

MODELING OF THE SURFACE/INTERFACIAL TENSION FOR THE LADLE REFINING OF SILICON METAL PROCESS

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ABSTRACT

Interfacial phenomena play an important role in the oxidative ladle refining (OLR) of metallurgical grade silicon (MG-Si). The removals of Al and Ca impurities are ultimately determined by the metal/slag exchange reactions. The refining process involves the mass transfer across the metal/slag interface. Phase separation occurring in the process is also dependent upon the interfacial properties of the metal and slag phases. In order to model the OLR process by the computational fluid dynamics (CFD) techniques, the surface/interfacial tensions of the liquid Si-Al-Ca alloy and SiO_2 -CaO- Al_2O_3 slag phases have to be determined mathematically.

In the present investigation, surface tensions of the liquid metal and slag phases were modelled separately using the constricted Gibbs energy minimization algorithm. The basic assumption is that the monolayer thick surface layer is in equilibrium with the bulk phase. Thermochemical properties of the molten slag and the surface properties of pure components are the only required input for the calculation. Calculated surface tensions of the liquid Si-Al-Ca alloys and SiO_2 -CaO- Al_2O_3 slags are able to reproduce the experimental results within the error limits. Interfacial tensions between metal and slag phases were then modelled applying the modified Girifalco-Good equation. The proportional factor is expressed as function of SiO_2 concentration. The calculated equilibrium contact angles suggest that almost no mutual wetting occurs for the slags at the gas/metal/slag interface. The calculated equilibrium spread tendency is also in good agreement with the laboratorial and industrial observations. This provides the conceptual basis for the CFD simulations.

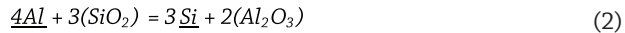
Key Words: *Surface tension, interfacial tension, modeling, SiO_2 -CaO- Al_2O_3 slag, metallurgical grade Si, oxidative ladle refining process.*

INTRODUCTION

In the oxidative ladle refining of metallurgical-grade silicon (MG-Si), oxygen is introduced either as gas or as SiO_2 oxidizing agent to the metal surface in order to remove the most common impurities, Al and Ca, in the silicon metal. No matter how the oxygen is introduced, reaction between oxygen and the most abundant element, Si, occurs immediately at the metal surface:



The dissolved Al and Ca impurities present at the metal/slag interface may react with SiO_2 as follows:



where underlined elements denote dissolved in the metal and brackets the slag phase. The impurities in MG-Si are ultimately depended on the metal/slag equilibria, Equation 2 and Equation 3. During the metal/slag reactions, the kinetics of the chemical interactions will exert a strong influence on the magnitude of the interfacial tension. Under conditions of intense metal/slag mass transfer, the interfacial tension will often decrease to very low values and remain at the low level until the system approaches its chemical equilibrium [29]. When the system approaches equilibrium, the interfacial tension will gradually increase and ultimately attain an essentially constant value. Extremely high rates of mass transport may lead to unstable interfacial tension gradients and the convective flow. Consequently, the emulsification can be occurred at the metal/slag interface. Alternatively, the presence of a highly surface-active component in either of the liquids can create a diffusion barrier at the interface and may impede mass transfer of other impurities.

Thermodynamic descriptions of the Si-Al-Ca alloy [1] and SiO_2 -CaO- Al_2O_3 oxide [8] systems have been reported in the literature. Using these thermodynamic descriptions, the measured element distribution equilibria [45] can be reproduced in an excellent manner, as shown in Figure 1.

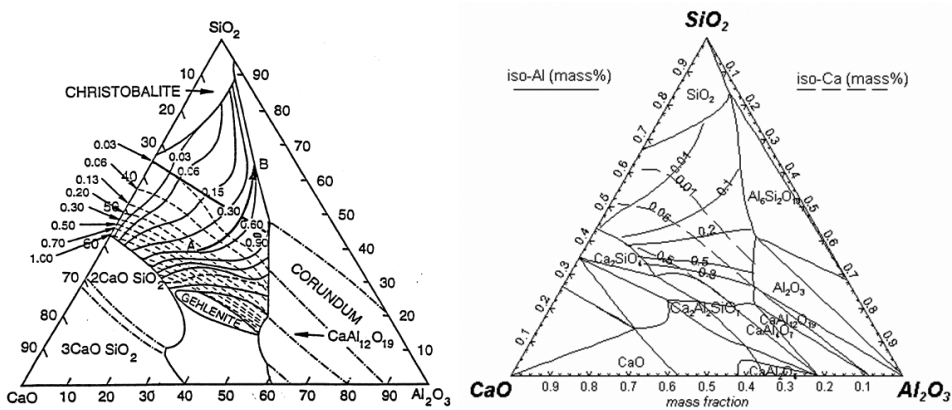


Figure 1: Comparison of the measured (left) and calculated (right) iso-concentration curves for Al and Ca in 75SiFe alloys in equilibrium with SiO_2 -CaO- Al_2O_3 slags at 1550°C

In recent years, it has become more popular to apply thermodynamic properties for the evaluation of the surface tensions of liquid alloys [40], molten slags [3, 24, 25, 26], salts [41, 40], mattes [46] and fluxes [33]. The Bulter's equation [2] and its modified formula [42] were used to couple the surface tension with the thermochemical properties. Tanaka and Hara [40] further used the Bulter's equation together with the Girifalco-Good equation [10] to calculate the interfacial tension between liquid steel and molten slag.

In this paper, a novel approach based on Gibbs energy minimization technique has been applied to determine the surface tensions of liquid Si-Al-Ca alloys and SiO₂-CaO-Al₂O₃ slags. The metal/slag interfacial tension has been estimated by the Girifalco-Good semiempirical equation. Based on the evaluated surface and interfacial tensions, the wettability (contact angle) of metal/slag and their spreading tendency have then been estimated. The present approach provides an efficient way for the simultaneous calculations of the metal/slag equilibria and the surface/interfacial properties.

THE SURFACE TENSION AND INTERFACIAL MODEL

The Surface Tension Model

Based on Guggenheim's statistic thermodynamic treatment [12], equilibria between the bulk phase and a monolayer thickness surface phase is first assumed. Following suggestions by Koukari and Pajarre [17], a fictitious species, *Area*, is introduced to the imaginary *surface* phase. The liquid metal, for instance, consists of a bulk and a surface phase. The components in bulk phase are Si, Al and Ca. The constituents of surface phase are SiArea_{x1}, CaArea_{x2} and AlArea_{x3}, where x1, x2 and x3 are, respectively, the stoichiometries of surface components determined by the molar surface area of pure components.

Figure 2 schematically shows the phase equilibria between the bulk and surface phases as well as the constituents of two phases in the Si-Al binary subsystem.

The chemical potentials of components in surface phase are determined by the relation $\mu_i^S = \mu_i^B + Area_i \sigma_i$, where $Area_i$ and σ_i are molar surface area (m³/mol) and surface tension of pure element *i*. The total Gibbs energy of the system is then expressed as:

$$G = \sum_i \mu_i^B n_i^B + \sum_i \mu_i^S n_i^S + \sigma \sum_i Area_i^S n_i^S \quad (4)$$

By introducing the material balance and an additional molar surface property constrains,

$$\sum_\alpha \sum_i a_{ik}^\alpha n_i^\alpha - b_i^\alpha = 0 \quad \text{and} \quad \sum_\alpha \sum_i Area_i^\alpha n_i^\alpha - Area = 0 \quad (5)$$

then minimization of the Gibbs energy of system results directly in the surface tension of the multicomponent melt as function of temperature, pressure and compositions [17]. In Equation 5, b_i is the total molar amount of component *i* and a_{ik} the stoichiometric coefficient between species *i* and component *k*. The chemical potential of the fictitious component, μ_{Area} , is equivalent to the surface tension of liquid melt, σ , in the unit of mN/m. As indicated by Pajarre *et al.* [30], the Bulter's equation [2] is actually satisfied when the constrained free energy minimization approach is applied. In this way, surface tension of a multicomponent melt can be directly determined using the commercial thermodynamic software, ChemSheet®, for example.

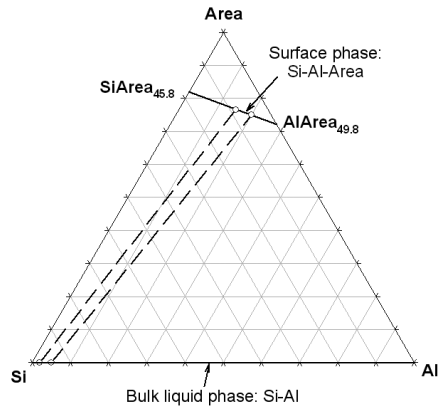


Figure 2: Schematic illustration of the bulk and surface phases in the Si-Al binary subsystem

The above surface tension model can readily be extended to the molten slag by introducing the fictitious surface oxide phase ($\text{SiO}_2\text{Area}_{y_1}$, CaOArea_{y_2} , $\text{Al}_2\text{O}_3\text{Area}_{y_3}$) into the oxide system. Again, the stoichiometries of surface components, y_1 , y_2 and y_3 , are determined by the molar surface area of pure SiO_2 , CaO and Al_2O_3 . Chemical potentials of components in the oxide surface phase can be determined similarly as in the metallic surface phase.

The basic data used to model the surface tension of liquid Si-Al-Ca alloy and SiO_2 -CaO- Al_2O_3 slag are listed in Table 1.

Table 1: Basic data for surface/interfacial tension calculations used in this work [24, 25, 26, 44]

Component	Phase	Molar area (m^2/mol)	Surface tension (mN/m)	Stoichiometric coefficient for area in surface phase
Si	Metal	45845	$994-0.145T$	45.8
Al	Metal	49829	$1042-0.18T$	49.8
Ca	Metal	93276	$474.5-0.1T$	93.3
SiO_2	Slag	77480	$243.2-0.031T$	77.5
Al_2O_3	Slag	78945	$1024-0.177T$	78.9
CaO	Slag	64088	$791-0.0935T$	64.1

It was first proposed by Speiser *et al.* [36] that the excess Gibbs energy in surface phase can be estimated from the excess Gibbs energy in the bulk phase: $G^{E,S} = \alpha G^{E,B}$, here α is the empirical proportional coefficient. The values of α vary significantly for different type of melts [39, 40, 41]. This implies that such kind of approximation is not physically sound. In the present work, the excess Gibbs energies in the surface phases have been reevaluated in order to fit the measured surface tension values. The evaluation is in principle similar to the optimization of thermochemical properties in the bulk phase.

The Slag/Metal Interfacial Tension Model

The relation between surface tensions of two liquids and their interfacial tension can be correlated by the work of adhesion:

$$E_A = \sigma_M + \sigma_S - \sigma_{MS} \quad (6)$$

The work of adhesion between liquid metal and molten slag is a measure of the bond

strength between the two media. Girifalco and Good [10] assumed that the work of adhesion is proportional to the geometric average of the surface tensions of two media:

$$E_A = 2\phi\sqrt{\sigma_M\sigma_S} = \sigma_M + \sigma_S - \sigma_{MS} \quad (7)$$

The proportionality factor, ϕ , is a semiempirical correction factor and general varies in the range between 0.32 and 1.15 [10].

RESULTS AND DISCUSSION

Surface Tension of the Liquid Si-Al-Ca Alloys

Surface tension of liquid silicon has been extensively studied by a number of researchers [32, 15, 34, 9, 21; 47] in the last decades. Comprehensive reviews on the surface properties of pure elements and Si-based binary alloy were given by Mills and Su [19] and Keene [14]. The surface tension assessed by Mills and Su for oxygen-saturated liquid silicon [19] is adopted in this work.

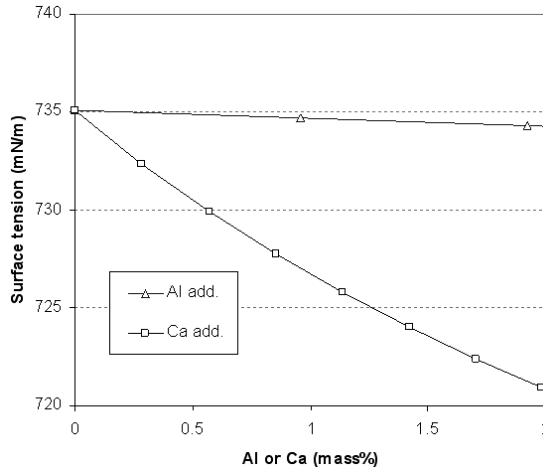


Figure 3: Effect of Al and Ca on the surface tension of liquid oxygen-saturated silicon at 1550°C

The contents of Al and Ca impurities appear in MG-Si are generally lower than 1-2 mass%. The effect of impurity on the surface tension of liquid silicon has extensively been studied by one of the present author [44]. The calculated surface tension in Si-Al and Si-Ca melts are in good agreement with the measured values [11, 14]. The effects of Al and Ca impurities on the surface tension of liquid silicon are shown in Figure 3.

Surface Tensions of the Molten $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$ Slags

Surface tensions of the molten $\text{SiO}_2\text{-CaO-Al}_2\text{O}_3$ slags were measured by Cooper and Kitchener [4], Gunji and Dan [13], Mukai and Ishikawa [20], Krinochkin *et al.* [18], Dragomir and Rusu [5], Sharma and Philbrook [35] Ono *et al.* [28], Elyutin *et al.* [7], King [16] and Muu *et al.* [22] In general, the experimental data are very scatter, especially in the $\text{CaO-Al}_2\text{O}_3$ system. A comparison between the calculated and measured surface tensions in $\text{SiO}_2\text{-CaO}$ binary silicates is shown in Figure 4.

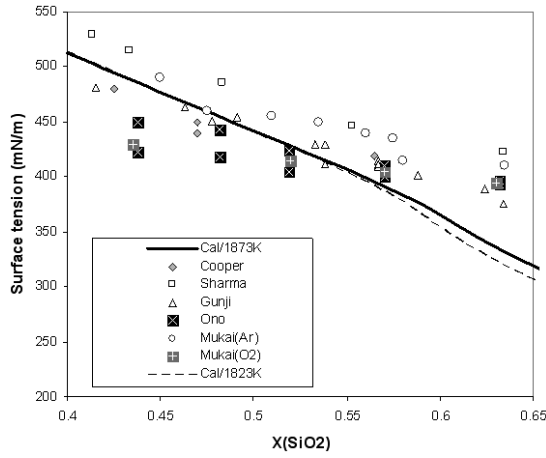


Figure 4: Surface tension of the SiO₂-CaO slag at 1550°C

Attempts for the modeling of surface tensions in the molten SiO₂-CaO-Al₂O₃ slags were made by several authors. Nakajima [23] used the simple regular solution approximation to estimate the surface tension of multicomponent slags. Choi and Lee [3] and Tanaka *et al.* [38, 39] used the Bulter’s equation [2] to calculate surface tension of molten slags. More recently, Nakamoto and coworkers [24] used the artificial neural network to simulate the surface tension of molten silicate slags. All the authors claimed that their models were able to represent the surface tension of molten SiO₂-CaO-Al₂O₃ slags.

The present calculated surface tensions of ternary SiO₂-CaO-Al₂O₃ slags, compared with the measured values, are shown in Figure 5. Good agreement exists between the present calculations and experimental values reported by Gunji and Dan [13], Mukai and Ishikawa [20]. Agreement with the calculation results by Nakajima [23] is only reasonable, probably due to the shortcoming nature of the simplified model.

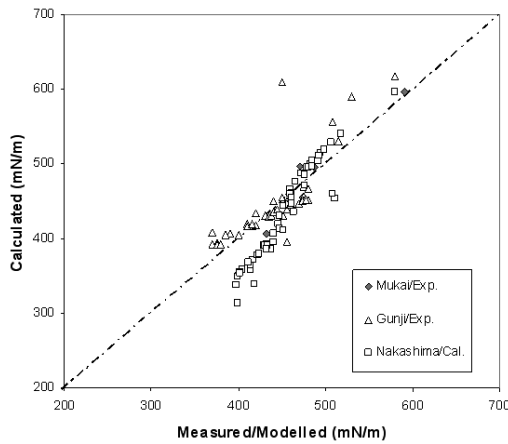


Figure 5: Surface tension of the SiO₂-CaO-Al₂O₃ slag at 1550°C

Interfacial Tension between Liquid Si-Al-Ca Metal and SiO₂-CaO-Al₂O₃ Slag Phases

Little experimental information exists about the interfacial tension between Si-based metal and molten SiO₂-CaO-Al₂O₃ slag. Dumay and Cramb [6] measured the interfacial tensions of Si-Fe alloys in contact with two SiO₂-CaO-Al₂O₃ covering slags using the sessile drop technique. They found that the measured interfacial tension values were close to the surface tension of liquid Si-Fe melts. The similar measurements by Popel *et al.* [31] were significant lower than those reported by Dumay and Cramb [6]. Dumay and Cramb attributed this to the different slags used in the measurements.

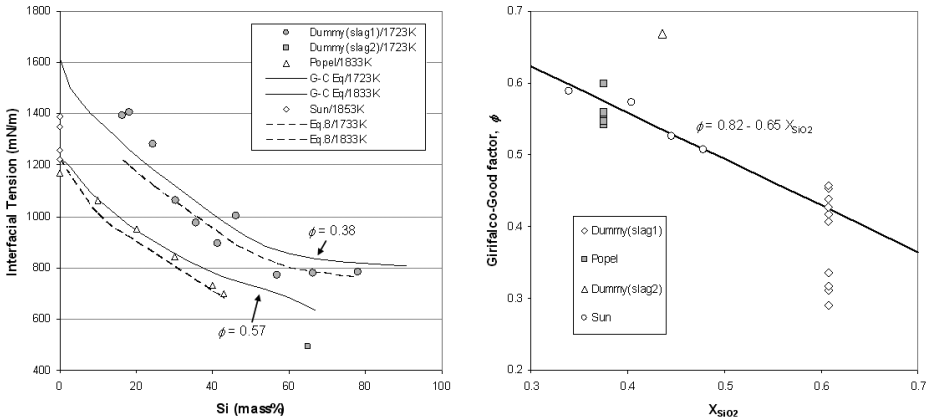


Figure 6: Measured interfacial tension and Girifalco-Good factor between liquid Si-Fe alloys and molten SiO₂-CaO-Al₂O₃ slags

The measured interfacial tensions have first been used to determine the proportional factor, ϕ , and shown in Figure 6. The experimental results by Dumay and Cramb [6] and by Popel *et al.* [31] can be reasonably reproduced by the Girifalco-Good equation using different ϕ values. As mentioned early, Tanaka and Hara [39] used the Girifalco-Good equation to calculate the interfacial tension between liquid steel and molten slag. They found that ϕ is related to FeO concentration: $\phi = 0.5 + 0.3X_{\text{FeO}}$. For the FeO-free slags, they used the constant, 0.5, to estimate the interfacial tensions.

As illustrated in Figure 6, ϕ is also dependent on the SiO₂ content in molten slag. The interfacial tensions between liquid Fe and SiO₂-CaO-Al₂O₃ slags reported by Sun *et al.* [37] have also been used to demonstrate the relation between ϕ and X_{SiO_2} . For the first approximation, following relation can be obtained from the experimental data:

$$\phi = 0.817 - 0.645X_{\text{SiO}_2} \quad (8)$$

Using this relation the measured interfacial tensions can be even better reproduced by the Girifalco-Good equation, as represented by the dashed lines in Figure 6.

As the surface and interfacial tensions have been modeled, the equilibrium wettability between liquid Si-Al-Ca alloys and SiO₂-CaO-Al₂O₃ slags can now be evaluated using the well-known Young equation:

$$\sigma_S - \sigma_{MS} = \cos(\theta)\sigma_M \quad (9)$$

where the surface tensions of metal and slag are determined using the Gibbs energy minimization technique. Substituting Equation 7 and Equation 8 into Equation 9 and rearrangement gives:

$$\cos(\theta) = (1.63 - 1.29X_{\text{SiO}_2}) \sqrt{\frac{\sigma_s}{\sigma_M}} - 1 \quad (10)$$

The spreading coefficient, a relative measure of the tendency of molten slag to spread across the surface of liquid alloy, is determined using following equation:

$$S = \sigma_M - \sigma_s - \sigma_{SM} = (1.63 - 1.29X_{\text{SiO}_2}) \sqrt{\sigma_M \sigma_s} - 2\sigma_s \quad (11)$$

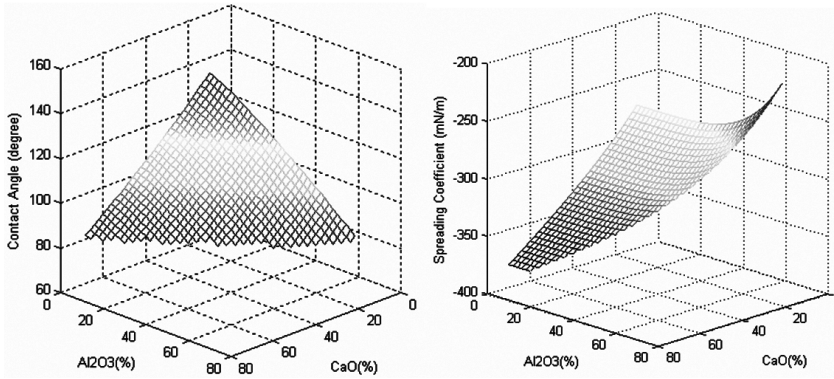


Figure 7: Calculated equilibrium contact angle (left) and spreading coefficient (right) between liquid Si-Al-Ca alloys and SiO_2 -CaO- Al_2O_3 slags at 1550°C

Calculated the equilibrium contact angle and spreading coefficient for the metal/slag system at 1550°C are shown in Figure 7. The calculated equilibrium contact angles suggest that no mutual wetting occurs for the slags at the gas/metal/slag interface. The calculated equilibrium spreading coefficient shows that increasing CaO content has the advantage of enhancing phase separation, while addition of Al_2O_3 has the opposite effect. This is in good agreement with the previous laboratorial and industrial observations [45]: Si metal and alumina silicate slags were difficult to be separated. The calculated wettability and spreading tendency provide the conceptual basis for the CFD simulations [27].

CONCLUSIONS

Surface tensions of the liquid Si-Al-Ca alloys and SiO_2 -CaO- Al_2O_3 slags have been modelled using the constricted Gibbs energy minimization approach. Equilibrium between the bulk phase and a monolayer thickness surface phase is assumed. Under this assumption, a fictitious species, Area, is introduced the imaginary surface phase. The chemical potentials of components in surface phase are determined by adding the surface energy to the bulk species, e.g. $\mu_i^S = \mu_i^B + \text{Area}_i \sigma_i$. Minimization of the Gibbs energy of surface phase containing system results directly in the surface tension of the multicomponent melt as function of temperature, pressure and compositions. The present calculated surface tensions of the metals and slags are able to reproduce the experimental results within the error limits.

Interfacial tensions between metal and slag phases were then modelled applying the modified Girifalco-Good equation. The proportional factor is expressed as function of

SiO₂ concentration. The calculated equilibrium contact angles suggest that almost no mutual wetting occurs for the slags at the gas/metal/slag interface. The calculated equilibrium spread tendency is also in good agreement with the laboratorial and industrial observations. The calculated wettability and spreading tendency provide the conceptual basis for the CFD simulations.

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