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Probabilistic Prediction of Acid Mine Drainage Generation Risk Based on Pyrite Oxidation Process in Coal Washery Rejects - A Case Study

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Abstract

In this paper, we investigate a probabilistic approach in order to predict how acid mine drainage is generated within coal waste particles in NE Iran. For this, a database is built based on the previous studies that have investigated the pyrite oxidation process within the oldest abandoned pile during the last decade. According to the available data, the remaining pyrite fraction is considered as the output data, while the depth of the waste, concentration of bicarbonate, and oxygen fraction are the input parameters. Then the best probability distribution functions are determined on each one of the input parameters based on a Monte Carlo simulation. Also the best relationships between the input data and the output data are presented regarding the statistical regression analyses. Afterward, the best probability distribution functions of the input parameters are inserted into the linear statistical relationships to find the probability distribution function of the output data. The results obtained reveal that the values of the remaining pyrite fraction are between 0.764% and 1.811% at a probability level of 90%. Moreover, the sensitivity analysis carried out by applying the tornado diagram shows that the pile depth has, by far, the most critical factors affecting the pyrite remaining.

1. Introduction

The pyrite oxidation process commonly results from the coal, metal waste, and tailing particles. Some of the critical factors such as O_2, Fe^{3+} ions, temperature, pH, Eh, and presence or absence of bacteria, which can affect this process, have been studied in the comprehensive studies for several decades. It is due to their role in mining environmental issues, especially acid mine drainage (AMD). AMD is generated when the pyrite or sulfidic minerals are exposed to atmospheric weathering. This process is always too complicated due to the substantial chemical, biological, and physical parameters that can be involved in it. As a result, the prediction and investigation approaches of AMD are categorized as a challenging topic, time-consuming, costly, and they always differ in every case. In general, monitoring the procedures are firstly conducted applying the geochemical and geophysical techniques including the laboratory and field tests. Subsequently, the prediction procedures are conducted by the analytical and numerical methods with different solving techniques of the numerical models that are governed mathematically in the oxidation process. Although the numerical approaches are the deterministic models, they only allow the researchers to evaluate the oxidation process by considering the deterministic parameters involving the pyrite oxidation process in long-term periods (US. EPA 1994). Moreover, the critical problems in these models, which are categorized as the traditional methods, are related to the in-situ measurements and intensive data. Hence, it always takes a lot of budget and time to have an appropriate database with a sufficient accuracy (Betrie et al., 2013).

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Over the last three decades, significant investigations have been performed to study the pyrite oxidation process, which results from mining waste or tailings. For example, Cathles and Apps (1975) have suggested a 1D non-steady-state model for the leaching process in a copper waste dump. They assumed that the dump was composed of the rock particles containing the pyrite and chalcopyrite particles. Their model was based on oxygen, heat balance, and air convection. Jaynes et al. (1984 a, b) have presented the POLS model for simulation of the pyrite oxidation and leaching process from the reclaimed coal strip mines during a long-term period. Both oxygen diffusion and ferric iron were the pyrite oxidants in the model. Davis and Ritchie (1986) have developed a numerical model for pyrite oxidation within the White's overburden dump at the Rum Jungle, Australia. Elberling et al. (1994) have supposed that oxygen diffusion is the dominant mechanism in the pyrite oxidation process, which results from tailings. They also applied the laboratory tests to evaluate the sulfide oxidation rate. Lefebvre and Gelinas (1995) have simulated AMD generation with their model, TOUGH AMD, within waste rocks. Some of the essential processes including hydrology, gas and heat transfer, geochemistry, and mass transport were involved in this model. Wunderly et al. (1996) have presented PYROX based on a finite element method (FEM), which is a coupled method of oxygen diffusion and sulfide mineral oxidation to simulate pyrite oxidation in the vadose zone of tailings in Nordic Main tailings impoundment near the Elliot Lake, Ontario. Mayer et al. (2001) have developed a 1D numerical of the multi-component reactive transport model (MIN3P) to simulate the evolution of pore water, pore gas, and mineralogical composition in mine waste. Fala et al. (2003) have used a numerical model to simulate the unsaturated flow within a waste pile according to the internal structure of the pile and grain size distribution of the waste particles. Molson et al. (2008) have developed a geochemical transport of AMD with a numerical solution based on a finite element discretization method called POLYMIN within the heterogeneous waste pile. Kleiv and Thornhil (2008) have predicted AMD neutralization in anoxic olivine drains. Their model was based on the kinetic rate expression for olivine dissolution and the sulfuric acid dissociation equilibrium. In 2008, Doulati Ardejani et al. detected AMD pollution, which resulted from a coal waste dump with a coupled method of geophysical techniques and a 1D FVM numerical method (Doulati Ardejani et al., 2008). Nevertheless, the roles of some of the most important parameters such as the values for the diffused oxygen in the waste particles and also the production values including the sulfate, HCO^{3-}, H^+, and ferrous and ferric irons werenot considered in the model. As a result, Jodeiri Shokri et al. (2016a) have developed the mathematical models of the pyrite oxidation process, presenting a 2D numerical simulation based on FVM. The mathematical models were firstly developed by adding appropriate boundary conditions, source and sink terms on non-linear equations including oxygen transportation, pyrite oxidation controlled by second-order kinetic rates, transporting the products based on multi-component advection-dispersion mechanism, and pH buffering. Moreover, for a better understanding of the pyrite oxidation process, in this case, a 3D geo-electrical inversed model was built by converting the results of the 2D resistivity measurements within the pile surface. Eventually, an innovative reclamation plan with supposing a capping process on the pile surface was suggested applying a 2D numerical modeling. Khosravi et al. (2017a) have presented a probability mapping of the distributions of some toxic elements including arsenic and chromium throughout the waste particles resulting from the Sarcheshemeh copper dump. For this purpose, they used a combination of the visible and near-infrared reflectance (VNIR) spectroscopy and geo-statistical analysis. In another research work in this case study, they found that VNIR spectroscopy could be applied for predicting the As concentration in the surrounding contaminated soils of the dump (Khosravi et al. b).

Despite the considerable research works in investigating the pyrite oxidation process through the waste and tailings, a literature review also revealed that no research work had not yet been presented to predict the process using a probabilistic method. Indeed, all the researchers have supposed that all the parameters involved in the pyrite oxidation process are deterministic. Hence, in this work, we evaluated the risk of the pyrite oxidation generation within a coal waste pile applying a probabilistic method.

2.1. Materials and Methods

2.1. Pile description

The Alborz Sharghi coal washing plant is located in the Semnan Province, NE of Iran

(Figure 1). The Alborz-Sharghi coal washing plant was established approximately 35 years ago; it produces 300,000 tons of washed coal annually. The coal extracted from the state mines including the Tazareh, Razi, and Tabas mines as well as some local private mines is washed in the plant (Jodeiri Shokri and Zare Naghadehi, 2018). There are several waste piles and two tailing impoundments nearby the plant. A previous investigation has shown that one of these piles, which is the oldest abandoned pile, has had the potential of AMD generation. The wastes had been dumped without considering any environmental concerns since 1999 (Jodeiri Shokri et al., 2016b). The wastes dumped with the pile are the result of the jig machine process in the plant. The waste pile is approximately 20 m in height, and covers an area of about 100 m × 150 m (Jodeiri Shokri et al., 2016a).

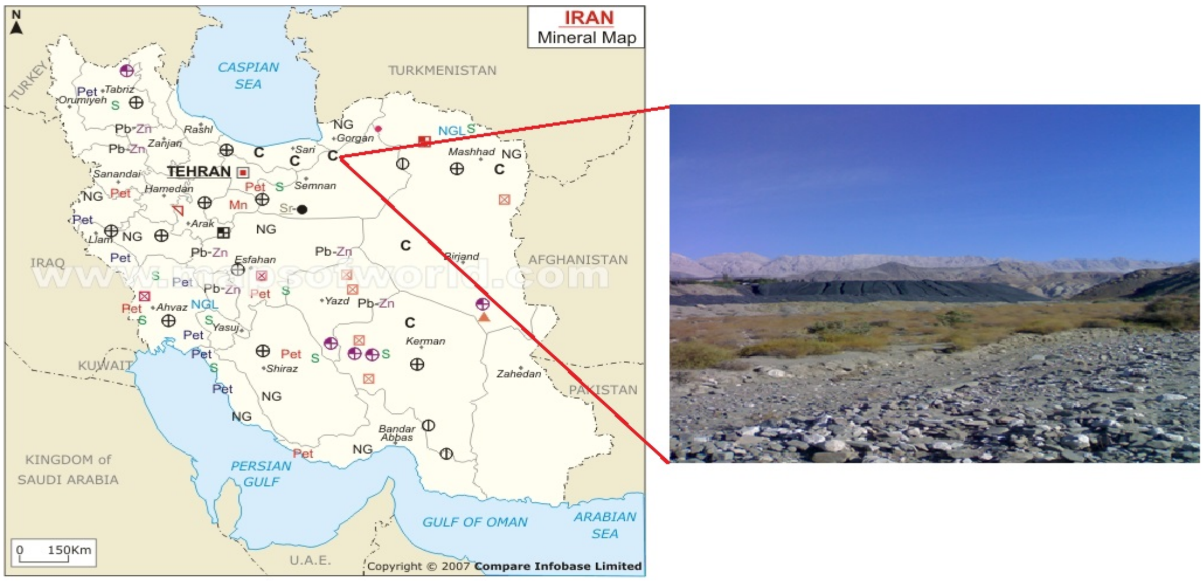

Figure 1. Geographical map of region and a view of pile

2.2. Methodology

As mentioned earlier, the AMD generation was predicted by a probabilistic method based on a Monte Carlo simulation. For this purpose, all the experimental data was collected from the pile in the previous studies such as Jodeiri Shokri *et al.* (2014 (a, b, c); 2016c), which was gathered to build a database. A brief database is presented in Table 1. Afterward, the input and output data was selected according to the available data. The depth of the waste within the pile, the fraction of diffused oxygen through the waste particles, and the concentration of bicarbonate within the pile as capability of AMD neutralization were considered as the input data. Moreover, the remaining pyrite fraction was selected as the output data. Then comprehensive statistical analyses were performed to find the best relationships between each output and all the inputs.

It should be mentioned that all the parameters involved in the pyrite oxidation process are supposed as uncertain. Therefore, it is necessary to describe their inherent uncertainties based on the probability distributions. Indeed, the risk for all parameters should be quantified by applying the distributions. In this research work, the @RISK software ver. 7 was employed for the description of data uncertainties in Excel worksheets. @RISK performs a risk analysis using a Monte Carlo simulation to show many possible outcomes in Microsoft Excel spreadsheet, and tells the user how likely they are to occur. This means that the user can judge which risks to take and which ones to avoid, allowing for the best decision-making under uncertainty (@Risk Manual, 2015).

It should be noted that several types of distributions such as Chi-Sq. Kolmogorov-Smirnov and Anderson-Darling are available in this software. Each one of these forms defines a series of possible values and the probability of occurrences (@RISK Manual, 2015).

After finding the best distribution functions for each input data with @RISK Ver. 7, a random sampling from the functions was done by a Monte Carlo simulation. Indeed, the risk analysis was executed by the Monte Carlo simulation. Different models of results are possibly built by applying a series of values, distribution functions, for those parameters, which have an inherent uncertainty in their nature. Then it repeats the calculation applying a different range of values from the functions that are taken randomly in each iteration. The number of uncertainties and the ranges specified can affect the simulation that may recalculate thousands or tens of thousands of times before the completion. The resulting outputs eventually from the simulation as the distribution functions (@RISK Manual, 2015). Finally, the risk analyses would be finished by finding the probability distributions functions of the outputs.

Table 1. Database of Alborz-Sharghi coal waste pile (Jodeiri Shokri *et al*. (2014(a, b, c); 2016)).

No.	Depth (m)	Fraction of diffused oxygen (%)	Concentration of bicarbonate (%)	Remaining pyrite fraction (%)	pH
1	0.00	0.21	4.00	0.52	4.80
2	0.10	0.19	3.80	0.57	4.75
3	0.20	0.17	3.60	0.63	4.70
4	0.30	0.16	3.45	0.70	4.62
5	0.40	0.14	3.20	0.79	4.55
6	0.50	0.13	3.00	0.86	4.50
7	0.60	0.11	2.80	0.97	4.42
8	0.70	0.10	2.63	1.06	4.30
9	0.80	0.09	2.45	1.17	4.25
10	0.90	0.07	2.40	1.25	4.20
11	1.00	0.05	2.50	1.33	4.15
12	1.10	0.04	2.90	1.38	4.30
13	1.20	0.03	3.50	1.44	4.45
14	1.30	0.02	4.15	1.47	4.65
15	1.40	0.014	4.80	1.54	4.80
16	1.50	0.005	5.50	1.58	5.00
17	1.60	0.002	5.85	1.60	5.10
18	1.70	0.00	5.86	1.62	5.20
19	1.80	0.00	6.45	1.63	5.25
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208	1.80	0.00	4.40	1.82	4.02
209	1.90	0.00	4.45	1.91	4.00
210	2.00	0.00	4.50	1.84	4.00

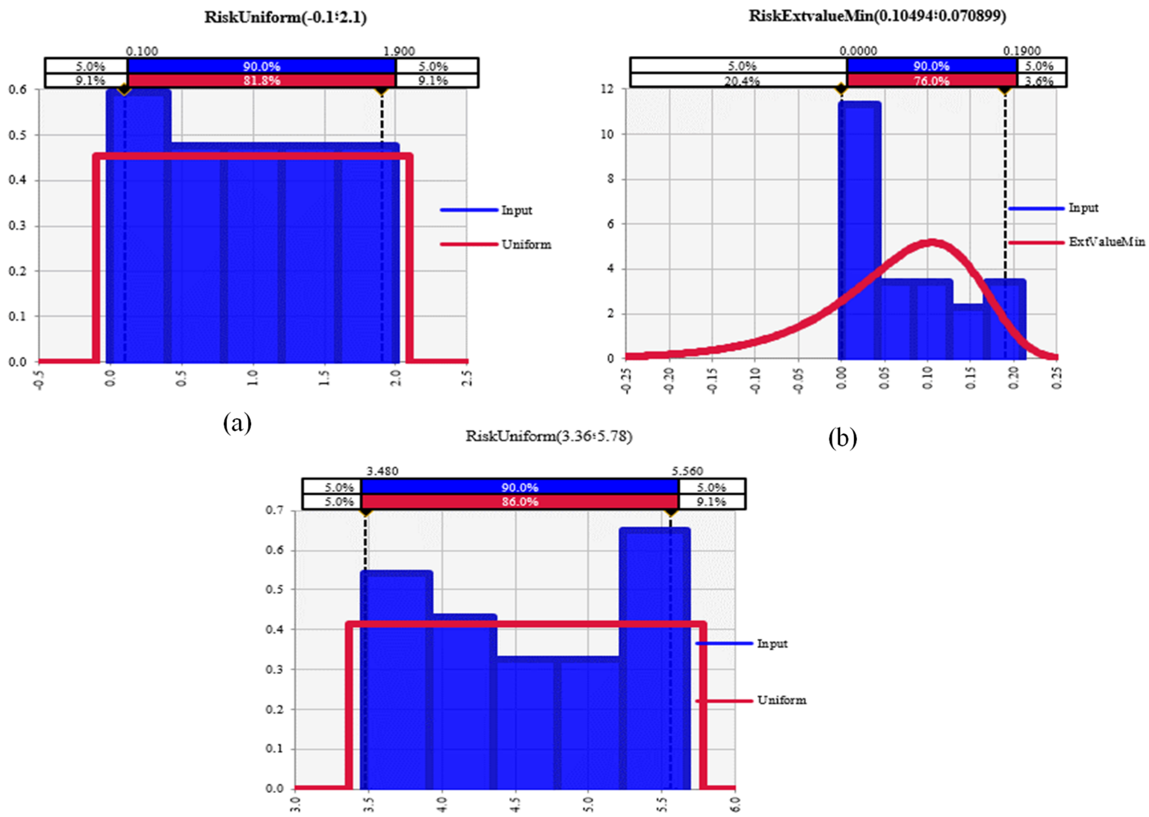
3. Results and Discussion

3.1. Probability distributions functions

The probability distribution functions were defined by applying @RISK, ver. 7, for each input. The best fittings of the distributions for each input including the pile depth, fraction of diffused oxygen, and bicarbonate concentration were obtained according to Akaike Information Criterion, Bayesian Information Criterion, Chi-Squared Statistics, Kolmogorov-Smirnov Statistics, and Anderson-Darling Statistics (Table 2 and Figure 2). Then the Chi-Squared Statistics was selected for further analyses.

Table 2. Best probability distributions for the input data.

Criterion	Depth	Fraction of diffused oxygen	Concentration of bicarbonate
AIC	Risk uniform (0.1;2.1)	Risk expon (0.069048) Risk shift (-0.003288)	Risk uniform (3.36;5.78)
BIC	Risk uniform (0.1;2.1)	Risk expon (0.069048) ;Risk shift (-0.003288)	Risk uniform (3.36;5.78)
Chi-Sq.	Risk uniform (0.1;2.1)	Risk extreme value Min (0.10494;0.070899)	Risk uniform (3.36;5.78)
K-S	Risk uniform (0.1; 2.1)	Risk logistic (0.062102;0.040455)	Risk uniform (3.36;5.78)
A-D	Risk uniform (-0.1;2.1)	Risk logistic (0.062102;0.040455)	Risk uniform (3.36;5.78)



(c)

Figure 2. Best probability distributions for input data; a) depth of the pile; b) Fraction of the diffused oxygen; c) Concentration of bicarbonate

3.2. Statistical analyses

3.2.1. Multiple linear regression

A regression model in which more than one regression variable is used is referred to as a multiple linear regression (MLR). In general, the response variable (the remaining pyrite fraction) may depend on the input data, i.e. the n variables (x). Equation (1) expresses an MLR with n regression variables (Shakeri *et al.* 2020):

$$C = \beta_0 + \beta_1 x_1 + \cdots + \beta_n x_n + \varepsilon \quad (1)$$

where:

C is a dependent variable (target variable);

x_i are the independent variables;

ε is the error of the model;

β = 0, 1, …, n, and β_j is considered as the regression coefficients.

The prediction process using this model resembles a super-plane in the n-dimensional space of the regression variables x_j. On the other hand, one may consider the prediction models of more complex structures (non-linear) than those expressed by Equation (1), for instance, in the following model:

$$C = \beta_0 + \beta_1 x_1 + \beta_2 x_2^3 + \beta_3 e^{x_3} + \beta_4 x_1 x_2 + \varepsilon \quad (2)$$

In order to simplify the analysis of the above equation, which is non-linear, one can simply substitute its variables with linear variables. Accordingly, taking $z_1 = x_1$, $z_2 = x_2^3$, $z_3 = e^{x_3}$, and $z_4 = x_1 x_2$, Equation (2) will take the following form (Shakeri *et al.* 2020):

$$C = \beta_0 + \beta_1 z_1 + \beta_2 z_2 + \beta_3 z_3 + \beta_4 z_4 + \varepsilon \quad (3)$$

3.2.2. Relationships between remaining pyrite fraction and all inputs

After describing the data, the best statistical relationship between each input and each output data including the remaining pyrite fraction has been suggested using the Table curve, v. 5.01, software, which is one of the most powerful statistical software available for curve and surface fitting the data. These relationships have been selected based on their R-squared coefficients. The best statistical relationships, 16, have been presented for both input data in Table 3. Indeed, this analysis provides an opportunity to consider the inter-relationship between the input and output data in the further proposed relationship. Also it should be noted that instead of the remaining pyrite fraction, the values of the squared root of the remaining pyrite (Sqrt (py)) have been considered due to being positive values for the remaining pyrite fraction.

Table 3. Relationships between the input variables and the remaining pyrite fraction.

Row	Relationship between the remaining pyrite and the input data	Parameters
1	$x_1 = Sqrt(d)$	$Sqrt(Py) \propto f(d)$
2	$x_2 = e^{-d}$	$Sqrt(Py) \propto f(d)$
3	$x_3 = e^{-o}$	$Sqrt(Py) \propto f(o)$
4	$x_4 = o^2$	$Sqrt(Py) \propto f(o)$
5	$x_5 = Sqrt(o)$	$Sqrt(Py) \propto f(o)$
6	$x_6 = bi^3$	$Sqrt(Py) \propto f(bi)$
7	$x_7 = e^{bi}$	$Sqrt(Py) \propto f(bi)$
8	$x_8 = bi^2 Lnbi$	$Sqrt(Py) \propto f(bi)$
9	$x_9 = \frac{o}{d}$	$Sqrt(Py) \propto f(d, o)$
10	$x_{10} = d \times Sqrt(o)$	$Sqrt(Py) \propto f(d, o)$
11	$x_{11} = d \times bi$	$Sqrt(Py) \propto f(d, bi)$
12	$x_{12} = e^{-d} + bi$	$Sqrt(Py) \propto f(d, bi)$
13	$x_{13} = d \times Ln(\text{bi})$	$Sqrt(Py) \propto f(d, bi)$
14	$x_{14} = bi \times o$	$Sqrt(Py) \propto f(o, bi)$
15	$x_{15} = o + \ln(bi)$	$Sqrt(Py) \propto f(o, bi)$
16	$x_{16} = d\ln(o)$	$Sqrt(Py) \propto f(o, d)$

3.2.3. Suggesting a statistical relationship for prediction of remaining pyrite fraction

After finding the best relationships between the input and the output data, each one of the relationships was considered as the independent variables, while the output was the remaining pyrite fraction. In order to find the best relationship for prediction of the remaining pyrite fraction, comprehensive statistical analyses were conducted by applying the IBM SPSS statistics software, ver. 25, based on the MLR method. For this, the back-propagation method was used in order to identify the best regression relationship among the selected relationships. Four relationships were taken after these analyses (Table 4). In order to achieve the best relationship, three statistical parameters were chosen as the best criteria: (1) R-square, (2) Adjusted R-square, and (3) Root mean squared error (RMSE). The principal goal of this process was to specify a statistical relationship exhibiting the maximum values of R-square and adjusted R-square, and the minimum value of RMSE. For this, all relationships were compared (see Table 3). Comparison of the statistical parameters of the relationships describe that relationship No. 4 is the best statistical relationship for predicting the remaining pyrite fraction. This relationship yields values of 0.922, 0.919, and 3.61 for the statistical parameters of R-square, Adjusted R-Square, and RMSE, respectively.

Table 4. Comparison of the statistical parameters of the models obtained using MLR.

Model	R Square	Adjusted R Square	RMSE
1	0.900	0.899	3.68
2	0.912	0.910	3.66
3	0.916	0.914	3.63
4	0.922	0.919	3.61

where:

Model 1: constant, x_2

Model 2: constant, x_2, x_9

Model 3: constant, x_2, x_7, x_9

Model 4: constant, x_2, x_4, x_7, x_9

Therefore, the best statistical relationship taken from MLR can be expressed as follows (Equation 4):

$$\text{Sqrt}(\text{Py}) = C_0 + C_1 x_2 + C_2 x_4 + C_3 x_7 + C_4 x_9 \quad (4)$$

where C_0 to C_4 denote the constants of the relationship with their values reported in Table 5.

Table 5. Values of the coefficients C_0 to C_7.

C_0	C_1	C_2	C_3	C_4
1.37	-0.55	-3.915	0.001	0.005

Eventually, the best statistical relationship for predicting the remaining pyrite fraction, which is a function of the depth, diffused oxygen fraction, and bicarbonate, can be expressed as follows (Equation 5):

$$\mathrm{Py} = (\mathrm{Sqrt}\ (\mathrm{Py}))^2 = (1.37 - 0.55e^{-d} - 3.915o^2 + 0.001e^{bi} + 0.005\frac{o}{d})^2 \quad (5)$$

where:

Py: Remaining pyrite fraction (%);
d: Pile depth (m);
o: Diffused oxygen (%);
bi: Concentration of bicarbonate (%).

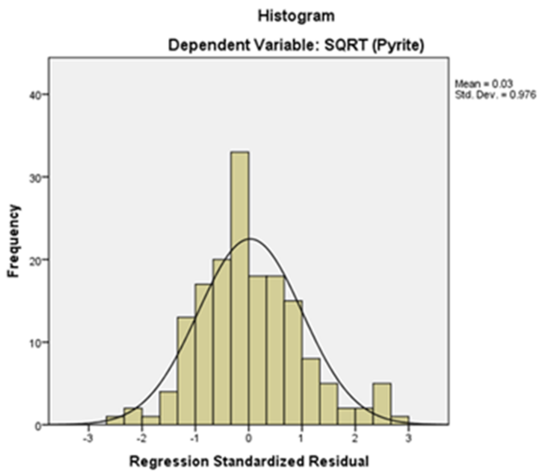


Figure 3. Histogram for the analysis of the modeling error

Figure 3 demonstrates a histogram for the analysis of the modeling error. The modeling error distribution function is a normal function, confirming that the regression test has been done correctly.

Also some data of the databases, 10%, were chosen as the validation data. Figure 4 shows the regression plot of the measured data and predicted data. As it can be seen, R-Squared is about 0.92, which shows that the MLR relationship can predict the remaining pyrite fraction successfully.

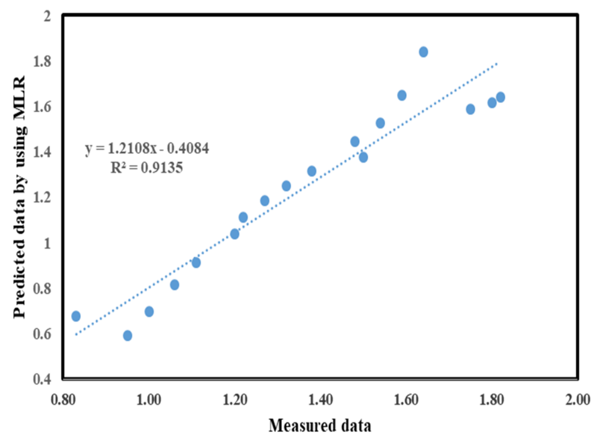


Figure 4. Linear regression plot of the output for the validation and measured data.

3.4. Predicting remaining pyrite fraction by applying probabilistic method

In the next step, the best probability distribution function of each input should be inserted in Equation (5) in order to estimate the remained pyrite. The probabilistic prediction of the remaining pyrite was simulated based on the Monte Carlo method. The number of the simulation iteration was 1000. Figure 5 and Table 6 show the meaningful occurrence probability of the events of the output data. The statistical parameters for the measured data are presented in Table 7.

As it could be seen in Tables 5 and 6, the results obtained from the simulation also corresponded to the field data very well. For instance, the meaningful occurrence probability of the simulation in 50% was 1.29, while the mean value of the measured data was 1.28. Moreover, the maximum value of the measured data was 2.08, while the meaningful occurrence probability of the simulation in 99% was 1.976, which revealed a reliable simulation.

Table 6. Meaningful occurrence probability of events of the remaining pyrite fraction.

Meaningful occurrence probability (%)	Values of the remaining pyrite fraction after simulation (%)
5	0.764
50	1.290
90	1.811
99	1.976

Table 7. Statistical parameters of the field data for the remaining pyrite fraction (%).

Statistical parameters	Values of the remaining pyrite fraction (%)
Mean	1.28
Median	1.34
Mode	1.63
Minimum	0.25
Maximum	2.08

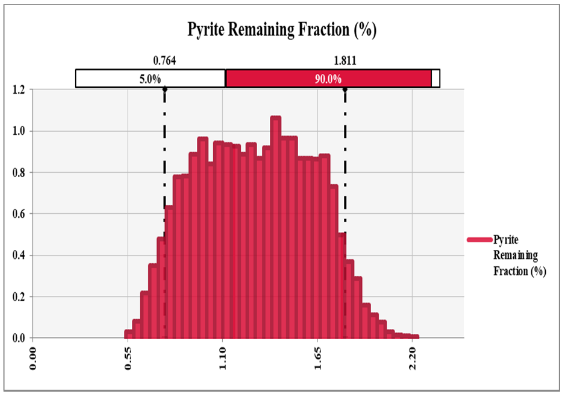


Figure 5. Meaningful occurrence probability of predicting the remaining pyrite fraction.

The histogram and the graph of the cumulative probability of the results are depicted in Figure 6. The results obtained show that the values of the remaining pyrite fraction are between 0.764% and 1.811% at a probability level of 90%. Also the values of the remaining pyrite fraction will be lower than 0.765 and 1.976 at the 5% and 99% probability levels, respectively. Figure 7 shows the best probability distribution function of the output of the Monte Carlo simulation. As it could be seen in this figure, the risk uniform function values (0.522, 2.246), which are 0.522 and 2.246, are the minimum and maximum values of the remaining pyrite fraction (%), respectively.

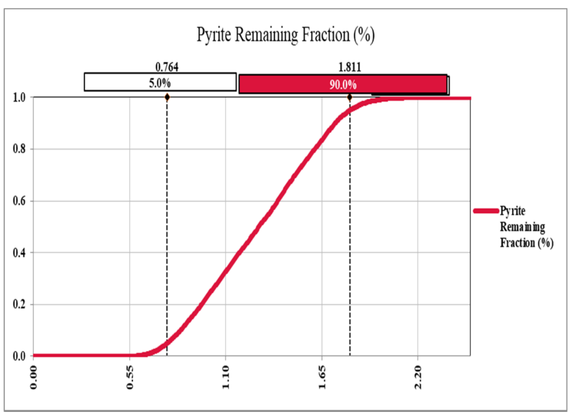


Figure 6. Comulative graph probability of predicting the remaining pyrite fraction.

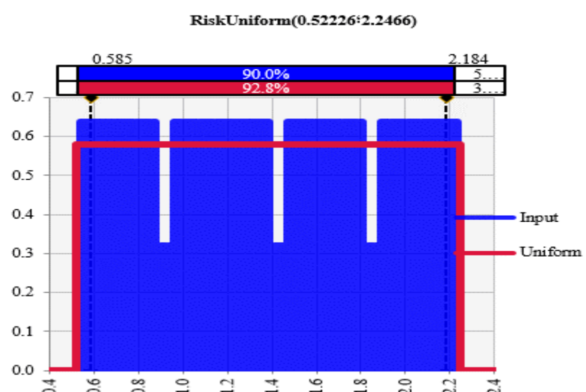


Figure 7. The best probability distribution function of the remaining pyrite fraction.

The tornado graphs show bars of each input, which affect the pyrite oxidation process. As it can be seen in Figure 8, the pile depth has the most important factor, which affects the pyrite remaining, while bicarbonate has the least effect on it.

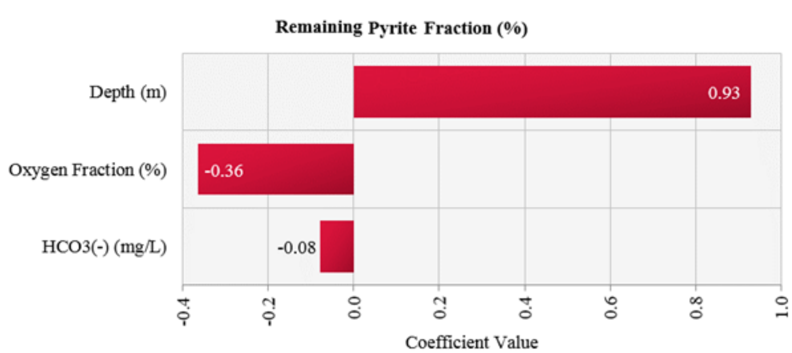


Figure 8. Tornado graphs for the remaining pyrite fraction.

4. Conclusions

In this research work, the pyrite oxidation process was investigated within the waste tailings based on the probabilistic method. For this, the remaining pyrite fraction and pH were considered as the output parameters, while the concentration of bicarbonate, depth of the waste, and oxygen fraction were the input data. The data was gathered based on the field data collected by the previous studies, which were done in the case study. After building a database, the best relationship between each output data and input data was determined by the linear regression method. The results obtained showed a high value of R-squared of 0.919 for the suggested statistical relationship. After finding the best relationship, the probability distributions functions were defined by applying @RISK, ver. 7, for the inputs. Then the distributions were inserted in the statistical relationships in order to find the probability distribution of the remaining pyrite fraction. The results obtained revealed that the remaining pyrite fraction was lower than 1.976% at a probability levelof 99%. Also the tornado diagram showed that the pile depth had the most critical effect on the remaining pyrite fraction, while the concentration of bicarbonate had the least effect on it.

5. Acknowledgment

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پیش‌بینی احتمالاتی ریسک تولید زهاب اسیدی معدن بر اساس فرآیند اکسیداسیون پیریت در بین ذرّات باطله‌های زغالی- مطالعه موردی

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چکیده:

در این مقاله، از یک روش احتمالاتی برای پیش‌بینی نحوه تولید زهاب اسیدی معدن در ذرّات باطله‌های زغالی در شمال شرقی ایران، استفاده شده است. برای نیل به این هدف، ابتدا، براساس پژوهش‌های قبلی انجام شده که در طول یک دهه گذشته در بررسی فرآیند اکسیداسیون پیریت در یکی از محل‌های متروکه انباشت باطله‌های زغالی انجام شده‌اند، پایگاه داده‌ای ساخته شده است. براساس نوع داده‌های دردسترس، میزان پیریت باقی‌مانده بعنوان داده خروجی در نظر گرفته شده است در حالی‌که، عمق قرارگیری باطله‌ها، غلظت بی‌کربنات و میزان فرکشن اکسیژن نفوذی در باطله‌ها، بعنوان داده‌های ورودی، فرض شده‌اند. سپس بهترین توابع توزیع، با استفاده از شبیه‌سازی مونت کارلو، برای هریک از این پارامترهای ورودی تعیین شده‌اند. همچنین، بهترین رابطه بین پارامترهای ورودی و خروجی براساس روش رگرسیون آماری ارائه شده‌است. در ادامه، بهترین توابع توزیع احتمالاتی هریک از پارامترهای ورودی، به رابطه خطی آماری وارد شدند تا با استفاده از آن، بتوان، بهترین تابع توزیع پارامتر خروجی را مشخص کرد. نتایج به‌دست آمده حاکی از آن است که میزان پیریت باقی‌مانده در سطح احتمال ۹۰ درصد، بین مقادیر ۰/۷۶۴ درصد و ۱/۸۱۱ درصد خواهد بود. از طرفی، تحلیل حساسیتی که با استفاده از نمودار تورنادو انجام شد، نشان می‌دهد که عمق قرارگیری باطله‌ها، اثرگذارترین فاکتور بحرانی بر میزان پیریت باقی‌مانده است.

کلمات کلیدی: زهاب اسیدی معدن، شبیه‌سازی مونت کارلو، تحلیل‌های آماری، باطله زغال.