Viscosity of CaO-aluminosilicate Slags

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ABSTRACT

Viscosities of CaO- aluminosilicate ternary melts were estimated using a structure-based model. The model considers depolymerization effects and related breakdown of the silicate network structure on the addition of metal oxides to the melts. The predicted values are in good agreement with the experimental values over the whole temperature and composition range.

INTRODUCTION

An understanding of the mechanism of the metallurgical process phenomena and an optimization of the processes require access to accurate data of the physical properties of the systems involved. Among them, the most important property in the case of molten systems is viscosity, in view of its direct effect on the kinetic conditions of the processes. Although, the viscosity measurements that have been conducted in the case of slag systems are numerous, the data available in literature are still too few to meet the technological demands. this is particularly true in case of complex slag systems, for which, experimental data are very often found to be available only for a few temperature and composition ranges. With the help of a suitable theoretical model, carefully assessed viscosity data can be stored as model parameters so that the values for relevant conditions can be calculated and even be retrieved online.

Quite a few models have been developed to evaluate viscosities of pure liquids as well as solutions. These models could be classified into two categories, that is, models based on fundamental molecular approach and those that employ semi-theoretical procedure. The former type of theories is still under development and hardly meet the technological requirements of today. On the other hand, the semi-theoretical or empirical procedures give satisfactory results. Among such models for high temperature slag systems, the structure-based model proposed by (Bockris and Reddy, 1977) considers depolymerization effects and related breakdown of the glass network structure, on the addition of metal oxides to the melts. The specific expressions of this model were developed and used to calculate the viscosities of various binary silicate systems (Hu and Reddy, 1990, Hu and Reddy, 1988, Raddy and Hebbar, 1991, Hebbar and Reddy 1991), binary borate systems (Shrivastava and Reddy, 1992, Reddy,et.al.,1992), and ternary borosilicate systems (Reddy,et.al.,1997, Zhang and Reddy, 1998, Zhang and Reddy, 1999, Zhang and Reddy, 2004), lead silicate slags (Zhang and Reddy, 2002, Mantha and Reddy, 2005, Yen, et.al., 2006), spent pot liner (Hu and Reddy, 1989), fluidity of slags from waste incinerators (Zhang, et.al., 2002), solid particle effect on viscosity of slags (Reddy and Yen, 1993), semisolid aluminum alloys and composites (Zhang, et.al., 2001, Zhang, et.al., 2001), structure based viscosity model for borate melts (Zhang and Reddy, 2001, Zhang and Reddy, 2005), titania aluminosilicate slags (Yan, et.al., 2018), iron oxide aluminosilicate melts, (Yan et.al., 2019), and structure based model for the aluminosilicate slags (Yan et.al., 2019), as a function of temperature and compositions. Excellent agreement was obtained between the calculated and experimentally determined viscosities. In this paper the viscosity model for SiO_2-Al_2O_3-CaO glass melts and calculated results are presented.

THEORETICAL CONSIDERATIONS

According to Flood's theory (Flood and Grjotheim, 1952), the mole fraction of cation is equal to "electrically equivalent fraction". Thus N_{Si4+} , and N_{Al3+} imply:

$$N_{Si^{4+}} = \frac{4n_{Si^{4+}}}{4n_{Si^{4+}} + 3n_{Al^{3+}}} \tag{1}$$

$$N_{Al^{3+}} = \frac{3n_{Al^{3+}}}{4n_{Si^{4+}} + 3n_{Al^{3+}}} \tag{2}$$

where $3nAl^{3+}$, $4nSi^{4+}$, are the number of equivalents of cations Al^{3+}, Si^{4+}. If expressing nAl^{3+}, and nSi^{4+} in term of X_{Al2O3}, and X_{SiO2}, then, one mole of Al_2O_3 consists of 2 mole Al^{3+} and one mole of SiO_2 consists of one mole Si^{4+}. Thus, $nAl^{3+}=2X_{Al2O3}$ and $nSi^{4+}=X_{SiO2}$. Then, the equation (1) and (2) can be expressed as:

$$N_{Si^{4+}} = \frac{4X_{SiO_2}}{4X_{SiO_2} + 3(2X_{Al_2O_3})} = \frac{2X_{SiO_2}}{2X_{SiO_2} + 3X_{Al_2O_3}} \quad (3)$$

$$N_{Al^{3+}} = \frac{3(2X_{Al_2O_3})}{4X_{SiO_2} + 3(2X_{Al_2O_3})} = \frac{3X_{Al_2O_3}}{2X_{SiO_2} + 3X_{Al_2O_3}} \quad (4)$$

The mole fraction of cation used to estimate several quantities of present model. For the ionic liquids containing holes, Bockris and Reddy (1977) presented the following expression for the viscosity:

$$\eta = \frac{2}{3} N_h R_h (6.28 mkT)^{\frac{1}{2}} \exp\left(\frac{E}{RT}\right) \quad (5)$$

where η: the viscosity (Ns/m^2),

N_h: the number of holes per unit volume (m^{-3}),

R_h: the average radius of the holes (m),

m: the mass of the ionic unit (kg),

k: Boltzmann constant, 1.38×10^{-23} (Joules/moles),

T: the absolute temperature (Kelvin),

E: the energy of ionic unit for viscous flow (Joules/moles),

R: Gas constant, 8.3144 (J/mol.K).

The equation (5) is a general expression, and its application requires calculation of N_h, R_h, m and E. A brief description of the calculation method for these quantities is given below.

Calculation of $(6.28mkT)^{1/2}$

The term $(6.28mkT)^{1/2}$ was rearranged as follows:

$$(6.28 mkT)^{\frac{1}{2}} = \left(6.28 \frac{m\,Av}{Av\,k\,T}\right)^{\frac{1}{2}} kT = \left(6.28 \frac{W}{R}\right)^{\frac{1}{2}} kT^{\frac{1}{2}} \quad (6)$$

where W is the molecular weight of an ionic unit and Av is the Avogadro constant (6.023×1023).

For $CaO-Al_2O_3$ melts, the molecular weight considered as the weight of AlO_6 unit. For $CaO-SiO_2$ melts, the molecular weight considered as the weight of SiO_4 unit. The combined molecular weight is obtained as:

$$W = N_{Si^{4+}} W_{SiO_4} + N_{Al^{3+}} W_{AlO_6} \quad (7)$$

where WSiO4=0.092(kg/mol), and WAlO6=0.123(kg/mol). Thus, the equation (6) can be rewritten as:

$$(6.28W/R)^{\frac{1}{2}} k\, T^{\frac{1}{2}} = (6.28/R)^{\frac{1}{2}} [N_{Si^{4+}}(0.092) + N_{Al^{3+}}(0.123)]^{\frac{1}{2}} k\, T^{\frac{1}{2}} \quad (8)$$

Calculation of R_h

Fürth(1941) has shown that the size of a typical hole in a liquid is the same as the ionic unit and the hole can accommodate an ionic unit. The basic building units are SiO_4 tetrahedra and AlO_6 octahedra for silicate and aluminate melts, respectively.

For SiO_4 tetrahedra, the minimum radii of holes should be equal to the radii of SiO_4 tetrahedra. The ionic radius of oxygen is 1.38Å and the radius of silicon is 0.26Å. According to this structure, the radius of SiO_4 tetrahedra can be deduced as 3.07Å. For AlO_6 octahedra, the minimum radii of holes should be equal to the radii of AlO_6 octahedra. The ionic radius of oxygen is 1.40Å and the radius of aluminum is 0.38Å. Therefore, the radius of AlO_6 octahedra can be calculated as 3.38Å.The radius of an ionic unit in the system containing SiO_2 and Al_2O_3 can be expressed by equation (9):

$$R_h = N_{Si^{4+}} R_{h,SiO_4} + N_{Al^{3+}} R_{h,AlO_6} \tag{9}$$

Calculation of N_h:

The number of holes per unit volume was expressed in terms of NO^0, where NO^0 is mole fraction of bridging oxygen in melts. This calculation involves that the number of holes is equal to SiO_4 and AlO_6 ionic units present in the melts and that all the holes are occupied by the ionic species. Thus,

$$N_h = NO^0 \cdot A_v = 6.023 \times 10^{23} \cdot NO^0 \tag{10}$$

Substituting (6.28mKT)1/2, Rh and Nh in equation (5), the following viscosity expression in Ns/m^2 for the system containing SiO_2 and Al_2O_3 as network formers can be obtained:

$$\begin{aligned}\eta = 4.8184 \times 10^{-10}(6.14X_{SiO_2} + 10.14X_{Al_2O_3}) \cdot (0.184X_{SiO_2} + 0.369X_{Al_2O_3})^{\frac{1}{2}} \\ \cdot (2X_{SiO_2} + 3X_{Al_2O_3})^{-\frac{3}{2}} \cdot NO^0 \cdot T^{\frac{1}{2}} \cdot \exp(\frac{E}{RT})\end{aligned} \tag{11}$$

estimate viscosity by eq. (11), NO^0 and E need to be calculated. The calculation of NO^0 and E are described below.

Calculation of NO^0:

If B mole of Al_2O_3 is mixing with S mole of SiO_2 and M mole of CaO, the charge and mass balance considerations, respectively, require that.

$$nO^0 = 3B + 2S - \frac{1}{2}nO^- \tag{12}$$

$$nO^{2-} = M - \frac{1}{2}nO^- \tag{13}$$

where nO^0: the number of bridging oxygen bonded to two silicon or aluminum atoms,

nO^-: the number of non-bridging oxygen bonded to only one silicon or aluminum atom,

nO^{2-}: the number of free oxygen ions,

The total number of anions can be calculated as:

$$nO^0 + nO^- + nO^{2-} = 3B + 2S + M \tag{14}$$

The mole fraction of bridging oxygen of total anions can be calculated by using Yokokawa's model (1969) and expressed as eq. (15).

$$NO^0 = \frac{nO^0}{total\ number\ of\ oxygen\ anions} = \frac{3B + 2S - \frac{1}{2}nO^-}{3B + 2S + M} \tag{15}$$

Considering the mole fraction in liquid melts, then

$$NO^0 = \frac{3X_{Al_2O_3} + 2X_{SiO_2} - \frac{1}{2}nO^-}{3X_{Al_2O_3} + 2X_{SiO_2} + X_{CaO}} = \frac{3X_{Al_2O_3} + 2X_{SiO_2} - \frac{1}{2}nO^-}{2X_{Al_2O_3} + X_{SiO_2} + 1} \tag{16}$$

The nO^- further can be calculated by eq. (17).

$$[1-\exp(\frac{\Delta G^0}{RT})](nO^-)^2 - 2(X_{SiO_2} + 2X_{Al_2O_3} + 1)(nO^-) + 4(2X_{SiO_2} + 3X_{Al_2O_3})(1 - X_{SiO_2} - X_{Al_2O_3}) = 0 \qquad (17)$$

ΔG^0 is the standard Gibbs energy of depolymerization reaction (Joules/mole). For SiO_2-CaO, and Al_2O_3-CaO melts, the depolymerization reactions can be described as reaction (18) and (19) respectively.

$SiO_2(l) + 2CaO(l) = 2CaO\cdot SiO_2(l)$ (18)

$Al_2O_3(l) + CaO(l) = CaO\cdot Al_2O_3(l)$ (19)

ΔG^0 can be expressed as:

$$\Delta G^o = N_{Al}3+\Delta G^o_{Al} + N_{Si}4+\Delta G^o_{Si} \qquad (20)$$

where ΔG^0_{Al} and ΔG^0_{Si} are the Gibbs energy for reactions (18) and (19) respectively.

Calculation of E

The energy of ionic unit for viscous flow is the function of composition and temperature. In the case of silicate melts the energy term is considered to be composed of energy required to break the silicate bonds in SiO_4^{4-} tetrahedral units and move the tetrahedral unit into the adjacent hole. Energy required for the jump is small. This has been determined to be equal to $3.3RT_m$, where R is the gas constant and T_m is the melting temperature of the slag system. Hence, E is dependent on the number of Si-O-Si bonds to be broken.

The value of E was calculated based on the experimental viscosity data choosing from literature (Urbain, et al., 1982, Kozakevitch, 1960, Machin and Yee, 1948) by using equation (11). A correlation of these E values as a function of temperature was made. As observed, the energy term can be expressed as equation (21):

$$E = f_0 + f_1 T + f_2 T^2 \qquad (21)$$

where f_i are constants and function of composition. The constants f_i are correlated with mole fraction of CaO and expressed by polynomial expression which are given by equation (22).

$$f_i = a_i + b_i X_{CaO} + c_i X_{CaO}^2 \qquad (22)$$

where a_i, b_i and c_i are function of r, the ratio between the molar fraction of SiO_2 and the sum of the molar fractions of SiO_2 and of Al_2O_3, and

$a_0 = 3050672.55 + 33225477.52r - 100908358.79r^2 + 67346588.18r^3$

$b_0 = -7755748.01 - 210431186.31r + 579249233.04r^2 - 369704483.2074r^3$

$c_0 = 6172363.82 + 282764773.05r - 741066602.32r^2 + 459301295.09r^3$

$a_1 = -2791.73 - 38511.50r + 114603.27r^2 - 75668.14r^3$

$b_1 = 7737.08 + 245431.66r - 668916.39r^2 + 423976.96r^3$

$c_1 = -6245.89 - 331028.64r + 863749.57r^2 - 533355.28r^3$

$a_2 = 0.696 + 10.914r - 31.981r^2 + 20.987r^3$

$b_2 = -1.916 - 70.170r + 189.968r^2 - 119.997r^3$

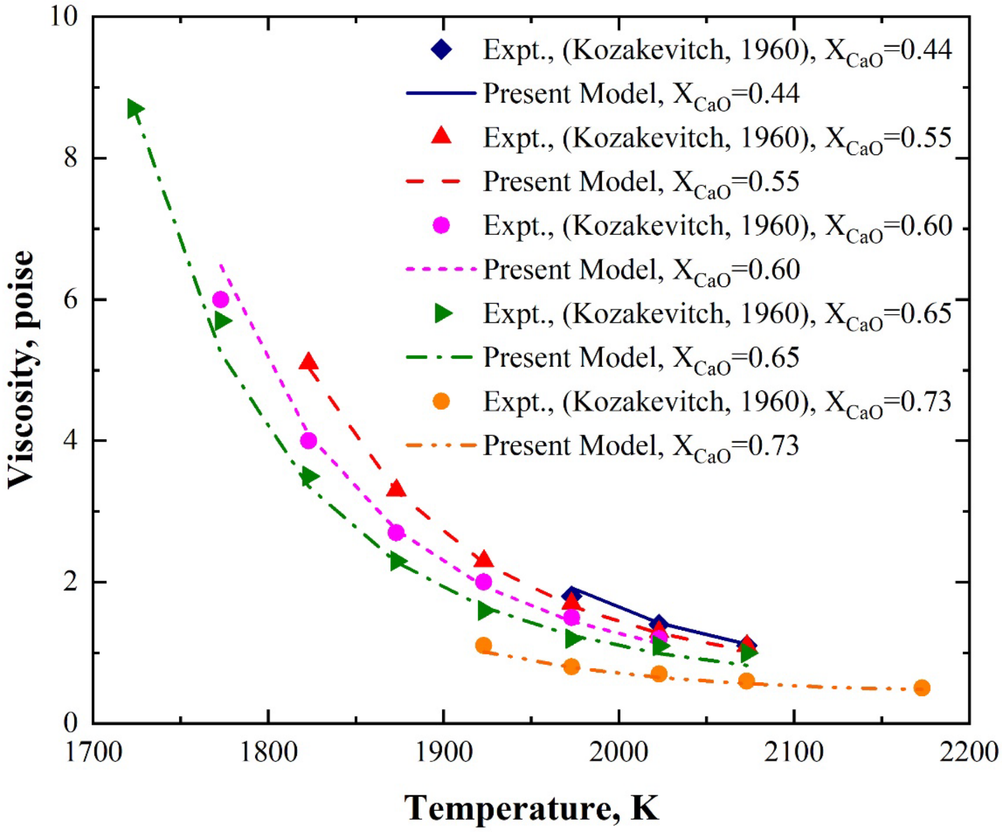
$c_2 = 1.571 + 95.057r - 247.522r^2 + 152.695r^3$

The calculated E value along with the NO^0 value were substituted into equation (11) to give the viscosity of the melt at a particular temperature and composition. The viscosity for multi component silicate melts was calculated using Weymann-Frenkel equation with empirical parameters (Kondratiev and Jak, 2001). In this paper, the calculated viscosities of several melts are given in the following results and discussion section.

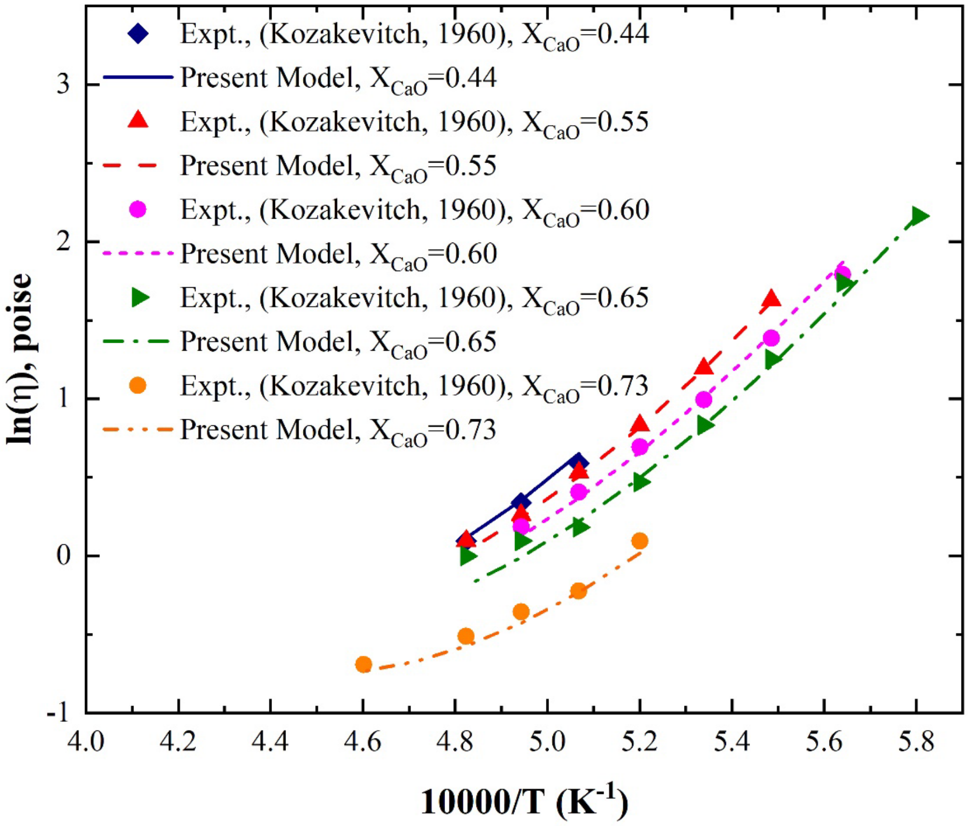
RESULTS AND DISCUSSION

Al_2O_3-CaO binary melts:

When r [SiO_2 / (SiO_2 + Al_2O_3)] = 0, the present model predicts viscosity values of the Al_2O_3-CaO binary melts. The experimental viscosity data and calculated results for r = 0 are shown in Fig. 1a and b. The calculated results are in good agreement with the experimental data. The viscosities decrease with the increase in temperature of the melts for all compositions. As can be seen from the figure, the viscosities decrease with increase in X_{CaO}.



(a)



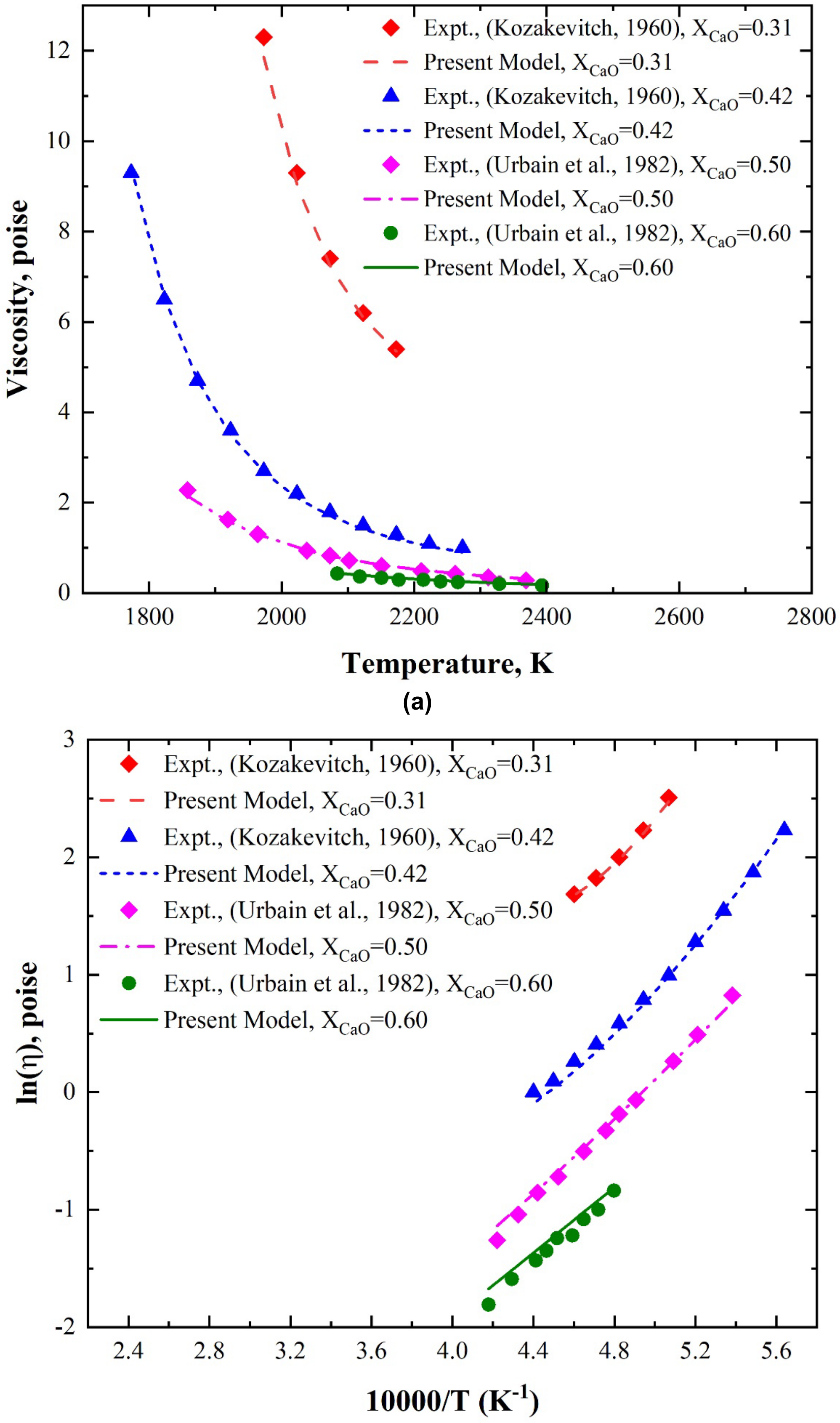
(b)

Fig.1:(a) Viscosity (η) vs temperature (T), and (b) ln(η) vs temperature (1/T) for CaO-Al_2O_3 melts. at r= [SiO_2 / (SiO_2 + Al_2O_3)] =0

SiO_2-CaO binary melts:

When r [SiO_2 / (SiO_2 + Al_2O_3)] = 1, the present model predicts viscosity values of the SiO_2-CaO binary melts. The experimental viscosity data and calculated results for r = 1 are shown in Fig. 2a and b. Again, the viscosities decrease with the increase in X_{CaO} and temperature of the melts for all compositions. The calculated results are in good agreement with the experimental data.

The viscosities of SiO_2-CaO and Al_2O_3-CaO binary melts as function of X_{CaO} at 2073 K is shown in Fig. 3. As can be seen at constant temperature, the viscosities of SiO_2-CaO melts decrease much significantly with increase X_{CaO} compared with that of Al_2O_3-CaO melts.



(a)

(b)

Fig.2: (a) Viscosity (η) vs temperature (T), and (b) ln(η) vs temperature (1/T) for CaO-SiO_2 melts. at r= [SiO_2 / (SiO_2 + Al_2O_3)] =1

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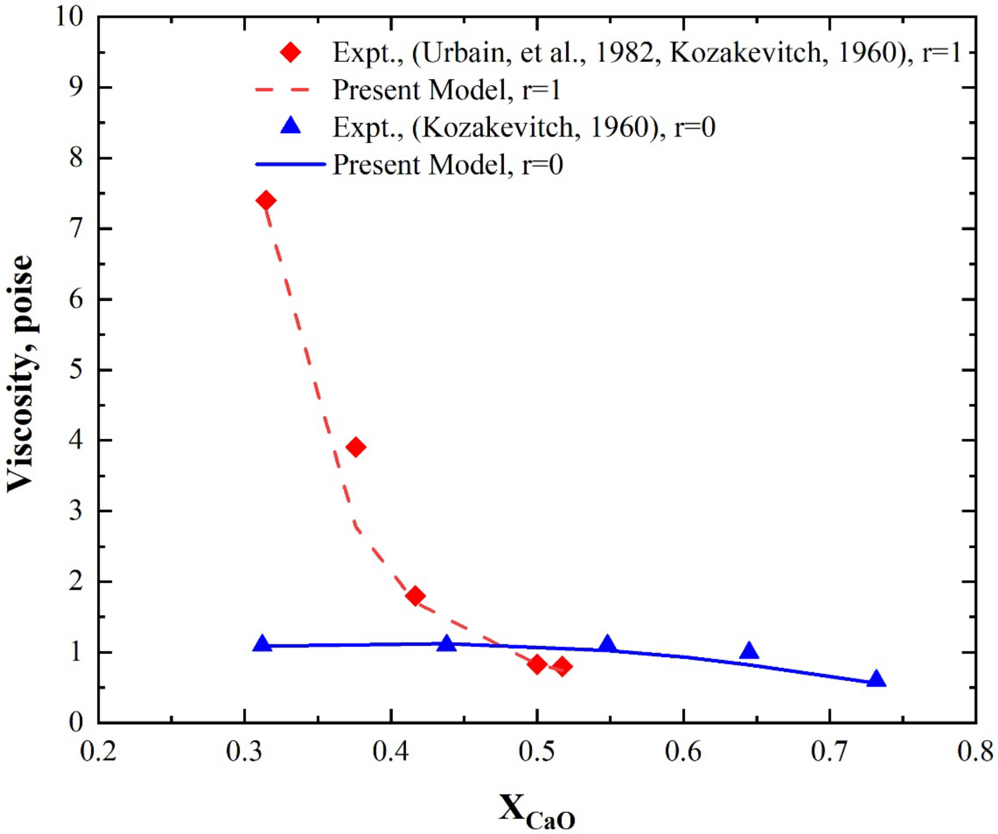
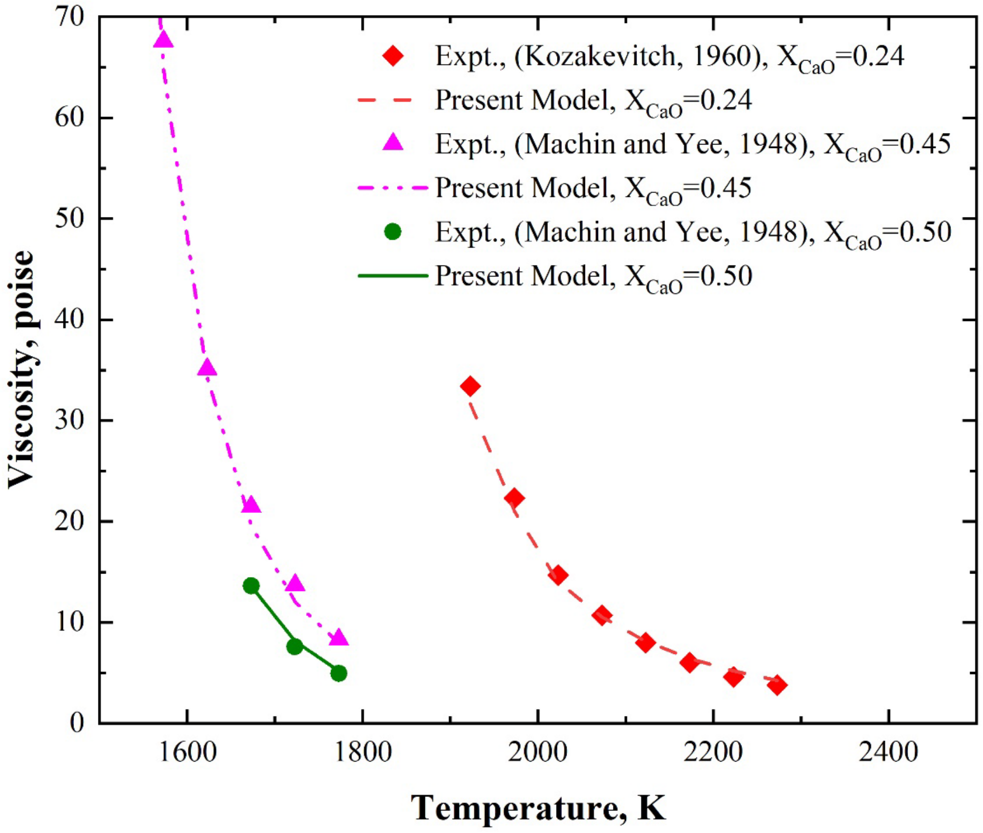


Fig.3: Viscosity (η) vs CaO mole fraction (X_{CaO}) for CaO-Al_2O_3 and CaO-SiO_2 melts at 2073K at r= [SiO_2 / (SiO_2 + Al_2O_3)]=0 and at r= [SiO_2 / (SiO_2 + Al_2O_3)]=1

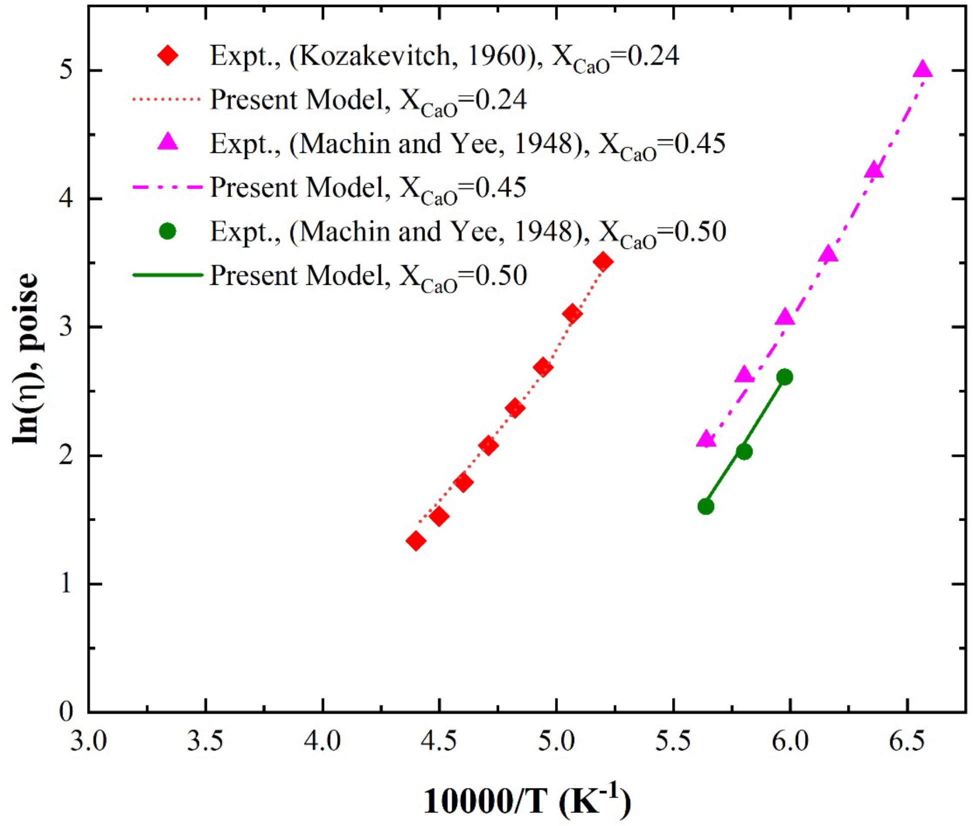
SiO_2-Al_2O_3-CaO ternary melts:

Fig. 4a and b and Fig. 5a and b show the experimental viscosity data and calculated data for r=0.75 and 0.82, respectively. As can be seen, viscosities decrease significantly with the increase in temperature for all compositions. Viscosities decrease with increase in X_{CaO}. Good agreement between calculated and experimental data can be seen in the temperature and composition range of experimental data.

The viscosities of SiO_2-Al_2O_3-CaO melts as function of (X_{Al2O3}/X_{CaO}) ratio at 1773K and X_{SiO2}=0.58 are shown in Fig. 6. As expected, it can be seen that at constant temperature and mole fraction of SiO_2, the viscosities of ternary melts increase significantly with increases in (X_{Al2O3}/X_{CaO}) ratio.

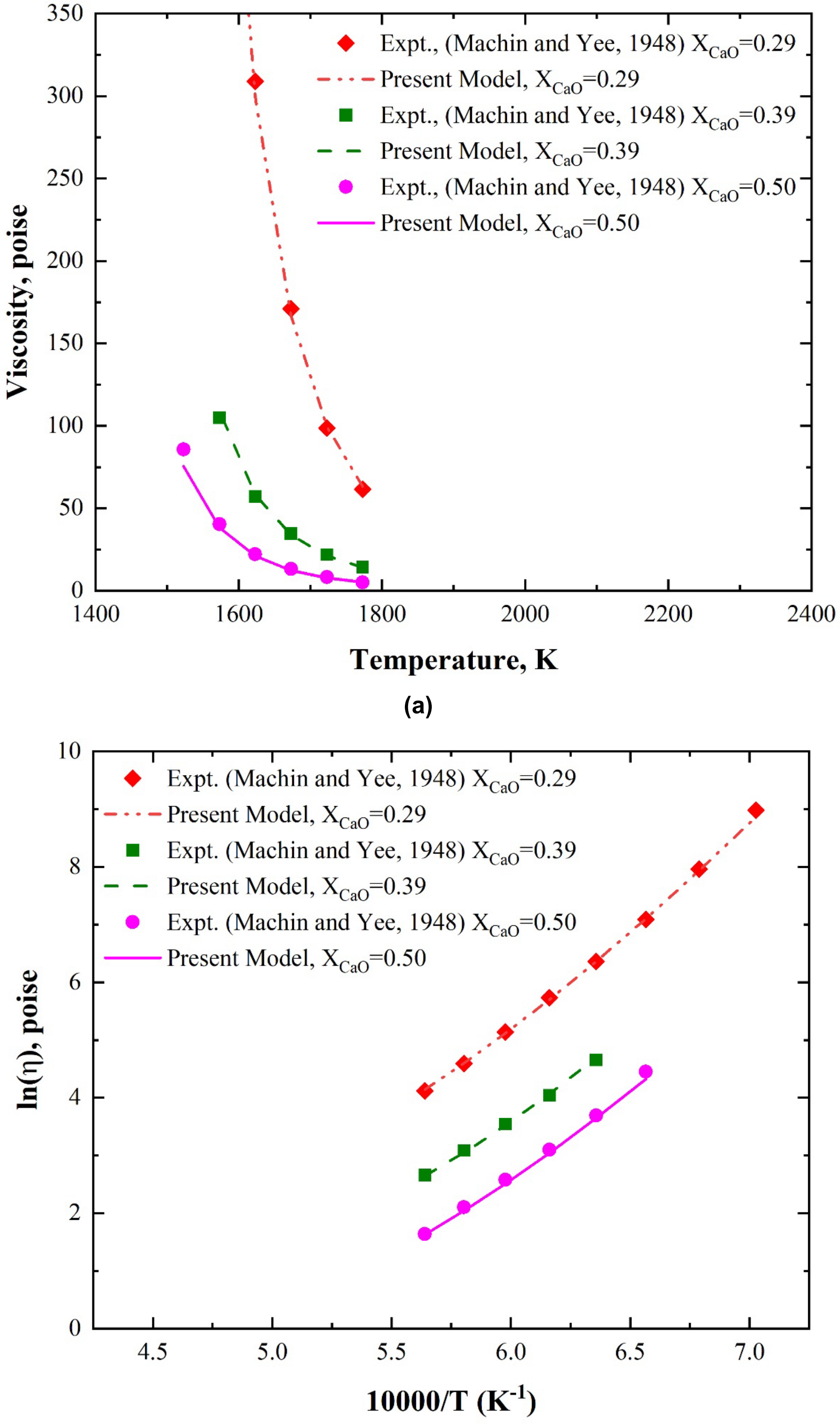


(a)



(b)

Fig.4: (a) Viscosity (η) as a function of temperature (T), and (b) ln(η) as a function of 1/T at r=0.75 for SiO_2-Al_2O_3-CaO ternary melts r= [SiO_2 / (SiO_2 + Al_2O_3)]



(a)

(b)

Fig.5: (a) Viscosity (η) as a function of temperature (T), and (b) ln(η) as a function of 1/T at r=0.82 for SiO_2-Al_2O_3-CaO ternary melts r= [SiO_2 / (SiO_2 + Al_2O_3)]

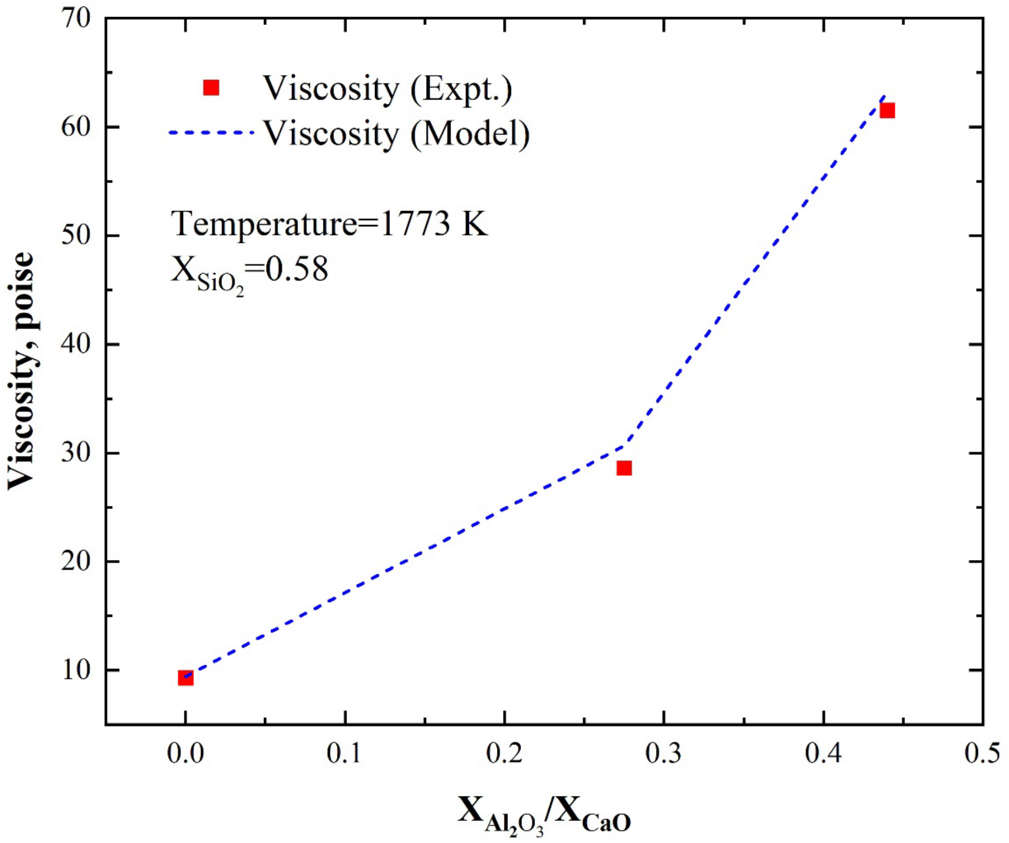


Fig.6: Viscosity vs (X_{Al2O3}/X_{CaO}) ratio for SiO_2-Al_2O_3-CaO ternary melts at 1773K and X_{SiO2}=0.58

Performance of Present Model

The model calculated viscosity values are compared with the experimental values in the SiO_2-Al_2O_3-CaO system and its subsystems over a wide composition and temperature ranges summarized in Fig. 7. The present model reproduces most of the experimental data within experimental uncertainties, the average of relative value for this model was found to be 18.2%. The model calculated data are in good agreement with the experimental data over a large temperature and composition range.

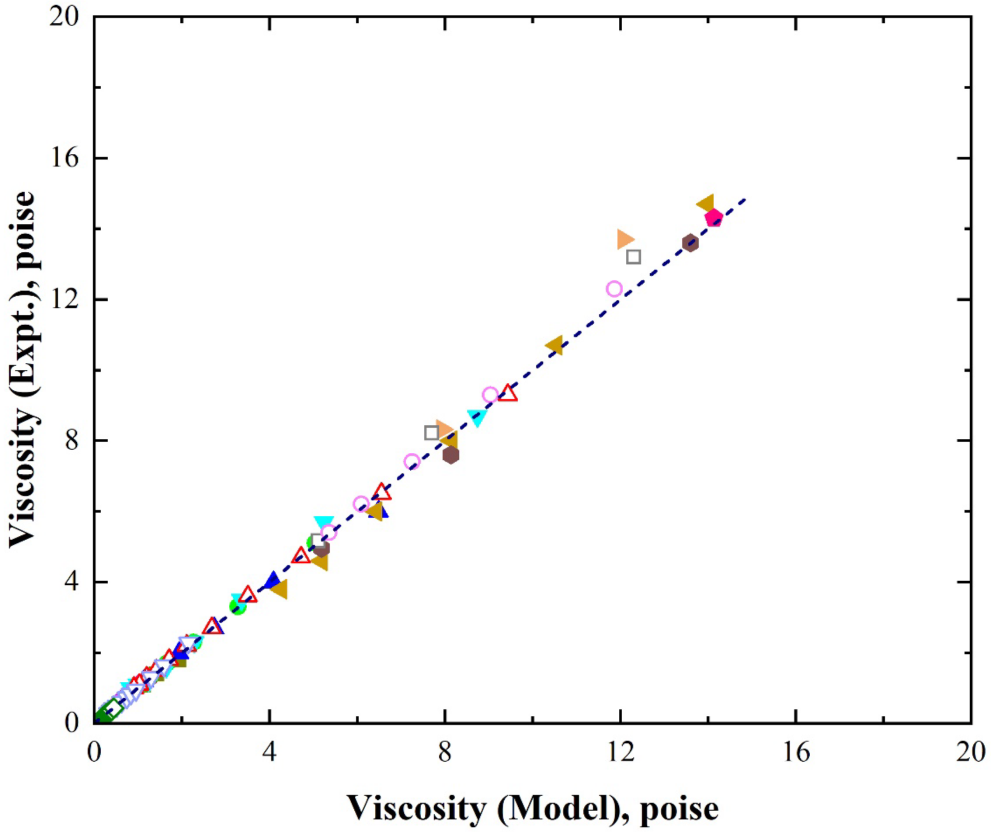


Fig 7. Comparison of experimental data and model calculated data for viscosities for SiO_2-Al_2O_3-CaO ternary melts for different ratios r= [SiO_2 / (SiO_2 + Al_2O_3)] and temperatures.

(! Expt., (Kozakevitch, 1960), X_{CaO}=0.44, r=0; , Expt., (Kozakevitch, 1960), X_{CaO}=0.55, r=0; 7 Expt., (Kozakevitch, 1960), X_{CaO}=0.60, r=0; B Expt., (Kozakevitch, 1960), X_{CaO}=0.65, r=0; Λ Expt., (Kozakevitch, 1960), X_{CaO}=0.73, r=0; Ω Expt., (Kozakevitch, 1960), X_{CaO}=0.24, r=0.75; β Expt., (Machin and Yee, 1948), X_{CaO}=0.45, r=0.75; μ Expt., (Machin and Yee, 1948), X_{CaO}=0.50, r=0.75; ξ Expt., (Machin and Yee, 1948) X_{CaO}=0.29, r=0.82; □ Expt., (Machin and Yee, 1948) X_{CaO}=0.39, r=0.82; ∀ Expt., (Machin and Yee, 1948) X_{CaO}=0.50, r=0.82; – Expt., (Kozakevitch, 1960), X_{CaO}=0.31, r=1; 8 Expt., (Kozakevitch, 1960), X_{CaO}=0.42, r=1; X Expt., (Urbain et al., 1982), X_{CaO}=0.50, r=1; M Expt., (Urbain et al., 1982), X_{CaO}=0.60, r=1; - - - - y=x)

CONCLUSIONS

A structure-based viscosity model was developed for CaO-aluminosilicate ternary melts, which can predict not only viscosities of SiO_2-Al_2O_3-CaO ternary glass melts. The experimental viscosity data and calculated results show that viscosities of SiO_2-Al_2O_3-CaO glass melts decrease with the increase in X_{CaO} and temperature of the melts. The model calculated data are in good agreement with the experimental data over a large temperature and composition range.

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