Design and Numerical Optimization of Gas Guidance System in Casting Silicon Furnace by the Orthogonal Experiment

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Received: 4 February 2021 / Accepted: 2 June 2021 / Published online: 7 June 2021
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Abstract
3D global simulations were performed to investigate the gas flow and impurities transport in casting silicon furnace under the influence of a unique designed spray-type gas guidance system (GGS). The simulation results show that, the intensity of backflow at crucible outlet had no obvious change with the application of this GGS, but the flow area of backflow was effectively inhibited above the melt free surface and the kinetic rate of reaction was weakened at the cover. Finally, the area-average concentration of CO at the melt free surface was decreased by 31%. The GGS was optimized by the orthogonal experiment, including the argon flow rate (Q), length of deflector (L) and distance between GGS and melt free surface (H). When Q = 40 L/min, H = 50 mm and L = 50 mm, the GGS had the most obvious effect on removing impurities.

Keywords Multi-crystalline silicon · Fluid flows · Impurities · Computer simulation · Heat and mass transfer

1 Introduction

Recently, multi-crystalline silicon (mc-Si) and casting seed-assisted mono-crystalline silicon (CSAM-Si) grown by directional solidification (DS) method has dominated 61% of the photovoltaic market due to the low production cost and relative high conversion efficiency [1]. However, as the major impurities in crystalline silicon ingot, oxygen (O) and carbon (C) significantly affect the efficiency of silicon solar cells [2]. High level of O may leads to the formation of O precipitates and thermal donors, which increase light degradation and seriously damage the electrical properties of silicon solar cells [3]. High concentrations of C in silicon can lead to the formation of silicon carbide (SiC), which will not only induce the formation of other defects, but also affect the subsequent slicing process [4]. Therefore, it is essential to control the O and C concentrations for the production of a high-quality casting silicon.

Many researchers have optimized the gas guidance system (GGS) to achieve impurity control in argon. Gao et al. [5] found that the concentration of O and C in the new-type argon tube furnace could be reduced by shortening the distance between the cover and melt free surface. Teng et al. [6, 7] showed that GGS could increase the gas velocity above melt free surface and reduce the concentration of SiO and CO. Bellmann et al. [8] proposed the argon injector which can control the flow in vertical and horizontal directions independently. Besides, horizontal gas injection is better than vertical injection in reducing impurities. Liu et al. [9] investigated that the flow pattern of argon and the distribution of SiO and CO on the melt free surface can be changed obviously by the u-shaped cover. In addition, the variation of process parameters also affect the impurities in argon. Li and Kakimoto et al. [2, 10] showed that the increase in argon flow can play a role in reducing oxygen and carbon impurities. Qi and Teng et al. [11, 12] explored the influence of furnace pressure on the transport of argon impurities.

The argon backflow at crucible outlet and the chemical reaction on the cover can significantly increase the concentration of O and C impurities. In order to suppress the backflow and weaken the kinetic rate of reaction, we designed a unique GGS and analyzed the impurities transport with fully 3D global simulations. Then the GGS was optimized by the orthogonal experimental design (OED). This study provides a possible way to produce high purity casting silicon ingot through the optimal design of the GGS in an industrial DS furnace for solar cell.
2 Model Description

3D global numerical model of G6 DS furnace based on the industrial scale was established by ANSYS Fluent software, including heat radiation, heat conduction, argon gas flow, chemical reaction and mass transfer. The furnace configuration and computational grids are shown in Fig. 1(a). The weight of the grown silicon ingot is 800 kg, and the size is about 1000 × 1000 × 345 mm³. The details of the establishment of this model have been shown in our previous published work [13]. In this paper, unique designed spray-type GGS was added in DS furnace. It has a plurality of side outlet holes and a bottom outlet hole, which is located in the argon region above the free surface of the silicon melt, to optimize the argon gas diversion and reduce oxygen and carbon impurities in the silicon. The structure is shown in Fig. 1(b).

| Case | Factors | Evaluation indexes |
|------|---------|-------------------|
|      | Q (L/min) | H (mm) | L (mm) | Q_{back} (kg/s) | R_{cover} (mol/s) | Y_{co} (ppm) |
| 1    | 20      | 20     | 0      | 6.04 × 10^{-5} | 1.43 × 10^{-7} | 42.1 |
| 2    | 20      | 50     | 50     | 6.08 × 10^{-5} | 1.33 × 10^{-7} | 40.4 |
| 3    | 20      | 80     | 100    | 6.12 × 10^{-5} | 1.25 × 10^{-7} | 43.5 |
| 4    | 20      | 110    | 150    | 5.94 × 10^{-5} | 1.20 × 10^{-7} | 42.8 |
| 5    | 30      | 20     | 50     | 5.18 × 10^{-5} | 1.64 × 10^{-7} | 25.4 |
| 6    | 30      | 50     | 0      | 5.24 × 10^{-5} | 1.49 × 10^{-7} | 26.3 |
| 7    | 30      | 80     | 150    | 5.29 × 10^{-5} | 1.28 × 10^{-7} | 28.1 |
| 8    | 30      | 110    | 100    | 5.23 × 10^{-5} | 1.33 × 10^{-7} | 28.5 |
| 9    | 40      | 20     | 100    | 4.61 × 10^{-5} | 1.81 × 10^{-7} | 24.1 |
| 10   | 40      | 50     | 150    | 4.52 × 10^{-5} | 1.49 × 10^{-7} | 18.6 |
| 11   | 40      | 80     | 0      | 4.59 × 10^{-5} | 1.53 × 10^{-7} | 18.5 |
| 12   | 40      | 110    | 50     | 4.43 × 10^{-5} | 1.46 × 10^{-7} | 18.2 |
| 13   | 50      | 20     | 150    | 3.95 × 10^{-5} | 1.99 × 10^{-7} | 21.4 |
| 14   | 50      | 50     | 100    | 3.88 × 10^{-5} | 1.59 × 10^{-7} | 13.4 |
| 15   | 50      | 80     | 50     | 3.69 × 10^{-5} | 1.67 × 10^{-7} | 14.1 |
| 16   | 50      | 110    | 0      | 3.99 × 10^{-5} | 1.54 × 10^{-7} | 14.6 |
3 Orthogonal Experimental Design

The OED is used to optimize the GGS design parameters, so as to reduce the effect of oxygen and carbon impurities [14, 15]. The core of OED is an orthogonal array composed of various factors and levels. In this paper, argon gas flow rate (Q), height between GGS and the melt free surface (H) and the length of the deflector (L) were determined as factors of the orthogonal experiment and each factor had four levels. It was assumed that any two factors did not interact with each other. Considering the limitation of space, the corresponding levels should not be too large or too small. Therefore, as seen from Table 1, four levels of Q: 20, 30, 40, 50 L/min; H: 20, 50, 80, 110 mm; L: 0, 50, 100, 150 mm have been selected. The evaluation indexes are backflow mass flow rate at crucible outlet (Q_{back}), kinetic rate of reaction on the cover (R_{cover}) and the area-average CO concentration on the melt free surface (Y_{co}). The orthogonal array of the 16 cases is shown in Table 1, designed according to the orthogonal design table L_{16} (4^3).

4 Results and Discussions

4.1 Effects on Flow Pattern

Figure 2 shows the particle tracking of argon gas above the melt free surface with original (a) and new-type (b) GGS.

By contrast, Fig. 2(b) shows the flow field with the new-type GGS. It can be seen that the argon gas is injected vertically from the bottom outlet and horizontally from the side outlets in different directions, respectively. Some part of argon gas ejected from the bottom outlet is first transported to the melt free surface and then flee away from the crucible, which is identical to the traditional way. The other part ejected from side outlets is beneficial for increasing the horizontal velocity and generate a fresh argon flow between the melt free surface and Vortex A, which will decrease the size of Vortex A and increase the distance between the big vortex A and the melt free surface. In this case, the probability of chemical reaction between SiO and carbon is decrease a lot, and the CO and SiO is hardly come back to the melt free surface, it has a relatively better effect on eliminating the impurities.

Figure 3 shows the distribution of argon velocity between point A and B (seen from Fig. 2b) with original and new-type GGS.
crucible outlet and the area of the backflow with the new designed GGS have almost no change. This is because the GGS changes the flow direction of argon, but the temperature and flow fields outside the crucible do not change, so the argon backflow caused by natural convection remains unchanged. However, the flow path of argon has changed obviously after flow into the crucible.

Figure 4 shows the particle tracking of argon flowing into the crucible interior within 10 s with original and new-type GGS. Results show that the distance between the bottom of the vortex and the melt free surface increases with the new designed GGS, and the lateral outflow effectively limits the size of the flow vortex formed by argon backflow above the melt free surface.

4.2 Effects on Chemical Reaction

Figure 5 shows the distributions of kinetic rate of reaction above the melt free surface with original and new-type GGS. The GGS did not affect the argon backflow at crucible outlet, so the area of low kinetic rate of reaction caused by the backflow at the cover has little change. However, the size of the Vortex A above the melt free surface and the concentration of SiO evaporated from the melt to the cover are reduced by the fresh argon gas flow from side outlets, thus reducing the kinetic rate of reaction at the cover.

Figure 6 shows the distribution of kinetic rate of reaction between point C and D with original and new-type GGS. The maximum kinetic rate of reaction between point C and D is $8.1 \times 10^{-7}$ mol/m$^2$·s, which is 52% lower than that of $1.7 \times 10^{-6}$ mol/m$^2$·s with the original GGS. In addition, the maximum reaction rate occurs at the bottom of the GGS, rather than at the junction of the cover and susceptor at diagonal plane, as shown in Fig. 5(b). This is because the SiO evaporated from the melt can react with the bottom of GGS under the influence of diffusion when the distance between the melt free surface and new-type GGS is small, thus it is necessary to further optimize the structure of the GGS.

4.3 Effects on Mass Transfer

Figures 7 and 8 show the distributions of SiO and CO concentration at diagonal plane and midcourt plane with original and new-type GGS. In Fig. 7(b), a fresh argon gas flow ejected from side outlets is limited to a smaller area above the melt free surface compared with the
original GGS. For CO, it is difficult to transport back to the melt free surface by convection and diffusion in the new-type GGS, seen from Fig. 8(b). The maximum concentration of SiO is located at the silicon-gas-crucible junction point for both two GGS, which is due to the less fresh argon gas reaching here. While the maximum concentration of CO is located at the junction of the cover and susceptor due to the accumulation of impurities induced by Vortex B, as shown in Fig. 2(a), which is mainly attributed to edge effects.

Figure 9 shows the distribution of CO on melt free surface with original and new-type GGS. The CO concentration is significantly reduced with the new-type GGS and the area-average CO concentration on the melt free surface is 34.1 ppm, which is 31% lower than that of 49.4 ppm with the original GGS. The newly design GGS can inhibit the chemical reaction at the graphite components and reduce the CO concentration by preventing it from being transported back to the melt free surface. However, the maximum CO concentration at the melt free surface is basically the same and both appear at the crucible side wall, which is due to the restraints of the backflow area around the side outlet, as seen from Fig. 2(b). The backflow still exists near the crucible side wall where the CO impurities are constantly accumulated, resulting in local high CO concentration area.

4.4 Analysis of the Orthogonal Experiment

The range analysis method is used to check the relative importance of each factor. The four ordered degree (OD) values of each factor in the same level \(i\) were summed, and the corresponding average value \(k_i\) and range \(R\) were calculated respectively as follows:

\[
K_i = \frac{\sum OD_i}{4}
\]

\[
R = k_{max} - k_{min}
\]

where \(k_i\) represents the impact of level \(i\) of each factor. The larger the \(R\) of a factor, the greater the effect of the factors on the evaluation indexes; that is to say, this factor is more sensitive to the experimental results.

In this section, we mainly studied the influence of new-type GGS on argon flow and impurity transport by regarding the kinetic rate of reaction, backflow at crucible outlet and CO concentration on the melt free surface as the evaluation indexes. The range analysis of the simulation results is shown in Table 2. It is concluded that the influences of the three factors on mass flow rate of backflow at crucible outlet follows the sequences: \(R_3 > R_1 > R_2\). With the increasing of \(Q\), the flow intensity of argon is increased, thus the flow rate of backflow increased. While \(H\) and \(L\) have little effect on it. In this case, the flow rate of backflow increases linearly with the increase of flow rate in the argon inlet.
The influences of the three factors on kinetic rate of reaction at graphite components follows the sequences: \( R_Q > R_H > R_L \). The intensity of surface chemical reaction is mainly affected by Q and H. The intensity of chemical reaction increases linearly with the increase of flow rate in the argon inlet. This is because with the increasing of flow rate, the flow intensity of argon is increased. Then the SiO evaporated from the melt free surface is more easily transport to the graphite surface, thus the intensity of chemical reaction increased. The chemical reaction intensity decreases first and then remains almost constant with the increase of H. When H is small, SiO evaporated from the melt free surface can easily transport to the graphite parts of the bottom new-type GGS under the effect of diffusion, thus the reaction intensity increased. While the length of deflector L have little effect on chemical reaction.

The influences of the three factors on CO concentration at melt free surface follows the sequences: \( R_Q > R_H > R_L \). Although the chemical reaction intensity increases with the increasing of Q, the backflow is seriously inhibited, which resulting in the reduction of CO concentration at melt free surface. However, the beneficial of additional increasing of Q decreases gradually. As the increase of H, the CO concentration decreases first and then remains unchanged. This is because the little distance will lead to the SiO reacts directly with the bottom of the new-type GGS. While the distance exceeds the concentration boundary layer, the chemical reaction intensity remains unchanged, which resulting in the CO concentration basically unchanged. With the increasing of L, the CO concentration decreases first and then increases. When L is small, the side effluent flows to the melt free surface under the effect of gravity, which cannot effectively decrease the size of Vortex A and increase the distance between the big vortex A and the melt free surface. When L is larger, the deflector will react with SiO evaporated from melt free surface, thus increasing the CO concentration. When \( H = 50 \text{ mm} \), the CO concentration is the lowest. According to the above discussion, the optimum parameters for the new-type GGS were determined as follows: \( Q = 40 \text{ L/min}, H = 50 \text{ mm}, L = 50 \text{ mm} \).
Table 2  Range analysis of the simulation results

| Q_{back} (kg/s) | R_{cover} (mol/s) | Y_{co} (ppm) |
|----------------|------------------|--------------|
| k₁     | 6.04 4.94 4.97 1.30 1.72 1.50 | 42.2 28.3 25.4 |
| k₂     | 5.24 4.93 4.85 1.44 1.47 1.55 | 27.1 24.7 24.5 |
| k₃     | 4.54 4.92 4.96 1.57 1.43 1.49 | 19.9 26.1 27.4 |
| k₄     | 3.88 4.90 4.92 1.69 1.38 1.49 | 15.9 26.0 27.7 |
| k₅     | 2.16 0.04 0.12 0.39 0.34 0.34 | 26.3 3.6 3.2 |
| R      | R_Q > R_L > R_H | R_Q > R_H > R_L | R_Q > R_H > R_L |

5 Conclusions

In order to evaluate the effect of a new designed spray-type GGS on the flow pattern and impurity distribution in an industrial casting silicon furnace, a 3D global model was established including thermal conduction, thermal radiation, argon flow, chemical reaction and mass transfer. The simulation results show that, the intensity of backflow at crucible outlet had no obvious change with the application of this GGS, but the backflow area was effectively inhibited above the melt free surface and the kinetic rate of reaction was weakened at the cover. Finally, the area-average concentration of CO at the melt free surface was decreased by 31%. The GGS was optimized by the orthogonal experiment. When Q = 40 L/min, H = 50 mm, L = 50 mm, the GGS had the most obvious effect on removing impurities.

Acknowledgements The Project is supported by Key Research and Development Program of Jiangsu Province of China (Grant No. BE2019009-003), Industry-University-Research Project (Wuxi Suntech Solar Power Co., Ltd. Grant No. 8421130025). The National Natural Science Foundation for Young Scholars of China (Grant No. 51206069).

Author Contributions Wenjia Su contributed to the conception of the study. Jiulong Li and Chen Li performed the simulation and modified the manuscript. Junfeng Wang helped perform the analysis with constructive discussion.

Funding The Project is supported by Key Research and Development Program of Jiangsu Province of China (Grant No. BE2019009-003), Industry-University-Research Project (Wuxi Suntech Solar Power Co., Ltd. Grant No. 8421130025). The National Natural Science Foundation for Young Scholars of China (Grant No. 51206069).

Data Availability All data generated or analyzed during this study are included in this published article.

Declarations

This manuscript is the authors’ original work and has not been published nor has it been submitted simultaneously elsewhere.

Conflict of Interest There are no conflicts of interest.

Consent to Participate The consent was obtained from individual or guardian participants.

Consent for Publication That all authors have checked the manuscript and have agreed to the submission.

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