A model for estimating viscosity of molten silicate slag based on slag structure theory

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Abstract: A structure based viscosity model has been proposed in the present study. In the model, the viscous activation energy, E_η is assumed to be inversely proportional to the total distance (bond length) among ions in molten slag according to the structural features of silicate slag, and E_η has been derived based on pure SiO_2 and inversely proportional to the same**.** The model formulation for correlating the viscosity of molten silicate system with its composition has been established where the mobility of an ion relative to pure SiO_2 has been considered The rules for calculating the concentrations of various ions existing in different composition region of the melts are involved piecewisely in the model. Experimental viscosity data for SiO_2-CaO-Al_2O_3-MgO system at different temperatures in literatures were used to fix the parameters in the present model. A reasonable agreement has been reached between the model estimated results and experimental data for SiO_2-CaO-Al_2O_3-MgO system. The average relative deviation obtained is approximating 20%. The model also shows satisfactory prediction for the corresponding binary and ternary systems. These indicate the model's good applicability to predict the viscosities of molten silicate slags.

Keywords: Viscosity, viscous activation energy, ionic theory, silicate

1. Introduction

As one of important physical properties of molten slag, viscosity has impact on the extraction, smelting and refining processes in metallurgy. For instance, as reported that the ability to absorb inclusions for slag improves with decrease of slag viscosity [1][2]. In addition, with mathematical model developments for the metallurgical process description and simulation, reliable viscosity data are urgently needed. However, the high temperature experiments for viscosity measurements is expensive work of time and labor consuming, thus the currently available viscosity data of molten slags are still not be able to meet the demands of the technological applications. Hence, it is essential to develop viscosity models for estimating the viscosities of various molten silicate slag systems.

A number of models have been proposed to estimate viscosity of molten slag based on Arrhenius equation and through establishing the connection between E_η and slag composition. Such as earlier Urbain model [3] which obtained equation extracted from limited experimental viscosity, thus, it is usually difficult to extend the model to other slag system, or lower order or higher order systems. In recent years, a series of models have been proposed on the basis of the structure of silicate slags, such as Mills *et al*[4] established a model which applied the optical basicity to predict the slag viscosity, Iida *et al*[5] used the modified quaternary basicity to predict slag viscosity and Zhang Lin *et al*[6][7] proposed a model exploring the relationship between slag viscosity and the concentration of bridge oxygen (O^0), non-bridge oxygen(O^-) and free oxygen (O^{2-}) in molten slag. Most of the models used the empirical or semi- empirical approaches, thus theoretical explanation between E_η and slag composition still needs to be clarified now. There have also been some viscosity models such as Koyana's model[8-10] and KTH model[11], trying to deduce E_η from theoretical consideration, however their calculations appeared somewhat complex. Masashi NAKAMOTO *et al*[12][113] proposed that "cutting-off" points owing to the addition of basic oxide caused viscous flow of silicate melts and deduced the relationship between E_η and slag composition by assuming E_η is inversely proportional to the distance of "cutting-off" points. As known, slag viscosity is determined by slag composition and temperature, thus it is proper approach to predict viscosity with structural consideration. In this paper, E_η is derived according to the structure of molten silicate and is assumed inversely proportional to the total distance among ions in molten slag. Furthermore, ionic theory is chosen to calculate the ion concentrations due to its simplicity and wide applicability.

2. Model

2.1 Model formulation

Silicate slags have network structure involving in SiO_4^{4-} units, where SiO_4^{4-} units are bonded with each other through bridge oxygen, the vertex oxygen in tetrahedron structure of SiO_4^{4-} units. When some basic oxide such as CaO etc, are added into silicate slag, the network structure was partially cut off by oxygen decomposed of basic oxide [4] to produce some numbers of non-bridge oxygen and/or free oxygen ions as shown in Fig. 1. As a result, the distance between ions in molten silicate increases. It has been suggested that as the degree of polymerization of melts diminishes owing to destructiveness of the network structure, there is corresponding decrease of slag viscosity [14][15].

According to the analysis above, assumptions have been proposed as follows: firstly, molten slag is made up of a variety of ions, such as Ca^{2+}, Mg^{2+}, O^{2-}, AlO_2^-, SiO_4^{4-} ……, as well as "SiO_2" that denotes the tetrahedron structure where the silicon is bonded with all bridge oxygen. Secondly, bond length controlled by different ions is different which can be expressed as their mobility correspondingly. A longer bond length indicates a weaker interaction between two ions, thus a greater mobility for the corresponding ions. Therefore, the viscous activation energy E_η is considered be inversely proportional to the distance between ions, which is denoted by S, and expressed as Eq(1). In order to analyze different effects of cation and anion on slag viscosity, the bond length between cation and anion in the present model has been divided into two parts controlled by the cation and anion respectively. The total bond length controlled by cation or anion is defined as its controlling bond length.

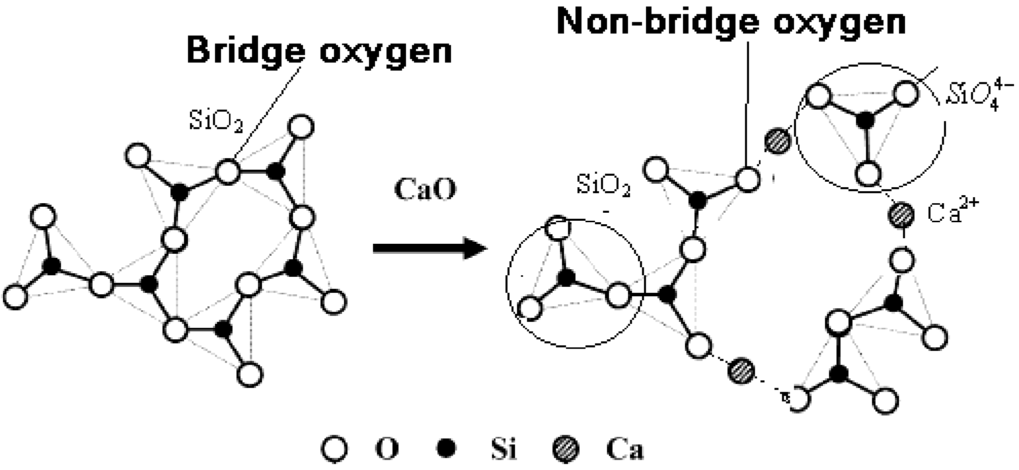

Fig. 1 Structure schematic diagram of silicate slag

$$E_\eta = \frac{E'}{S} = \frac{E'}{\sum_{i=1}^{n} x_i s_i} = \frac{E' / s_{"SiO_2"}}{x_{“SiO_2”} + \sum_{j=1}^{n} \frac{s_j}{s_{"SiO_2"}} x_j} \qquad (1)$$

In Eq(1), s_i and x_i represent the controlling bond length and mole fraction of ion i (i=Ca^{2+},Mg^{2+}, SiO_4^{4-},…as well as “SiO_2”) respectively. j and x_j represent the type of ion excluding “SiO_2” and its corresponding mole fraction respectively. $E'/ s_{"SiO_2"}$ is written as E and $s_j / s_{"SiO_2"}$ is written as λ_j. Substituting them into Eq(1), it gives

$$E_\eta = \frac{E}{x_{“SiO_2”} + \sum_{j=1}^{n} \lambda_j x_j} \qquad (2)$$

Where λ_j is representing the relative mobility of ion j relative to pure SiO_2. The term of ($x_{“SiO_2”} + \sum_{j=1}^{n} \lambda_j x_j$) is a dimensionless factor indicating the destructive degree of network tetrahedron structure of SiO_2 after adding basic oxide. There have been two extreme cases: Eq(2) turns to pure SiO_2 form when

$x_{"SiO_2"} = 1$ and the SiO$_2$ structure is destroyed completely when $x_{"SiO_2"} = 0$.

Substituting Eq(2) into Arrhenius equation, the viscosity for molten slag gives as

$$\eta = A \bullet \exp\left(\frac{E}{(x_{"SiO_2"} + \sum_{j=1}^{n} \lambda_j x_j)RT}\right) \qquad (3)$$

Where *A* is pre-exponential factor，*R* is the gas constant and *T* is the Kelvin temperature. In the pure SiO_2 slag,，equation (3) becomes as viscosity formula of pure SiO_2 $\eta = A \bullet \exp(E / RT)$. Therefore, the parameters A and E in the present model represent pre-exponential factor and viscous activation energy of pure SiO_2.

2.2 Ion concentration calculation

According to ionic theory of molten slags, acidic and basic oxide will be combined with each other as much as possible, to destroy each other's network structure when they are mixed in molten state. Assumed SiO$_2$ is dissociated as SiO_3^{2-} and SiO_4^{4-}, Al$_2$O$_3$ is dissociated as AlO_2^-. Basic oxides would dissociated as the cations (Ca^{2+}, Mg^{2+}, *etc*) and free oxygen ions. For the sake of simplicity, a complete dissociation process has been assumed, as the temperature is very high in molten slag. There exist different types of ions in molten slag with different slag components, thus the ion concentration should be in a calculated piecewise manner. The $SiO_2 - Al_2O_3 - CaO - MgO$ quaternary slag here is taken as an example for description of the ion concentration calculations as follows.

(a) $x_{SiO_2} + x_{Al_2O_3} > x_{CaO} + x_{MgO}$

In this case, the SiO$_2$ content existing in the slag is enough high, so that their network structure in the slag cannot be destroyed completely. Therefore, “SiO_2” exist in the molten slag as well as Ca^{2+}, Mg^{2+}, AlO_2^- and SiO_3^{2-}. Other types of ions are negligible. The concentration of each ion can be calculated as follows:

$$x_{Ca^{2+}} = x_{CaO} / \left(1 + x_{Al_2O_3}\right); \qquad (4)$$

$$x_{Mg^{2+}} = x_{MgO} / \left(1 + x_{Al_2O_3}\right); \qquad (5)$$

$$x_{\mathrm{AlO}_2^-} = 2\bullet x_{Al_2O_3} / \left(1 + x_{Al_2O_3}\right)\text{;} \tag{6}$$

$$x_{SiO_3^{2-}} = \left(x_{CaO} + x_{MgO} - x_{Al_2O_3}\right) / \left(1 + x_{Al_2O_3}\right)\text{;} \tag{7}$$

$$x_{\text{"}SiO_2\text{"}} = (x_{SiO_2} - x_{CaO} - x_{MgO} + x_{Al_2O_3}) / \left(1 + x_{Al_2O_3}\right) \tag{8}$$

(b) $x_{Al_2O_3} + x_{SiO_2} < x_{CaO} + x_{MgO} < x_{Al_2O_3} + 2\, x_{SiO_2}$

Generally, the composition of blast furnace (BF) slag is in this range. In this case, all the SiO_2 combine with basic oxide into SiO_3^{2-} and/or SiO_4^{4-} and thus there are nearly no "SiO_2" in the molten slag. Consequently, there are Ca^{2+}, Mg^{2+}, AlO_2^-, SiO_3^{2-} and SiO_4^{4-} existing in the molten slag. Hence the concentration values of ions in molten slag can be calculated as follows:

$$x_{Ca^{2+}} = x_{CaO} / \left(1 + x_{Al_2O_3}\right)\text{;} \tag{9}$$

$$x_{Mg^{2+}} = x_{MgO} / \left(1 + x_{Al_2O_3}\right)\text{;} \tag{10}$$

$$x_{\mathrm{AlO}_2^-} = 2\bullet x_{Al_2O_3} / \left(1 + x_{Al_2O_3}\right)\text{;} \tag{11}$$

$$x_{SiO_4^{4-}} = (x_{CaO} + x_{MgO} - x_{Al_2O_3} - x_{SiO_2}) / \left(1 + x_{Al_2O_3}\right) \tag{12}$$

$$x_{SiO_3^{2-}} = (2x_{SiO_2} + x_{Al_2O_3} - x_{CaO} - x_{MgO}) / \left(1 + x_{Al_2O_3}\right)\text{;} \tag{13}$$

(c) $x_{CaO} + x_{MgO} > x_{Al_2O_3} + 2\, x_{SiO_2}$

There are many of refining slags and fluxes whose chemical composition values fall in this range. Similarly with the analysis above, the types of ions existing in the molten slag in this case are Ca^{2+}, Mg^{2+}, AlO_2^-, SiO_4^{4-} and O^{2-}. The concentration values of each type of the ions can be attained from the following equations.

$$x_{Ca^{2+}} = x_{CaO} / \left(1 + x_{CaO} + x_{MgO} - 2\bullet x_{SiO_2}\right)\text{;} \tag{14}$$

$$x_{Mg^{2+}} = x_{MgO} / \left(1 + x_{CaO} + x_{MgO} - 2\bullet x_{SiO_2}\right)\text{;} \tag{15}$$

$$x_{\mathrm{AlO}_2^-} = 2\bullet xAl_2O_3 / \left(1 + xCaO + xMgO - 2\bullet xSiO_2\right)\text{;} \tag{16}$$

$$x_{SiO_4^{4-}} = x_{SiO_2} / \left(1 + x_{CaO} + x_{MgO} - 2\bullet x_{SiO_2}\right)\text{;} \tag{17}$$

$$x_{O^{2-}} = (x_{CaO} + x_{MgO} - x_{Al_2O_3} - 2 \bullet x_{SiO_2}) / (1 + x_{CaO} + x_{MgO} - 2 \bullet x_{SiO_2}) \tag{18}$$

Fig. 2 shows composition dependence of the fraction of three types of oxygen, bridge oxygen (O^0) , non-bridge oxygen(O^-) and free oxygen ions(O^{2-}), calculated by applying the calculation equations above into $CaO - SiO_2$ binary slag and compared with Frohberg's cell model [16] . When $x_{CaO} > 2x_{SiO_2}$ (x_{SiO_2} <0.33), there are only free oxygen O^{2-} and non-bridge O^- but bridge oxygen O^0 (SiO_4^{4-} contains $4O^-$, SiO_3^{2-} contains 2 O^- and 1 O^0 and SiO_2 includes 2 O^0) in the molten slag. When $x_{CaO} < 2x_{SiO_2}$ (x_{SiO_2} >0.33), there are only O^- and O^0 but free oxygen O^{2-} . When $x_{CaO} = 2x_{SiO_2}$ (x_{SiO_2} =0.33), the concentration of non-bridge O^- reaches its maximum as the SiO_2 most change into SiO_4^{4-}. As shown in Fig. 2, the present calculated results are in good agreement with those by using Frohberg's cell model, that indicates the assumption and the calculation rules in the present model is very reasonable.

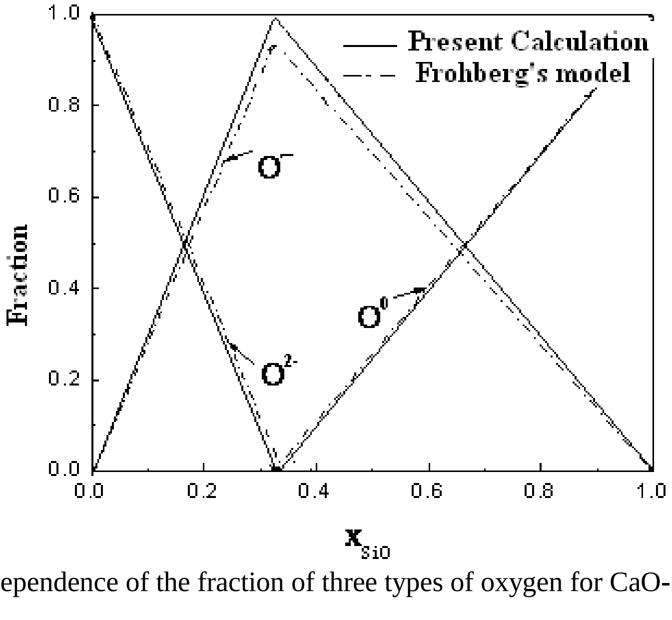

Fig. 2 Composition dependence of the fraction of three types of oxygen for CaO-SiO$_2$ binary slag.

2.3 Model parameters

The available bond length data in high temperature is not complete yet now, thus the value the parameters λ_j could only be fixed by using the experimental data. Ref [4] provides a number of experimental data with detailed slag composition. The parameters of the present model are obtained by fitting the experimental viscosity of slag of Ref[4] at the temperature of 1500℃，1450℃ and 1400℃ (105 points in total) respectively as follows: $\lambda_{SiO_4^{4-}}=13.0$, $\lambda_{SiO_3^{2-}}=7.0$, $\lambda_{AlO_2^{-}}$ =4.5, $\lambda_{O^{2-}}$ =6.0, $\lambda_{Ca^{2+}}$ =4.0, $\lambda_{Mg^{2+}}$ =3.8, $A=10.812e^{-0.0048T}$ and E=423kJ/mol

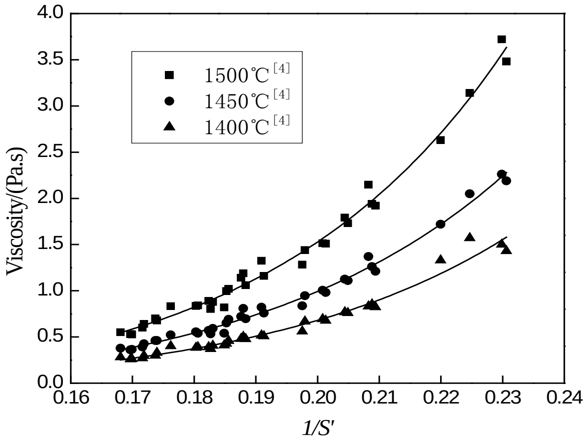

Fig. 3. The relationship between the dimensionless factor S′ and experimental viscosity

$$(S'=x_{\text{“}SiO_2\text{”}}+\sum_{j=1}^{n}\lambda_j x_j)$$

As shown in Fig.3, good fitting relationship exists between dimensionless factor S’ and viscosity at different temperature. The viscosity rises near monotonously with S’ decrease that indicates dimensionless factor S’ could well describe destructive degree of network tetrahedron structure of SiO_2 due to basic oxide addition, and further characterizes slag viscosity. Larger S’ implies network tetrahedron structure of SiO_2 is destroyed more completely, thus the corresponding slag possesses lower viscosity.

3. Results and discussion

In order to verify the reliability and applicability of the present model, large numbers viscosity experimental data of binary, ternary and multi-component slag systems from the published articles were collected and compared with the calculated results of the present model. Equation (19) and (20) were used to calculate the relative deviation and average relative deviation respectively.

$$\Delta=\frac{|\eta_{mea}-\eta_{cal}|}{\eta_{mea}}\times 100\% \tag{19}$$

$$\Delta_{av}=\frac{1}{N}\sum_{i=1}^{N}\Delta_i \tag{20}$$

In Eqs.(19) and (20), η_{mea} and η_{cal} are the measured viscosity value and calculated result of this model, N is the number of viscosity data respectively.

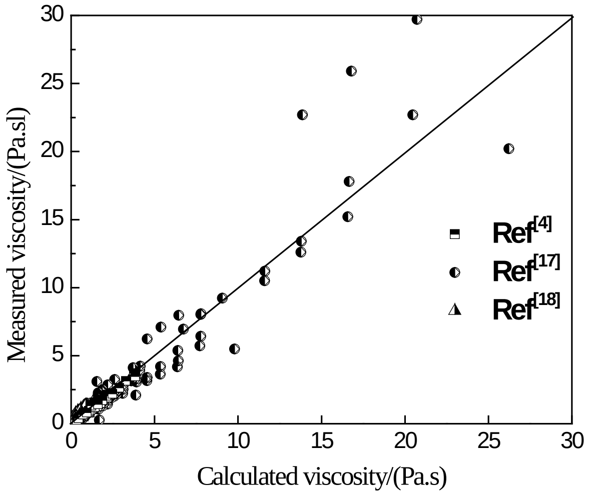

Fig. 4 Comparison between calculated and experimental viscosity for SiO_2-CaO-MgO-Al_2O_3 system.

Fig. 4 shows the comparison of the calculated results of the present model and the experimental viscosity values in references [4,17-18] in SiO_2-CaO-MgO-Al_2O_3 slag, where 281 data are presented in total. The temperature range of slag in the references is from 1573K to 1773K.and $x_{(CaO+MgO)}/x_{(SiO_2+Al_2O_3)}$ is between 0.50 to 1.60. The number of data with the relative deviation between calculated values and experimental viscosity within 20% is 161 among the total number of 281, which gives a ratio of 57.3%. The number of data with the relative deviation within 30% reaches 76.9% (216/281). The average relative deviation is 21.8%.

Fig.5 shows the comparison of the calculated result of the present model and the experimental viscosity value in references [19-20] in SiO_2-CaO-MgO ternary system slag, where 180 data are demonstrated in total. The temperature range of slag in the references is from 1623K to 2372K.and $x_{(CaO+MgO)}/x_{SiO_2}$ is between 0.75 and 1.87. The number of data with the relative deviation within 20% reaches 130 among the total data of 176, which gives a ratio of 73.9%. The number of data with relative deviation within 30% reaches 148, which gives a ratio of 84.1%. The average relative deviation is 20.0%.

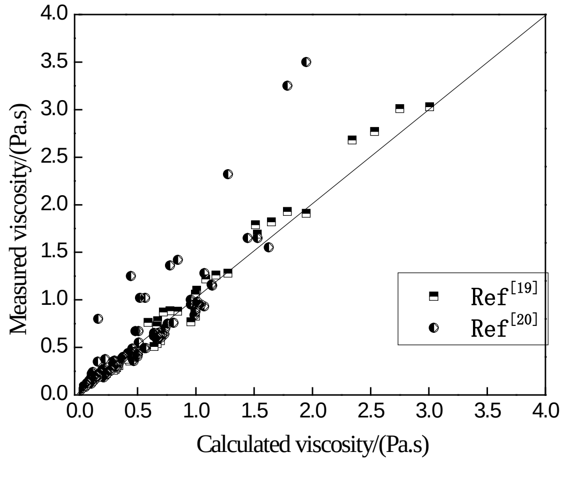

Fig 5 Comparison between calculated results and experimental viscosity for SiO_2-CaO-MgO system.

Fig. 6 shows the comparison of the calculated result of the present model and the experimental viscosity value in references[19, 21-24] in SiO_2-CaO binary system slag, where 245 data are presented in total. The temperature range of the slags in the references is from 1713K to 2393K.and x_{CaO} / x_{SiO_2} is between 0.67 to 2.5. There are 71.7% of the data with a relative deviation within 20%, and 91.7% of the data with a relative deviation within 30% in the temperature range from 1713K to 1973K. The average relative deviation is 15.3%. Therefore, the predictions of the model in the temperature range are accurate. While, if the temperature is higher than 1973K(1973-2398K), the majority of the data have a relative deviation greater than 30%, indicating that the model deviation is getting lowered down but still in a reasonable range. The reason perhaps could be that the increasing rate of bond length between different ions differ with temperature rise, resulting in the values of parameter $\lambda_j = s_j / s_{"SiO_2"}$ variation obviously when temperature increase sharply. Of course there are a few data within the temperature range in 1713K to 1973K whose relative deviation is greater than 30%. It is found that the majority of them are the slag system with high melting point and a sharp rise point has turned out in their viscosity-temperature curve. This might be due to the presence of a solid phase in the slag at the temperature but the model does not take it into account.

As shown in Fig. 5 and Fig. 6, the model also shows a better deviation between model prediction and experimental data for the corresponding binary and ternary systems when the temperature is within 1973K indicating that the new model has wide applicability.

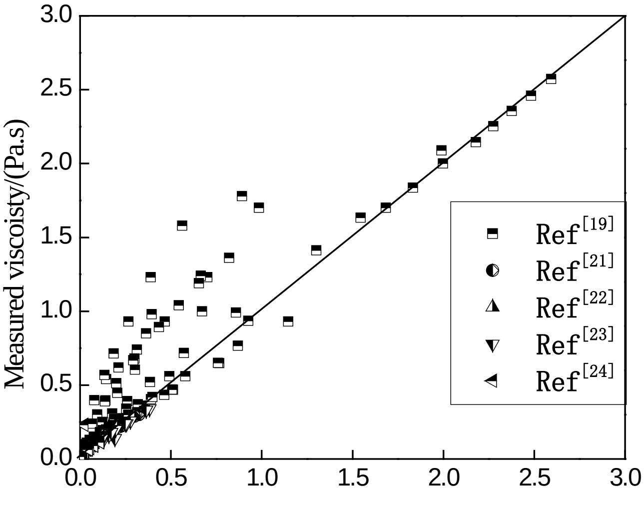

Fig. 6 Comparison between model calculated results and the experimental viscosity for SiO_2-CaO system.

The high temperature experiment is very complex so that the experiment deviation is about 20% usually. Further deviations could be caused due to different equipment and operators. Therefore, it is reasonable that the deviation between the model calculated result and the measured viscosity is within 30%. The relative deviations between calculated results using present model and experimental value are mostly within 30% that shows the reliability of the calculation results of the model from the statistics. The reference [4] compared the average relative deviation between series of viscosity model and experimental results at the temperature of 1673K，1723K，1773K and 1803K, where Iida's model's average deviation is 26.6%, Riboud's model's is 43%, Koyana's model's is 33.2% and Urbain's model's is 101.5%.T he average relative deviation of present model is about 20% thus this model accuracy is quite well.

When the temperature is higher than 1973K, the present model is not suitable any more. As explained above, this is perhaps because the increase rate of bond length between different ions differs with temperature rise, resulting in the values of parameter λ_j variation when temperature rises sharply. The further research is needed to study the relationship between the parameters λ_j and the temperature. But it does not preclude the model to apply in ironmaking and steelmaking as their temperature are usually lower than 1973K.

4. Conclusion

In this paper, the relationship between viscous activation energy and silicate slag composition was established in a structure-based consideration. A series of ion concentrations in the molten slag systems were formulated in the model. A better agreement reaches between the model estimated results and previously published experimental viscosity data for SiO2-CaO-Al2O3-MgO system. The average relative deviation is approximating 20%. Moreover, the model also shows a better accuracy and precision in estimating the viscosities of the corresponding binary and ternary systems, therefore indicating that the new model has good applicability.

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