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DERIVATION OF $CaO-SiO_2-Al_2O_3$ SYSTEM SLAG VISCOSITY EQUATION BY GP

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Abstract

Slag viscosity is essential in high-temperature metallurgical processes. However, a slag viscosity model is difficult to exactly interpret as it has a strong nonlinear relation with its composition and temperature. In this paper, genetic programming (GP) was employed to derive a $CaO-SiO_2-Al_2O_3$ slag viscosity equation. The equation was automatically described as a simple algebraic equation with the basicity and content of Al_2O_3 and temperature. The average relative error between the values obtained by the equation and the experimental data used for its derivation was as low as 17.1%. Computer simulations were performed to evaluate the accuracy of the derived viscosity equation and were then compared with many experimental viscosities and calculated values of other researchers. Slag compositions and temperatures for simulation calculations were the experimental data which were not used for deriving a viscosity equation. The results showed that the viscosity equation was relatively exact. The viscosities of $CaO-SiO_2-Al_2O_3$ system slag could be simply and expediently predicted within the wide range of compositions and temperatures by using the derived viscosity equation.

Keywords: *Slag; Viscosity equation; Genetic programming; Prediction*

1. Introduction

Slag viscosity is a key property that determines the stability and productivity in the metallurgical furnace operation, and it is of great importance in understanding the fluid dynamic of molten slags and slag-metal reaction kinetics during the pyro-metallurgy process. Therefore, fundamental understanding of slag viscosity and the various factors that influence this property have significant theoretical and practical meaning. Up to now, there have been many experiments to measure, and models to estimate slag viscosities [3-11]. Most models are based on the Arrhenius equation, Weymann-Frenkel equation, Eyring equation, Vogel-Fulcher-Tamman equation, and empirical relation equation etc [10]. These equations have constants depending on the slag's structure activation energy for viscous flow and temperature.

Viscosity models can be classified into explicit models, structural models, and miscellaneous models according to the way in which they are connected to description of the slag structure [3]. Explicit models only distinguish compounds according to their network of modifying or amphoteric character. Various parameters of the explicit model are explicit functions of the mole fractions of these various compounds. Structural models use the same concepts as the thermodynamic quasi-chemical models of slag. These thermodynamic models start with the structural description of the slags given before and develops a description expression of the Gibbs free energy through the quasi-chemical approach proposed by Guggenheim [3]. Therefore, all the models in this category refer to one of the two thermodynamic models of slags based on cell model or the modified quasi-chemical model. Factsage (thermodynamic calculation software) viscosity module predicts well the viscosity of slag within the experimental error limits because the structural change in the multicomponent slag is automatically counted in the viscosity model by calculating the structural units (SUs) from the modified quasi-chemical model [3]. The Miscellaneous models such as Hanao et al.'s network model [12] do not take into account the structure of the slags.

Mills [13] and Pal's [14] models are based on structurally related optical basicity. Using these models, viscosities can be predicted in a limited composition range of slag, and the errors are relatively high. Slag viscosity is strongly related to its melting structure; it is difficult to entirely interpret its structural features. Anyhow, it is an obvious fact that slag viscosity can be described as a function of slag composition and temperature.

Slag viscosity equation describes that only slag

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composition and temperature are to be derived by genetic programming (GP). GP is a technique for exploring the unknown function relationships of objects by imitating the principle of Darwinian natural evolution. A neural network (NN) builds an approximated function that matches inputs to target outputs. GP seem to have the same functionality as NN does.

However, GP can dynamically explore complex functions describing the system so that its fitting error is minimum, while NN just optimizes the coefficients in the given functions to learn the matching between inputs and outputs. In a word, GP is a structure optimization approach, while NN is a parameter optimization approach. GP is an evolutionary algorithm-based methodology inspired by biological evolution to find computer programs that perform a user-defined task, which can create model with minimal human efforts and background knowledge [15].

Traditional modeling techniques such as nonlinear regression models typically require the model to have a generic structure substantially specified in advance, like the thermodynamic quasi-chemical model of slag. Other techniques such as NN and autoregressive models are predictive of unseen data, but can be difficult to interpret [16, 17].

In contrast, GP places minimal constraints on model structure, requires little or no domain knowledge, and is capable of automatically inferring parsimonious explanatory models like those that would ordinarily be hand-crafted from first principles by a human expert [18, 27].

GP has been successfully applied in many scientific fields; but no report has yet been made of applying GP in the metallurgical field [19-27]. In particular, viscosity of metallurgical slag is difficult to interpret because it has a strong nonlinear relation with composition and temperature of slag. However, Using GP, the viscosity equation can simply be derived with no need of interpretation of the structure of slag. The viscosities of $CaO\text{-}SiO_2\text{-}Al_2O_3$ system within the wide composition and temperature range are listed in references [1-4, 6].

In this work, viscosity equation of $CaO\text{-}SiO_2\text{-}Al_2O_3$ system slag was derived using the Kozanevitch's experimental data [1] and GP. Then, in order to evaluate its accuracy, the results were compared with the experimental data and model calculation results in previous research [1-4, 6].

2. Model

2.1. General Model Description

For derivation of slag viscosity equation, firstly the GP was discussed. Based on the collected experimental data, GP automatically generates the models without any pre-definition of the structure of models, automatically evolves both the structure and parameters of models and finally obtains the rational model.

GP can be divided into preparatory steps and executional steps [15, 24]. The preparatory phase contains the following five major steps:

a) The set of terminals for each branch in tree,

b) Selection of primitive functions for creating tree,

c) The set of the fitness function,

d) The set of parameters for the run, and

e) The set of termination criterion and display of the result.

The step a) and step b) specify the ingredients for composing the individual trees. Individual functions are decoded in the form of tree in order to perform the genetic operation [15]. A run of GP is a competitive search among a diverse population of functions describing the system which composed of selected primitive functions and terminals.

For the problem of estimating slag viscosity equation, the primitive functions may consist of the '+', '−', '×', '/', '^', 'log', 'exp', and 'sin' functions. The independent variables in terminals are composition and temperature of slag.

The third step concerns the fitness calculation for the individual functions in population. The fitness calculation is used for estimating the performance of individual functions. Fitness is the driving force of Darwinian natural selection [24]. In this work, the goal is to get GP to automatically derivate a slag viscosity equation, and the fitness function is the mechanism for telling GP to derivate viscosity equation with composition and temperature of slag.

The first two steps a) and b) define the exploration space and the third step c) implicitly specifies the desired exploration target. The steps d) and e) are administrative. The fourth step d) defines the control parameters. It consists of population size, maximum depth of individual tree, and probabilities of genetic operations [15]. The fifth step e) defines the termination criterion and the output of the final result. The termination criterion in step e) may include a maximum generation number as well as success of exploration [16]. After the steps mentioned above are performed, the run of GP can be started.

GP starts with a population of randomly generated individual trees which consist of terminals and primitive functions (preparatory steps a) and b)). GP iteratively transforms a population of individual trees into a new generation of the population by randomly selected genetic operations [15]. Individuals are probabilistically selected from the population based on their fitness valves (preparatory step c)), GP executes reproduction, crossover, and mutation operations, resulting in creating a new population.

The executional steps of GP are as follows [24]:

1. Randomly create an initial population of individual trees that consists of the mathematical operators, primitive functions, and terminals (independent variables and constants).

2. Iteratively execute the following sub-steps (a-c) on the population until the termination criterion mentioned in preparatory step e) is satisfied.

(a) Calculate the fitness values of each individual functions in the population by using the fitness function mentioned in preparatory step c).

(b) Select one or two individual trees from the population with a probability based on fitness values to take part in the genetic operations (sub-step (c)).

(c) Create new individual(s) for the new population by performing the following genetic operations.

•Reproduction: Copy the selected individuals in old population into the new population.

•Crossover: Create two new offspring for the new population by swapping randomly chosen sub-trees from two selected individuals in the old population.

•Mutation: Create one new offspring for the new population by randomly mutating a randomly chosen sub-tree in one selected individual in the old population.

3. When the termination criterion is satisfied, the best individual in the population is outputted.

Figure 1 shows individual tree generated by random choices of the primitive functions and terminals. Mathematical expressions of individual trees are described as following:

$Individual_1 = A + t^w - C$

$Individual_2 = C - \exp(A/w_1) + C \cdot A/(t + w_2)$

The primitive functions of $Individual_1$ are '−', '+', and '^', terminals are C, A, t, w. C, A, t: independent variables, w: constant;

A maximum depth of tree is related to complexity of mode [15], and each individual in the population is measured or compared in terms of how well it performs the task at hand (using the fitness value provided in the third preparatory step). The creation of the initial population is a blind random search in the search space for modeling the system, and fitness of most of individuals are very poor, while some individuals in the population are fitter than others. Differences of fitness value provide a criterion to decide future exploring direction.

The crossover operation is exemplified in Figure 2. Mathematical expressions of parent in the old population and offspring are as following:

$Parent_1 = (C \cdot w - A) \cdot (C/t)$,

$Parent_2 = \exp(\cos(A)) + w - C + A$

$Offspring_1 = (C + A) \cdot (C/t)$,

$Offspring_2 = \exp(\cos(A)) + w_1 - (C \cdot w_2 - A)$

Traditional mutation consists of randomly selecting a mutation point in a tree and substituting the subtree rooted there with a randomly generated subtree, as illustrated in Figure 3. Mathematical expressions of parent and offspring are as following:

$Parent = (w \cdot C + A) \cdot (w/t)$, $Offspring = (C + A) \cdot (w/t)$

Reproduction involves simple copy of certain individuals into the new population. After the genetic operations are performed in the old population (current), the new offspring population replaces the old population. This iterative processes of fitness calculation and performance of the genetic operations are repeated until termination criterion is satisfied.

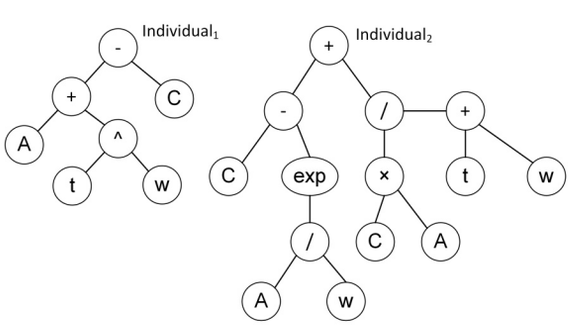

Figure 1. *Randomly generated individual trees*

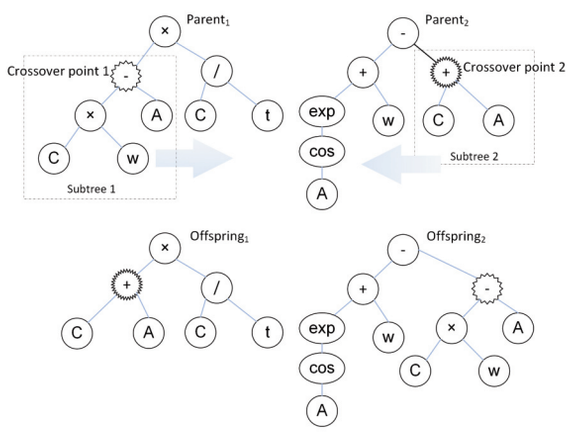

Figure 2. *Example of crossover operation*

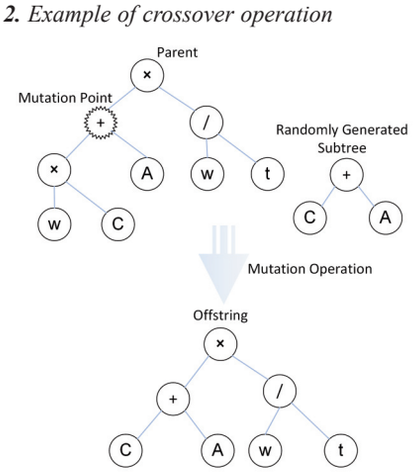

Figure 3. *Example of mutation operation*

2.2. Parameters for CaO-SiO_2-Al_2O_3 system Slag Viscosity Model

As mentioned above, viscosity of CaO-SiO_2-Al_2O_3 system slag is considered to be related to its composition and temperature. Therefore, the set of terminals was defined as following:

$$T = \{C , A , t , W\} \quad (1)$$

'C' and 'A' are related to CaO, Al_2O_3 content in slag, and 't' is related to temperature of slag. 'w' denotes constant. Independent variables were set as 'C', 'A' and 't' because content of SiO_2 in slag could be determined by CaO and Al_2O_3 contents.

The set of primitive functions was defined as following:

$$F = \{+, -, \times, /, \hat{}, \cos, \sin, \exp, \ln\} \quad (2)$$

Fitness of individual was calculated by following Eq (3).

$$fit = 1 / \left(1 + \sum_{i=1}^{n} \left(\eta_{i,calc} - \eta_{i,exp}\right)^2\right) \quad (3)$$

where n is total number of experimental data used in GP, $\eta_{i,Calc}$ is viscosity calculated by individual at i-th experimental point, and $\eta_{i,Expe}$ was viscosity measured at i-th experimental point. Fitness value was bigger for better individuals in population. As for the fitness measurement, it was very important to determine the constants contained in individual. The constants were determined by genetic algorithm and Levenberg-Marquardt method. Genetic algorithm was used for a global search in the wide range and Levenberg-Marquardt method [25] was used for a local search of constants. Penalty was assigned on the individuals that could not be evaluated to acceptable fitness. In other words, in the cases where fitness was not a real number or remarkably small. Individuals that did not include all independent variables were discarded. Initial individuals were created by using the ramped half-and-half generative method, and depth of an individual tree was specified in the range 3-7. Maximum depth of an individual was limited to 16 considering calculation time in evolutionary process. Individuals with more than 16 in maximum depth were discarded.

Selection of individuals was performed by employing the fitness-proportionate selection method. [15] In this case, selection probability is expressed by following Eq (4).

$$P_j = fit_j / \sum_{j=1}^{M} fit_j \quad (4)$$

where p_j is a selection probability of j-th individual, fit_j is a fitness of j-th individual, and M is an initial population number.

The numbers of initial population and final generation were set as 150 and 200, respectively. Initial data to run were Kozakevitch's experimental values [1]. In Kozakevitch's experimental data, contents of CaO, SiO_2 and Al_2O_3 all were in the range of 10-60%, temperature was in the range of 1500-1900°C.

Rational result was not obtained in run process, i.e. the problem of premature convergence appeared. Hornby proposed a novel approach for overcoming the problem of premature convergence, where the age was applied to individuals [26]. In this work, individual's age structure was applied, and the age of newly generated individual was defined as 0. Its age was increased by one after its reproduction. Each population did not involve any individual older than maximum age. Individuals above maximum age were discarded. Maximum age was set as 10. **Figure 4** shows the flowchart of GP for derivation of CaO-SiO_2-Al_2O_3 Slag viscosity equation. Program for derivation of CaO-SiO_2-Al_2O_3 system slag viscosity equation was implemented in Matlab code.

3. Result and discussion

3.1. CaO-SiO_2-Al_2O_3 Slag Viscosity Equation

Eq (5) showed a result obtained by run of program based on the model mentioned in **Section 2**. As expected, as a viscosity equation of slag, Eq (5) was described as form of exponential function, which involved not only general algebra operator but trigonometric function.

$$\begin{aligned}\eta = {} & 0.1 \cdot exp((C/t - A - 104.8481 - ((C + 170.9252 - \\ & (t + A + sin(t)) / (0.131 \cdot t \cdot exp(1.8097 - C - A)\hat{}0.4655 + \\ & C + A + C) \cdot t - A - t - 9.3319 \cdot C) \cdot \\ & (((8.077 \cdot C - 10.1089 \cdot t) \cdot C + 32.9407) / t + 36.5922) / \\ & ((0.2116 \cdot t \cdot exp(1.1714 - C - A)\hat{}1.0614 + A + A + C) \cdot \\ & (C - 372.9156) / (-2.837 + C)) + (C - 10.1034 + C - A / \\ & ((97.502 + t) \cdot (-35.5062)) + (1.6103 - A) / (5.3582 \cdot t \cdot \\ & exp(0.3063 - C - A)\hat{}(-15.9357) + C)) \cdot \begin{pmatrix} A - C - \\ C / t - 4.1632 \end{pmatrix} / \\ & (C + t)) \cdot (-584.11 - C) + 210.916 \cdot (551.7789 / t - 42.9855) \\ & + 165.0859 \cdot t) / 263.2423), \ Pa \cdot s \end{aligned} \quad (5)$$

where C=0.1·CaO; A=0.1·Al_2O_3; t=0.01·T; CaO: content of CaO in slag, wt %; Al_2O_3: content of Al_2O_3 in slag, wt %; T: absolute temperature, K;

Content of CaO was determined by slag basicity (R) and Al_2O_3 content, it was described by the following equation (7).

$$R = CaO/SiO_2 \quad (6)$$

$$CaO = R\,(100 - Al_2O_3)/(R+1), \% \quad (7)$$

$$C = 0.1 \cdot CaO \quad (8)$$

$A=0.1\cdot Al_2O_3$ (9)

$t=0.01\cdot T$ (10)

Eq (5) is described as a form of general algebraic equation, however, it is relatively complex. This fact implies that viscosity of slag has a strong nonlinear relation with composition and temperature of slag. Viscosity of slag can be simply and expediently calculated in a wide range of composition and temperature on $CaO\text{-}SiO_2\text{-}Al_2O_3$ system slag by using Eq (5-10).

3.2. Validation of Viscosity Equation

To evaluate accuracy of derived viscosity equation, the experimental values listed in references and calculation results by other viscosity models were compared.

Viscosities calculated by Eq (5) were compared with Kozanevitch's experimental values [1] and calculated results by Factsage7.0, which were showed in Figure 5. Basicity (R) and content of Al_2O_3 of Kozanevitch's slag [1] were in the range of 0.16-6 and 10-65%, respectively, and temperatures were in the range of 1500-1900°C. As shown in Figure 5, Viscosities calculated by Eq (5) were fairly fit with the experimental values and more accurate than calculated results by Factsage7.0. The agreement of the calculated by Eq (5) with experimental values was evaluated by the average of relative error between them, described by the following equation:

$$\text{error}=\frac{100}{N}\sum_{i=1}^{N}\left|\frac{\eta_{Calculations,i}-\eta_{Experiments,i}}{\eta_{Experiments,i}}\right| \quad (11)$$

where N denotes the total number of experimental data points.

The average of relative error between Eq (5) and Kozanevitch's experimental values was 17.1%.

Suzuki calculated the viscosities of $CaO\text{-}SiO_2\text{-}Al_2O_3$ system slag by using revised QCV (quasi-chemical viscosity) model and compared with experimental values of many researchers [6]. The model enables the viscosities of fully liquid slag in $CaO\text{-}SiO_2\text{-}Al_2O_3$ system to be predicted within experimental uncertainties over a wide range of composition and above liquidus temperatures. Viscosities calculated by Eq (5) were compared with Suzuki's calculation results and experimental viscosities of other researchers, which is shown in Figure 6 and Figure 7. Figure 6 shows change of viscosity with mole $Al_2O_3/(CaO+Al_2O_3)$ at different SiO_2 mole fractions and temperatures. Figure 7(a) shows change of viscosity with SiO_2 mole fractions at different temperatures in mole $Al_2O_3/CaO=50/50$, and Figure 7(b) shows the changes of basicity and contents of Al_2O_3 with SiO_2 mole fraction in mole $Al_2O_3/CaO=50/50$. As shown in Figure 6 and Figure **7,** viscosities calculated by Eq (5) were fit relatively

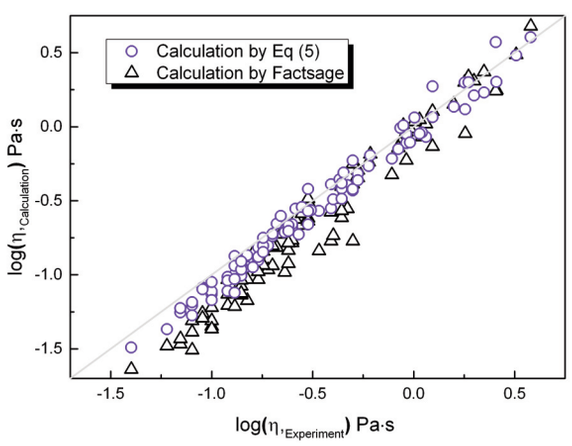

Figure 5. *Comparison between Kozakevitch's experimental values [1] and viscosities calculated by Eq (5), Factsage in $CaO\text{-}SiO_2\text{-}Al_2O_3$ system slag*

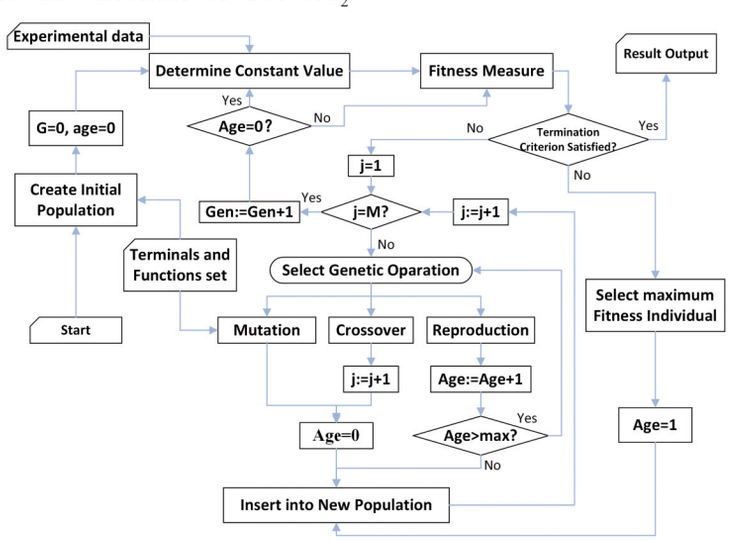

Figure 4. *Flowchart of GP for derivation of $CaO\text{-}SiO_2\text{-}Al_2O_3$ Slag viscosity equation*

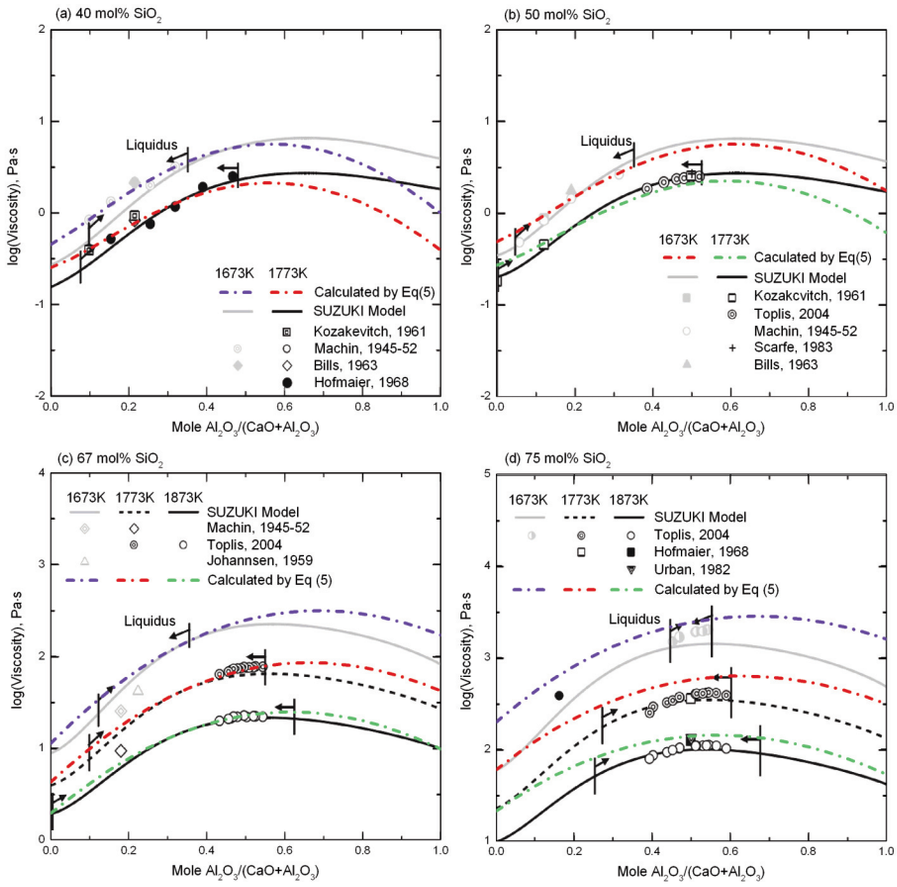

Figure 6. *Comparison of viscosities calculated by Eq (5) and Suzuki's revised QCV model, experimental viscosities in $CaO-SiO_2-Al_2O_3$ system slag: (a) 40 mol % SiO_2, (b) 50 mol % SiO_2, (c) 67 mol % SiO_2, (d) 75 mol % SiO_2*

well with viscosities calculated by Suzuki's model within the composition range of measure values and other researcher's experimental values.

Voskoboynikov measured many experimental viscosities of $CaO-SiO_2-Al_2O_3$ system slag in reference [2]. To evaluate the reliability of Eq (5) once more, viscosities calculated by Eq (5) were compared with Voskoboynikov's experimental values and viscosities calculated by Factsage7.0, which is shown in Figure 8. Basisity and Al_2O_3 of Voskoboynikov's slag were in the range of 0.16-3.7 and 0-35% respectively, and temperatures were in the range of 1250-1550℃. As can be seen in the Figure 8, calculated viscosities were relatively well fit with experimental values and it was more accurate than calculated results by Factsage 7.0. The average of relative error between values calculated by Eq (5) and experimental values was 20.48%.

Viscosities calculated by Eq (5) were compared with calculation values and experimental data of other researchers listed in reference [3], which is illustrated in Figure 9. Viscosities were calculated according to the change with weight ratio $Al_2O_3/(Al_2O_3+CaO)$ at different weight contents SiO_2 at 1500℃. It can be seen that viscosities calculated by Eq (5) were relatively well fit with experimental and calculated viscosities.

Finally, viscosities calculated by Eq (5) were compared with experimental results and model calculation results in reference[4] and calculation results by Factsage7.0, which is shown in Figure 10. The average of relative errors between viscosities calculated by Eq (5), Factsage and experimental data were 21.3% and 36.5%, respectively.

It is very important to exactly indicate the application range of the Eq (5). In slag compositions compared above, Eq (5) could be regarded to be relatively exact. Slag compositions of all viscosity measurement points mentioned above (see Figure 5-10) are shown in Figure 11. For convenience, slag compositions were represented by basicity and content of Al_2O_3. As shown in Figure 11, lines (1-4) and line 5 corresponded to slag compositions in Figure 6(a-d) and Figure 7 respectively, and lines (6-9) corresponded to slag compositions in Figure 9 (SiO_2 40, 50, 60, 70%). As shown in Figure 11, Kozanevitch's composition range was the widest, while Voskoboynikov's composition range was

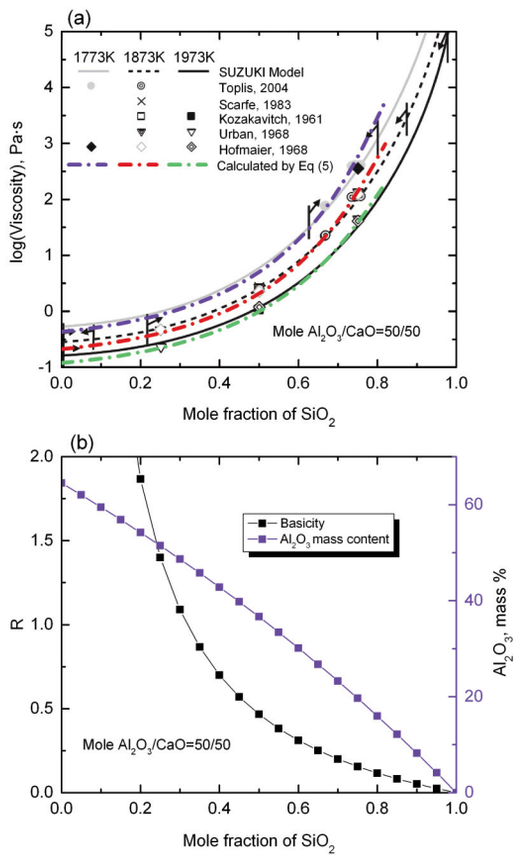

Figure 7. *(a) Comparison of viscosities calculated by Eq (5) and Suzuki's model, experimental viscosities with changes of SiO_2 mole fraction at different temperatures in CaO-SiO_2-Al_2O_3 system slag, (Al_2O_3/CaO=50/50) (b) Relationship between mole fraction of SiO_2 and basicity, Al_2O_3 mass content in CaO-SiO_2-Al_2O_3 system slag (Al_2O_3/CaO=50/50)*

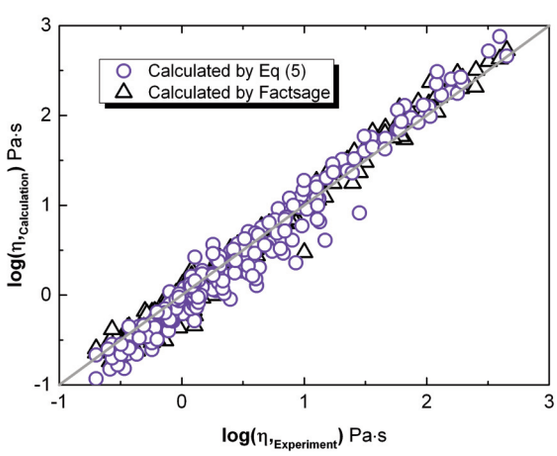

Figure 8. *Comparison between experimental values and experimental values, viscosities calculated by Eq (5), Factsage in CaO-SiO_2-Al_2O_3 system slag (experimental viscosities was listed in reference [2])*

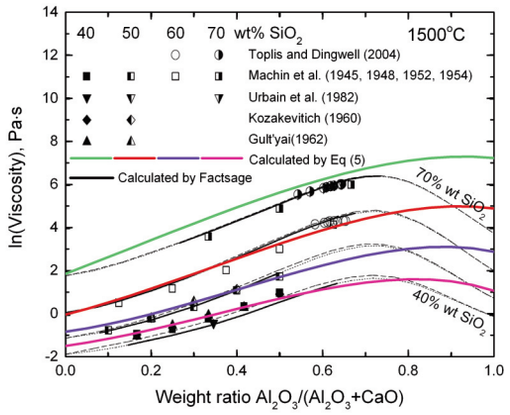

Figure 9. *Comparison of viscosities calculated by Eq (5) and experimental data, calculated by Factsage [3]*

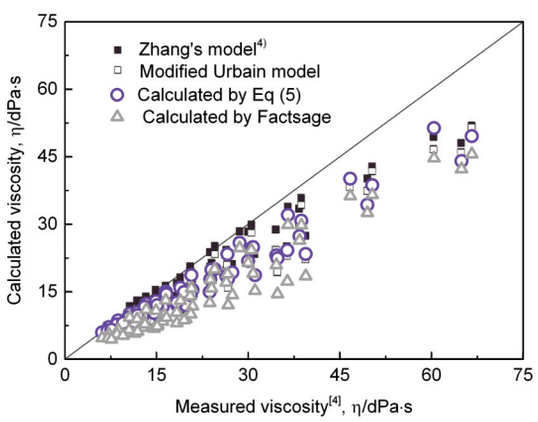

Figure 10. *Comparison of viscosities calculated by Eq (5) and Zhang's experiment data [4], model calculation results*

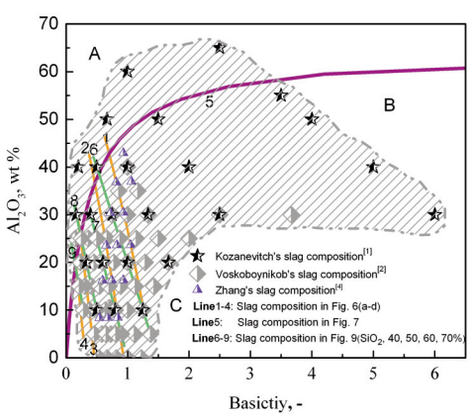

Figure 11. *Compositions of slag used in this work*

narrower than Kozanevitch's one. However, Voskoboynikov' experimental points were much higher than others. Slag composition ranges of other researchers were relatively narrow, which were contained within Kozanevitch and Voskoboynikov's composition ranges. Moreover, as shown in Figure 11,

viscosities for slag compositions were not measured in areas A, B, and C, because melting points of slag were relatively high in these areas.

All in all, Eq (5) can be recognized to depict the viscosity of $CaO-SiO_2-Al_2O_3$ system slag in wide composition and temperature range, i.e. in the area of gray oblique lines showed in Figure 11.

4. Conclusions

In this paper, genetic programming (GP) was employed to derive viscosity equation of $CaO-SiO_2-Al_2O_3$ system slag and its algorithm was listed. By applying GP, it was automatically described as form of simple algebraic equation with slag composition (or basicity and Al_2O_3 content) and temperature. The viscosities of $CaO-SiO_2-Al_2O_3$ system slag can be simply and expediently calculated in a wide range of compositions and temperatures by using the derived viscosity equation. To evaluate accuracy of viscosity equation of $CaO-SiO_2-Al_2O_3$ system slag, experimental data and calculated results of many researchers which were not used for derivation of the viscosity equation were compared, and a reasonably good agreement was demonstrated between calculated results by viscosity equation Eq (5) and other researcher experimental and calculated data. It is estimated that genetic programming can be used for distilling free-form nature laws from not only slag viscosity but also other complex metallurgical processes.

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IZVOĐENJE JEDNAČINE VISKOZNOSTI ŠLJAKE SISTEMA $CaO-SiO_2-Al_2O_3$ POMOĆU GP

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Apstrakt

Viskoznost šljake je od suštinskog značaja u metalurškim procesima koji se odvijaju na visokim temperaturama. Međutim, model viskoznosti šljake je teško tačno protumačiti zbog postojanja snažne nelinearne veze između sastava šljake i temperature. U ovom radu je korišćeno genetsko programiranje (GP) za izvođenje jednačine viskoznosti $CaO-SiO_2-Al_2O_3$ šljake. Jednačina je automatski opisana kao jednostavna algebarska jednačina koja uključuje baličnost i sadržaj Al_2O_3 i temperaturu. Prosečna relativna greška između vrednosti dobijenih jednačinom i eksperimentalnih podataka korišćenih za njeno izvođenje je iznosila samo 17,1%. Računarske simulacije su izvedene zbog procene tačnosti izvedene jednačine viskoznosti, a zatim su rezultati upoređeni sa mnogim eksperimentalno dobijenim vrednostima za viskoznost, kao i sa izračunatim vrednostima koje su dobili drugi istraživači. Sastav šljake i temperature korišćene za simulacione proračune su bili eksperimentalni podaci koji nisu korišćeni za izvođenje jednačine viskoznosti. Rezultati su pokazali da je jednačina viskoznosti relativno tačna. Viskoznost sistema $CaO-SiO_2-Al_2O_3$ šljake se može jednostavno i jasno predvideti na osnovu sastava i temperature pomoću izvedene jednačine za viskoznost.

Ključne reči: *Šljaka; Jednačina za viskoznost; Genetsko programiranje; Predviđanje*