Mathematical Modelling to Control the Chemical Composition of Blast Furnace Slag Using Artificial Neural Networks and Empirical Correlation

Wandercleiton Cardoso 1, Danielle Barros 2, Raphael Baptista 3, Renzo di Felice 1

1 Chemical Engineering at Genoa University, Liguria, Italy
2 Civil Engineering at UFRGS, Porto Alegre/RS, Brazil
3 Materials Engineering at Federal Institute of Espirito Santo, Brazil
wandercleiton.cardoso@dicca.unige.it

Abstract. Portland cement additions have been used for many years with the main objective of reducing the amount of clinker. Among the additions, blast furnace slag, resulting from the production of pig iron, that is, reusing this by-product, reduces the emission of carbon dioxide as well as decreases the exploitation of natural limestone and clay reserves, which are raw materials for Portland clinker. In order to reduce these emissions and increase the availability of raw materials, research has been directed to study clinker-free binders, as is the case with activated alkali cements and supersulfated cements. In this way, alkali-activated cements can only involve the reuse of industry by-products and do not require the calcination of the raw material, thus reducing the emission of polluting gases into the atmosphere. Supersulfated cement are composed of up to 90% blast furnace slag, in addition to 10 to 20% calcium sulfate. One of the most important characteristics of blast furnace slag is the ratio of the content of CaO and SiO₂, also known as the simplified basicity index (B2). This paper proposes the mathematical modeling of an artificial neural network to predict the final chemical composition of the blast furnace slag to be produced based on the operational parameters of the blast furnace aiming its use in the production of special cements such as alkali-activated cements and supersulfated cements. The high values of (R) associated with low values (RMSE) show the good statistical performance of ANN demonstrating that the mathematical model is efficient to carry out the forecast of the production of blast furnace slag.

1. Introduction

Portland cement production is aggressive to the environment, consuming large amounts of limestone and energy for burning and crushing clinker and, mainly, generates large amounts of carbon dioxide. Portland cement additions have been used for many years with the main objective of reducing the amount of clinker. Among the additions, blast furnace slag, resulting from the production of pig iron, that is, reusing this by-product, reduces the emission of carbon dioxide as well as decreases the exploitation of natural limestone and clay reserves, which are raw materials for Portland clinker.

However, during the production of pig iron the main objective is to adjust the chemical composition of the material produced (pig iron) to be sent to the next stage for steel production, that is, the blast furnace slag is a by-product that doesn’t exist rigorously of chemical composition control. In order to reduce these emissions and increase the availability of raw materials, research has been
directed to study clinker-free binders, as is the case with activated alkali cements and supersulfated cements.

Activated alkali cements are produced by mixing products composed of alumina, calcium oxides and silicates, such as blast furnace slag and an alkaline activating solution, usually metal hydroxides, carbonates or silicates.

Due mainly to the chemical composition of the activated raw material, the activated alkali cement can present very different characteristics and properties. For example, a low calcium content provides, as a reaction product, a gel composed of hydrated aluminates and silicates. However, when the calcium content is high, it provides the formation of hydrated calcium silicate, with a moderate degree of Al substitution and low Ca/Si ratio, C-(A)-S-H, which is amorphous to partially crystalline.

In this way, alkali-activated cements can only involve the reuse of industry by-products and do not require the calcination of the raw material, thus reducing the emission of polluting gases into the atmosphere. The use of blast furnace slag has an advantage in its application due to its chemical composition similar to that of Portland cement. In addition, due to the high pig iron production, the slag is generated on a large scale, which makes its consumption necessary.

The reactivity of the slag depends on its properties, that is, chemical composition, fineness, raw material used, method and cooling rate. Fineness is a conditioning factor for reactivity, since it is known that the greater its specific surface, the greater the contact with the activator, which is responsible for the initial activation and reaction.

The detailed chemistry of alkali activation is still the subject of much debate in the scientific literature; because for the manufacture of alkali-activated cements, the quality of the blast furnace slag is a determining factor in the final quality of the cement produced. Blast furnace slag with higher amounts of MgO provided an increase in the development of compressive strength and hydrotalcite formation (Mg$^{2+}$xAl$^{3+}$y(OH)$^2$ (x+y)(CO$_3$)$^2$−y/2mH$_2$O), in addition to a decrease in porosity in the microstructure.

However, a greater amount of Al$_2$O$_3$ in the raw material decreases the rate of heat released in the initial hydration of the slag, but has no significant influence on the final strength of the mortars. In this context, we can also mention supersulfated cement composed of up to 90% blast furnace slag, in addition to 10 to 20% calcium sulfate, usually in anhydrous form (CaSO$_4$) and up to 5% alkaline activator, which can be clinker or Portland cement, or hydroxides. The chemical characteristics required for the slag, however, are not the same as those required for use in Portland Cement, requiring greater control of the produced slag.

The chemical composition of the slag is highly dependent on the type of fuel (mineral or vegetable coal) used in the production of pig iron. Its main compounds are silica oxide (SiO$_2$), aluminum oxide (Al$_2$O$_3$) and calcium oxide (CaO), in different proportions than Portland cement. SiO$_2$ and Al$_2$O$_3$ come from ore, while CaO comes from limestone used as a flux.

One of the most important characteristics of blast furnace slag is the ratio of the content of CaO and SiO$_2$, also known as the simplified basicity index (B2). The importance of CaO and its proportion is shown in the removal of sulfur from mineral coal. In blast furnaces with charcoal, its importance is secondary, since this type of fuel contains low sulfur content.

Therefore, slag obtained from blast furnaces with charcoal generally have a low CaO/SiO$_2$ ratio, thus being considered acidic. Slag from coke blast furnaces is considered basic, in view of the need for
a high percentage of CaO to remove the sulfur present. Commonly, the following relationship is found in the literature: (\(\text{CaO}/\text{SiO}_2 < 1\)) acid slag and (\(\text{CaO}/\text{SiO}_2 > 1\)) basic slag.

The effect of the physical and chemical variability of the slag, as well as the control of the chemical composition of the blast furnace slag produced has not been well understood, providing a strong impetus for research in this area. In this context, this paper proposes the mathematical modeling of an artificial neural network to predict the final chemical composition of the blast furnace slag to be produced based on the operational parameters of the blast furnace aiming its use in the production of special cements such as alkali-activated cements and supersulfated cements.

2. Materials and methods

2.1 Data collect

The data used comes from the operation of a blast furnace of a Brazilian steelmaker that has an average daily production of 7200 tonnes. The operational data correspond to 105 records (105 days of operation) related to the average daily values of operation of 18 input variables and 4 output variables.

In the period selected to carry out the data collection, the blast furnace operated without major operational variations, with central gear and with practically the same reactivity value as the coke. The input variables used in the model are shown in Table 1.

| Input data        | Unit | Mean    | Input data        | Unit | Mean    |
|-------------------|------|---------|-------------------|------|---------|
| Pellet            | kg/t | 754.4 ± 62.4 | Blowing flow     | Nm³/min | 6228.1 ± 587.2 |
| Sinter            | kg/t | 754.4 ± 50.2 | Coke ash content  | %     | 8.9 ± 0.9 |
| Iron ore          | kg/t | 37.1 ± 27.4 | Coke moisture     | %     | 3.9 ± 0.7 |
| Coke rate         | kg/t | 300.2 ± 26.4 | Nitrogen          | Nm³/t | 17.3 ± 11.1 |
| PCI rate          | kg/t | 198.1 ± 17.2 | Oxygen flow       | Nm³/t | 14371 ± 419 |
| Fuel rate         | kg/t | 498.3 ± 21.8 | Oxygen enrichment | %     | 4.1 ± 0.9 |
| Dolomite          | kg/t | 7.1 ± 4.6   | Flame temperature | °C    | 1203 ± 21  |
| Slag basicity (B2)| %    | 1.19 ± 0.03 | Airspeed tuyère   | m/s   | 221 ± 21   |
| Slag basicity (B4)| %    | 1.06 ± 0.03 | Permeability      | -     | 4.21 ± 0.21 |

The output variables analyzed in the mathematical model were the chemical compounds: aluminum oxide (\(\text{Al}_2\text{O}_3\)), calcium oxide (\(\text{CaO}\)), magnesium oxide (\(\text{MgO}\)) and silicon dioxide (\(\text{SiO}_2\)) which represent an approximate percentage of 98% of the chemical composition of the granulated blast furnace slag used in Portland cement production.

These elements are also used to calculate the binary (B2) and quaternary (B4) basicities of granulated blast furnace slag. Table 2 shows the descriptive statistics of the 105 records in the model.

| Variable                  | Minimum | Maximum | Mean  | Standard deviation |
|---------------------------|---------|---------|-------|--------------------|
| (%\(\text{SiO}_2\)) Silicon dioxide | 35.8    | 38.7    | 37.1  | 0.7                |
| (%\(\text{MgO}\)) Magnesium oxide   | 5.1     | 6.4     | 5.9   | 0.3                |
| (%\(\text{CaO}\)) Calcium oxide     | 43.1    | 45.3    | 44.5  | 0.5                |
| (%\(\text{Al}_2\text{O}_3\)) Aluminum oxide | 9.3     | 11.2    | 10.3  | 0.4                |
Cross-validation is a technique to assess the generalizability of a model, based on a set of data. This technique is widely used in problems where the purpose of modeling is prediction. We then try to estimate how accurate this model is in practice, that is, its performance for a new set of data.

The central concept of cross-validation techniques is the partitioning of the data set into mutually exclusive subsets, and later, the use of some of these subsets to estimate the model parameters (training data), with the remaining subsets (validation data test) used in the validation of the model.

To perform cross-validation of the neural network data, 30 more variables were selected in addition to the 105 initially selected.

2.2 Data normalization
The normalization of the input data was performed in the MINITAB software according to equations (1) and (2):

\[
delta_i = \frac{(max - min)}{(max _ {imput}_i - min _ {imput}_i)}
\]

\[
norm _ {imput}_i = min - (\delta_i \times min _ {imput}_i) + (\delta_i \times imput_i)
\]

The pre-processing step, which consists of normalizing the input variables, was used to match the order of magnitude of the input variables between 0 and 1, avoiding numerical problems during the training phase, in addition to improving the performance of the backpropagation training algorithm.

Considering the variety of characteristics of the data to be obtained, data standardization was performed aiming at optimization during modeling and the reduction of the convergence time of the model, due to its versatility and wide possibility of solving linear and non-linear problems with function linear rectified activation and supervised learning, using the backpropagation algorithm.

2.3 Artificial neural network configuration
Artificial neural networks are characterized as artificial intelligence techniques inspired by the structure of the human brain, simulating mathematical operations in computer systems in an efficient and simplified way.

Artificial neural networks perform three essential operations: learning and storing knowledge; application of knowledge acquired in solving proposed problems, in addition to acquiring new knowledge of constant learning.

The artificial neuron is the basic processing element of an ANN being formed by a set of input connections \((x_j)\), synaptic weights \((w_{kj})\), where, \((k)\) is the number of input neurons and \((j)\) corresponds to the input stimulus, and the bias \((b_k)\) which is a weighting parameter that increases or decreases the value of the linear combination of inputs of the neuron activation function \((f)\).

Figure 1 below illustrates the simplified model of an artificial neuron, where it presents a simplified model of an artificial neuron, where \((u_k)\) represents the linear combination of the input signals, and \((y_k)\) corresponds to the output value of the neuron.

Thus, the entry weighting process represents the learning rate acquired by an ANN. Weights are adjusted as the input data set is presented to the network. The supervised learning process in an ANN
is based on adjusting the synaptic weights so that the output value is as close as possible to the expected value.

![Figure 1. Architecture of an artificial neural network](image)

The activation function \( f \) aims to limit the input signals from the network to a specific range, normally varying between (0 and 1) or (-1 and 1), generating an output neuron from the values of input \( (x_1, x_2, x_j) \) of the neural network and the adjusted synaptic weights. The most used functions in engineering research are the linear, log-sigmoid and tan-sigmoid functions.

Artificial neural networks have unique and specific arrangements and characteristics that adjust to the type of problem to be solved, and may have a single hidden layer or in several layers. In architecture, multilayer perceptron (MLP), the artificial neural network is composed of multilayers with nonlinear activation of the sigmoidal type in hidden layers giving the network a genuinely nonlinear mathematical modeling.

In this type of architecture, the error \( e \) is not obtained simply through the difference between the desired output and the output calculated by the network because there are now intermediate layers.

The backpropagation algorithm is one of the most used in practical ANN applications, since it corrects the error of the intermediate layers each time, estimating the effect caused in the output layer error, using the descending gradient, that is, the error is, therefore, back-paid in the network correcting the synaptic weights of the hidden layers.

The Levenberg-Marquardt algorithm is an optimization of the backpropagation algorithm, using an iterative numerical optimization technique capable of locating the minimum of a function expressed as the sum of squares of other non-linear functions. The Levenberg-Marquardt back-up is an adaptive network that uses the Jacobian matrix for calculations that assume performance as an average or sum of squared error.

In this paper, the potential of ANN was investigated to estimate the quality of the chemical composition of the blast furnace slag used in cement production by developing different models of ANNs. The simulations in this study were performed in the MATLAB R2020b environment using the “nftool” toolbox and the Levenberg-Marquardt training algorithm.

The number of neurons used in the middle layer depends on the complexity of the problem to be modeled, making it difficult to estimate the number to be considered, which sometimes requires several attempts to obtain the ideal quantity, thus not having an exact solution, to determine the number of neurons in the middle layer.
The number of neurons in the middle layer is generally defined empirically and depends on several factors, such as: a) number of training examples; b) amount of noise in the examples; c) complexity of the function to be learned; d) statistical distribution of training data.

In most metallurgical problems, the architecture of an ANN is usually obtained by trial and error, however, a single hidden layer is sufficient to approximate any continuous function. The number of hidden neurons \( h \) in a single-layer network is a function of the number of input variables \( I \), as shown in Equation (3);

\[
H < (2I + 1)
\]

In this paper, neural networks were trained with a single hidden layer, with the number of neurons in the hidden layer varying (50, 60, 70, 100 and 200) and using 18 input variables with a function of log-sigmoid activation in the hidden layer and the linear function in the output layer.

The number of neurons used in the middle layer depends on the complexity of the problem to be modeled, making it difficult to estimate the number to be considered, which sometimes requires several attempts to obtain the optimum quantity, thus not having an exact solution, for determining the number of neurons in the middle layer.

3. Results and discussions

Developing a mathematical model capable of predicting the chemical composition of the slag to be produced in a blast furnace is not really easy, so choosing the best estimation method to be used suggests the use of statistical techniques to evaluate the different methods used in investigations involving the production process of a blast furnace. The artificial neural network uses 2 statistical criteria to assess the efficiency of the mathematical model: root mean square error (RMSE) and regression R Values (R).

The RMSE is the root mean square error of the difference between the estimated and measured values, which consequently attributes greater weight to the largest errors. Values close to zero indicate better model performance. The RMSE is calculated as follows in Equation (4);

\[
RMSE = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (C_{\text{neural}} - C_{\text{real}})^2}
\]

Where \( n \) is the number of observations, \( C_{\text{neural}} \) is the value calculated by the artificial neural network, and \( C_{\text{real}} \) is the value measured in a chemical laboratory.

The performance is also evaluated through the coefficient of correlation for equations (R). In general, \( R \) value has the objective of evaluating the relationship between two variables, from \( n \) observations of those variables, indicating how much the independent variable can be explained by the fixed variable. The coefficient of correlation for equations (R) is calculated as follows in Equation (5)

\[
R = \left[ \frac{\sum_{i=1}^{n} (C_{\text{neural}} - C_{\text{real}})^2}{\sum_{i=1}^{n} (C_{\text{real}} - \bar{C}_{\text{neural}})^2} \right]^{\frac{1}{2}}\left[ \frac{\sum_{i=1}^{n} (C_{\text{real}} - \bar{C}_{\text{real}})^2}{\sum_{i=1}^{n} (C_{\text{neural}} - \bar{C}_{\text{neural}})^2} \right]^{\frac{1}{2}}
\]
The statistical analysis was performed using the Minitab statistical software. Figures 2 to 5 show the measured values of the compounds aluminum oxide (Al$_2$O$_3$), magnesium oxide (MgO), calcium oxide (CaO) and silicon dioxide (SiO$_2$) analyzed in the laboratory (real) and the values calculated by the artificial neural network.

**Figure 2.** Behavior of the output variable aluminum oxide: laboratory (real) and ANN (neural)

**Figure 3.** Behavior of the output variable calcium oxide: laboratory (real) and ANN (neural)

**Figure 4.** Behavior of the output variable magnesium oxide: laboratory (real) and ANN (neural)
Figure 5. Behavior of the output variable magnesium oxide: laboratory (real) and ANN (neural)

In addition, in order to verify the model's functionality, hypothesis tests were done, which was performed with a 99% confidence interval to verify whether the real and neural sampling groups were identical. The real and neural sample groups of the variables aluminum oxide ($\text{Al}_2\text{O}_3$), magnesium oxide (MgO), calcium oxide (CaO) and silicon dioxide (SiO$_2$) do not have atypical data points (outlier) in both groups (real and neural) of the samples.

After analyzing the 4 groups ($\text{Al}_2\text{O}_3$, MgO, CaO and SiO$_2$) in the "Minitab® software”, we can conclude that the data (real and neural) provide sufficient evidence that the neural measurements don’t differ in the 4 groups.

In the industry, a very important control operational parameter is the slag basicity ($B_2$ and $B_4$), which are represented by the following Equations 6 and 7:

$$B_2 = \frac{\text{CaO}}{\text{SiO}_2}$$  \hspace{1cm} (6)

$$B_4 = \frac{\text{CaO} + \text{MgO}}{\text{SiO}_2 + \text{Al}_2\text{O}_3}$$  \hspace{1cm} (7)

The slag basicity is also an important control element, aiming to retain a greater amount of alkali in the slag, avoiding the formation of pellets and improving the hearth permeability of the reactor. The calculated ($B_2$) basicity of the real and neural sampling was 1.199 and 1.200, respectively, while the calculated ($B_4$) basicity was 1.061 and 1.052 for real and neural sampling respectively. The behavior of the slag basicity ($B_2$ and $B_4$) is shown in Figure 6.

Figure 6. Behavior of the slag basicity ($B_2$ and $B_4$): laboratory (real) and ANN (neural)
The slag basicity is also an important control element, aiming to retain a greater amount of alkali in the slag, avoiding the formation of pellets and improving the hearth permeability of the reactor. The calculated (B2) basicity of the real and neural sampling was 1.199 and 1.200, respectively, while the calculated (B4) basicity was 1.061 and 1.052 for real and neural sampling respectively. The behavior of the slag basicity (B2 and B4) is shown in Figure 5.

The high values of \( (R) \) associated with low values (RMSE) show the good statistical performance of ANN demonstrating that the mathematical model is efficient to carry out the forecast of the production of blast furnace slag. The results obtained in conjunction with the cross-validation of the data prove the ANN's capacity to generalize the acquired knowledge. The final values of the mathematical correlation \( (R) \) and the root mean square error (RMSE) are shown in Figure 7.

![Figure 7. Final result of training the artificial neural network](image)

It’s evident that this research doesn’t intend to exhaust the study about the importance of the production of blast furnace slag, however, it’s common knowledge that the discussion of this argument, blast furnace slag, is a relevant factor that influences the consumption of fuels, pig iron productivity and quality, gas flow by the charge, calculation of the melting bed, operational stability and thermal losses.

The product of blast furnace slag impacts the total fixed carbon consumption; therefore, the composition of the slag must be controlled so that it’s sufficiently fluid to flow out of the blast furnace and easily separate from the pig iron in conditions suitable to promote the removal alkali.

The chemical composition and temperature of the slag depends on the thermodynamic conditions in force in the blast furnace production area, therefore, the quality of the coal fines injected in blast furnaces that deserves mention is the ash content or more precisely the amount of aggregated impurities. These impurities, in large quantities, harm the blast furnaces, as they interfere with the rate of coal substitution, the slag volume, the energy consumption and the viscosity and fluidity of the slag produced.

The levels of sulphur and phosphorus in the coke charged are further important elements in terms of blast furnace operations and productivity. Approximately 60-80% of the sulphur in the coal remains in the coke, the rest being expelled during coke making. Costly desulphurisation practices prior to steel making will be necessary for hot metal high in sulphur. Phosphorus is also a detrimental contaminant in steel and as such requires removal either prior to or during steel making. However,
both alternatives are expensive. Greater than 90% of the phosphorus charged to the blast furnace reports to the hot metal.

The ash from the injection of coal fines reacts with 15 to 20% of the dripping slag produced in the blast furnace, causing an increase in viscosity and melting temperature. Improving the performance of the reactor, it’s suggested to increase the oxygen enrichment rate by stabilizing the flame temperature and improving the permeability of the blast furnace, and also to reduce the pouring temperature of pig iron and slag and also the silicon content. These two suggestions will work by reducing the total carbon consumption of the process and improving productivity and producing a slag of better quality.

4. Conclusions
The model of artificial neural networks, in this work, was developed to predict the production and quality control of blast furnace slag aiming at high quality slag for the production of alkali and Supersulfated cements;

Obtaining liquid iron and slag in stable conditions is a very hard task, because the blast furnace is a complex machine, conjugating several sub-processes. Some of them are continuous, some transient, occurring in the same reactor and still subject to oscillations in raw material composition;

The analysis of alternative raw materials or practice standards can be held also with the support of the model as long as the variables are kept inside the operating range studied; It could be concluded that the neural model is a relevant tool to support an iron Blast Furnace operation since some corrections and retraining are carefully carried out by expert human operators in a systematic way. These procedures are crucial for adopting the neural model as a standard operating practice.

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