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Genetic algorithm-artificial neural network model for the prediction of germanium recovery from zinc plant residues

S. Akkurt, S. Ozdemir & G. Tayfur

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Synopsis

A multi-layer, feed-forward, back-propagation learning algorithm was used as an artificial neural network (ANN) tool to predict the extraction of germanium from zinc plant residues by sulphuric acid leaching. A genetic algorithm (GA) was used for the selection of training and testing data and a GA-ANN model of the germanium leaching system was created on the basis of the training data. Testing of the model yielded good error levels ($r^2 = 0.95$). The model was employed to predict the response of the system to different values of the factors that affect the recovery of germanium and the results facilitate selection of the experimental conditions in which the optimum recovery will be achieved.

World production of germanium, which is widely used in the chemical, electronic and optical materials industries, is about 100 t/year. It is obtained mainly as a by-product of the extraction of other metals, and zinc smelter flue systems are the principal source. Cinkur has been stockpiling solution purification residues at its zinc plant in Turkey and has built up a stock of more than 300 t with a germanium concentration in excess of 1000 ppm. In a previous study¹ this residue, also known as 'copper cake', was treated with 0-250 g/l H₂SO₄ to bring the germanium into a solution that could later be processed by tannin precipitation² or solvent extraction;³ the solution remaining after germanium extraction could then be processed to recover its Cu, Ni, Co, etc. The effects of such factors as time, temperature, solids content and acid concentration on the leach recovery of germanium were reported. Data obtained in that work have now been employed to create a model that describes germanium recovery from zinc plant residues by feeding them via a genetic algorithm (GA) into an artificial neural network (ANN).

ANN are used to model complex systems in a wide range of fields.^{4–6} In many mining and extractive metallurgical projects the effects of processing parameters (e.g. time, temperature, mixing speed, the solids content in a leaching reactor, rotation speed of a tumbling ball-mill, media/charge ratio in the mill, etc.) on the response of the system (e.g. per cent leach recovery of zinc or energy consumption in a ballmill) are subjects of study. ANN can be used to model such processes by using plant data as feed in the setting up of the model. The model can then be used to predict the response of the system to different combinations of parameters. In other words, plant engineers can utilize such models for optimization purposes.

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Model development

The common, three-layer, feed-forward type of ANN, as shown in Fig. 1, was adopted in the present study. In a feedforward network the input quantities are fed into input layer

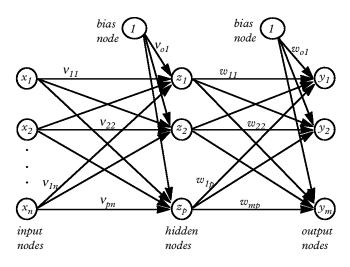


Fig. 1 Typical architecture of back-propagation ANN model

neurons, which pass them on to the hidden-layer neurons after application of a weight. A hidden-layer neuron adds up the weighted input received from each input neuron, associates it with a bias and then passes the result through a nonlinear transfer function (Fig. 2). The output neurons perform the same operation as a hidden neuron. In this study the sigmoid function was employed as an activation function.⁷

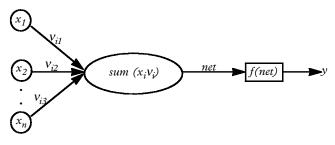


Fig. 2 Weighted output from each node passes through nonlinear transfer function

Learning is accomplished in ANN by a back-propagation (BP) algorithm. The objective of a BP network is, by minimizing a predetermined error function, to find the optimal weights that would generate an output vector, $\mathbf{Y} (= y_1, y_2, ..., y_p)$, as close as possible to the target values of the output vector, $\mathbf{T} (= t_1, t_2, ..., t_p)$, with a selected accuracy. A predetermined error function has the form^{4,7}

$$E = \sum_{P} \sum_{m} \left(y_i \uparrow t_i \right)^2 \tag{1}$$

where y_i is a component of an ANN output vector, **Y**; t_i is a component of a target output vector, **T**; *m* is number of output neurons; and *P* is number of training patterns.

Table 1 Conditions for copper cake leaching experiments¹

Data collection

The data used in the GA–ANN modelling had been collected previously from the leaching of germanium-containing zinc plant residues denoted 'old copper cake' (OCC) and 'new copper cake' (NCC),¹ which had been stockpiled at different periods during operation of the plant. Both cake samples

| Test run | Type of copper cake | Solids, % | Temperature, °C | Concentration of H ₂ SO ₄ , g/l | Time, h | MnO ₂ addition g/25g CC | Recovery of Ge in leach, % |
|-------------|------------------------|--------------|--------------------|---|------------|---------------------------------------|-------------------------------|
| 1 | OCC | 25 | 85 | 0 | 2.5 | 0 | 0.4 |
| 2 | OCC | 25 | 25 | 150 | 2.5 | 0 | 81.0 |
| 3 | OCC | 25 | 55 | 150 | 2.5 | 0 | 84.7 |
| 4 | OCC | 25 | 85 | 150 | 2.5 | 0 | 89.3 |
| 5 | OCC | 25 | 85 | 50 | 2.5 | 0 | 38.1 |
| 6 | OCC | 25 | 85 | 200 | 2.5 | 0 | 72.5 |
| 7 | OCC | 25 | 85 | 250 | 2.5 | 0 | 74.4 |
| 8 | OCC | 50 | 85 | 150 | 2.5 | 0 | 32.4 |
| 9 | OCC | 12.5 | 85 | 150 | 2.5 | 0 | 88.2 |
| 10 | OCC | 6.25 | 85 | 150 | 2.5 | 0 | 92.8 |
| 11 | OCC | 25 | 85 | 150 | 0.5 | 0 | 88.0 |
| 12 | OCC | 25 | 85 | 150 | 3.5 | 0 | 85.3 |
| 13 | NCC | 25 | 85 | 150 | 0.5 | 0.43 | 98.9 |
| 14 | NCC | 25 | 85 | 150 | 2.5 | 0.43 | 99.3 |
| 15 | NCC | 25 | 85 | 150 | 2.5 | 0 | 88.8 |
| 16 | NCC | 25 | 85 | 150 | 3.5 | 0.43 | 98.9 |
| 17 | OCC | 25 | 96 | 150 | 2.5 | 0 | 91.6 |
| 18 | OCC | 25 | 85 | 100 | 2.5 | 0 | 80.6 |
| 19 | OCC | 25 | 85 | 150 | 1.5 | 0 | 89.0 |
| 20 | NCC | 25 | 85 | 150 | 1.5 | 0.43 | 99.3 |

OCC, old copper cake; NCC, new copper cake.

The least-square error method, along with a generalized delta rule, was used to optimize the network weights. The gradient-descent method, along with the chain rule of derivatives, was employed to modify network weights:

$$v_{ij}^{\text{new}} = v_{ij}^{\text{old}} \uparrow \delta \frac{\partial E}{\partial v_{ij}}$$
(2)

where δ is learning rate used to increase the chance of avoiding the training process being trapped in a local minimum instead of a global minimum.

Quantitative input data of various types and magnitudes are normalized at the beginning to a common scale of 0.1-0.9. Equation 3 compresses all the data in this range:

$$x_i = 0.1 + 0.8 \times \frac{x_i \uparrow x_{\min}}{x_{\max} \uparrow x_{\min}}$$
(3)

where x_{max} and x_{min} are the maximum and minimum values of the *i*th node in the input layer for all the feeding data vectors $(1 \le i \le n)$. At the beginning, also, the weights v_{ij} and w_{ij} were assigned a random value between -1 and 1. Details of the ANN can be found elsewhere.^{4,7,10}

Development of an ANN model involves the use of training and testing data-sets. When the training and testing data-sets are not separated to equal averages and similar ranges the resulting model could be biased. To overcome these shortcomings of ANN a GA was used in this study. A GA can be described as a guided random search method. It is made up of four important steps: initialization, selection, reproduction and mutation.^{8,9} Detailed information on the GA used can also be found elsewhere.^{4,8,9} were dried at 110°C, ground to pass 100 mesh and subjected to leaching in sulphuric acid solution under the conditions reported in Table 1. Leaching was carried out in a glass reactor that was heated externally from beneath by a hotplate. Mixing was performed with an immersed, Teflon-coated magnet at a fixed rotation speed of about 150 rev/min. The temperature was measured by an immersed mercury thermometer.

Model application

The three-layer, feed-forward ANN was constructed with six neurons in the input layer for six input variables. In the hidden layer six neurons were chosen by trial and error. Finally, in the output layer one neuron was used for the output variable per cent germanium recovery from the leaching solution. The input variables were x_1 , per cent solids in the leach; x_2 , leaching temperature, °C; x_3 , acid concentration, g H₂SO₄/l; x_4 , duration of leaching, h; x_5 , MnO₂ added, g/25 g copper cake; and x_6 , type of copper cake (0 for OCC and 1 for NCC).

These six input variables and one output variable form a special case of the more general model illustrated in Fig. 1, in which the number of output-layer neurons is more than one. Neurons in each layer are fully connected to every node in the neighbouring layers. No bias term was used during modelling, but a momentum term was used to help to obtain better convergence during iterations. A total of 20 data-sets were employed, of which 16 were used for training of the ANN and the remainder for testing of the model. Each data-set had seven components, comprising the six input variables and the output variable. The program operated for 40 000 iterations

and the optimal weights were calculated with an average learning error of 7.69% for the output variable.

The results of the training runs are plotted in Fig. 3, in which the high r^2 value (0.910) indicates that the training was successfully performed. Ideally, such graphs should have an intercept value of 0 and a slope of 1, i.e. if the data were on the diagonal, the learning error would be zero.

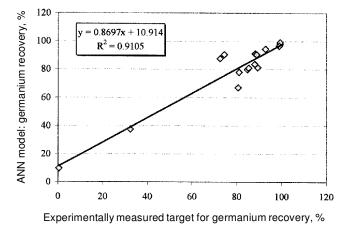


Fig. 3 Results of learning run

Because of the limited number of experimental data only four testing outputs were compared with real, measured data and the results are given in Table 2. As can be seen in Table 2, the performance of the model is quite satisfactory.

Table 2 Results of testing runs

| No. | Targeted output, % Ge recovery | Output by NN model, % Ge recovery | Absolute error, % Ge recovery | Relative error, % |
|-----|--------------------------------------|---|-------------------------------------|-------------------------|
| 1 | 38.1 | 39.9 | -1.7 | -4.6 |
| 2 | 89.0 | 82.3 | 6.7 | 7.5 |
| 3 | 98.9 | 100.5 | -1.6 | -1.6 |
| 4 | 91.6 | 81.7 | 9.8 | 10.8 |

Results

The experimental data for the effects of single factors are presented below and the results of the ANN sensitivity analysis are then discussed.

Single factor effects

Figs. 4-7 display the effects of individual factors on the ger-

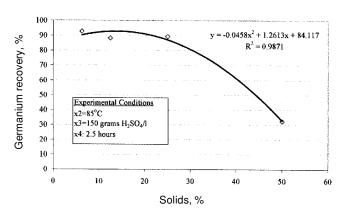


Fig. 4 Effect of per cent solids content on germanium recovery

manium recovery. The data points were obtained from Table 1. The polynomial equations for the best-fit curves are shown in each figure along with the r^2 values. Increasing the percentage of solids led to a sharp decrease in the leach recovery (Fig. 4). In contrast, increase in the acid concentration brought a substantial boost in germanium leach recovery (Fig. 5), but at very high acid concentration the germanium recovery decreased by about 15%. This may have been the result of precipitation from the solution in extremely acidic concentrations.

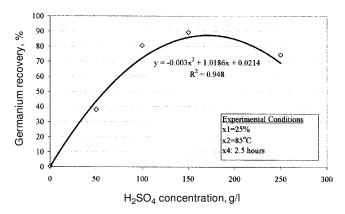


Fig. 5 Effect of acid concentration on germanium recovery

Germanium dissolution from the cake reached its peak within about half an hour and further mixing of the system did not help to increase recovery (Fig. 6). Increases in temperature had only a slight beneficial effect in achieving higher leach recoveries (Fig. 7).

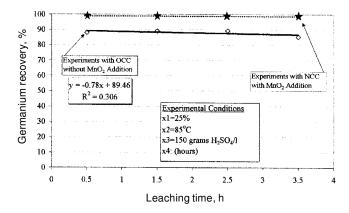


Fig. 6 Effect of leaching time on germanium recovery

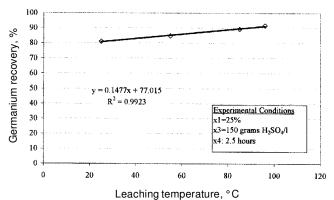


Fig. 7 Effect of leaching temperature on germanium recovery

The combined effects of more than one factor at varying levels can be seen on surface plots produced by use of the ANN model developed here.

Sensitivity analysis

The GA–ANN model was used to predict outputs (per cent germanium recovery) at various levels of the input factors, i.e. time, temperature, acid concentration and per cent solids. For the purposes of sensitivity analysis the complete range of each input factor was discretized into 10-15 subdivisions. For example, the temperature range $25-90^{\circ}$ C was divided into ten subdivisions and the acid concentration (0-250 g/l) was divided into 11 subdivisions, such that a total of 110 predicted per cent germanium recovery values was obtained. The utility of this sensitivity analysis is that it enables the researcher or plant operator to identify the experimental conditions that give higher germanium recovery.

Figs. 8–13, based on the results of prediction runs with the model, show the effects on germanium recovery of two factors at a time, in the form of surface plots. The effect of time and temperature on germanium recovery is displayed in Fig. 8. It can be seen that increasing temperature led to a gradual linear increase of germanium recovery at all lengths of time. The duration of leaching was not important because germanium was taken into solution within half an hour and further leaching did not increase germanium recoveries. Insignificant interaction is observed between the time and temperature factors.

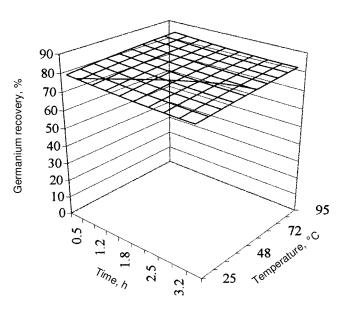


Fig. 8 Combined effects of leaching time and temperature on germanium leach recovery at constant values of x_1 , per cent solids in leaching process (25%), and x_3 , acid concentration (150 g H₂SO₄/l)

Fig. 9 is a surface plot of the effects of time and per cent solids on germanium recovery. Leaching time was not a strong factor in increased germanium recovery, but solids loading of the leaching system, on the other hand, had a major effect. Fifty per cent solids loading entailed the loss of more than half of the germanium recovery. However, more than 80% recovery was achieved at 25% solids at almost every length of leaching time.

Fig. 10 shows the effect of acid concentration and time on germanium recovery. The germanium recovery reached a plateau at around 80–90% once a 150 g/l acid concentration

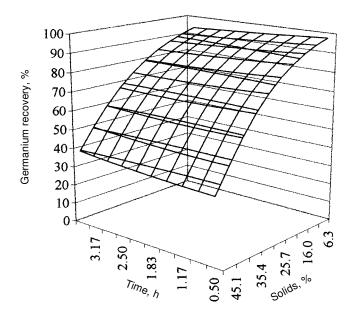


Fig. 9 Combined effects of leaching time and per cent solids on germanium leach recovery at constant values of x_{2} , leaching temperature (85°C), and x_{3} , acid concentration (150 g H₂SO₄/l)

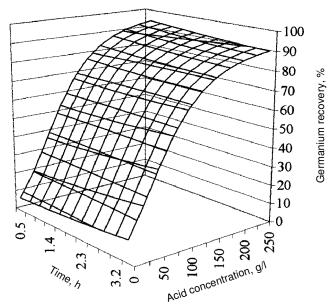


Fig. 10 Combined effects of leaching time and acid concentration on germanium leach recovery at constant values of x_1 , per cent solids in leaching process (25%), and x_2 , leaching temperature (85°C)

was exceeded. This behaviour altered little with different lengths of time.

Fig. 11 illustrates the effects of temperature and acid concentration; increasing acid concentration led to an increase in germanium recovery at all levels of temperature.

Fig. 12 shows the effects of per cent solids and acid concentration. Increase in acid concentration and decrease in per cent solids brought about increased recoveries. The germanium recovery was highest at low per cent solids (6.25-25%)and high acid concentrations—between about 100 and 200 g/l. A high solids loading with a low acid concentration was clearly the least productive combination.

Fig. 13 shows the effects of temperature and per cent solids. Increase in per cent solids decreased the recovery

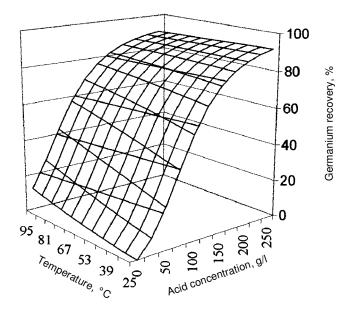


Fig. 11 Combined effects of leaching temperature and acid concentration on germanium leach recovery at constant values of x_1 , per cent solids in leaching process (25%), and x_4 , leaching time (2.5 h)

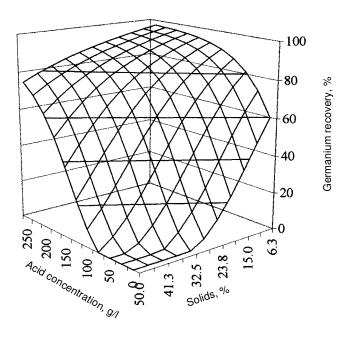


Fig. 12 Combined effects of per cent solids in leaching process and acid concentration on germanium leach recovery at constant values of x_2 , leaching temperature (85°C), and x_4 , leaching time (2.5 h)

sharply at all temperatures. Increases in temperature at high per cent solids led to a gradual increase in germanium recovery, but this effect was not observed at low solids loadings.

The relationships observed between factor effects and the germanium recovery conform to the general understanding of leaching systems. The plots are especially useful when a researcher seeks interactions between factor effects and a visual means of inspecting system behaviour.

Conclusions

A three-layer GA–ANN model was developed for the prediction of per cent germanium recovery from the solution purification residues of a Turkish zinc plant. The satisfactory predictions of the observed recovery by the model indicate

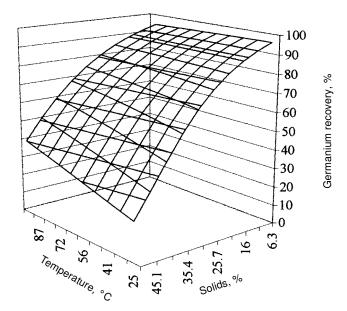


Fig. 13 Combined effects of leaching temperature and per cent solids in leaching process on germanium recovery at constant values of x_3 , acid concentration (150 g H₂SO₄/l), and x_4 , leaching time (2.5 h)

that ANN could be a useful tool for the modelling of leaching systems.

Genetic algorithms provided a balanced means of separating training and testing data at the beginning of the modelling task. The learning rate was adjusted on the basis of a practical formula developed in-house, which gave good results.

Factors affecting the leaching recovery of germanium were investigated quantitatively. Data obtained through a classical 'one-factor at a time' approach could be presented on singlefactor plots, but such plots are unable to show the effects of combinations of factors. However, GA–ANN modelling is able to produce surface plots that facilitate understanding of the system by providing a visual representation of interactions within it. Thus, the model lends itself to use by plant operators as a guide to the conditions that give maximum recovery.

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Authors

S. Akkurt was awarded BS and MS degrees from the Middle East Technical University, Ankara, Turkey, and a Ph.D. from Clemson University, U.S.A., before joining Izmir Institute of Technology, Turkey, in 1998, where he is an assistant professor.

Address: Mechanical Engineering Department, Izmir Institute of Technology, Izmir, 35437 Turkey; e-mail sedatakkurt@iyte.edu.tr

S. Ozdemir gained his BS at Dokuz Eylul University, Izmir, Turkey, MS at Illinois Institute of Technology, Chicago, U.S.A., and Ph.D. from the University of Florida, U.S.A., before joining Izmir Institute of Technology in 2000.

G. Tayfur was awarded a B.Sc. degree from Istanbul Technical University, Turkey, and M.Sc. and Ph.D. degrees from the University of California, Davis, U.S.A., before joining Izmir Institute of Technology in 1995.

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