Modelling Viscosity of Alumina-containing Silicate Melts

Ling Zhang and Sharif Jahanshahi
G K Williams CRC for Extractive Metallurgy
CSIRO-Division of Minerals,
Box 312, Clayton South, Vic. 3169, Australia
Fax: 61-3-95628919
Email: ling.zhang@minerals.csiro.au

ABSTRACT

A structurally related model for the viscosity of silicate melts developed by the present authors [1,2] has been extended to melts containing alumina. In the binary Al_2O_3-SiO_2 system viscosity was modelled by treating Al_2O_3 as a metal oxide, i.e., as a network modifier based on the similarities in the general behaviour of the binary systems. The ternary and higher order systems were modelled using only the parameters for binary silicate melts without introduction of high order parameters. The model has been validated for ternary and higher order (upto six component) aluminosilicate melts over broad temperature and composition ranges against an extensive collection of experimental data. Good agreement between the experimental data and the viscosity values calculated by the model was obtained.

INTRODUCTION

Alumina is one of the major components in silicate glasses and melts which is important to glass and metallurgical industries and the earth science. As a result there has been considerable effort directed towards investigating the structure and various properties of aluminosilicate melts and glasses [3,4]. One of the important findings is that the influence of Al_2O_3 on physical and transport properties at a given silica mole fraction depends on the ratio of R (alumina)/(metal oxide) for systems. For example, Kozakevitch [5] measured viscosity of the CaO-Al_2O_3-SiO_2 system over a temperature range of 1873 to 2373 K and found that at and above 2173 K and a silica mole fraction above 0.3, viscosity increases with increasing Al_2O_3 content when R<1. Maxima were observed at $Al_2O_3/CaO=1$, viscosity then decreases as Al_2O_3/CaO ratio increases. Similar behaviour occurs for both viscosity and electrical conductivity of sodium aluminosilicate glasses [4,6,7]. It is believed that the changes in properties with R ratio are due to structural changes in the systems, caused by the so-called amphoteric role played by Al_2O_3 in the network structure. Given that the melts of interest to metallurgical processes often have low silica concentration where extensive three dimensional network structures are not formed, then the network notion maybe generalised to include various sizes and shapes of anions formed by connection of SiO_4^{4-} and AlO_4^{5-} tetrahedra.

There have been mainly two types of structural models proposed to explain the physical changes observed in the aluminosilicate glasses [4]. One is the coordination model, which holds the traditional view of a change of Al coordination from being tetrahedral to being both tetrahedral and octahedral as R increases above unity. The other is the tricluster model, which proposes that Al remains tetrahedrally coordinated over the entire composition range and the formation of triclusters (three tetrahedra meeting at a common vertex to a three coordinated oxygen) for R>1 could account for the property changes. In the paper by Zirl and Garofalini [4] the experimental evidence in support of both models has been discussed. The results of their molecular dynamics simulations of sodium aluminosilicate glasses favour the tricluster model. However, it may be difficult to judge which model provides the better picture of the structure based on current knowledge. It is clear that as R becomes greater than unity, there are not enough metal cations to act as charge compensators and Al atoms bring some disorder or weak points to the network structure, which may cause the observed changes in physical and transport properties.

A review by Turkdogan and Bills in 1960 [8] analysed viscosity behaviour of alumina containing silicate melts in the CaO-MgO-Al_2O_3-SiO_2 system and sub-systems. A concept of the silica equivalence of alumina, N_a, was introduced to correlate data in binary and higher order systems. Although useful in representing data and showing the relationships between R and N_a, the formula can not be readily used to calculate viscosity. Another finding from their work is that the plots of log(viscosity) versus 1/T(K) do not show a linear relationship over the temperature range of 1423 to 2273 K, where data are available. The slope of the curves increases with decreasing temperature and the degree of the curvature becomes more severe as silica and alumina content increases. A structural change with temperature was considered to be responsible for such behaviour.

There are a few viscosity models for slags containing Al_2O_3 [9-11]. In particular the Urbain model [9] has been considered as a general model for multicomponent systems. The Weymann equation [12] was used to describe the temperature dependence of viscosity. In the Urbain model

Al_2O_3 was treated as amphoteric component and up to twelve parameters were needed for calculating viscosity of the melts containing Al_2O_3.

In the present paper the work on extending the structurally related viscosity model to Al_2O_3 containing silicate melts will be discussed. Based on the general viscosity behaviour of the binary Al_2O_3-SiO_2 system [5,13], which is similar to other binary metal oxide silicate melts, the contribution of Al_2O_3 to the network structure was not considered for the binary. In ternary or higher order systems where other metal oxides, such as CaO and MgO are present, the contribution by Al_2O_3 to the network structure was assumed to be proportional to the sum of the mole fraction of metal oxides. No additional parameter was introduced to account for the effect of Al_2O_3. The modelling strategy and results will be discussed by comparing the calculated viscosity with the experimental data. The calculated iso-viscosity curves in the CaO- Al_2O_3-SiO_2 system are also compared with the Urbain model [9].

THE MODEL

The model was based on the analysis of the general viscosity behaviour of silicate melts observed by experimental studies. The experimental data of the binary Al_2O_3-SiO_2 system [5,13] show that the viscosity behaviour is similar to other binary metal oxide silicate melts. As a result, the modelling procedures described in detail by the present authors [1,2] were used for the Al_2O_3-SiO_2 system.

The temperature dependence of viscosity was calculated using the Weymann equation [12] or Frenkel equation [14]

$$h = A^W T \exp(E_h^W / RT), \qquad (1)$$

where η is viscosity, T is temperature in K, R is the gas constant, A^W and E_h^W are the pre-exponential term and the activation energy, respectively.

The composition dependence of viscosity is described in terms of structural parameters, i.e., the activation energy is a function of fractions of bridging (or doubly bonded) oxygen N_O^0 and free oxygen N_O^{2-};

$$E_h^W = a + b(N_{O^0})^3 + c(N_{O^0})^2 + d(N_{O^{2-}}), \qquad (2)$$

where *a, b, c* and *d* are fitting parameters optimized against experimental data. The values of N_O^0 and N_O^{2-} were obtained by the Cell model formulism [15]. The pre-exponential term A^W was calculated by using the equation

$$\ln(A^W) = a' + b' E_h^W, \qquad (3)$$

where *a'* and *b'* are fitting parameters.

As described in our previous papers [1,2], the model parameters *a, b, c* and *d* in Eq.(2), and *a'* and *b'* in Eq.(3) were only required to be determined for binary silicate systems. For higher order silicate systems, the model parameters are assumed to be linear functions of those of the binary

silicate systems. More details and examples of calculation of the parameters for higher order systems have been provided in previous papers [1,2].

The maximum viscosity values observed in the CaO-Al_2O_3-SiO_2 system at a fixed silica content and Al_2O_3 /CaO ratio near unity [5] suggest that the effect of Al_2O_3 cannot be treated as a simple addition of a metal oxide. For most metal oxides, such as CaO or MgO, the cations are not linked to the SiO_4^{4-} units to form the network. On the other hand Al_2O_3 can not be considered as equivalent silica addition either, in which Al_2O_3 joined the network as additional structural units in the form of AlO_4^{5-} acting similar as SiO_4^{4-} but with less stability. The viscosity behaviour showed both structural characteristics. However, to model such behaviour correctly, a physical picture of the structural changes brought about by Al_2O_3 is required.

Starting with the CaO-SiO_2 binary at a fixed silica mole fraction (X_{SiO2}=0.4), initial substitution of Al_2O_3 for CaO will promote AlO_4^{5-} to join the silicate anions as part of chains, branches, sheets or the network comfortably as there are enough Ca^{2+} cations around to act as charge compensators. Viscosity is expected to increase with the substitution of Al_2O_3 for CaO. Further substitution, i.e., increase R, will cause the viscosity to rise further till R=1. When R>1, the number of charge compensators are less than the number of AlO_4^{5-} tetrahedra. In order to maintain the local charge balance, it is expected that some degree of re-arrangement of AlO_4^{5-} units inside the anions occur which will introduce disorder or weak points to the network structure. When this effect becomes dominating, further substitution causes viscosity to decrease. The exact nature of the disorder in the structure can be discussed by using the structural models mentioned earlier. However, for the purpose of the modelling, the argument above provides a physical picture which suggests that the network former nature of Al_2O_3 depends on the existence and the relative amount of the charge compensator. It is assumed in the present model that the contribution of Al_2O_3 to the network is proportional to the sum of the mole fraction of the metal oxides.

As mentioned earlier, non-linear behaviour of the activation energy was found for alumina containing slags [8]. For example, data by Urbain et al [13] which covered a wide temperature range (1578-2456 K) in the CaO-Al_2O_3–SiO_2 system showed the curves of log(η) versus 1/T(K) bending up towards low temperatures implying higher activation energy values. As a result temperature dependent terms of the activation energy were introduced for ternary and higher order Al_2O_3 containing slags.

RESULTS AND DISCUSSION

A. Al_2O_3 –SiO_2

Viscosity in the Al_2O_3–SiO_2 system has been investigated by Kozakevitch [5] and Urbain et al [13]. The composition range covered X_{SiO2}=0.298 to 0.938 and the temperature range was 1923 to 2373 K. The variation of viscosity with composition and temperature are plotted in Figure 1 and 2. The general behaviour of viscosity in the Al_2O_3–SiO_2 is similar to other binary silicate melts, i.e., viscosity increases with increasing silica content and the plots of log(η) versus 1/T(K) are linear. It can be seen that the two sets of measurement are in good agreement with slightly lower values from Kozakevitch [5]. The calculated viscosities by the present model (solid lines) in Figures 1 and 2 represent the experimental data well.

B. Ternary and Higher Order Systems

The model has been tested for viscosity of alumina containing slags from ternary up to six component systems by using a comprehensive collection of published data. Good agreement between the model calculations and the experimental data was obtained for most systems. A full discussion of the modelling will be reported in a separate publication [16]. Examples of the systems examined are given below. These systems are of practical importance, i.e., the base system for many metallurgical processes, $CaO\text{-}Al_2O_3\text{–}SiO_2$, coal ash type slag of $CaO\text{-}Al_2O_3\text{–}FeO\text{-}SiO_2$ and a six component system $CaO\text{-}MgO\text{-}FeO\text{-}Al_2O_3\text{–}Fe_2O_3\text{-}SiO_2$.

1. $CaO\text{-}Al_2O_3\text{–}SiO_2$

The viscosity of the $CaO\text{-}Al_2O_3\text{–}SiO_2$ system has been studied extensively due to its practical importance and theoretical interests in examining the structural properties [5,13,17-31]. The experimental points can be found in most of liquid region from 1573 to 2273 K at 50 K intervals. The most comprehensive sets of data are available from work by Machin, Hanna and Yee [17-20] and Kozakevitch [5]. A systematic approach was taken in these studies, which provided a clear picture of viscosity behaviour.

Our model was validated using the data on this ternary system. A comparison of results is shown in Figure 3 where the calculated values versus the experimental data are plotted. The data shown in this figure are from 16 papers [5,13,17-31] with 1083 data points. The fit to most of the data is excellent considering such a wide range of values. This also suggests that the different measurements gave a consistent picture of the viscosity behaviour in this system, considering the 30% error bar common to viscosity measurements.

Most published measurements were carried out at relatively lower temperatures (1573-1873 K) where the liquid region is confined to low alumina contents. The data by Kozakevitch [5] at 2173 K covered a liquid region from low to high alumina content and provides a more complete picture of effect of alumina on viscosity. Based on these data points, Kozakevitch proposed “U” shape iso-viscosity curves for this system (Figure 6-7 in Ref [5]) which showed the maxima viscosity value at R=1. This observation can be considered as one of the early evidence of amphoteric nature of alumina. In order to show if the model can reproduce such behaviour, the calculated iso-viscosity curves are plotted together with data from Ref. [5] in Figure 4. The iso-viscosity curves calculated by the Urbain model [9] are also shown. It can be seen that the “U” shape iso-viscosity curves were obtained and the fit to the data is close. The iso-viscosity curves by the Urbain model all lie below the ones by the present model and this suggests that the Urbain model tends to overestimate viscosity in these systems.

2. $CaO\text{-}FeO\text{-}Al_2O_3\text{-}SiO_2$

Recently, the viscosity in the $CaO\text{-}FeO\text{-}Al_2O_3\text{-}SiO_2$ system has been extensively studied by Hurst et al [32-34]. Their studies were aimed at investigating the behaviour of coal ash type slag. The measurements were carried out for temperatures at 1673, 1723 and 1773 K. The level of “FeO” was less than 2 wt% for fluxed coal ash, 5 and 10% wt for synthetic slag. The modelling results are compared with their data in Figure 5. It can be seen that the model is applicable to coal ash type slags characterised by high Al_2O_3 (15-36 wt%) and high SiO_2 (34-61 wt%) contents. The figure also shows that the fit to the fluxed coal ash data is not as good as those of the synthetic slags. The model tends to overestimate viscosity which may be due to re-normalisation of the slag composition by omitting small amount of alkali oxides and other impurities. It is evident that

the effect of alkali components that are much more effective in reducing viscosity than alkali earth oxides can not be ignored in such slags.

3. $CaO-MgO-FeO-Fe_2O_3-Al_2O_3-SiO_2$

It has been found that there are less data available for the higher systems. The viscosity data found for highest order system were six component system of the $CaO-MgO-FeO-Fe_2O_3-Al_2O_3-SiO_2$ system by Bill [23] and the $CaO-MgO-MnO-FeO-Al_2O_3-SiO_2$ system by Mishra et al [35]. The composition and temperature covered for these systems are relatively limited when compared with lower order systems, only the region of practical interest have been experimentally investigated. As an example of the modelling results for the higher order systems, the fit of the model to the data of the $CaO-MgO-FeO-Fe_2O_3-Al_2O_3-SiO_2$ system by Bills [23] is shown in Figure 6. The Fe_2O_3 content in the melts is less than 1 wt%, therefore it is basically a five component systems with CaO: 17-27, MgO: 4.5-7.7, FeO: 4-31, Al_2O_3: 6.6-18 and SiO_2: 30-56 wt%. It can be seen the calculated viscosity values agree well in general with the experimental data.

C. General Features of the Model

The viscosity model for alumina-containing and alumina-free silicate melts developed by the present authors [1,2,36] has been aimed to be general, accurate and with physical meaning. The approach was to collect as much as possible published data and analyse the experimental observations on the general behaviour of viscosity with varying temperature and composition. At the same time the theories on liquid viscosity were reviewed, existing models evaluated and some models examined in detail [37].

A general picture of the behaviour of silicate melts emerged during the modelling process and it may be summarised as follows:

- Effect of different oxide components on viscosity can be described by the notion of network former (SiO_2), modifier (CaO, MgO, FeO, MnO, PbO, Fe_2O_3) and amphoteric (Al_2O_3) oxides, similar to that as originally proposed by Urbain [9].
- The network former SiO_2 has strongest effect on viscosity. High silica content results in an increase in the number of strongly bonded structure unit, SiO_4^{4-}, and linkage of these units make it difficult for the viscous flow. As a result viscosity increases with increasing silica content (in mole fraction).
- The modifiers CaO, MgO, FeO, MnO, PbO and Fe_2O_3 tend to reduce viscosity. However their effectiveness are different. As has been discussed by the authors [1,2], the viscosity data suggested that the stability of the metal oxides in terms of the melting point could be used as a guide to estimate their relative effectiveness in reducing viscosity. This can be considered that the movement of atoms depends on the bonding environment, i.e., the interaction between oxygen and metal cations. The stronger the bonding, the more difficult the movement and therefore higher viscosity is expected. Similarly, Bills [23] has observed the difference in the effect of MgO and FeO on viscosity and she attributed this to the difference in the electrostatic binding force between Fe^{2+} and Mg^{2+} to silicate anions.
- The effect of amphoteric components, such as Al_2O_3 depends on the presence of the charge compensators. The structure unit AlO_4^{5-} is not as stable as SiO_4^{4-} due to the longer bond length of Al-O than that of Si-O. As a result, the increase in viscosity cause by equal mole fraction of alumina is less than that of silica. Furthermore the effectiveness of Al_2O_3 in increasing

viscosity reduces as the ratio of R (Al_2O_3/CaO) increases. In the Al_2O_3 - SiO_2 binary where charge compensators are not available, viscosity behaviour is similar to other binary silicate melts, except the viscosity is much higher than for other binary silicate melts.

Based on the above understanding, the composition dependent part of the model was treated by using structural parameters, while the temperature dependence of viscosity was described by Arrhenius type equations. Although the model is still empirical in nature, i.e., fitting parameters are needed, it provides accurate descriptions of effect of different components on viscosity. Therefore it can be used to predict viscosity beyond the region where data are not available.

A limitation of the model is for slags with low SiO_2 content, say $X_{SiO2}<0.1$. The lower part of the iso-viscosity curves in the CaO-Al_2O_3-SiO_2 system (Figure 4) suggest viscosity in the CaO-Al_2O_3 system will reach a minimum around $X_{Al2O3}=0.6$ which does not seem to be possible. A separate model for silica free aluminate slags is needed for the CaO-Al_2O_3 binary system. Fortunately the silica content in most practical metallurgical melts is greater than $X_{SiO2}=0.2$ and the model has been shown to be valid in this region.

It should be mentioned the model in the current form is only applicable to homogeneous liquid slags. The validation discussed above includes only data measured in the liquid region. Quite often the published data show a sudden jump of the viscosity values below a certain temperature. This is an indication of precipitation of crystal particles. By referring to the phase diagram it was often found that the measurements were below liquidus. Such data were not included in the validation. Approaches for modelling viscosity of melts with solid particles have been initiated at our laboratory, and experimental investigations to quantify the effect of volume fraction and size of the solid particles on viscosity carried out [38,39].

Future improvements of the model include incorporation of additional elements such as alkali elements and extension to calculate viscosity for melts with solid particles. Validation and refinements will continue as new data become available. The model has been invited as one of the participants of a Round Robin test of viscosity models conducted by the NPL [40]. The findings of the test are expected to be published in the near future.

CONCLUSIONS

A structurally related model for viscosity of molten slags has been successfully extended to aluminosilicate melts containing CaO, MgO, MnO, FeO, Fe_2O_3 and PbO. The effect of alumina on viscosity was treated based on the understanding of the structural role played by alumina. The viscosity of both alumina-free and alumina-containing multicomponent slags can be calculated using only the parameters obtained for their sub-binary systems. Good agreement between calculated values and extensive collections of published data over a wide temperature and composition range was obtained. Close analysis of viscosity behaviour in relation to the findings on the structure of the melts is very useful in building up models with more physical meanings and therefore a better description of phenomenon under investigation.

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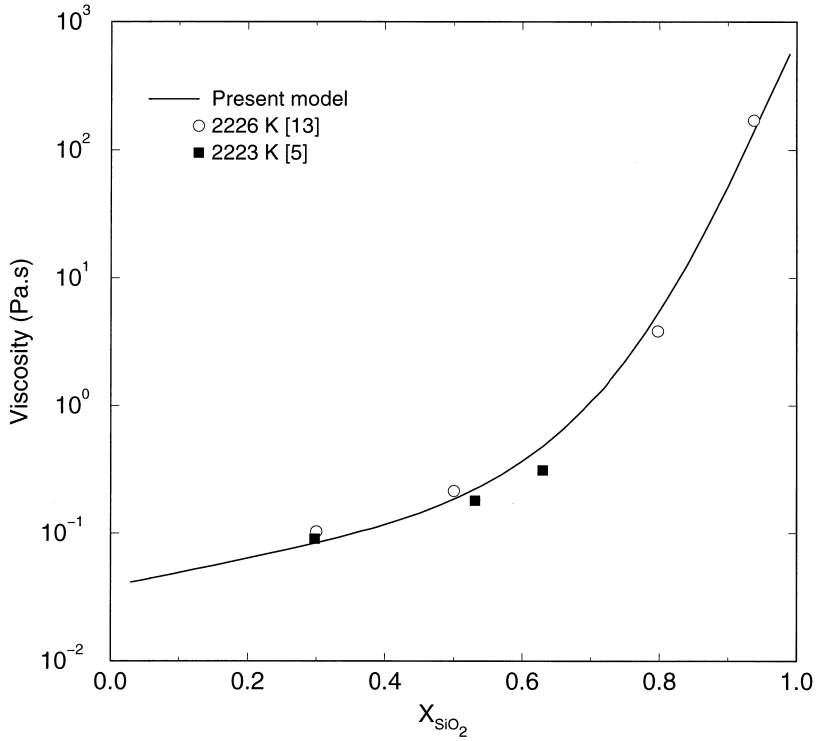

Figure 1. Comparison between the calculated values and the experimental data on the composition dependence of viscosity in the Al_2O_3-SiO_2 system at 2223 and 2226 K.

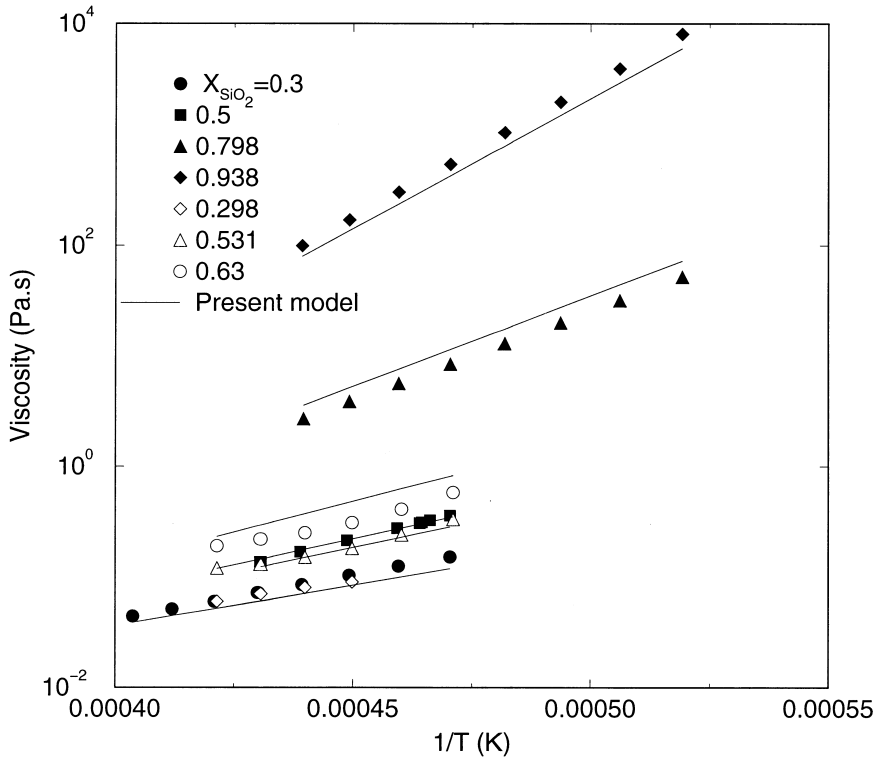

Figure 2. Variation of viscosity with temperature and composition in the Al_2O_3-SiO_2 system. Solid symbols are data from Ref.[13] and open symbols from Ref.[5].

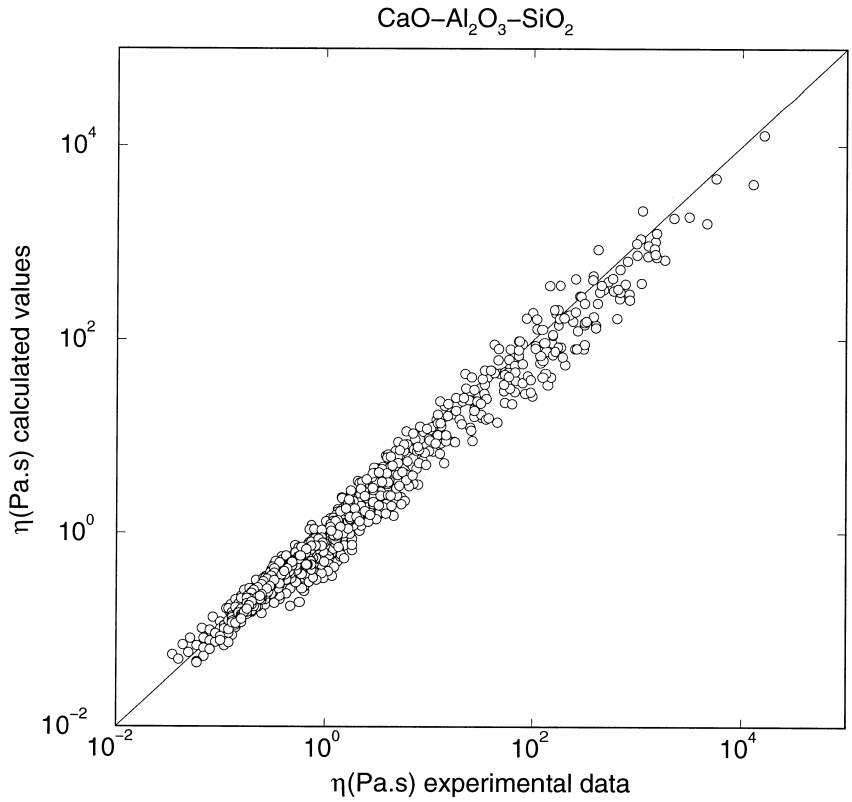

Figure 3. Comparison between the calculated viscosity values and the experimental data [5,13,17-31] for the CaO-Al_2O_3-SiO_2 system.

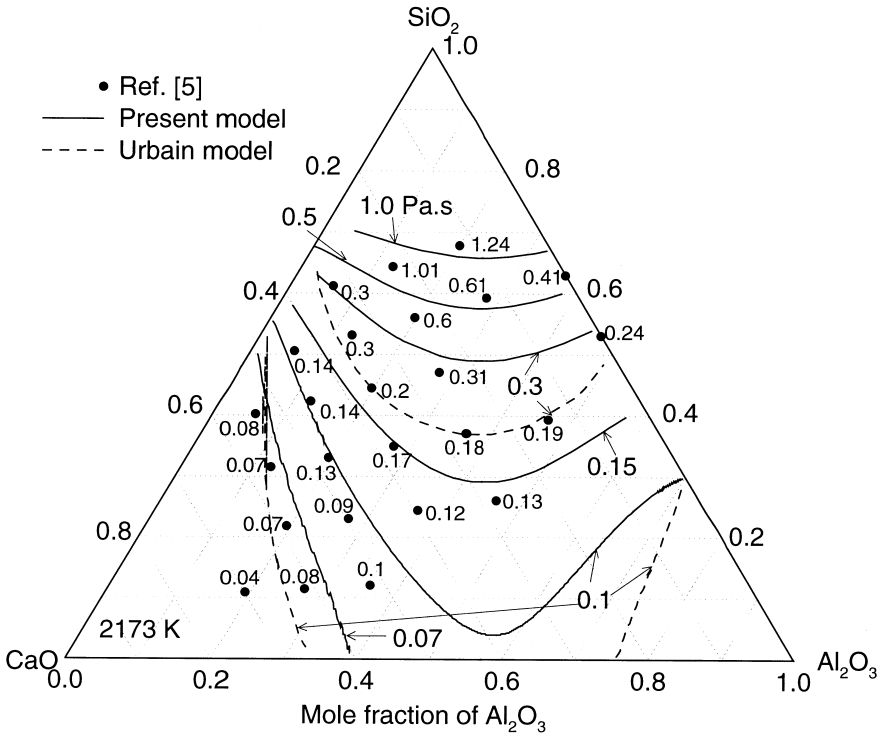

Figure 4. Comparison between the calculated values and the experimental data [5] on the composition dependence of viscosity of the CaO-Al_2O_3-SiO_2 system at 2173 K.

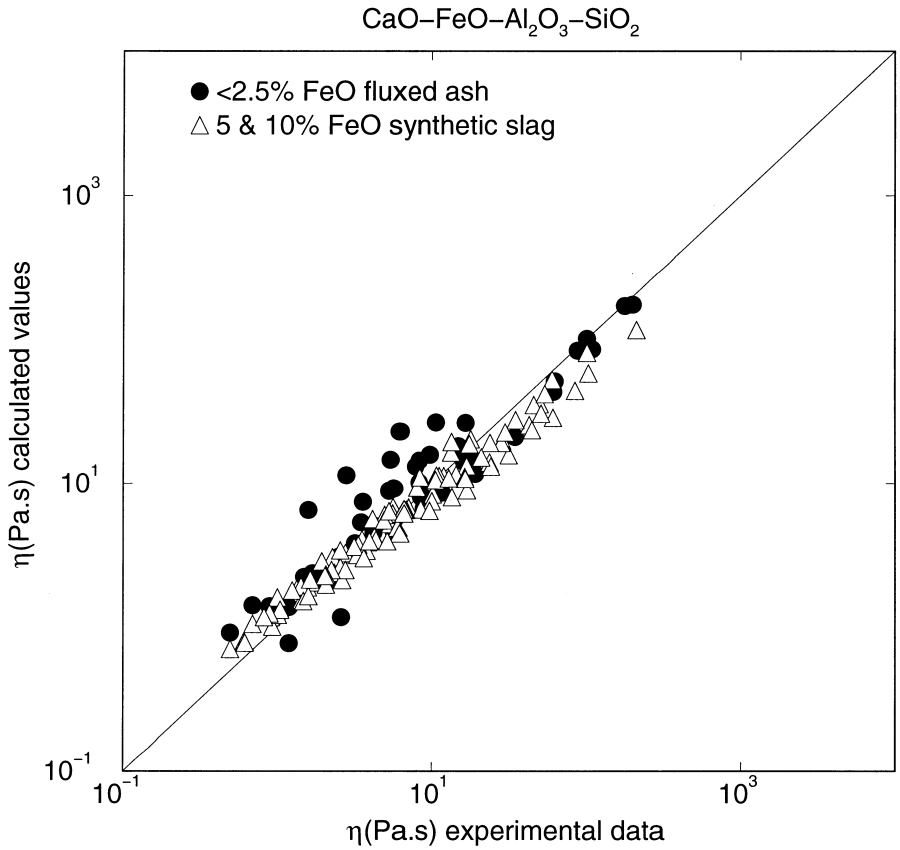

Figure 5. Comparison between the calculated viscosity values and the experimental data of fluxed coal ash at 1723 and 1773 K [32] and synthetic slag CaO-FeO-Al_2O_3-SiO_2 at 1673,1723 and 1773 K [34].

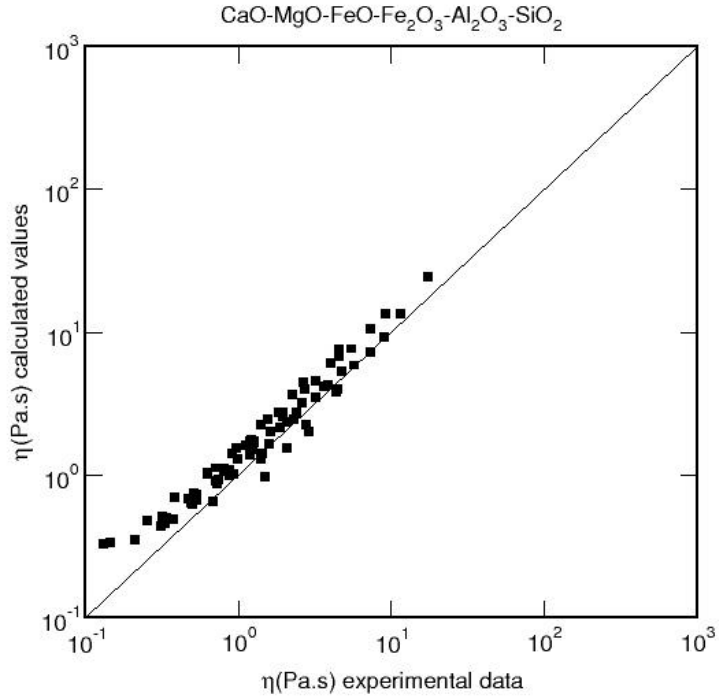

Figure 6. Comparison between the calculated viscosity values and the experimental data [23] for the CaO-MgO-FeO-Fe_2O_3-Al_2O_3-SiO_2 system.