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Prediction of the Silicon Yield on using Different SiO_2/C/SiC Ratios

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ABSTRACT

Silicon is produced by carbothermal of SiO_2 at high temperatures from 1600°C to 3000°C in an electric arc furnace. During production, significant amounts of carbon monoxide are observed.
In this paper, the effect of carbon on the yield of silicon is discussed in order to reduce the amount of carbon monoxide (CO) production without affecting the yield of silicon. That discussing and illustrating by drawing the results of prediction of silicon has production using different ratios of SiO_2/C/SiC.
The results showed that the increase in carbon moles leads to an increase in the yield of carbon monoxide. Specifically, 9 moles of carbon can be replaced with 1 mole of silicon carbide to get half the carbon monoxide output and to maintain the yield of silicon at 2200 K.

1. Introduction

Silicon with a purity of 98.5% is produced in electric arc furnaces. The process is greatly improved with more efficient material handling and improved process control resulting in energy savings. An electric arc furnace (EAF) consists of a vessel filled with quartz and carbonaceous materials [1]. Electric arc furnaces operate at high temperatures between 1000°C and 3000°C, and electric arc furnaces in the past decade have been trying to focus on making the process more efficient and environmentally friendly. The increase in productivity in electric arc furnaces was directly related to the increase in the use of oxygen, as the amount of electrical input replaced the exothermic reaction energy [2]. Therefore, the dynamic model was studied with all the features of the static model in addition to containing information from the reaction kinetics and process dynamics. The mechanism of interactions and kinetics of interactions for this complex system must first be studied [1]. Because experiments are difficult at high temperatures above 1500°C, the reactions that occur in the electric arc furnace for the production of silicon are not agreed upon, and because of the multiplicity of these experiments and about 17 potential reactions are involved in the electric arc furnace. Therefore, the mechanisms of the reaction of SiO_2 + SiC and the effect of additive gases in the temperature range of 1270-1400 ° C were studied [1]. Bekker et al. [3] developed a model of EAF of basic kinetic relationships and thermodynamic with the end goal to simulate closed-loop control. Simplifying assumptions and empirical relationships are used to describe mechanisms which are not measurable or well understood. Osthuizen et al. [4] expanded the model to predict the depth of slag foam and the temperature of natural gas. The next models [5, 6] assume that the process consists of equilibrium zones but there are limitations of mass transit. Cameron et al. [5] have developed the EAF model for dynamic simulation to identify improvements in EAF operating practices, assumed that it consisted of four stages (metal, slag, organic steel and gas) and analyzed from six interfaces between metal, carbon, gas and slag. While the kinetics and mechanisms of liquid Si – C were presented and discussed [6]. Furthermore, the thermodynamic carbon synthesis chemistry of β-SiC has been shown [7]. Furthermore, a model has been developed for the rapid carbohydrate synthesis of silicon carbide micro powders [8]. SiO gas production in electric arc reactor is discussed [9]. Selected reaction mechanisms in carbothemal reduction to produce Si are shown in [10]. Finally, the carbonic reactions to reduce silica to produce silicon were determined and the reaction kinetics was determined [11].

A computer program called SOLGASMIX was used to calculate the chemical equilibrium at high temperature using the free energy reduction method. Carbothermal reduction of SiO_2 to Si was simulated as a function of the molar ratio of SiO_2, SiC and C using a thermodynamic calculation of Gibb's free energy minimization [12]. Silicon metal formed in the SiO_2-C system at 1250 °C and completed its formation at 1500-2000 °C with an increase of C. In addition, SiC to SiO_2-C system completes the conversion of SiO_2 to Si which catalyzes the lowering the temperature of the system and excess carbon. In this paper, a different computational technique, such as MATLAB® simulation, was used to simulate Carbothermal reduction of SiO_2 to Si using Gibb's free energy minimization. Then focus on the best conditions for the best productivity of silicon while preserving the environment by reducing moles of carbon.

2. Simulation

The thermodynamic equilibrium calculations for Gibb's free energy minimization were performed using a MATLAB® simulator. For the simulation, the following assumptions were used (1) Substances that can be reactants or products – SiO_2, SiC,

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C, Si, SiO, and CO; (2) Feed material – SiO_2, C and/or SiC; (3) The temperature range is 1800 - 3000°C; (4) Pressure 1 atm.

MATLAB® has also been used with the assumption that the gases are real. Filsinger and Bourric [11] presented the mechanism of the Carbothermal reduction reaction of SiO2 by carbon used in this study, as shown in equations (1)–(7).

$$\mathrm{SiO_2 + C \rightarrow SiO + CO} \qquad (1)$$

$$\mathrm{SiO_2 + 3C \rightarrow SiC + 2CO} \qquad (2)$$

$$\mathrm{2SiO_2 + SiC \rightarrow 3SiO + CO} \qquad (3)$$

$$\mathrm{SiO + 2C \rightarrow SiC + CO} \qquad (4)$$

$$\mathrm{SiO + SIC \rightarrow 2Si + CO} \qquad (5)$$

$$\mathrm{SiO_2 + 2Si \rightarrow 2SO} \qquad (6)$$

$$\mathrm{3SiO_2 + 2SiC \rightarrow Si + 4SiO + 2CO} \qquad (7)$$

Janaf tables [12] are used to get the perfect Gibbs free energy (Gi) and then used, but the real Gibbs free energy is computed by:

$$\mathbf{G_{1=}G_{if} + RTLny_1 + RTLn\acute{\emptyset}_1} \qquad (\mathbf{8})$$

R is the gas constant; T is the temperature; y1 is the mole ratio of the compound 1; Ǿ: is the fugacity coefficient. Solids and liquids Ǿ =1. When y1 = zero RTLny1 is discarded. To calculate Ǿ for a mixture of two gases (SiC, SiO) the equations in the literature [13] were used. Table 1 shows the physical properties of SiO and SiC

Table 1 Physical properties of SiO and SiC

Physical properties	P_C (bar)	T_C (K)	T_b (K)	w	References
SiO	**990.58**	**3672.52**	**2150**	**1.136**	**[16]**
SiC	**717.512**	**3384.30**	**1908**	**0.876**	**[16] and [17]**

$$\mathrm{Ln}\acute{\emptyset}_1 = \frac{P}{RT}\left(B_{11} + y^2{}_2\delta_{12}\right) \qquad (9)$$

$$\mathrm{Ln}\acute{\emptyset}_2 = \frac{P}{RT}\left(B_{22} + y^2{}_1\delta_{12}\right) \qquad (10)$$

$$\delta_{12} = 2B_{12} - B_{11} - B_{22} \qquad (11)$$

B11, B22: values of pure-species virial coefficients, B12: cross coefficients. Calculate B11, B22 and B12 in [18]:

$$B_{ij} = \frac{RT_{c_{ij}}}{P_{c_{ij}}}\left(B^{\circ} + w_{ij}B^{1}\right) \qquad (12)$$

Calculate w_{ij} was used the equations in references [16] and [19].

3. Results and discussion

Different mole ratios of carbon, silica, and silicon carbide were selected in real case. This is to determine the best productivity of silicon, taking into account reducing the productivity of carbon monoxide to preserve the environment without affecting the productivity of silicon.

Figure (1) represents the amounts of carbon monoxide produced at different temperatures.

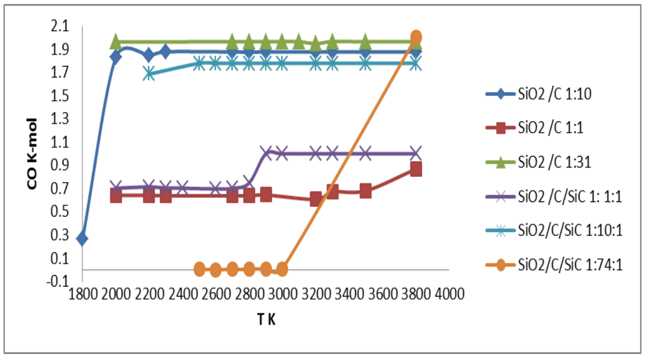

Figure 1: Carbon monoxide production at different temperatures

Figure (2) represents the amounts of silicon produced at different temperature.

Figure (2) shows the highest amounts of silicon produced at SiO_2/ C/ SiC 1:10:1 and SiO_2/ C/ SiC 1:1:1. When comparing the quantities of carbon monoxide and silicon, it was found that the best in terms of environmental preservation and also with a high productivity of silicon at SiO_2/ C/ SiC 1:1:1 and a temperature of 2200 K.

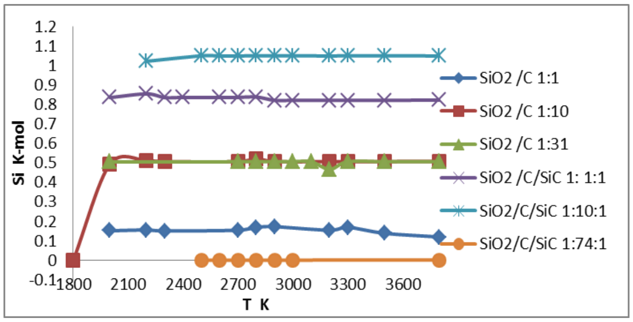

Figure 2: Silicon production at different temperatures

It has been observed that increasing the number of moles of carbon more than 10 moles for each mole of silica does not increase the productivity of silicon, but may even stop the production of silicon, as in the case of SiO_2/ C/ SiC 1:74:1.

It was found that replacing 9 moles of carbon with 1 mole of silicon carbide gives a higher yield of silicon in case SiO_2/ C 1:10 to SiO_2/ C/ SiC 1:1:1. The amounts of carbon monoxide are less than half at the presence of silicon carbide.

These tables (2, 3) show what was explained above.

Table 2: Amounts of carbon monoxide and silicon produced (k-mol) at SiO_2 /C 1:10

T K	CO	Si
1800	0.263	0
2000	1.832	0.494
2200	1.847	0.511
2300	1.878	0.507
2700	1.877	0.507
2800	1.874	0.521
2900	1.877	0.507
3200	1.877	0.507
3300	1.877	0.507
3500	1.877	0.507
3800	1.877	0.508

Table 3: Amounts of carbon monoxide and silicon produced (k-mol) at SiO_2 /C/SiC 1:1:1

T K	CO	Si
2000	0.703	0.836
2200	0.714	0.855
2300	0.704	0.836
2400	0.703	0.837
2600	0.697	0.837
2700	0.702	0.837
2800	0.748	0.839
2900	0.999	0.822
3000	0.999	0.822
3200	0.999	0.822
3300	0.999	0.822
3500	0.999	0.822
3800	1	0.823

Table 4: Amounts of carbon monoxide and silicon produced (k-mol) at SiO_2 /C/SiC 1:10:1

T K	CO	Si
2200	1.686	1.023
2500	1.779	1.051
2600	1.779	1.05
2700	1.779	1.05
2800	1.779	1.051
2900	1.779	1.051
3000	1.779	1.051
3200	1.778	1.05
3300	1.779	1.051
3500	1.779	1.051
3800	1.778	1.05

Table No. 4 represents the amounts of carbon monoxide and silicon at mole ratio is SiO_2 /C/SiC 1:10:1, it was observed that the amounts of carbon monoxide more than doubled. Although the amounts of silicon increased only 0.1 k-mol more than the case of SiO_2 /C/SiC 1:1:1 .

The results were compared with other paper as "A simulation study on the direct carbothermal reduction of SiO_2 for Si metal" [13]. The results were identical between them in the production of silicon and the release of carbon monoxide.

4. Conclusion

The electric arc furnace has shown a distinguished performance in the manufacture of silicon and this was done by testing the results in the MATLAB® program. The best amount of silicon production and the lowest amount of carbon monoxide was obtained in the ratio SiO_2/C/SiC 1: 1:1. Whereas, the productivity of silicon at ratio SiO_2/C/SiC 1:1:1 reaches 0.855 kmol, and carbon monoxide decreases to 0.714 kmol, compared to the productivity of silicon at ratio SiO_2/C/SiC 1:10:1 it reaches 1.023 kmol, and carbon monoxide increases to 1.686 kmol at the same temperature . So, by comparison, it is found that, the best in terms of environmental friendliness at SiO_2 / C / SiC 1: 1: 1 and a temperature of 2200 K. It has been observed that increasing the number of moles of carbon more than 10 moles per mole of silica does not increase the yield of silicon, but may even stop the production of silicon, in the case of SiO_2 / C / SiC 1:1:1, that is, the presence of one carbon, the productivity of silicon reaches 0.836 kmol, while in the case of SiO_2 / C / SiC 1:10:1, that is, the presence of ten carbons, the productivity of silicon reaches 1.023 kmol, and with it the presence of carbon monoxide increased to more than double in the case of SiO_2 / C / SiC 1:1:1, when trying to raise carbon to more than 74 carbon, the results became very bad, the yield of silicon reaches zero kmol. It was found that replacing 9 moles of carbon with 1 mole of silicon carbide gives a higher yield of silicon in the case SiO_2 / C 1:10 to SiO_2 / C / SiC 1: 1: 1.The amounts of carbon monoxide are less than half in the presence of silicon carbide. Whereas, the productivity of silicon at SiO_2 / C 1:10 reaches 0.511 kmol and carbon monoxide reaches 1.847 kmol at a temperature of 2200 K. While the productivity of silicon at ratio SiO_2/C/SiC 1:1:1 reaches 0.855 kmol, and carbon monoxide decreases to 0.714 kmol.

References

[1] L.P.Hunt; Silicon Precursors :Their manufacture and Properties; Handbook of Semiconductor silicon technoloogy ; Noyes PuB. (1990).

[2]J. G. Bekker, I. K. Craig, and P. C. Pistorius, "Modeling and simulation of an electric arc furnace process," ISIJ international, vol. 39, pp. 23-32, 1999.

[3]R. D. MacRosty and C. L. Swartz, "Dynamic modeling of an industrial electric arc furnace," Industrial & engineering chemistry research, vol. 44, pp. 8067-8083, 2005.

[4] D. J. Oosthuizen, J. H. Viljoen, I. K. Craig, and P. C. Pistorius, "Modelling of the off-gas exit temperature and slag foam depth of an Electric Arc Furnace," ISIJ international, vol. 41, pp. 399-401, 2001.

[5]A. Cameron, N. Saxena, and K. Broome, "Optimizing EAF operations by dynamic process simulation," in ELECTRIC FURNACE CONFERENCE, 1998, pp. 689-696.

[6]M. Modigell, A. Traebert, and P. Monheim, "A modeling technique for metallurgical processes and its applications," AISE Steel Technology(USA), vol. 78, pp. 45-47, 2001.

[7] Y.-M. Chiang, R. P. Messner, C. D. Terwilliger, and D. R. Behrendt, "Reaction-formed silicon carbide," Materials Science and Engineering: A, vol. 144, pp. 63-74, 1991.

[8] F. Van Dijen and R. Metselaar, "The chemistry of the carbothermal synthesis of β-SiC: Reaction mechanism, reaction rate and grain growth," Journal of the European Ceramic Society, vol. 7, pp. 177-184, 1991.

[9] J. A. Johnson, C. M. Hrenya, and A. W. Weimer, "Intrinsic Reaction and Self‐Diffusion Kinetics for Silicon Carbide Synthesis by Rapid Carbothermal Reduction," Journal of the American Ceramic Society, vol. 85, pp. 2273-2280, 2002.

[10] S. Bao, M. Tangstad, K. Tang, and E. Ringdalen, "Production of SiO gas in the silicon process," Infacon XIII, At Kazakhstan, Almaty, 2013.

[11] E. Ringdalen and M. Tangstad, "Reaction mechanisms in carbothermic production of silicon, study of selected reactions," The Minerals, Metals & Materials Society (TMS), pp. 195-203, 2012.

[12] D. H. Filsinger and D. B. Bourrie, "Silica to silicon: key carbothermic reactions and kinetics," Journal of the American Ceramic Society, vol. 73, pp. 1726-1732, 1990.

[13] H.-C. Lee, S. Dhage, M. S. Akhtar, D. H. Kwak, W. J. Lee, C.-Y. Kim, et al., "A simulation study on the direct carbothermal reduction of SiO2 for Si metal," Current Applied Physics, vol. 10, pp. S218-S221, 2010.

[14] C. L. Yaws, The Yaws handbook of vapor pressure: Antoine coefficients: Gulf Professional Publishing, 2015

[15] J. M. Smith, H. C. Van Ness, M. M. Abbott, and M. T. Swihart, Introduction to chemical engineering thermodynamics, 2018.

[16] E. P. Bruce, M. P. John, and P. O. C. John, Properties of Gases and Liquids, Fifth Edition, 5th ed. ed. New York: McGraw-Hill Education, 2001.

[17] O. Ruff, "The Formation and dissociation of silicon carbide," Transactions of The Electrochemical Society, vol. 68, pp. 87-109, 1935.

[18] Y. K. Yeo, Chemical Engineering Computation with MATLAB®: CRC Press, 2017.

[19] J. H. Perry, "Chemical engineers' handbook," ed: ACS Publications, 1950.

Abbreviation and Symbols

SiO_2	**Silicon dioxide**
CO	**Carbon monoxide**
Si	**Silicon**
SiC	**Silicon carbide**
SiO	**Silicon monoxide**
C	**Carbon**
G_i	**Gibbs free energy**
G_{if}	**Real gibbs free energy**
y_1	**The mole ratio of the compound 1**
Ǿ	**Fugacity coefficient**
R	**Gas constant**
P_C	**Critical pressure**
T_C	**Critical temperature**
T_b	**Boiling point**
w	**Acentric factor**
B_{ij}	**Species virial coefficients**