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| Author(s)     | Nakamoto, Masashi; Miyabayashi, Yoshitugu; Holappa, Lauri; Tanaka, Toshihiro      |
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A Model for Estimating Viscosities of Aluminosilicate Melts Containing Alkali Oxides

Masashi NAKAMOTO,1) Yoshitugu MIYABAYASHI,2) Lauri HOLAPPA3) and Toshihiro TANAKA3)

1) Laboratory of Metallurgy, Helsinki University of Technology, P.O. Box 6200, FI-02015 TKK, Espoo, Finland.
2) Nikko Environmental Services Co., Ltd., 3453 Miyata, Hitachi, Ibaraki 317-0055 Japan.
3) Division of Materials and Manufacturing Science, Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871 Japan.

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A model to estimate the viscosity of aluminosilicate melts, including alkali oxides, was constructed in the present study. The model is based on an earlier model, and considers the bonding state of oxygen in molten silicate and the flow mechanism of melts with a network structure. The straightforward method of Susa et al. to evaluate bonding states of oxygen in chemical compositions was applied to the present model since their method can easily be extended to any multi-component system. The calculation results of our model can express the dependence on viscosity in quaternary aluminosilicates containing alkali oxide and their subsystems.

KEY WORDS: viscosity; silicate melts; structure; aluminosilicate melts; alkali oxides.

1. Introduction

Demand of smelting furnaces for waste treatment has been increasing in the respect of detoxification as well as recycling of industrial waste such as asbestos1,2) shredder residue,3,4) and glass dust containing lead.5) The essential issue in smelting furnace operations is to elucidate adequate conditions with the aim of stabilizing processes due to the wide variety in the composition of industrial waste. In this regard, Miyabayashi et al.1) pointed out that the fluidity of slag is one of the crucial operating components. Although the fluidity of slag is typically evaluated by the viscosity measurement, the estimation of viscosity by models is considered to be much more helpful in evaluating viscosity in melting furnaces due to the diversity in composition of industrial waste. This is because of the considerable difficulty in measuring viscosity of all kinds of slags in melting furnaces due to the diversity in composition of industrial waste. The system of the slag in melting furnaces for the waste including asbestos is the multi-component aluminosilicate containing CaO, FeO, Fe2O3, MgO, K2O, Na2O, etc.1)

Many models to estimate the viscosity of slag melts have been developed in the past.6–8) It has been accepted that the estimation of viscosity of molten slags is a difficult task since the abrupt and complicated structure changes of silicate melts are reflected by the viscosity. From this viewpoint, successful models based on the structure of molten slags have been proposed by Zhang and Jahanshahi,9–11) and Kondratiev and Jak.12–15) The three bonding states of oxygen in slag melts, which are (i) bridging oxygen, (ii) non-bridging oxygen ion, and (iii) free oxygen ion, are considered in their viscosity model. The authors9–21) have also constructed a structurally-based viscosity model by considering the flow mechanism of silicate melts within the network structure as well as the bonding states of oxygen in molten silicate. In this model, the concentrations of non-bridging oxygen ions and free oxygen ions are calculated as bonding states of oxygen by the IRSID thermodynamic ‘cell model.’22,23) This viscosity model can represent the composition dependence of viscosity in a wide composition range in the multi-component silicate system SiO2–CaO–MgO–FeO–Al2O3 in blast furnace process. However, the IRSID “cell model” is not applicable to slag containing alkali oxides, which cannot be ignored in the slag of melting furnace, in spite of its wide validity system SiO2–TiO2–Ti2O3–Cr2O3–Al2O3–Fe2O3–CrO–FeO–MgO–MnO–CaO–CaF2–S.

On the other hand, a predictive equation for the refractive indices of aluminosilicate melts was proposed by Susa et al.24) Assuming that the structure of aluminosilicate melts consists of only three kinds of chemical bonds, i.e. Si–BO (bridging oxygen in Si tetrahedral unit), Al–BO (bridging oxygen in Al tetrahedral unit with a charge-compensating cation), and Si–NBO (NBO: non-bridging oxygen connected with Si) with a charge-compensating cation, it is possible to calculate the bonding states of oxygen in molten aluminosilicate by the chemical composition of slag without any thermodynamic parameters. Owing to a simple method, the evaluation of the bonding states of oxygen can be conducted in various kinds of silicate systems including alkali oxides as well as alkaline earth oxides. Their equation succeeds in predicting the refractive index values with small error in six component silicate melts in spite of its simplicity.

In the present work, the earlier viscosity model developed by the authors was modified to the viscosity of alumi-
nosilicate melts containing alkali oxides in melting furnaces, $\text{SiO}_2$–CaO–MgO–FeO–K$_2$O–Na$_2$O–Al$_2$O$_3$ system, by applying the method of Susa et al. to evaluate the bonding states of oxygen in the molten silicate instead of thermodynamic models.

2. Model

2.1. Equation

The equation to describe the viscosity of molten silicate slag in the present model is based on the equation in the earlier model which was proposed considering the bonding state of oxygen in the network structure of silicate melts and its flow mechanism.$^{16-21}$ The concept of the model is as follows: Silicate slag has a network structure made of bonding $\text{SiO}_4^{2-}$ units and Si tetrahedral ions, as shown in Fig. 1(a). Si tetrahedral ions are combined through bridging oxygen $\text{O}^\circ$, the convex oxygen in the structure of Si tetrahedral ions. The non-bridging oxygen ion $\text{O}^-$ and the free oxygen ion $\text{O}^{2-}$ are generated by partially cutting off the network structure of Si tetrahedral in $\text{SiO}_2$ when adding some basic oxides such as CaO and Na$_2$O in silicate slag, as shown in Fig. 1(b). The non-bridging oxygen ion and the free oxygen ion have a larger mobility than the bridging oxygen in the network structure because there are “cutting-off” points. These are bonds of oxygen not connected to Si, just next to the non-bridging oxygen ion and the free oxygen ion due to the breaking of the network structure. Assuming that the movements of “cutting-off” points cause a viscous flow, therefore, an increase in the number of non-bridging oxygen ions and free oxygen ions yields a lowering of the activation energy of viscosity. In addition, it is assumed that these “cutting-off” points move in a random direction along the network structure of silicate melts. The equation of viscosity $\eta$ in binary silicate system derived from the above concept in earlier studies$^{16-21}$ is given by the following equations:

$$\eta = A \cdot \exp \left( \frac{E_v}{RT} \right) \quad \text{(1)}$$

$$E_v = \frac{E}{1 + \alpha \cdot (N_{\text{O}^-} + N_{\text{O}^\circ})} \quad \text{(2)}$$

where $A (=4.80 \times 10^{-5})$ is a constant, $E_v$ is the activation energy for viscosity, $R$ is the gas constant, and $T$ is the temperature. $E (=5.21 \times 10^4 (J))$ is the activation energy for viscosity of pure $\text{SiO}_2$, and $\alpha$ is a parameter relating to the weakness of the bonding between the cation and oxygen ion at the “cutting-off” point, which depends on the oxide component. $N_{\text{O}^-}$ and $N_{\text{O}^\circ}$ are fractions of the non-bridging oxygen ion and free oxygen ion, respectively. The $N_{\text{O}^-}$ and $N_{\text{O}^\circ}$ are evaluated by the IRSID ‘cell model’.\textsuperscript{22,23}

In the evaluation of chemical bonds of oxygen, Susa et al.$^{24}$ assumed that the structure of the aluminosilicate melts is composed of only three kinds of chemical bonds. Their assumption results in the calculation of chemical bonds of oxygen solely by the concentration of slag components. In addition, by considering the Al–BO bond in their equation, the oxygen states can be estimated in case that aluminum oxide works as network former.

The equation to calculate the viscosity of silicate melts is modified to the following equations in order to apply the method of Susa et al. instead of the IRSID ‘cell model’ to the evaluation of the bonding states of oxygen:

$$\eta = A \cdot \exp \left( \frac{E_v}{RT} \right) \quad \text{(3)}$$

$$E_v = \frac{E}{1 + \sum_i \alpha_i \cdot N_{\text{BO}+\text{FO}_i} + \sum_j \alpha_j \cdot M_{\text{BO}} \cdot N_{\text{BO}+\text{BO}_j}} \quad \text{(4)}$$

where the activation energy $E_v$ is a function of the sum of the fractions of non-bridging oxygen ion (NBO) and free oxygen ion (FO) $N_{\text{BO}+\text{FO}_i}$ and the fraction of the bridging oxygen (BO) in the Al tetrahedral unit $N_{M_{\text{BO}}+M_{\text{BO}_j}}$ is the same as the $(N_{\text{O}^-} + N_{\text{O}^\circ})$ in Eq. (2). $\alpha_i$ and $\alpha_j$ are parameters. $i$ is the component of melt except $\text{SiO}_2$, i.e. CaO, MgO, FeO, K$_2$O, Na$_2$O, Al$_2$O$_3$, and $j$ is the charge-compensating ion from component $i$ except Al$_2$O$_3$, i.e. Ca$^{2+}$, Mg$^{2+}$, Fe$^{3+}$, K$^+$ and Na$^+$. $A$ and $E$ have the same values in the original equation, i.e. $4.80 \times 10^{-5}$ and $5.21 \times 10^4(J)$ respectively. It is assumed that there are no differences between non-bridging oxygen ions and free oxygen ions in the respect of the influence of those ions on viscosity. The effect of the bridging oxygen in the Al tetrahedral unit on the activation energy is treated in the same manner as the effect of the non-bridging oxygen by assuming that the bridging oxygen in the Al tetrahedral unit has a somewhat different mobility from that of the bridging oxygen in the Si tetrahedral unit.

The fractions of two kinds of oxygen $N_{\text{BO}+\text{FO}_i}$ and $N_{M_{\text{BO}}+M_{\text{BO}_j}}$ were calculated with reference of the method of Susa et al.$^{24}$ as follows:

(i) Binary system

(i-1) $a\text{SiO}_2 + b(M_2O_3) \quad \{a + b=1 \text{ (mol)}\}$

$$((M_2O_3) = \text{CaO, MgO, FeO, K}_2\text{O or Na}_2\text{O}) \text{ system}$$

The addition of basic oxide to silicate provides non-bridging oxygen ions and free oxygen ions by breaking the Si–O bonds in the network structure of silicate; i.e. the number of oxygen bonds not connected to Si increases. When $b$ mol of $(M_2O_3)$ is added to silicate the number of the bonds of oxygen not connected to Si on basic oxide
(M\textsubscript{2}O\textsubscript{3}) \(n_{O-Mi}\) is 2\(b_i\) mol because 1 mol of (M\textsubscript{2}O\textsubscript{3}) produces 2 mol of oxygen bonds not connected to Si. The total number of the bonds of oxygen in the system \(\alpha\text{SiO}_2-b_1(M\textsubscript{2}O\textsubscript{3})\), \(n_{\text{total-O}}\) is \(4a+2b\) mol. In our model, the ratio of the number of oxygen bonds not connected to Si \(n_{O-Mi}\) to the total number of the oxygen bonds in the system \(n_{\text{total-O}}\) is treated as the sum of the fractions of non-bridging oxygen ions and free oxygen ions \(N_{\text{NBO+FO}}\). In case of a binary system, the content of bridging oxygen in the Al tetrahedral \(N_{\text{Al-BO}_3}\) is zero. Subsequently,

\[ N_{\text{NBO+FO}}=n_{O-Mi}/n_{\text{total-O}}=b_1/(2a+b_1) \] .........................................(5)

\[ N_{\text{Al-BO}_3}=0 \] .............................................(6)

(ii-2) \(\alpha\text{SiO}_2-b_1(M\textsubscript{2}O\textsubscript{3})-c\text{Al}_2\text{O}_3\) system \(\{a+b+c=1\text{ (mol)}\}\)

When the amount of basic oxide (M\textsubscript{2}O\textsubscript{3}), \(b_1\) is higher than the amount of aluminum oxide \(c\), aluminum oxide works only as network former since there is a sufficient charge-compensating cation to produce the Al tetrahedral in the melt as mentioned above. Here, it is assumed that the supply of charge-compensating ions in each basic oxide is proportional to the content of basic oxide, meaning that the number of oxygen bonds in the Al tetrahedral \(n_{O-Al(Al)}\) with the charge-compensating cation \(j\) is \(8c(b_1/\sum b_i)\) mol and that the number of the bonds of oxygen not connected to Si is \(2b_1-2c(b_1/\sum b_i)\) mol. Subsequently,

\[ N_{\text{NBO+FO}}=n_{O-Mi}/n_{\text{total-O}}=4c\left(b_1/\sum b_i\right)\left(2a+\sum b_i+3c\right) \] .........................................(13)

\[ N_{\text{Al-BO}_3}=n_{O-Al(Al)}/n_{\text{total-O}}=b_1-c\left(b_1/\sum b_i\right)\left(2a+\sum b_i+3c\right) \] .........................................(14)

(iii-2) \(\sum b_i<c\)

When the sum of the amount of basic oxide (M\textsubscript{2}O\textsubscript{3}), \(b_1\) is lower than the amount of aluminum oxide \(c\), the equation is derived in the same manner as the equation in the ternary system. Subsequently,

\[ N_{\text{NBO+FO}}=n_{O-Mi}/n_{\text{total-O}}=4b_1\left(2a+\sum b_i+3c\right) \] .........................................(15)

\[ N_{\text{Al-BO}_3}=n_{O-Al(Al)}/n_{\text{total-O}}=3\left(c-\sum b_i\right)\left(2a+\sum b_i+3c\right) \] .........................................(16)

3. Results and Discussion

3.1. Determination of Parameters

There are two kinds of parameters, \(\alpha_x\) and \(\alpha_y_{\text{inAl}}\), in the present equation (4) for expressing the viscosity of molten aluminosilicates. The former parameter relates to the non-bridging oxygen ion and the free oxygen ion, and the latter is the parameter relating to the bridging oxygen in the Al tetrahedral. The \(\alpha_x\) and \(\alpha_y_{\text{inAl}}\) relate to the weaknesses in the bonds of non-bridging oxygen ion, and the weakness of bridging oxygen in the Al tetrahedral, respectively. In the present equation, therefore, the larger parameter values lead to a lower viscosity. These weaknesses of \(\alpha_x\) and \(\alpha_y_{\text{inAl}}\) depend on the cation next to the non-bridging oxygen ion and...
the free oxygen ion, which is the cation produced by the addition of basic oxide \((\text{M}_x\text{O}_y)\) to silicate, and the charge-compensating cation near Al tetrahedral, which is also the cation produced by adding basic oxide \((\text{M}_x\text{O}_y)\) to silicate, respectively.

In this study, the parameters \(\alpha_i\) and \(\alpha_{\text{in,Al}}\) were determined from experimental data in binary silicate systems\(^{25-34}\) and ternary aluminosilicate systems.\(^{26,34-36,39-41}\) The way to determine the parameters is as follows: (i) First, the value of \(\alpha_i\) in each oxide \(i\) was obtained from the application of the above equation to fit the experimental values of the viscosity in each binary \(\text{SiO}_2-\text{(M}_x\text{O}_y)\) system, where \((\text{M}_x\text{O}_y)\) is CaO, MgO, FeO, K\(_2\)O, Na\(_2\)O or Al\(_2\)O\(_3\). (ii) Subsequently, the assessment of the value \(\alpha_{\text{in,Al}}\) in each charge-compensating cation \(j\) was conducted by fitting the calculated viscosity with parameter \(\alpha_i\) determined in the binary system to the experimental data in each ternary \(\text{SiO}_2-\text{(M}_x\text{O}_y)\)-Al\(_2\)O\(_3\) system, where \((\text{M}_x\text{O}_y)\) is CaO, MgO, FeO, K\(_2\)O or Na\(_2\)O. The experimental data\(^{25-41}\) used for the determination of these parameters are shown in Table 1, and the assessed parameters are listed in Table 2.

### 3.2. Calculation Results

At first, the ability of the present model to represent viscosity using the evaluation manner for oxygen state with reference to Susa et al.'s model\(^{24}\) was examined by comparing the calculated values with the literature data, which were used for the parameter determinations.

**Table 1.** Experimental data used for determining model parameters.

| System          | Temperature (K) | References |
|-----------------|-----------------|------------|
| **Binary**      |                 |            |
| \(\text{SiO}_2-\text{CaO}\) | 1723 - 2373     | 25-27      |
| \(\text{SiO}_2-\text{MgO}\) | 1823 - 2273     | 27,28      |
| \(\text{SiO}_2-\text{FeO}\) | 1473 - 1723     | 29,32      |
| \(\text{SiO}_2-\text{K}_2\text{O}\) | 1373 - 1973     | 28,33      |
| \(\text{SiO}_2-\text{Na}_2\text{O}\) | 1373 - 1973     | 28,33      |
| \(\text{SiO}_2-\text{Al}_2\text{O}_3\) | 1973 - 2373     | 26,34      |
| **Ternary**     |                 |            |
| \(\text{SiO}_2-\text{CaO-}\text{Al}_2\text{O}_3\) | 1423 - 2273     | 26,35      |
| \(\text{SiO}_2-\text{MgO-}\text{Al}_2\text{O}_3\) | 1673 - 2073     | 36-39      |
| \(\text{SiO}_2-\text{FeO-}\text{Al}_2\text{O}_3\) | 1523 - 1573     | 40         |
| \(\text{SiO}_2-\text{K}_2\text{O-}\text{Al}_2\text{O}_3\) | 1523 - 1673     | 39         |
| \(\text{SiO}_2-\text{Na}_2\text{O-}\text{Al}_2\text{O}_3\) | 1523 - 1773     | 41         |

**Table 2.** Model parameters.

| \((\text{M}_x\text{O}_y)\)_i | \(\alpha_i\) | \(\alpha_j\) | \(\alpha_{\text{in,Al}}\) |
|------------------------------|--------------|--------------|--------------------------|
| CaO                          | 4.00         | Ca\(^{2+}\)  | 1.46                     |
| MgO                          | 3.43         | Mg\(^{2+}\)  | 1.56                     |
| FeO                          | 6.05         | Fe\(^{3+}\)  | 3.15                     |
| K\(_2\)O                     | 6.25         | K\(^{+}\)    | -0.60                    |
| Na\(_2\)O                    | 7.35         | Na\(^{+}\)   | 0.27                     |
| Al\(_2\)O\(_3\)              | 1.14         |              |                          |

The calculation results with the experimental data in binary systems, \(\text{SiO}_2-\text{CaO}, \text{SiO}_2-\text{Na}_2\text{O}\) and \(\text{SiO}_2-\text{Al}_2\text{O}_3\), are shown in Figs. 2-4. The present model represents the com-
position dependences of viscosity in these binary systems. The calculated values tend to deviate slightly downward from the experimental values with higher CaO and Na2O contents in SiO2–CaO and SiO2–Na2O systems. This deviation could be due to the assumption that there are no differences between the influence of non-bridging oxygen ions and free oxygen ions on viscosity. In the previous studies,16–21 the content of non-bridging oxygen ion and the content of free oxygen ion were individually calculated by the IRSID ‘cell model’, and the sum of these oxygen contents described the composition dependence of viscosity over a wide composition range. The thermodynamically calculated \((N_{\text{NBO}} + N_{\text{FO}})\) in a binary system, for example, increases until a certain composition, and then does not change so much. This behavior corresponds to the composition dependence of viscosity in binary systems, which shows a sharp decrease in viscosity in the range from pure SiO2 to a certain composition, after which the viscosity decreases only slightly. In the present model, on the other hand, the sum of the fractions of non-bridging oxygen ions and free oxygen ions \(N_{\text{NBO+FO}}\) was estimated only from the number of all oxygen bonds not connected to Si except for the bonds in Al tetrahedral, leading to a continuous increase of \(N_{\text{NBO+FO}}\) with increasing the basic oxide in the whole composition range, i.e. the viscosity continues to decrease to some extent even at higher basic oxide content.

Figures 5 and 6 show the viscosities in ternary aluminosilicate systems, SiO2–CaO–Al2O3 at 1 973 K and SiO2–K2O–Al2O3 at 1 673 K, respectively. In these figures, the lines are the iso-viscosities calculated by the present model and the dots represent experimental data. The iso-viscosity line breaks at a certain point in Fig. 5. This is the point in which the content of CaO is equal to the content of Al2O3 in mol. When the amount of CaO is higher than the amount of aluminum oxide, aluminum oxide works only as network former due to a sufficient amount of charge-compensating ion Ca2+ for producing the Al tetrahedral in the melts as mentioned above. On the other hand, at the Al2O3 side of the above point, aluminum oxide works as network modifier as well as network former because of the lack of charge-compensating ion Ca2+. The iso-viscosity lines calculated relating to this point by the present model describe the composition dependency of experimental viscosity over a wide composition region in the SiO2–CaO–Al2O3 system at 1 973 K, although the calculated results show a slightly lower viscosity than the experimental data at low SiO2 contents, as well as in binary systems. In Fig. 6, the calculation results in SiO2–K2O–Al2O3 at 1 673 K are in agreement with the experimental data although the composition region targeted in the present calculation is narrow.

The viscosity in quaternary aluminosilicate slags containing MgO or FeO was shown in Figs. 7 and 8. Figure 7 presents the viscosity of SiO2–CaO–MgO–10mass%Al2O3 slag at 1 723 K. The calculated iso-viscosity lines well express the composition dependency in viscosity measured by Machin et al.42,43 showing the lowering of viscosity with decreasing SiO2. The viscosity in quasi-ternary SiO2–CaO–FeO system at 20mass%Al2O3 by Gimmelfarb44 indicates an unusual trend in Fig. 8, where the viscosity decreases by increasing CaO or FeO at relatively high SiO2 content, and more CaO or FeO yields the increase in viscosity. As can be appreciated from the liquidus line drawn with “FactSage” in this figure, the data at low SiO2 concentration exist in the zone of coexistence of solid and liquid, leading to a viscosity rise. Comparing the calculated viscosity with the literature data in a liquid region, it was found that the present model reproduces the viscosities in SiO2–CaO–MgO–10mass%Al2O3 system.

Finally, the prediction of viscosity in the quaternary aluminosilicate system SiO2–CaO–Al2O3–K2O or Na2O was attempted by using the present model. The viscosity measurements of the molten aluminosilicates containing alkali oxide have been conducted by the group of Nakashima et al.45 Figure 9 shows the viscosity of quaternary aluminosilicate melts SiO2–CaO–Al2O3–K2O or Na2O at 1 873 K. Figure 9(a) shows the measured viscosity, whose values were taken from data points from Ref. 45, and Fig. 9(b) shows the calculated values by the present model. In Figs. 9(c) and 9(d), in addition, the calculations were conducted by the two generally-known models, (i) Riboud model46 and (ii) modified Iida model,47 which can be applied to the aluminosilicate systems containing alkaline oxides and have been recognized as models suited for estimating the
viscosity of industrial slags.\textsuperscript{48}) In Fig. 9(a), the viscosity of SiO\textsubscript{2}–CaO–Al\textsubscript{2}O\textsubscript{3}–K\textsubscript{2}O melts increases with increasing the content of K\textsubscript{2}O, while the viscosity of SiO\textsubscript{2}–CaO–Al\textsubscript{2}O\textsubscript{3}–Na\textsubscript{2}O melts decreases with increasing content of Na\textsubscript{2}O. On the other hand, the addition of Na\textsubscript{2}O to molten SiO\textsubscript{2}–CaO–Al\textsubscript{2}O\textsubscript{3}–Na\textsubscript{2}O system lowers the viscosity, and the viscosity in SiO\textsubscript{2}–CaO–Al\textsubscript{2}O\textsubscript{3}–K\textsubscript{2}O indicates a slight decrease with the addition of K\textsubscript{2}O in the calculation of the results in Fig. 9(b). As shown in Fig. 9(c), Riboud model has a similar trend as the present model, but with stronger decreasing influence on viscosity by increasing K\textsubscript{2}O or Na\textsubscript{2}O than the present model. In the calculation results of modified Iida model in Fig. 9(d), the additions of K\textsubscript{2}O and Na\textsubscript{2}O yield a similar composition dependence of viscosity. From this result, it was found that the present model clearly expresses the difference between the effect of K\textsubscript{2}O addition and the effect of Na\textsubscript{2}O addition to SiO\textsubscript{2}–CaO–Al\textsubscript{2}O\textsubscript{3} melts on viscosity in comparison with the other models, although the
calculation values don’t describe the increment of viscosity of adding K₂O. In the present model, the following two parameters determine the behavior of viscosity with varying composition in aluminosilicate system: (i) the parameter \( \alpha_{\text{inAl}} \), which was introduced as the weakness of bridging oxygen in the Al tetrahedral with charge-compensating cation \( j \), and (ii) the amount of each charge-compensating ion supplied to form Al tetrahedral. In the former case, the \( \alpha_{\text{inAl}} \) of K₂O is smaller than the \( \alpha_{\text{inAl}} \) of Na₂O, which means the bond of bridging oxygen in the Al tetrahedral with K⁺ is stronger than Na⁺. The calculated results in two quaternary systems SiO₂–CaO–Al₂O₃–K₂O and SiO₂–CaO–Al₂O₃–Na₂O reflect the parameter \( \alpha_{\text{inAl}} \). In the latter case, it is reasonable to suppose that some of cations preferentially or with higher possibility work as charge-compensating ion since each cation has specific properties like interactions with other ions. Due to a lack of this information, however, in this study it was assumed that the supply of charge-compensating ion from each basic oxide was proportional to the content of basic oxide, which causes a poor reproducibility of composition dependence of experimental values in SiO₂–CaO–Al₂O₃–K₂O system. For example, if K⁺ works as charge-compensating ion strongly in comparison with Ca²⁺, i.e. the amount of K⁺ as charge-compensating ion is larger than that estimated with the present assumption and the amount of Ca²⁺ is smaller, the viscosity is higher than the viscosity calculated in the present work since \( \alpha_{\text{inAl}} \) of K⁺ is smaller than \( \alpha_{\text{inAl}} \) of Ca²⁺. As a result, this gives a better description of viscosity in SiO₂–CaO–Al₂O₃–K₂O system.

4. Conclusions

The earlier viscosity model derived by considering the bonding states of oxygen in molten silicate slag and its flow mechanism was modified in order to estimate the viscosity of aluminosilicate melts including alkali oxides. In this modification, the concentration of non-bridging oxygen ions and free oxygen ions used in the calculation of bonding states of oxygen were evaluated with reference to the method of Susa et al. instead of a thermodynamic model. The present model can present the composition dependence of viscosity in ternary aluminosilicate systems as well as quaternary aluminosilicate systems containing MgO or FeO, and the distinction of the effects of K₂O and Na₂O on viscosity in quaternary aluminosilicate systems containing each alkali oxide without thermodynamic parameters.

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