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Control of melt-crystal interface shape during sapphire crystal growth by heat exchanger method

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ABSTRACT

We numerically investigate the melt-crystal interface shape during the early stage of the solidification process when the crystal diameter increases. The contact angle between the melt-crystal interface and the crucible bottom wall is found obtuse during this stage, which is unfavorable for the crystal quality. We found that the obtuse contact angle is caused by the thermal resistance difference between the sapphire crystal and melt as well as the insufficient cooling effect of the crucible bottom. Two approaches are proposed to suppress the obtuse contact angle. The first approach is to increase the emissivity of the outer surface of crucible bottom. The second approach is to install a heat shield near the crucible bottom. The reduction of the emissivity of the heat shield is also favorable for the suppression of the obtuse contact angle. Compared with the increase of the emissivity of the crucible bottom, the installation of a heat shield is a more effective approach to prevent the appearance of an obtuse contact angle for the sake of reliability since a molybdenum heat shield can be reused and will not induce other impurities.

1. Introduction

Sapphire crystal has been widely used in diverse fields, such as silicon-on sapphire, RF devices, optical window materials, active luminescent media and substrates for LEDs [1,2]. With the booming development of LED market in recent years, the demand of sapphire crystal has increased rapidly. There are many methods to produce sapphire crystal in industry, such as the Czochralski (Cz), Kyropoulos (KY), edge-defined film-fed growth (EFG) and heat exchanger methods (HEM) [3]. Among these methods, HEM is very promising since it can produce large-size crystals with low dislocation density and bubbles because of the low temperature gradient during solidification. Moreover, the controlled heat extraction system (CHES) which is a modification of HEM, can directly produce sapphire crystal in c-axial direction. Therefore, the yield of the wafers of the target orientation is larger because of geometric efficiency gains [4].

During the sapphire crystal growth by HEM method, the temperature distribution, melt flow and melt-crystal interface shape in the solidification process have important effect on the crystal quality since they determine the thermal stress, dislocation and impurity distributions in the crystal. The temperature distribution, melt flow and melt-crystal interface shape in the solidification process of HEM method have been studied by many researchers. Lu and his coworkers developed 2D [5] and 3D [6] local models to investigate the heat transfer in the HEM furnace for sapphire growth. They also investigated the effect of the anisotropic conductivity of the sapphire crystal [7] on the melt–crystal interface shape. Xiong et al. [8] designed a modified HEM furnace combining the advantages of the traditional HEM systems and Bridgman systems, and simulated the growth process of optical crystals with a transient model. Zhang et al. [9] established quasi-steady-state and transient models to investigate the effects of transport mechanisms and cold finger design in a modified HEM system. Hoshikawa and his coworkers [10] used numerical simulation to guide the growth of c-axial sapphire by vertical Bridgman method, which is very relevant with HEM. They also investigated the formation of low-angle grain boundaries [11,12] and metallic inclusions [13] by experiments. Liu et al. [14] developed a transient global model to study the heat transfer in sapphire crystal growth by HEM method and validated their results by experiment. They also investigated the effect of crucible cover [15] and crucible location [16] on heat transfer during crystal growth.

The solidification process of the HEM method has two stages: the early stage and the later stage. In the early stage, the diameter of the sapphire crystal enlarges when growing. In the later stage, the diameter of the crystal keeps constant. Most of the existing studies [6,8,14–16] focus on the later stage since the major part of the crystal is grown in

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this stage. However, the early stage is also important since the dislocations generated in this stage can extend to the whole crystal. Few studies have focused on the early stage of the solidification process during sapphire crystal growth by HEM method. Viechnicki et al. [17] found that the crucible corner is a "hot spot", and that the melt in this region could not solidify until the end of the solidification process. Therefore, the dislocations in this region increased. Lu et al. [5,18] found that the contact angle between the melt-crystal interface and the crucible bottom was always obtuse during the early stage of the solidification process, resulting in the "hot spot" at the crucible corner. They applied a crucible with curved base to suppress the appearance of the "hot spot" [19]. However, their model is quasi-steady and only the crystal, melt and crucible were included in the computation domain. Zhang et al. [9] investigated the heat transfer mechanism for the early stage of growth. However, their simulation model is quasi-steady and the applied system is small-scale. Therefore, it is necessary to conduct further investigation on the early stage of the solidification process during sapphire crystal growth by HEM method.

In this study, the early stage of the solidification process is numerically investigated by a transient global model. The purpose of our study is the control of the melt-crystal interface shape during the early stage of the solidification process.

2. Model description

The schematic of an HEM furnace for sapphire crystal growth is shown in Fig. 1 (left side). The furnace mainly consists of a tungsten crucible, a resistant heater, a heat exchanger and insulations. The diameter of the grown crystal is 275 mm, and the height is 200 mm. A helium-cooled heat exchanger is placed under the crucible bottom to extract heat from the crystal and melt domains. The insulations are made up of parallel-placed molybdenum slices to prevent heat loss and are considered as a solid with effective conductivity in the simulations. The chamber is charged with argon gas at a very low pressure and the chamber wall is cooled by water.

We developed a transient global model to study the heat transfer in sapphire crystal growth by HEM method [14]. The numerical simulations are conducted for a 2D cylindrical system. The assumptions are the same as that in our previous work [14]. In order to predict the melt-crystal interface, an enthalpy formulation based on fixed grid methodology is developed for the numerical solution of this phase-change problem [20]. The governing equations for the thermal and flow fields in the sapphire domain are as follows:

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

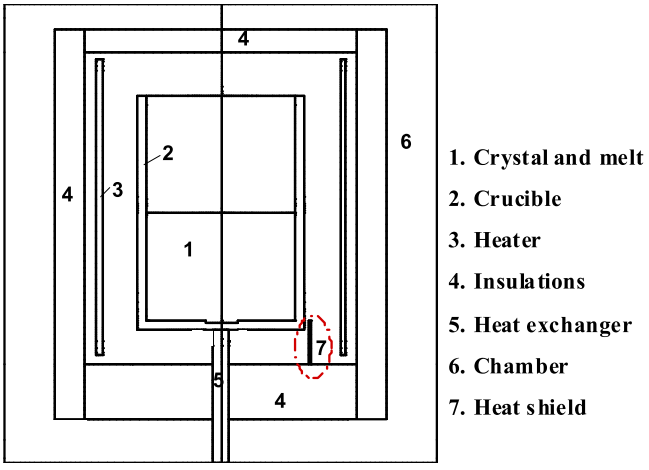

Fig. 1. Schematic of the original (left) and modified (right) HEM furnaces.

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

$$\rho\frac{\partial h}{\partial t} + \rho\vec{u}\cdot\nabla h = \nabla\cdot(\frac{k}{C_p}\nabla h) + \dot{q}_{rad} + S_h \tag{3}$$

For the other solid components, the energy conservation equation can be expressed as follows:

$$\rho C_p\frac{\partial T}{\partial t} = \nabla\cdot(k_s\nabla T) + \dot{q}_s \tag{4}$$

In Eqs. (1)–(3), $\vec{u}$ is the velocity, p is the pressure, μ is the dynamic viscosity, $\vec{g}$ is the gravity acceleration vector, β is the thermal expansion coefficient, k is the thermal conductivity, h is the sensible enthalpy with $h = C_pT$, and h_{ref} is the reference sensible enthalpy, while $\dot{q}_{rad}$ is the heat source caused by radiation. In Eq. (4), $\dot{q}_s$ is the heat generation rate per unit volume, which equals zero for all solid components except the heater, and k_s is the solid component thermal conductivity.

In addition, $\vec{S}_m$ is an artificial source term in the momentum conservation Eq. (2) whose form is as follows:

$$\vec{S}_m = \frac{C_1(1-f_l)^2}{f_l^3 + C_0}\vec{u} \tag{5}$$

where C_0 is a small constant (10^{-3}) and C_1 is a big constant (10^6). f_l is the liquid fraction in each cell defined as:

$$f_l = \begin{cases} 0 & T < T_m \\ 1 & T \geq T_m \end{cases} \tag{6}$$

where T_m is the melting point of sapphire. In the melt region, $\vec{S}_m$ equals zero and consequently has no influence on the melt flow. In the crystal region, the source term $\vec{S}_m$ dominates all other terms in the momentum conservation equation. Therefore, the velocity $\vec{u}$ approaches zero. In this way, the source term $\vec{S}_m$ is used to guarantee zero velocity when a cell in the sapphire domain is fully crystallized.

Then in the Eq. (3), S_h is the phase related source term described as:

$$S_h = -\frac{\partial}{\partial t}(\rho\Delta H) \tag{7}$$

where ΔH is the latent heat expressed as:

$$\Delta H = f_l L \tag{8}$$

where L is the latent heat of phase change. Therefore, the source term S_h in the energy conservation equation represents the effect of latent heat on heat transfer.

Temperature continuity and heat flux conservation constitute the thermal boundary conditions between any two sub-domains. For the outer surface of the crucible bottom, the thermal boundary condition can be expressed as:

$$-k_g(\nabla T)_g\cdot\vec{n} + k_{cru}(\nabla T)_{cru}\cdot\vec{n} + \vec{q}_{rad}\cdot\vec{n} = 0 \tag{9}$$

$$T_g = T_{cru} \tag{10}$$

where subscripts g and cru represent gas (helium or argon) and crucible, respectively. $\vec{n}$ is the unit normal vector at the boundaries. $\vec{q}_{rad}$ is the radiative heat flux at the gas side. For the interface between the crucible bottom and the sapphire region, the thermal boundary condition is given as:

$$-k_{sap}(\nabla T)_{sap}\cdot\vec{n} + k_{cru}(\nabla T)_{cru}\cdot\vec{n} + \vec{q}_{rad}\cdot\vec{n} = 0 \tag{11}$$

$$T_{sap} = T_{cru} \tag{12}$$

where subscript sap denotes sapphire. $\vec{q}_{rad}$ is the radiative heat flux at the sapphire side. The other boundary conditions of the thermal and flow fields and the evolutions of the gas cooling flow rate and the heating power are the same as that in our previous work [14]. The physical properties of the constituents in the HEM furnace are shown