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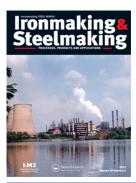
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RESEARCH ARTICLE

Silicon in hot metal from a blast furnace, the role of FeO

J. L. T. Hage^a, J. van der Stel^a and Y. Yang ^b

^aDepartment of Ironmaking and Steelmaking, Tata Steel Research and Development, IJmuiden, Netherlands; ^bDepartment of Ironmaking and Steelmaking, Delft University of Technology, Delft, Netherlands

ABSTRACT

Silicon [Si] in hot metal is an impurity acting in the steel shop as an energy source that is released by oxidation during oxygen blowing in the converter. Preferred silicon concentration in hot steel is typically 0.4 wt-% (± 0.1 wt-%), helping predictable and low-cost processing. In practice, the Si concentration is difficult to control and may occasionally reach levels higher than 1 wt-%. The authors studied data from observations, samples and autopsies of a chilled blast furnace, a core drill and a furnace in operation. In addition, production data from a melting reduction pilot plant (Hlsarna) and FactSage® calculations were used. The amount of FeO in the raceway, the area in which hot gas and powdered coal (PCI) are introduced in the blast furnace, in relation to [Si] in hot metal, is observed. The goal of this paper is to contribute to a better understanding about the mechanism of the dissolution of silicon in hot metal.

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Blast furnace; Hot metal quality; Silicon; Raceway; Process control; SiO; FeO; Sicontrol

Introduction

The challenge: keeping [Si] constant at low concentrations

The heating processes of products melting in the raceway and in the liquid bath of the hearth and the effect of pulverized coal injection (PCI) on heat exchange in the oxidizing zone and its extension are described by Formoso et al. [1]. Adequate amounts of silicon dissolved in hot metal are important for steelmaking as it releases energy when oxidised in the oxygen furnace. Too much silicon generates excessive heat and additional slag, causing additional wear of the refractory. The optimal Si-concentration is around 0.4 wt-%. In practice, it is difficult to achieve a constant level (Figure 1), which seems related to the thermal state of the furnace. When [Si] drops too much, additional carbon is charged and vice versa. The response time for coke is around 5 h, while adjusting the PCI is faster. A rapid change from 0.2 wt-% Si to 0.7 wt-% within an 8-h shift is not unusual and more extreme values can occur, which cannot be explained by just the thermal state.

During the 6th European Coke and Ironmaking Conference ECIC in Dusseldorf (July 1st 2011), the concept for a 2D-model for silicon prediction was presented by Hage et al. [2]. The approach is based on calculating fluxes of iron and silicon in the blast furnace and it proved to be useful in explaining silicon variations. To understand, predict and control the silicon level in hot metal, it is important to understand the silicon transport phenomena in the furnace. Since the 1970s, gasification of silica (SiO₂) to SiO downstream of the tuyeres has been suggested as an important intermediate process [3]. The SiO gas is transported upwards by the hot blast and interacts with hot metal and subsequently dissolves as elemental silicon. In the dripping zone, contact between hot metal and slag is intimate, and thus silicon can interact with FeO in the slag phase and can be re-oxidised to silica.

The commonly used theory

Kinetics are required for understanding the dissolution reactions of silicon. Steiler and others [4–6] described a theoretical kinetic model, in which SiO2 is gasified in the raceway to SiO gas, which subsequently dissolves in the metal near the cohesive zone. In the lower part of the dripping zone and in the raceway, where oxygen is available, re-oxidation of [Si] takes place. Figure 2 shows the silicon reactions below the cohesive zone. It is interesting to lower the SiO₂ input with alternative injection materials like biomass [7]. Models were developed based on this concept, simplifying the blast furnace model into sub-models, like the model of temperature against silicon content [8–12].

Dissolving silicon in hot metal is a combination of phenomena controlled by several mechanisms, which can be interpreted differently:

- SiO gas production in the raceway
- Transport of SiO gas
- Dissolution of SiO gas in hot metal
- · Mixing of hot metal in the hearth of the furnace

Geerdes et al. [13] described the blast furnace in a popular scientific format: 'Modern Blast Furnace Ironmaking'. The theory from Steiler [4–6] is further adapted by Geerdes:

- Formation of SiO in the raceway. The first reduction step takes place at the very high flame temperatures in the raceway (>2000°C) (SiO₂ + CO → SiO + CO₂) (Equation (1)). The silica originates from the ash of the coke and coal (gangue).
- Further reduction by means of direct reduction with carbon in iron. The SiO gas in contact with the liquid iron can be reduced as follows: SiO+[C] → [Si] + CO (Equation (2)).

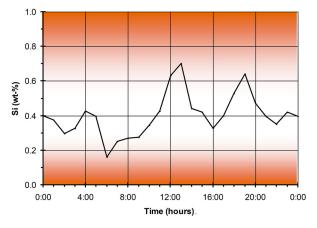


Figure 1. Possible Si-variations in hot metal within 24 h in a blast furnace.

- 3. The more intimate the contact between iron and gas, the higher the hot metal silicon content. The higher the height of the dripping zone, the more contact time between hot gasses and the liquid metal, leading to higher hot metal temperatures. Longer contact allows more SiO gas to react with the carbon in the hot metal, leading to higher hot metal silicon content. Therefore, a high-located melting zone corresponds with high hot metal temperature and high hot metal silicon.
- 4. The hot metal silicon is in equilibrium with the slag. Important aspects are:
 - a. When iron droplets descend and pass through the slag layer, the silicon can be re-oxidised if FeO is present in the slag, according to: $[Si] + 2 FeO \rightarrow$ SiO2 + 2 Fe (Equation (3)).
 - b. Low ${\rm SiO_2}$ values in slag lead to low Si in hot metal.
 - c. The hot metal formed in the centre of the furnace has a high silicon content, while the hot metal formed near the wall has lower hot metal silicon content. The cast result is an average value.

The question addressed in this paper is whether these theories need updating. Besides measurements described in the

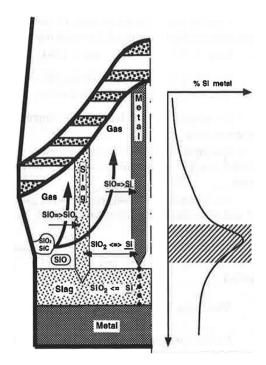


Figure 2. Reactions described by Steiler [6].

literature, this paper includes recent data from an alternative ironmaking process: HIsarna® [14,15].

Method: analyses of data from literature and calculations

Three sampling trials in existing literature describe the interaction between FeO and silicon. Here are the described data and observations:

- 1. Data from an excavation of a frozen experiment (by cold gas injection) in the so-called experimental blast furnace (EBF), located in Lulea (Sweden) with 1.5-m diameter;
- 2. Core drill through a tuyere from an off-wind industrial-scale blast furnace from Ruukki (Finland) with 8-m diameter;
- 3. Sampling an industrial scale blast furnace in operation from Kawasaki Steel (Japan) with 11.1-m diameter.

Data from hot metal from Hisarna, a melting-reduction process, confirmed the SiO gas theory only applies at higher temperatures. The HIsarna pilot plant is in IJmuiden (The Netherlands).

Finally, FactSage® calculations helped identify the main source for SiO gas. A comparison is made of 'free' SiO₂ (quartz) present in both PCI and coke, and SiO₂ present in the slag phase from sinters or pellets.

A 2D figure based on these data shows the mechanisms relevant for the dissolution of Si in hot metal.

Describing the results

Data from a small frozen furnace (1.5 m diameter) [16]

A small blast furnace was quenched after completing a campaign. Hot metal was sampled in the area around the tuyeres and analysed for its silicon content. Samples were taken from the wall area, the middle, and the centre. The raceway effect in a small furnace is much larger than in full-scale furnaces. The results published by Omori (1982) are presented in Figure 3 [17].

The residence time in this small furnace is about 4 h and the height is about 10 m. Based on these assumptions, the average speed for solid material is about 25 min/m, but for liquid material, it will be shorter. The silicon concentration rises from about 1.5 wt-% above the tuyere with a maximum of 7 wt-% in the middle of the furnace. A 0.7 m further down, the dissolved Si was subsequently re-oxidised to become part of slag. The overall concentration was 1.2 wt-% Si, which is shown in the graph.

Data from a full-scale blast furnace during a stop (8 m diameter) [18]

Core drilling through the tuyeres can be done during a furnace stop (off wind situation). It delivers materials at tuyere level from the raceway towards the centre of the furnace. The hot metal found in these cores can be analysed for its chemical composition. Figure 4 shows an example of silicon and FeO data from an 8-m diameter furnace from Ruukki [18]. This work allowed studying coke and hot metal properties in this area. The peak matches results from

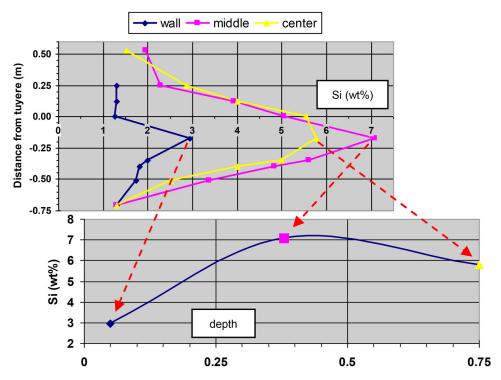


Figure 3. Vertical and lateral variations of silicon in an experimental blast furnace [16]. On the first graph, the height is plotted on the Y-axis, the Si content on the X-axis. On the second graph, the Si content is plotted on the y-axis and on the x-axis the depth in the furnace. Samples were taken 20 cm below the tuyeres.

Omori: dissolving silicon is fast and the highest concentrations are found in or near to the raceway.

The high amount of FeO in the first metre from the wall seems typical for the current burdening methods of the blast furnace. Silicon reaches its highest level in the first metre from the wall as well. Near the middle of the furnace, the silicon concentration is very low. The amount of FeO appears only enough to oxidise part of the silicon. However, this ratio may change rapidly since FeO is reduced by carbon and CO-gas as well. The reported concentration of silicon in the hot metal was 0.4 wt-%. FeO in slag is in the order of 0.1 wt-%.

The data from the small furnace and the large furnace illustrate various phenomena:

- Silicon concentrations can locally reach high levels.
- Kinetics for dissolving SiO is fast (within fractions of a second); otherwise, the relatively high concentrations from 50 cm till around 1 m cannot occur.
- The highest value of silicon is measured at the end of the raceway.
- Kinetics for reoxidation of Si are relatively fast as well but depend on the slag/hot metal interactions.

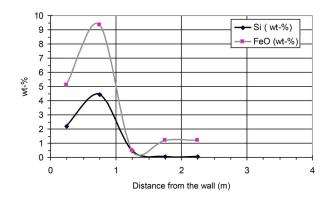


Figure 4. Hot metal analyses taken by a core drill through the raceway [18].

Data from a full-scale blast furnace in operation (11.1 m diameter) [19]

Measured higher silicon values were already reported by Kawasaki Steel in 1988 by in situ sampling in the raceway. The study is based on a probe that penetrates sideways through a tuyere (Figure 5). The probe was used during operation of the furnace.

The measured silicon in hot metal data (Figure 6) confirmed that in the raceway, the silicon reaches levels above 1.0 wt-%, as was expected. Between the tuyeres, the hot metal contains relatively low silicon values of around 0.2 wt-%. It shows that despite the temperature of around 1500°C, the dissolved amount of silicon is limited. FeO data were not reported in the paper.

Data from a smelting-reduction plant: the HIsarna®process [20]

Data from the Hisarna® process proves the mechanisms described in the previous paragraph. The HIsarna® process is a development of the ULCOS (Ultra Low CO₂ Steelmaking) [21] project in cooperation with HIsmelt®. Ore agglomeration

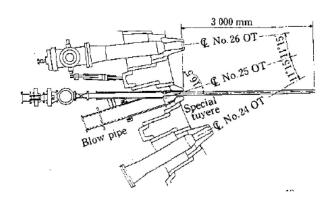


Figure 5. The sideway probe from Takeda et al. [19].

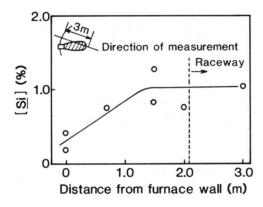


Figure 6. Silicon data from Takeda's sideway probe [19].

and coke making are completely avoided. Characteristics of this one-step process are no tuyeres and no raceway and temperatures of around 1500°C. If the hypothesis of the SiO gas formation is correct, the Si content in the hot metal will be very low.

The HIsarna® process is characterized by intensive mixing of slag containing relatively high amounts of FeO (typically 5 wt-%), droplets of Fe and traditional slag components like SiO₂, MgO and CaO. The only route for silica to dissolve directly in the hot metal is via thermodynamic equilibrium. Any silicon present in the hot metal will be re-oxidised in the slag phase. Once the hot metal is tapped from the process, the concentration of silicon values will be low. Table 1 shows the typical hot metal and slag analyses from a blast furnace and Table 2 shows the typical hot metal and slag analyses from HIsarna® pilot plant. As expected, the silicon level from HIsarna® is very low.

Thermodynamic calculations: SiO gas generation

The SiO gas production was calculated with FactSage® (Figure 7), the input data are shown in Table 3. The results are presented in arbitrary units and should be used as relative values. SiO gas can originate from two phases: pure SiO₂ (quartz) in coal or coke and from slag in pellets or sinters. This figure shows that SiO gas originating from quartz is much more effective than from slag. The polymer structure of slag lowers the so-called activity of silica significantly, which was also concluded by Gustavsson et al. [16]. The

Table 1. Typical hot metal and slag compositions from a blast furnace [13].

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BF hot metal	Typical (wt-%)	BF slag	Typical (wt-%)
Fe	94.5	CaO	40
C	4.5	MgO	10
Si	0.40	SiO ₂	36
Mn	0.30	Al_2O_3	10
S	0.03		
P	0.07	FeO	0.1

Table 2. Typical hot metal composition from Hlsarna® [20].

Typical (wt%)	HIsarna Slag	Typical (wt%)
94.5	CaO	38
4.0	MgO	9
0.006	SiO ₂	35
0.026	Al_2O_3	9
0.05		
0.04	FeO	2
	94.5 4.0 0.006 0.026 0.05	94.5 CaO 4.0 MgO 0.006 SiO ₂ 0.026 Al ₂ O ₃ 0.05

presence of CaO in a SiO₂ containing slag decreases the activity of SiO₂ even further.

Also shown in Figure 7 is that at lower theoretical flame temperatures, less SiO gas is produced. Consequently, less silicon is dissolved, resulting in lower silicon values in hot metal. This is confirmed by Niwa et al. [22]. Fukuyama No. 5 blast furnace operated with a monthly [Si] average of 0.1%, by lowering the flame temperature and maintaining a stable operation. Controlling the input of Si in the furnace and lowering the flame temperature lowers the [Si] in hot metal.

SiO gas can be suppressed by the oxygen partial pressure [23,24]. A higher oxygen partial pressure will result in higher FeO levels. SiO is subsequently dissolved in hot metal.

Construction of a 2D figure

Figure 8 shows the most important reactions in the blast furnace. Above the cohesive zone, only gas reduction occurs. Below the cohesive zone, the final reduction of the liquid FeO can be either gas reduction or direct reduction with coke; below the tuyere level, it is only direct reduction. SiO gas is generated in the raceway. Adding more PCI results in less FeO and, as a result, silicon increases in the hot metal.

Figure 9 shows the concept of the mechanism. In the first layer, several processes are happening simultaneously. Hot metal is produced first, more so in the raceway area than in the middle. SiO gas is formed in the tuyere and subsequently dissolves. The iron is not fully reduced, especially in the raceway. Directly below the tuyere, silicon reacts with FeO. The interaction is enhanced by coke in the so-called dripping zone. Hot metal and slag are separated, and slag is floating on top of the iron. The Si content in the dripping iron is inhomogeneous, with higher concentrations near the wall and lower concentrations in the middle. Finally, the hot metal is tapped, which causes mixing and determines the final Si concentration in hot metal. Especially this tapping practice seems to result in the Si fluctuations.

The model is based on the calculation of fluxes. Possibly this approach can be incorporated in existing blast furnace models, like Mogador [25]. The flux of melted iron and iron oxides is based on burdening of the furnace. The Si and FeO fluxes are based on silica input and gasification characteristics around the tuyeres. Reoxidation occurs in the dripping zone, below the tuyeres. In the slag layer, the liquid iron is separated from the slag. In the hearth below is the transportation of the hot metal to the taphole.

The hot metal flow with relatively high silicon content near the wall will mix with the hot metal flow with low silicon content from the middle due to transport. Coupling this flux model with a flow model of the hearth could possibly be used to explain or even predict silica variations. A 2D model for hearth flow is developed by Post et al. [26,27] (Figure 10).

Discussions

Discussion of the FeO mechanism

The hot metal analyses from the core drill [18] from the Ruukki furnace show that SiO gas interacts with liquid close to the raceway area. The probe measurements by Takema

SiO₂ gasification - result of FactSage calculations

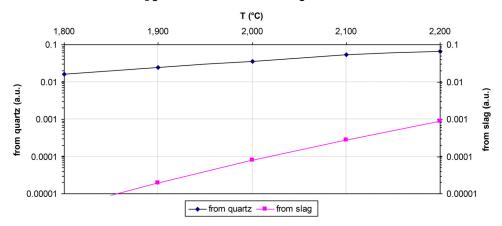


Figure 7. These results are based on the input file from Table 3. SiO gas was originated from either pure SiO₂ (quartz) or from the slag phase. The temperature is the flame temperature in the raceway (RAFT), the a.u. are based on mass (g).

Table 3. Input used for FactSage®.

Species	Mass (g) SiO from slag	Mass (g) SiO from pure SiO ₂
Al_2O_3	13	0
SiO ₂	20 (= 0.3 mol)	20
MgO	10	0
CaO	50	0
FeO	7	0
Fe	10	10
C	1	1
N_2	1	1
CO	100	100
CO ₂	0	0

[19] show that between the tuyeres, the silicon concentration is significantly lower at the end of the raceway. [Si] concentrations may reach levels of 7 wt-% and seem not to be limited by any slag composition. FeO also reaches its highest level at the end of the raceway. The hypothesis is that FeO is the only significant oxygen source below the tuyeres and as such able to re-oxidise silicon. Increasing the amount of FeO will decrease the amount of silicon in hot metal.

FeO and SiO will react forming iron (Fe) and silica (SiO₂). Below the raceway, the amount of CO/CO₂ gas is limiting, so the only oxygen source available to re-oxidise [Si] is FeO. In the dripping zone below the raceway, the slag and hot metal drip over the coke bed, enhancing the interaction between both phases.

A calculation is made using the data presented in Figure 4, assuming that all FeO reacts with silicon (Table 4). The silicon concentration is still high (from 4.3 to 2.6 wt-%). The measured concentration at the tap is 0.41 wt-%, so mixing and dilution with hot metal fluxes with lower silicon content take place in the blast furnace hearth. Increasing the FeO content at the end of the raceway will lower silicon concentration. Methods like ore injection (sinter fines) will increase the level of FeO at the end of the tuvere; the silicon content was reported to drop from 0.57 to 0.38 wt-% [20]. It is recommended to investigate other methods to control FeO in the blast furnace. In the HIsarna® plant FeOrich slag is continuously mixed with liquid iron.

Discussion of the commonly used theory

The data show that most SiO gas is captured in the raceway. For this reason, the effect of a higher dripping zone causing a higher concentration of Si is not likely. A higher dripping zone is the result of more energy input by increasing the hot blast, coke or PCI, reflecting the so-called thermal state of the furnace. This results in more SiO₂ input via these carbon sources. More SiO gas will be produced and most likely less FeO, resulting in higher Si concentrations in the hot metal. According to the theory described in Section 'The commonly used theory', the hot metal in the centre of the furnace will

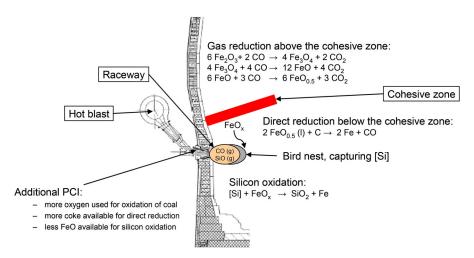


Figure 8. Schematic view of the blast furnace, showing the mechanisms for reduction of FeO.

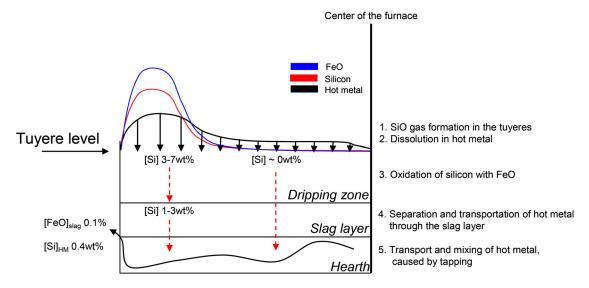


Figure 9. Concept for the 2D model.

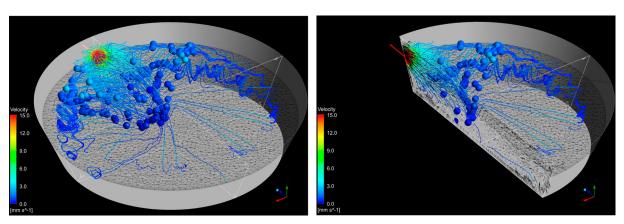


Figure 10. Distributed flow case: 80% of the inflow flow through the outer ring. Streamlines from the outlet into the interior of the hearth, arrows indicate flow direction. Colouring indicates hot metal velocity. Symbols indicate 1 h residence time away from outlet [27].

have high Si content, while the hot metal in the outer rim will have lower concentrations, but no evidence was found for this theory.

The cast will result in an average value but will depend on the flow patterns in the hearth, explaining the Si variations. SiO₂ in slag will not affect the dissolution process due to its low activity.

Thermal state of the furnace

The thermal state of the furnace is the amount of energy relative to the burden. A so-called high thermal state means higher energy input than required, whereas a low thermal state means lower energy input than required. The thermal state also reflects the Si content in the hot metal: a high thermal state leads to higher Si and vice versa. Adding more coal or coke increases the flame temperature and

Table 4. Calculations of reoxidation of [Si] by FeO – (based on Kerkkonen et al. [18])

Compound	Max in the raceway (wt-%)	After the FeO + [Si] reaction (wt-%)
FeO	9.3	0.0
Si	4.3	2.6
Fe ⁰ (metallic iron)	82	93
С	4.0	4.2

increases the input of SiO2, so that more SiO gas will be formed. The additional carbon will lead to additional reduction capacity or less FeO. Both mechanisms lead to higher Si concentrations in hot metal.

Global mass balance

Using global data, a simplified mass balance can be constructed:

- Input via pellets/sinter, PCI and coke:
 - ∘ Si from ~1.5 ton/thm slag in pellets and sinters is 26 kg/ thm (kg per ton hot metal)
 - Si from quartz in \sim 0.2 ton/thm PCI is 4.3 kg/thm (\sim 11%)
 - Si from quartz in ~0.3 ton/thm coke is 7.5 kg/thm $(\sim 20\%)$
- Output via hot metal and slag:
 - ∘ Si in hot metal is around 4.0 kg/thm (~10%)
 - ∘ Si in ~0.2 ton/thm blast furnace slag is around 34 kg/ thm
 - Some dust

Based on these numbers, around 1/3 of the total Si-input via the quartz phase ends up in hot metal, the rest of the silica ends up in the slag.



Conclusions

The goal of this paper is to investigate the mechanism how silicon is introduced in hot metal and to develop an online 2D silicon prediction model. The mechanism to transfer silicon into hot metal was studied via data from the literature, data from HIsarna® and thermodynamic calculations. The main conclusions:

- The process for silicon formation is via SiO gas, which can only be formed at temperatures above 2000°C from quartz. Data from HIsarna shows that at lower temperatures, no silica is dissolved in hot metal, confirming this statement.
- A large part of the pure silica (quartz) entering the raceway is gasified.
 - Lowering the silica input from coal and coke will lower the silicon in hot metal.
 - Extra PCI will not only generate more SiO gas, but less FeO will be available in the raceway area as well. Both mechanisms result in increasing silicon content in hot
 - Silica from sinters and pellets is present in the slag phase. This silica will remain as slag and will not take part in the gasification process.
- SiO is not transported far into the furnace but dissolves in hot metal in the raceway area.
 - Silicon levels may reach relatively high values between 4 and 7 wt-% just behind the raceway, indicating fastdissolving kinetics.
 - The adapted theory from Steiler [10] may need revision (Section 'The commonly used theory', point 4c)
- Reoxidation of silicon is done by FeO. FeO is the only oxygen source below the raceway.
 - FeO is closely related to silicon concentrations in hot metals.
- The remaining silicon is further diluted by transport phenomena in the hearth, resulting in a relatively low average value during casting.
 - Iron fluxes at the end of the raceway start high in silicon (4.3 wt-%), while the intermediate state after reoxidation is still relatively high (2.6 wt-%). The result during casting is an average silicon concentration of around 0.4 wt-%, due to mixing with liquid iron low in silicon.

Disclosure statement

No potential conflict of interest are expected by the authors.

ORCID

Y. Yang http://orcid.org/0000-0003-4584-6918

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