Numerical Simulation of Polysilicon Solid-liquid Interface Transmogrification in Heat Transfer Process

Xi Yang 1, Wenhui Ma 2,*, Guoqiang Lv 2, Mingyu Zhang 1,*

1Yunnan Provincial Energy Research Institute Co.Ltd, Kunming 650093, China
2State Key Laboratory of Complex Nonferrous Metal Resources Cleaning Utilization in Yunnan Province/The National Engineering Laboratory for Vacuum Metallurgy, Kunming University of Science and Technology, Kunming 650093, China

Abstract. The shape of solid-liquid interface during the directional solidification process, which is difficult to be observed and measured in actual processes, controls the grain orientation and grain size of polysilicon ingot. We carried out numerical calculations of the directional solidification progress of polycrystalline silicon and invested the means to deal with the latent heat of solidification in numerical simulation. The distributions of the temperature field of the melt for the crystallization progress as well as the transformation of the solid-liquid interface were obtained. The simulation results are consistent with the experimental outcomes. The results show that the curvature of solid-liquid interface is small and stability, larger grain sized columnar crystal can be grown in the laboratory-scale furnace at a solidification rate of 10 μm•s\(^{-1}\). It shall provide important theoretical basis for metallurgical process and polysilicon production technology.

1. Introduction

Solid-liquid interface (s/l) has a decisive effect on the crystal growth and material orientation, which is the prerequisite to get ideal ingot as well as the purpose for analysis and optimization of process parameters. The s/l interface is determined by the heat loss of the horizontal and vertical during the solidification process, usually appear three cases, namely convex, flat and concave. It makes the crystal deviates from the axial growth with not flat interface, result in silicon ingot increase grain boundary, refine grains, and also affect the distribution of impurities on the front edge of solidified, influence the purification effect [1-4]. Therefore, research on heat transfer characteristics of polysilicon DS process to controlling and adjusting suitable s/l interface has important theoretical significance and practical application value. Through experiment results to discuss the solid-liquid interface is a feasible method, but there are still cycle longer, higher cost and process factors beyond their control [5-9]. For this reason, many researchers study by the method of numerical simulation [9-11].

In this paper, we described the s/l interface representation and processing method of latent heat. The transient numerical simulations were carried out to study the DS process of polysilicon based on laboratory DS furnace. The temperature distribution, s/l interface of polysilicon in the DS process was performed and compared with the experimental results.
2. Model descriptions

2.1. System configuration and assumptions
Directional solidification system of polysilicon is shown in Fig. 1. The solidification process can be described as: charge the MG-Si feedstock in a graphite crucible, and melt it by resistance heater; after holding for a period of time, cool the bottom of the crucible and pull the crucible downward slowly with a certain rate while keeping the resistance heater still, such that the molten Si directionally solidifies into crystal upward from the bottom as a result of axial thermal gradient. Since this process involves complicated heat transfer problems, several reasonable assumptions or simplifications are applied in the numerical simulation:

1. all units inside the furnace are columnar, so the model is 2-D axial symmetric;
2. the physical properties of silicon are functions of temperature and are isotropic;
3. the system is vacuum, so the gas convection effect in the furnace is ignored; the heat transfer between crucible exterior and the chamber is through radiation;
4. flow pattern of melt silicon inside the crucible is mainly natural convection caused by density variation and has little effect on the macro solid-liquid interface shape, so such flow of melt silicon is ignored.

2.2. Governing equations and properties
There would be large releases of latent heat accompanying with the phase change of silicon. Currently, main methods to deal with latent heat are temperature recovery method, enthalpy method, and equivalent heat capacity method [12]. Temperature recovery method needs a great deal of storage space and computation time to supervise and treat the temperature of every point during the iteration process; enthalpy method takes enthalpy and temperature as variables to establish conservation equations of liquid region, mushy zone, and solid region simultaneously and obtain the distribution of enthalpy, and then work out the temperature depending on enthalpy of each point. Both these two methods are quiet complicated. In this paper, we choose the simple and effective equivalent heat capacity method which is fit for finite element method (FEM) to calculate the latent releases.

Equivalent heat capacity method replaces released latent heat with equivalent specific heat in the energy equation. The specific heat capacity of silicon is $C_p$, and the latent heat is considered through the change of $\Delta H$. Define a Gaussian curve as follow:

$$\delta = \frac{1}{\Delta T \sqrt{\pi}} \exp\left[-\left(T - T_m\right)^2 / \left(\Delta T\right)^2\right]$$  \hspace{1cm} (1)

where $T_m$ is the melting point, and $\Delta T$ is half of the transition temperature span which denotes the temperature range of mushy zone and was set to 0.01 K in this case since we are treating crystal of pure substance and the grid is very fine. Then we can replace $C_p$ in the heat equation with ($C_p + \delta \Delta H$). There are specific heat changes during phase transition which can be approximated as $\Delta C_p = \Delta H / T$, and
represented by using the software’s built-in smoothed Heaviside step function. The dynamic interface tracking method is implemented by the fraction of liquid phase $B$, given by

$$B = \begin{cases} 1, & T > T_m + \Delta T \\ \frac{(T - T_m + \Delta T)}{(2\Delta T)}, & (T_m - \Delta T) \leq T \leq (T_m + \Delta T) \\ 0, & T < T_m - \Delta T \end{cases}$$ \hspace{1cm} (2)$$

The heat transfer in vacuum directional solidification system is mainly conductive and radiative. The governing equations of the conductive heat transfer in all components can be described as follow:

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k \nabla T) + Q$$ \hspace{1cm} (3)$$

where $\rho$, $C_p$, and $T$ are density, specific heat capacity, and temperature respectively. The parameter $k$ is thermal conductivity, and $Q$ is the heat source term. The radiative heat exchanges between all gray and diffuse surfaces in the furnace were calculated by following equations:

$$(1 - \varepsilon)G = J - \varepsilon\sigma T^4$$ \hspace{1cm} (4)$$

$\varepsilon$ is the emissivity, $G$ is irradiation, $\sigma$ is called the blackbody radiation constant; $J$ is radiosity. In the DS process, the crucible will move downward away from the hot zone, thus the radiative heat transfer between crucible wall and furnace inner surface changes with their relative position, so does the radiation angle factor $f$, which is involved in the calculation of $G$ and $J$. The radiation angle factor between two columnar surfaces is

$$f_{1,2} = \int_A \int_A \frac{\cos \theta_1 \cos \theta_2 dA_1 dA_2}{\pi ((R-r)^2 + (x-z)^2)}$$ \hspace{1cm} (5)$$

which depends on only the relative position of two surfaces: $R$ and $r$ are radius of furnace inner surface and crucible exterior wall, respectively; $x$ and $z$ are heights of furnace inner surface and crucible exterior wall, respectively. The typical thermophysical properties used in the current simulation were provided by manufacturers.

3. Results and discussions

Different pulling-down rates will change the ratio of radial and axial heat dissipation, affect the s/l interface shape, and lead to different solidification state [13]. In our model as shown in Fig. 1, the inner diameter of the crucible was 0.13 m, the charged height of silicon material was 0.15 m. Under the same heating and cooling condition, the crucible was pulled downwards with different rates namely 5 \(\mu\)m/s, 10 \(\mu\)m/s and 15 \(\mu\)m/s. The s/l interface shapes and temperature profiles at different solidification fractions of 0.2, 0.5 and 0.8 under different pulling-down rates were compared and researched, as shown in Figs. 2 ~ 4.
From Figs. 2 ~ 4 we can draw that as the pulling-down rate increased, the temperature gradient inside the silicon material also increased, and the s/l interface shapes varied. The shapes of the s/l interface for 0.2, 0.5 and 0.8 solidification fractions under different pulling-down rates are shown in Fig. 5. According to Fig. 5, under the pulling-down rate of 5 μm/s, the s/l interface was concave in the beginning of solidification, then gradually became flat, and convex; at 0.8 solidification fraction, the convexity was strongly marked; such s/l interface would make the grain growth direction change a lot, which is not conducive to formation of columnar crystal and may even cause grain fracture under the effect of stresses, resulting in grain refinement and defects increase. As for the pulling-down rate of 10 μm/s, the s/l interface was relatively flat in the early stages of the process and then became slightly convex gradually, and the variation as well as the curvature was smaller, as shown in Fig. 5(b); such slightly convex and steady interface shape would provide conditions for grain enlarging and meets the crystal morphology requirement of solar-grade Si. While the pulling-down rate was 15 μm/s, the s/l interface remained a slightly convex shape during the whole solidification process; but it can be seen from Fig. 5(c) that the s/l interface is more convex at the 0.25 solidification fraction than that at the 0.5 solidification fraction, so the s/l interface experience a convex to flat and again to convex change, and this change is not favorable to grain growth yet.
Fig. 5  The solid-liquid interface morphology under different pulling rate:

(a) 5 μm/s, (b) 10 μm/s, (c) 15 μm/s

A pilot-scale vacuum DS furnace was used to produce polysilicon with different pulling-down rates of 5 μm/s, 10 μm/s and 15 μm/s based on the above simulation results. The experimental s/l interface shapes were detected by analyzing the grain morphology in the longitudinal section of the polysilicon ingot, as shown in Fig. 6.

Fig. 6  The crystal morphology of the silicon ingots in a longitudinal sectional at different pulling-down rates: (a) 5 μm/s, (b) 10 μm/s, (c) 15 μm/s

It can be found from Figs. 6 that silicon ingots produced by different pulling-down rates are quite different in crystal morphology and grain direction. The grains are refined at the beginning of the solidification and the crystals grow from the bottom to top in a divergent shape for 5 μm/s along the normal direction of its s/l interface based on thermal field (Fig. 5(a)), thus there are a lot of fine grains grown from the crucible bottom in the silicon ingots. For crystal grown under 10 μm/s, its crystal grains grow bigger along the growth direction, and much less grains grow from the sidewall due mainly to more desirable s/l interface (Fig. 5(b)). What can be noticed firstly in Fig. 6(c) is that growth direction is divergent but average grain sizes in the Si ingot are minimized due to the s/l interface as show in Fig. 5(c). The experimental outcomes shown in Fig. 6 and the above simulation results are in accordance with each other, indicating that such numerical model can reflect the invisible solidification process visually.

4. Conclusion

Time-dependent numerical calculations of the vacuum directional solidification progress of polycrystalline silicon are carried out and compared with experimental results in this paper. The method to deal with the latent heat of solidification in numerical simulation are validated, which indicates that numerical simulation can reflect straightforward the solid-liquid interface transmogrification in polysilicon directional solidification process. The results show that the curvature of solid-liquid interface is small and stability, and larger grain sized columnar crystal can be grown in
the laboratory-scale furnace under a solidification rate of 10 \( \mu \text{m/s} \). Such simulation method is suitable for researching the s/l interface features of polycrystalline silicon and other metallic material in directional solidification process.

References

[1] Mei X. Y., Ma W. H., Wei K. X., et al. Columnar crystals of multi-crystalline silicon in directional solidification [J]. Acta Scientiarum Naturalium Universitatis Sunyatseni, 2009, (48) 2:89-91.

[2] Guo T. M., Li C. X. Characteristics of irregular interface pattern during unidirectional solidification [J]. Journal of Synthetic Crystals, 2003, 32 (5):495-501.

[3] Yuge N., Hanazawa K., Kato Y. Removal of Metal Impurities in Molten Silicon by Directional Solidification with Electron Beam Heating [J]. Materials transactions, 2004, 45 (3):850-857.

[4] Kvande R., Mjøs Ø., Ryningen B. Growth rate and impurity distribution in multicrystalline silicon for solar cells [J]. Materials Science & Engineering A, 2005, Vol.413:545-549.

[5] Liu Q. D., Lin A. Z., Lin X. B. Preparation of polysilicon ingot and study on its morphology and microstructure [J]. Chinese Rare Metals, 2002, 26 (06): 416-419.

[6] Tan Y, Sun S. H., Dong W., et al. Research of solid-liquid interface property during directional solidification process for multicrystalline silicon [J]. Journal of Materials Engineering, 2012, (08):33-38.

[7] Chen G. H., Wang Z. Y., Xiao Y.B., et al. Numerical simulation of directional solidification process for the polysilicon ingot [J]. Equipment for Electronic Products Manufacturing, 2009, (173):40-46.

[8] Luo Y. F., Hu Y., Zhang F. Y., et al. Research on numerical simulation for the directional growth of polycrystalline silicon [J]. Foundry Technology, 2011, (32)10:1368-1371.

[9] Ma X., Zheng L. L., Zhang H, et al. Thermal system design and optimization of an industrial silicon directional solidification system [J]. Journal of Crystal Growth, 2011, 318 (11):288-292.

[10] Shur J. W., Kang B. K., Moon S. J., et al. Growth of multi-crystalline silicon ingot by improved directional solidification process based on numerical simulation [J]. Solar Energy Materials & Solar Cells, 2011, 95 (12): 3159–3164.

[11] Chen L. G., Dai B. Optimization of power consumption on silicon directional solidification system by using numerical simulations [J]. Journal of Crystal Growth, 2012, 354 (11):86–92.

[12] Gao X. Different processing modes of latent heat in temperature field numerical simulation [J]. Xinjiang Youse Jinshu, 2011, 34 (z1):105-107.

[13] Chang C. E., Wilcox W. R. Control of interface shape in the vertical bridgman-stockbarger technique [J]. Cryst. Growth, 1974, (21)1:135–140.