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Artificial neural network prediction of aluminum extraction from bauxite in the Bayer process

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Abstract: This paper presents the results of statistical modeling of the bauxite leaching process, as part of the Bayer technology for alumina production. Based on the data collected during the period 2008–2009 (659 days) from the industrial production in the Alumina Factory Birač, Zvornik (Bosnia and Herzegovina), the above-mentioned process was statistically modeled. The dependant variable, which was the main target of the modeling procedure, was the degree of Al_2O_3 recovery from boehmite bauxite during the leaching process. The statistical model was developed as an attempt to define the dependence of the degree of Al_2O_3 recovery on the input variables of the leaching process, *i.e.*, the composition of the bauxite, the composition of the sodium aluminate solution and the caustic module of the solution before and after the leaching process. As statistical modeling tools, multiple linear regression analysis (MLRA) and artificial neural networks (ANNs) were used. The fitting level obtained using MLRA, was $R^2 = 0.463$, while the ANN resulted in an R^2 value of 0.723. In this way, the model defined using the ANN methodology could be used for the efficient prediction of the degree of recovery of Al_2O_3 as a function of the process inputs, under the industrial conditions of the Alumina Factory Birač, Zvornik. The proposed model also has a universal character and, as such, is applicable in other factories employing the Bayer technology for alumina production.

Keywords: leaching; bauxite; Bayer process; statistical modeling; neural networks.

INTRODUCTION

The Bayer process of alumina extraction is a basic commercial procedure and more than 90 % of the world alumina production is obtained in this way. Despite the fact that this process has been used for alumina production for a long

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period, there are still attempts to improve the process further.[1] Bauxite is a complex heterogeneous material used in the Bayer process of alumina production. Aluminum is usually present in bauxite in the form of hydroxide minerals, such as gibbsite (hydrargillite) ($Al(OH)_3$), boehmite ($AlO(OH)$) or diaspore ($HAlO_2$). Besides aluminum minerals, bauxite ore contains various combinations of silica (SiO_2), aluminosilicates, iron oxide (Fe_2O_3), titanium oxide (TiO_2) and other impurities, such as carbonates and sulfides, in trace amounts.[2,3]

The Bayer process includes the leaching of bauxites in a concentrated sodium hydroxide (caustic) solution at temperatures ranging from 373 K (100 °C) to 523 K (250 °C), depending on the mineralogical form of the aluminum hydroxide in the bauxite. The process includes reactions with soluble silica compounds and titan dioxide under certain conditions.[4] Silicon is usually present in bauxite in two potential forms: *i*) soluble, which is usually in the form of kaolinite ($Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$) or the amorphous form of silicon dioxide (silica) and *ii*) insoluble, in the form of quartz. In a caustic solution, the dissolved kaolinite and amorphous silicon dioxide form a sodalite-type product, with the general formula $3(Na_2O \cdot Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O)$ 2NaX, where X could be: OH^-, Cl^-, CO_3^{2-} or SO_4^{2-}.[5–8] The rate of the aluminum hydroxide leaching process depends on its mineral form in the bauxite. The trihydrate bauxite type gibbsite can be dissolved in a caustic solution in the temperature range 373–453 K (100–180 °C). Monohydrate bauxite forms (boehmite and diaspore) are dissolved in the temperature ranges 403–453 K (130–180 °C) and 473–523 K (200–250 °C), respectively.[9,10]

Sodium aluminosilicate is precipitated in red mud, which is the source of aluminum and sodium hydroxide losses during the Bayer process. Sedimented red mud also presents an environmental problem.[11] This problem has attracted much attention recently, especially because of global environmental protection problems, which demand the compliance with global principles during local actions.[12]

During the process of bauxite leaching in alkaline sodium aluminate, the aluminum ions in the solution are hydrolyzed in the aqueous environment, forming numerous mononuclear and polynuclear hydroxo complex ions. Finally, in mild-to-strong alkaline solutions, $Al(OH)_4^-$ is predominant at pH values higher than 10.[11] Kinetic parameters indicate that the bauxite leaching process is conducted in the so-called “kinetic area”, which suggests that the temperature is the main parameter influencing the overall rate of the process.[10,13] The process parameters influencing the leaching rate and the degree of Al_2O_3 recovery are: the mineralogical and chemical composition of bauxite, grain size distribution, caustic module of the starting solution and its Na_2O (caustic) mass fraction, leaching process temperature, stirring speed and duration of the process.[1,10,11,13]

The process of bauxite leaching, under industrial conditions of Bayer technology for alumina production is highly complex. The ability to predict the reco-

very of Al_2O_3 during leaching as the result of modeling of the input process variables presents a great challenge for the management of the process.[4]

The main objective pursued in this work was to create a mathematical model for the prediction of the degree of Al_2O_3 recovery (output of the process), during bauxite leaching, as the function of the input variables of the process. The obtained model presents a great advantage due to its ability to predict accurately the output of the investigated process.

EXPERIMENTAL

A data set suitable for the calculations and presented in this paper was formed according to the data collected during the industrial production in the Alumina Factory Birač, Zvornik (Bosnia and Herzegovina). This factory has a production capacity of 600 000 tons of alumina per year. The important process parameters included in the obtained data set were the chemical composition of the bauxite (including Al_2O_3, SiO_2, Fe_2O_3, TiO_2, CaO and H_2O, and the mass loss during calcination); the composition of the starting aluminate solution (including Al_2O_3, Na_2O and the starting caustic module); the chemical composition of the residual autoclave mud – red mud (including Al_2O_3, $Na_2O_{(total)}$, SiO_2, TiO_2 and CaO); the composition of the aluminate solution at the end of the leaching process (including Al_2O_3, Na_2O and the final caustic module). The chemical composition of the samples was expressed as the mass fraction of the constituents. All of these process parameters were measured daily during the years 2008 and 2009, and in this way, 659 samples were collected during each day of stabile production in the factory. All the samples were examined for potential outliers before any further modeling procedure. No strong extreme behavior of the variables was detected. Thus, the obtained results could be considered as true representatives of the investigated process.

The output of the investigated process was the "Al_2O_3 leaching recovery", which is presented as Y in the further text, and it refers to the alumina recovery in the digestion process that was calculated using the following equation:

$$Y = 100(1 - Al_2O_{3(rm)} \cdot Fe_2O_{3(b)} / Al_2O_{3(b)} \cdot Fe_2O_{3(rm)}) \quad (1)$$

where $Al_2O_{3(b)}$ and $Fe_2O_{3(b)}$ are the mass fraction in the bauxite (%) and $Al_2O_{3(rm)}$ and $Fe_2O_{3(rm)}$ are the mass fraction in the residual autoclave mud (red mud) (%).

On application of Eq. (1) for calculating the degree of recovery Al_2O_3 during the leaching process, which is based on adopting the "inert" Fe_2O_3, acceptable results (accuracy above 99 %) were obtained.

The following process parameters were selected as the inputs included in the statistical modeling procedure:

X_1 – Na_2O (caustic) concentration in the starting solution (g dm^{-3}),
X_2 – Al_2O_3 concentration in the starting solution (g dm^{-3}),
X_3 – starting caustic ratio of the solution,
X_4 – moisture mass fraction in the bauxite (%),
X_5 – Al_2O_3 mass fraction in the bauxite (%),
X_6 – SiO_2 mass fraction in the bauxite (%),
X_7 – Fe_2O_3 mass fraction in the bauxite (%),
X_8 – TiO_2 mass fraction in the bauxite (%),
X_9 – CaO mass fraction in the bauxite (%),
X_{10} – mass loss during calcinations of the bauxite (%) and
X_{11} – final caustic ratio of the solution at the end of the leaching process.

During the period when these parameters were measured and the corresponding data set formed, the operation of the factory was in a stable mode. The bauxite used for alumina production was from the ore body Vlasenica (Bosnia and Herzegovina). This ore body has large reserves of boehmitic bauxites. The temperature of the leaching process was kept constant at 518 K (245 °C). The pressure in the autoclave reactors was 35 bar. The size distribution of the bauxite grains was 100 % –74 μm, obtained after the hydrocyclone classification. The solid to liquid ratio of the autoclave charge was *S*:*L* = 1:5. The solid phase concentration was 160–200 g dm^{-3}, depending on the Na_2O (caustic) concentration in the returned aluminate solution. The rate of mechanical stirring was 31 rpm.

RESULTS AND DISCUSSION

For modeling the bauxite leaching process, the data were collected by measuring the above-defined input and output process variables. The values of the measured input variables of the technological process as well as the process quality indicators, the output of the process (Y) in the form of descriptive statistics results, are presented in Table I.

TABLE I. Descriptive statistics of the input (X_i) and output (Y) variables of the bauxite leaching process including 659 data sets

Parameter	Range	Minimum	Maximum	Mean		Std. deviation	Variance
				Statistic	Std. error		
X_1	90.19	129.66	219.86	2.040E2	0.4050	10.398	108.131
X_2	48	81	129	109.79	0.329	8.442	71.274
X_3	1.898	2.075	3.972	3.069	0.0080	0.2059	0.042
X_4	7.21	8.11	15.32	11.533	0.056	1.439	2.073
X_5	4.58	50.08	54.66	51.975	0.034	0.866	0.750
X_6	3.54	4.88	8.42	6.3208	0.022	0.562	0.316
X_7	4.39	22.42	26.81	24.833	0.031	0.803	0.645
X_8	0.63	2.22	2.85	2.540	0.003	0.078	0.006
X_9	2.82	0.29	3.11	1.253	0.014	0.370	0.137
X_{10}	3.17	11.40	14.57	12.459	0.023	0.581	0.337
X_{11}	0.524	1.116	1.639	1.414	0.00122	0.0314	0.001
Y	13.077	76.293	89.370	84.427	0.06574	1.687	2.848

It should be noted that X_8 and X_{11} have a small variance (Table I). However, these variables are important for the investigated technological process, especially X_{11}. The variable X_{11} presents the caustic ratio of the solution at the end of the leaching process; it is one of the most important parameters of the Bayer process; thus, it cannot be omitted from the analysis.

Linear regression analysis

For defining the correlation dependence in the form of output of the process $Y = f$ input of the process (X_1–X_{11}), a bivariate correlation analysis was performed. In this analysis, the Pearson correlation coefficients (PCC) with responding statistical significance were calculated (Table II) using statistical software

TABLE II. Correlation matrix for the input (X_1–X_{11}) and output variables (Y) of the investigated process (number of data points for each variable was 659)

Parameter	Correlation	X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}	X_{11}	Y
X_1	PC[a]	1											
	S[b]												
X_2	PC	0.534[c]	1										
	S	0.000											
X_3	PC	0.151[c]	–0.752[c]	1									
	S	0.000	0.000										
X_4	PC	–0.368[c]	–0.561[c]	0.367[c]	1								
	S	0.000	0.000	0.000									
X_5	PC	–0.379[c]	–0.463[c]	0.238[c]	0.570[c]	1							
	S	0.000	0.000	0.000	0.000								
X_6	PC	–0.378[c]	–0.240[c]	–0.012	0.107[c]	–0.105[c]	1						
	S	0.000	0.000	0.758	0.006	0.007							
X_7	PC	0.223[c]	0.425[c]	–0.320[c]	–0.393[c]	–0.431[c]	–0.282[c]	1					
	S	0.000	0.000	0.000	0.000	0.000	0.000						
X_8	PC	0.251[c]	0.030	0.164[c]	0.127[c]	0.049	–0.353[c]	–0.026	1				
	S	0.000	0.439	0.000	0.001	0.210	0.000	0.499					
X_9	PC	0.153[c]	0.055	0.063	–0.127[c]	–0.380[c]	–0.108[c]	–0.247[c]	–0.011	1			
	S	0.000	0.155	0.108	0.001	0.000	0.005	0.000	0.771				
X_{10}	PC	0.474[c]	0.205[c]	0.123[c]	–0.220[c]	–0.384[c]	–0.307[c]	–0.277[c]	0.233[c]	0.346[c]	1		
	S	0.000	0.000	0.002	0.000	0.000	0.000	0.000	0.000	0.000			
X_{11}	PC	–0.327[c]	–0.479[c]	0.312[c]	0.367[c]	0.300[c]	0.163[c]	–0.237[c]	–0.082[d]	–0.022	–0.180[c]	1	
	S	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.035	0.573	0.000		
Y	PC	0.233[c]	–0.069	0.265[c]	0.169[c]	0.311[c]	–0.494[c]	–0.289[c]	0.424[c]	0.126[c]	0.325[c]	0.103[c]	1
	S	0.000	0.076	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000	0.008	

[a]Pearson correlation; [b]Sig. (2-tailed); [c]correlation is significant at the 0.01 level (2-tailed); [d]Correlation is significant at the 0.05 level (2-tailed)

SPSS v.18 (PASW Statistics). The results obtained revealed considerable statistical significance of the correlations ($p \leq 0.01$) for most of the coupled variables, Table II. To define the dependence of the output variable as a function of input variables, using the multiple linear regression analysis (MLRA) with acceptable level of fitting, it is necessary that the value of PCC between Y and X_i should be with statistical significance ($p \leq 0.01$).[14,15] The analysis of the data presented in Table II revealed that this constraint was attained in all cases except for Y–X_2: $PCC = -0.069$ ($p = 0.076$).

As most of the variables had an acceptable level of correlation and statistical significance, it was concluded that the MLRA approach should be considered as an adequate tool for modeling the investigated process. For the purpose of MLRA analysis, the assembly of 659 input and output samples was divided into two groups. The first group consisted of 458 (70 %) randomly selected samples. This group was used for the creation of the model, whereas the second group, consisting of the remaining 201 (30 %) samples from the starting data set, was used for testing the model. The selection of the variables for these two stages was performed using a random number generator.

Using multivariable regression, the following equation was developed to describe the relationship between the leaching recovery and the selected input variables:

$$Y = 38.437 - 0.024X_1 + 0.098X_2 + 3.603X_3 - 0.022\,X_4 + 0.478X_5 - 1.101X_6 - \\ - 0.562X_7 + 4.892X_8 + 0.203X_9 + 0.270X_{10} + 6.586X_{11}\ (R^2 = 0.555) \quad (2)$$

Equation (2) represents the complete linear model developed during the first phase of the MLRA modeling procedure.

The results of the ANOVA tests of the developed model are presented in Table III. The small value of the F statistics (Table III) indicates that the developed model was below the level for an accurate prediction of the value of dependent variable (Y) based on the values of input variables (X_i). The ratio of the regression to the residual was 55:45 %, advocating that only 55 % of the variance in the dependant variable (Y) values, is explained by the model.

TABLE III. Results of the ANOVA[a] test (predictors (constant): X_{11}, X_8, X_9, X_7, X_3, X_6, X_1, X_4, X_{10}, X_5 and X_2; dependent variable: Y) performed during the training of the model

Parameter	Sum of squares	Degree of freedom (df)	Mean square	F	Sig.
Regression	708.744	11	64.431	50.511	.000[a]
Residuals	568.913	446	1.276	–	–
Total	1277.657	457	–	–	–

The summary results describing the MLRA model in the development phase, are presented in Table IV. The relatively small value of the correlation coefficient ($R = 0.745$), resulting by a low coefficient of determination ($R^2 = 0.555$),

suggests that the model developed according to MLRA seems unacceptable for the prediction of alumina extraction from bauxite in the Bayer process.

TABLE IV. MLRA summary of the model developed during the training phase

Model	R	R^2	Adjusted R^2	Standard error of the estimate
1	0.745	0.555	0.544	1.1294

However, some further validation of the model was performed in the testing stage using the second part of the data set (total 201 vectors). During the testing phase of the MLRA model, the calculated coefficient of determination (R^2), as expected, further decreased in comparison to the testing phase and was now 0.463. A comparative presentation of the measured and the calculated values using the MLRA approach for the investigated process is illustrated in Fig. 1.

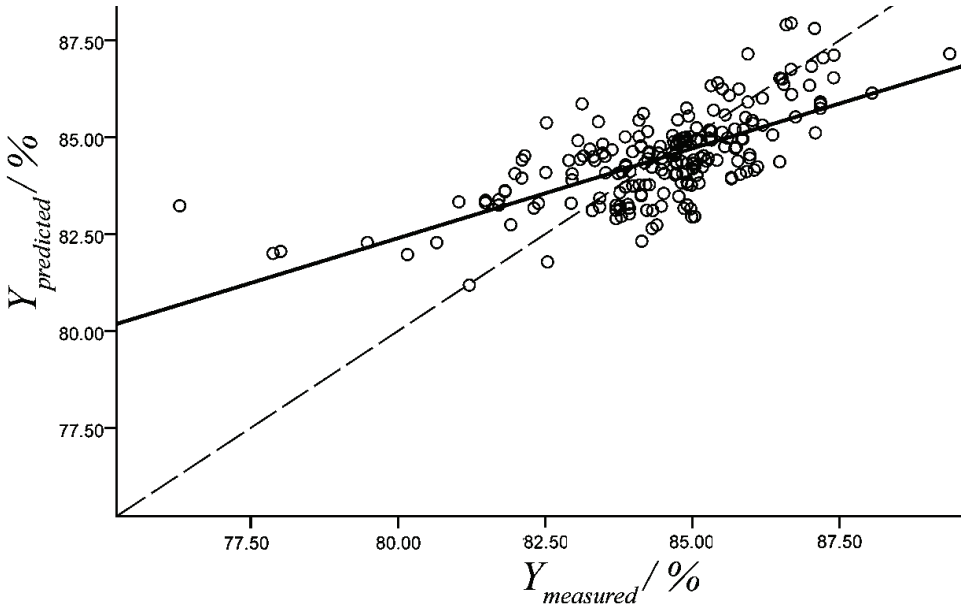

Fig. 1. Dependence between the calculated and measured values of the Al_2O_3 recovery (Eq. (2)); - - - = ideal position; ⎯ = regression lines; ○ = values calculated using the MLRA model in the testing stage).

The difference distribution between the recoveries calculated from Eq. (2) and the actually determined amounts of recovery is shown in Fig. 2. The multivariable regression analysis predicted the alumina leaching recovery with a determination coefficient $R^2 = 0.463$ and differences of – 7.20 to 2.50 from actually determined recoveries in the plant, which does not represent a large significance. Thus, the results showed that the MLRA modeling methodology lead to no acceptable correlation between the selected variables and the Al_2O_3 recovery.

Artificial neural network

Artificial neural networks can be viewed as nonlinear approaches to multivariate statistical methods, not bound by assumptions of normality or linearity. Although neural networks originated outside the field of statistics and have even been seen as an alternative to statistical methods in some circles, there are signs that this viewpoint is initiating an appreciation of the manners in which neural networks complement classical statistics.[16,17]

The ANN used in the development of the model described in this paper is depicted in Fig. 3. As shown, this network consists of three layers of nodes. The layers, described as input, hidden and output layers, comprise i, j and k numbers of processing nodes, respectively. Each node in the input (hidden) layer is linked to all the nodes in the hidden (output) layer using weighted connections. In addition to the i and j numbers of input and hidden nodes, the ANN architecture also houses a bias node (with a fixed output +1) in its input and hidden layers and they provide additional adjustable parameters (weights) for model fitting. The number of the nodes (i) in the ANN network input layer is equal to the number of inputs in the process, whereas the number of output nodes (k) equals the number of process outputs. However, the number of hidden nodes (j) is an adjustable parameter the magnitude of which is determined by issues, such as the desired approximation and generalization capabilities of the network model.[18,19]

The employment of an ANN usually comprises three phases. First is the training phase, which is achieved using 70–80 % of randomly selected data from the starting data set. During this phase, the correction of the weighted parameters of the connections is achieved through the necessary number of iterations, until the mean squared error between the calculated and measured outputs of the network is minimal. During the second phase, the remaining 20–30 % of the data are used for testing the "trained" network. In this phase, the network uses the weighted parameters determined during the first phase. These new data, excluded during the network learning stage, are now incorporated as the new input values (X_i) that are then transformed into the new outputs (Y_i). The third phase is a validation of the network on a new data set. This data set usually consists of the data from the new experimental measurements of the same process. The validation phase presents the final level of a successful or unsuccessful prediction obtained by using the network developed in the two previous stages on a new data set.[17,20,21]

In this study, the ANN methodology was applied for modeling the process of bauxite leaching under industrial conditions using the available data, the descriptive statistics of which is presented in Table I. The assembly of 659 input and output samples was divided into two groups. The first group consisted of 458 (70 %) randomly selected samples, which was used for training the network, while the second group consisted of the 201 (30 %) remaining data, which was used for testing the network.

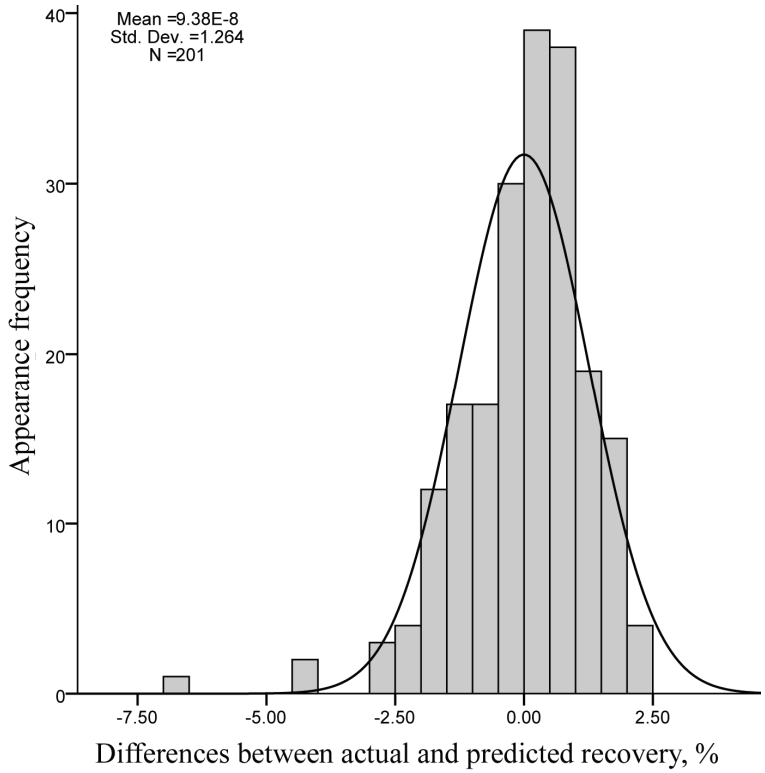

Fig. 2. Distribution of the differences between the actual Al_2O_3 recovery and the Al_2O_3 recovery estimated from Eq. (2) for 659 samples.

For the development of a relational ANN configuration, the previously defined input variables X_1–X_{11} and the output variable Y (described in the previous text), were used as the elements of the network architecture, Fig. 3.

As previously discussed, the ANN presented in Fig. 3 consists of three layers: the input, output and hidden layers. The neurons of the input layer present the information on the process input variables – X_i (independent variables), while the only neuron in the output layer generates the output information – the process quality indicator – Y (dependent variable). The methodology of choosing the appropriate number of neurons in the hidden layer as well as the procedure of the back-propagation learning algorithm is described in detail in the literature.[17]

In the phase of the network training, the necessary number of iterations was performed until the error between the measured output of the bauxite leaching process, Al_2O_3 leaching recovery, Y and the calculated values did not minimize and remained constant. The obtained results from the training stage could be evaluated by comparison of the calculated values Y with the measured ones. The obtained coefficient of the determination ($R^2 = 0.773$) show a large degree of fit-

ting among calculated and measured values, obtained during the training phase and could be used in the subsequent testing and validation.

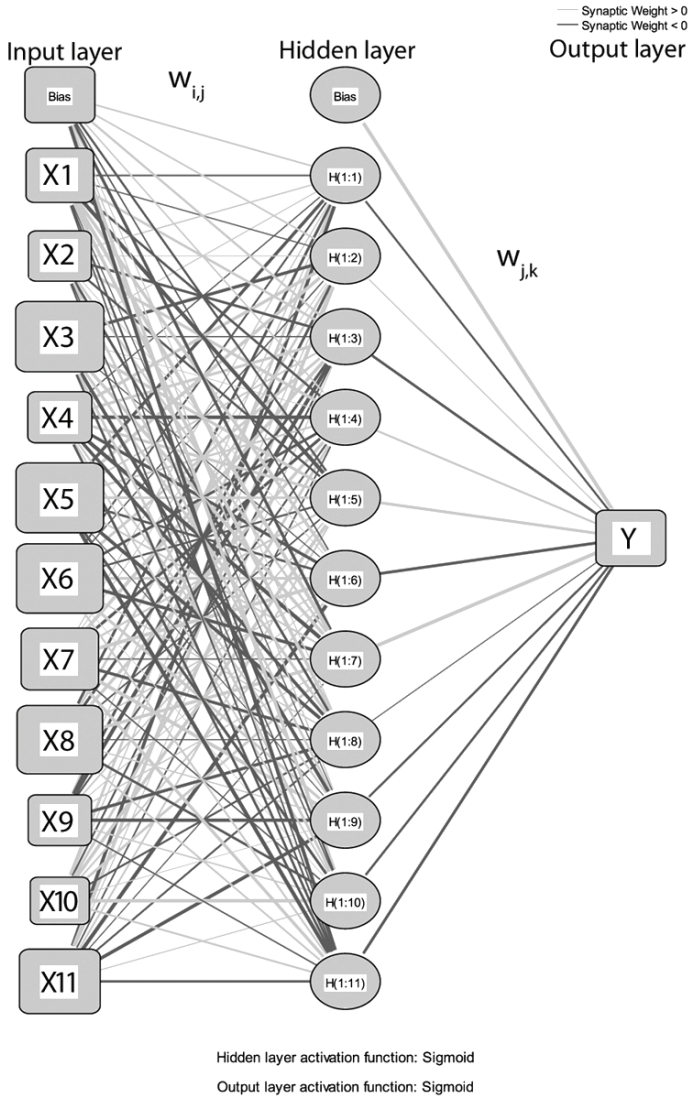

Fig. 3. The ANN architecture for the determination of the Al_2O_3 degree of recovery during bauxite leaching.

The test set (total 201 vectors), which examines the fidelity of the model, showed that the model could be used to estimate the leaching recovery quite satisfactorily. The value of the determination coefficient (R^2) for the test set was to some extent smaller 0.723 (Fig. 4). The differences of –3.20 to 2.25, between

predicted and the actually determined Al_2O_3 recovery (Fig.5), which were calculated by the (X_1–X_{11}) input sets in the ANN model, are proof that the listed variables could be considered as reliable inputs for the prediction of the Al_2O_3 recovery in the Birač Alumina Plant.

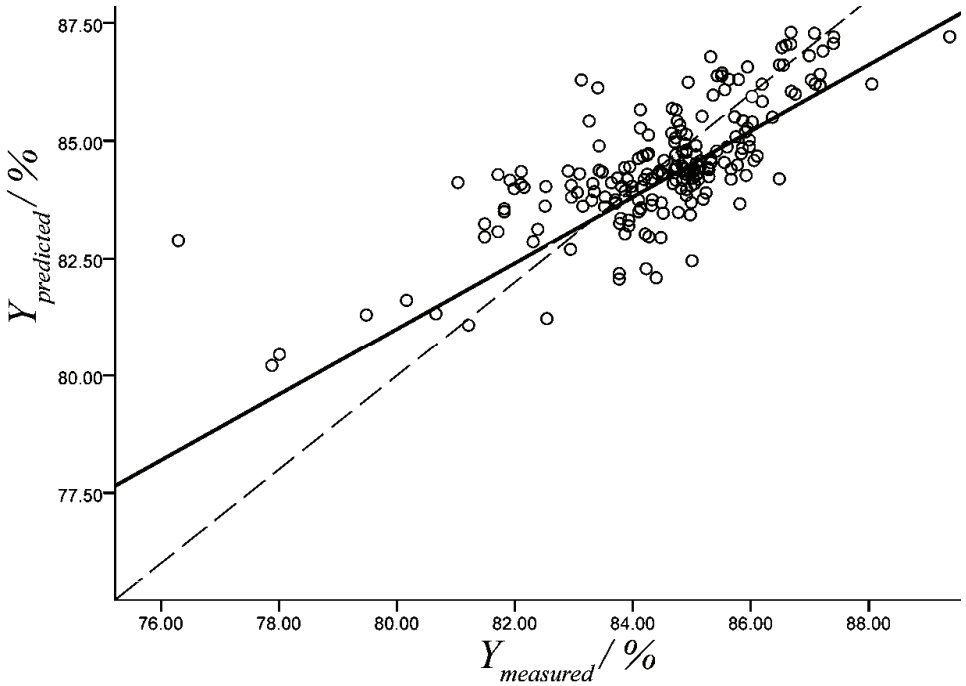

Fig. 4. The recovery predicted by neural network *vs*. the actual measured Al_2O_3 recovery (- - - = ideal position; ─ = regression lines; o = values calculated using the ANN model in the testing stage).

CONCLUSIONS

Values of the correlation analysis of the degree of Al_2O_3 recovery from the leaching of boehmitic bauxite under industrial conditions in the factory Birač, Zvornik (Bosnia and Herzegovina) were determined using the MLRA and ANN methodologies. The values of the coefficient of determination (R^2) were 0.463 and 0.723, respectively. These results indicated a highly acceptable degree of fitting of the dependence $Y = f(X_1–X_{11})$ obtained using ANN procedure as part of the SPSS software application, version 18 (PASW Statistics).[22]

The ANN procedure predicted the Al_2O_3 leaching recovery with good accuracy; it achieved a determination coefficient of R^2 = 0.723 and differences of –3.20 to 2.25 from actual determined recoveries in the plant. The selected ANN structure consisted of 458 (70 %) samples for training and 201 (30 %) for testing.

The defined elements of the ANN structure could be applied generally to conditions in any factory that employs the Bayer technology for alumina production.

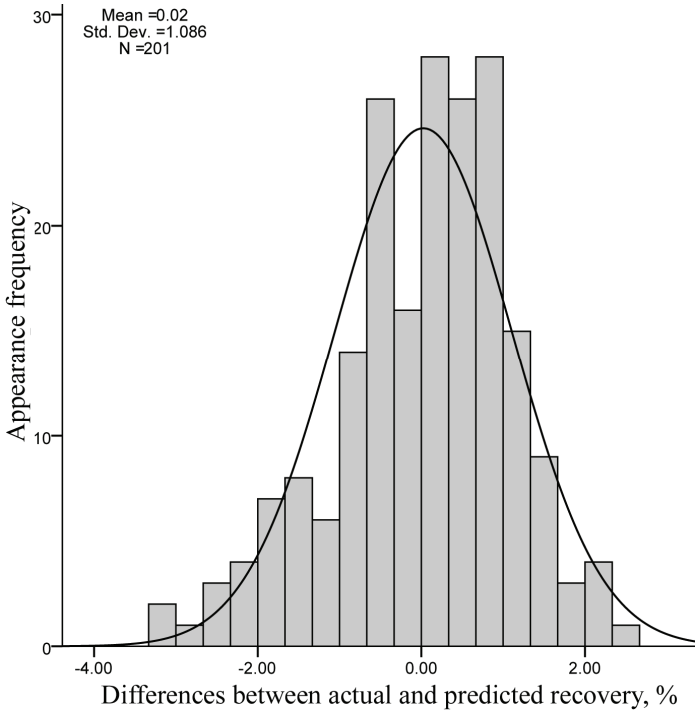

Fig. 5. Distribution of the differences between the actual Al_2O_3 recovery and that estimated by the neural network.

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И З В О Д

ПРЕДВИЂАЊЕ ЕКСТРАКЦИЈЕ АЛУМИНИЈУМА ИЗ БОКСИТА У БАЈЕРОВОМ ПРОЦЕСУ ПРИМЕНОМ ВЕШТАЧКИХ НЕУРОНСКИХ МРЕЖА

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Овај рад представља резултате статистичког моделовања процеса лужења боксита као дела Бајеровог процеса производње алуминијума. Засновано на подацима, сакупљеним током периода 2008–2009 (659 дана), који потичу из индустријске производње у фабрици алуминијума Бирач – Зворник (Босна и Херцеговина), извршено је статистичко моделовање наведеног процеса. Зависна промењива, чије одређивање је и основни циљ процедуре моделовања, представља степен издвајања Al_2O_3 из бемитног боксита током процеса лужења. Статистички модел је добијен као покушај да се дефинише зависност

степена издвајања Al_2O_3 од улазних промењивих процеса лужења: састав боксита, састав раствора натријум-алумината као и каустичног модула раствора пре и након процеса лужења. Као алати статистичког моделовања коришћени су вишеструка линеарна регресиона анализа (MLRA) и вештачне неуронске мреже (ANN). Ниво фитовања, добијен употребом MLRA, износио је $R^2 = 0{,}463$, док је ANN резултовала са $R^2 = 0{,}723$. На овај начин, модел дефинисан употребом ANN методологије, може се користити за ефикасно предвиђање нивоа издвајања Al_2O_3 у функцији процесних промењивих, у условима индустријске производње у фабрици Бирач – Зворник. Предложени модел, такође има и универзални карактер и као такав је примењив у другим фабрикама које примењују Бајерову технологију за производњу алуминијума.

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