Slags in Production of Manganese Alloys

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Abstract: The paper analyses the equilibrium partitioning of manganese and silicon between slag and alloy during the

production of high carbon ferromanganese (HC FeMn) and silicomanganese using FACTSage software. The results of

this modeling are compared with industrial and experimental data and used to elucidate the nature of the reduction

processes.

In production of HC FeMn, molten slag co-exists with a monoxide phase (Ca,Mg,Mn)O in which MnO is the major

constituent. Analysis of industrial data shows that slag and monoxide are close to equilibrium, but manganese

slag-metal partitioning is far from equilibrium. It is evident that the rate of MnO reduction from the slag is slower than

the rate of MnO dissolution into the slag. A previous laboratory study confirmed that MnO reduction from HC FeMn

slag is slow and that equilibrium is not reached.

In the production of silicomanganese, the charge consists of manganese ore, ferromanganese slag, quartzite and

fluxes. Excavation of industrial furnaces has revealed the presence of quartzite in the reaction zone, so there are four

condensed phases in the reaction zone; molten slag, molten silicomanganese alloy, quartzite and coke. FACTSage

modeling showed that during the production of silicomanganese, manganese and silicon partitioning between the metal

and slag is close to equilibrium at 1600°C. The concentration of silica in silicomanganese slags is much lower than the

silica-saturation value. It can be concluded that the reduction of silica from these slags is faster than the dissolution of

quartzite into the slag.

**Keywords:** reduction, slag, equilibrium, ferromanganese, silicomanganese

1. Introduction

Manganese alloys are mainly produced in Submerged Arc Furnaces (SAF), in which metal oxides are reduced by

carbon from coke or char, while heat needed for the reduction and smelting reactions is supplied by electrical energy.

Smelting/reduction processes require high temperatures, 1400-1500°C for the production of high-carbon

ferromanganese (HC FeMn) and 1600-1650°C for the production of silicomanganese (SiMn). Olsen et al. [1] have

given a detailed description of the technology for the production of manganese alloys.

Charge materials for the production of ferromanganese include manganese ore, coke, char and fluxes. Sometimes iron

ore is added to achieve an appropriate Mn/Fe ratio in the ferromanganese. The charge for silicomanganese production

includes ferromanganese slag, formed during the production of ferromanganese, as well as manganese ore, coke, char,

fluxes, and quartzite. Manganese in the manganese ore exists in the Mn<sup>3+</sup> or Mn<sup>4+</sup> valency states in pyrolusite,

hausmanite, cryptomelane and other compounds. Iron oxides are reduced to metallic iron and higher manganese oxides

are pre-reduced to MnO in the solid state by CO in the furnace gas atmosphere. Pre-reduced manganese ore starts to melt at 1150-1250°C depending on the ore composition. Upon melting it includes two main phases; liquid slag (MnO-SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-CaO-MgO system) and a monoxide solid phase (Mn,Ca,Mg)O which contains MnO as the major component.

In ferromanganese smelting, the reduction mechanism involves MnO or (Mn,Ca,Mg)O dissolution into the molten slag followed by reduction of MnO from the slag by carbon from coke or char or by carbon dissolved in ferromanganese. The solubility of MnO in molten slag depends on temperature and slag composition, increasing with increasing temperature and decreasing slag basicity.

In silicomanganese production, MnO is first reduced by reaction (1), forming liquid Mn-Fe-C alloy with a high manganese concentration. Silica is then reduced by slag-metal reaction (2).

$$(MnO) + C = [Mn] + CO$$
 (1)

$$(SiO2) + [Mn] = [Si] + (MnO)$$
 (2)

Reaction (1) is a solid/liquid/gas reaction while in reaction (2) both slag and metal phases are molten and the reaction is fast.

In the coke bed of the ferroalloy furnace, there is a mixture of pre-reduced ore, slag, metal and coke. Thermodynamic equilibrium between the condensed phases in production HC FeMn, therefore, involves four phases; molten slag, solid monoxide (Mn,Ca,Mg)O, molten Mn-Fe-C alloy and carbon (coke). HC FeMn slag is typically tapped at temperatures above 1450°C. In the temperature range 1450-1500°C, slag at equilibrium with metal contains 20-35 wt% MnO, depending on the ore composition, while the MnO content in industrial slag is higher, 37-43 wt% [1].

Excavation of a silicomanganese furnace reveals lumps of quartz, so there are four condensed phases in the reaction zone; molten slag, molten silicomanganese alloy, quartzite and coke. Silicomanganese slag contains 6-12 wt% MnO and 38-44 wt%  $SiO_2$  [1]. Manganese and silicon slag-metal partitioning is close to equilibrium; however, these phases are not at equilibrium with quartzite.

The aim of this paper is to analyse equilibrium manganese and silicon partitioning during the production of high carbon FeMn and SiMn. Equilibrium phase composition and manganese and silicon partitioning were calculated using the commercial software FACTSage, Version 6.2 with the database for manganese alloys developed by Tang *et al.* [2]. Comparison with industrial data and results of a laboratory study will be helpful to understanding of reduction reactions in the production of ferroalloys.

### 2. Slag in Production of High Carbon Ferromanganese

Manganese ores of different origin have different chemical composition, mineralogy and morphology. Sorensen *et al.* [3] studied the phase compositions of Wessels (South Africa), Groote Eylandt (North Australia), CVRD (Brazil) and Gabonese manganese ores and how they changed during the process of calcination in oxidising and reducing atmospheres. All ores exhibited a common property; upon heating in CO or H<sub>2</sub> to 800-1200°C they formed a solid

monoxide phase, whose main component was MnO. Wessels ore contains relatively high amounts of iron and calcium, but is low in silicon and alkalis. Groote Eylandt (GE) ore is a high-grade ore with a manganese content of 55 wt% (predominantly pyrolusite). Apart from pyrolusite, GE ore also contains iron silicate and silica. Both CVRD and Gabonese ores contain a high concentration of alumina.

In the production of manganese alloys, a mixture of different ores is charged into the ferroalloy furnace. In this paper, the reduction behaviour of GE and Wessels ores, and 50:50 mixture of both will be examined. Industrial slag, whose chemical composition was reported in [1], will also be studied.

FACTSage was used to calculate carbothermal reduction in the presence of an excess of carbon in a pure CO atmosphere. The chemical composition of slag formed during the reduction process strongly depends on the ore. This is illustrated by calculation of the compositions of slag at 1400 °C, 1450 °C and 1500 °C presented in Table 1, which also includes the chemical compositions of the ores before reduction. The chemical compositions of metal are given in Table 2.

Table 1. Equilibrium chemical composition (wt pct) of liquid slag during the reduction of Groote Eylandt ore, Wessels ore and their mixture, calculated using FACTSage.

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	Temperature, °C	MnO	FeO	SiO <sub>2</sub>	$Al_2O_3$	CaO	MgO	K <sub>2</sub> O
Groote Eylandt ore	Before reduction	81.9	5.80	6.09	3.90	0.18	<0.1	1.53
	1400	49.9	<0.01	26.6	19.4	1.07	0.60	2.39
	1450	42.5	<0.01	26.8	26.9	1.74	0.96	1.02
	1500	32.3	<0.01	26.7	34.4	3.70	2.03	0.91
Wessels ore	Before reduction	70.0	14.4	4.18	1.51	8.34	0.73	-
	1400	12.2	<0.01	18.6	20.6	44.8	3.75	-
	1450	7.64	<0.01	19.6	20.8	46.8	5.20	-
	1500	6.27	<0.01	20.8	19.8	49.0	6.28	-
Groote Eylandt &	Before reduction	76.0	10.1	5.14	2.70	4.26	0.41	0.77
Wessels ores mixture	1400	27.6	<0.01	29.2	15.4	24.4	2.28	1.15
(50:50)	1450	20.7	<0.01	31.4	17.2	27.2	2.54	0.93
	1500	14.1	< 0.001	31.8	19.6	30.9	2.87	0.57

Table 2. Equilibrium chemical composition (wt pct) of liquid metal during the reduction of different manganese ores, calculated using FACTSage.

Mn source	Temperature, °C	Mn	Fe	Mn/Fe	С	Si
Groote Eylandt ore	1400	85.8	6.86	12.5	7.27	0.08
	1450	85.7	6.54	13.1	7.26	0.47
	1500	85.0	6.31	13.5	6.92	1.74
	1400	76.7	16.3	4.7	7.03	< 0.001
Wessels ore	1450	76.6	16.2	4.7	7.14	<0.01
	1500	76.5	16.3	4.7	7.26	<0.01
Groote Eylandt – Wessels ores	1400	81.2	11.7	6.9	7.14	0.03
mixture (50:50)	1450	81.1	11.5	7.1	7.23	0.15
	1500	80.9	11.4	7.1	7.22	0.50

Phases formed during the reduction of Groote Eylandt ore at  $1400-1500^{\circ}$ C in addition to slag, metal and graphite included a small amount of leucite  $KAlSi_2O_6$  while at  $1500^{\circ}$ C equilibrium phases also included corundum. Reduction of Wessels ore featured the formation of dicalcium silicate and monoxide, whose main component was CaO. Only liquid metal and slag were observed during the reduction of the Groote Eylandt-Wessels ore mixture.

The results in Table 1 show a strong effect of the slag composition on the equilibrium MnO content of the slag. The activity of manganese in the ferromanganese and the activity of MnO in the slags at equilibrium are presented in Table 3,

as calculated with FACTSage. Changes with temperature in the activities of MnO in slags at equilibrium are defined by the change in the equilibrium constant for the reduction reaction (1) which is given below

$$MnO(1) + C(gr) = Mn(1) + CO(g)$$
  $K = 2.08 \text{ at } 1400^{\circ}\text{C}$   
= 3.38 at 1450°C  
= 5.35 at 1500°C

Table 3. Manganese activity in ferromanganese and MnO activity in slag (both referred to the liquid state) at equilibrium during reduction.

	Groote Eylandt ore		Wessels ore			Groote Eylandt – Wessels ore mixture			
Temperature, °C	1400	1450	1500	1400	1450	1500	1400	1450	1500
$a_{Mn(l)}$ in alloy	0.412	0.403	0.389	0.365	0.359	0.352	0.388	0.381	0.373
a <sub>MnO(I)</sub> in slag	0.189	0.113	0.068	0.169	0.102	0.063	0.178	0.107	0.066

The differences in MnO activities in slags formed during the reduction of different ores at equilibrium can be attributed to the variations in manganese activity in the metal phases as a result of the differing Mn/Fe ratios in the resulting ferroalloys (Table 2). The Mn/Fe ratio and manganese activity in ferromanganese are the highest during the reduction of Groote-Eylandt ore and the lowest during the reduction of Wessels ore.

At the same temperature, the MnO activity and its concentration in the slag were also the highest for the reduction of Groote Eylandt ore and the lowest for the reduction of Wessels ore. However, the differences in MnO activities were much smaller than the differences in MnO concentrations. The MnO activity in the slag at equilibrium depends only on temperature and metal composition at a constant partial pressure of CO(g), while the MnO activity coefficient also depends on the slag composition. The slag basicity, defined as;

$$B = (\%CaO + \%MgO)/(\%SiO_2 + \%Al_2O_3)$$

was the lowest for the Groote Eylandt ore and the highest for the Wessels ore. The MnO activity coefficient increases with increasing slag basicity [1]. Low slag basicity during the reduction of Groote Eylandt ore explains the low MnO activity coefficient and high MnO concentration in the slag. The MnO activity coefficient in slag formed during reduction of Wessels ore is high and the MnO concentration is consequently low. The smelting of Wessels ore is very similar to the "fluxed process" in which fluxes are added to the charge to increase the slag basicity and decrease the MnO content in the slag [1]. The slag formed in smelting of Wessels ore is in equilibrium with a monoxide phase (Ca, Mg, Mn)O. However the MnO content in the monoxide phase and its thermodynamic activity are low.

In the production of manganese alloys, a mixture of different ores is charged into the ferroalloy furnace and a typical slag is reported [1] to have the following composition:

This "typical" slag, as calculated using FACTSage at 1400-1500°C, contains two phases; liquid slag and monoxide (Ca,Mg,Mn)O containing 80-81 wt% MnO. The typical industrial slag is therefore in equilibrium with the monoxide phase. The MnO activity in this typical slag, from FACTSage, is 0.329 at 1400°C, 0.366 at 1450°C and 0.403 at 1500°C. These activities are all well above the values for MnO in the three slags presented in Table 3 which are in equilibrium with ferromanganese alloy. Equilibrium calculations show that when this "typical" slag is reduced further by reaction

with carbon, the MnO concentration decreases from 40.3 wt% to 26.2 wt% at 1400°C, 19.8% at 1450°C and 9.60 wt% at 1500°C. It is clear that the MnO concentration in the "typical" slag is above the MnO concentration that would be in a slag which was in equilibrium with ferromanganese.

It can be concluded from this thermodynamic analysis that the rate of dissolution of MnO from the ore into the slag is faster than the rate of MnO reduction from the slag.

## 3. Carbothermal Reduction of MnO from Ferromanganese Slag

Analysis of MnO reduction from slag in the industrial production of high carbon ferromanganese demonstrated that manganese slag-metal partitioning is far from equilibrium. This section reviews laboratory studies of MnO reduction from slag. Sun *et al.* [4] and Safarian *et al.* [5] examined the reaction between ferromanganese slag and substrates manufactured from different carbonaceous materials, while Yastreboff *et al.*[6] studied the carbon-slag reaction in an intimate mixture of graphite or char with ferromanganese slag.

The degree of reduction of MnO from synthetic or industrial slag by a graphite or coke substrate at  $1450^{\circ}$ C was quite minor even though the activity of MnO in both synthetic and industrial slags calculated using FACTSage were above the equilibrium value of  $a_{\text{MnO}} = 0.10$ -0.11 (see Table 3). Even at  $1550^{\circ}$ C the reduction of MnO from synthetic slag by a substrate made from graphite was very slow [4] but increased significantly when graphite was replaced by coke. A strong effect of a type of substrate on the rate of MnO reduction from ferromanganese slag was also observed in [5].

**Figure 1** shows a cross-sectional view of the slag-substrate interface after the reduction of industrial slag by graphite (from [4]). EDS analysis of the slag and metal drop formed on this interface is shown in **Figure 2**. EDS counts and atomic ratios of concentrations of the major components of the slag to the calcium concentration in the slag bulk phase, slag-metal interface and metal droplet are given in **Table 4**. The composition of slag very close to the interface is given by point 1. Point 2 is even closer to the interface but the EDS counts at this point are unreliable because they are affected by the proximity of the metal phase.

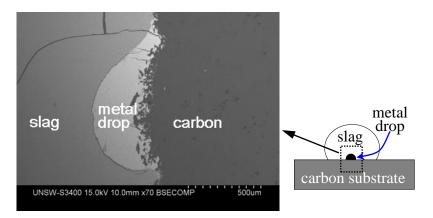
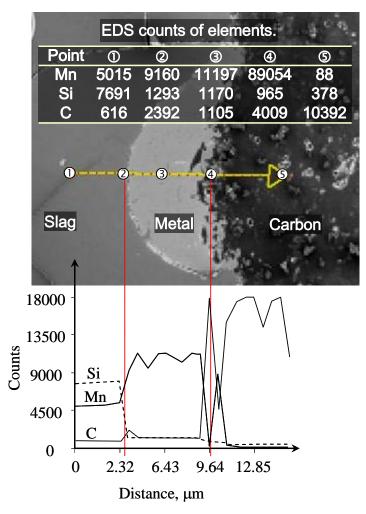


Figure 1. Cross-sectional view of the slag-carbon interface after the reaction of industrial slag with a graphite substrate [4].

Calcium, aluminium and magnesium oxides were not reduced under the given experimental conditions and are

uniformly distributed in the slag phase. The concentration of manganese in the bulk slag phase is significantly higher than at the interface, which indicates that MnO mass transfer in liquid slag can contribute to controlling the reduction rate.



**Figure 2**. EDS of slag-graphite interface in reduction of industrial slag [4].

Table 4. EDS counts and atomic ratios of elements to calcium in the vicinity of interface (point 1) and in the slag bulk phase in reduction of industrial slag by graphite

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Element		Ca	Mn	Si	Al	Mg		
EDS count	s at point 1	3985	5015	7691	3942	918		
Atomic ratio	Bulk	1	2.67	1.82	0.99	0.24		
	Interface	1	1.26	1.93	0.99	0.23		

Carbothermal reduction of MnO from the slag into the metal phase in argon, which was studied in [4], has no thermodynamic constraint as the CO partial pressure in the inert gas can be very low to provide a thermodynamic driving force for the reaction to proceed. However, this condition only applies where the three phases (slag, carbon and gas) are present i.e. along the slag-substrate perimeter. The reduction reaction actually occurs on the slag-substrate interface where the CO pressure should be above 1 atm for a bubble to nucleate.

The slow reduction of MnO from slag by the substrate can be explained by a relatively small slag-substrate interfacial area. Wetting of carbonaceous materials by ferromanganese slag is quite poor [4], therefore a rough surface of coke and

other carbonaceous materials does not secure a high interfacial reaction area in the industrial production of manganese alloys.

MnO reduction from ferromanganese slag was much faster when ferromanganese slag was intimately mixed with graphite [6]. In these experiments, the mixture of solid graphite and liquid slag had a high interfacial area and was well-exposed to the gas phase. As a result, MnO reduction at 1400°C in CO was close to completion within 2 hours.

Kinetic analysis of laboratory and industrial data on MnO reduction in [7] demonstrated that the chemical reaction can be the rate limiting stage. The MnO concentration profile in the slag in Figure 4 shows that MnO mass transfer in the slag can also contribute to the rate control. This analysis leads to the conclusion that the rate of MnO reduction from slag in the production of HC FeMn is mix-controlled by the interfacial carbon-slag reaction and the mass transfer of MnO. The slag-coke interfacial area is not expected to be large because coke is in the form of large lumps in the reaction zone and the coke is poorly wetted by the slag.

# 4. Slag in Production of Silicomanganese

The charge for the production of silicomanganese consists of manganese ore, ferromanganese slag, quartzite and dolomite or calcite fluxes. A typical charge comprises, per tonne of silicomanganese [1]; 1029 kg MnO, 105 kg FeO, 337 kg SiO<sub>2</sub>, 191 kg Al<sub>2</sub>O<sub>3</sub>, 248 kg CaO, 108 kg MgO and 79 kg of Si sculls. The product of smelting is a silicomanganese alloy containing 67 to 70 wt% manganese, 17 to 20 wt% silicon and 1.5 to 2 wt% carbon. The process temperature for the production of silicomanganese is 1600-1650°C. The temperature within the coke bed in a silicomanganese furnace in which the reduction of MnO and SiO<sub>2</sub> occurs is not known with certainty. In study [8], the coke bed temperature was found to be between 1566°C and 1607°C. It was also found that this temperature was dependent on the manganese source used for silicomanganese production. At these high temperatures, both manganese and silicon oxides are reduced. Reduction of silica proceeds via dissolution of quartz into the slag and reduction of silica from the slag. The slag produced from smelting contains [1];

6-12 wt% MnO, 38-44 wt% SiO<sub>2</sub>, 10-25 wt% Al<sub>2</sub>O<sub>3</sub>, 20-35 wt% CaO and 5-15 wt % MgO.

FACTSage was used to calculate slag-metal equilibrium in which the "typical" charge was reacted with excess graphite in a CO atmosphere. The chemical compositions of silicomanganese slag and metal formed at equilibrium by carbothermal reduction of the "typical" charge at 1600°C and 1650°C are given in **Tables 5** and **6** respectively. These tables also present the chemical compositions of slag and metal produced from such charge materials in an industrial 27 MW submerged arc furnace [1].

Table 5. Chemical composition of charge and silicomanganese slag for an industrial 27 MW submerged arc furnace [1] and at equilibrium calculated using FACTSage

Condition			Chemical comp	osition, wt%		
	MnO	FeO	SiO <sub>2</sub>	$Al_2O_3$	CaO	MgO
Initial charge	39.2	4.00	35.9	7.29	9.45	4.12
27 MW furnace [1]	8.1	-	43.3	15.0	20.2	8.6
1600 °C (equilibrium)	6.38	-	42.8	17.8	23.1	9.89
1650°C (equilibrium)	2.89	-	37.2	21.4	27.7	10.8

All charge materials are presented in the form of oxides (charge mixture is taken from [1], Table 4.4).

The chemical composition of the slag in the production of silicomanganese in a 27 MW submerged arc furnace is close to the chemical composition of the slag at equilibrium with metal and carbon (graphite) in the carbothermal reduction of a "typical" charge at 1600°C. However, the equilibrium MnO content in the slag during reduction at 1650°C is significantly lower than in the industrial slag.

Table 6. The chemical composition of silicomanganese from a 27 MW submerged arc furnace [1] and at equilibrium with slag and graphite calculated using FACTSage

Condition	Chemical composition, wt%					
	Mn	Fe	Si	С		
27 MW furnace [1]	70.5	8.1	19.2	1.5		
1600 °C (equilibrium) <sup>1)</sup>	72.9	8.63	15.7	2.78		
1650 °C (equilibrium) 1)	72.4	8.65	16.1	2.94		

<sup>&</sup>lt;sup>1)</sup>Reduction products included graphite and SiC.

In smelting of silicomanganese, high temperatures (in excess of 1600°C) facilitate fast reduction of MnO from the slag, while silica is reduced by reaction (2) between molten metal and slag:

$$(SiO_2) + [Mn] = [Si] + (MnO)$$

The reaction between liquid slag and liquid metal in the production of silicomanganese is relatively fast and approaches equilibrium.

The thermodynamic activities of MnO and  $SiO_2$  in the industrial (27 MW furnace) slag and at equilibrium at  $1600^{\circ}$ C, as calculated by FACTSage, are given in **Table 7** and are also not very different.

Table 7. MnO and SiO<sub>2</sub> activities in industrial slag in 27 MW submerged arc furnace and at equilibrium in the reduction process at 1600°C and 1650°C (from FACTSage)

Slag	27 MW furnace,	Equilibrium, 1600°C	27 MW furnace,	Equilibrium, 1650°C
	1600°C		1650°C	
MnO(I) activity	0.0216	0.0173	0.0243	0.0110
SiO <sub>2</sub> (I) activity	0.219	0.150	0.215	0.0545

As it is mentioned above, excavation of a silicomanganese furnace revealed the presence of quartzite in the reaction zone, so at thermodynamic equilibrium in the reaction zone the condensed phases are; liquid slag, liquid metal, coke (char) and quartzite. However, the data in Table 7 show that liquid slag is far from equilibrium with quartzite. The activity of silica in silica-saturated slag for the standard state of pure liquid silica is 1.04 at 1600°C and 1.02 at 1650°C (the standard state for solid silica is cristobalite with melting temperature 1723°C).

It can be concluded that the slag and silicomanganese alloy are close to equilibrium, but that equilibrium with quartzite is not reached. It can also be concluded that the rate of dissolution of quartzite into the slag is much slower than the rate of silica reduction from the slag.

# 5. Conclusions

Condensed phases in the reaction zone during the production of high carbon ferromanganese include coke, molten slag, metal, and monoxide (Ca,Mg,Mn)O. The reaction zone in the production of silicomanganese consists of coke, molten slag, metal and quartzite. The following conclusions can be drawn from thermodynamic analysis of

slag-metal-carbon equilibrium in the production of manganese alloys.

The thermodynamic activity of MnO in slag in equilibrium with liquid metal and carbon in the production of high carbon ferromanganese from different manganese ores is in a relatively narrow range (Table 3). Differences in the thermodynamic activity of MnO in slag are due to differing compositions of manganese alloys (manganese to iron ratio). However, the MnO activity coefficient and MnO content in the slag formed in processing of different ores show significant differences (Table 1) which depend on the slag basicity. Thus at 1450°C the MnO concentration in the slag formed during smelting reduction of Groote Eylandt ore is 42.5 wt% while slag formed in the processing of Wessels ore contains 7.6 wt% MnO. Typical industrial slag (from [1]) contains monoxide (Ca,Mg,Mn)O phase. MnO activity in the industrial slag is above its equilibrium value. Therefore, in the production of HC FeMn, slag-metal-carbon equilibrium is not reached while slag is close to equilibrium with the monoxide phase. The rate of MnO reduction from the slag is mix-controlled by diffusion in the slag phase and the rate of the chemical reaction.

In the production of silicomanganese, quartzite is dissolved into the slag and silica is reduced from the slag into the liquid metal alloy. The chemical composition of slag in the production of silicomanganese in an industrial submerged arc furnace is close to the chemical composition of the slag at equilibrium with metal and carbon at 1600°C. However, the thermodynamic activity of silica in the slag is below the value expected for slag in equilibrium with quartzite. Therefore, liquid slag formed in production of silicomanganese is far from equilibrium with quartzite. This implies that the rate of quartzite dissolution into the slag can be a rate controlling step in the production of silicomanganese.

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