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Convective Interactions and Heat Transfer in a Czochralski Melt with a Model Phase Boundary of Two Different Shapes

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***Abstract*—**Implicit in most large-scale numerical analyses of the crystal growth from the melt is the assumption that the shape and position of the phase boundary are determined by the transport phenomena coupled strongly to the melt hydrodynamics. In the present numerical study, the interface shape-effect on the convective interactions in a Czochralski oxide melt is described. It was demonstrated that thermocapillary flow affects inversely the phase boundaries of distinct shapes. The inhomogenity of heat flux and the location of the stagnation point at the crystallization front were investigated. The forced convection effect on the point displacement at the boundary found to be much stronger for the flat plate interface compared to the cone-shaped one with and without the Marangoni flow.

***Keywords*—**Computer simulation, fluid flow, interface shape, thermocapillary effect.

I. Introduction

PROCESSES developed to grow large single crystals from the melt are among the most challenging and precise in engineering practice, and large-scale numerical analyses has been vital to advance crystal growth technology to current level. Refractory oxides such as gadolinium gallium garnet (GGG) are widely used as solid-state laser hosts and materials for epitaxial films in magneto-optical devices [1], [2]. As the most commonly used technique, garnet crystals are grown by the Czochraslki (Cz) method during which melt flow instabilities can lead to serious problems. These hydrodynamic instabilities cause morphological changes as spiral formation arising after the onset of symmetry breaking in an initially steady and axisymmetric melt flow [3].

The Cz growth process of garnet single crystals shows as well the so-called interface-inversion as an abrupt change in the shape of the phase boundary [4]. The crystallization front shape, which is initially convex to the melt, can be suddenly changed to a flat or even concave interface. The interface inversion, particularly occurs when the crystal diameter, $2r_x$ and so the rotationally-driven flow predominates in the flow field. The influence of the convective interactions on the shape and position of the phase boundary is well known by some experimental and many numerical works. However, the details of the interface inversion and the dynamical processes during the inversion of the shape boundary are not yet understood.

In the present hydrodynamic model of Cz oxide melt flow, the computational results of a model with cone-shaped interface is compared to those of a flat crystallization front. That is, particular attention is paid to reveal the impact of the interface deflection itself on the convective interactions and heat transfer in the melt.

II. Description of the Model

Hydrodynamics of a high Prandtl number ($\mathrm{Pr} \geq 1$) fluid affects strongly the heat transport at the phase boundary in a Czochraslki (Cz) growth configuration. In the present two dimensional and steady-state finite volume method calculations, the flow field of an oxide ($Gd_3Ga_5O_{12}$) melt is characterized by similarity parameters $\mathrm{Pr} = 4.69, \mathrm{Gr} = 8.22 \times 10^4, \mathrm{Ma} = 1.93 \times 10^4$ and/or $\mathrm{Ma} = 0$ for the cases with and without Marangoni effect, based on the crucible radius, $r_c = 60\mathrm{mm}$, and the driving temperature difference, $\Delta T_{max} = T_w - T_{mp} = 72\ \mathrm{K}$ between the crucible wall and the melting point. The nondimensional parameters are defined as the Grashof number, $\mathrm{Gr} = g\beta\Delta T_{max} r_c^3/\nu^3$, the Marangoni number, $\mathrm{Ma} = [\partial\sigma/\partial\, T]\Delta T_{max} r_c/\mu\alpha_T$ and the Prandtl number, $\mathrm{Pr} = \nu/\mu$ where g is the gravitational acceleration, β is the coefficient of volumetric thermal expansion, ν is the kinematic viscosity, α_T is the thermal diffusivity, σ is the surface tension coefficient and μ is the dynamic viscosity of the melt. The physical properties of the GGG melt used in the present simulations all are given in [5]. The crucible of height $h_c = 2r_c$ is filled with the melt of a meniscus configuration ($h_m \sim 4\mathrm{mm}$) close to the edge ($r = r_x, z = h_c + h_m$) of crystal-dummy ($r_x/r_c \cong 0.4$) rotating uniformly with the angular velocity Ω (rad /s) around the symmetry axis (OZ). The intensity of the rotationally-driven flow is given by $\mathrm{Re} = 81.4\ \Omega$ for both the flat plate ($k/r_x = 0$) and the cone-shaped ($2\alpha = 120^\circ, k/r_x = 0.58$) phase boundaries. The melt is generally assumed to be opaque ($\varepsilon_l = 0.3$), the ambient temperature and heat transfer coefficient are $T_a = T_{mp} - \Delta T_{max}$. Here, k/r_x denotes the convexity of interface.

The steady-state convective interaction regimes in a Cz melt are defined by the ratio between the intensity of buoyancy-driven flow and the rotationally-driven forces (see Fig. 2), represented as $\mathrm{Gr}/\mathrm{Re}^2$. For $\mathrm{Gr}/\mathrm{Re}^2 > 1$ the thermal convection, that is, the buoyancy-and the surface tension driven flows, predominates and the heat balance at the phase

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boundary leads to a convex to melt interface with $(k/r_x) > 0$ as the interface deflection. We define k as the height of the cone-shaped crystallization front. When the swirl velocity, $u_\theta = r_x \Omega$ increased and so Gr/Re^2 approached to the unity, the isotherms just beneath the growing crystal correspond to a flat interface with $(k/r_x) \cong 0$. The crystal rotation drives a flow which streams upward below the crystallization front, outward along the meniscus, and down along the boundary between the thermal convection and the rotationally-driven convection cells. The shear layer between the two cells, known as the Stewartson layer, is a thin region of high vorticity separating the regions of low vorticity which can be modelled as a line across which the velocity field is discontinuos [6]. The Stewartson layer has a contact point, $K(r_{sp}, z)$ on the crystallization front $0 < r_{sp} < r_x$ for the case $Gr/Re^2 > 1$. The point K lies near the tri-junction point at $r_{sp} \cong r_x$ when the governing parameter of the flow field (Gr/Re^2) is around the unity, and the further enhancement of the forced convection intensity leads to a larger displacement of the stagnation point, K radially outwards and the interface inversion occurs for the case $(Gr/Re^2) < 1$.

The geometry of the problem to be studied is depicted in Fig. 1. The cylindrical crucible is stationary and the crystal rotates at the rate $\Omega(rad/s)$. The oxide melt is assumed to be incompressible, Newtonian, Boussinesq fluid. The temperature of the melt-crystal interface is maintained at the melting point, T_{mp}. The heat loss from the melt free surface is due to convection and radiation to an ambient temperature, T_a. The sidewall of the crucible is kept at a constant temperature, T_w. The bottom of the crucible is adiabatic. The no-slip condition is applied for all physical boundaries of the melt except for its free surface. The melt free surface is assumed to be free of stresses or not when the thermocapillary effect is taken into account.

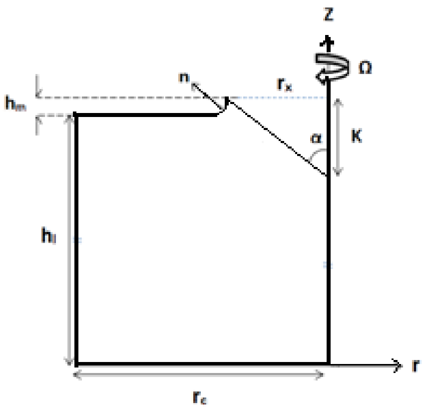


Fig. 1 Sketch of open crucible model with a convex to melt interface

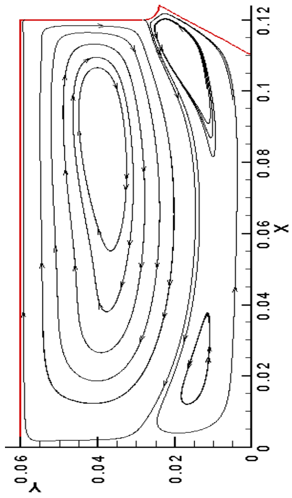


Fig. 2 The flow field in the Cz model subjected to the boundary conditions of the problem $(Pr = 4.69, Gr = 8.22 \times 10^4, Ma = 2.0 \times 10^4, Re = 325, k/r_x = 0.58)$

III. Governing Equations and Boundary Conditions

The Cz melt flow is governed by the momentum, continuity, and energy equations in the Boussinesq approximation. These equations are given as follows:

$$\nabla . u = 0 \quad (1)$$

$$\rho(\nabla . u) = -\nabla P + \mu \nabla^2 u + \rho g \quad (2)$$

$$\nabla . (uT) = \alpha_T \nabla^2 T \quad (3)$$

where $u = (u_r, u_\theta, u_z)$ is the flow velocity vector, P, T and ρ are the pressure, the melt temperature and the melt density. The boundary conditions which define the melt free surface can be written in cylindrical coordinates as:

$$\mu(\partial u_r / \partial n) = -(\partial \sigma / \partial T)(\partial T / \partial r) \quad (4)$$

$$(\partial u_\theta / \partial n) = 0, \qquad u_z = 0 \quad (5)$$

$$-\lambda_l(\partial T / \partial n) = h_a(T - T_a) + \varepsilon_l \sigma_B (T^4 - T_a^4) \quad (6)$$

Equation (4) describes the Marangoni effect depending on the radial temperature difference $\Delta T_{max} = T_w - T_{mp}$. The coefficient $(\partial \sigma / \partial T)$ is assumed to be vanished for the case without the thermocapillary effect. $n = (n_r, n_z)$ is a normal vector from the free surface outwards to the ambient with $h_a = 10\ W.m^{-2}.K^{-1}$ as its coefficient of heat transfer. The boundary conditions at the melt centerline are $u_r = u_\theta = 0$, $\partial u_z / \partial r = 0$ and $\partial T / \partial r = 0$. At the crucible sidewall, $u = 0$ and $T = T_w$. The crucible bottom is conditioned by $u = 0$ and $\partial T / \partial u_z = 0$. The crystal-melt interface is defined by $T = T_{mp}$, $u_r = u_z = 0$ and $u_\theta = r_x \Omega$.

IV. Results and Discussion

It was found that thermocapillary effect on the heat removed from the melt free surface $Q_l(W)$ results in a constant increment of $25.5 \pm 1.5\ W$ for Ω in the range $2{\sim}6\ rad/s$. This means that, the rotationally-driven flow does not affect the heat transfer at the melt free surface. Contrarily, the heat removed from the phase boundary $Q_x(W)$ depends on the shape and rotation rate of the crystal dummy. It was shown that, the influence of Marangoni (Ma) convection on $Q_x(W)$ is decreasing (for $k/r_x = 0$) and/or vanishing (for $k/r_x = 0.58$) with the crystal rotation rate.

The ratio between the heat fluxes, $\eta = q_x/q_l$ was calculated for the crystal rotation rate in the range $2 \leq \Omega\ (rad/s) \leq 6$. It was shown that η is larger for the convex to melt interface with $L'_x = r_x/\sin\alpha$ if compared to the flat plate phase boundary of $L_x = r_x$. The marangoni flow which carries hot fluid to the cold spots in the meniscus region, affects inversely the ratio η for the cone-shaped and the flat plate phase boundaries, respectively. The inverse effect of Marangoni convection on η, found to be enhanced by increasing the rotation rate. This has been shown in Fig. 3 that increasing the forced convection intensity the Marangoni effect on η found to be enhanced for the convex to melt interface while the effect is vanishing with Re for the flat interface.

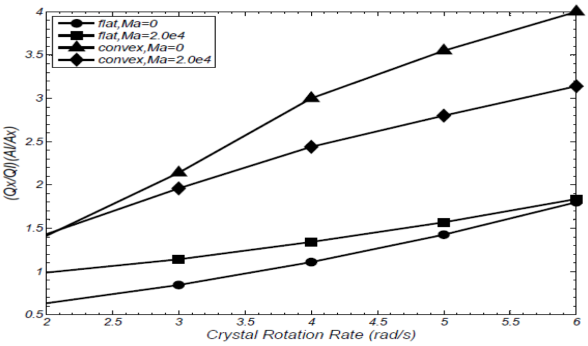


Fig. 3 The inverse effect of Marangoni convection on $\eta = (Q_x/Q_l)(A_l/A_x)$

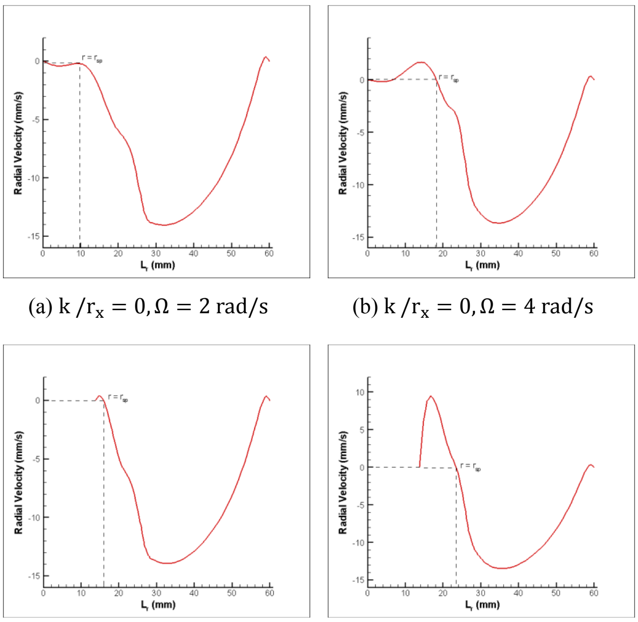
The most important characteristic which affects the inhomogenity of the heat flux, $q_x\ (W/m^2)$ at the phase boundary is the location of the contact point, $K\ (r_{sp}, z)$ of the thermal and forced convection flows on the melt boundaries. The radial position, r_{sp} of the stagnation point, K at which the flow velocity, u_r changes the sign, was measured on a horizontal line, L_r (2 mm below the melt free surface) for different cases. The manner after which we determined the radial position of $K\ (r_{sp}, z)$ is illustrated by two sets of figures, Figs. 4 (a)-(d) and 5 (a)-(d) for the cases with and without Marangoni flow, respectively. Note that only the left moiety of the system is considered. Hence, close to the melt free surface, $u_r < 0$ belongs to the thermal convection (clockwise circulation) and $u_r > 0$ is the radial velocity due to the rotationally-driven flow (ccw).

It was shown that, by increasing the rotation rate ($2 \leq \Omega\ (rad/s) \leq 4$), the point $K\ (r_{sp}, z)$ moves outward from its location on the phase boundary. As given in Table I, the forced convection effect on the displacement of the stagnation point is much stronger for the flat phase boundary compared to the convex to melt ($k/r_x = 0.58$) interface.

TABLE I
Convective Interaction and Interface Shape Effect on the Radial Position of the Stagnation Point $K\ (r_{sp}, z)$ with $r_{sp} = r_x \pm \Delta r, r_x = 23.75$ mm

Cone-shaped interface ($k/r_x = 0.58$)		Flat plate interface ($k/r_x = 0$)	
$2 \leq \Omega\ (rad/s) \leq 4$		$2 \leq \Omega\ (rad/s) \leq 4$	
$Ma = 0$	$Ma \cong 2 \times 10^4$	$Ma = 0$	$Ma \cong 2 \times 10^4$
$22.3 \leq r_{sp}(mm) \leq 28.3$	$16.0 \leq r_{sp}(mm) \leq 23.5$	$15.8 \leq r_{sp}(mm) \leq 27.0$	$10.0 \leq r_{sp}(mm) \leq 18.2$
$\Delta r = 27\%$	$\Delta r = 47\%$	$\Delta r = 71\%$	$\Delta r = 82\%$

This is a noticeable result because a cone-shaped interface exerts more shear onto the melt due to its larger boundary ($L' = r_x/\sin\alpha$) compared to a flat interface. It was shown (Table I) that, in the presence of thermocapillary forces ($Ma \neq 0$), the point $K\ (r_{sp}, z)$ displaces largely but remains on the crystal dummy ($r_{sp} \leq r_x$): Even for the flow field characterized by $Gr/Re^2 = 0.775$, the radial position of the point K was found to be equal to $r_{sp} = 18.2$ and $r_{sp} = 23.5$ for the flat and cone-shaped interfaces, respectively.

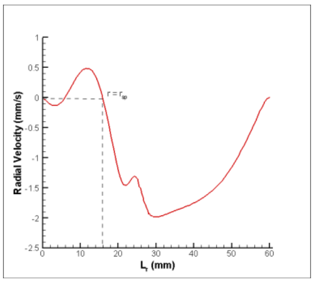


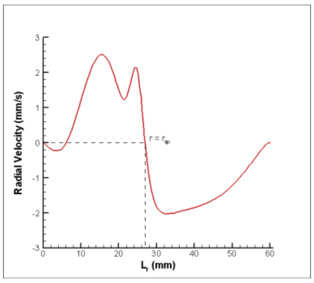
(a) $k/r_x = 0, \Omega = 2$ rad/s (b) $k/r_x = 0, \Omega = 4$ rad/s

(c) $k/r_x = 0.58, \Omega = 2$ rad/s (d) $k/r_x = 0.58, \Omega = 4$ rad/s

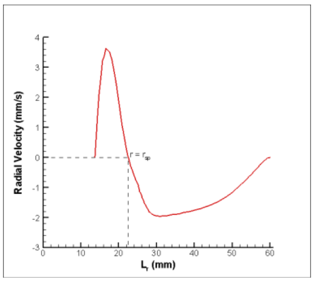
Fig. 4 Radial velocity of the flow along the horizontal line L_r (2mm below the melt free surface) for the case $Ma \neq 0$

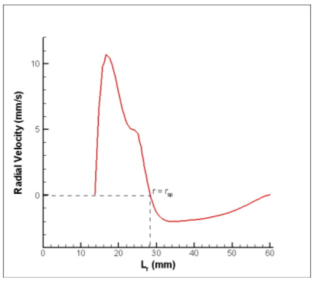
For the convex to melt phase boundary ($k/r_x = 0.58$), increasing Ω (or Re) the inhomogenity of heat flux at the interface $\Delta q = (q_x - q'_x)/(q_x + q'_x)$ is vanishing. That is, for $r_{sp} < r_x$, the local heat flux at the center, q_x $(r = 0)$ is larger than the heat flux near the tri-junction point, $q'(r = r_x)$ and, hence $\Delta q > 0$ .Contrarily, if the stagnation point K locates at $r_{sp} \geq r_x$, the inhomogenity of heat flux is $\Delta q \leq 0$. In the case of lower rotation rate ($\Omega = 2$ rad /s , $Gr/Re^2 = 3.1$), the point K locates on the crystal dummy and, therefore, $\Delta q > 0$ for both of the interface shapes.





(a) $k/r_x = 0, \Omega = 2$ rad/s (b) $k/r_x = 0, \Omega = 4$ rad/s





(c) $k/r_x = 0.58, \Omega = 2$ rad/s (d) $k/r_x = 0.58, \Omega = 4$ rad/s

Fig. 5 Radial velocity of the flow along the horizontal line L_r (2mm below the melt free surface) for the case $Ma = 0$

REFERENCES

[1] Y. Fei, M.M.C. Chou, B.H.T. Chai, "Crystal growth and morphology of substituted gadolinium gallium garnet" J. Cryst. Growth, vol. 240, 2002, pp. 185-189.

[2] Z. Jia, X. Tao, C. Dong, X. Cheng, W. Zhang, F. Wu, M. Jiang, "Study on crystal growth of large size Nd: Gd3 Ga5 O12 single crystals by Czochralski method" J. Cryst. Growth, vol. 292, 2006, pp. 386-390.

[3] N. Crnogorac, H. Wilke, K. A. Cliffe, A. Yu. Gelfgat, E. Kit, "Numerical modeling of instability and supercritical oscillatory state in a Czochralski model system of oxide melt" Cryst. Res. Technol. vol. 43, 2008, pp.606-615.

[4] O. N. Budenkova, M.G. Vasiliev, V.S. Yuferev, I. A. Ivanova, A.M. Ivanov, A. M. Bul'kanov, V.V. Kalaev, "Investigation of the variation in the crystallization front shape during growth of gadolinium gallium and terbium gallium crystals by the Czochralski method" Crystallography Reports, vol.53, 2008,pp.1181-1190.

[5] N. Crnogorac, H. Wilke, G. O. Young, "Measurement of physical properties of D_yScO_3 melt" Cryst. Res, Technol, vol. 44, 2009, pp. 581-589.

[6] A. Yeckel, J.J. Derby, "Computational simulations of the growth of crystals from liquids" in *Crystal Growth Technology*, H. J. Scheel and T. Fukuda, Eds. New York: John wiley and sons, 2003, pp. 115–137.