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Numerical simulation of impurity transport under the effect of a gas flow guidance device during the growth of multicrystalline silicon ingots by the directional solidification process

Ying-Yang Teng [a], Jyh-Chen Chen [b,*], Bo-Siang Huang [b], Ching-Hsin Chang [b]

[a] *Chung Shan Institute of Science and Technology (CSIST), Taiwan, ROC*
[b] *Department of Mechanical Engineering, National Central University, Jhongli 320, Taiwan, ROC*

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ABSTRACT

A numerical simulation has been performed to determine the concentration of oxygen and carbon in a Si melt during the mc-Si ingot growth process under the influence of a gas guidance device. With the application of this gas guidance device, the gas velocity above the free surface may increase, followed by a decrease in the SiO and CO concentrations flux at the free surface. As a consequence oxygen and carbon concentrations in the melt may decrease. The effectiveness of the gas flow guidance device mainly depends on the gap between it and the free surface. A lower oxygen concentration in the melt may be obtained with a smaller gap. On the other hand, the carbon concentration in the melt also decreases as the gap decreases until the gap reaches a certain small value, after which it increases.

1. Introduction

Multicrystalline silicon (mc-Si) is an important material in the PV market with demand growing year by year. The directional solidification (DS) method is the most common method for the production of mc-Si ingots. The DS technique is advantageous for the production of mc-Si crystal/ingots because of its lower production costs and larger crystal/ingot weights [1]. However, the impurity content and concentration affect the quality of the as-grown ingot. Such impurities may be divided into two kinds: metal and non-metal impurities. The phenomenon of minority carrier recombination is caused mainly by metal impurities. To preserve solar cell efficiency, the feedstock must have an extremely low concentration of metal impurities. The most important non-metal impurities are oxygen and carbon. A high concentration of oxygen impurity induces thermal donors in the mc-Si wafer which decrease the electrical performance [2] and also cause a problem with light degradation of the solar cell [3]. The oxygen impurity from the heated quartz crucible and the feedstock [4] is dissolved in the silicon melt and then is either incorporated into the ingot or evaporates as silicon monoxide (SiO) gas at the gas/melt interface [5]. During mc-Si ingot growth, argon gas at a predetermined flow rate passes through the gas pipe to carry the evaporated SiO gas away from the furnace. The SiO gas in the furnace may react chemically with the graphite components to form carbon monoxide (CO) gas. If the CO gas is transferred back to the gas/melt interface, it may dissolve into the melt through the gas/melt surface. The carbon and oxygen impurities (atoms) in the melt may segregate into the crystal or form silicon carbine (SiC) particles.

There have been many research studies examining the transport of oxygen and carbon impurities during the mc-Si ingot growth process. Matsuo et al. [5] proposed an oxygen transport model that included the chemical reaction at the interface between the quartz crucible and the Si_3N_4 liner, and between the Si_3N_4 liner and the Si melt, while studying the heat, flow and oxygen impurity transport in a directional solidification furnace. The Schmidt number Sc represents the relative ability of momentum diffusivity and mass diffusivity, approximately 8 and 6.3 with SiO impurity and 13.1 and 1.6 with CO impurity in argon gas at 300 K and 2000 K, respectively. In other words, the transport of SiO and CO impurities in the argon gas depends strongly on the gas flow motion. Gao et al. [6] performed a numerical simulation to investigate the transfer of oxygen and carbon impurities in a small directional solidification furnace without a cover on the crucible. They have shown that the oxygen and carbon concentrations can be reduced by using a higher argon flow rate and furnace pressure. The reduction in the carbon concentration is more significant for a higher argon flow rate, while the oxygen concentration is more sensitive to a higher furnace pressure. Teng et al. [7] showed the difference in effect of the furnace pressure on the oxygen impurity distribution in mc-Si ingot for different solidification ratios. The application of a gas flow guidance device in the DS furnace leads to a reduction in the oxygen and carbon

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* Corresponding author. Tel.: +886 3425 0476.
E-mail addresses: jcchen@cc.ncu.edu.tw, jcchen@ncu.edu.tw (J.-C. Chen).

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concentration in the silicon melt. Gao et al. [8] showed that the oxygen and carbon concentrations in the crystal can be lowered by the installation of a gas flow guidance device. Teng et al. [9] have further shown that the oxygen concentration in the crystal can be significantly lessened when the vertical distance between the gas flow guidance device and the gas/melt interface, and the gap between the crucible sidewall and the tip of the guidance device decreases. Due to cost considerations, the gas flow guidance device is usually made of graphite [8]. When the gas flow guidance device approaches the gas/melt interface, we can expect that the possibility of CO gas, which originates from the reaction of SiO gas with the graphite, being dissolved into the melt will increase. The returned CO gas may increase the oxygen and carbon concentrations in the melt. Therefore, it is possible that there is an optimum vertical position for placing the graphite gas flow guidance device to obtain the lowest oxygen and carbon concentrations.

Although there have been some numerical studies on DS furnaces with gas guidance devices, we still have only a limited understanding of the physical mechanisms of the SiO and CO gas transfer in the argon gas. The purpose of this study is to clarify the physical mechanisms of the SiO and CO gas transfer in the argon gas, and to find the optimum position for the gas guidance device so as to obtain the lowest oxygen and carbon concentrations.

2. Mathematical model

Fig. 1(a) shows a schematic diagram of the industrial DS furnace used in the present study. The total amount of silicon in the crucible for growing the mc-Si ingots is 240 kg. Fig. 1(b) illustrates the crucible with a gas flow guidance device, where the symbols h and d represent the vertical distance between the guidance device and the free surface, and the gap between the crucible sidewall and the device tip, respectively. A resistance heater is used in this DS furnace.

The furnace is assumed to be axially-symmetric. The Si melt is assumed to be a Newtonian fluid, and the argon gas is considered to be an ideal gas. Since the velocity of argon gas in the DS furnace is less than 1 m/s, the Mach number of the gas in the furnace is less than 3×10^{-3}. Therefore, the viscous dissipation can be neglected. Since the capillary number of the Si melt is smaller than 6×10^{-6}, the effect of surface deformation of the free surface is not considered. The global heat transfer in the furnace during the growth process is computed by an axially-symmetric model. The computations in the furnace involve the radiative heat transfer in the furnace cavity, conduction in the solid blocks, and gas and melt convection. The differential equations governing heat transfer, flow transport, and concentration of impurities are as follows:

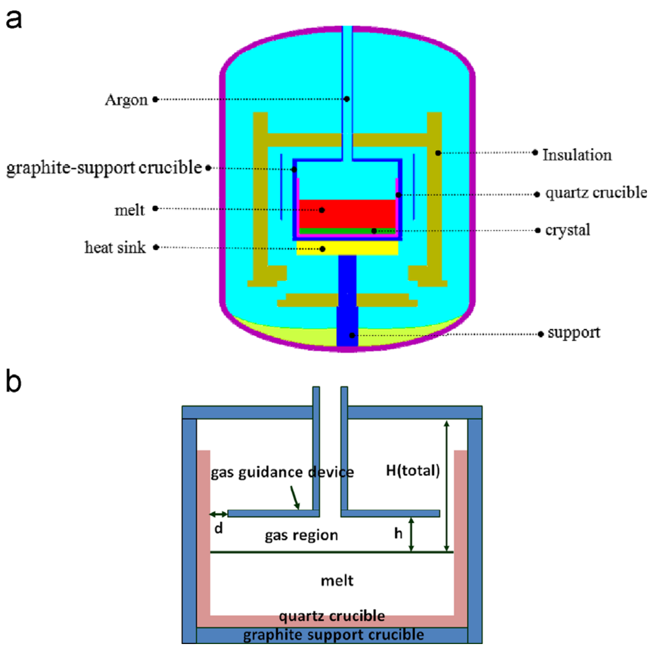

Fig. 1. Schematic diagram of the DSS furnace: (a) conventional furnace and (b) crucible with gas flow guidance device.

$$\frac{\partial\rho_i}{\partial t}+\nabla\cdot(\rho_i\vec{u}_i)=0, \tag{1}$$

$$\frac{\partial(\rho_i\vec{u}_i)}{\partial t}+(\vec{u}_i\cdot\nabla)\rho_i\vec{u}_i=-\nabla p_i+\nabla\cdot f_i+(\rho_i-\rho_{i,0})\cdot\vec{g}, \tag{2}$$

$$\frac{\partial(\rho_i Cp_i T_i)}{\partial t}+\nabla\cdot(\rho_i Cp_i\vec{u}_i T_i)=\nabla\cdot(k_i\nabla T_i), \tag{3}$$

$$\frac{\partial(\rho_i C_j)}{\partial t}+\nabla\cdot(\rho_i\vec{u}_i C_j)=\nabla\cdot(D_j\nabla C_j), \tag{4}$$

$$\rho_g=\frac{p_0 m}{RT} \tag{5}$$

where t, ρ, $\rho_{i,0}$, Cp, $\vec{u}$, T, k, p, f, $\vec{g}$, C, D, p_0, m and R are the time, density, reference density, heat capacity, velocity vector, temperature, thermal conductivity, pressure, stress tensor, gravitational acceleration, concentration, diffusion coefficient, reference pressure, molecular weight and universal gas constant, respectively. Subscript i may be g or l, indicating the argon gas and the melt, respectively. Subscript j may be SiO, CO, O or C, indicating silicon oxide and carbon oxide in the argon gas and the oxygen and carbon impurity in the melt, respectively. The concentration fields for SiO, CO, O or C can be computed by Eq. (4) using the velocity and temperature distributions obtained from Eqs. (1)–(3). The Reynolds average Navier–Stoke equation (RANS) is used with one equation model to describe the effect of turbulent motion in the melt. The normal velocity along the free surface is zero, and the tangential velocity component must satisfy the Marangoni condition.

The wall of the quartz crucible is coated with a Si_3N_4 liner to reduce the dissolution activity of oxygen. Both the chemical reactions at the interface between the quartz crucible and the Si_3N_4 coating, and at the interface between the Si_3N_4 coating and the silicon melt are considered. The boundary conditions indicating the oxygen concentration at the Si_3N_4 liner surface immersed in the silicon melt and the relationship between the oxygen and SiO concentrations at the free surface are adopted from our earlier study [7]. The evaporated SiO gas reacts chemically to carbon monoxide (CO) gas at the hot graphite surface in the furnace. The reaction can be expressed as

$$\mathrm{SiO_{(g)}}+2\mathrm{C_{(s)}}\rightarrow\mathrm{CO_{(g)}}+\mathrm{SiC_{(s)}}, \tag{6}$$

where subscript s indicates a solid. The ratio of equilibrium partial pressure values for CO_g and SiO_g for reaction (6) is adopted from Smirnov and Kalaev [10] and can be expressed as

$$\frac{P_{\mathrm{CO}}}{P_{\mathrm{SiO}}}=\exp\left(\frac{9529}{T}-0.156\right). \tag{7}$$

The conservation of concentration flux of CO and SiO at the graphite surface is

$$C_{\mathrm{Ar,SiO}}=KC_{\mathrm{Ar,CO}}, \tag{8}$$

$$\frac{(\rho D)_{\mathrm{SiO}}}{m_{\mathrm{SiO}}}\cdot\frac{\partial C_{\mathrm{SiO}}}{\partial n}=\frac{(\rho D)_{\mathrm{CO}}}{m_{\mathrm{CO}}}\cdot\frac{\partial C_{\mathrm{CO}}}{\partial n}, \tag{9}$$