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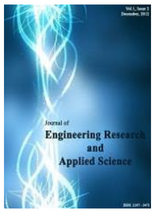

Classification of Pb recovery effecting factors with artificial ıntelligence based algorithms

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Abstract
Lead (Pb) is one of the most widely used non-ferrous materials. Many methods have been developed in order to produce the lead metal from initial raw materials or secondary resources. Process economy of the non-ferrous metal production is one of the major problems for the industry. In this study, an artificial neural network (ANN) based system was developed in order to provide detailed information regarding classification and estimation of Pb recovery effecting materials. The ANN structure was proposed with 4 inputs for predicting Pb recovery percentage. Four input parameters: temperature, NaCl concentration, time, and solid to liquid ratio values were selected based on physical considerations and the experimental test results. Analysis results showed that NaCl concentration and solid to liquid ratio are the most effective inputs on the recovery of Pb element from leach residue.

Keywords: Pb recovery; artificial neural network; conjugate gradient algorithm.

1. Introduction

Nowadays, zinc and lead are the most widely used base-metals after aluminum and copper among non-ferrous metals. Zinc is employed significantly in corrosion protection of iron-steels and in brass alloys due to its proper chemical, physical properties and process economy. On the other hand, lead with its low melting point, high formability and with its characteristically high density is preferred by many sectors [1,2]. Both metals could be produced by alternative methods depending on the initial raw materials. Although lead production relies on pyrometallurgical treatments such as BF, ISP or QSL, sulfide type zinc ores is generally treatment by roasting-leaching-electrowinning sequence. However, in Turkey, due to the complex nature of the existing ores, oxidized type zinc and lead ores is processed by Waelz – leaching – electrowinning sequence at Çinkur zinc plant. The leach residues of the plant have been deposited in stockpile area since 1980s, so huge amount of leach residue is accumulated. According to the researchers [2-4], metallic content of the residues is so high that it has an eloquently high economical potential. In addition, the environmental studies of these residues also indicated that weathering of such rich base-metal containing wastes at open air was causing a critically high pollution risk.

Cinkur leach residues containing high ratio of zinc and lead have already been an interest to both metallurgists and mineral engineers from the first production of them. All researchers have focused to increase the recovery zinc and lead from this type of residue by taking into consideration process economy. They tried several applications such as physical and chemical enrichment methods and processes to recover metallic zinc and/or lead from the beginning of the operations of the plant [2-8]. Among the researches carried out on Çinkur leach residues, physical concentration studies such as flotation tests at various pH; concentration by slime tables and hydrocyclone methods did not yield good results [2]. Beneficiation and treatment methods of lead and zinc by pyro or hydrometallurgical route commonly separated the metals and individually dealt with each other. It is generally accepted that zinc is recovered from the residues by hot sulphuric acid leaching [1]. The remaining secondary residue is found rich in terms of lead content as $PbSO_4$, so it should be converted into metallurgical treatable compounds like $PbCl_2$, $PbCO_3$, $Pb(OH)_2$, PbS [9-11]. A number of these studies on leach residue indicate that several parameters such as reagent concentration, temperature, Solid/Liquid ratio, duration etc. affect the recovery of lead, but none of them has clearly demonstrated the effect of the parameters on the leaching efficiency of lead.

Therefore, in the present study, optimization of the

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crucial parameters (concentration of solvent, temperature, reaction duration, solid/liquid ratio) affecting the leaching efficiency of the lead from waste, and significant levels of the leaching parameters are statistically investigated.

2. Material and method

2.1. Used materials

Representative sample of the leach residue was obtained from plant's disposal area, where they have been kept under uncontrolled atmospheric conditions for long years. Therefore, it was assumed that open surface had been weathered so samples were taken from deeper portions of the residue heaps as mentioned previous studies [2,6]. Bulk density, specific gravity and particle size analyses were applied to the leach residue sample. Results of the tests indicated that the residue had 0.79 g/cm^3 bulk density with 4.38 and 3.93 specific gravity values which were measured by water and air pycnometers, respectively. The discrepancy in these two values was thought to be due to the dissolution of water-soluble components in the residue during measurement. Then, the physical characterization was concluded by particle size analysis given in Figure 1.

According to the wet sieve analysis of the sample, as seen in Figure 1, about 76% of the residue was under 100 µm. Moreover, calculations showed that the residue dissolved in water approximately by 8.4%. Chemical analysis of the sample for Zn, Pb and Fe contents was performed by Perkin-Elmer Model 2380 Atomic Absorption Spectrophotometer, and obtained as 12.59%, 15.21% and 6.45%, respectively. Researchers studied on chemical and physical characterization of the residues in detail have reported that the main phases in the bulk were $PbSO_4$ and $CaSO_4.0.5H_2O$. In addition, zinc containing phases were claimed to be hydrous sulphates ($ZnSO_4.7H_2O$) and partially ferritic or silicate bearing complexes like $ZnO.Fe_2O_3$, $2ZnO.SiO_2$.

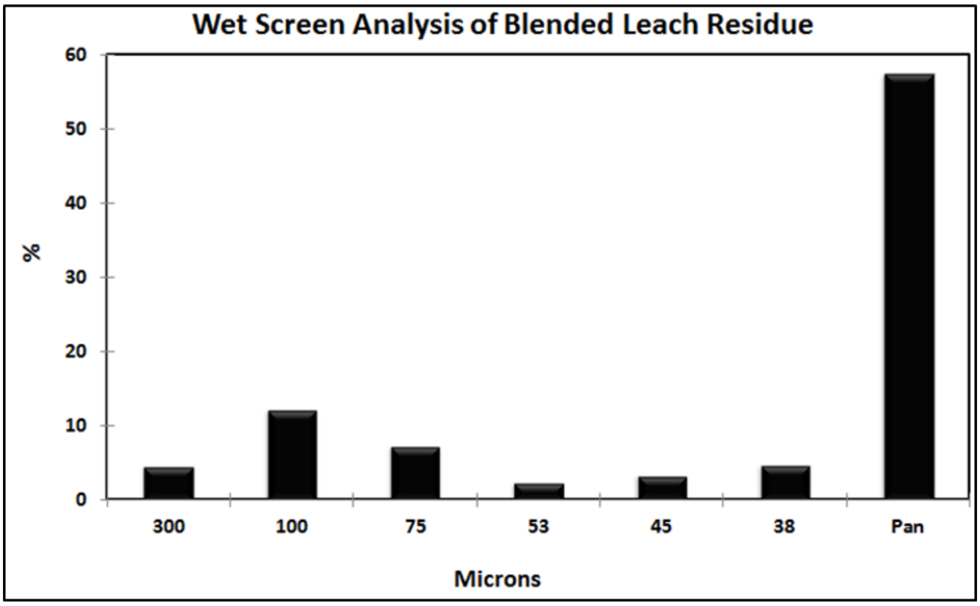

Figure 1. Wet Screen Analysis of the Leach Residue.

2.2. Hydrometallurgical experiments

In this study, inputs used to analyze statistically the lead recoveries were based on previous study [2], these recovery ratios were obtained from the residue after hydrometallurgical brine leach process. In this research, leaching trials were performed in de-ionized water with brine solutions in a borosilicate glass reaction flask to which magnetic stirring and heat were applied. Experimental variables were chosen to be temperature, dissolution time, solvent (NaCl) concentration and solid to liquid ratio.

Metal recoveries, as weight percent were calculated by using the following formulae, as the analyses were done on remaining solids (secondary leach residue):

$$\%\Re = \left(1-\frac{M_{SLR}*W_{SLR}}{M_{LR}*W_{LR}}\right)*100$$

from undissolved solid where M_{LR} and M_{SLR} are the analyzed metal contents of primary and secondary leach residues in wt %, respectively and W_{LR}, W_{SLR} are the weights of initial feed and undissolved portion of leach residue [2].

2.3. Neural network development

The neural networks represent the predictive model. In Neural Designer neural networks allow deep architectures, which are a class of universal approximator. Conjugate gradient method was utilized in this research. In the conjugate gradient algorithm search is performed along conjugate directions, which produces generally faster convergence than gradient descent directions. Training algorithm is presented in Table 1.

Table 1. Training algorithm.

Description		Value
Training direction method		Value
Training rate method		FR
Training rate tolerance		Brent Method
Min. parameters increment form	Norm of the parameters increment vector at which training stops.	0.0005
Min. loss increase	Minimum loss improvement between two successive iterations.	1e-009
Gradient norm goal		1e-012
Max. iterations number		0.001
Maximum time	Maximum training time.	1000

The ANN structure was proposed with 4 inputs for predicting Pb recovery percentage. Four input parameters: temperature, NaCl concentration, time, and solid to liquid ratio values were selected based on physical considerations and the experimental test results. The proposed ANN structure is given in Figure 2. Basic statistics are very valuable information when designing a model, since they might alert to the presence of spurious data. It is a must to check for the correctness of the most important statistical measures of every single variable. Table 2 shows the minimums, maximums, means and standard deviations of all the variables in the data set.

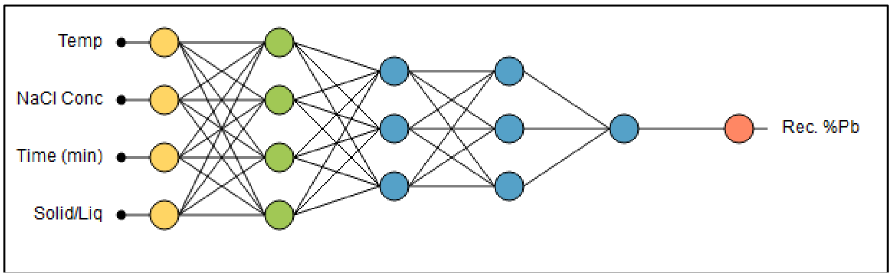

Figure 2. Proposed ANN structure.

Table 2. Data set.

	Minimum	Maximum	Mean	Deviation
Temperature	0	103	11.5597	25.4812
NaCl conc.	0	300	64.7799	116.898
Time (min)	0	300	10	31.6277
Solid / Liq.	0	0.25	0.0296122	0.0644186
Recovered Pb %		98.28	16.7502	32.4278

The correlation is a numerical value between 0 and 1 that expresses the strength of the relationship between two variables. When it is close to 1 it indicates a strong relationship, and a value close to 0 indicates that there is no relationship. The correlation between all input variables is given in Table 3. Principal components analysis allows to identify underlying patterns in a data set, so it can be expressed in terms of other data set of lower dimensions without much loss of information. The resulting data set should be able to explain most of the variance of the original data set by making a variable reduction. The final variables can be named principal components (Table 4).

Table 3. Input variables correlation.

	Temperature	NaCl conc.	Time (min)	Solid / Liq.
Temperature	1	0.824	0.618	0.82
NaCl conc.		1	0.586	0.86
Time (min)			1	0.598
Solid / Liq.				1

Table 4. Principle component.

	Relative variance	Cumulative variance
1	79.2227	79.2227
2	12.5725	91.7952
3	4.70966	96.5048
4	3.49518	100

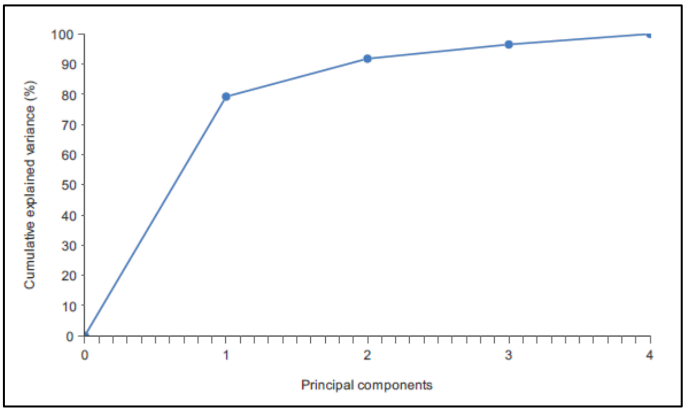

Figure 3. Variance chart.

The next plot (Figure 3) shows the cumulative explained variance for the principal components. The x-axis represents each of the principal components and the y-axis depicts the cumulative explained variance. As it can be seen, the total explained variance for all the principal components is 100% but if the number of chosen principal components decreases also makes it the total explained variance.

All prediction studies have been conducted with the aid of Neural Designer software. The size of the scaling layer is 4, the number of inputs. The scaling method for this layer is the Mean Standard Deviation. The number of layers in the neural network is 3. The following table depicts the size of each layer and its corresponding activation function. The architecture of this neural network can be written as 4:3:3:1. The norm of the parameters gives a clue about the complexity of the predictive model. If the parameters norm is small, the model will be smooth. On the other hand, if the parameters norm is very big, the model might become unstable. Also note that the norm depends on the number of parameters. Proposed parameters norm was obtained as 4.19.

3. Discussions

Input values and their corresponding values are given in Table 5. The input variables are Temp., NaCl concentration, Time (min) and Solid / Liq.; and the output variable is Rec. %Pb. The predictive model takes the form of a function of the outputs with respect to the inputs. The mathematical expression represented by the model can be used to embed it into another software, in the so-called production mode. The mathematical expression represented by the neural network is written below (Figure 4). It takes the inputs Temp., NaCl Concentration, Time (min) and Solid/Liq. to produce the output Rec. %Pb. For classification problems, the information is propagated in a feed-forward fashion through the scaling layer, the perceptron layers and the probabilistic layer.

Table 5. Principle component.

	Values
Temperature	98.7514
NaCl concentration	64.7799
Time (min.)	10
Solid / Liq.	0.0296122
Recovery amount of Pb (%)	0.987915861

```
scaled_Temp=(Temp-11.5597)/25.5079;
scaled_NaCl_Conc=(NaCl_Conc-64.7799)/117.021;
scaled_Time__min_=(Time__min_-10)/31.661;
scaled_Solid_Liq=(Solid_Liq-0.0296122)/0.0644863;
principal_component_1=(0.518613*scaled_Temp+0.520893*scaled_NaCl_Conc+0.432663*scaled_Time__min_+0.522028*scaled_Solid_Liq);
principal_component_2=(-0.179022*scaled_Temp-0.298939*scaled_NaCl_Conc+0.898128*scaled_Time__min_-0.268239*scaled_Solid_Liq);
principal_component_3=(0.834745*scaled_Temp-0.347343*scaled_NaCl_Conc-0.0748585*scaled_Time__min_-0.420653*scaled_Solid_Liq);
principal_component_4=(0.0468156*scaled_Temp-0.72018*scaled_NaCl_Conc-0.0237625*scaled_Time__min_+0.691798*scaled_Solid_Liq);
y_1_1=Logistic(-0.653454
-0.951606*principal_component_1
-0.510492*principal_component_2
+0.198641*principal_component_3
+0.344856*principal_component_4);
y_1_2=Logistic(1.32944
+0.215547*principal_component_1
-0.293948*principal_component_2
+0.710974*principal_component_3
-1.77833*principal_component_4);
y_1_3=Logistic(0.0118592
+0.209758*principal_component_1
-0.954104*principal_component_2
+1.02946*principal_component_3
+0.478226*principal_component_4);
y_2_1=tanh(-0.637585
-0.606519*y_1_1
-0.570129*y_1_2
-0.274573*y_1_3);
y_2_2=tanh(0.946404
+0.592686*y_1_1
+0.407883*y_1_2
-0.495361*y_1_3);
y_2_3=tanh(-1.55112
+0.252093*y_1_1
-0.174191*y_1_2
-1.14839*y_1_3);
scaled_Rec._%Pb=Logistic(1.87177
-0.403293*y_2_1
+0.936251*y_2_2
-1.45754*y_2_3);
(Rec._%Pb) = Probability(non_probabilistic_Rec._%Pb);

Logistic(x){
    return 1/(1+exp(-x))
}

Probability(x){
    if x < 0
        return 0
    else if x > 1
        return 1
    else
        return x
}
```

Figure 4. Mathematical expression.

A classifier can be well calibrated when the portion of positive events that it predicts is equal to the portion of positive events that occur. The calibration

plot shows how well calibrated a classifier is. The following chart (Figure 5) represents the true probability versus the estimated probability. The base line represents a classifier with a perfect calibration. If a classifier has a good discrimination capacity, it must be well calibrated. However, a good calibration of the classifier does not imply a good discrimination capacity. Obtained results showed that proposed model both have good calibration and discrimination.

Proposed model performance was evaluated with Sum Squared Error (SSE), Mean Squared Error (MSE), Root Mean Squared Error (RMSE), Normalized Squared Error (NSE) and Minkowski Error (ME) operators. These errors are presented in Table 6. All error results of the ANN classification system were obtained as within the acceptable limits.

According to the results of the classification, the most consistency is observed between the Solid to liquid ration and NaCl concentration on the Pb recovery in the model (Figure 6 and Figure 7). By analyzing the results, it can be concluded that both inputs have effect on the analyzed Pb recovery amount. These results show parallelism with those of the previous studies on lead recovery [2,6].

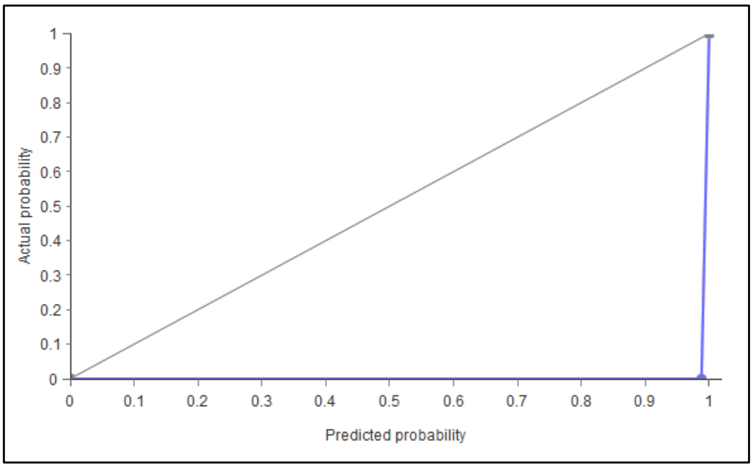

Figure 5. Calibration chart.

Table 6. Proposed network errors.

	Training	Selection	Testing
Sum squared error	3.1721	1.4508	1.2966
Mean squared error	0.0139	0.0187	0.0162
Root mean squared error	0.1149	0.1352	0.1284
Normalized squared error	0.1322	0.2374	0.1939
Minkowski error	7.2718	3.1872	2.6223

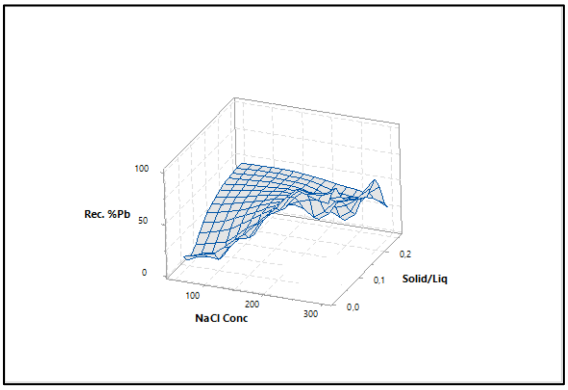

Figure 6. Pb recovery vs Solid/Liq and NaCl conc.

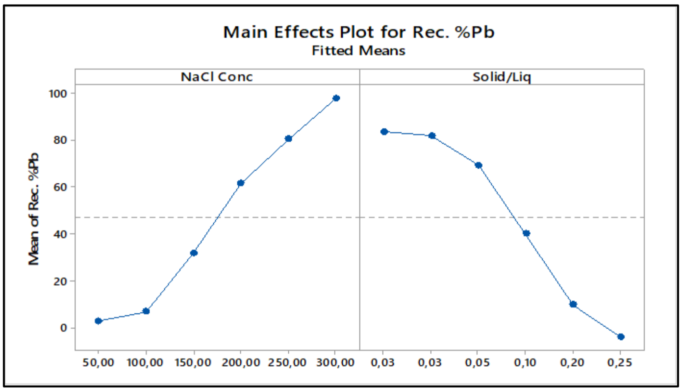

Figure 7. Main effects plot.

4. Conclusions

According to the physical, chemical and mineralogical investigations, the leach residue showed a complex nature. Having defined the characteristics, as the process economy is the basic limitation for production, hydrometallurgical treatment methods were assessed.

The aim of this research is to develop an ANN based Pb recovery systems in order to provide a classification of effecting materials. For this purpose, the system was developed using conjugate gradient algorithm. The outcomes of this research can be summarized as follows:

- The ANN model approves the strong correlation between the Pb recovery amount with the Solid to liquid ratio and NaCl concentration.
- The outcomes of the study can be assessed by other artificial and mathematical systems for better understanding of inputs effects on Pb recovery amounts.
- This statistical analysis indicates that lead recoveries up to 98 % could be obtained at a low pulp density with considerable NaCl concentration in laboratory scale. Compared with previous studies, these results yielded very consistent findings in terms of Pb leach efficiency.

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