation (Schubert & Schneider, 1970). Barite is suppressed with depressants such as  $\text{Na}_2\text{SiO}_3$ , Tannic acid, and Quebracho to clean from impurities such as quartz and calcite, at  $\text{pH} = 8-11$ . In the literature, low-grade barite ores have been effectively concentrated by flotation using collectors such as alkyl sulfate, petroleum sulfonate, and their mixture (Martinez et al., 1975; Lamont & Sullivan, 1982; Harris, 1988; Lenzo & Sarquis, 1995; Guimaraes & Peres, 1998; Hadjiev et al., 2000; Bulatovic, 2015; Kecir & Kecir, 2016).

Barite flotation is generally carried out using fatty acids and their salts or sulfate/sulfonate type collectors as mentioned above. Since the fatty acids also float the calcite mineral, they give a lower grade concentrate than the sulfonates. Meanwhile, fatty acids are difficult to remove after the flotation process is complete. Therefore, barite cannot be used in many industries such as drilling mud because barite cannot be made hydrophilic with water. It has been found that the long-chain organic sulfates used in barite flotation are excellent collector materials and are also easily separated from the concentrated mineral (Gullett et al., 1964). However, the sulfate-type collectors are quite expensive compared to other collectors such as Na-oleate and fatty acid.

Consequently, considering the importance of collector adsorption in barite flotation, flotation experiments should be performed with collectors that will separate the barite mineral from the gangue mineral more selectively and more effectively.

The measure of success in flotation experiments is determined by the percentage values of the recovery and grade of the concentrate obtained. Recovery and grade determination in ore processing is a routine practice for any ore processing plant. Therefore, estimating the recovery and grade of the barite concentrate using mathematical methods based on the variation of various operating parameters can help the concentration plants operate optimally. Different statistical methodologies such as multivariable linear regression (*MLR*), multivariable nonlinear regression (*MNLR*), and artificial neural network (*ANN*) models can be applied to estimate the grade and recovery of the rougher barite concentrate. Among these, *MLR* has been the most preferred one because it can be easily used by the mineral engineers in the concentrate plant (Deniz, 2020; 2021).

An artificial neural network (*ANN*) is quite similar to *MNLR*, and it can use a larger number of the model parameters and the multivariable response data, and therefore it is preferred in modeling. Although there are many types, the most used is multilayer feed-forward networks, which use the back-propagation learning algorithm. An *ANN* can learn complex nonlinear relationships using a training algorithm with a set of input and output data (Smith, 1996; Hagan et al., 1996; Umucu et al., 2016). *ANN* models have some advantages over traditional statistical models such as *MLR* and *MNLR*, and it is performed better than other models. *ANNs* have been used for optimization and estimation since the late 1980s, and have also started to be used as a preferred modeling method in mineral processing for

the last 10 years. Although the estimation of the recovery and grade of the concentrate obtained depending on different operating parameters with different concentration methods of different ores has been studied by some researchers using *ANN*, no one has been modeling on barite flotation. As the grades of barite ores decrease around the world, it is thought that more studies in the future will focus on barite concentration by flotation.

This study primarily aimed to obtain a barite concentrate with a minimum 90%  $\text{BaSO}_4$  requirement of the market by concentrating on a barite tailing by flotation. For this purpose, first, laboratory-scale flotation tests were carried out to determine the effects of some operating parameters on the rougher flotation of the barite tailings. Second, it was targeted to estimate the recovery and grade of barite concentrate by *ANN* and *MLR* models using a statistical software package (*SPSS 22.0*) depending on the selected operating parameters.

## 2. Materials and methods

### 2.1. Materials

Low-grade barite tailings used in this study were collected from the storage area belonging to Baser Mining I.C., Isparta, Turkey. Firstly, the contents of major oxide elements in the sample were determined by a Philips/Panalytical PW2400 model X-ray fluorescence (*XRF*) spectrometer, and the results are presented in Table 1.  $\text{BaSO}_4\%$  grade was also determined as 37.23% by wet method solubilisation and weight loss methods. As seen in Table 1, the  $\text{SiO}_2$  value of 33.22% comes not only from the quartz mineral but also from the chlorite, sericite, and orthoclase minerals. Also, the  $\text{SO}_3$  value comes not only from  $\text{BaSO}_4$  but also from other sulfide-containing minerals.

Table 1. Chemical compositions of the barite tailings by *XRF*

Components (%)	$\text{SiO}_2$	$\text{CaO}$	$\text{Al}_2\text{O}_3$	$\text{Fe}_2\text{O}_3$	$\text{MgO}$	$\text{Na}_2\text{O}$	$\text{K}_2\text{O}$	$\text{SO}_3$	LOI*
	33.22	8.07	7.37	1.48	0.63	1.37	1.89	44.63	0.35

\*LOI: Loss on ignition

Then the mineralogical analysis of the tailing sample was carried out with a Jeol JSDX-100S4 model X-ray diffractometer (*XRD*). According to the *XRD* result of the barite tailing seen in Fig. 1(a), orthoclase, quartz, calcite, chlorite, and sericite clay minerals were encountered in addition to the barite mineral. Then, in order to determine how the barite tailings are distributed in the sieve size fractions according to both their weight ratios and  $\text{BaSO}_4$  grade, the sample was wet screened, and the cumulative undersize distributions and  $\text{BaSO}_4$  grade changes as a particle size are shown in Fig. 1(b). As seen in Fig. 1(b) that the  $\text{BaSO}_4$  grade increases with decreasing particle size. These results indicate that barite minerals are more brittle than gangue minerals (calcite, quartz, orthoclase, etc.). However, since clay minerals such as chlorite and sericite are much more easily brittle than barite minerals, they were collected at the lowest sieve size ( $\sim 38 \mu\text{m}$ ), hence reducing the barite grade.

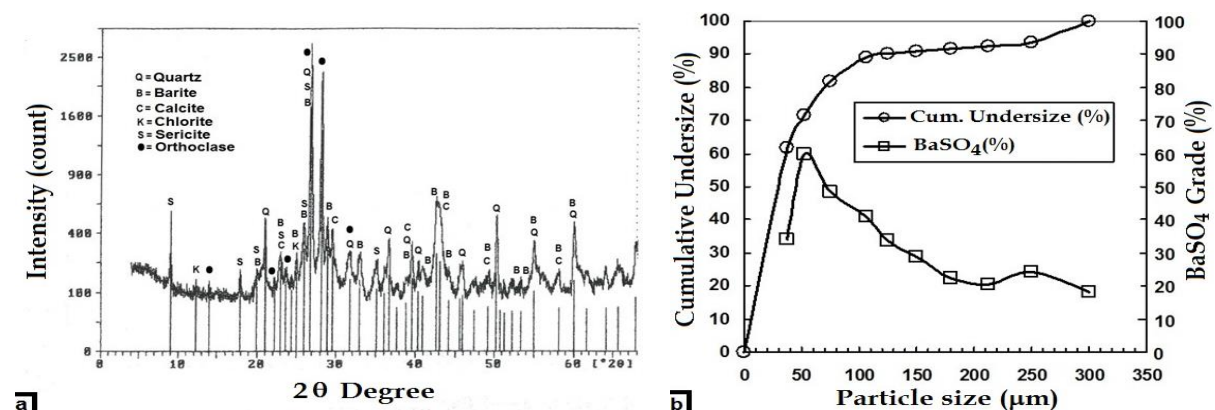


Fig. 1. Result of (a) the *XRD* pattern and (b) variation of the cumulative undersize distribution and  $\text{BaSO}_4$  as a function of particle size of the barite tailing

## 2.2. Methods

While Na-Oleate is one of the salts of oleic acid, the A845 is a petroleum sulphonate produced by Cytec. Sulfosuccinamate collectors such as A845 manufactured by CYTEC are alkylated succinate acids with a sulphonate group attached between the groups. A845 was developed to be similar to oleates, but more selective than oleates (Cytec Industries, 2004). In addition, the short conditioning times with A845 have favored the best flotation.

In this study, firstly, the performance of collectors called Na-oleate from oleic acid salts and A845 from sulfonate collectors were tested to determine the effect of collector type on the barite flotation. While determining the effect of the collector type, other operating conditions such as the collector dosage of 800 g/Mg, the pulp pH 9 with CaO, the depressant ( $\text{Na}_2\text{SiO}_3$ ) amount of 1000 g/Mg, the frother (pine-oil) dosage of 150 g/Mg, the pulp solid ratio of 20%, the agitation speed of 1500 rpm, and the conditioning and flotation times of 3 min were kept constant.

After determining the appropriate collector type, the laboratory scale flotation experiments in a Denver flotation cell were carried out in 5 steps. First, the barite tailings were dispersed in suspension at a 20% solid pulp ratio using the mineral processing plant's own tap water. Second, the pulp was conditioned for 5 min by adjusting the pH with CaO and monitoring by a pH meter. Third, a depressant ( $\text{Na}_2\text{SiO}_3$ ) of 500 to 3000 g/Mg was added to the suspension for 5 min conditioning. Fourth, froth was done with a sulfonate collector (A845) of 200 to 1000 g/Mg with 5 min of conditioning followed by a coarser flotation time of 3 min. Fifth, in the three-stage cleaning of the rougher concentrate, only CaO was added as a pH regulator without A845 in all three cleaner stages. Finally, the barite tailings were combined with the new rougher feed for the cleaner flotation, and steps second through fourth are repeated.

Then, a statistical experimental program was started to reveal the effect of some operating parameters on the flotation recovery and grade of the barite concentrate obtained from the experiments as a result of rougher flotation. This experimental program was prepared with four different process variables (pH, collector dosage, depressant dosage, and flotation time) by keeping all other operating conditions constant. The ranges of these operating parameters are given in Table 2.

Table 2. Values applied in the rougher flotation experiments for four different operating variables

Experimental No	Operating variables			
	pH	Collector dosage (g/Mg)	Depressant dosage (g/Mg)	Flotation time (min)
	$X_1$	$X_2$	$X_3$	$X_4$
1	8	800	1000	3
2	9	800	1000	3
3	10	800	1000	3
4	11	800	1000	3
5	8	200	1000	3
6	8	400	1000	3
7	8	600	1000	3
8	8	800	1000	3
9	8	1000	1000	3
10	8	400	500	3
11	8	400	1000	3
12	8	400	1500	3
13	8	400	2000	3
14	8	400	2500	3
15	8	400	3000	3
16	8	400	1500	1
17	8	400	1500	2
18	8	400	1500	3
19	8	400	1500	4

Recovery and grade percentage (as a response to rougher flotation) as a function of these four explanatory variables were investigated by *MLR* and *ANN* modeling techniques using the *SPSS 22.0* package program. The estimation equations of the recovery and grade of barite concentrate with *MLR* and *ANN* modeling techniques were examined by considering the  $R^2$ , *MSE*, and *RMSE* for the significance of the relationship.

### 3. Results and discussion

#### 3.1. Rougher flotation experiments

In the rougher flotation experiment, firstly, the performance of two different collector types, Na-oleate and A845 was investigated.

The experimental conditions were kept constant for both collector types, i.e., the pulp solids ratio of 20%, the pH= 9,  $\text{Na}_2\text{SiO}_3$  dosages of 1000 g/Mg, and collector dosages of 800 g/Mg, pine oil of 160 g/Mg, and the conditioning and flotation times of 5 min. In barite flotation, the A845 collector gave better results than Na-oleate, similar to other researchers. It was determined from the results that a rougher concentrate containing 48.37%  $\text{BaSO}_4$  with 93.83% recovery could be obtained with the use of A845 as the collector. This result turned out to be much better than Na-oleate, which provided a rougher concentrate containing 47%  $\text{BaSO}_4$  with 79% recovery. It was determined that both recovery and selectivity were high in sulfonate flotation of barite ore in an alkaline pulp. Also,  $\text{Na}_2\text{SiO}_3$ , which is widely used as a depressant of calcite minerals, had a significant effect on the selectivity of the concentrate.

One of the most important parameters in the success of flotation is the pH value of the pulp. For this reason, the flotation experiments were carried out on the pH value first, and the pH value was changed from 8 to 11 for the 5-min conditioning time. The results indicated the best result at pH 9. As the pH increased above 9, it also showed a significant decrease in both grade and recovery of the barite concentrate. However, pH 8, which is the natural pH of pulp, was accepted as the optimum pH value since there was no problem in terms of both recovery and barite grade of the concentrate.

Secondly, it was aimed to determine the optimum A845 dosage, keeping other operating conditions constant. It was observed that optimum barite flotation occurred at 400 g/Mg collector dosage as a result of changing the A845 dosage from 200 to 1000 g/Mg. The results of the tests showed that while there was a decrease in the  $\text{BaSO}_4$  content of the concentrate with the increase in the collector dosage, the maximum barite recovery was achieved at the 400 g/Mg A845 dosage, followed by a slight decrease in the barite recovery.

Third, the depressant ( $\text{Na}_2\text{SiO}_3$ ) dosage was varied between 500 and 3000 g/Mg keeping other conditions constant, and the optimum depressant dosage of 1000 g/Mg produced the best result. While there was a significant increase in the  $\text{BaSO}_4$  content with the increase in depressant dosage, the maximum barite recovery was achieved at the 1000 g/Mg  $\text{Na}_2\text{SiO}_3$  dosage, followed by a slight decrease in the barite recovery. A similar situation was stated by Bulatovic (2015) that sodium silicate acts as an important dispersant during barite flotation, and dosages higher than  $\text{Na}_2\text{SiO}_3$  of 1500 g/Mg significantly increase the barite recovery.

Fourth, the effect of flotation time on concentrate grade and recovery was investigated, keeping other conditions constant. The experiments were carried out for 1-min intervals from 1 min to 4 min to determine optimum flotation time, and the flotation time of 3 min gave the best result. It was observed that the flotation recovery increased as the flotation time increased, but the  $\text{BaSO}_4$  content decreased.

According to the rougher flotation results, the maximum recovery and grade of a rougher barite concentrate could be obtained by flotation from the barite tailings containing 37.23%  $\text{BaSO}_4$  were found to be 98.26% and 53.41%, respectively. Using the results from the three-stage cleaner flotation experiments, the recovery and grade of the final barite concentrate were simulated using a procedure similar to that described for the locked-cycle tests and could be produced a cleaner concentrate containing 94.27%  $\text{BaSO}_4$  with a recovery of 91.63%.

As a result of this study, it was revealed that the flotation method was more effective in terms of selective than MGS, a gravity method, previously used by Deniz (2021) for a similar barite tailing. Moreover, the consumption of collectors and depressants was very high due to the presence of calcite in the barite tailing. In future studies, it is planned to work on different collector and depressant types and quantities in terms of economically.

### 3.2. MLR model analysis

The relationship between one dependent variable and one or more independent variables can be investigated using multivariable linear (MLR) or nonlinear regression (MNL) analyses. MLR Models express the value of an estimated dependent variable as a linear function of one or more independent variables, while MNL Models express it as a nonlinear function. A choice between the two regression models is determined by the strength of the relationship to be obtained. In addition, if there is no problem in the strength of the relationship, MLR models, which are easier to understand and reveal by many researchers, are preferred. The basic assumption of the MLR is that the association between  $Y$  and the  $\beta_n$  vector of regression  $X_i$  is linear. Eq. 1 given below represents the MLR model (Kutner et al., 2004; Zhou & Li, 2011; Nakhaei et al., 2012).

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_4 X_4 + \dots + \beta_n X_n \quad (1)$$

where  $Y$  is the dependent variable (recovery or grade);  $X_1, X_2, \dots, X_n$  are the independent variables;  $\beta_0$  is the constant, where the regression line intercepts the  $Y$ -axis, and  $\beta_1, \beta_2, \dots, \beta_n$  are regression coefficients that express how much the dependent variable changes when the independent variables change by 1 unit (Kutner et al., 2004; Zhou & Li, 2011).

The purpose of MLR analysis is to estimate a dependent variable with more than one independent variable. On the other hand, since MLR analysis involves very complex calculations, it can only be done using computer software that uses statistical methods. A specific set of operating variables is required to determine the grade and recovery of the concentrate with an MLR analysis. Therefore, in this study, the four independent variables (flotation time, pH, collector, and depressant dosages) and one output dependent variable (*Recovery* or *Grade*) values were used to generate the statistical relationships. In this study, the IBM SPSS 22 statistical software package, a computer simulation programming that applies the least-square method, was used to generate the multivariable linear regression (MLR) functions. In both regression functions, *Recovery* (%) and *Grade* (BaSO<sub>4</sub>, %) of the barite concentrate were introduced as dependent variables, and  $X_1$  (pH value),  $X_2$  (collector dosage, g/Mg),  $X_3$  (depressant dosage, g/Mg), and  $X_4$  (flotation time, minute) as independent variables.  $\beta_n$  coefficients were estimated from the experimental results using the SPSS program. As a result, we also uncovered the statistical relationship between the same four variables and the recovery or grade variable. The regression equations that can most reliably estimate the recovery and grade of the barite concentrate with MLR models are given in Eqs. 2 and 3:

$$\text{Recovery} = 66.861 - (1.999 \cdot X_1) + (0.006 \cdot X_2) + (0.003 \cdot X_3) + (11.713 \cdot X_4) \quad R^2 = 0.828 \quad (2)$$

$$\text{Grade} = 92.145 - (1.135 \cdot X_1) - (0.001 \cdot X_2) + (0.003 \cdot X_3) - (12.250 \cdot X_4) \quad R^2 = 0.977 \quad (3)$$

The relationships between the experimental results and the estimated values using these equations are shown in Fig. 2 as the output of the IBM SPSS 22 package program. As a result, it was found that the grade and recovery of the concentrate obtained as a result of the rougher flotation of the barite tailings was in a good relationship between the experimental values and the estimated values.

Whether the regression equation explains a statistically significant part of the variance is determined by SPSS ANOVA and coefficients tables. In order to evaluate the performance of the developed equations, the root mean square error (RMSE),  $R^2$ , and  $F$  value are the most important factors that determine how well the obtained regression models fit the experimental data. While the RMSE is a type of error-index, and the closest and lowest value to 0 indicates the validity of the model, the  $R^2$  value lies between 0 and 1, and a value of 1 represents fully matched data. Also, the  $F$  value is a test based on  $F$ -test used to determine the significance of an  $R^2$ . The  $F$  value tells whether the model is statistically significant. The higher the  $F$  value, the more significant the model (Hair et al., 1998; Paulson, 2007).

In addition, the variance increases factor (VIF), tolerance ( $T$ ), and Durbin Watson ( $D-W$ ) factors are the statistical tool that determines whether there is autocorrelation between data. It is known that if each VIF value is less than 10.0 and the mean VIF value is less than 3.0, it means that the obtained regression equations are much more reliable and the independent variables are independent of each other. The same is true for tolerance ( $T$ ) values, meaning that the variables are independent of each other for values greater than 0.10. The  $T$  values approaching zero means that each estimated independent variable is highly estimated with other estimated independent variables (Hair et al., 1998; Paulson, 2007).

The  $D-W$  value ranges from 0 to 4, and when  $D-W = 2$  it means that there is no autocorrelation. The absolute values of  $\beta$  (beta) indicate the order of importance of each independent variable that affects the

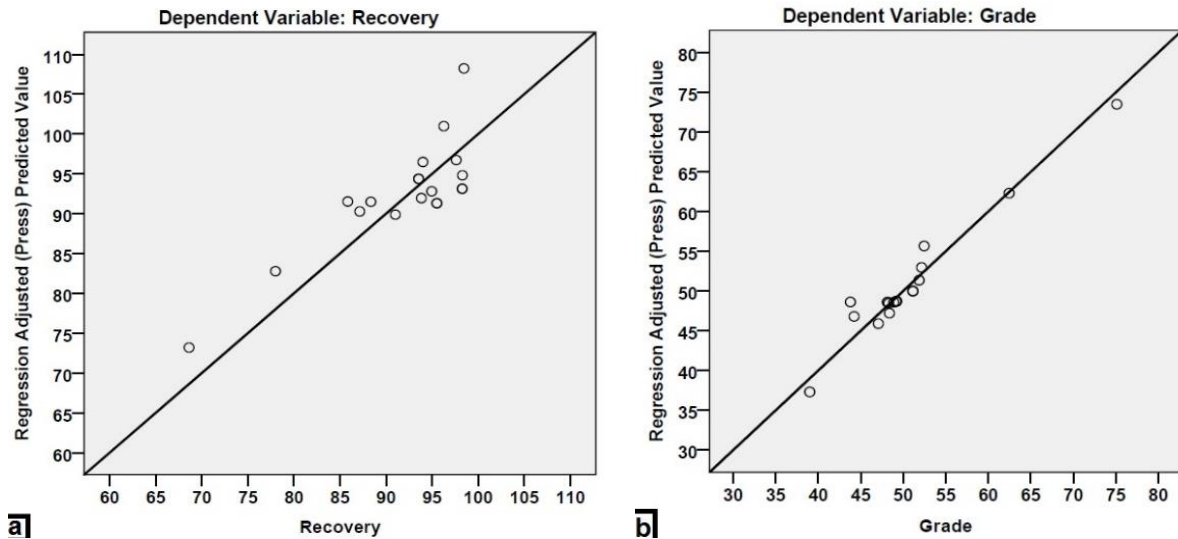


Fig. 2. Relationships between experimental and estimated values for recovery (a) and grade (b) of the concentrate

dependent variable. The greater the absolute value of a  $\beta$  (beta) value, it means that it is the independent variable that is in the relationship that most affects the dependent variable (Hair et al., 1998; Paulson, 2007). In this study, these statistical factors were used to control the data from the model equations with the experimental data.

The ANOVA for two equations is given in Tables 3 and 4.  $R^2$  values between the values estimated and the experimental values of the recovery and grade of the barite concentrate were 0.828 and 0.977, respectively. Also, the  $F$  values for the recovery and grade of the barite concentrate were 16.818 and 147.23 ( $p = 0.000$  or less than 0.0005), respectively, highly significant for both equations, especially for the  $\text{BaSO}_4\%$  grade. Therefore, it is easy to see that each of the variables added to all independent variables is significant at a  $p$ -value of 0.05 (5%) at the 95% confidence level. Additionally, the root mean squared error (RMSE) values for estimation of the recovery and grade of the barite concentrate were 2.169 and 0.440, respectively. The statistical factors given in Tables 3 and 4 showed a strong correlation between the experimental and estimated data.

As shown in Tables 3 and 4, the Durbin-Watson (D-W) factors for grade and recovery of the barite concentrate were found close to 2, being 1.884 and 1.945, respectively, so it can be said that there is no autocorrelation between the data. It also showed that there was no multicollinearity between the four independent variables, as the  $VIF$  values were considerably smaller than 3.0 and the  $T$  values considerably higher than 0.10 for both recovery and grade of the barite concentrate. Moreover, the fact that all  $T$  and  $VIF$  values were close to 1.00 showing that the independent variables were not affected by each other. Therefore, there is no cause for concern for either equation obtained.

The absolute value of  $\beta$  shows the order of importance of the independent variables among the dependent variables. When the  $\beta$  values from Tables 3 and 4 are examined to determine the contributions of the independent variables in the models, it was determined that the flotation time ( $X_4$ ) made the greatest contribution to the barite concentrate with both  $\beta = 0.860$  for the recovery and  $\beta = -0.925$  for the grade. As a result, there was a significant relationship between recovery and grade of the barite concentrate and dependent variables (flotation time, pH, collector, and depressant dosages).

Table 3. The statistical factors for recovery

	$B$	Std. Error	$\beta$	Tolerance ( $T$ )	$VIF$	$F$	D-W	$R^2$	RMSE
Const.	66.861	10.482							
$X_1$	-1.999	1.195	-0.212	0.764	1.310				
$X_2$	0.006	0.005	0.171	0.691	1.448	16.818	1.945	0.828	2.169
$X_3$	0.003	0.002	0.267	0.859	1.164				
$X_4$	11.713	1.522	0.860	0.985	1.015				



Table 4. The statistical factors for grade

	<i>B</i>	Std. Error	$\beta$	Tolerance ( <i>T</i> )	<i>VIF</i>	<i>F</i>	<i>D-W</i>	<i>R</i> <sup>2</sup>	<i>RMSE</i>
Const.	92.145	3.742							
<i>X</i> <sub>1</sub>	-1.135	0.427	-0.124	0.764	1.310				
<i>X</i> <sub>2</sub>	-0.001	0.002	-0.018	0.691	1.448	147.230	1.884	0.977	0.440
<i>X</i> <sub>3</sub>	0.003	0.001	0.214	0.859	1.164				
<i>X</i> <sub>4</sub>	-12.250	0.543	-0.925	0.985	1.015				

In addition, the three-dimension (3D) plots are the graphical representations of the regression equation and the type of interactions between the variables to deduce the optimum conditions. Barite grades (Fig. 3) and recoveries (Fig. 4) of the barite flotation concentrates were studied as a function of the interaction between the pH (*X*<sub>1</sub>), the collector dosage (*X*<sub>2</sub>), the depressant dosage (*X*<sub>3</sub>), and the flotation time (*X*<sub>4</sub>), respectively. While one dependent variable (*Recovery* or *Grade*) was kept constant in each graph, the interaction of 2 of the 4 dependent variables was examined.

Fig. 3 shows the three-dimension (3D) relationship between barite recovery and the binary of other independent variables (*X*<sub>1</sub>, *X*<sub>2</sub>, *X*<sub>3</sub>, and *X*<sub>4</sub>). The highest barite recovery can be achieved with a higher collector dosage (*X*<sub>2</sub>), higher depressant dosage (*X*<sub>3</sub>), and higher flotation time (*X*<sub>4</sub>) at pH=8 (*X*<sub>1</sub>). It is obvious from Figs. 3a-3f that the increase in flotation time had a particularly great effect on the barite recovery. In addition, Fig. 3d shows the 3D relationship between barite recovery with collector dosage (*X*<sub>2</sub>) and depressant dosage (*X*<sub>3</sub>), and it can be concluded that the depressant dosage is not a determinate factor for the barite recovery.

Fig. 4 shows the 3D relationship between BaSO<sub>4</sub>% grade and the binary of other independent variables (*X*<sub>1</sub>, *X*<sub>2</sub>, *X*<sub>3</sub>, and *X*<sub>4</sub>). It is clear that the grade of barite concentrate is most affected when the flotation time (*X*<sub>4</sub>) changes, but is least affected by the depressant dosage.

### 3.3. ANN model analysis

Artificial neural networks (ANNs), are similar to multivariate nonlinear regression (MNL) analyses and are widely used to optimize complex nonlinear processes and estimate the output of desired results with the given input (experimental data) (Smith, 1996). An ANN can learn complex nonlinear relationships using a training algorithm with a set of input and output data, and then it can model the relationship. ANNs have consisted of interconnected neurons that might have several inputs, hidden, and output layers working sequentially and parallel. When input data (*X*<sub>*n*</sub>) are introduced into the neural network, they are multiplied by the synaptic weights (*W*<sub>*i,n*</sub>) between neurons, and these neurons propagate between the layers and generate the output data (*Y*<sub>*n*</sub>) with the help of an activation function.

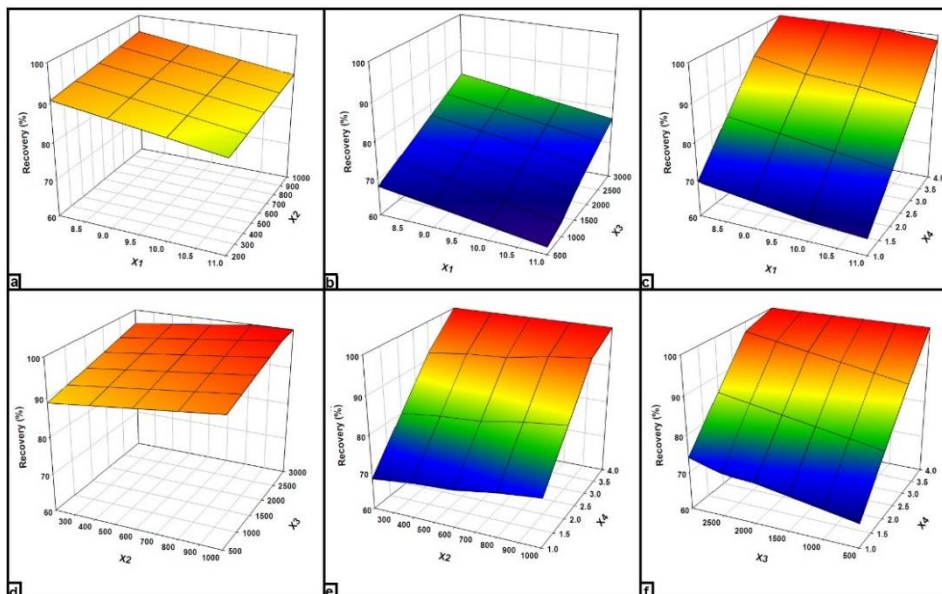


Fig. 3. Changes in the flotation recovery of barite concentrate depend on some operating parameters



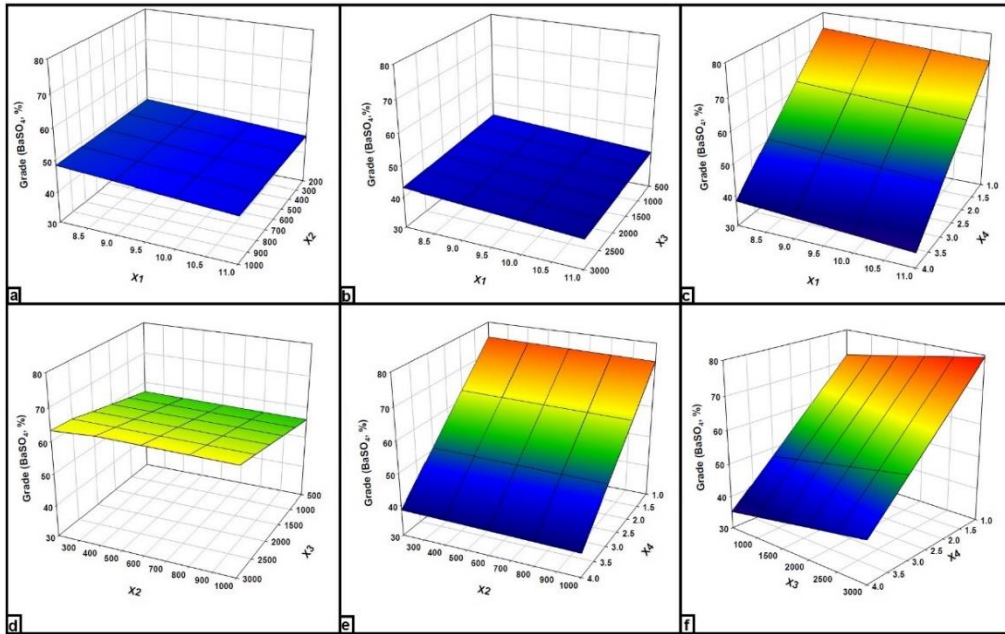


Fig. 4. Changes in the grade of barite concentrate depend on some operating parameters

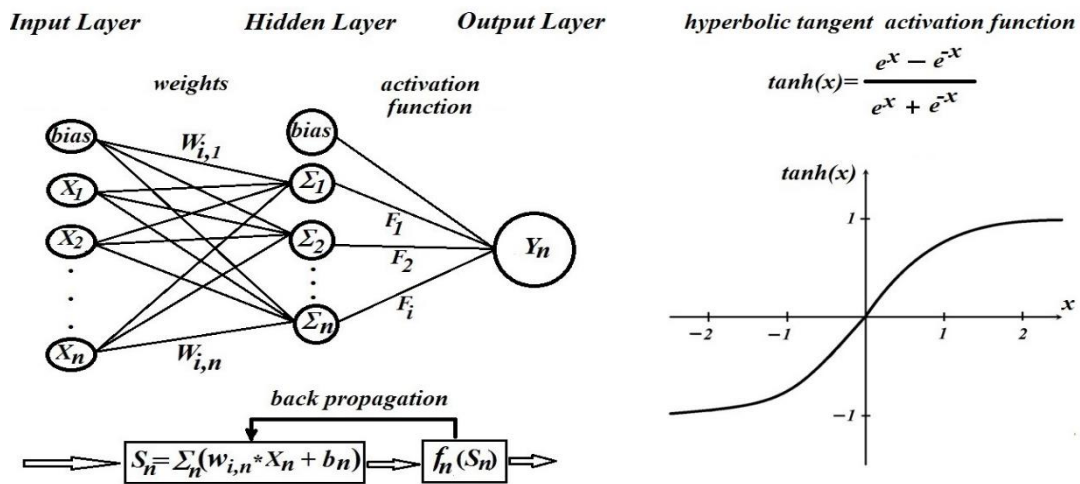


Fig. 5. The architecture of an ANN model structure and hyperbolic tangent activation function

When training this input data, they are iterated until the most reliable empirical relationship is established between the input and output data. ANN provides learning by minimizing the errors of the outputs by adjusting the weights (Zhu et al., 1999; Umucu et al., 2016). A multilayer perceptron network, working with feed-forward logic, is the most popular network architecture for ANN. Fig. 5 shows the system structure of a feed-forward ANN and a hyperbolic tangent activation function.

In this study, a back propagation ANN was developed using the Neural Network function in SPSS version 22.0. The ANN architecture included two hidden layers and a multilayer perceptron network with back propagation architecture. However, an algorithm was not used to determine the number of hidden neurons in the network. An ANN model usually has three layers: input layers, hidden layers consisting of interconnected processing units namely neurons, and output layers. The first is the input layer, and in this study, it was formed from four source points such as pH, collector dosage, depressant dosage, and flotation time. The second layer was the hidden layer of nonlinear processing units and consisted of two points. Finally, the output layer was run according to the *Hyperbolic Tangent*, an activation function used in the input layer. However, a function was not used in the output layer. In this study, since the training set is relatively small, it was performed 10-fold-cross-validation to avoid problems, and the final accuracy obtained from the average of 10 tests, each using a different test set, is considered. 70% of the data is set to be assigned to the training stage and 30% of the data to the testing stage. In this study, it was automatically set to reach the best ANN architecture. The automatic

architecture finds the best number of neurons in hidden layers and uses the default activation functions. As a result of the tests, the ANN models with one input layer of 4 input variables, one hidden layer of 2 neurons, and one output layer of one output variable (4-2-1 structure) (as seen in Fig. 6) were selected according to the highest coefficient of determination ( $R^2$ ), the mean square error ( $MSE$ ) and the lowest root mean square error ( $RMSE$ ) to estimate the recovery and grade of the barite concentrate. Since the SPSS automatically put the estimators in order of their importance, no formula was used. The statistical parameters of the best ANN models for the estimation of the recovery and grade of the barite concentrate are presented in Table 5.

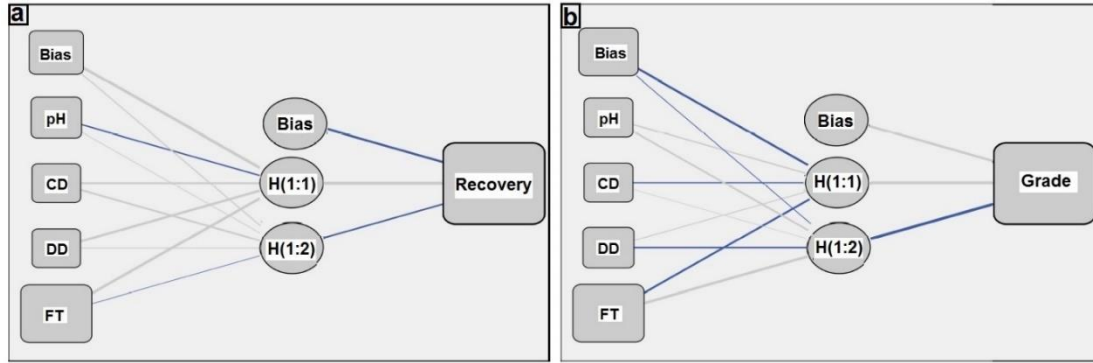


Fig. 6. The architecture of ANN models used to determine the recovery (a) and grade (b) of the barite concentrate

In this study, the hyperbolic tangent function with an output range of  $[-1, 1]$  is used as the activation function and is defined in Eq. 4. In addition, the  $R^2$ ,  $MSE$ , and  $RMSE$  equations given in Eqs. 5, 6, and 7 were used to evaluate the effectiveness of the selected models.

$$\tanh(x) = \frac{e^x - e^{-x}}{e^x + e^{-x}} \quad (4)$$

$$R^2 = 1 - \frac{\sum(y_i - \hat{y}_i)^2}{\sum(y_i - \bar{y})^2} \quad (5)$$

$$MSE = \frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{n} \quad (6)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{n}} \quad (7)$$

where  $y_i$  and  $\hat{y}_i$  are the experimental values and the estimated values, respectively.  $\bar{y}$  is the mean of the experimental values  $y_i$ , and  $n$  is the number of experiments.

To analyze the accuracy of the ANN models, the estimated values of the recovery and grade of the barite concentrate were plotted against the experimental values, as seen in Fig. 7. The  $R^2$  values obtained by the ANN model for recovery and grade were 0.995 and 0.960, respectively, it is seen that the models have a very realistic approach. The estimation error values for the recovery and grade of the barite concentrate were 0.514 and 0.080 for the  $RMSE$  and 0.265 and 0.006 for the  $MSE$ , respectively. It was determined from these results that the ANN models can estimate both the recovery and grade of the barite concentrate with very smaller errors and very high accuracy.

According to the normalized importance values obtained by the ANN model, it was observed that the flotation time ( $X_4$ ) had a great effect of 100% on both the recovery and grade of the barite concentrate. In addition, SPSS results revealed two hidden neurons that had significant effects on the recovery and grade of barite concentrate. It was found that the H (1:1), a hidden neuron, has the most significant effect on both recovery and grade according to the hidden neurons' weights.

### 3.4. Comparison of the MLR and ANN models

Fig. 8 shows a comparison of the ANN and MLR estimated values for each of the 19 experimental values of recovery and grade of barite concentrate. The results also showed that both the MLR and ANN models were able to estimate the  $BaSO_4$  grade very well within the acceptance limit. However, the estimated values of the MLR were significantly lower accuracy according to all experimental data on the recovery of the barite concentrate. On the other hand, the ANN model with the least  $RMSE$  and the highest  $R^2$  values showed that it was much more suitable than the MLR model in estimating the recovery of the barite concentrate.

As seen from Table 5 that when the estimation ability of the ANN and MLR models are compared,  $R^2$  values higher than 0.80 indicate that there are relations with acceptable accuracy for the two models. RMSE and MSE values showed that ANN gives almost better estimates with smaller errors than MLR. Therefore, it appears possible to use both models, although the ANN model is a more reliable tool for estimating the recovery and grade of the barite concentrate. In addition, the estimation errors of the MLR model for recovery of barite concentrate were acceptable due to the complexity of the flotation process.

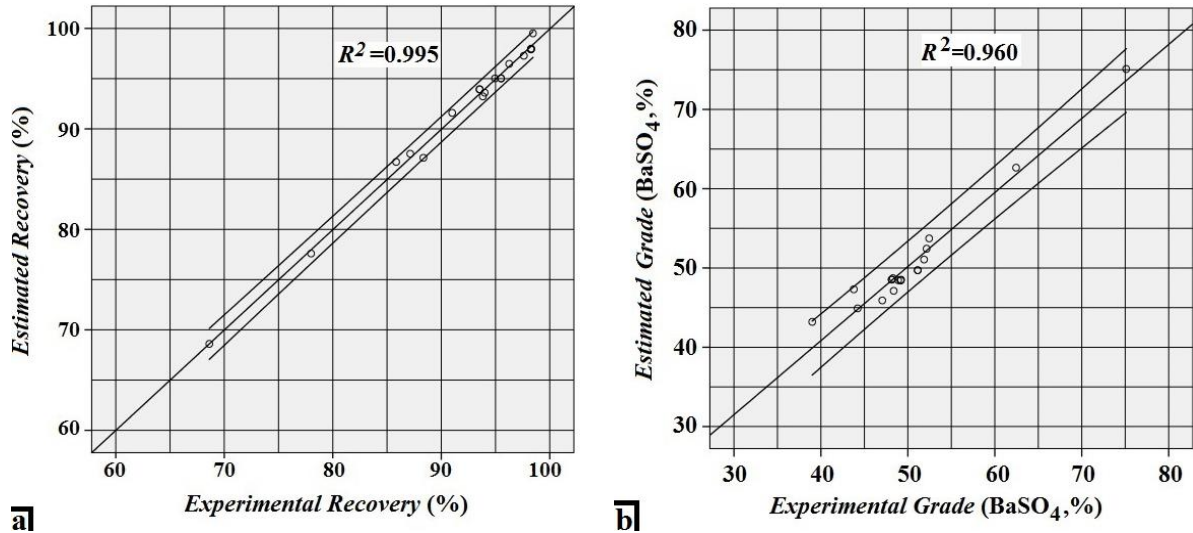


Fig. 7. Comparison of the ANN estimated values and the experimental values of the recovery (a) and grade (b)

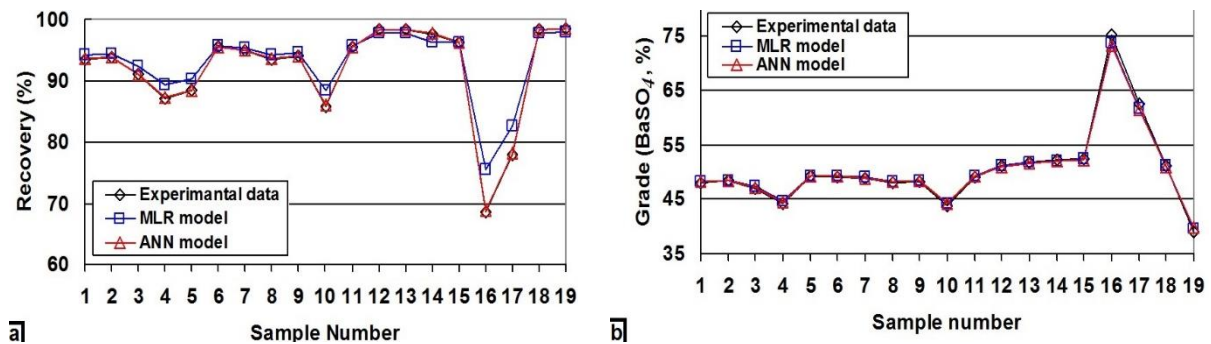


Fig. 8. Comparison plots of the estimated values by ANN and MLR with values of experimental recovery (a) and grade (b) of the barite concentrate

Table 5. Statistical parameters of the MLR and ANN models

Model Types	Grade			Recovery		
	$R^2$	RMSE	MSE	$R^2$	RMSE	MSE
MLR	0.977	0.440	0.194	0.828	2.169	4.704
ANN	0.960	0.514	0.265	0.995	0.080	0.006

#### 4. Conclusions

In order to control the process in a mineral processing plant and to understand the behavior of the operating variables in the process, there is a need to create equations that can estimate the grade and recovery values to be obtained. These model equations allow us to take preventive measures for possible undesirable situations in the process and to avoid unnecessary spending and squandering. Creating a mathematical model for flotation processes can help optimize the process, thus saving time and effort spent performing experiments.

In this study, it was proposed a numerical application of the flotation process optimization model based on the recovery and grade of the barite concentrate at the rougher flotation on the barite tailings using multivariable linear regression (MLR) and artificial neural network (ANN) models. Therefore, recovery and grade ( $\text{BaSO}_4$ , %) of the barite concentrate were introduced as dependent variables, and  $X_1$  (the pH value),  $X_2$  (the collector dosage, g/Mg),  $X_3$  (the depressant dosage, g/Mg), and  $X_4$  (the flotation time, minute) as independent variables.

The  $R^2$  values of MLR models developed to estimate the recovery and grade of the barite concentrate of the rougher flotation were found to be 0.828 and 0.977, respectively, while the  $R^2$  values of ANN models were found to be 0.995 and 0.960, respectively. It shows that the ANN models describe much better than the MLR models. In addition, according to each two models, the flotation time ( $X_4$ ) had a major effect on both the recovery and grade. The concentration mechanism is difficult to understand because of the complexity of physicochemical adsorption in flotation processes, hence, it is difficult to model compared to other physical concentration methods (shaking table, magnetic separator, etc.). Therefore, it is an important result that the model equations in this study were presented with very high validity.

As a result of this study, more operating parameters such as solid content, frother dosage, and conditioning times can be added to the grade-recovery models obtained with MLR and ANN to increase the barite concentrate to a  $\text{BaSO}_4$  grade above 94%. Additionally, similar studies should be carried out for cleaner and scavenger flotation circuits for obtaining cleaner concentrate in the concentration of the barite tailings.

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