A Thermodynamic Study of Silicon Containing Gas around a Blast Furnace Raceway

J. GUSTAVSSON, A. M. T. ANDERSSON and P. G. JÖNSSON
Div. of Applied Process Metallurgy, Royal Institute of Technology, SE-100 44 Stockholm, Sweden.
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The equilibrium conditions for silicon transfer from ash to the liquid metal phase via SiO gas have been discussed by several authors. However, no published calculations have been found using the most modern thermodynamic models available. Since there are major differences in the results of calculations using different thermodynamic models and the models are continuously being improved, new equilibrium calculations on SiO and SiS gas formation have been performed using the recently developed models. Different ingoing compositions of coke ash, coal powder ash and blast air were used in the calculations. The compositions chosen represent blast furnace no. 3 at SSAB in Luleå, Sweden. Temperature was found to be the major factor influencing the equilibrium silicon level in the gas phase. At low temperatures (below 1600°C) the total gas pressure was also seen to influence the silicon content in the gas phase. The main reason for this is that below 1600°C, the amount of liquid slag at equilibrium increases with the total gas pressure. Liquid slag contains large amounts of silica that then can not be found in the gas phase. Higher carbon activity is usually expected to result in higher SiO gas levels in the blast furnace. The equilibrium calculations show that increased carbon activity increases the amount of silicon in the gas phase at temperatures up to about 1600°C, but that at higher temperatures SiC is formed that decreases the equilibrium silicon level in the gas phase.

KEY WORDS: ironmaking; blast furnace chemistry; thermodynamics; calculation; slags; cast iron; gases; silicon compounds.

1. Introduction

The formation of silicon monoxide gas from ashes and slag in the blast furnace followed by its reduction to dissolved silicon in hot metal is regarded as the main route for silicon transfer in the blast furnace.1–24) During the operation of Swedish blast furnaces it has been noticed that the final silicon content in iron is dependent on the total gas pressure in the lower part of the blast furnace shaft. In a previous publication it was shown that the calculated equilibrium content of silicon in hot metal is higher at lower blast pressures. 25) It was therefore deemed interesting to carry out an in-depth study of the equilibrium conditions for silicon monoxide gas formation.

The first part of the paper describes the thermodynamic calculation procedure. Thereafter, the effect of i) coke and coal powder qualities, ii) total gas pressure, and iii) graphite activity on the silicon distribution are presented and discussed.

2. Theoretical Considerations

Silicon monoxide is usually assumed to form from coke ash and ash from injectants as a result of any of the following reactions5–8,10,12,13,15–17,19,20,22–24):

\[
\text{SiO}_2 \text{(in ash)} + \text{CO} \rightarrow \text{SiO} + \text{CO}_2 \quad (1)
\]

\[
\text{SiO}_2 \text{(in ash)} + \text{C} \rightarrow \text{SiO} + \text{CO} \quad (2)
\]

\[
2\text{SiO}_2 + \text{SiC} \rightarrow 3\text{SiO} + \text{CO} \quad (3)
\]

\[
\text{SiC} + \text{CO} \rightarrow \text{SiO} + 2\text{C} \quad (4)
\]

Usually reaction (2) is given as the dominating reaction5,6,12,13,15–17,19,20,22–24). Some sources also give reaction (1) as the dominating reaction7,8,10). The difference between reactions (1) and (2) is Boudouard's reaction:

\[
\text{C} + \text{CO}_2 \rightarrow 2\text{CO} \quad (5)
\]

If reaction (1) occurs, the CO2 gas formed will react with carbon (from coke or injectants) according to reaction (3) resulting in the net reaction (2). Most of the coke in the blast furnace process is consumed in the raceway. Before the coke is burnt in the raceway, it is exposed to high tem-
temperatures. At that time coke ash can react with carbon in the coke or carbon monoxide, forming SiO gas according to reaction (1) or (2). Coke ash silica can also react with coke carbon to form SiC. This SiC can further react according to reaction (3) or (4), forming SiO gas.\(^{3,17,19}\) It is important to point out that coal powder is injected through the tuyeres without prior heating, and to a great extent is burnt inside the raceway. When the coal powder has been burnt off, unburned char and ash will remain. The remaining ash will partly be collected in the region around the raceway and be burned char and ash will remain. The remaining ash will partly be collected in the region around the raceway and be able to react according to reaction (1) or (2).\(^{21}\) Some of the ash around the raceway area also can form SiC followed by reaction (3) or (4).\(^ {21}\)

Equilibrium conditions for silicon monoxide gas formation have been calculated using input data from the blast furnace no. 3 at SSAB Tunnplåt AB in Luleå, Sweden. Similar calculations have been performed by Mathieson \textit{et al.}, though representing other ash compositions and less species considered.\(^ {26}\) Calculations have been carried out to examine how much silicon can be present in the gas at equilibrium conditions. The following assumptions were made in the calculations:

- The only components included in the calculation were:
  - Blast air, including \(O_2\), \(N_2\) and \(H_2O\).
  - Coke, including Fe, CaO, SiO\(_2\), AlO, MgO and S. The oxidation degrees given in the SSAB Tunnplåt database were used.
  - Coal powder, including Fe, CaO, SiO\(_2\), MnO, AlO, MgO, NaO and H\(_2O\).

- The reason for not including other factors in the calculation, such as iron from the pellets and primary slag, is that most SiO gas is assumed to form from the ashes, which has higher silica activity than other components. The formation of SiO gas is assumed to occur before the ashes are mixed with other components. The amount of different phases and the phase compositions at equilibrium have been calculated. There is no possibility to differentiate between different ashes and slags, at equilibrium they will mix. This also includes other phases that can be mixed with the ashes (for instance slags) and that would lower the silica activity and therefore result in less SiO gas formation. In data to the calculations are the amount of different elements per metric ton of hot metal, temperature and total gas pressure. Instead of giving the amount of carbon, the activity level has been given.

- A reference period measure was created by recording the total consumption of coke and coal powder in the blast furnace, as well as their source and quality. Figures representing the total consumption in the blast furnace during one week of operation were used as the reference period. The figures were expressed in kg per metric ton of hot metal.
  - When different coke and/or coal powder sources were used than under the reference period the amount of coke or coal powder was calculated so that the total dry amount of components other than ash was kept constant.

- The carbon activity was assumed to be unity with reference to graphite, except for some calculations that examine the effect of different carbon activities on the SiO formation process. The amount of carbon in the system was calculated so that the carbon activity in the system was equal to the carbon activity given as in data. In the case that graphite was given to be stable, the amount of carbon in the system was calculated to the minimum possible amount to obtain graphite stability (\textit{i.e.} the amount of graphite was calculated to be zero at equilibrium).

The oxidation level of the components in the ashes was estimated by SSAB Tunnplåt based on the chemical composition. More specifically, the elemental composition has been determined and thereafter expressed using a standardized oxygen potential. It should be noted that this simplification might result in values that differ from the real oxygen potential. However, it is seen to be of less importance for this study, since the degree of oxidation was found to have only a small influence on the total amount of oxygen in the system. In the reference case 5.6% of the oxygen in the system comes from oxides in coke and coal powder ash. Since most ash components are added with the correct oxygen potential the difference in oxygen in the system resulting from incorrect oxygen potential in ingoing components will be much less than 5.6%. Additionally, most of the oxygen will exist as a component in the CO gas at equilibrium conditions. The amount of carbon in the CO gas is based on the assumption that graphite is stable, as mentioned earlier. Therefore, small changes in the total amount of oxygen in the system will only have a small influence on the result of the calculation. In fact, the change is so small that it can be ignored, being negligible when compared with the errors that may occur due to uncertainties in the thermodynamic models that have been used.

### 2.1. Method of Solution

The calculations were performed using the computer software Thermo-Calc. Almost the same databases, including thermodynamic data that have been used previously to calculate equilibrium between hot metal and slag,\(^ {25}\) have been used in these calculations. However, in this case some preliminary calculations showed that the equilibrium silicon content in the liquid metal phase exceeded the maximum amount of 5 wt% that the database TCFe3\(^ {27}\) can handle. It was therefore decided to use the database SSol2\(^ {28}\) to describe the liquid metal phase. This database is not as precise as TCFe3 when used to describe high iron content melts, but was the best available choice for equilibrium calculations of high silicon metallic melts. The reason for getting silicon contents above 5 wt% is that only iron from ash has been included in the calculations, which means that only small amounts of iron have been included. The databases TCFe3 and SSol2 have been compared with each other for a tapped hot metal composition at SSAB Oxelösund AB. The reason of choosing a hot metal from SSAB Oxelösund AB instead of a hot metal from SSAB Tunnplåt AB is that the silicon content is higher at SSAB Oxelösund AB. The composition of the hot metal, the calculated activities and the reference state for the activities can be found in Table 1. The temperature of the hot metal was 1493°C. It can be seen that the relative difference in sulfur activity is extremely large and that there also is a significant difference for titanium. The difference in activity for other elements can be ignored. Titanium is not included in the slag phase, and was not included in significant
amounts in phases other than the liquid metal phase. For this reason the difference in titanium activity between the databases does not show significant influence on the result of the calculation. Preliminary calculations using the TCFe3 database showed that most sulfur was present in the gas phase at temperatures where liquid metal phase exists. For this reason no major differences in the calculation result could be expected when using incorrect sulfur activities in the liquid metal phase. As mentioned earlier, the use of the TCFe3 database would give big problems to calculate the behavior of silicon in the liquid metal phase. This can be handled by the SSol2 database and since the use of the SSol2 database does not give any large deviations in calculation results because of the bad handling of sulfur, it was seen as a better choice than using the TCFe3 database.

For the slag phase, the database Slag1 has been used. The equilibrium calculations in the gas phase were made using the database SSub3. Furthermore, the description of solid phases was mainly taken from the SSub3 database, but also data from the SSol2 database. The SSub3 and SSol2 databases are reported to be compatible with each other.

As has been pointed out earlier, different databases have been used for different phases. More specifically, the best available database to express each phase has been selected. However, it should be noted that there are still no databases that are completely relevant for blast furnace conditions. Also, it should be noted that Thermo-Calc AB as well as other software suppliers do not make any guarantees for the error ranges of the databases. The results of the calculations may therefore differ from the real equilibrium conditions. In order to get more reliable calculations better experimental data is needed. Such experiments would, however, be extremely expensive and almost certainly have large error ranges.

In these preliminary equilibrium calculations, several databases have been combined in different ways and the calculation results for different combinations have been compared. No major differences in the amount of silicon calculated in the gas phase have been noticed. Despite this, other phases than gas that were calculated to be stable at equilibrium, differed between different database combinations. Some database combinations showed completely different stable phases than other combinations. Calculations have also been performed to study the effect of different carbon activities and different total gas pressure on the silicon distributions. These calculations may contain large errors as a result of the uncertainty of the databases.

### 3. Results and Discussion

The influence of i) coke and coal powder qualities, ii) total gas pressure, and iii) graphite activity on the equilibrium silicon distribution between phases has been evaluated using thermodynamic calculations. The amount of each element in the system per unit of produced hot metal has been used as input, with the exception for carbon where unity graphite activity has been assumed. The composition of the raw materials used is expressed in kg per metric ton of hot metal as presented in Table 2. The table is calculated based on data from blast furnace no. 3 at SSAB Tunnplåt AB in Luleå, Sweden. Since the amount of carbon is not used as input it is not presented in the table.

#### 3.1. Effect of Coke and Coal Powder Qualities on Silicon Distribution

Thermodynamic calculations of the effect of coke and coal powder qualities on predicted silicon distributions were made. Figure 1 shows the resulting silicon distribution between phases at calculated equilibriums for different coke qualities and temperatures. The predictions were made for the injection of coal quality 1 and a blast composition as described in Table 2. Furthermore, the total gas pressure

![Table 1. Comparison of activities calculated with the TCFe3 database and the SSol2 database for a high silicon hot metal. The temperature is 1493°C.](image)

| Element | wt-% | Activity TCFe3 | Activity SSol2 | Reference state |
|---------|------|----------------|----------------|-----------------|
| C       | 4.35 | 7.58E-01       | 7.72E-01       | Graphite        |
| Si      | 0.02 | 3.70E-05       | 4.22E-05       | Solid          |
| Mn      | 0.31 | 1.17E-03       | 1.14E-03       | Mol           |
| S       | 0.042| 2.11E-08       | 1.72E-03       | S2(g)         |
| V       | 0.28 | 8.00E-05       | 8.44E-05       | VI             |
| Ti      | 0.16 | 5.61E-06       | 1.16E-05       | Ti(s)         |
| Fe      | 93.94| 6.60E-01       | 6.58E-01       | Fe(s)         |

![Fig. 1. Equilibrium silicon distribution between phases at different temperatures using different coke qualities. Coal 1 is injected.](image)
was set to 3.7 atm in all calculations and other parameters were set as indicated in the figure. As can be seen in Fig. 1, the differences in silicon distribution between coke quality 1 and coke quality 2 are small. However, the behavior of coke quality 3 differs from the other qualities at temperatures up to 1 600°C. This is mainly due to coke quality 3 containing more aluminum than coke quality 1 and coke quality 2, see Table 2. Despite these differences in silicon distribution, the predicted equilibrium amounts of silicon in the gas phase are relatively constant at each temperature for the different coke qualities.

Figure 2 shows the effect of different coal powder qualities on the silicon distribution in the different phases at different temperatures. The calculations were carried out in a way that would simulate the use of coke quality 1 and blast air with data as indicated in Table 2. As in the previous case, the total gas pressure was set to 3.7 atm in all calculations and all other parameters were set as indicated in the figure. From the results in Fig. 2 it can be seen that there are almost no differences in the results of the calculation between coal qualities 2 and 3. However, coal quality 1 contains much more iron than coal quality 2 and 3, which result in more metallic melt.

3.2. Effect of Total Gas Pressure on Silicon Distribution

As mentioned earlier, practical experience from operation of blast furnaces has indicated that the total gas pressure has an influence on the silicon content in the hot metal.

Table 2. Consumption of coke, coal powder and blast used in the calculations. The consumption is given as kg/thm. The table is calculated based on data from SSAB Tunnplåt AB in Luleå, Sweden.

| Coke 1 | Coke 2 | Coke 3 | Coal 1 | Coal 2 | Coal 3 | Blast |
|--------|--------|--------|--------|--------|--------|-------|
| Fe     | 1.989  | 1.013  | 1.676  | 4.704  | 0.493  | 0.5  |
| CaO    | 0.729  | 0.020  | 1.475  | 0.994  | 0.188  | 0.268 |
| SiO₂   | 21.047 | 19.371 | 21.053 | 5.683  | 5.327  | 5.566 |
| Al₂O₃  | 9.380  | 8.134  | 13.838 | 2.018  | 1.806  | 2.002 |
| MgO    | 0.398  | 0.131  | 0.379  | 0.371  | 0.147  | 0.204 |
| Na₂O   | 0.133  | 0.131  | 0.235  | 0.039  | 0.036  | 0.541 |
| S      | 1.471  | 1.731  | 2.548  | 1.054  | 1.038  | 0.865 |
| MnO    | -      | -      | -      | 0.074  | 0.006  | 0.007 |
| H₂O    | -      | -      | -      | 1.335  | 1.263  | 1.418 |
| O₂     | -      | -      | -      | -      | -      | -540.284 |
| N₂     | -      | -      | -      | -      | -      | 914.281 |

For this reason, it was believed to be of interest to calculate how the total gas pressure influences the equilibrium distribution of silicon between the gas phase and other phases. Figure 3 shows a plot of the percentage of the predicted total amount of silicon in the system found in the gas phase at equilibrium as a function of the total gas pressure. The different lines in the figure represent different temperatures. The calculation is performed using the ingoing amounts of coke quality 1, coal quality 1 and blast as defined in Table 2 and by setting the graphite activity equal to unity. Note that in the calculation different solid and/or liquid components are stable in different temperature and pressure regions. This explains the characteristics of the curves in Fig. 3. The increases in the calculated equilibrium silicon content in the gas phase at 1.1 atmospheres gas pressure and 1 700°C as well as at 2.7 atmospheres gas pressure and 1 800°C is a result of the same phenomena. Increasing the total gas pressure from these points decreases the amounts of SiC and CaS and increases the amount of (Al₂O₃)(CaO)₂(SiO₂) (gehlenite). Decreasing the amount of CaS leads to that sulfur has to be transferred to another phase. The calculation shows that most sulfur is transferred to SiS gas. The transfer mechanism can not be found in equilibrium calculations but the sum reaction can be calculated. The sum reaction at 1 800°C when the total gas pressure changed from 2.47
to 2.84 atm has been calculated. The reaction would be extremely long if all constitutions in the gas phase were given. Therefore, only the elements in the gas phase are given. Elements dissolved in liquid iron are indicated with a line under the element. The calculated sum reaction can be written as:

\[
\begin{align*}
11.81 \text{ AlN (s)} + 1.03 \text{ Al (g)} + 44.01 \text{ C (g)} + 2.13 \text{ Ca (g)} + 0.16 \text{ Fe (l)} + 16.77 \text{ SiC (s)} + 10.71 \text{ CaS (s)} & \\
\rightarrow 6.42 (\text{Al}_2\text{O}_3)(\text{CaO})_2(\text{SiO}_2) (\text{s}) + 11.81 \text{ N (g)} + 10.71 \text{ S (g)} + 10.22 \text{ Si (g)} + 0.06 \text{ C} + 0.16 \text{ Fe(l)} + 0.13 \text{ Si} + 60.72 \text{ C (graphite)}
\end{align*}
\] (6)

As can be seen in reaction (6), silicon from the decrease of SiC partly goes to the gas and partly to formation of \((\text{Al}_2\text{O}_3)(\text{CaO})_2(\text{SiO}_2)\). The total effect is that the calculated equilibrium amount of silicon in the gas phase increases.

The solid and liquid phases existing at different total gas pressures at 1 800°C are shown in Fig. 4. AlN, liquid metal and SiC are stable at all gas pressures. CaS is stable between 1.48 atm and 2.71 atm. Gehlenite is also stable at 2.59 atm and higher pressures.

It was seen in Fig. 3 that silicon decreases in the gas when increasing the gas pressure at low temperatures (1 400 and 1 500°C). According to the calculations, most of the decrease is due to liquid slag formation. Liquid slag contains large amounts of silica. However, it should be noted that no slag is formed at higher temperatures at the gas pressures studied. As can be seen in Fig. 5, the amount of silicon in the gas is relatively constant at a temperature of 1 500°C at low total gas pressures. At higher gas pressures a slag phase begins to form that includes high levels of silicon.

From Fig. 3 it can be seen that the calculated equilibrium amount of silicon in the gas phase at 1 600°C is almost constant at all total gas pressures. This does not mean, however, that the behavior of silicon in the system is independent of the total gas pressure. From Fig. 6 it can be seen that the calculated equilibrium silicon content in other phases than gas are influenced by a change of the total gas pressure. At low total gas pressures it can be seen that much silicon is present as SiC and at higher total gas pressures silicon is present in different Al–Ca–Si–O compounds.

According to the calculations there also exists a liquid metal phase that is stable at all examined temperatures and total gas pressures. Since the only ingoing components are coke ash, coal powder ash and blast, the liquid metal phase contains only small amounts of iron. As a result the liquid metal phase is not especially representative for a real blast furnace case. However, the results of the calculations indicate that it is thermodynamically possible to form metal droplets with high silicon content within the coke pieces without adding iron from outside the coke pieces. The calculated equilibrium amount of the liquid metal phase can be found in Fig. 7. It can be seen that the amount of liquid metal increases with temperature in all examined cases.

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**Fig. 4.** Solid and liquid phases calculated at different total pressures and with a constant temperature of 1 800°C.

**Fig. 5.** Solid and liquid phases present in the system at calculated equilibrium conditions at 1 500°C for different total gas pressures.

**Fig. 6.** Silicon distribution between phases at 1 600°C.

**Fig. 7.** Amount of calculated equilibrium liquid metal phase at different total gas pressures for different temperatures. The temperature is given in °C.
except for at low total gas pressures at 2000°C. The calculated equilibrium silicon content of the liquid phase can be found in Fig. 8. As can be seen, the equilibrium silicon content of the metal melt is extremely high. Figure 8 also shows that the silicon composition in the liquid metal phase increases with temperature in all examined cases. The effect of the total gas pressure on the calculated equilibrium amount of liquid metal differs at different temperatures, see Fig. 7. At low temperatures, the amount of liquid metal decreases when total gas pressure increases. This is most probably due to liquid slag that is formed due to oxidation of silicon and some iron, which are the main elements in the liquid metal phase. At high temperatures, the amount of liquid metal phase slightly increases when total gas pressure increases. At lower temperatures silicon is present in different solid and/or liquid phases. At the highest temperatures, 1900°C and 2000°C, all silicon is found in the gas phase and in the liquid metal phase. Since the silicon content in the liquid metal phase, as well as the amount of liquid metal, usually increases with temperature, more silicon will be found in the liquid metal phase at higher temperatures. This is the reason for the line at 2000°C in Fig. 3 being slightly lower than the line at 1900°C. At high temperatures, more gas will form, especially at low total gas pressures. This is probably the reason for the amount of liquid metal increasing with the total gas pressure at high temperatures, see Fig. 7. That is also the reason why less liquid metal phase is formed at low total gas pressures and 2000°C than at 1900°C.

3.3. Effect of Graphite Activity on Equilibrium Silicon Distribution

In previous calculations in this report the carbon activity has been assumed to be unity with reference to graphite. In reality this may not be the case. Therefore, the calculations of the reference case have been performed at various carbon activities. The percentage of silicon in the system that is found in the gas phase at calculated equilibrium conditions can be found in Fig. 9. Looking at reaction (2) it can be expected that higher carbon activity values will result in higher amounts of silicon in the gas. This effect can be noticed for the data, for instance at 1500°C. At this temperature most silicon is found in the slag phase. The amount of slag decreases with increasing carbon activity which increases the amount of silicon in gas, liquid metal and \( \text{Al}_2\text{O}_3\text{SiO}_2 \). At higher temperatures than 1500°C it can be seen that the calculated equilibrium amount of silicon in the gas decreases at high carbon activity levels. This is due to formation of solid SiC at high carbon activity levels and also because more silicon goes to the liquid metal phase at higher temperatures, as seen in previous section. Stable solid and liquid phases at different carbon activities at 1600°C are presented in Fig. 10. From the 1600°C line in Fig. 9 it can be seen that the amount of silicon in the gas phase increases with carbon activity at low carbon activity levels. From Fig. 10 it can be seen that thismostly is as a result of decreasing amounts of slag. Furthermore, it can be seen that the decreasing amounts of silicon in the gas phase at 1600°C and high carbon activities that can be observed in Fig. 9, is a result of the formation of SiC.

Figure 11 shows a plot valid for 1800°C, all other parameters are equal to Fig. 10 which is valid for 1700°C. SiC forms at equilibrium at higher levels of carbon activity, decreasing the amount of silicon in the gas.

In Fig. 12 it can be seen that several phase transformations occur between different carbon activities. Between the carbon activities 0.6 and 0.7, \( \text{Al}_2\text{O}_3\text{SiO}_2 \) disappears and AlN is formed. Between the carbon activities 0.77 and 0.78, liquid slag disappears and solid \( \text{Al}_2\text{O}_3\text{CaO}_2\text{SiO}_2 \) is
formers. At carbon activities close to unity, SiC forms, decreasing the equilibrium amount of silicon in the gas phase.

All the reasoning above is based on calculated thermodynamics. Kinetics have also to be considered in order to calculate what actually happens inside the blast furnace. This is, however, outside the scope of this publication.

4. Conclusions

A thermodynamic study of the effect of i) coke and coal powder qualities, ii) total gas pressure, and iii) graphite activity on the silicon distribution in a blast furnace was carried out. Calculations were made for different temperatures. The specific conclusions from the study are:

1. More silicon can dissolve in the liquid metal phase in the tuyere level than measured in hot metal at tapping.

2. According to thermodynamics, silicon can be transferred to hot metal through the gas phase (mainly as SiO and SiS gas).

3. The amount of silicon in the gas phase may be influenced by the total gas pressure, giving less silicon in the gas phase at high total gas pressures.

4. Much silicon can exist in the gas phase even at lower carbon activities (0.5 relative to graphite) at temperatures of 1600°C and above. At temperatures higher than 1600°C, higher carbon activity levels do not necessarily give higher equilibrium amounts of silicon in the gas.

5. It is thermodynamically possible to find high silicon content metal droplets inside the coke pieces around the raceway.

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