# EQUILIBRIUM MODEL OF THE FERROSILICON PROCESS IN THE SUBMERGED ARC FURNACE

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## Abstract

Based on the non-stoichiometric Gibbs free enthalpy minimization algorithm (FEM), an equilibrium physicochemical model of the ferrosilicon FeSi75 smelting process has been elaborated where the influence of process parameters was determined. In the above model in the submerged arc furnace, the basic parameters are temperatures of the upper and lower reactors and a fraction of the reducer in reaction mixture expressed by the molar ratio $C/SiO_2$. In addition, considering the carbon content and the content of admixtures in the metallic phase, small amounts of oxides: $Al_2O_3, CaO, MgO, TiO_2$ in the simulation calculations were included in the reaction mixture. The following temperature ranges were taken into account: 1630 to 1650 ℃ for the upper reactor and 1920 to 2010 ℃ for the lower one. The presented calculation results show a good compliance with the observations of the increased quality ferrosilicon FeSi75 (used in the production of special steels) smelting process under industrial conditions. This concerns in particular the influence of temperature conditions and the fraction of the reducer in the reaction mixture on the silica reduction process efficiency as well as the yields of basic component Si and the trace elements Al, Ca, Mg, Ti.

**Keywords:** Ferrosilicon, equilibrium model, Gibbs free enthalpy minimization, submerged arc furnace

## 1.    INTRODUCTION

Ferrosilicon is smelted in the submerged arc furnaces with Søderberg self-baking electrodes immersed in the charge material [1,2]. Ferrosilicon smelting is a continuous process. Raw materials are fed into the furnace from its top as a mixture of quartzite, carbon reducers (coal, pea coke, and wood chips) and iron-bearing materials (mill scale or steel chips). Periodically, liquid metal is tapped into the ladle through one of the tap holes located next to the furnace hearth. The heat, necessary for the highly endothermic carbothermic silica reduction process, is generated directly in the charge as a result of electric current flow (resistive heating) and due to electric arc radiation in the gas chambers placed near the electrode tips. The efficiency of carbothermic silica reduction process depends on temperature and energy conditions within the reaction zones near the electrodes. Ferrosilicon smelting process in the submerged arc furnaces is considered to be a slag-free process although small amounts of slag are always seen in the ferrosilicon furnace. This is associated with the quality of raw materials and the presence of small amounts of Al₂O₃, CaO, MgO and TiO₂ in the charge mixture. A physicochemical model of ferrosilicon smelting process has been presented, using the non-stoichiometric algorithm of Gibbs free energy minimization (FEM) [3,4]. As the input data, the algorithm requires the initial composition of the reaction mixture, process parameters (*T* - temperature, *P* - pressure) and a list of components that may appear in the equilibrium composition of each system phase. In this approach, chemical reactions or stoichiometric equations do not have to be determined. For calculations, the HSC 7.1 [4] thermochemical software was applied, enabling the use of a thermochemical database and solution of FEM problems directly in the Excel spreadsheet. The presented model is an extension of models with two isothermal reactors [5,6] where equilibrium conditions in Fe-Si-O-C systems are taken into account. In this paper, a Fe-Si-O-C-Al-Ca-Mg-Ti system was added to the equilibrium model so that the presence of Al₂O₃, CaO, MgO, TiO₂ admixtures, related to ferrosilicon smelting in the submerged arc furnaces, can be taken into consideration.

## 2. EQUILIBRIUM MODEL FOR THE ELECTROCHEMICAL PROCESS OF FERROSILICON SMELTING

As the physicochemical model of the process, a system of two closed isothermal reactors [6] was selected: an upper reactor with a lower temperature $T_1$ and a lower reactor with a higher temperature $T_2$. Between the reactors and the environment as well as between the in-system reactors, the cyclic mass transfer occurs when the equilibrium state is reached. Simulation of continuous ferrosilicon process was performed recursively. In Cycle 1, a portion of the reaction mixture is fed into the upper reactor (**Table 1**). The molar composition of the mixture corresponds approximately to the composition of charge mixture used for FeSi75 ferrosilicon smelting. In Reactor 1, chemical reactions occur until the equilibrium state is reached at the temperature $T1$. Then, the gaseous phase products leave the reactor and, when cooled to $T_w = 850\,°C$, they do not participate in the further chemical process. At the same time, the products of condensed phases are transferred down from Reactor 1 o Reactor 2 where they react until the equilibrium state is reached

**Table 1** Molar composition of the reaction mixture in the model of ferrosilicon smelting process

| i | Component | Atomic mass | $m_i$ (kmol) |
|---|---|---|---|
| 1 | $Al_2O_3$ | 101.96 | 0.001 |
| 2 | $Al_2O_3*SiO_2(A)$ | 162.05 | 0.01 |
| 3 | CaO | 56.08 | 0.015 |
| 4 | $FeO*SiO_2$ | 131.93 | 0.01 |
| 5 | MgO | 55.85 | 0.005 |
| 6 | $SiO_2$ | 60.08 | 0.98 |
| 7 | $TiO_2$ | 79.90 | 0.0005 |
| 8 | Fe | 55.85 | 0.13 |
| 9 | C | 12.01 | 1.9 -2.0 |

at $T_2$. Next, the condensed phase products leave Reactor 2 and, following the temperature decrease to $T_1$, they form metallic and slag phases, leave the system and do not participate in the further process. Simultaneously, the gaseous phase products leave Reactor 2 and, after being cooled to the temperature $T_1$, they are transferred back to Reactor 1 where they react during the next cycle. Cycle 2 and all further cycles begin with a new portion of the reaction mixture being fed into Reactor 1 where it reacts with gaseous phase components (that remain after the previous cycle) until the equilibrium state is reached at the temperature $T_1$. Next, during Cycle 2 and each further cycles, the process is the same as in Cycle 1. Considering the real conditions, the temperature $T_w$ corresponds to the temperature of gases (waste products of silica reduction with carbon) observed immediately after leaving the inner part of the ferrosilicon furnace, close to the upper surface of the charge. Using a simplifying assumption, Reactor 1 corresponds to charge zones in the operating space of a submerged arc furnace where the heat is generated as a result of electric current flow (resistive heating). Zone 2 corresponds to the gas chambers that surround the electrodes tips where the heat is generated due to the electric arc radiation. Based on the model, the mass and energy balances of the process, theoretical indexes of raw material and energy consumption as well as element yields were determined. The non-stoichiometric algorithm of Gibbs minimization is a useful approach for solving the non-linear optimization problem with contrains resulting from the mass balances for individual elements in the system:

$$\left.\begin{aligned} &\min_{\boldsymbol{n}} \frac{G(T_i, P, \boldsymbol{n}_i)}{RT_i} \\ &A \cdot \boldsymbol{n}_i = B \\ &\boldsymbol{n} \geq 0 \end{aligned}\right\} \tag{1}$$

where:

$G(T_i, P, \boldsymbol{n}_i,)$    - Gibbs free enthalpy function for the reactor, $i = 1,2$

$\boldsymbol{n}_i = \left(\boldsymbol{n}_i^1, \boldsymbol{n}_i^2, \dots, \boldsymbol{n}_i^f\right)^T, \ \ \boldsymbol{n}_i^r = \left(n_{1,i}^r, n_{2,i}^r, \dots, n_{k^r,i}^r\right)^T$

$\boldsymbol{n}_i^r$ - vectors whose components are numbers of moles of individual phase components, $r = 1,2,\dots,f$

$\boldsymbol{A}$ - atomic matrix, sized $N \times n$, whose elements are numbers of individual atoms of a specific phase components (**Tables 2,3**)

$n = k^1 + k^2 + \cdots + k^f$, a number of components in the system

$\boldsymbol{B}$ - vector $N \times 1$: a vector whose components are numbers of gram atoms of $\mathrm{Fe, Si, O, C, Al, Ca, Mg, Ti}$ elements that are present in the system during each cycle

$f$ - a number of phases in the system

$N = 8$ - a number of atoms that form the Fe-Si-O-C-Al-Ca-Mg-Ti system

$R$ - gas constant (J/(kmol·K))

For each cycle, the equilibrium composition of the system and the energy balance are determined. The energy balance includes the following components:

$$Q_R = \Delta H_{298}^{T1}(\boldsymbol{n^0}) + \Delta H_{T1}^{R1}(\boldsymbol{n^0}, \boldsymbol{n_2^1}) + \ \Delta H_{T1}^{T2}(\boldsymbol{n_1^2}, \boldsymbol{n_1^3}, \boldsymbol{n_1^4}, \boldsymbol{n_1^5}) + \Delta H_{T1}^{T_w}(\boldsymbol{n_1^1}) + \ \Delta H_{T2}^{T1}(\boldsymbol{n_2^1}), \tag{2}$$

$$Q_{Arc} = \Delta H_{T2}^{R2}(\boldsymbol{n_1^2}, \boldsymbol{n_1^3}, \boldsymbol{n_1^4}, \boldsymbol{n_1^5}) + \Delta H_{T2}^{T1}(\boldsymbol{n_2^2}, \boldsymbol{n_2^3}, \boldsymbol{n_2^4}, \boldsymbol{n_2^5}), \tag{3}$$

$$Q = Q_R + Q_{Arc} \tag{4}$$

where:

$\Delta H_{298}^{T1}(\boldsymbol{n^0})$ - enthalpy change of the substrates during heating from the ambient temperature up to the temperature $T_1$ (MJ/kmol)

$\Delta H_{T1}^{R1}(\boldsymbol{n^0}, \boldsymbol{n_2^1})$, $\Delta H_{T2}^{R2}(\boldsymbol{n_1^2}, \boldsymbol{n_1^3}, \boldsymbol{n_1^4}, \boldsymbol{n_1^5})$ - thermal effects of the chemical reactions in Reactor 1 and Reactor 2, respectively (MJ/kmol)

$\Delta H_{T1}^{T2}(\boldsymbol{n_1^2}, \boldsymbol{n_1^3}, \boldsymbol{n_1^4}, \boldsymbol{n_1^5})$ - enthalpy change of the condensed phases while heating during the transfer from Reactor 1 down to Reactor 2 (MJ/kmol)

$\Delta H_{T2}^{T1}(\boldsymbol{n_2^1})$ - enthalpy change of the gaseous phase while cooling during the transfer from Reactor 2 up to Reactor 1 (MJ/kmol)

$\Delta H_{T1}^{T_w}(\boldsymbol{n_1^1})$ - enthalpy change of the gaseous phase components after leaving Reactor 1 and cooling from the temperature $T_1$ to the temperature $T_w$ (MJ/kmol)

$\Delta H_{T2}^{T1}(\boldsymbol{n_2^2}, \boldsymbol{n_2^3}, \boldsymbol{n_2^4}, \boldsymbol{n_2^5})$ - enthalpy change of the condensed phase components after leaving Reactor 2 and cooling from the temperature $T_2$ to $T_1$ (MJ/kmol)

$Q$ - overall energy demand for the reduction process (MJ/kmol)

The components of energy balance, $Q_R, Q_{Arc}$, describe the distribution of heat in reaction zones surrounding the electrodes in the submerged arc furnace. The component $Q_R$ corresponds to the charge zones where the heat is generated due to resistive heating while the component $Q_{Arc}$ corresponds to the gas chambers located near the electrode tips where the heat is generated as a result of the electric arc radiation. The heat distribution within the operating space of the furnace was determined with percent fractions $Q_1$, $Q_2$ related to the components $Q_R, Q_{Arc}$ of the energy balance as follows:

$$Q_1 = 100 \cdot \frac{Q_w}{Q}, \qquad Q_2 = 100 \cdot \frac{Q_{Arc}}{Q}. \tag{5}$$

Based on the mass and energy balances, the yields of the basic component Si and the trace elements $Al, Ca, Mg, Ti$ were calculated and the energy consumption index $E$ was determined:

$$E = \frac{1000 \cdot Q}{3.6 \cdot \eta \cdot m} \ \ (kWh/t) \tag{6}$$

where:

$m$ - weight of the metallic phase that leaves Reactor 2 (kg/kmol SiO₂)

$\eta$ - coefficient related to the energy efficiency of the process, $\eta \approx 0.88$

3.6 - Energy Unit Converter, $1 \ kWh = 3.6 \ MJ$

Using the thermochemical database HSC 7.1 [4], 150 components in the Fe-Si-O-C-Al.-Ca-Mg-Ti system were considered. They form five phases in the 1200 to 2500 ℃ temperature range. Due to a large number of the components, **Table 2** presents the components of individual phases limited to the Fe-Si-O-C-Al system. For comparison, a full list of the metallic phase components in the Fe-Si-O-C-Al-Ca-Mg-Ti system is presented in **Table 3**.

**Table 2** Components of individual phases of the Fe-Si-O-C-Al. system ($N = 5, f = 5, n = 84, k^1 = 28, k^2 = 25, k^3 = 1, k^4 = 1, k^5 = 13$ )

| Phase 1 | $C_{5(g)}$ | $C_{6(g)}$ | $C_{7(g)}$ | $C_{8(g)}$ | | | $CO_{(g)}$ | $CO_{2(g)}$ |
|---|---|---|---|---|---|---|---|---|
| | $C_2O_{(g)}$ | $C_3O_{2(g)}$ | $Fe_{(g)}$ | $Fe_{2(g)}$ | $Fe(CO)_{5(g)}$ | $FeO_{(g)}$ | $FeO_{2(g)}$ | $O_{(g)}$ |
| | $O_{2(g)}$ | $O_{3(g)}$ | $Si_{(g)}$ | $Si_{2(g)}$ | $Si_{3(g)}$ | $Si_{4(g)}$ | $SiC_{(g)}$ | $SiC_{2(g)}$ |
| | $Si_2C_{(g)}$ | $SiO_{(g)}$ | $SiO_{2(g)}$ | $Si_2O_{2(g)}$ | | | | |
| Phase 2 | $Al_4CO_4$ | $AlO$ | $Al_2O_3$ | | | | | |
| | | | | | | | | |
| | $FeO$ | | $Fe_2O_3$ | | | | | |
| | $SiC_2$ | | | | | | | |
| Phase 3 | $C$ | | | | | | | |
| Phase 4 | $SiC$ | | | | | | | |
| Phase 5 | $Al_4C_3$ | $Al_4SiC_4$ | $Fe_2C$ | $Fe_3C$ | $Al$ | $Fe$ | $FeSi$ | $FeSi_2$ |
| | $Fe_3Si$ | $Fe_5Si_3$ | $Si$ | | | | | |

**Table 3** Components of the metallic phase in the Fe-Si-O-C-Al.-Ca-Mg-Ti system ($N = 8, f = 5, n = 150, k^1 = 47, k^2 = 72, k^3 = 1, k^4 = 1, k^5 = 29$)

| Phase 5 | $Al$ | $Al_3Ti$ | $Al_3C_3$ | $Al_4SiC_4$ | $AlTi$ | $Ca$ | $CaAl_2$ | $CaAl_4$ |
|---|---|---|---|---|---|---|---|---|
| | $CaC_2$ | $CaSi$ | $Fe$ | $Fe_2C$ | $Fe_2Ti$ | $Fe_3C$ | $Fe_3Si$ | $Fe_5Si_3$ |
| | $FeSi$ | $FeSi_2$ | $FeTi$ | $Mg$ | $Mg_2C_3$ | $Mg_2Si$ | $MgC_2$ | $Si$ |
| | $Ti$ | $Ti_5Si_3$ | $TiC$ | $TiSi$ | $TiSi_2$ | | | |

## 3.    EFFECTS OF PARAMETERS ON EFFICIENCY OF THE SILICA REDUCTION PROCESS

Basic parameters in a model of the silica reduction process in the submerged arc furnace are temperatures $T_1, T_2$ of the upper and lower reactors as well as the fraction of reducer in the reaction mixture expressed by

the molar ratio $m_9 = C/SiO_2$ (**Table 1**). In addition, simulation calculations included a small fraction of oxides: $Al_2O_3, CaO, MgO, TiO_2$ in the reaction mixture that are always present in the ferrosilicon smelting process. Temperature ranges of $T_1$: 1630 to 1650 °C, $T_2$: 1920 to 2010 °C were included in the calculations. The data on reaction mixture components are presented in **Table 1**. Schematic results following 25 cycles of the reaction system differing in the temperature conditions of reaction zones are presented in **Figures 1 and 2**. The heat balance data for the **Figure 1** process are presented in **Table 4**. The calculation results are well consistent with observations of the industrial process of ferrosilicon smelting. This particularly refers to the effects of temperature conditions and the fraction of reducer in the charge mixture on the efficiency of the reduction process. Th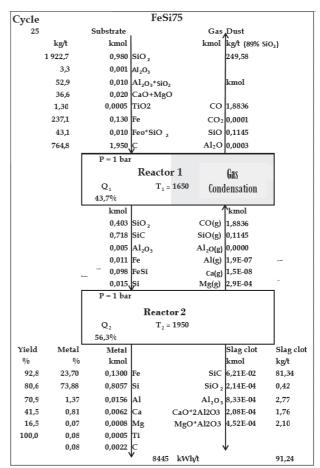e results shown in **Figures 1 and 2** confirm a favourable influence of the lower temperature of the upper zone ($T_1$) and of the higher temperature of the lower zone ($T_2$) on the reduction process efficiency. The following values of the process efficiency are well consistent with the observations of ferrosilicon smelting industrial process: specific energy consumption index $E$, basic component Si yield, trace element (Al, Ca, Mg, Ti) yields, metallic phase chemical composition and amounts of waste products of the process (dust, SiC phase, slag phase).

**Figure 1 — Cycle 25, FeSi75**

| Substrate kg/t | Substrate kmol | | Gas Dust kg/t (89% SiO₂) |
|---|---|---|---|
| 1 781,6 | 0,980 | SiO₂ | 169,59 |
| 3,1 | 0,001 | Al₂O₃ | |
| 49,0 | 0,010 | Al₂O₃*SiO₂ | kmol |
| 33,9 | 0,020 | CaO+MgO | |
| 1,21 | 0,0005 | TiO2 | CO 1,9276 |
| 219,7 | 0,130 | Fe | CO₂ 0,0001 |
| 39,9 | 0,010 | Feo*SiO₂ | SiO 0,0840 |
| 708,7 | 1,950 | C | Al₂O 0,0002 |

P = 1 bar

**Reactor 1** — $Q_1$, $T_1$ = 1630, 37,3% — Gas Condensation

| kmol | | | kmol (gas) |
|---|---|---|---|
| 0,443 | SiO₂ | CO(g) | 1,9276 |
| 0,758 | SiC | SiO(g) | 0,0840 |
| 0,005 | Al₂O₃ | Al₂O(g) | 0,0000 |
| 0,012 | Fe | Al(g) | 1,2E-07 |
| 0,089 | FeSi | Ca(g) | 1,0E-08 |
| 0,011 | Si | Mg(g) | 2,4E-04 |

P = 1 bar

**Reactor 2** — $Q_2$, $T_2$ = 1960, 62,7%

| Yield % | Metal % | Metal kmol | | Slag clot kmol | Slag clot kg/t |
|---|---|---|---|---|---|
| 92,8 | 21,96 | 0,1300 | Fe | SiC 1,68E-02 | 20,36 |
| 88,4 | 75,11 | 0,8839 | Si | SiO₂ 1,03E-04 | 0,19 |
| 85,9 | 1,54 | 0,0189 | Al | Al₂O₃ 3,92E-04 | 1,21 |
| 62,0 | 1,13 | 0,0093 | Ca | CaO*2Al2O3 9,75E-05 | 0,77 |
| 18,5 | 0,07 | 0,0009 | Mg | MgO*Al2O3 1,91E-04 | 0,82 |
| 100,0 | 0,07 | 0,0005 | Ti | | |
| | 0,12 | 0,0032 | C | 7941 kWh/t | 24,42 |

**Figure 2 — Cycle 25, FeSi75**

| Substrate kg/t | Substrate kmol | | Gas Dust kg/t (89% SiO₂) |
|---|---|---|---|
| 1 922,7 | 0,980 | SiO₂ | 249,58 |
| 3,3 | 0,001 | Al₂O₃ | |
| 52,9 | 0,010 | Al₂O₃*SiO₂ | kmol |
| 36,6 | 0,020 | CaO+MgO | |
| 1,30 | 0,0005 | TiO2 | CO 1,8836 |
| 237,1 | 0,130 | Fe | CO₂ 0,0001 |
| 43,1 | 0,010 | Feo*SiO₂ | SiO 0,1145 |
| 764,8 | 1,950 | C | Al₂O 0,0003 |

P = 1 bar

**Reactor 1** — $Q_1$, $T_1$ = 1650, 43,7% — Gas Condensation

| kmol | | | kmol (gas) |
|---|---|---|---|
| 0,403 | SiO₂ | CO(g) | 1,8836 |
| 0,718 | SiC | SiO(g) | 0,1145 |
| 0,005 | Al₂O₃ | Al₂O(g) | 0,0000 |
| 0,011 | Fe | Al(g) | 1,9E-07 |
| 0,098 | FeSi | Ca(g) | 1,5E-08 |
| 0,015 | Si | Mg(g) | 2,9E-04 |

P = 1 bar

**Reactor 2** — $Q_2$, $T_2$ = 1950, 56,3%

| Yield % | Metal % | Metal kmol | | Slag clot kmol | Slag clot kg/t |
|---|---|---|---|---|---|
| 92,8 | 23,70 | 0,1300 | Fe | SiC 6,21E-02 | 81,34 |
| 80,6 | 73,88 | 0,8057 | Si | SiO₂ 2,14E-04 | 0,42 |
| 70,9 | 1,37 | 0,0156 | Al | Al₂O₃ 8,33E-04 | 2,77 |
| 41,5 | 0,81 | 0,0062 | Ca | CaO*2Al2O3 2,08E-04 | 1,76 |
| 16,5 | 0,07 | 0,0008 | Mg | MgO*Al2O3 4,52E-04 | 2,10 |
| 100,0 | 0,08 | 0,0005 | Ti | | |
| | 0,08 | 0,0022 | C | 8445 kWh/t | 91,24 |

**Figure 1** Results of simulation calculations based on the model of ferrosilicon smelting process. Process parameters: temperature of reactors: $T_1 = 1630$ °C, $T_2 = 1960$ °C, $P$ = 1 bar, charge mixture composition (kmol): $m_1 = 0.001$, $m_2 = 0.01$, $m_3 = 0.015$, $m_4 = 0.01$, $m_5 = 0.005$, $m_6 = 0.98$, $m_7 = 0.0005$, $m_8 = 0.13$, $m_9 = 1.95$

**Figure 2** Results of simulation calculations based on the model of ferrosilicon smelting process. Process parameters: temperature of reactors: $T_1 = 1650$ °C, $T_2 = 1950$ °C, $P$ = 1 bar, charge mixture composition (kmol): $m_1 = 0.001$, $m_2 = 0.01$, $m_3 = 0.015$, $m_4 = 0.01$, $m_5 = 0.005$, $m_6 = 0.98$, $m_7 = 0.0005$, $m_8 = 0.13$, $m_9 = 1.95$

**Table 4** Heat balance of the process presented in **Figure 1**

| No. | Heat balance component | (kcal/mol) | (MJ/kmol) | (kWh/Mg) | (%) |
|---|---|---|---|---|---|
| 1 | Substrate heating from 25°C up to $T_1$ | 46,68 | 195,29 | | |
| 2 | Thermal effect of reactions in Reactor 1 | 38,21 | 159,86 | | |
| 3 | Cooling of gas from Reactor 2: $T_2 \rightarrow T_1$ | -4,13 | -17,27 | | $Q_1$ |
| 4 | Heating of products leaving Reactor 1 up to $T_2$ | 9,62 | 40.24 | | |
| 5 | Cooling of gas from Reactor 1: from $T_1$ up to 850 °C | -16,24 | -67,95 | | |
| | Overall heat in Reactor 1, $Q_R$ | 74,13 | 310,17 | 2606,8 | 37,3 |
| 6 | Reaction of Reactor 1 products in Reactor 2 | 127,30 | 532,61 | | |
| 7 | Cooling of metal and slag from $T_2$ to the tapping temp. $T_1$ | -2,70 | -11,32 | | $Q_2$ |
| | Overall heat in Reactor 2, $Q_{Arc}$ | 127,59 | 521,29 | 4381,3 | 62,7 |
| 8 | Overall energy consumption, $Q = Q_R + Q_{Arc}$ | 198,72 | 831,46 | | 100,0 |
| | Energy consumption, $E = Q/\eta$ $\eta = 0,88$ | 225,82 | 944,84 | 7941,0 | |

## 4.     CONCLUSIONS

1)     The presented model enables a better understanding of the ferrosilicon smelting process in the submerged arc furnace. It gives a potential for determining the effects of reducer fraction in the charge and temperature conditions of the reactions zones on the efficiency of silica reduction with carbon.

2)     A small deficiency of reducer in the charge mixture in relation to the stoichiometry of the silica reduction reaction $SiO_2+2C=Si+2CO$ has a favorable effect on a smaller trend for SiC phase formation and on the efficiency of the silica reduction process.

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