ELSEVIER

Available online at www.sciencedirect.com

SCIENCE @ DIRECT®

Journal of Crystal Growth 247 (2003) 597–612

JOURNAL OF
**CRYSTAL
GROWTH**

www.elsevier.com/locate/jcrysgro

Three-dimensional simulation of floating-zone crystal growth of oxide crystals

C.W. Lan*

Department of Chemical Engineering, National Taiwan University, Taipei, Taiwan 10617, ROC

Received 20 June 2002; accepted 9 October 2002

Communicated by D.T.J. Hurle

Abstract

The floating-zone (FZ) method is a popular technique for the growth of high-temperature oxide crystals. However, the growth usually requires skillful control of the zone stability, which is strongly coupled with heat flow, interfaces, and the grown crystal morphology. In this report, we present a three-dimensional self-consistent simulation of floating-zone oxide growth in a mirror furnace by taking these factors into account simultaneously. This model is based on an efficient finite volume method with multigrid acceleration and interface tracking. The steady growth of a YAG crystal in a double-ellipsoid mirror furnace is taken as an example to illustrate the intricate coupling of convection, interfaces, meniscus, and the grown crystal shape.

© 2002 Elsevier Science B.V. All rights reserved.

PACS: 44.25.+f; 47.27.Te; 81.10.Fq; 02.60.c6; 02.70.Fj

Keywords: A1. Computer simulation; A1. Convection; A1. Crystal morphology; A1. Fluid flows; A1. Heat transfer; A2. Floating zone technique

1. Introduction

The floating-zone (FZ) method is a popular technique for growing single crystals. Particularly, it is a crucibleless process, so that the contamination from the crucibles could be avoided. For oxides and semiconductors that absorb infrared, the use of light heating is convenient and efficient. However, the heating, especially through double ellipsoid mirrors, is in general asymmetric, and can lead to asymmetric molten zone as well as a non-

uniform temperature distribution. Although rotation of the feed rod and the crystal improves the zone uniformity, the heat flow remains to be three-dimensional (3D), even with an axisymmetric heating [1]. The 3D zone shape is strongly affected by heating power and convection, as well as the intricate coupling of these factors with interfaces. Indeed, maintaining a stable zone is always required for a successful growth, and this is often regarded as the most difficult task for the FZ technique. Another problem for FZ growth is the high thermal gradient along the free surface. Significant Marangoni flow can be induced leading to unstable heat flows and highly distorted

*Tel./fax: +886-2-2263-3917.

E-mail address: cwlan@ntu.edu.tw (C.W. Lan).

interfaces, even in the microgravity environment [2,3]. Severe growth striations due to the unstable flow are typical, which are believed to be the cause of the unstable growth rate (even with back melting) [1]. Computer simulation has been an effective way to get a deeper insight of the interplay heat flow and non-linear phenomena, e.g. [4–7]. However, 3D simulation for FZ growth considering all these factors, particularly the zone shape, interfaces, and the grown crystal morphology, in a self-consistent manner has not yet been reported before.

Most of the numerical models for FZ crystal growth are two-dimensional, e.g. [8–12]. Duranceanu and Brown [8] first introduced the conduction thermocapillary model in a self-consistent manner. Lan and Kou [9] further considered melt convection showing that the highly convex interfaces are induced by the Marangoni flow. In their latter work, the effects of rotation were also considered and compared with experimental observations [10]. From there, high-speed rotation was proposed for interface inversion. The effects of magnetic fields on dopant transport were also considered by Lan [11], in which the non-uniform magnetic fields and multiple steady states were investigated. The modeling of the needle-eye configuration was conducted by Mühlbauer et al. [12], and this excellent work could be regarded as a milestone for 2D FZ simulation considering most of the physical features, even including the wetting line of the meniscus with the feeding rod. Lan and Liang [13] further extended the thermocapillary model to 3D; however, the melt convection was not considered. Their model was also integrated with a finite lamp model to simulate the 3D heat transfer in a double-ellipsoid mirror furnace [14]. Recently, the 3D simulation of time-dependent Marangoni convection has been carried out [15,16]. The coupling of time-dependent convection with the growth rate has also been considered, where back melting due to rotational oscillatory Marangoni flow was also illustrated [15]. Ratnieks et al. [16] conducted 3D simulation using the commercial CFD-code FLUENT for the needle-eye configuration, where the interfaces and the meniscus were taken from their 2D self-consistent model [12]. The effect of rotation on growth striations was investigated, and good agreement

with experiments was found. Unfortunately, so far, the 3D meniscus and the grown crystal shape have not yet been taken into account simultaneously in the 3D model. Since the FZ growth is a meniscus-controlled process, to model the growth in a self-consistent manner, the 3D zone, the growth angle, and the crystal morphology cannot be ignored in the simulation. However, such a simulation has not been reported.

In this report, we present a 3D self-consistent FZ model and illustrate some simulation results for YAG growth in a double-ellipsoid mirror furnace. The steady heat flow, interfaces, meniscus, and the crystal shape are simulated simultaneously. The effects of convection modes and growth orientation considering the anisotropic growth angle on the crystal shape are illustrated. In Section 2, the mathematical model and numerical solution are described briefly. Section 3 is devoted to results and discussion, where some benchmark comparisons with previous models are performed. Conclusions and comments are drawn in Section 4.

2. Mathematical model and numerical simulation

A generic FZ crystal growth in a double-ellipsoid mirror furnace is illustrated in Fig. 1; the mesh used for calculation (after convergence) is also illustrated. Since axisymmetry is no longer assumed here, the system is described by a Cartesian coordinate (x, y, z) . The point-source model [11,17,18] for the mirror furnace is adopted for the heat input condition. Other types of heating, e.g. [8,9] can be implemented easily as well. This furnace model is simple and consistent, which can capture most of features of lamp heating, even though it considers only primary-light reflection. The feed rod is steadily moving downward at speed U_f , while the crystal pulling speed is U_c . For a pseudo-steady state growth, for the same density of the feed and the crystal, $A_f U_f = A_c U_c$, where A_f and A_c are the cross section area the feeding rod and the crystal, respectively. However, the shape of the grown crystal, which can be represented by a local radius $R_c(\theta)$, remains unknown and needs to be calculated; θ is the

Fig. 1. Schematic of floating-zone crystal growth in a double-ellipsoid mirror furnace; a sample mesh for simulation is also shown; a small portion of the melt is removed to illustrate the mesh inside the molten zone.

azimuthal angle. Furthermore, the flow and temperature fields, as well as the melt/feed ($h_f(y, z)$) and melt/crystal ($h_c(y, z)$) interfaces, and the meniscus ($R_m(x, y, z)$) are also represented in the Cartesian coordinate.

The dimensionless variables are defined by scaling length with the feed rod diameter D_f , velocity with α_m/D_f , pressure with $\rho_m \alpha_m^2/D_f^2$, and temperature with the melting point T_m , where α_m is the thermal diffusivity and ρ_m the melt density. The steady-state governing equations describing the convection and heat transport in the melt are required as follows:

$$\nabla \cdot \mathbf{v} = 0 \quad (1)$$

$$\mathbf{v} \cdot \nabla \mathbf{v} = -\nabla P + Pr \nabla^2 \mathbf{v} - Pr Ra_T (T - 1) \mathbf{e}_g, \quad (2)$$

$$\mathbf{v} \cdot \nabla T = \nabla^2 T, \quad (3)$$

where $\mathbf{v}$, P and T are the dimensionless velocity, pressure, and temperature, respectively. Also, $Pr = \nu_m/\alpha_m$ is the Prandtl number, where ν_m is the kinematic viscosity. The gravity direction $\mathbf{e}_g$ is the unit vector of gravity, which can be aligned ($\alpha = 0$) with or tilted ($\alpha > 0$) from the growth axis on the x - y plane, as shown in Fig. 1. Furthermore, T_m also serves as a reference temperature. The

associated dimensionless number Ra_T in the source term of Eq. (2) is the thermal Rayleigh number, defined as follows:

$$Ra_T = \frac{\beta_T g T_m D_f^3}{\nu_m \alpha_m},$$

where β_T is the thermal expansion coefficients and g the gravitational acceleration. In the crystal (c) and the feed rod (f) only heat transfer needs to be considered:

$$(\mathbf{v}_i \mathbf{e}_x + r \Omega_i \mathbf{e}_\theta) \nabla T = \nabla \cdot \kappa_i \nabla T, \quad i = (c, f) \quad (4)$$

where r is the radial distance, Ω_i the rotation speed, and $\kappa_i = \alpha_i(T)/\alpha_m$ the dimensionless thermal diffusivity of feed and crystal; α_i is the thermal diffusivity of the feed rod ($i = f$) or the crystal ($i = c$). Also, $\mathbf{e}_x$ and $\mathbf{e}_\theta$ are the unit vectors in the axial and azimuthal angle directions, respectively. In this study, rotation is not considered ($\Omega_i = 0$), because the cross section of the grown crystal is not round, and time dependent heat flow will be induced by rotation. Nevertheless, we shall discuss the effect of rotation in the near future. In addition, most of the oxide crystals are often quite transparent to infrared. Therefore, internal radiation cannot be ignored. We have chosen the simplest model, i.e., the no-slip Rosseland

diffusion model used by Vizman et al. [19], for the crystal. Because the feed rod is usually prepared by sintering, the optical distance is small so that opacity is assumed; the melt is assumed to be opaque as well. One may consider a better model for the internal radiation [20], but it requires more computational effort, which is not the major interest of this study. Nevertheless, based on the same computer code, we can add a more sophisticated model for radiation if necessary.

The no-slip condition is used for the velocity at solid boundaries. At the free surface, the shear stress balance is imposed:

$$\boldsymbol{\tau} : \mathbf{n}\mathbf{s} = Ma\partial T/\partial s, \quad (5)$$

where $\boldsymbol{\tau} : \mathbf{n}\mathbf{s}$ is the shear stress at the n - s plane of the free surface; $\mathbf{n}$ and $\mathbf{s}$ are the unit normal and tangential vectors at the free surface, respectively. Also, Ma is the Marangoni number, which is defined as:

$$Ma = \frac{|\partial\gamma/\partial T| T_m D_f}{\rho_m \nu_m \alpha_m},$$

where $\partial\gamma/\partial T$ is the surface-tension-temperature coefficient of the melt. Two tangential directions need to be considered for the stress balance. In addition, the kinematic condition ($\mathbf{n} \cdot \mathbf{v} = 0$) at the free surface and the normal stress balance (the Young–Laplace equation) are also satisfied, i.e.,

$$\boldsymbol{\tau} : \mathbf{n}\mathbf{n} = (2H)Bo + \lambda_0, \quad (5a)$$

where $2H$ is the mean curvature, $Bo = \gamma/(\rho_m g D_f^2)$ is the static Bond number; γ is the surface tension of the melt. The detailed way for calculating the mean curvature can be found elsewhere [13]. Also, λ_0 is a reference head that needs to be determined to satisfy the growth angle constraint for the steady growth or the global mass conservation for stationary melting. For the steady growth, the growth angle constraint needs to be satisfied at the melt/gas/crystal tri-junction, i.e.,

$$\mathbf{n}_m \cdot \mathbf{n}_c = \cos(\phi_0(\theta)), \quad (6)$$

where $\mathbf{n}_m$ and $\mathbf{n}_c$ are the unit vectors at the melt and crystal surfaces, respectively, at the tri-junction line $\phi_0(\theta)$ is the growth angle for the growing crystal having a constant local radius, which can also depend on the crystallographic

orientation. During calculation, the radius and the shape of the crystal are not known *a priori*. Therefore, the growth angle constraint is used for the calculation of the local crystal radius $R_c(\theta)$. However, the static head λ_0 is used to satisfy the global mass conservation, $A_c = A_f U_f / U_c$.

The thermal boundary conditions at the growth and feeding fronts are set by the heat flux balances:

$$\begin{aligned} Q_{|m} - Q_{|f} + \gamma_c [v_i \mathbf{e}_x - r \Omega_i \mathbf{e}_\theta] St \cdot \mathbf{n} &= 0, \\ i &= (c, f), \end{aligned} \quad (6a)$$

where $\mathbf{n}$ is the unit normal vector at the feeding or growth interface pointing to the melt. $Q_{|m}$, $Q_{|c}$, and $Q_{|f}$ are the dimensionless total heat fluxes at the melt, the crystal, and the feeding sides, respectively. Also, γ_c is the density ratio of the crystal to the melt. The Stefan number $St = \Delta H / C_{p_m} T_m$ scales the heat of fusion ΔH released during solidification to the sensible heat in the melt; C_{p_m} is the specific heat of the melt. The component for $(r \Omega \mathbf{e}_\theta \cdot \mathbf{n})$ is particularly important for 3D interfaces during rotation. Clearly, with a normal rotation speed, the melting ($\mathbf{e}_\theta \cdot \mathbf{n} > 0$) and growth ($\mathbf{e}_\theta \cdot \mathbf{n} < 0$) rates could be larger than the pulling rate if $(\mathbf{e}_\theta \cdot \mathbf{n})$ is not trivial. Furthermore, this term also increases the interface distortion along the rotational direction, which can further reduce the asymmetry. Although we do not consider rotation in this study, some discussion and experiments can be found for a similar configuration [21]. The heat transfer between the sample surface and the surrounding is similar to that in the 2D case described before [11].

The above governing equations and their associated boundary conditions can only be solved numerically. We have developed an efficient finite volume method (FVM) scheme using the primitive variable formulation and multigrid acceleration [22] for the free and moving boundary problems. In the present study, both for NaNO_3 and YAG, steady-state results are always obtained. Two levels of grids are used. The overall iteration scheme for getting a steady-state solution is straightforward. Temperature and velocity calculations are placed in the inner iterations. After having a number of inner iterations, we find the interface shape by locating the melting point isotherm, which also provides the position for

the upper and lower tri-junction rims. Then, the meniscus is calculated based on the normal stress balance, in which there is an outer iteration loop for moving the local radii and getting the reference pressure to satisfy the growth angle or constant mass constraint and the overall mass conservation. The iterations continue until all variables converge. Usually, the total inner iteration number for the fine grid is about 3000 to reduce the relative errors about five orders. This also includes about 120 iterations for the interfaces. One calculation takes about 2 h of CPU time in a Pentium-4 (2 GHz) PC. A sample converged mesh for calculation is shown in Fig. 1. As shown, finer grid spacing is placed near the interfaces to enhance the accuracy of calculation. Detailed description of the numerical method can be found elsewhere [22].

3. Results and discussion

3.1. Benchmarking and heating flux distribution

Before presenting the results, we have performed extensive benchmark comparisons with previous works for a self-consistent 2D model and a simplified 3D model. The FZ melting of a 4-mm-diameter NaNO_3 rod is used for the comparison with the 2D model. The sample length is 4 cm and its both ends are insulated. The same Gaussian-type temperature distribution is used as that in [23]; constant mass constraint is used for calculation. The comparison is summarized in Table 1 for the maximum melt temperature and the outer zone length. As shown, they are in good agreement. More importantly, our 3D solution is perfectly axisymmetric for the 2D cases. For the simplified 3D model, the upper and lower interfaces are

assumed flat and fixed, and the half-zone configuration is considered for silicon with an aspect ratio of one and $Pr = 0.01$; the free surface is adiabatic. Detailed physical properties used are similar to that in Ref. [5], but melt viscosity is adjusted slightly to match the Prandtl number (0.01). Good agreement is found as well. The detailed comparison for the onset of symmetry breaking and oscillatory solution has been reported previously [15]. However, for the oxide growth, both NaNO_3 and YAG, we have not yet found any time-dependent flows. This is owing to their larger melt viscosity and higher Prandtl number as compared with silicon. After building the confidence for the computer code, the following calculations are performed for YAG crystals. The physical properties and some input parameters are listed in Table 2 [24].

For a sample calculation with lamp power of 550 W (each lamp), the calculated heat flux distribution from the lamps on the sample surface and the resulted surface temperature distribution are shown in Figs. 2a and b, respectively. Because of the point-source and primary-reflection assumptions, the heat flux distribution on the sample is very steep. Most of energy is deposited on the molten zone facing to the lamps. As a result, the temperature gradient in the axial direction, especially near the melt/solid interface is very high being about 1513 K/cm in the crystal side. For a finite lamp size, the heating will be more diffusive [14]. Meanwhile, the higher-order reflections could further reduce the thermal gradients [25,26]. To compensate the higher-order reflections, we have purposely used an effective ambient temperature that is much higher than the mirror temperature, which is close to room temperature, for simulation. As will be discussed shortly, the superheating in the molten zone is very high being about 439°C

Table 1
Comparison of 2D and 3D calculations for the FZ melting of a 4-mm NaNO_3 rod.

	Conduction only		Buoyancy flow		Buoyancy + Marangoni flows	
	(K)	(mm)	(K)	(mm)	(K)	(mm)
2D model	616.53	3.726	599.16	4.04	589.27	4.74
3D model	616.12	3.73	599.53	4.06	589.30	4.64

Table 2
Physical properties and some input parameters [24]

<i>YAG</i>
$\rho_c = 3.685 \text{ g cm}^{-3}$
$\rho_m = 4.3 \text{ g cm}^{-3}$
$a_R = 5 \text{ cm}^{-1}$ (Rosseland absorption coefficient of the crystal)
$n = 1.8$ (refractive index of the crystal)
$T_m = 1970.0^\circ\text{C}$
$\Delta H = 455.5 \text{ J g}^{-1}$
$h = 1.1 \times 10^{-3} \text{ W cm}^{-2}^\circ\text{C}^{-1}$ (heat transfer coefficient)
$k_c^{\text{cond}} = 0.1 \text{ W cm}^{-1}^\circ\text{C}^{-1}$ (molecular thermal conductivity)
$k_m = 0.1 \text{ W cm}^{-1}^\circ\text{C}^{-1}$
$Cp_c = 1.0 \text{ J g}^{-1}^\circ\text{C}^{-1}$
$Cp_m = 0.39 \text{ J g}^{-1}^\circ\text{C}^{-1}$
$\beta_T = 6.5 \times 10^{-5} \text{ K}^{-1}$
$\mu_m = 0.4 \text{ g cm}^{-1} \text{ s}^{-1}$
$\gamma = 780 \text{ erg cm}^{-2}$
$\partial\gamma/\partial T = 0.035 \text{ erg cm}^{-2} \text{ K}$
$\phi_0 = 5-10^\circ + \Delta\phi_0 \cos(m\theta)$
$\Delta\phi_0 = 0-10^\circ; m = 3, 4$
<i>Other input parameters</i>
$L = 6.8 \text{ cm}$
$D_c = 0.68 \text{ cm}$
$a = 12.5 \text{ cm}$ (long-axis length of the mirror)
$b = 11.5 \text{ cm}$ (short-axis length)
$P_{\text{Lamp}} = 450-650 \text{ W}$
$T_{\text{amb}} = 500^\circ\text{C}$
$U_f = U_c = -1 \times 10^{-4} \text{ cm s}^{-1}$
$\alpha = 0-30^\circ$
<i>Dimensionless groups</i>
$Pr = 4; Ra_T = 1.53 \times 10^4; Ma = 4917.9$

in Fig. 2b, while 323°C in the angular direction. For a more accurate simulation, a better model [14,25,26] for the lamp heating should be used, but they require much more computational effort. In this report, we prefer to use a simpler model, but focus our study on the 3D heat flow and meniscus-controlled features.

3.2. Effect of convection modes

Fig. 3 shows the calculated results for the effect of convection modes; each lamp power is 550 W . For the case of conduction only in Fig. 3a, except the flow due to sample movement, because of the asymmetric heating, the molten zone is much longer facing to the lamps (see the $x-y$ plane), while much heat loss occurs in the z direction. As a

result, the growth interface has a saddle shape; it is convex in the $x-y$ plane and concave in the $x-z$ plane. The $y-z$ plane ($x = 3.4 \text{ cm}$) also shows that the higher temperature is located at the surface facing to the lamp, while the lower temperature at the other two sides. Because the thermal conductivity of the melt is very low, the superheating in the molten zone is very high being 971°C here. The maximum temperature difference in the angular direction is up to 745°C . From the isotherms at the $x-z$ plane, the maximum temperature appears at the center. As the buoyancy flow is considered, the temperature fields and zone length are slightly affected. The temperature at the $y-z$ plane remains similar. However, a clear buoyancy flow appears at the $x-y$ plane; the melt floats upward near the hot spot and sinks at the center; the maximum melt velocity is about 0.97 cm/s upward and the downward velocity is about 0.52 cm/s . Accordingly, the isotherms are slightly distorted in the flow direction. This convection pattern is somewhat different from the 2D one. For 2D cases, the flow sinks at the centerline leading to the maximum melt velocity at the center. However, for the 3D case, the flow sinks at the cooler region, which is widely distributed in the z direction. As shown by the flow pattern in the $x-z$ plane, all the flows are flowing downward. In addition, due to the flow, the upper interface becomes much more concave. In practice, the concave upper interface often causes problems. Bubbles can easily accumulate there and this could lead to catastrophic zone collapse when the bubble is grown too big. Furthermore, although radial thermal gradients are very high, the buoyancy flow is still very weak here. This is simply due to the very viscous melt. Nevertheless, the maximum superheating (777°C) is significantly lower than that without the buoyancy flow.

With thermocapillary flow, the zone length is increased dramatically at the melt surface, while reduced at the zone center, as shown in Fig. 3c. The convection shown in the $x-y$ plane is similar to that in the axisymmetric case [9]. Because the melt flow near surface is pushed toward the upper and lower interfaces, significantly distorted isotherms and very convex interfaces are obtained

Fig. 2. (a) Heat flux and (b) temperature distributions on the sample from the furnace; $P = 550 \text{ W}$ and $\phi = 5^\circ$; $q_{\max} = 1473.33 \text{ W/cm}^2$; $T_{\min} = 773 \text{ K}$ and $T_{\max} = 2682.5 \text{ K}$; $\Delta q = q_{\max}/40$ and $\Delta T = (T_{\max} - T_{\min})/40$.

(viewed from the x – y plane). The maximum melt velocity is 3.5 cm/s . The maximum superheating is reduced to 439°C . The thermocapillary flow in the x – z plane is slightly weaker due to the smaller thermal gradients in the axial direction at the surface. Again, due to the similar flow, the lower interface viewed from the x – z plane becomes slightly convex. Interestingly, if we examine the flow on the y – z plane (at $x = 3.4 \text{ cm}$), significant angular melt flow is induced leading to four flow

vortices; the maximum angular melt velocity is about 2 cm/s . Indeed, this is due to the angular thermal gradients caused by the two-lamp heating. Due to the significant angular melt motion, the temperature distribution in the angular direction becomes much more uniform, as compared with previous two cases. The maximum angular temperature difference is only 323°C , as compared with 573°C in (b) and 745°C in (a). The zone length uniformity is greatly improved as well.

Fig. 3. Effect of convection modes on the heat flow and zone shape: (a) conduction only; (b) buoyancy convection; (c) buoyancy and Marangoni convection; $P = 550\text{ W}$ and $\phi_0 = 5^\circ$; $\Delta T = (T_{\max} - T_{\min})/20$ in the 2D plot and $\Delta T = (T_{\max} - 773)/40$ in the 3D plot.

Rotating the sample, counter-rotation or corotation, could significantly reduce the angular temperature gradients. Also, the interface shape could become much more axisymmetric. However, significant angular growth and melting is another concern [21].

3.3. Effect of heating power

Beside 550 W, a lower and a higher heating powers are also examined to illustrate the power

sensitivity on the zone shape and heat flows. Figs. 4a and b show the calculated results for 500 and 600 W, respectively. As shown, from the shape and the isotherms on the x-y plane, the zone length is not very sensitive to the heating power. Both molten zones are quite stable. The superheating is 380°C at 500 W and 512°C at 600 W. However, the interface convexity decreases with the increasing zone length. In general, the power ranging from 450 to 650 W (each lamp) has been examined by simulation showing that a stable zone

Fig. 3 (continued).

can be obtained. However, in practice a too short or a too long molten zone will make the growth difficult. Especially, when the molten zone increases, zone necking near the upper interface increases. To some extent the molten zone will collapse due to the melt dewetting from the upper tri-junction rim. A suitable molten zone usually requires well-separated interfaces and less deformed meniscus. By judging from this, the power of 550 W is a suitable choice.

Nevertheless, the interfaces (in the x – y plane) for all cases are still very convex and such severe interface deformation has been quite typical in

practice, even with crystal rotation, e.g. [27]. In general, the calculated interfaces are consistent with the previous experiments [27,28]. On the other hand, the distortion of the feeding interface is different from the growing one, and this is due to necking and the opaque feeding rod. Crystal rotation is helpful in reducing interface convexity [27]. However, for a crystal with a non-circular cross section, there is, in principle, no steady state for rotation. As will be illustrated shortly, even for constant growth angle, due to the requirement of growth angle constraint in Eq. (6), the grown crystal shape is not perfectly round. Having

Fig. 3 (continued).

anisotropic interfacial energy, the grown crystal surface could also be faceted. Due to this difficulty, even though some approximations can be made, in this report, we do not attempt to investigate the effect of rotation. However, from our preliminary calculations, rotation can greatly improve the zone uniformity as well as interface shape, which is consistent with the experimental observation [27].

The melt convection also increases with the increasing heating power. For 600 W in Fig. 4b, the maximum melt velocity is about 4 cm/s, while

it is 3.5 and 3 cm/s for 550 and 500 W, respectively. However, the angular melt velocity is not affected much by the heating power. This is simply due to the improvement of angular temperature uniformity; the larger the flow, the smaller the thermal gradient.

3.4. Anisotropic growth angle and crystal morphology

It has been well known for oxides that the grown crystal morphology, i.e., the cross section

Fig. 4. Effect of heating power: (a) $P = 500$ W; (b) 600 W.

shape, is often not round [29–32]. The crystal facet at the surface depends on the growth orientation. This is believed to be the cause of the anisotropy of the crystal/vapor interfacial energy [33,34]. For example, Dreeben et al. [32] measured the growth angle at the growth tri-junction for sapphire during FZ growth. In the $\langle 0001 \rangle$ growth, $\phi_0 = 35^\circ$ at the faceted tri-junction, while $\phi_0 = 17^\circ$ at the non-faceted junction rim. In other words, the growth angle ϕ_0 is not constant along the tri-junction rim. For YAG growth, the cross section of the grown crystal is quite round during the growth in the $\langle 111 \rangle$ direction [30,31]. However,

a slightly near square shape is often obtained for the $\langle 100 \rangle$ growth. On the other hand, for YIG growth, it is more obvious; a triangular shape is obvious in $\langle 111 \rangle$ growth, while a square shape in $\langle 100 \rangle$ growth [29]. In order to illustrate this effect, we also consider the anisotropic growth angle in the model.

For the lamp power of 550 W, the calculated results for $\langle 111 \rangle$ growth are shown in Fig. 5a for $\phi_0 = 10^\circ + 10^\circ \cos(3\theta)$, i.e., with a 3-fold symmetry (3m). Similarly, for $\langle 100 \rangle$ growth is shown in Fig. 5b for $\phi_0 = 10^\circ + 10^\circ \cos(4\theta)$, i.e., with a 4-fold symmetry (4m); $\theta = 0$ at the $-y$ direction.

Fig. 4 (continued).

In the x - y plot of Fig. 5a, due to the larger growth angle in the $-y$ direction being 20° , the melt bulges outward, while the growth angle is zero at the left corner. For the $\langle 100 \rangle$ growth in Fig. 5b, the melt surface bulges outward in both y and z directions. Besides the crystal and zone shapes, the flow and thermal profiles in Figs. 5a and 5b are very close. The grown crystal shapes (dashed lines) at different growth angle distributions are summarized in Fig. 6. For constant growth angle in Fig. 6a, the grown crystal is not perfectly round, and this is due to the non-uniform zone length. The side with longer zone length (in y

and $-y$ directions) tends to grow outward more, while the other two sides ($+z$ and $-z$ directions) grow inward. This is particularly obvious for non-uniform growth angle with 3-fold symmetry; the growth angle is larger at 0° , 120° , and 240° . However, for 4-fold symmetry, the result is just the opposite. The crystal facets appear at angles 0° , 90° , 180° , and 270° , while the crystal surface grows outward at 45° , 135° , 225° , and 315° . Interestingly, this is consistent with the measurement of Dreeben et al. [32] for sapphire fibers having a rectangular cross section; the growth angle at the faceted side is larger.

Fig. 5. Effect of anisotropic growth angle: (a) $\phi_0 = 10^\circ \pm 10^\circ \cos(3\theta)$; (b) $\phi_0 = 10^\circ \pm 10^\circ \cos(4\theta)$.

3.5. Tilted growth

Although the growth axis in practice is always aligned with the gravity, the effect of tilting could be an interesting problem to investigate because the zone shape can be significantly affected. Fig. 7 shows the heat flow and the zone shape due to 18° tilting (by changing the gravity orientation α). The heating power is kept at 550 W and the growth angle (uniform) at 5° . Because the molten zone shape is affected to the $-y$ direction, the grown crystal is shifted to satisfy the growth angle constraint. Besides the

slightly larger melt velocity (3.9 cm/s), the calculated heat flow and growth front shape are very similar to that without tilting. In other words, the contribution from the buoyancy flow and the meniscus shape is very little here. It is also observed that there is a symmetry breaking in the $x-z$ plane. Through extensive mesh refinement in the angular direction, it is believed that the symmetry breaking is not affected much by the mesh number. Increasing tilting also reduces the zone stability. The necking at the upper tri-junction rim in the y direction increases with the tilting.

Fig. 5 (continued).

The crystal shifting from the feeding axis due to tilting is illustrated in Fig. 8. Also, the crystal shape lightly deviates from a perfect circle. The diameter in the y -direction is longer than that in the z -direction. Again, this is also due to the longer zone length in the x – y plane. With tilting in Figs. 8b and c, the crystal shape is similar. In fact, there is no example in practice of growing the crystal with tilting. From the present calculations, we do not find any advantages, beside the stronger melt flow, that might be helpful to the growth.

4. Conclusions

A self-consistent 3D model considering heat flows, interfaces, meniscus, and the grown crystal shape is developed for FZ crystal growth. For the 2D and half-zone models, our results agree very well with previous calculations. The growth of YAG crystals in a double-ellipsoid mirror furnace is illustrated. The effects of convection, heating power, anisotropic growth angle, and tilting are discussed. Due to the high melting point and the low thermal conductivity, superheating and

Fig. 6. Effect of anisotropic growth angle on the crystal shape: (a) $\phi_0 = 5^\circ$; (b) $\phi_0 = 5^\circ \pm 5^\circ \cos(3\theta)$; (c) $\phi_0 = 10^\circ \pm 5^\circ \cos(3\theta)$; (d) $\phi_0 = 5^\circ \pm 5^\circ \cos(4\theta)$; (e) $\phi_0 = 10^\circ \pm 10^\circ \cos(4\theta)$; solid line is for the feed rod and dashed for the crystal; $P = 550$ W.

thermal non-uniformity is very high leading to non-uniform interfaces. The interface convexity and deformation decrease with the increasing heating power and zone length. With the thermocapillary effect, the axial and angular surface flows are significantly enhanced and they reduce significantly the thermal non-uniformity. The anisotropic growth angle with 3- or 4-fold symmetry also leads to triangular or square cross section of the growing crystal depending on the growth direction. However, the faceting planes for the 4-fold symmetry appear at the direction with a lower growth angle, which is consistent with the experimental observation. Although rotation is often used in practice, it is not considered in this

Fig. 7. Heat flow and zone shape of a tilted growth at $\alpha = 18^\circ$; $P = 550$ W and $\phi_0 = 5^\circ$.

Fig. 8. Effect of ample tilting on the crystal position and shape: (a) $\alpha = 0^\circ$; (b) $\alpha = 18^\circ$; (c) $\alpha = 36^\circ$. $P = 550$ W and $\phi_0 = 5^\circ$.

study due to the non-steadiness of the model nature. However, the effect of rotation will be illustrated in the near future.

Moreover, due to the extremely high temperature during crystal growth, in-situ measurements are difficult. Therefore, the self-consistent numerical model provides a tool to better understand the process. Especially, the model also provides useful information for theory, such as the facet formation and growth mechanisms. Nevertheless, a better furnace model and a better simulation of the internal radiation are necessary for a more quantitative simulation.

References

- [1] P. Dold, A. Croll, K.W. Benz, J. Crystal Growth 183 (1998) 545.
- [2] M. Schweizer, A. Croll, P. Dold, Th. Kaiser, M. Lichtensteiger, K.W. Benz, J. Crystal Growth 203 (1999) 500.
- [3] Th. Kaiser, A. Cröll, P. Dold, K.W. Benz, Proc. of the 2nd European Symp. Fluids in Space, Naples, 1996, p. 245.
- [4] A. Muhlbauer, A. Muiznieks, G. Raming, H. Riemann, A. Ludke, J. Crystal Growth 198/199 (1999) 107.
- [5] M. Levenstam, G. Amberg, J. Fluid Mech. 297 (1995) 357.
- [6] M. Prange, M. Wanschura, H.C. Huhlmann, H.J. Rath, J. Fluid Mech. 394 (1999) 281.
- [7] S. Nakamura, T. Hibiya, K. Kakimoto, N. Imaishi, S. Nishizawa, A. Hirata, K. Mukai, S. Yoda, T.S. Morita, J. Crystal Growth 186 (1998) 85.
- [8] J.L. Duranceau, R.A. Brown, J. Crystal Growth 75 (1986) 367.
- [9] C.W. Lan, S. Kou, J. Crystal Growth 108 (1991) 351.
- [10] C.W. Lan, S. Kou, J. Crystal Growth 114 (1991) 517.
- [11] C.W. Lan, J. Crystal Growth 169 (1996) 269.
- [12] A. Muhlbauer, A. Muiznieks, J. Virbulis, A. Ludge, H. Riemann, J. Crystal Growth 180 (1997) 372.
- [13] C.W. Lan, M.C. Liang, Int. J. Numer. Methods Eng. 40 (1997) 621.
- [14] C.W. Lan, C.H. Tsai, J. Crystal Growth 173 (1997) 561.
- [15] C.W. Lan, C.H. Chian, J. Crystal Growth 230 (2001) 172.
- [16] G. Ratnieks, A. Muiznieks, A. Muhlbauer, G. Raming, J. Crystal Growth 230 (2001) 48.
- [17] P.J. Hart, J. Opt. Soc. Am. 48 (1958) 673.
- [18] D. Rivas, J. Sanz, C. Vazquez, J. Crystal Growth 47 (1979) 219.
- [19] D. Vizman, I. Nicoara, G. Muller, J. Crystal Growth 212 (2000) 334.
- [20] S.A. Korpela, J. Ni, A. Chait, M. Kassemi, J. Crystal Growth 165 (1996) 455.
- [21] C.W. Lan, C.H. Chian, T.Y. Wang, J. Crystal Growth 218 (2000) 115.
- [22] C.W. Lan, M.C. Liang, J. Comp. Phys. 152 (1999) 55.
- [23] C.W. Lan, Int. J. Numer. Methods Fluids 19 (1994) 41.
- [24] V.J. Fratello, C.D. Brandle, J. Crystal Growth 128 (1993) 1006.
- [25] A. Eyer, R. Nitshe, H. Zimmermann, J. Crystal Growth 47 (1979) 219.
- [26] D. Rivas, R. Haya, J. Crystal Growth 241 (2002) 249.
- [27] K. Kitamura, I. Shindo, S. Kimura, J. Crystal Growth 46 (1979) 277.
- [28] K. Kitamura, S. Kimura, K. Watanabe, J. Crystal Growth 57 (1982) 475.
- [29] H. Tsuiki, K. Kitazawa, T. Masumoto, K. Shiroki, K. Fueki, J. Crystal Growth 49 (1980) 71.
- [30] K. Kitamura, Y. Miyazawa, Y. Mori, S. Kimura, M. Higuchi, J. Crystal Growth 64 (1983) 207.
- [31] K. Kitamura, S. Kimura, M. Higuchi, T. Oshikiri, K. Komatsu, J. Crystal Growth 69 (1984) 537.
- [32] A.B. Dreeben, K.M. Kim, A. Schujko, J. Crystal Growth 50 (1980) 126.
- [33] T. Surek, J. Appl. Phys. 47 (1976) 4384.
- [34] W. Bardsley, F.C. Frank, G.W. Green, F.T.J. Hurle, J. Crystal Growth 23 (1974) 341.