Surface Tension Calculation of Molten Slag in $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ Systems Based on a Statistical Modelling Approach

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(Received on November 7, 2018; accepted on December 20, 2018)

A calculation model of surface tension for molten slags in $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ systems has been developed in the present study, based on a statistical modelling approach. A total of 832 experimental data of slags have been critically selected and used for the optimization of model parameters, and the present model has a fairly lower average deviation with 4.60% within the applicable composition and temperature ranges. And also, the composition and temperature dependences of surface tension have been discussed using the model. The results indicate that the surface tensions always decrease with increasing SiO_2 content and increase with increasing MgO content. The surface tension will decrease with increasing temperature.

KEY WORDS: surface tension; statistical modeling approach; molten slag; $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ systems.

1. Introduction

Surface tension is one of important physical properties of molten slags. It not only is applied to the research of melt structure characteristics and thermodynamic properties, but also is closely related to many metallurgy phenomena such as slag foaming, emulsification and kinetics of slag-metal reactions. However, the experimental measurement on the surface tension of multi-component system is usually difficult and time-consuming and laborious at elevated temperature, and there are too few existing surface tension data to meet the needs in industrial applications. Therefore, it is indispensable to determine the surface tension of molten slag by developing more accurate and applicable models.

Many kinds of surface tension calculation models for melts have been reported in literatures over the years. An additive model was proposed in early by Boni and Derge¹⁾ based on the additivity rule, and the surface tension was obtained by employing the surface tension of each pure oxide. Later, the model was further modified by Mills²⁾ and Nakajima³⁾ by considering the effect of surface-active components on surface tension. Although additive model has been widely used in practical applications owing to its convenience, it tends to cover a narrow composition ranges, at least for $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ system and its subsystems.^{3,4)} Another model was developed by Tanaka *et al.*⁵⁾ based on Butler's equation to calculate surface tension and was found to estimate successfully the surface tension values of ternary systems. However, due to the lack

of thermodynamic data of multi-component system, the applicability is limited for quaternary and multisystems. Based on the neural network of human brain, Nakamoto and Hanao⁶⁾ proposed a neural network model for calculation of surface tension. This method is usually applied to the prediction of certain binary and ternary systems. When the model is used to multi-component systems, there is an unsatisfactory reproducibility between the measured values and calculated results.

The $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ systems are of considerable importance for controlling and optimization of metallurgical process. In a previous study,⁷⁾ a density calculation model for the $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ system and its subsystems has been proposed by present authors successfully. In the present work, the model will be applied to the prediction of surface tension of molten slag in $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ systems and the model parameters will be optimized based on the published reliable data. The ultimate aim of this study is to explore the reliable methods for developing applicable calculation model of surface tension in molten slag systems.

2. Method

2.1. Experimental Data

Reliable and accurate experimental data are closely related to the model prediction performance. Totally more than 950 surface tension data were collected through open literature sources. However, it was found that some data were clearly inconsistent with other measurements. For example, the measurements obtained in graphite crucible were often found to have a large experimental uncertainties due to the reaction between graphite and molten slag,

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DOI: <https://doi.org/10.2355/isijinternational.ISIJINT-2018-746>

and thus these data were removed. In this way, roughly 12 percent of the collected data were removed. In this work, the surface tension data obtained by different methods were thought in good agreement. A total of 832 published experimental data used to calibrate the present model has been summarized in **Table 1**, and the composition ranges of these data have been shown in **Fig. 1**.

2.2. Model Description

It is well known that the surface tension is mainly related to chemical composition and temperature of molten slag. To calculate surface tension according to the composition and temperature, the expanded approximation rules³⁾ was used to estimate the surface tension of $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ systems based on the regular solution approximation rules

Table 1. Summary of selected experimental data of surface tension in $\text{SiO}_2\text{--Al}_2\text{O}_3\text{--CaO--MgO}$ systems from various references.

System	Reference	Temperature (K)	Number	Method*
SiO_2	Parikh. (1958) ⁸⁾	1 361 to 1 536	8	Fibre elongation
	Kingery. (1959) ⁹⁾	2 073	3	PD
Al_2O_3	Kingery (1959) ⁹⁾	2 327	1	PD
	Wartenberg, <i>et al.</i> (1936) ¹⁰⁾	2 327	1	DW
	Bartlett and Hall (1965) ¹¹⁾	2 327	1	SD
	Maurakh, <i>et al.</i> (1967) ¹²⁾	2 327	1	PD
	McNally, <i>et al.</i> (1968) ¹³⁾	2 327	1	DW
	Zubarev, <i>et al.</i> (1969) ¹⁴⁾	2 800	1	SD
	Rasmussen and Nelson (1971) ¹⁵⁾	2 327	3	PD
	Elyutin, <i>et al.</i> (1971) ¹⁶⁾	2 616	1	MBP
	Elyutin, <i>et al.</i> (1974) ¹⁷⁾	2 523 to 2 473	2	MBP
	Anisimov, <i>et al.</i> (1977) ¹⁸⁾	2 327	1	MBP
	Lihmann and Haggerty (1985) ¹⁹⁾	2 327	3	PD
	Ikemiya, <i>et al.</i> (1993) ²⁰⁾	2 340 to 2 407	16	MBP
	Glorieux, <i>et al.</i> (2002) ²¹⁾	2 327	1	Aerodynamic levitation
$\text{SiO}_2\text{--Al}_2\text{O}_3$	Rasmussen and Nelson (1971) ¹⁵⁾	2 070 to 2 500	17	PD
$\text{SiO}_2\text{--CaO}$	King. (1951) ²²⁾	1 843	3	CD
	Cooper and Kitchener (1959) ²³⁾	1 873	5	MBP
	Swisher and McCabe (1964) ²⁴⁾	1 873	3	MBP
	Popel, <i>et al.</i> (1969) ²⁵⁾	1 823	4	MBP
	Ono, <i>et al.</i> (1969) ²⁶⁾	1 748 to 1 873	38	MBP
	Sokolov, <i>et al.</i> (1969) ²⁷⁾	2 000	1	MBP
	Ejima and Shimoji (1970) ²⁸⁾	1 873	3	MBP
	Sharma and Philbrook (1970) ²⁹⁾	1 873	5	MBP
	Mikhailikov, <i>et al.</i> (1970) ³⁰⁾	1 873	1	SD
	Gunji and Dan (1974) ³¹⁾	1 823 to 1 873	20	MBP
	Staronka and Piekarska (1978) ³²⁾	1 773 to 1 873	2	/
	Mukai and Ishikawa (1981) ³³⁾	1 773 to 1 873	30	PD
$\text{SiO}_2\text{--MgO}$	King. (1951) ²²⁾	1 843	3	CD
$\text{Al}_2\text{O}_3\text{--CaO}$	Eitel. (1954) ³⁴⁾	1 753 to 1 853	6	/
	Popel and Esin (1957) ³⁵⁾	1 773	1	MBP
	Ershov and Popova (1964) ³⁶⁾	1 773 to 1 973	3	MBP
	Yakobashvili, <i>et al.</i> (1965) ³⁷⁾	1 753	1	MBP
	Ogino, <i>et al.</i> (1966) ³⁸⁾	1 773	3	MBP
	Krinochkin, <i>et al.</i> (1971) ³⁹⁾	1 853 to 1 988	15	SD
	Sikora and Zielinski (1974) ⁴⁰⁾	1 723 to 1 923	5	MBP
	Elyutin, <i>et al.</i> (1974) ¹⁷⁾	2 473	8	MBP
	Zielinski and Sikora (1977) ⁴¹⁾	1 723 to 1 923	5	MBP

	Mukai and Ishikawa (1981) ³³⁾	1 723 to 1 873	4	PD
	Muu, <i>et al.</i> (1985) ⁴²⁾	1 873	2	/
	Oliveira, <i>et al.</i> (1999) ⁴³⁾	1 843 to 1 883	18	MBP
	Dou, <i>et al.</i> (2009) ⁴⁴⁾	1 723 to 1 773	9	CD
	Wegener, <i>et al.</i> (2013) ⁴⁵⁾	1 743 to 1 894	4	PD
Al ₂ O ₃ –MgO	Elyutin, <i>et al.</i> (1974) ¹⁷⁾	2 523	7	MBP
SiO ₂ –Al ₂ O ₃ –CaO	Boni and Derge (1956) ¹⁾	1 603 to 1 835	23	MBP
	Barrett and Thomas (1959) ⁴⁶⁾	1 631 to 1 913	40	MBP
	Elliott, <i>et al.</i> (1963) ⁴⁷⁾	1 623 to 1 773	4	/
	Ershov and Popova (1964) ³⁶⁾	1 873 to 1 973	9	MBP
	Ogino, <i>et al.</i> (1966) ³⁸⁾	1 703 to 1 823	13	MBP
	Evseev and Filippov (1967) ⁴⁸⁾	1 873 to 1 943	6	MBP
	Sokolov, <i>et al.</i> (1969) ²⁷⁾	2 000	5	MBP
	Gunji and Dan (1974) ³¹⁾	1 823 to 1 873	72	MBP
	Mukai and Ishikawa (1981) ³³⁾	1 723 to 1 873	78	PD
	Koshida, <i>et al.</i> (1981) ⁴⁹⁾	1 623 to 1 773	7	SD
	Vaisburd and Brandon (1999) ⁵⁰⁾	1 623 to 1 773	4	MBP
	Dobrovská, <i>et al.</i> (2011) ⁵¹⁾	1 630 to 1 798	55	SD
	Sukenaga, <i>et al.</i> (2011) ⁵²⁾	1 723 to 1 823	5	CD
SiO ₂ –CaO–MgO	Mikhailikov, <i>et al.</i> (1970) ³⁰⁾	1 873	2	SD
	Staronka and Piekarska (1978) ³²⁾	1 673 to 1 973	12	/
SiO ₂ –Al ₂ O ₃ –MgO	Boni and Derge (1956) ¹⁾	1 687 to 1 809	10	MBP
Al ₂ O ₃ –CaO–MgO	Askari and Cameron (1991) ⁵³⁾	1 873	1	MBP
	Xu, <i>et al.</i> (2012) ⁵⁴⁾	1 823	13	CD
SiO ₂ –Al ₂ O ₃ –CaO–MgO	Benesch, <i>et al.</i> (1976) ⁵⁵⁾	1 673 to 1 773	36	MBP
	Koshida, <i>et al.</i> (1981) ⁴⁹⁾	1 623 to 1 773	63	SD
	Askari and Cameron (1991) ⁵³⁾	1 873	6	MBP
	Peng, <i>et al.</i> (2014) ⁵⁶⁾	1 691 to 1 773	21	CD
	Sukenaga, <i>et al.</i> (2015) ⁵⁷⁾	1 723 to 1 823	20	CD
	Yan, <i>et al.</i> (2018) ⁵⁸⁾	1 723 to 1 823	45	MBP
	Xu, <i>et al.</i> (2012) ⁵⁴⁾	1 773 to 1 873	12	CD

SD: sessile drop; MBP: maximum bubble pressure; PD: pendant drop; DW: Drop weight; CD: Cylinder method

of excess molar quantities for binary system. The surface tension of molten slag was calculated as follows:

$$\sigma = \sum X_i \sigma_i(T) + \sigma^{EX} \quad \text{..... (1)}$$

where σ is the surface tension of molten slag, X_i is the mole fraction of component i , $\sigma_i(T)$ is the temperature-dependent partial molar surface tension of component i , and σ^{EX} is the temperature-independent excess surface tension of molten slag, by simply considering the binary interactions between each component.

Accordingly, the surface tension of pure oxides $\sigma_i(T)$ and excess surface tension σ^{EX} are expressed as:

$$\begin{aligned} \sigma_i(T) &= \sigma_{i,R} + \frac{\partial \sigma_i}{\partial T}(T - T_R) \\ &= \sigma_{SiO_2} + \frac{\partial \sigma_{SiO_2}}{\partial T}(T - T_R) + \sigma_{Al_2O_3} + \frac{\partial \sigma_{Al_2O_3}}{\partial T}(T - T_R) + \dots (2) \\ \sigma_{CaO} &+ \frac{\partial \sigma_{CaO}}{\partial T}(T - T_R) + \sigma_{MgO} + \frac{\partial \sigma_{MgO}}{\partial T}(T - T_R) \end{aligned}$$

$$\begin{aligned} \sigma^{EX} &= \sum_{i < j} a_{ij} X_i X_j \\ &= a_{12} X_{SiO_2} X_{Al_2O_3} + a_{13} X_{SiO_2} X_{CaO} + a_{14} X_{SiO_2} X_{MgO} + \dots (3) \\ &+ a_{23} X_{Al_2O_3} X_{CaO} + a_{24} X_{Al_2O_3} X_{MgO} + a_{34} X_{CaO} X_{MgO} \end{aligned}$$

where $\sigma_{i,R}$ is partial molar surface tension of component i at reference temperature T_R , and $\partial \sigma_i / \partial T$ is the temperature coefficient of component i , a_{ij} represents the interaction factor between components i and j .

The values of fitting parameters were obtained by minimization of the function.

$$\sum_{i=1}^N \frac{(\sigma_{mea} - \sigma_{cal})^2}{\sigma_{mea}} \quad \text{..... (4)}$$

where σ_{mea} and σ_{cal} are measured and calculated surface tension, respectively.

All of the 832 surface tension measurements were used equally to derive the optimized values. The optimization was performed using non-linear least-squares fitting based

Fig. 1. Component contents of molten slags in database.

on the well-known Levenberg–Marquardt algorithm.

3. Results and Discussions

3.1. Model Performance

The obtained optimized parameter values were listed in **Tables 2** and **3** respectively. The performance of the model on surface tension was evaluated by the difference between the measured and calculated surface tension value, which is expressed by:

$$\delta_i = \frac{|\sigma_{i,\text{cal.}} - \sigma_{i,\text{mea.}}|}{\sigma_{i,\text{mea.}}} \quad \dots\dots\dots (5)$$

The accuracy of the model was represented by the average relative error (Δ), for N measurements, which was calculated by

$$\Delta = \frac{1}{N} \sum_{i=1}^N \delta_i \times 100\% \quad \dots\dots\dots (6)$$

The root mean square error (RMSE), which represents the average absolute error, was also calculated

$$\text{RMSE} = \sqrt{\frac{\sum_{i=1}^N (\sigma_{i,\text{cal.}} - \sigma_{i,\text{mea.}})^2}{N}} \quad \dots\dots\dots (7)$$

As shown in **Fig. 2**, it is found that there is an excellent agreement between the calculated and measured surface tension, with overall accuracy of 4.60% and the average

Table 2. Optimized parameter values of $\sigma_{i,R}$ and $\partial\sigma_i/\partial T$.

Component	$\sigma_{i,R}$ (1 773 K) ($\text{mN}\cdot\text{m}^{-1}$)	$\partial\sigma_i/\partial T$ ($\text{mN}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)
SiO_2	293.93	-4.55×10^{-3}
Al_2O_3	759.12	-277.27×10^{-3}
CaO	672.09	-264.93×10^{-3}
MgO	1 177.56	-789.32×10^{-3}

Table 3. Optimized parameter values of interaction coefficient for excess surface tension.

Parameter value a_{ij} ($\text{mN}\cdot\text{m}^{-1}$)	System
a_{12}	68.24 $\text{SiO}_2\text{-Al}_2\text{O}_3$
a_{13}	-97.27 $\text{SiO}_2\text{-CaO}$
a_{14}	-1 229.35 $\text{SiO}_2\text{-MgO}$
a_{23}	-350.59 $\text{Al}_2\text{O}_3\text{-CaO}$
a_{24}	-60.68 $\text{Al}_2\text{O}_3\text{-MgO}$
a_{34}	-857.40 CaO-MgO

absolute error of 25.78 mN/m for 832 experimental data in $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaO-MgO}$ systems.

The present model is developed based on statistical modelling approach, and the calculation results are accordingly in higher accuracies. For most of the subsystems, the present model also results in very high accuracy, as shown in detail in **Fig. 3**. By comparison, the three previously reported simi-

lar models¹⁻³⁾ are also used to predict the surface tension, but tend to have an unsatisfactory results in the viewpoint of achieving highly accurate estimation, as shown in Fig. 4. However, it must be noted that the present model can only be used within the selected composition ranges. Therefore, more reliable experimental data in wide composition and temperature range are fundamental for developing the statistical model to predict the surface tension of molten slags.

3.2. Composition and Temperature Dependences of Surface Tension

3.2.1. Unary System

Surface tension of pure component SiO_2 was measured by Parikh⁸⁾ using fiber elongation method in vacuum, dry N_2 and Air respectively and the effect of atmosphere on

surface tension was investigated at different temperatures. Another measurement was performed by Kingery⁹⁾ using PD method at 2 073 K and three points were obtained. As shown in Fig. 5(a), the measured values are compared with the calculated results and the surface tension slightly decreases with increasing temperature.

Many studies for surface tension of Al_2O_3 have been carried out at different temperatures ranging from 2 342 K to 2 500 K. As shown in Fig. 5(b), the measured values are compared with the calculating results and show a good agreement with a mean deviation of 3.8%. It can be seen that the surface tension of Al_2O_3 decreases obviously with increasing temperature and there is a discrepancy at the melting point for experimental results from different research groups. Because of high melting temperature for

Fig. 2. Comparison between measured and calculated surface tension data by using the present model for all SiO_2 – Al_2O_3 – CaO – MgO systems.

Fig. 3. Performances of present model in surface tension calculation for each system (S, A, C and M represent SiO_2 , Al_2O_3 , CaO , MgO , respectively).

Fig. 4. Prediction of surface tension of molten slag using previously similar models.

 Fig. 5. Temperature dependence of surface tensions of SiO_2 and Al_2O_3 .

Fig. 6. Composition dependences of surface tension for binary systems.

pure CaO and MgO, there is no literature of pure liquid CaO and MgO published has not been found until now.

3.2.2. Binary Systems

The composition and temperature dependences of surface tension for the binary systems are shown in Fig. 6. It is found that the calculated values agree well with the measurements. The surface tension always decreases with the increment of SiO₂ content in SiO₂-M systems, as shown in Figs. 6(a), 6(b) and 6(c). In cases of Al₂O₃-MgO system, currently only one group of measured data at 2473 K by Elyutin *et al.*¹⁷⁾ was found, as presented in Fig. 6(d). The results indicate that the surface tension slightly decreases as the Al₂O₃ content grows. For the Al₂O₃-CaO system, a great quantity of studies have been published, as presented in Fig. 6(e). The surface tension decreases at first and then increases with the increase of Al₂O₃ at different temperatures, showing the amphoteric nature of Al₂O₃. Besides, it should be noted that the compositional dependence of the surface tension shows a similar trend with the experimental results by Elyutin *et al.*¹⁷⁾ at 2473 K.

Generally, the temperature tends to depend weakly on surface tension of melts. On the one hand, with the increases of temperature, the distance between particles increases and the interaction force decreases, leading to a decrease in the surface tension of the melt. On the other hand, the increase of temperature will also result in a decrease of the polymerization degree of silica anions, so that the surface tension will increase. The two effects are superimposed, resulting in

a decrease of surface tension.

3.2.3. Ternary Systems

The iso-surface tension curves are plotted at 1873 K using the present model and compared with the measured values for SiO₂-Al₂O₃-CaO, SiO₂-CaO-MgO, SiO₂-Al₂O₃-MgO and Al₂O₃-CaO-MgO systems, respectively. As shown in Fig. 7, the results show a similar trend with binary systems. For instance, the surface tension always decreases with increasing SiO₂ content for silicate systems. As a surface-active component of molten slag, the increase of SiO₂ content will lead to an increase of the polymerization degree of silica anions, thus reducing the surface tension of molten slag. For the Al₂O₃-CaO-MgO system, the increase of CaO will result in a decrease of surface tension and show a large influence on surface tension in high CaO region.

3.2.4. Quaternary Systems

As an important base system, SiO₂-Al₂O₃-CaO-MgO quaternary system is commonly applied to the studies of blast furnace slag, ladle slag, mold flux, and so on. So the surface tension of SiO₂-Al₂O₃-CaO-MgO quaternary system was extensively studied by Sukenaga *et al.*⁵⁷⁾ and Yan *et al.*⁵⁸⁾ at 1723 K to 1823 K. The measured results are compared with the calculated values, as presented in Fig. 8. It is seen that the present model can accurately predict the surface tension and the surface tension decreases with increasing SiO₂ content and temperature, which is consistent with the trends in binary and ternary systems.

Fig. 7. Composition dependences of surface tension for several ternary systems.

Fig. 8. Composition and temperature dependences of surface tension for SiO₂-Al₂O₃-CaO-MgO system.

Fig. 9. Iso-surface tension curves for quaternary SiO₂-Al₂O₃-CaO-MgO system (mN/m): (a) MgO=10%; (b) MgO=20%; (c) MgO=30%; and (d) MgO=40%.

Using the present model, the iso-surface tension curves for quaternary systems of SiO₂-Al₂O₃-CaO-MgO with constant MgO content (10%, 20%, 30% and 40%, respectively) at 1873 K are calculated, as shown in Fig. 9. It is found that the surface tension increase when the amount of MgO is increased from 10% to 40%, which is consistent with the trends in binary and ternary systems.

4. Conclusions

In the present work, a surface tension calculation model in SiO₂-Al₂O₃-CaO-MgO systems and its subsystems was developed based on a statistical modelling approach. A total number of 832 experimental data have been used for

the optimization of model parameters. The main summary remarks can be obtained as follows:

(1) Present model covers wide composition ranges in SiO₂-Al₂O₃-CaO-MgO systems, and calculates the surface tension of molten slag systems with remarkable accuracy of relative average error (Δ) of 4.60% and overall absolute error (RMSE) of 27.57 mN/m.

(2) For all of binary, ternary and quaternary systems, the surface tensions of SiO₂-Al₂O₃-CaO-MgO molten slags always decrease with increasing SiO₂ content and increase with increasing MgO content.

(3) Temperature tends to have a double effect on surface tension of molten slag, and usually, the increase of temperature will result in a reduce of surface tension.

Acknowledgements

This work was supported by the National Natural Science Foundation of China [Grant numbers 51574066, 51774072, 51574065 and 51774073] and the National Key R&D Program of China [Grant numbers 2017YFB0304201, 2017YFB0304203 and 2016YFB0300602].

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