Sulphur Transfer between Liquid Iron and Basic Slags
Using Carbon and Silicon as Deoxidizers

By Martin G. FROHBERG** and Arman NILAS**

Synopsis
The sulphur transfer from metal to slag was studied by thermogravimetric and volumetric methods. It has been observed that both silicon and carbon were involved in special reactions, which influence the desulphurization process. The attainment of equilibrium of silicon in metal with silica in slag stops further transfer of sulphur from metal to slag as brought about by silicon.

I. Introduction
Sulphur transport from liquid metal to slag, which proceeds during the removal of oxygen in metal by various deoxidizers has not yet been investigated thoroughly. During the desulphurization of iron the reduction of iron (II) oxide formed requires reducing agents. Research workers could provide no satisfactory answer to date in the case of carbon-saturated iron containing silicon, as to whether carbon or silicon or both play a major role as a reducing agent in the desulphurization process. The aim of present experiments was to elucidate by means of thermogravimetric and volumetric methods the role of the said elements during desulphurization.

The sulphur transport relation in liquid iron in presence of excess lime in slag is represented as follows:

\[ [\text{FeS}] + \text{CaO} = \text{CaS} + \text{FeO} \]  \(\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdOTS04
determined by chemical analysis.

### III. Experimental Results

The results of the experiments carried out by thermogravimetric method are given in Tables 1 and 2. Table 1 contains the results of the runs carried out at 1490°C employing a rate of heating of 10°C/min. These results plotted in Fig. 1 show that the percent sulphur transfer according to reaction (2) ($A_s$) decreases with the increase of the initial silicon content of the metal. As the heating rate in these runs was too slow, the dissolved sulphur in the liquid iron had enough time to react with the solid slag. The dotted line in Fig. 1 shows $A_s$ for liquid iron and solid slag and is found to follow a similar curve.

Table 2 contains the results of the runs carried out at 1550°C employing a higher rate of heating of 50°C/min. The percentage of reaction between solid slag and liquid iron according to reaction (2) decreased in this case as shown in Fig. 2. Figures 3 and 4 show the original records of run 5 and run 12 which were carried out at different heating rates. A sulphur balance conducted in each experiment showed that part of the sulphur was removed from the system. This can be due to the sulphur evaporation and the formation of volatile silicon monosulphide, which leaves the system in form of vapour and condenses in the colder part of the apparatus. The formation of silicon monosulphide and its volatilization has already been reported by various investigators.23–26 The loss in weight of run 9 in Table 1, which was carried out without any slag, indicates decreases of 0.15%.

![Percent sulphur transport through carbon.](image)

**Fig. 1.** The effect of initial silicon content on the sulphur transport through carbon at 1490°C (rate of temperature rise 10°C/min). Thermogravimetric investigation.

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**Table 1.** Experimental results of the thermogravimetric investigations at 1490°C. Slag/metal ratio 1:1. Rising rate of temperature 10°C/min.

<table>
<thead>
<tr>
<th>Run No</th>
<th>Fe (g)</th>
<th>Silicon in iron, (initial) (%)</th>
<th>Sulphur in iron, (initial) (%)</th>
<th>Sulphur in iron, (final) (%)</th>
<th>Silica in slag, (final) (%)</th>
<th>Sulphur in slag, (final) (%)</th>
<th>CO calc. accord. Eq. (2) (mg)</th>
<th>$W_{act.}$ (mg)</th>
<th>$W_{act. (c)}$ (mg)</th>
<th>$A_s$ (%)</th>
<th>$A_{s,c}$ (%)</th>
<th>Total sulphur loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
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<td>0.81</td>
<td>0.01</td>
<td>1.38</td>
<td>46.5</td>
<td>46</td>
<td>44.7</td>
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<td>96</td>
<td>13.8</td>
<td>13.8</td>
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</tr>
<tr>
<td>2</td>
<td>6.619</td>
<td>0.81</td>
<td>0.01</td>
<td>1.43</td>
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<td>44</td>
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<tr>
<td>3</td>
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<td>29</td>
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<tr>
<td>4</td>
<td>6.864</td>
<td>0.54</td>
<td>0.40</td>
<td>0.013</td>
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<tr>
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<td>23</td>
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<td>6.846</td>
<td>6.87</td>
<td>0.80</td>
<td>6.71</td>
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<td>19</td>
<td>—</td>
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</tr>
</tbody>
</table>

**Nomenclature**

$W_{act.}$: actual weight decrease of liquid metal-solid slag and liquid metal-liquid slag system

$W_{act. (c)}$: actual weight decrease of liquid metal-solid slag

$A_s$: $W_{act.}/CO_{calc.}$

$A_{s,c}$: $W_{act. (c)}/CO_{calc.}$

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**Table 2.** Experimental results of the thermogravimetric investigations at 1550°C. Slag/metal ratio 1:1. Rising rate of temperature 50°C/min.

<table>
<thead>
<tr>
<th>Run No</th>
<th>Fe (g)</th>
<th>Silicon in iron, (initial) (%)</th>
<th>Sulphur in iron, (initial) (%)</th>
<th>Sulphur in iron, (final) (%)</th>
<th>Silica in slag, (final) (%)</th>
<th>Sulphur in slag, (final) (%)</th>
<th>CO calc. accord. Eq. (2) (mg)</th>
<th>$W_{act.}$ (mg)</th>
<th>$W_{act. (c)}$ (mg)</th>
<th>$A_s$ (%)</th>
<th>$A_{s,c}$ (%)</th>
<th>Total sulphur loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>6.808</td>
<td>0.81</td>
<td>0.006</td>
<td>1.38</td>
<td>48.5</td>
<td>45</td>
<td>13.1</td>
<td>93</td>
<td>27</td>
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<tr>
<td>11</td>
<td>6.862</td>
<td>0.29</td>
<td>0.26</td>
<td>0.005</td>
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<td>45.0</td>
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<td>62</td>
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<tr>
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<td>1.74</td>
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<td>50</td>
<td>12</td>
<td>+12.2</td>
</tr>
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<td>1.78</td>
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<td>1.05</td>
<td>1.14</td>
<td>43.0</td>
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<td>6.80</td>
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<td>2.06</td>
<td>1.63</td>
<td>51.0</td>
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<td>26</td>
<td>8.82</td>
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<td>16</td>
<td>6.827</td>
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<td>0.68</td>
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<td>1.10</td>
<td>1.22</td>
<td>40.5</td>
<td>19</td>
<td>5.26</td>
<td>48</td>
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</tbody>
</table>

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**Research Article**
Sulphur transport combining the reactions liquid metal-solid slag and liquid metal-liquid slag

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Sulphur transport through carbon at 1 550°C (rate of temperature rise 50°C/min). Thermogravimetric investigation.

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Table 3. Experimental results of the volumetric investigations at 1 600°C. Lime-alumina slag (ratio 1:1).

<table>
<thead>
<tr>
<th>Row No.</th>
<th>Run No.</th>
<th>17</th>
<th>18</th>
<th>19</th>
<th>20</th>
<th>21</th>
<th>22</th>
<th>23</th>
<th>24</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Sulphur in iron, (initial) (%)</td>
<td>1.913</td>
<td>1.913</td>
<td>1.830</td>
<td>1.913</td>
<td>1.913</td>
<td>1.913</td>
<td>1.810</td>
<td>1.913</td>
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<tr>
<td></td>
<td>Sulphur in iron, (final) (%)</td>
<td>0.139</td>
<td>0.194</td>
<td>0.168</td>
<td>0.010</td>
<td>0.014</td>
<td>0.010</td>
<td>0.120</td>
<td>0.010</td>
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<td>2</td>
<td>Silicon in iron, (initial) (%)</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>0.4</td>
<td>0.8</td>
<td>1.3</td>
<td>—</td>
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</tr>
<tr>
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<td>Silicon in iron, (final) (%)</td>
<td>—</td>
<td>—</td>
<td>—</td>
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<td>0.299</td>
<td>0.58</td>
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<tr>
<td>3</td>
<td>Slag/metal ratio (%)</td>
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<td>0.15</td>
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<td>0.15</td>
<td>0.13</td>
<td>0.08</td>
<td>0.15</td>
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<tr>
<td>4</td>
<td>Sulphur in slag, (final) (g)</td>
<td>7.14</td>
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<td>6.65</td>
<td>7.04</td>
<td>6.9</td>
<td>13.6</td>
<td>6.6</td>
</tr>
<tr>
<td>5</td>
<td>Silica in slag, (final) (g)</td>
<td>1.07</td>
<td>1.265</td>
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<td>1.0</td>
<td>1.055</td>
<td>0.9</td>
<td>1.08</td>
<td>0.98</td>
</tr>
<tr>
<td>6</td>
<td>Increase in pressure (mmHg)</td>
<td>32.0</td>
<td>32.4</td>
<td>29.5</td>
<td>25.0</td>
<td>13.6</td>
<td>4.7</td>
<td>29.0</td>
<td>27.5</td>
</tr>
<tr>
<td>7</td>
<td>Quantity of evolved CO-gas calculated according to ideal gas law (mg)</td>
<td>1 225</td>
<td>1 240</td>
<td>1 130</td>
<td>957</td>
<td>521</td>
<td>179</td>
<td>1 185</td>
<td>1 050</td>
</tr>
<tr>
<td>8</td>
<td>Quantity of transported sulphur in slag calculated according to Eq. (2) (mg)</td>
<td>1 400</td>
<td>1 420</td>
<td>1 290</td>
<td>1 090</td>
<td>595</td>
<td>205</td>
<td>1 350</td>
<td>1 200</td>
</tr>
<tr>
<td>9</td>
<td>Quantity of transported sulphur in slag calculated according to Eq. (3) (mg)</td>
<td>—</td>
<td>—</td>
<td>—</td>
<td>372</td>
<td>897</td>
<td>1 410</td>
<td>—</td>
<td>300</td>
</tr>
<tr>
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<td>Sulphur loss according to the calculations of CO-values (mg)</td>
<td>374</td>
<td>299</td>
<td>372</td>
<td>440</td>
<td>407</td>
<td>288</td>
<td>340</td>
<td>410</td>
</tr>
<tr>
<td></td>
<td>Sulphur loss according to the calculations of CO-values (%)</td>
<td>19.6</td>
<td>15.6</td>
<td>20.3</td>
<td>23.0</td>
<td>21.3</td>
<td>15.1</td>
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<td>21.4</td>
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<tr>
<td>11</td>
<td>Percent sulphur transport through carbon (A_d) (%)</td>
<td>100</td>
<td>100</td>
<td>100</td>
<td>74</td>
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<td>100</td>
<td>80</td>
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<td>Total sulphur loss (%)</td>
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<td>44.4</td>
<td>52.8</td>
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</table>

Research Article
Table 4. Experimental results of the volumetric investigations at 1600°C. Lime-alumina-silica slag with an initial silica content of 6.9%.

<table>
<thead>
<tr>
<th>Row No.</th>
<th>Run No.</th>
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Fig. 5. The CO evolution during the reaction of liquid carbon-saturated iron with lime-alumina slag (ratio 1:1) at 1600°C.

Fig. 6. The CO evolution during the slag metal reaction of liquid carbon-saturated iron with lime-alumina slag (ratio 1:1) at 1600°C and its dependence from initial silicon content.

centration in iron. The total amount of sulphur transported in the slag without considering the sulphur loss due to evaporation is attained through carbon, as no other deoxidizer is contained in the metal. On the other hand, the results in the Fig. 6 show that the carbon monoxide evolution decreases with an increasing content of silicon in the metal. This means that the sulphur transfer through carbon according to

Eq. (2) decreases with increasing silicon in the metal, if the initial slag does not contain silica. The observation of a constant pressure at the end of a run leads to the fact that the equilibrium of the reaction was reached. The concentrations of silica in slag and of silicon in metal at the end of heats given in Table 3 show that silica equilibrium is attained according to
IV. Discussion

The results obtained by the two experimental methods given in Figs. 1, 2, and 8 show a large difference of the rate of sulphur transfer through carbon. This can be due to either of following reasons during the volumetric experiment:

1. The relatively lower initial pressure,
2. The higher temperature,
3. The larger quantities of reactants used, and/or
4. The induction heating (stirring effect).

The above factors influence also the evaporation rate of sulphur from the slag and so there is a relatively high sulphur loss during the volumetric procedure. But the results obtained by both experimental procedures show that the sulphur transfer proceeds through both carbon and silicon, provided silicon acts as a deoxidizer. To have a better understanding of the sulphur transfer mechanism from metal to slag, it is necessary to find out in which form the elements involved in the reactions are present in the system. The nature of sulphur transfer at present is assumed to be electrochemical. The dissolved sulphur in metal reacts with the free oxygen ions in the slag, whereas the oxygen ions are an indicator of the basicity in the slag. The general equation for sulphur transfer is then given as follows:

\[ [\text{S}] + (\text{O})_{\text{slag}} = [\text{S}]_{\text{slag}} + [\text{O}] \]  \quad (5)

Splitting the above reaction in its transport steps gives:

\[ [\text{S}] + 2e^- = [\text{S}]_{\text{slag}} \]  \quad (6)

\[ (\text{O})_{\text{slag}} = [\text{O}] + 2e^- \]  \quad (7)

The above Eq. (6) shows that a transfer of one sulphur atom to the slag requires two electrons. These electrons come from the oxygen ions in the slag according to reaction (7). As soon as reaction (7) reaches its equilibrium state, no more sulphur can be transported to the slag. But a disturbance in the state of equilibrium may cause a new reflow of the sulphur from metal to slag. The present metalloids carbon and silicon in the metal can disturb the state of equilibrium through the reactions:

\[ [\text{C}] + (\text{O})_{\text{slag}} \rightarrow \text{CO}_{\text{gas}} + 2e^- \]  \quad (8)

\[ \frac{1}{2} [\text{Si}] + 4(\text{O})_{\text{slag}} \rightarrow \frac{1}{2}(\text{SiO}_2)_{\text{slag}} + 4e^- \]  \quad (9)

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The above reactions supply the electrons, which affect the further sulphur transfer, and the quantity of electrons thus set free depends on the rates of the reactions (8) and (9).

In the first part of the experiments carried where no silicon was present in the metal, the sulphur transfer was caused only through the reaction (8). The rate of the reaction (8) is given by the growth of the carbon monoxide bubbles at the active sites. When both carbon and silicon are present in liquid iron, both reactions (8) and (9) take place. The silicon transfer from metal to slag is relatively faster than the reaction (8). Reaction (9) can carry on until the difference of chemical potential of silicon in metal and in the slag is reduced to zero. When there is no chemical potential difference as shown in run 26 (Fig. 7) sulphur transfer proceeds according to reaction (8) only.

V. Conclusions

Following conclusions have been drawn from the present investigations:

(1) The reactions between solid slag and liquid metal during the thermogravimetric procedure show the similarity in the influence capability of carbon and silicon to the sulphur transfer.

(2) Sulphur losses from the slag are due to evaporation of sulphur and the formation of silicon monosulphide.

(3) Carbon and silicon take part in the desulphurization process.

(4) Under the equilibrium state of silica in slag and silicon in metal, sulphur transfer occurs only through carbon monoxide evolution.

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