International Journal of Heat and Mass Transfer 108 (2017) 2355–2364

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

# Global simulation of coupled oxygen and carbon transport in an industrial directional solidification furnace for crystalline silicon ingots: Effect of crucible cover coating

Lijun Liu [a,*], Wencheng Ma [a], Xiaofang Qi [a], Zaoyang Li [a], Yunfeng Zhang [b]

[a] Key Laboratory of Thermo-Fluid Science and Engineering, Ministry of Education, School of Energy and Power Engineering, Xi'an Jiaotong University, Xi'an, Shaanxi 710049, China
[b] Yingli Green Energy Holding Co. Ltd., Baoding, Hebei 071051, China

ARTICLE INFO

*Article history:*
Received 13 October 2016
Received in revised form 23 January 2017
Accepted 23 January 2017
Available online 6 February 2017

*Keywords:*
Transient global simulation
Oxygen and carbon transport
Impurity segregation
Directional solidification
Silicon

ABSTRACT

For accurate prediction of oxygen (O) and carbon (C) distributions in the crystalline silicon ingots grown by the industrial directional solidification (DS) furnace, we first performed transient global simulations of heat transfer based on a fully coupled calculation of the thermal and flow fields. The phase change problem was handled by an enthalpy formulation technique based on a fixed-grid methodology. The coupled C and O transport in the DS furnace was then carried out, taking into account five chemical reactions. Special attention was devoted to modeling the O and C impurity segregation during the entire solidification process. It was found that the developed model can successfully simulate the impurity segregation at the growth interface and the obtained boundary layer thickness is similar to that estimated by analytical calculation. The effect of the crucible cover coating on the coupled O and C transport was investigated. The numerical results show that the C concentration in the grown silicon ingot can be reduced by about 60% if the pure graphite cover was replaced by a graphite cover with an inert material coating on it. However, the cover coating did not significantly affect the O concentration in the grown ingot. The numerical predictions of the C concentration showed satisfactory agreement with the experimental measurements.

## 1. Introduction

Multi-crystalline silicon (mc-Si) ingot is the dominant material in the photovoltaic market at present and is expected to remain so in the foreseeable future. Directional solidification (DS) method is the leading technique for producing mc-Si ingots owing to its good feedstock tolerance, low production cost and easy operation. However, the DS process is accompanied by the transport of impurities, such as oxygen (O), carbon (C) [1], and their related reactants. O and C impurities in the grown ingots can significantly deteriorate the conversion efficiency of solar cells [2,3]. Effective control of O and C impurities in the silicon ingot is therefore essential to produce high quality solar cells. Numerous studies of the O and C transport in the DS process for mc-Si ingots have been carried out [4–13]. Many of these comprise local simulations [4–7] that just calculate the transport of impurities in the silicon melt. Some global simulations [8–13] take into account the gas transport of impurities. However, these simulations either neglected the C transport in the silicon melt [8–10] or didn't directly include the O and C segregation during the entire solidification process [11–13] so that the final distributions of O and C impurities in the silicon ingot cannot be predicted.

With the aid of the aforementioned numerical models, the effects of several process parameters and local furnace configurations on the impurity transport in the DS furnace have been investigated. Trempa et al. [14] studied the influence of growth rate on the C distribution in the silicon melt and crystal. Li et al. [8] analyzed the effects of argon flow rate and furnace pressure on the evaporation of SiO and concentration of CO at the melt surface in a DS furnace. Nakano et al. [15] pointed out that the heater positions could change the flow direction of the melt near the crucible wall and ultimately affects the O concentration in the melt. Bellmann et al. [16] designed a novel method for gas flow in a DS furnace and numerically evaluated its effect on impurity transport using a global impurity transport model [17]. Gao et al. [18,19] proposed a crucible cover (or a gas flow guidance device) to control the O and C transport in a small laboratory-scale DS furnace. Teng et al. [10,12] extended this idea to a large-size industrial DS furnace and numerically reported that the O and C concentrations in the silicon melt were affected by the position and geometry of the gas flow guidance device. Liu et al. [13] also numerically

---

* Corresponding author.
*E-mail address:* ljliu@mail.xjtu.edu.cn (L. Liu).

http://dx.doi.org/10.1016/j.ijheatmasstransfer.2017.01.092
0017-9310/© 2017 Elsevier Ltd. All rights reserved.

investigated the effect of crucible cover on the O and C transport in the large-size industrial DS furnace and reported that the C concentration in the silicon melt by using a graphite cover can be reduced by two orders of magnitude by using an inert material crucible cover or a graphite cover with an inert material coating on it. However, experimental results for the C concentration in the finally grown silicon ingot did not show the same benefits as those numerically reported [13] when the graphite crucible cover with coating was adopted in the DS furnace. For an accurate prediction of O and C concentrations in the grown ingot in the large-size industrial DS furnace, it is necessary to improve the global models of O and C transport as mentioned above.

In this study, transient global simulations of coupled O and C transport were implemented for the entire environment of an industrial DS furnace. Special attention was focused on modeling of the phase change as well as the O and C segregation during the silicon solidification process. Five chemical reactions were taken into account. The effect of crucible cover coating on the O and C concentrations in the grown ingot was investigated. The numerical results of C concentration in the ingot were compared with the experimental measurements obtained using Fourier transform infrared (FTIR) spectrometry.

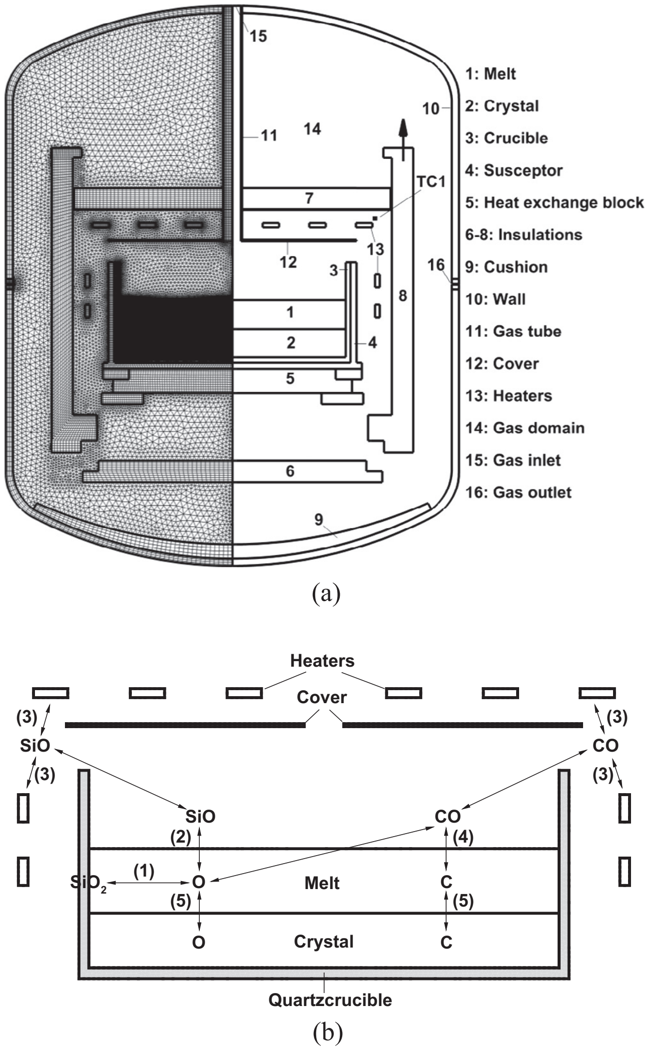

**Fig. 1.** Configuration/computation grids of the industrial DS furnace (a) and pathway of the coupled O/C transport in the furnace chamber (b).

## 2. Experimental setup

Fig. 1(a) shows the configuration of the industrial DS furnace. The DS system mainly consists of quartz crucible, susceptor, heat exchange block, graphite resistance heater, insulations and chamber wall. The silicon feed material is loaded into a quartz crucible with a volume of 84 × 84 × 42 $cm^3$. The height of the silicon ingot is 24 cm. The crucible walls are supported by susceptors to avoid deformation at high temperature. The furnace is well sealed with a water-cooled wall and operates at a low pressure. Inert argon gas is used for purifying the growth environment. Thermocouple 1 (TC1) is installed near the top graphite resistance heater to monitor the temperature, which is used for controlling the heater power. The side insulation, labeled 8 in the figure, moves up in a prescribed way to first activate and then maintain the solidification process. Mc-Si ingots were grown in the same industrial DS furnace with a graphite cover with and without coating, respectively, using the same processing recipe and feedstock in the production line. The substitutional C in the grown ingots was measured by FTIR spectrometry.

## 3. Model description

### 3.1. Global model of heat transfer

A transient global model of heat transfer, taking into account the melt convection, argon flow, solid thermal conduction, thermal radiation, and phase change, was developed for the DS furnace. The entire furnace was equivalently simplified to be axisymmetric and divided into a number of sub-domains, as shown in the right part of Fig. 1(a). To compromise the computational convenience and accuracy, the major assumptions in the global model are as follows: (1) all radiative surfaces are assumed to be diffuse-gray; (2) the silicon melt flow is laminar and incompressible; (3) the Boussinesq assumption is applied to the melt flow; (4) the low Mach approximation and the ideal gas law are applied to the argon gas. The Reynolds number of the argon gas flow in this industrial DS furnace is of the order of 4320 and the standard $k-\varepsilon$ turbulence model [20] was used. It is worth noting that the DS furnace under consideration in this study is in three dimension (3D). However, the silicon melt structure presents some symmetric characteristics in the square shaped crucible except at the corner region [21]. Besides, the thermal field in the silicon domain, the melt-crystal interface shape and the silicon melt flow are dominated by the radial interaction between the silicon and the quartz crucible rather than its asymmetry in the circumferential direction because of the huge difference in the thermal properties of the silicon material and quartz crucible. Therefore, to release computational load, the 3D systems were equivalently simplified to two dimensional (2D) axisymmetric under the condition that the thermal resistance remained unchanged in the DS furnace. This simplification is widely used to study the characteristics of heat and mass transfer in the industrial DS furnaces [12,22–24].

To track the melt-crystal (m-c) interface evolution during the entire solidification process, an enthalpy formulation technique based on a fixed-grid methodology [25] was used to model the phase change problem in the silicon region during the growth process. The governing equations for the silicon domain are expressed in the following forms:

$$\nabla \cdot \vec{u} = 0, \tag{1}$$

$$\rho \frac{\partial \vec{u}}{\partial t} + \rho \vec{u} \cdot \nabla \vec{u} = -\nabla p + \nabla \cdot [\mu(\nabla \vec{u} + \nabla \vec{u}^T)] - \beta \rho \vec{g}(h - h_{ref})/C_p + \vec{S}_u, \tag{2}$$