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Evaluation of the Surface Tension of Ternary Silicate Melts Containing Al_2O_3 , CaO , FeO , MgO or MnO

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A thermodynamic model for determining the surface tension of molten ionic mixtures derived by considering the ionic radii was extended to ternary silicate melts. The composition dependence of the surface tensions of molten silicates in ternary systems, was reproduced by the present model using surface tension information, the molar volumes of pure oxides, as well as the anionic and cationic radii of the component oxides in the system.

KEY WORDS: surface tension; thermodynamics; silicate; ionic radius; molten slag.

1. Introduction

The surface tension of molten silicates is a key parameter in industrial processes at high temperatures, and a large volume of experimental surface tension data has been reported for silicate melts.¹⁾ Despite the development of several models^{2–4)} for evaluating the surface tension of molten oxide mixtures, which were in fact thermodynamically-based on Butler's equation⁵⁾ with a procedure proposed by Speiser *et al.*,⁶⁾ a number of problems still remain. These problems include for example discrepancies between the calculated results of the surface tension of molten slag and literature values, a lack of thermodynamic data in multi-component oxide and fluoride systems, *etc.*

Tanaka *et al.*⁷⁾ recently developed a model for evaluating the surface tension of molten ionic mixtures, based on the ratio of cationic and anionic radii as the model parameters. This model is characterized by the simplicity with which it can be extended to multi-component systems, because the calculations are based only on the surface tension, molar volume and ionic radii of the pure components in the system. This model has also been applied to binary silicate melts.⁷⁾ The calculated results reproduced the composition dependence of the surface tension of silicate melts in certain binary systems. In this calculation, however, the hypothetical surface tensions of certain pure molten oxides below their melting points are necessary, because oxides such as CaO and MgO have rather high melting points. In addition, there is a lack of reliable experimental data for the surface tension of some pure oxide melts. Therefore, the surface tensions of some pure oxides have to be treated as model parameters. Furthermore, the cation to anion ratio in

SiO_2 was also used as a parameter in this model, since the minimum anion unit in SiO_2 was considered to be SiO_4^{4-} , although, the effective radius of SiO_4^{4-} is unknown.

In this work, the temperature dependence of the surface tension was evaluated for pure oxides not only in the liquid phase, but also below their respective melting points, to extend the above model to address the surface tension calculations in molten ternary silicates. The calculated surface tensions were compared with the experimentally obtained data for binary and ternary silicate melts composed of SiO_2 , Al_2O_3 , CaO , FeO , MgO or MnO . Here, the ratio of the cationic and anionic radii for SiO_2 was also discussed, along with the evaluation above.

2. Model for Evaluating Surface Tension of Molten Silicates

2.1. AX–BY System

The surface tension (σ) of the AX–BY melt is calculated from Eqs. (1) and (2)⁷⁾:

$$\sigma = \sigma_{\text{AX}}^{\text{Pure}} + \frac{RT}{A_{\text{AX}}} \ln \frac{M_{\text{AX}}^{\text{Surf}}}{M_{\text{AX}}^{\text{Bulk}}} \dots\dots\dots(1)$$

$$\sigma = \sigma_{\text{BY}}^{\text{Pure}} + \frac{RT}{A_{\text{BY}}} \ln \frac{M_{\text{BY}}^{\text{Surf}}}{M_{\text{BY}}^{\text{Bulk}}} \dots\dots\dots(2)$$

where

$$M_{AX}^P = \frac{\frac{R_A}{R_X} \cdot N_{AX}^P}{\frac{R_A}{R_X} \cdot N_{AX}^P + \frac{R_B}{R_Y} \cdot N_{BY}^P},$$

$$M_{BY}^P = \frac{\frac{R_B}{R_Y} \cdot N_{BY}^P}{\frac{R_A}{R_X} \cdot N_{AX}^P + \frac{R_B}{R_Y} \cdot N_{BY}^P}.$$

Subscripts A and B are cations, while X and Y are anions. Superscripts ‘Surf’ and ‘Bulk’ indicate the surface and bulk, respectively. R is the gas constant, T is the absolute temperature, σ_i^{Pure} is the surface tension of pure molten oxide i ($i=AX$ or BY), which is treated as a model parameter. $A_i = N_0^{1/3} \cdot V_i^{2/3}$ corresponds to the molar surface area in a monolayer of pure molten oxide i (N_0 : Avogadro’s number, V_i : molar volume of pure molten oxide i). N_i^P is the mole fraction of oxide i in phase P (P=Surf or Bulk). R_A and R_B are the radii of cations A and B, while R_X and R_Y are the radii of anions X and Y. For example, in the case of the SiO_2 –CaO system,

$$R_A = R_{\text{Si}}^{4+}, \quad R_B = R_{\text{Ca}}^{2+} \dots\dots\dots(3)$$

$$R_X = R_{\text{SiO}_4}^{4-}, \quad R_Y = R_{\text{O}}^{2-} \dots\dots\dots(4)$$

where it is assumed that Si^{4+} is cation, and SiO_4^{4-} is anion unit in order to evaluate the ratio of cationic and anionic radii for SiO_2 because the complex ion SiO_4^{4-} exists in molten SiO_2 system as mentioned in Ref. 7). However, $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}$ must be treated as a parameter in this model since there is no precise information on the apparent effective radii of complex anion $R_{\text{SiO}_4}^{4-}$.

The above Eqs. (1) and (2) have been derived from Butler’s equation⁶⁾ by considering assumptions [1] and [2].⁷⁾

- [1] It is well known that molten ionic mixtures readily undergo surface relaxation processes, such as the spontaneous changes in ionic distance at the surface, enabling the energetic state of the surface to approach the bulk state. Thus, the contribution from excess Gibbs energy terms is neglected in Butler’s equation.
- [2] In ionic substances, it is well known that the ionic structures depend upon the ratio of the cationic to anionic radii. In order to evaluate the ionic structures and physico-chemical properties of ionic materials, we should consider the ratio of the cationic and anionic radii.

2.2. AX–BY–CZ System

The surface tension (σ) equations for the AX–BY–CZ melt are expressed on the basis of Eqs. (1) and (2) as follows:

$$\sigma = \sigma_{AX}^{\text{Pure}} + \frac{RT}{A_{AX}} \ln \frac{M_{AX}^{\text{Surf}}}{M_{AX}^{\text{Bulk}}} \dots\dots\dots(5)$$

Table 1. Ionic radii of cations and oxygen ion (Å).

Si^{4+}	0.42
Al^{3+}	0.51
Ca^{2+}	0.99
Fe^{2+}	0.74
Mg^{2+}	0.66
Mn^{2+}	0.80
O^{2-}	1.44

Table 2. Molar volume of pure components (m^3/mol).

Oxide	Temperature (K) dependence of molar volume (m^3/mol)
SiO_2	$27.516 \{1 + 1 \cdot 10^{-4} (T - 1773)\} \cdot 10^{-6}$
Al_2O_3	$28.3 \{1 + 1 \cdot 10^{-4} (T - 1773)\} \cdot 10^{-6}$
CaO	$20.7 \{1 + 1 \cdot 10^{-4} (T - 1773)\} \cdot 10^{-6}$
FeO	$15.8 \{1 + 1 \cdot 10^{-4} (T - 1773)\} \cdot 10^{-6}$
MgO	$16.1 \{1 + 1 \cdot 10^{-4} (T - 1773)\} \cdot 10^{-6}$
MnO	$15.6 \{1 + 1 \cdot 10^{-4} (T - 1773)\} \cdot 10^{-6}$

$$\sigma = \sigma_{BY}^{\text{Pure}} + \frac{RT}{A_{BY}} \ln \frac{M_{BY}^{\text{Surf}}}{M_{BY}^{\text{Bulk}}} \dots\dots\dots(6)$$

$$\sigma = \sigma_{CZ}^{\text{Pure}} + \frac{RT}{A_{CZ}} \ln \frac{M_{CZ}^{\text{Surf}}}{M_{CZ}^{\text{Bulk}}} \dots\dots\dots(7)$$

where

$$M_{AX}^P = \frac{\frac{R_A}{R_X} \cdot N_{AX}^P}{\frac{R_A}{R_X} \cdot N_{AX}^P + \frac{R_B}{R_Y} \cdot N_{BY}^P + \frac{R_C}{R_Z} \cdot N_{CZ}^P},$$

$$M_{BY}^P = \frac{\frac{R_B}{R_Y} \cdot N_{BY}^P}{\frac{R_A}{R_X} \cdot N_{AX}^P + \frac{R_B}{R_Y} \cdot N_{BY}^P + \frac{R_C}{R_Z} \cdot N_{CZ}^P},$$

$$M_{CZ}^P = \frac{\frac{R_C}{R_Z} \cdot N_{CZ}^P}{\frac{R_A}{R_X} \cdot N_{AX}^P + \frac{R_B}{R_Y} \cdot N_{BY}^P + \frac{R_C}{R_Z} \cdot N_{CZ}^P}.$$

Subscripts A, B and C are cations, and X, Y and Z are anions. Superscripts P=‘Surf’ and ‘Bulk’ indicate the surface and bulk, respectively. R_A , R_B and R_C correspond to the radii of cations A, B and C, and R_X , R_Y and R_Z are the radii of anions X, Y and Z. The ionic radii data were taken from the data compiled by Shannon,⁸⁾ while the molar volumes of the pure oxides, as recommended by Mills and Keene,⁹⁾ were used in the present calculations. These values are listed in **Tables 1** and **2**, respectively.

Table 3. Systems and temperatures used for evaluation of model parameters.

System	Temperature (K)
SiO ₂ -CaO	1843
SiO ₂ -FeO	1843
SiO ₂ -MgO	1843
SiO ₂ -MnO	1843
SiO ₂ -Al ₂ O ₃ -CaO	1873
SiO ₂ -Al ₂ O ₃ -FeO	1473, 1573
SiO ₂ -Al ₂ O ₃ -MnO	1873
SiO ₂ -CaO-FeO	1573, 1623, 1673
SiO ₂ -CaO-MgO	1873
SiO ₂ -CaO-MnO	1773
SiO ₂ -FeO-MgO	1623, 1673, 1823
SiO ₂ -FeO-MnO	1673

3. Results and Discussions

The surface tensions of the pure oxides used as parameters in the present model were evaluated by minimizing the differences between the calculated σ results and the literature values.^{1,10-31} For this purpose, it is advisable to use the literature data in as many systems as possible, and for as many temperatures. Therefore, for evaluation purposes, almost all of the available literature data for ternary silicate systems containing Al₂O₃, CaO, FeO, MgO or MnO, were collected over the wide composition and temperature ranges in SLAG ATLAS 2nd Edition.^{1,13-31} The selected oxide systems and targeted temperatures are listed in **Table 3**. The total number of literature values is 457. In this study, the value σ_{SiO_2} (mN/m)=243.2+0.031*T* (K) was used as the surface tension of molten pure SiO₂, as suggested by NIST.³²

In addition, the ratio of the cationic to anionic radii for SiO₂ was reassessed through the above evaluation. It is generally known that SiO₄⁴⁻ ions exist as the minimum anionic unit in SiO₂ in silicate slags.³³⁻³⁵ In our previous work,⁷ the $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}$ ratio in binary SiO₂-based slags was assumed to be 0.5 by considering the structural features of the silicates. Here, we compared the results calculated using $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ with those obtained using $R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$. The latter ratio ($R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$) signifies that there is no consideration of the anion unit SiO₄⁴⁻ in molten silicate systems.

Figure 1 shows the calculated results obtained for the SiO₂-Al₂O₃-CaO system at 1873 K using $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ and $R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$, taken after evaluating the surface tensions of pure oxides, as explained in detail below. The iso-surface tension curves corresponding to $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ reproduce the composition dependence of the surface tension^{10,11} better than those of $R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$. The same trends were observed in other silicate systems. The average errors in all systems assessed using Eq. (8) are 4.2% for $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ and 4.8% for $R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$.

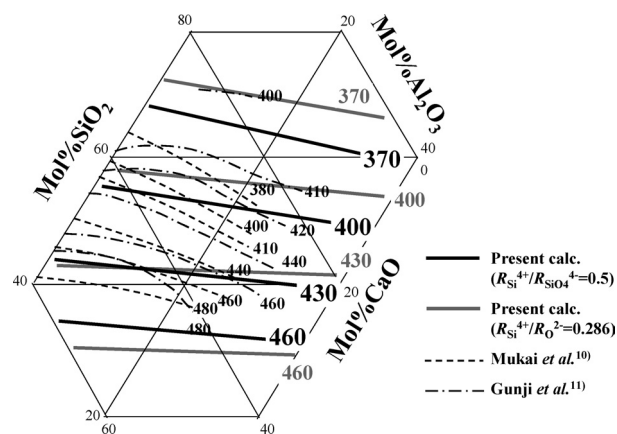
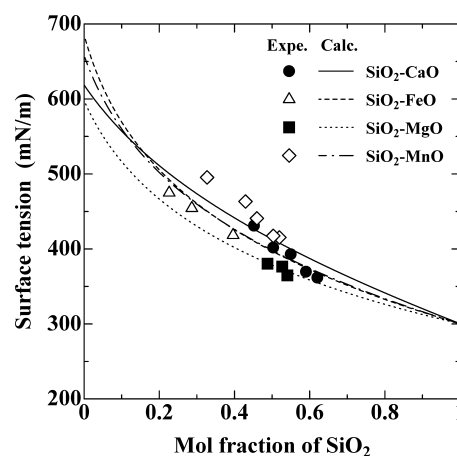

Fig. 1. Surface tension (mN/m) of molten SiO₂-Al₂O₃-CaO system at 1873 K. Comparison between the calculated results obtained for $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ and for $R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$.

Table 4. Model parameters (temperature dependence of surface tension for pure oxides).

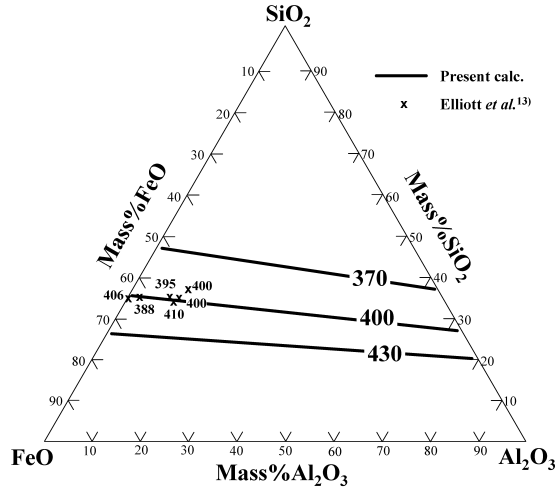
Oxide	Temperature (K) dependence of surface tension (mN/m)	
Al ₂ O ₃	1024 - 0.177 <i>T</i>	1473 - 1873 K
CaO	791 - 0.0935 <i>T</i>	1573 - 1873 K
FeO	504 + 0.0984 <i>T</i>	1473 - 1843 K
MgO	1770 - 0.636 <i>T</i>	1623 - 1873 K
MnO	988 - 0.179 <i>T</i>	1673 - 1873 K


Fig. 2. Surface tension of molten SiO₂ based binary system at 1843 K.

$$\text{Average error} = \frac{1}{N} \sum_{i=1}^N \left| \frac{\sigma_{\text{Calc}} - \sigma_{\text{Expe}}}{\sigma_{\text{Expe}}} \right| \times 100 (\%) \dots (8)$$

In Eq. (8), the σ_{Expe} and σ_{Calc} values correspond to the experimental surface tension values in the literature^{1,10-31} and the calculated surface tension, respectively. *N* corresponds to the number of the literature data. It is found from these results that $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ is adequate for the present calculation of the surface tension of silicate melts. On the other hand, $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}$ is considered to be smaller than $R_{\text{Si}}^{4+}/R_{\text{O}^{2-}}=0.286$ when taking into account the sizes of the complex ion SiO₄⁴⁻ and O²⁻ from the geometric viewpoint. $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}$, however, may be far bigger than the ratio esti-

(a)



(b)

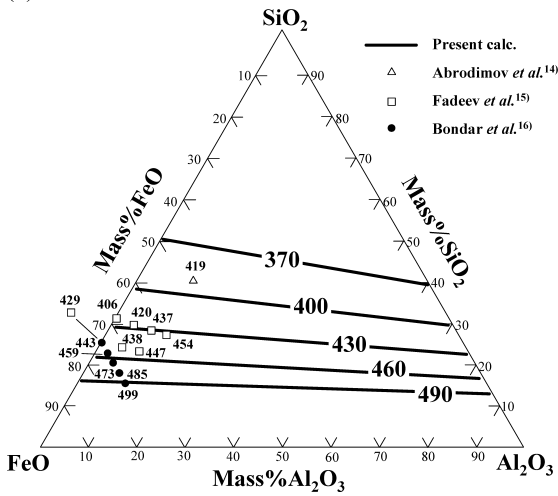


Fig. 3. Surface tension (mN/m) of molten SiO_2 - Al_2O_3 -FeO system at (a) 1473 and (b) 1573 K.

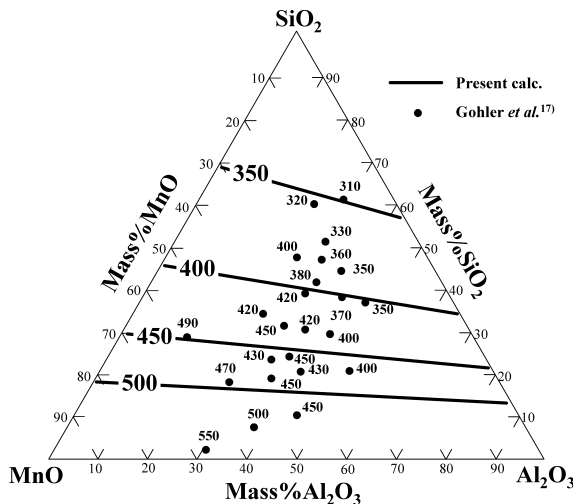
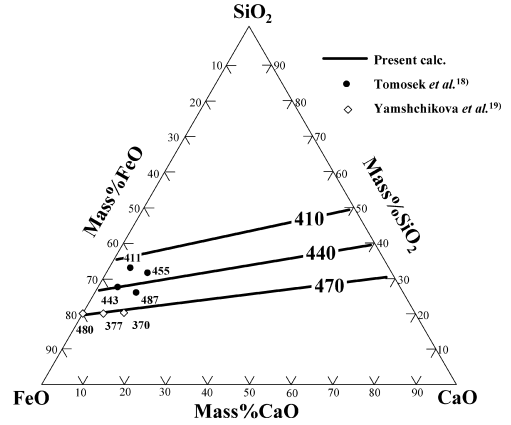


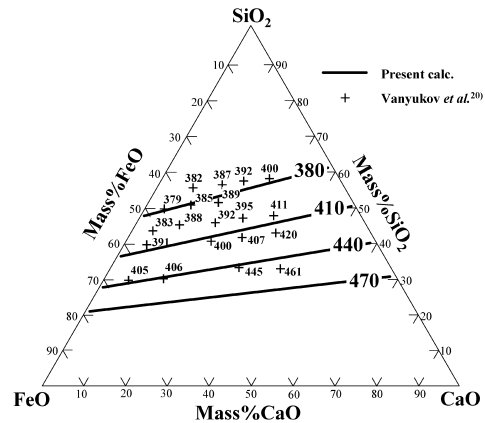
Fig. 4. Surface tension (mN/m) of molten SiO_2 - Al_2O_3 -MnO system at 1873 K.

mated geometrically since the apparent effective radii of complex anions reported in the literature³⁶⁾ are generally much smaller than the radii predicted geometrically. In addition, it is not clear whether $R_{\text{Si}^{4+}}/R_{\text{SiO}_4^{4-}}$, which is obtained by assuming that Si^{4+} is cation and SiO_4^{4-} is anion unit in SiO_2 , is the ratio that actually reflects the micro-structure of

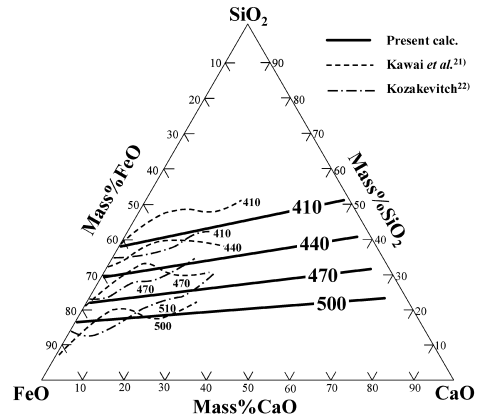
(a)



(b)



(c)



(d)

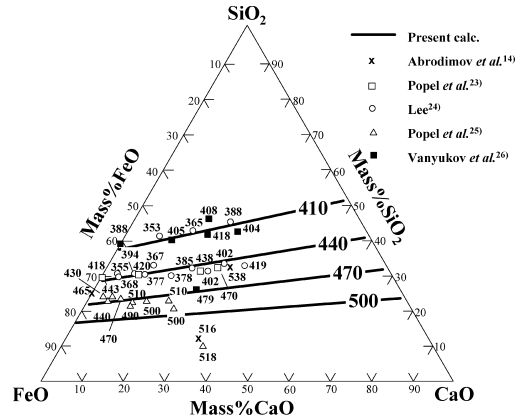


Fig. 5. Surface tension (mN/m) of molten SiO_2 -CaO-FeO system at (a) 1573, (b) 1623, (c) and (d) 1673 K.

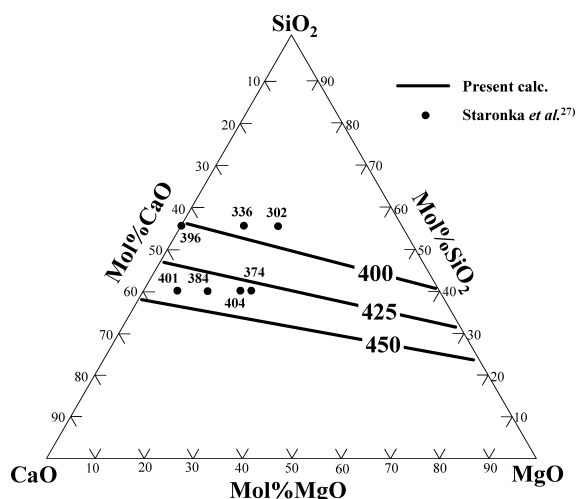


Fig. 6. Surface tension (mN/m) of molten SiO_2 -CaO-MgO system at 1873 K.

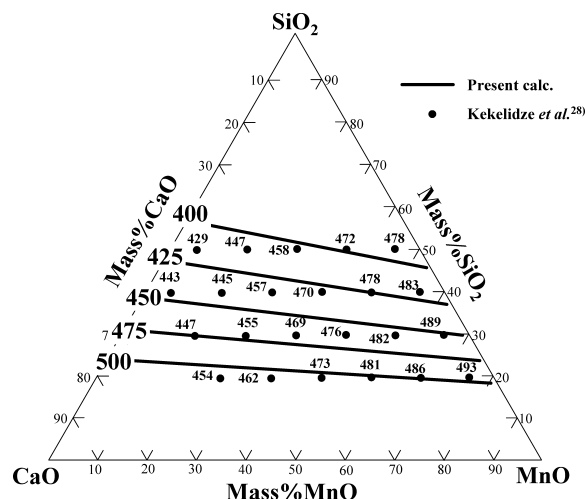


Fig. 7. Surface tension (mN/m) of molten SiO_2 -CaO-MnO system at 1773 K.

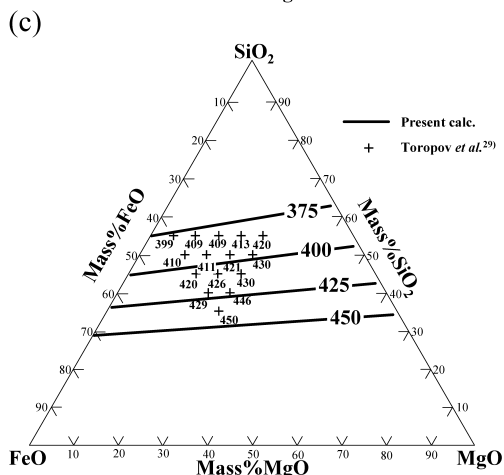
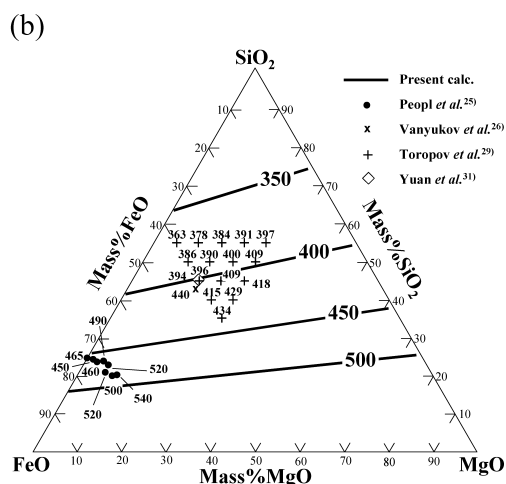
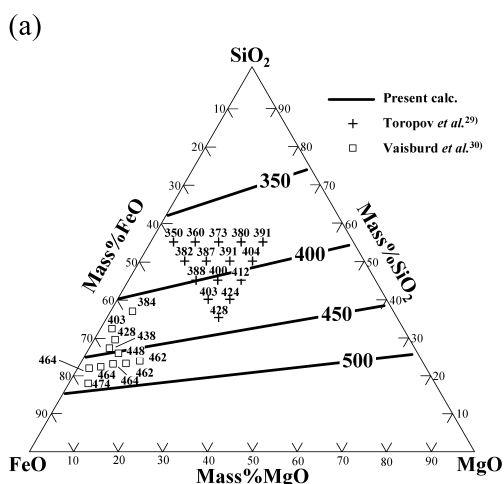


Fig. 8. Surface tension (mN/m) of molten SiO_2 -FeO-MgO system at (a) 1623, (b) 1673 and (c) 1823 K.

molten SiO_2 system. Therefore, $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$ was selected as a parameter for the calculations in the present work as well as in the previous work.⁷⁾

Table 4 shows the equations corresponding to the temperature dependence of the surface tension values for pure oxides obtained in this evaluation with $R_{\text{Si}}^{4+}/R_{\text{SiO}_4}^{4-}=0.5$. The surface tension values calculated in the binary and ternary silicates melts are shown in **Figs. 1** through **9**. It is found that the experimental surface tensions^{1,10-31)} in the ternary

systems targeted in this study decrease mainly with increasing the content of SiO_2 while the effects of the exchange of other two components on the surface tension depend on the systems. For example, SiO_2 -CaO-FeO system in **Fig. 5** shows a decrease in the surface tension with increasing FeO at constant SiO_2 and the replacement of MnO by FeO in SiO_2 -FeO-MnO system gives hardly any variation of the surface tension in **Fig. 9**. Although the present model is the simplest model in which the surface tension of molten

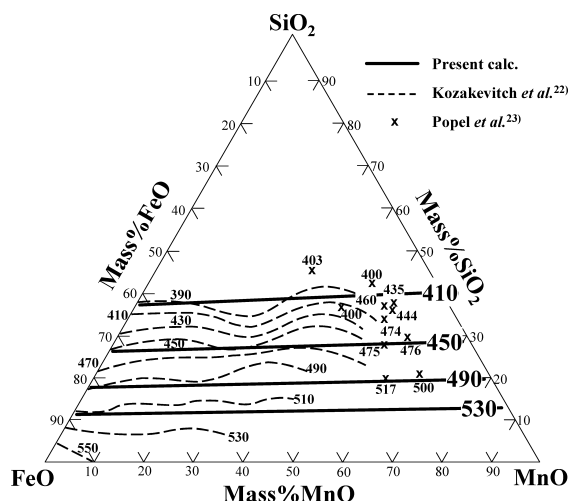


Fig. 9. Surface tension (mN/m) of molten SiO_2 -FeO-MnO system at 1673 K.

slag can be calculated from information relating to the surface tension and molar volume of pure oxides, as well as from the cationic and anionic radii in the system, the results reproduce the composition dependence of the experimental data at various temperatures in binary and ternary silicates.

4. Conclusions

A thermodynamic model for evaluating the surface tension of molten ionic mixtures by considering their ionic radii was extended to silicate melts in various ternary systems. The calculated results obtained using the present model reproduced the composition dependence of the surface tension of molten silicates in ternary systems.

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