RELATIONSHIP BETWEEN THE CALCULATED OXYGEN ACTIVITY AND THE SULFUR PARTITION RATIO FOR CaO-Al2O3-SiO2-MgO SLAG DURING LADLE REFINING

Zoran Slović1, Karlo T. Raić2, Ljubomir Nedeljković2, Tatjana Volkov-Husović2

1The Key-to-Metals, d. o. o., Zrenjaninski put 13P, 11000 Belgrade, Serbia
2University of Belgrade, Faculty of Technology and Metallurgy, Department of Metallurgical Engineering, Karnezijska 4, 11120 Belgrade, Serbia

A slag-metal equilibrium study was carried out to investigate the effect of oxygen activity on the sulfur partition for CaO-Al2O3-SiO2-MgO slag. The sulfide capacity prediction models by Sosinsky-Sommerville based on the optical basicity and the KTH model in terms of the defined interaction coefficient of the component i to j, are used in this work for both a comparison and an estimation of the sulfur partition ratio between ladle-treated slag and liquid steel. From the obtained results, it was shown that the Sosinsky-Sommerville optical basicity approach gives higher values for the sulfur partition ratio (Ls) compared with the KTH model.

Keywords: sulfide capacity, sulfur partition ratio, oxygen activity, optical basicity, KTH model

1 INTRODUCTION

Over the past few decades the sulfur level in steel has been improved enormously. Therefore, the more intensive development industries, such as automotive and pipelines for the transportation of gas and oil, require good control of the sulfur level in steel products. Due to these facts, close control of the sulfur level is essential for the production of good-quality steel. One of the main subjects when investigating the slag/metal interface is the behavior of the oxygen in the liquid steel. The state of oxidation of a bath is of vital importance in controlling the reactions between the slag and the metal in steelmaking. It influences both the metal losses in the slag and the quality of the produced steel. The present paper is focused on predicting the oxygen activities calculated using the sulfur equilibrium between the top slag and the steel.

1.1 Sulfide capacity models

The concept of sulfide capacity was proposed by Finchman and Richardson, and it was defined as:

\[ \text{C}_s = (\%S)(P_{O2}/P_{S2})^{1/2} \] (1)

The sulfide capacity is a property of the slag that is dependent only on the temperature and the slag’s composition. The sulfide capacity can be used to describe the potential ability of an arbitrary homogeneous molten slag to remove sulfur and to compare the desulfurization characteristics of different slags.

The slag desulfurization capacity in the system slag-metal may be expressed as the slag sulfide capacity:

\[ [S] + (O^2-) = (S^{2-}) + [O] \] (2)

Some authors have preferred to define it with reference to the slag metal reaction, in which case the definition becomes:

\[ \text{C}'_s = (\%S)[a_i]/[a_s] \] (3)

Using Turkdogan’s formulation the relation between \( C_s \) and \( \text{C}'_s \) can be written as

\[ \text{C}_s = \text{C}'_s / K_{os} \] (4)

The equilibrium constant \( K_{os} \) for the above equation is:

\[ \lg K_{os} = -935/T + 1.375 \] (5)
1.2 Calculation of the activity of oxygen in the slag and the steel

It is well known that control of the desulfurization process is impossible if the oxygen activity is not known. When the steel is deoxidised with aluminum and silicon, the reactions deciding the oxygen content are the Al/O/Al₂O₃ and the Si/O/SiO₂ equilibrium. Table 1 summarises the chemical reactions used in this work that take part in the deoxidation of the steel melt, in the slag-melt equilibrium during ladle treatment, their mole free-energy changes in the standard state, as well as their reaction constants.

Also, the Ohta and Suito expressions were used to calculate the Al₂O₃ and the SiO₂ activities in the slag, while Wagner’s expressions in equations (6,7) were used to calculate the alumina [a]Al and silicon [a]Si activities in the steel. All the used oxides are in weight percent.

The activity coefficients of the elements in the metal are calculated by using Wagner’s equation, as follows:

\[
\lg f_i = \Sigma (e_i \cdot \% j)
\]  (6)

where:

\( e_i \) – interaction coefficient of j on i

\( f_i \) – Henry’s activity coefficient for the species i in the metal

From Equation 6, the a activity of element i in the steel can be calculated as:

\[
a_i = f_i \cdot [\% i] \quad (i = Si, Al, S)
\]  (7)

The interaction coefficients used in this work are as follows:

\( e_{Si} = 0.0153 + 233/T, \quad e_{Si} = 0.113, \quad e_{Si} = 0.063, \quad e_{Al} = 0.035, \quad e_{Mn} = 0.026, \quad e_{2Al} = 0.011 + 63/T, \quad e_{Si} = 0.091, \quad e_{2Si} = 0.056, \quad e_{Si} = 0.030, \quad e_{Mn} = 0.089 + 34.5/T, \quad e_{Si} = 0.056, \quad e_{Mn} = 0.058, \quad and \ e_{Mn} = 0.002.\)

From the equations of the equilibrium constants given in Table 1, it was possible to derive an expression for the oxygen activity:

\[
a_{O_s} = \sqrt{\frac{a_{SiO_2}}{a_{Al}_2O_3 \cdot e^{-\frac{\Delta G°}{RT}}}}
\]  (9)

The sulfur partition ratio between the slag and the metal may be expressed by combining equations (4) to (9) as follows:

\[
\lg L_s = -935/T + 1.375 + \lg C_s + \lg f_s - \lg [a_o]
\]  (10)

where:

\( C_s \) – sulphide capacity

\( L_s \) – sulfur partition between the slag and the metal,

\( [a_o] / [a_s] \) – oxygen and sulfur activity in the molten metal,

\( \% S \) – sulfur weight percent in the steel

\( \% S \) – sulfur weight percent in the slag

1.3 Optical basicity concept

The optical basicity of the molten slag can be calculated using the following relationships:

\[
\Lambda = \sum \frac{N_i}{X_i \cdot n_{i0}}
\]  (11)

\[
N_i = \sum X_i \cdot n_{i0}
\]  (12)

where:

\( \Lambda \) – optical basicity of the slag

\( \Lambda \) – optical basicity value of the component "i"

\( N_i \) – compositional fraction

\( X_i \) – mole fraction of component "i" in the slag

\( n_{i0} \) – number of oxygen atoms in the component "i"

Sosinsky and Sommerville (S–S) derived the following empirical correlation between the optical basicity, the temperature and the sulfide capacity of the slag at temperatures between 1400 °C and 1700 °C:

\[
\lg C_s = \frac{22690 - 54640\Lambda}{T} + 436\Lambda - 25.2
\]  (13)

The values of the optical basicity for the common steelmaking oxides used in this work have been taken from the literature.

Table 1: Thermodynamic data on chemical reactions taking place in deoxidation, and slag-metal equilibrium

<table>
<thead>
<tr>
<th>Chemical reactions</th>
<th>Mole free energy changes, ( \Delta G^0 ) (J/mol)</th>
<th>Constants, ( K = \exp(-\Delta G°/RT) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2[Al]+3[O] = (Al₂O₃)</td>
<td>( \Delta G^0_{Al} = -1205115 + 386.7T )</td>
<td>( \frac{a_{Al}<em>2O_3}{a</em>{Al}_3O_4} ) (Eq.8)</td>
</tr>
<tr>
<td>[Si]+2[O] = (SiO₂)</td>
<td>( \Delta G^0_{Si} = -581900 + 2218T )</td>
<td>( \frac{a_{SiO_2}}{a_{Si}_2O_3} ) (Eq.9)</td>
</tr>
</tbody>
</table>

\[
\lg a_{Al2O3} = (-0.275(\% CaO) + 0.167(\% MgO)) / (\% SiO₂) + 0.033(Al₂O₃) - 1.560 \]

\[
\lg a_{SiO2} = 0.036(\% MgO) + 0.061(Al₂O₃) + 0.123(\% SiO₂) - (\% SiO₂) / (\% CaO) - 6.456 \]
1.4 The KTH model

The KTH model was developed by the Department of Metallurgy in the Royal Institute of Technology (Sweden).\textsuperscript{17} According to the definition of the sulfide capacity, \( C_s \), can be expressed as:

\[
C_	ext{s} = \exp \left( \frac{-\Delta G^0}{RT} \left( \frac{a_{S^{2-}}}{f_{S^{2-}}} \right) \right) = \exp \left( \frac{-\Delta G^0}{RT} \right) \exp \left( \frac{-\Delta G^0}{RT} \right) (14)
\]

where \( a_{S^{2-}} \) is the activity of the oxygen ions in the slag, \( f_{S^{2-}} \) is the activity coefficient of the sulfide ions in the slag, \( R \) is the gas constant, \( T \) is the temperature in K, and \( \Delta G^0 \) is the standard Gibbs energy change. In the model, the ratio of the activity of \( O_2^- \) to the activity coefficient of \( S^{2-} \), \( a_{O_2^-}/f_{S^{2-}} \), is expressed as

\[
a_{O_2^-}/f_{S^{2-}} = \exp \left( \frac{-\xi}{RT} \right) (15)
\]

In the case of unary systems, \( \xi \) is a function of the temperature only. However, in a multicomponent system, \( \xi \) is described as a function of both the temperature and the composition:

\[
\xi = \Sigma (X_i \xi_i + \xi_{\text{mix}}) (16)
\]

where the subscript \( i \) denotes the component \( i \), \( X_i \) is the mole fraction of this component, \( \xi_i \) is expressed as a linear function of the temperature for each component in the slag in the absence of an interaction between the different species, \( \xi_{\text{mix}} \) is the mutual interaction between the different species.

According to the model,\textsuperscript{17} the sulfide capacities of the six-component slags can be expressed as follows:

\[
RT \ln C_s = \sum -X_{i=S^{2-}} \cdot \ln (a_{S^{2-}}/f_{S^{2-}}) = \sum -X_{i=S^{2-}} \cdot \ln (a_{S^{2-}}/f_{S^{2-}}) (17)
\]

Although the KTH model is valid for the atmosphere-slag interaction, in this paper it will be used to compare the values of \( C_s \).

2 EXPERIMENTAL

The plant data of 12 heats of low-carbon, Al-Si killed steel from the Dillinger Hütte Steel-plant in Dillingen, Germany, taken after the vacuum-degassing operation in the ladle-refining process, were used in this study. The liquid steel samples were taken using an automatic sampling system, while the slag samples were manually collected with a spoon and subjected to a chemical analysis. Because the oxygen analyses of the steel samples were not available, a logical way to estimate the oxygen levels in the steel bath was to estimate the activities of the \( Al_2O_3 \) and \( SiO_2 \) in the slag by thermodynamic calculations from the contents obtained by sampling and chemical analysis, and use them to estimate the oxygen potential in the steel bath, assuming a slag/metal equilibrium.\textsuperscript{18}

3 RESULTS AND DISCUSSION

The average chemical compositions of the analyzed metal and slag phases are summarized in Table 2.

The optical basicity for all the analyzed slags was in the narrow range \( \Lambda = 0.77-0.79 \).

3.1 Comparison of the sulfide capacity results

In this work, we applied equations (3) to (7) in order to calculate the measured values of the sulfide capacity with the calculated oxygen activity \( [a_o] \) in the steel according to Eq. (8) (subsequently called Case A) and Eq. (9) (subsequently called Case B). Then the results were compared with the calculated values of the sulfide capacity by Sosinsky-Sommerville (S-S) and the KTH model. Table 3 shows the calculated values of optical basicity, the measured and calculated values of the sulfide capacity, the oxygen activities \( [a_o] \) in the steel and alumina \( a_{(Al_2O_3)} \) and the silica \( a_{(SiO_2)} \) activities in the slag for the analyzed heats. As can be seen in Table 3, the calculated values for the alumina activities \( a_{(Al_2O_3)} \) are generally very low, i.e., below mass fractions \( w = 10^{-3} \% \) and \( 10^{-4} \% \) and sometimes even lower than \( w = 10^{-5} \% \). Comparatively, in the case of the calculated values of the silica activities \( a_{(SiO_2)} \) after the VD treatment, the calculated values of the silicon activity in the slag are stable at the level of \( w = 10^{-4} \% \) which corresponds to a \( SiO_2 \) content of a few per cent. This is in accordance with the published results.\textsuperscript{19}

Table 2: Chemical composition of analysed metal and slag

Tabela 2: Kemijska sestava analizirane kovine in lindre

<table>
<thead>
<tr>
<th>%</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>S</th>
<th>Al</th>
<th>T</th>
</tr>
</thead>
<tbody>
<tr>
<td>Average</td>
<td>0.10</td>
<td>0.35</td>
<td>1.51</td>
<td>0.0004</td>
<td>0.027</td>
<td>1855</td>
</tr>
<tr>
<td>Range</td>
<td>0.03-0.18</td>
<td>0.22-0.46</td>
<td>1.37-1.61</td>
<td>0.0002-0.0005</td>
<td>0.011-0.037</td>
<td>1838-1869</td>
</tr>
<tr>
<td>%</td>
<td>CaO</td>
<td>SiO₂</td>
<td>MgO</td>
<td>S</td>
<td>Al₂O₃</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>54.64</td>
<td>4.38</td>
<td>7.46</td>
<td>0.70</td>
<td>30.64</td>
<td></td>
</tr>
<tr>
<td>Range</td>
<td>52.49-57.00</td>
<td>2.38-8.20</td>
<td>4.10-12.16</td>
<td>0.46-1.03</td>
<td>27.51-34.38</td>
<td></td>
</tr>
</tbody>
</table>
Table 3: The calculated optical basicity, oxygen activities \([a_o]\) in steel and alumina \(a_{Al2O3}\) and silica \(a_{SiO2}\) activities in slag for analysed heats

<table>
<thead>
<tr>
<th>Heats</th>
<th>(a_{Al2O3})</th>
<th>(a_{SiO2})</th>
<th>([a_o]_{\text{Al}})%</th>
<th>([a_o]_{\text{Si}})%</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.78</td>
<td>0.000442</td>
<td>1.26E-05</td>
<td>5.77E-05</td>
</tr>
<tr>
<td>2</td>
<td>0.78</td>
<td>0.003583</td>
<td>7.34E-05</td>
<td>9.84E-05</td>
</tr>
<tr>
<td>3</td>
<td>0.77</td>
<td>0.000044</td>
<td>9.27E-06</td>
<td>7.04E-06</td>
</tr>
<tr>
<td>4</td>
<td>0.78</td>
<td>0.000007</td>
<td>5.68E-06</td>
<td>1.04E-04</td>
</tr>
<tr>
<td>5</td>
<td>0.78</td>
<td>0.000012</td>
<td>5.01E-06</td>
<td>7.94E-05</td>
</tr>
<tr>
<td>6</td>
<td>0.77</td>
<td>0.000150</td>
<td>2.03E-05</td>
<td>1.22E-04</td>
</tr>
<tr>
<td>7</td>
<td>0.78</td>
<td>0.000222</td>
<td>1.24E-05</td>
<td>6.37E-05</td>
</tr>
<tr>
<td>8</td>
<td>0.79</td>
<td>0.000616</td>
<td>1.92E-05</td>
<td>7.58E-05</td>
</tr>
<tr>
<td>9</td>
<td>0.78</td>
<td>0.000446</td>
<td>3.29E-05</td>
<td>1.05E-04</td>
</tr>
<tr>
<td>10</td>
<td>0.79</td>
<td>0.000060</td>
<td>8.77E-06</td>
<td>5.86E-05</td>
</tr>
<tr>
<td>11</td>
<td>0.78</td>
<td>0.000249</td>
<td>1.54E-05</td>
<td>7.72E-05</td>
</tr>
<tr>
<td>12</td>
<td>0.79</td>
<td>0.000024</td>
<td>1.25E-05</td>
<td>9.27E-05</td>
</tr>
</tbody>
</table>

Also, it can be seen from the results in Table 3 that the changes in the obtained values of the sulfide capacities at the end of ladle treatment were relatively small for the S-S model and the measured values derived by a calculation of the sulfide capacities from a prediction of the oxygen activity \([a_o]\) (Case A) and the KTH model results compared with those derived by the calculation of the sulfide capacities by a prediction of oxygen activity \([a_o]\) (Case A). Moreover, the obtained values of the sulfide capacities of the analyzed slags are in good agreement with earlier published results.

3.2 Comparison of the sulfur distribution ratio

The slag-metal sulfur distribution ratio \(L_s\) after desulfurization is another important parameter in the modeling of sulfur removal in steel making. In this work the predicted values of \(L_s\) were calculated using equation (8), suggested by M. T. Andersson et al. For the purpose of a comparison, the sulfide capacity was also calculated using the Sosinsky-Sommerville (S-S) approach based on the optical basicity concept according to be original parameters, as well as the KTH model. Then, the results were compared with the calculated values of the sulfur distribution ratio with the calculated oxygen activity \([a_o]\) (Case A) and the oxygen activity \([a_o]\) (Case B).

The comparison of the calculation determined sulfur distribution ratio \(L_{s\text{calc}}^\text{msr}\) using both cases with the measured \(L_{s\text{msr}}\) is shown in Table 4 and Figures 1 and 2.

As can be seen from Table 4 and Figures 1 and 2, both models generally exhibit higher discrepancy for the values of \(L_s\) in Case B compared with the results obtained in Case A. Also, it is evident that the correlation between the measured \(L_{s\text{msr}}\) and the calculated values of the sulfur distribution ratios \(L_{s\text{calc}}\) are more or less scattered. In contrast, it is clear that the predictions with the KTH model agree well with the measured results in Case A. The optical basicity concept, which is represented here by the Sosinsky-Sommerville (S-S) model, gives much higher predicted values. This is in good agreement with earlier published results.

The exceptions are the results obtained when the oxygen activity data is used in the metal phase \([a_o]\), (Case B). It is obvious that the data points show larger scattering, but both models give better \(L_s\) prediction compared with Case A.

Based on thermodynamic calculations, it can be summarised that the equilibrium state was possibly not

Table 4: The values of sulfur partition ratio for different calculated oxygen activities \([a_o]\) and \(L_{s\text{msr}}\) calculated by Sosinsky-Sommerville (S-S) and KTH models

<table>
<thead>
<tr>
<th>Heats</th>
<th>(L_{s\text{msr}})</th>
<th>(L_{s\text{calc}}) Case A</th>
<th>(L_{s\text{calc}}) Case B</th>
<th>(L_{s\text{msr}}) Case A</th>
<th>(L_{s\text{msr}}) Case B</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>3290</td>
<td>3940</td>
<td>1100</td>
<td>1945</td>
<td>543</td>
</tr>
<tr>
<td>2</td>
<td>2967</td>
<td>964</td>
<td>719</td>
<td>801</td>
<td>597</td>
</tr>
<tr>
<td>3</td>
<td>2233</td>
<td>8373</td>
<td>854</td>
<td>7454</td>
<td>760</td>
</tr>
<tr>
<td>4</td>
<td>1867</td>
<td>19479</td>
<td>1067</td>
<td>7491</td>
<td>410</td>
</tr>
<tr>
<td>5</td>
<td>1723</td>
<td>16515</td>
<td>1043</td>
<td>7049</td>
<td>445</td>
</tr>
<tr>
<td>6</td>
<td>1527</td>
<td>3463</td>
<td>579</td>
<td>1555</td>
<td>260</td>
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<tr>
<td>7</td>
<td>2060</td>
<td>7104</td>
<td>1381</td>
<td>3723</td>
<td>724</td>
</tr>
<tr>
<td>8</td>
<td>1984</td>
<td>5343</td>
<td>1353</td>
<td>3766</td>
<td>954</td>
</tr>
<tr>
<td>9</td>
<td>1520</td>
<td>2865</td>
<td>897</td>
<td>1813</td>
<td>567</td>
</tr>
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<tr>
<td>11</td>
<td>1228</td>
<td>5263</td>
<td>1048</td>
<td>2066</td>
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<td>12</td>
<td>1094</td>
<td>11944</td>
<td>1614</td>
<td>5167</td>
<td>698</td>
</tr>
</tbody>
</table>

Table 5: The differences between measured \(L_{s\text{msr}}\) and calculated values of sulfur partition ratio \(L_{s\text{calc}}\) for different calculated oxygen activities \([a_o]\) and \(L_{s\text{msr}}\) calculated by Sosinsky-Sommerville (S-S) and KTH models

<table>
<thead>
<tr>
<th>Heats</th>
<th>(\Delta) Case A (S-S)</th>
<th>(\Delta) Case B (S-S)</th>
<th>(\Delta) Case A (KTH)</th>
<th>(\Delta) Case B (KTH)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-650</td>
<td>2190</td>
<td>1345</td>
<td>2747</td>
</tr>
<tr>
<td>2</td>
<td>-2002</td>
<td>2247</td>
<td>2166</td>
<td>2370</td>
</tr>
<tr>
<td>3</td>
<td>-6140</td>
<td>1379</td>
<td>-5220</td>
<td>1473</td>
</tr>
<tr>
<td>4</td>
<td>-17613</td>
<td>800</td>
<td>-5624</td>
<td>1456</td>
</tr>
<tr>
<td>5</td>
<td>-14792</td>
<td>680</td>
<td>-5326</td>
<td>1278</td>
</tr>
<tr>
<td>6</td>
<td>-1936</td>
<td>948</td>
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<td>1267</td>
</tr>
<tr>
<td>7</td>
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<td>1336</td>
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<td>623</td>
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<td>953</td>
</tr>
<tr>
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<td>-8561</td>
<td>-63</td>
<td>-3926</td>
<td>629</td>
</tr>
<tr>
<td>11</td>
<td>-4035</td>
<td>180</td>
<td>-838</td>
<td>817</td>
</tr>
<tr>
<td>12</td>
<td>-10850</td>
<td>-520</td>
<td>-4073</td>
<td>396</td>
</tr>
</tbody>
</table>

Case A = \(L_{s\text{msr}} - L_{s\text{calc}}\)\[/\text{[a_o]_Al}\]
Case B = \(L_{s\text{msr}} - L_{s\text{calc}}\)\[/\text{[a_o]_Si}\]
obtained, since the vacuum degassing time for all the heats was almost the same.

Table 5 shows the differences between the measured and calculated values of the sulfur partition ratio $L_s$ for the analysed cases. The "−" means that the calculated values of $L_s$ are higher than the measured values.

It is clear that in Case A both models give calculated values that are higher than the measured values for almost all the analyzed heats. One possible reason for the increased deviation between the measured and calculated values of the sulfur distribution ratio $L_s$ could be the fact that using the well-known Ohta and Suito equations to calculate the alumina activity might not be appropriate for the slags whose silica content was too far away from the specified lower limit of $w = 10\%$. The second reason could be the relatively small number of analysed samples.

4 CONCLUSIONS

1. The equilibrium sulfur partition ratio, calculated by considering the reaction $2[\text{Al}] + 3[\text{O}] = (\text{Al}_2\text{O}_3)$ in equilibrium for the calculation of the oxygen activity in the steel during ladle treatment, was much higher compared with the reaction $[\text{Si}] + 2[\text{O}] = (\text{SiO}_2)$ with the measured sulfur partition ratio.

2. The Sosinsky-Sommerville optical basicity model gives higher values of the sulfur partition ratio $L_s$ compared with the KTH model.

3. The possible reason for the increased deviation between the measured and the calculated values of sulfur partition ratio $L_s$ was that the use of the well-known Ohta-Suito equations to calculate the alumina activity might not be appropriate for the slags whose silica content was too far away from the specified lower limit of $w = 10\%$.

4. The KTH model is an applicable tool to predict the sulfur partition ratio $L_s$.

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