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Sulphide Capacity Prediction of Molten Slags by Using a Neural Network Approach

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In the present study, the neural network approach was applied for the estimation of sulfide capacities (Cs) in binary and multi-component melts at different temperatures. The calculated results obtained using neural network computation were plotted against the experimental values for comparison comparative purposes. Besides, iso-sulfide capacity contours on liquid regions of some ternary melt phase diagrams were generated and plotted by using neural network model results. It was found that calculated results obtained through neural network computation agree very well with the experimental results and more precise than those of some models.

KEY WORDS: sulfide capacities; molten melts; neural network computation; estimation.

1. Introduction

Sulfide capacity (Cs) modeling is of great interest in metallurgical engineering, since its aim is to predict the desulfurization power of slags which directly affect the metal quality during the production stage. There are many empirical, semi-empirical and theoretical sulfide capacity models in the literature.^{1–5)} For example, Reddy and Blander (RB) developed a model, a priori, based on a simple solution model and on knowledge of the chemical and solution properties of sulfides and oxides.^{2,3)} Moretti and Ottonello proposed a model for calculating the sulfide capacity of simple and complex silicate melts using different polymeric solution theory.⁵⁾ Both models are purely theoretical and do not require any experimental data. Sosinsky and Somerville first proposed a Cs prediction model using optical basicity concept which is related with the ratio of electron donor power of the oxides in the glass to the electron donor power of “free” oxide ion defined by Duffy and Ingram.^{1,6)} KTH model is based on an optimization of the experimentally determined Cs values of simple systems in order to obtain those of multicomponent slags.⁷⁾ However, above models are in good agreement with experimental results only within a certain composition and/or temperature range due to either lack of thermodynamic data or complexity of the mathematical equations which are required in the model. There are also a number of derived equations mostly based on simple regressions of experimentally obtained Cs values of simple as well as complex melt systems.^{8–10)} However, these equations are not flexible enough to produce reliable data far from the experimental zone, such as for predicting Cs iso-contours in complex systems. In the present study, an advanced empirical approach, neural network computation will be described briefly and applied for

the estimation of sulfide capacities (Cs) in multicomponent slags and flux systems which consist of SiO₂, CaO, Al₂O₃, FeO, MgO, MnO, Na₂O, CaF₂, CaCl₂ and TiO₂ at different temperatures.

2. Neural Network Computation for Cs Calculations

Neural network computation can be defined as a processing method which imitates the features of the human brain.^{11–13)} The human brain contains approximately 10 billion nerve cells (neurons). A biological neuron which is a functioning unit of the nervous system consists of dendrite, soma, axon and synapse (**Fig. 1(a)**). A simplified artificial equivalent of a biological neuron is shown in **Fig. 1(b)**. Neurons communicate *via* input signals. A neuron accepts inputs associated with different weights from multiple neurons. The summation of the inputs (intensity) is multiplied by their associated weight. When the intensity of the signal is high enough to pass over a certain critical value (threshold), then an output signal is transmitted through axon and synapse to the next neuron. The state that the intensity of signal exceeds the threshold is called “ignition” and it can be expressed in a sigmoid function shown in Eq. (1) in the neural network concept.

$$y = f(x) = 1/[1 + \exp(-\eta \cdot x)] \dots\dots\dots(1)$$

Here, x is an input value and y is the output. η is a coefficient which determines the shape of the sigmoid curve. **Figure 2** represents a schematic diagram of the back propagation method in a three layers-type neural network computation which consists of an input layer, a middle (hidden) layer and an output layer. The units in the middle layer are connected with the input and output units. However, there are no connections within a layer. The final result “ y ” in the

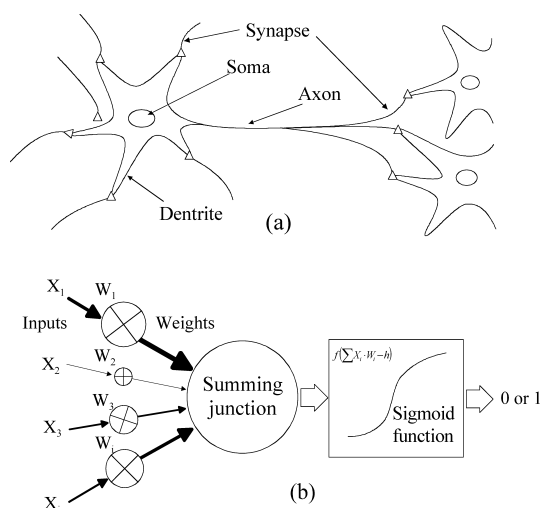


Fig. 1. Schematic diagrams of a) a biological neuron and b) a simple artificial neuron.

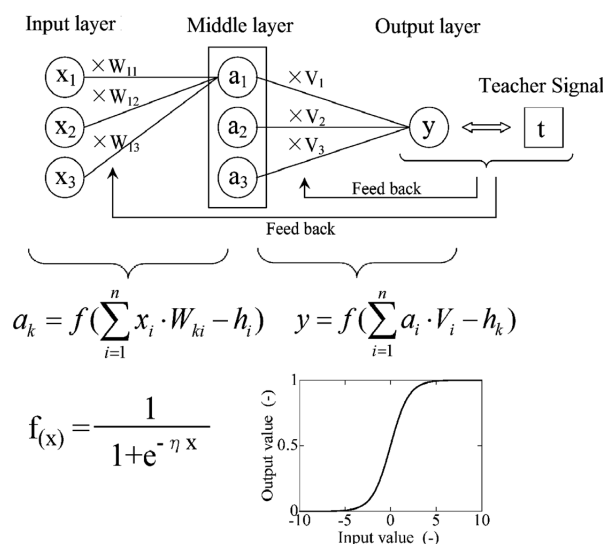


Fig. 2. Schematic diagram of the back-propagation approach in 3 layers-type neural network structure with 3 units in the input layer and 3 units in the middle layer.

output layer is expressed by using the above sigmoid function as follows:

$$a_k = f\left(\sum x_i \cdot W_{ki} - h_i\right) \dots\dots\dots(2)$$

$$y = f\left(\sum a_k \cdot V_k - h_k\right) \dots\dots\dots(3)$$

where x_i is an input value of unit i in the input layer, W_{ki} is a connection weight between unit i in the input layer and unit k in the middle layer, h_i is a critical value for unit i . y is the final output, V_k is a connection weight between unit k in the middle layer and the final output, h_k is a critical value for unit k . After values are applied to the units in the input layer, signals propagate through the middle layer to the output layer. Each link between neurons contains a unique weight value. A comparison is made between output values and the teaching values. The errors are calculated for each output unit and then propagated backwards through the network to correct the connection weights and the critical values in each unit. This “learning” process is repeated until the overall error value drops to acceptable levels.

Some studies exist on the estimation of some physical properties of molten slags by neural network approach in the literature. Tanaka *et al.* predicted viscosity and solidification temperature of mold fluxes in multi-component systems using neural network computing.^{14,15)} Nakamoto *et al.* applied this approach to estimate the surface tension in ternary silicate melts.¹⁶⁾ Both studies also discussed on the criteria for designing the number of units in the middle layer in order to obtain optimum results.

In the present study, the computation was carried out by using SlagVis software. The SlagVis was designed by Research Center of Computational Mechanics Inc., Osaka University, and Sumitomo Metal Industries Ltd. to estimate first the physical properties of multi-component slags by neural network model.^{14–17)} Since the calculation method is the same as that for those physical properties, the program was also found applicable for the Cs predictions. The following initial values were selected in the software for all calculations; number of middle unit 5, maximum iteration 1 000 000, learning rate 1.5 which is a constant used in artificial neural network learning algorithms to affect the speed of learning, and target relative error 0.02. These values, which were selected as the optimal parameters for the present study, were obtained by trial and error until an adequate match was achieved between experimental and calculated Cs values. In calculations, input values were mole fraction of the components, whereas the experimental Cs results in logarithmic scale were served as teaching values that provide feedback. Then, the sulfide capacity predictions of molten slags were calculated using this network.

3. Results and Discussions

Since the neural network model is based on an empirical approach, the consistency of the experimentally determined Cs values to be used as teaching values in the model is very essential.

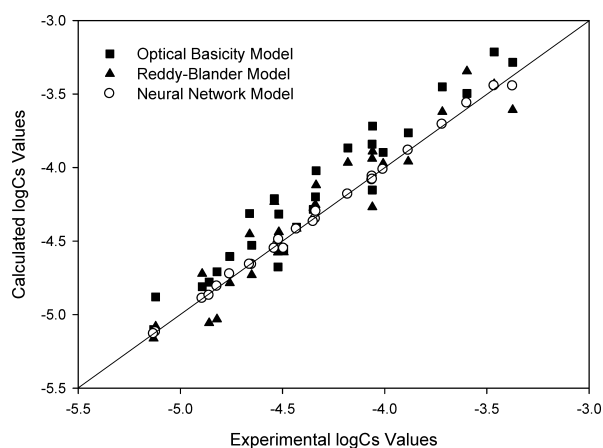
The main difficulties in the present study were the lack of experimental data and/or inconsistent Cs values of some similar slag compositions carried out by different authors. For example, Cs values of MgO–SiO₂ slags at 1923 K found by Sharma and Richardson¹⁸⁾ (7 data points) are not only almost three times higher but also much more scattered than the findings of Nzotta *et al.*¹⁹⁾ (12 data points) which were obtained more than 30 years later. In that case, if both data are used in the same neural network calculations as teaching value, then the predicted results would inevitably be a failure. When the above concerns were taken into account, where possible, comparatively reliable and/or new data obtained with advanced measuring techniques were selected from the literature in order to evaluate much consistent results in the present calculations.

The experimental data collected from the literature was used for the present neural network estimation of sulfide capacities of binary and multi-component melts at different temperatures and listed in Table 1.

Initially, neural network estimation method was compared with Sossinky and Sommerville’s optical basicity equation and Reddy–Blander model which are often used for predicting Cs values in silicate slag systems. For this aim, experimental Cs values of CaO–MgO–SiO₂ slags at

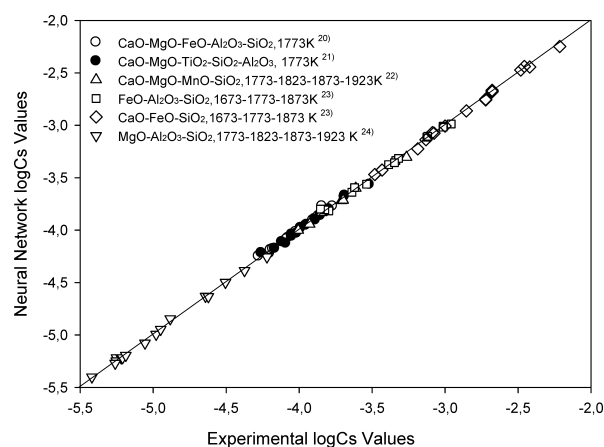
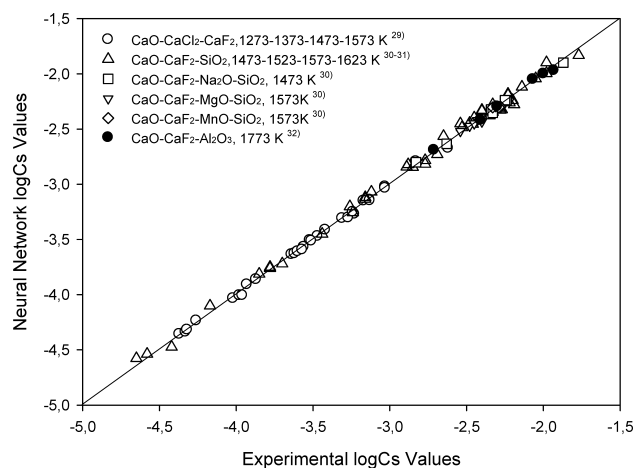
Table 1. Experimental data used for the Neural Network Cs estimation.

Slag System	Temperature, K	Ref.
CaO-MgO-FeO-Al ₂ O ₃ -SiO ₂	1773	20
CaO-MgO-TiO ₂ -Al ₂ O ₃ -SiO ₂	1773	21
CaO-MgO-MnO-SiO ₂	1773, 1823, 1873, 1923	22
CaO-MgO-SiO ₂	1773, 1823, 1873	19
FeO-Al ₂ O ₃ -SiO ₂	1673, 1773, 1873	23
CaO-FeO-SiO ₂	1673, 1773, 1873	23
MgO-Al ₂ O ₃ -SiO ₂	1773, 1823, 1923	24
CaO-MgO-Al ₂ O ₃	1873	25
CaO-Al ₂ O ₃ -SiO ₂	1773	26
CaO-SiO ₂	1773	27
FeO-SiO ₂	1773	27
FeO-CaO	1773	27
FeO	1773	28
CaO-CaF ₂ -CaCl ₂	1273, 1373, 1423, 1473	29
CaO-CaF ₂ -SiO ₂	1473, 1523, 1573, 1623	30, 31
CaO-CaF ₂ -Na ₂ O-SiO ₂	1473	30
CaO-CaF ₂ -MgO-SiO ₂	1573	30
CaO-CaF ₂ -MnO-SiO ₂	1573	30
CaO-CaF ₂ -Al ₂ O ₃	1773	32
CaO-CaCl ₂	1773	33

**Fig. 3.** Comparison between experimental and calculated Cs values obtained by different models for CaO-MgO-SiO₂ slags at the temperatures of 1 723–1 873 K.

the temperatures of 1 723, 1 773, 1 823 and 1 873 K were taken from the study of Nzotta *et al.*¹⁹⁾ It is noted that for Cs calculation of each composition, whereas optical basicity model required mole fraction, temperature and theoretical or optimized optical basicity (Λ) values, these requirements were mole fraction, equilibrium constant, K_{eq} , activity of metal oxides, a_{MeO} , and temperature values for the Reddy-Blander model. However, only one calculation was carried out for neural network method to predict the whole Cs values just inserting mole fraction and temperature as input values and experimental Cs values as teaching values. As a result of comparison of the these models with experimental Cs values, it was shown in **Fig. 3** that better regression can be obtained with neural network approach compared to optical basicity and Reddy-Blander models.

Figures 4 and 5 represent the comparison between experimental and calculated values of multicomponent silicate melts and halide containing multi-component oxide melts, respectively. As seen in both figures, very good compliance between neural network predicted values and the experimental data points were found. Especially, in point of halide containing melts view, only an optical basicity model

**Fig. 4.** Comparison between experimental and calculated Cs values obtained by neural network computation for multicomponent silicate melts at different temperatures.**Fig. 5.** Comparison between experimental and calculated Cs values obtained by neural network computation for halide containing multicomponent melts at different temperatures.

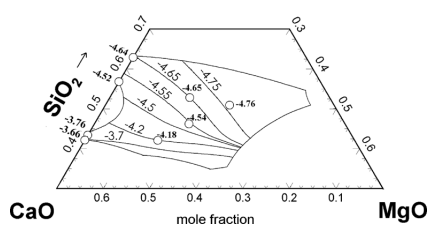
was tried for Cs prediction in the literature. However, the results were not in good agreement with the experimental data.³¹⁾ This situation makes the neural network approach a good engineering tool to estimate much reliable results.

Iso-sulfide capacities of the ternary slag systems for different compositions and temperatures were generated using the neural network model. Some experimental Cs values in Table 1, not only ternary but also of lower sub-systems neighbor to liquidus region were taken into account for each calculation. The experimental Cs values of the melt compositions which are not in liquid region were neglected to prevent erroneous Cs results. The iso-sulfide capacity contours were inserted to phase diagrams which were generated by FactSage 6.0 using “Phase Diagram” module with FToxide database.³⁴⁾

Experimental Cs values of CaO-SiO₂ and CaO-MgO-SiO₂ were used to generate iso-sulfide capacity contours in liquid region of the ternary melt at 1 773 K. As seen in **Fig. 6**, logarithmic scaled capacity contours are in good agreement with experimental data and vary between -3.7 and -4.75 . In order to perform more complicated regression example, CaO-FeO-SiO₂ melt at 1 773 K was chosen for

the prediction. For this aim, experimentally obtained Cs values of molten state pure FeO, CaO–SiO₂, FeO–SiO₂, CaO–FeO, and CaO–FeO–SiO₂ melts were used. **Figure 7** shows that an increase in the SiO₂ content results in a sharp decrease of the desulfurization power of slags and predicted iso-sulfide contour values within the large liquid region decreases from –1.8 to –4.5. Experimental sulfide capacity data of molten state CaO–CaCl₂ and CaO–CaCl₂–CaF₂ melts at 1 273 K were used as teaching value in order to evaluate iso-sulfide capacity contours on the ternary phase diagram. According to the calculations, the contours were found almost parallel to the CaCl₂–CaF₂ axis, since CaO is the only Cs determinant in this melt.

In the calculations, the number of parameters strongly depends on the component number of the slag. Temperature



4. Conclusions

In the present work, we applied the neural network calculations to the sulfide capacity predictions in multi-component melts. The computation results were found in good agreement with the experimental values. It was also constructed iso-sulfide counters on ternary phase diagrams establishing a link among experimental Cs values of molten ternary and lower sub-systems. It can be concluded that neural network based computation is a very useful technique for predicting Cs values in molten melts, but attention needs to be paid to the quantity and accuracy of the experimental data.

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