NATURAL AND MARANGONI CONVECTIONS IN A FLOATING ZONE

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Using an order-of-magnitude evaluation, a quantitative evaluation was performed of the natural and Marangoni convections produced in the melt during single-crystal growth by the floating zone method.

The Reynolds number, which forms a standard for free interfacial velocity, was described as a function of the Grashof number, Marangoni number and Prandtl number. The results of this theoretical study showed good agreement with previous experimental works using model liquids and a glass melt.

Introduction

The floating zone method is a widely used technique for growing single crystals. When single crystals are grown by this method, natural and Marangoni convections are formed in the melt due to the temperature distribution, and have an important effect on the quality of the single crystals. For this reason, much research has been carried out on the convection processes occurring in a floating zone, both in theoretical studies³⁻⁶⁾ and also in experimental work using model liquids^{1,7,10)} and a glass melt.⁸⁾ However, these works concentrated on the velocity and temperature distributions for convections in melts under certain growth conditions; there have been no reports regarding a quantitative evaluation of the relationship between single-crystal growth conditions and melt convection velocity.

The authors carried out a quantitative evaluation of the natural and Marangoni convections for the melt inside a two-dimensional rectangular open boat.⁹⁾ In this study, an order-of-magnitude evaluation^{9,11)} was performed for the Marangoni and natural convections in a floating zone, thus making possible a quantitative evaluation of the effect of natural and Marangoni convections on the interfacial velocity at the free interface.

1. Configuration and Basic Equations

A diagram of the theoretical model and the coordinate system are shown in Fig. 1. The following assumptions were made:

- (1) the fluid is incompressible and Newtonian;
- (2) flow is steady, symmetrical and two-dimensional;
- (3) the free interface and the crystal-melt interface are

both flat; (4) $T_H > T_C$; (5) there is no rotation or movement of the upper and lower rods; (6) the values of all physical properties are constant except for the interfacial tension in the stress balance equation and the density in the buoyancy force term.

The basic equations are as follows:

Continuity equation

$$\frac{1}{r} \frac{\partial}{\partial r} (ru_r) + \frac{\partial u_z}{\partial z} = 0 \quad (1)$$

Navier-Stokes equations

$$u_r \frac{\partial u_r}{\partial r} + u_z \frac{\partial u_r}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial r} + \nu \left[\frac{\partial}{\partial r} \left(\frac{1}{r} \frac{\partial}{\partial r} (ru_r) \right) + \frac{\partial^2 u_r}{\partial z^2} \right] \quad (2)$$

$$u_r \frac{\partial u_z}{\partial r} + u_z \frac{\partial u_z}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial z} + \nu \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_z}{\partial r} \right) + \frac{\partial^2 u_z}{\partial z^2} \right] + g\beta(T_H - T_C) \quad (3)$$

Energy equation

$$u_r \frac{\partial T}{\partial r} + u_z \frac{\partial T}{\partial z} = \alpha \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) + \frac{\partial^2 T}{\partial z^2} \right] \quad (4)$$

Stress balance equation (at $r = r_c$)

Fig. 1. Configuration for analysis and coordinate system

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Table 1. Results-of-Order-of-Magnitude Evaluation of Natural and Marangoni Convections in a Floating Zone

Reynolds number, Re_{zs}	$o[\text{viscous force}] \gg o[\text{inertial force}]$ $Re_{zs} \ll o[1]$		$o[\text{viscous force}] \sim o[\text{inertial force}]$ $Re_{zs} \sim o[1]$	
Prandtl number, Pr	$Pr < o[1]$	$Pr > o[1]$	$Pr < o[1]$	$Pr > o[1]$
Boundary layer thickness, δ	$o[l]$	$o[l]$	$o\left[\left(\frac{\nu l}{u_{zs}}\right)^{1/2}\right]$	$o\left[\left(\frac{\alpha l}{u_{zs}}\right)^{1/2}\right]$
	$o\left[\frac{Ma}{Cr}\right] < 1$		$o\left[\frac{Ma^{2/3}}{Gr^{1/2}}\right] < 1$	$o\left[\frac{Ma^{2/3}}{Gr^{1/2}} Pr^{1/6}\right] < 1$
Natural convection-dominant regime	$o\left[\nu \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_z}{\partial r}\right)\right] = o[g\beta(T_H - T_C)]$	$o\left[u_z \frac{\partial u_z}{\partial z}\right] = o[g\beta(T_H - T_C)]$	$o\left[\nu \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_z}{\partial r}\right)\right] = o[g\beta(T_H - T_C)]$	$o\left[\nu \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial u_z}{\partial r}\right)\right] = o[g\beta(T_H - T_C)]$
	$u_{zs} = o\left[\frac{g\beta(T_H - T_C)l^2}{\nu}\right]$	$u_{zs} = o[g\beta(T_H - T_C)l^{1/2}]$	$u_{zs} = o\left[\left(\frac{g\beta(T_H - T_C)l}{Pr}\right)^{1/2}\right]$	$u_{zs} = o\left[\left(\frac{g\beta(T_H - T_C)l}{Pr}\right)^{1/2}\right]$
	$Re_{zs} = o[Gr]$	$Re_{zs} = o[Gr^{1/2}]$	$Re_{zs} = o\left[\frac{Gr^{1/2}}{Pr^{1/2}}\right]$	$Re_{zs} = o\left[\frac{Gr^{1/2}}{Pr^{1/2}}\right]$
	$o\left[\frac{Ma}{Gr}\right] > 1$	$o\left[\frac{Ma^{2/3}}{Gr^{1/2}}\right] > 1$	$o\left[\frac{Ma^{2/3}}{Gr^{1/2}} Pr^{1/6}\right] > 1$	$o\left[\frac{Ma^{2/3}}{Gr^{1/2}} Pr^{1/6}\right] > 1$
Marangoni convection-dominant regime	$o\left[\mu \frac{\partial u_z}{\partial r}\right] = o\left[\left \frac{\partial \sigma}{\partial T} \frac{\partial T}{\partial z}\right \right]$			
	$u_{zs} = o\left[\left \frac{\partial \sigma}{\partial T}\right (T_H - T_C)/\mu\right]$	$u_{zs} = o\left[\frac{ \partial \sigma / \partial T ^{2/3}(T_H - T_C)^{2/3}\nu^{1/3}}{\mu^{2/3}l^{1/3}}\right]$	$u_{zs} = o\left[\frac{ \partial \sigma / \partial T ^{2/3}(T_H - T_C)^{2/3}\alpha^{1/3}}{\mu^{2/3}l^{1/3}}\right]$	$u_{zs} = o\left[\frac{ \partial \sigma / \partial T ^{2/3}(T_H - T_C)^{2/3}\alpha^{1/3}}{\mu^{2/3}l^{1/3}}\right]$
	$Re_{zs} = o[Ma]$	$Re_{zs} = o[Ma^{2/3}]$	$Re_{zs} = o\left[\frac{Ma^{2/3}}{Pr^{1/3}}\right]$	$Re_{zs} = o\left[\frac{Ma^{2/3}}{Pr^{1/3}}\right]$

$$\mu \frac{\partial u_z}{\partial r} = -\frac{\partial \sigma}{\partial T} \frac{\partial T}{\partial z} \quad (5)$$

Using an order-of-magnitude evaluation for the equations above,^{9,11)} an evaluation was made of the effect of natural convection, Marangoni convection and Prandtl number on interfacial velocity at the free interface.

2. Order-of-Magnitude Evaluation

In Eqs. (1)–(5), r and ∂r were replaced by δ , ∂z by l , u_r and ∂u_r by u_{r0} , u_z and ∂u_z by u_{z0} (although in Eq. (5), ∂u_z was replaced by u_{zs}), and ∂T by $(T_H - T_C)$, and an analysis was performed in the same manner as reported in the previous paper.⁹⁾ The results are shown in Table 1. In Table 1, Ma/Gr , $Ma^{2/3}/Gr^{1/2}$ and $Pr^{1/6}Ma^{2/3}/Gr^{1/2}$ are the parameters determining whether it is the natural or Marangoni convection that will have an effect on the interfacial velocity.

When the order of magnitude of these parameters is less than 1, natural convection is dominant; conversely, when the value is greater than 1, Marangoni convection is dominant. Also, when the order of magnitude of the parameters is approximately equal to 1, the effects of natural and Marangoni convections on interfacial velocity are approximately equal. In previous research, comparisons of the buoyancy and thermocapillary forces, and the magnitudes of the natural and Marangoni convections, in a floating zone have been carried out using the following “dynamic Bond number”:^{2,7,10)}

$$Bo = \rho g \beta L^2 / |\partial \sigma / \partial T| = (Ma_L / Gr_L)^{-1} \quad (6)$$

However, the results of the present study have shown that the “dynamic Bond number” can only be used to determine which melt convection is dominant when $Re_{zs} \ll o[1]$, and that when $Re_{zs} \gg o[1]$ the important parameters are $Ma^{2/3}/Gr^{1/2}$ and

Fig. 2. Comparison of results in Table 1 with previous experimental results

$Pr^{1/6} Ma^{2/3}/Gr^{1/2}$. In addition, it can be seen from Table 1 that when $Re_{zs} \geq 0[1]$ and also $Pr > 0[1]$, the interfacial velocity is a function not only of the Grashof and Marangoni numbers but also of the Prandtl number.

3. Comparison with Previous Experimental Results

Preisser *et al.*¹⁰⁾ and McNeil *et al.*^{7,8)} carried out experimental research on Marangoni convection in a vertical capillary liquid bridge using molten sodium nitrate, and silicone oils and molten sodium borate, respectively, and observed the interfacial velocity induced by Marangoni convection by means of a tracer technique. Their experimental results, after adjustment to comply with the form of the results of Table 1, are shown in Fig. 2. Good agreement was obtained between the results shown in Table 1 and previous experimental results.

From Fig. 2 it was possible to determine the constants of proportionality for Table 1, and as a result the following experimental equations were obtained:

When $Re_{zs} \ll 0[1]$,

$$Re_{zs} = 0.106 Ma \quad (7)$$

$$u_{zs} = 0.106 |\partial \sigma / \partial T| (T_H - T_C) / \mu \quad (8)$$

When $Re_{zs} > 0[1]$,

$$Re_{zs} = 0.470 Ma^{2/3} \quad (9)$$

$$= 0.974 Ma^{2/3} / Pr^{1/3} \quad (r_c/l = 0.732) \quad (9')$$

$$u_{zs} = 0.974 \left(\frac{|\partial \sigma / \partial T| (T_H - T_C) \alpha^{1/2}}{\mu l^{1/2}} \right)^{2/3} \quad (10)$$

Also,

$$Re_{zs} = 0.311 Ma^{2/3} \quad (11)$$

$$= 0.645 Ma^{2/3} / Pr^{1/3} \quad (r_c/l = 1.36) \quad (11')$$

(a) McNeil *et al.*⁷⁾

Fig. 3(a). Interfacial velocity induced by Marangoni convection (comparison with experimental results by McNeil *et al.*⁷⁾)

(b) Preisser *et al.*¹⁰⁾

Fig. 3(b). Interfacial velocity induced by Marangoni convection (comparison with experimental results by Preisser *et al.*¹⁰⁾)

$$u_{zs} = 0.645 \left(\frac{|\partial \sigma / \partial T| (T_H - T_C) \alpha^{1/2}}{\mu l^{1/2}} \right)^{2/3} \quad (12)$$

Figure 3 shows the relationship between the interfacial velocity, u_{zs} , and the temperature difference, $(T_H - T_C)$. In Fig. 3, points represent previous experimental results and lines show the results given by Eqs. (8), (10), and (12). Figure 3(b) shows an interfacial velocity of approximately 20 mm/sec. when the temperature difference between the upper and lower rods is approximately 30 K. Considering that the values of l used in the experiments were 2.2 mm

and 4.1 mm, this is an extremely high velocity, and shows that the flow induced by Marangoni convection is extremely fast.

Conclusion

Using an order-of-magnitude evaluation of Marangoni and natural convections in a floating zone, the effects of the natural and Marangoni convections on interfacial velocity at the free interface were investigated. The theoretical results obtained showed good agreement with previous experimental results for Marangoni convection in a vertical capillary liquid bridge using model liquids and a glass melt.

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Nomenclature

Bo	= dynamic Bond number ($=\rho g \beta L^2 / \partial \sigma / \partial T $)	[—]
g	= gravitational acceleration	[m/s ²]
Gr	= Grashof number ($=g \beta \Delta T l^3 / \nu^2$)	[—]
Gr_L	= Grashof number ($=g \beta \Delta T L^3 / \nu^2$)	[—]
L	= reference length	[m]
l	= half-length of free interface of floating zone	[m]
Ma	= Marangoni number ($= \partial \sigma / \partial T \Delta T l / (\mu \cdot \nu)$)	[—]
Ma_L	= Marangoni number ($= \partial \sigma / \partial T \Delta T L / (\mu \cdot \nu)$)	[—]
o	= order	[—]
p	= pressure	[Pa]
Pr	= Prandtl number ($=\nu / \alpha$)	[—]
r	= radial coordinate	[m]
r_c	= radius of floating zone	[m]
Re	= Reynolds number ($=ul / \nu$)	[—]
T	= temperature	[K]
u	= velocity	[m/s]

u_{r0}	= reference velocity of u_r	[m/s]
u_{z0}	= reference velocity of u_z	[m/s]
z	= axial coordinate	[m]
α	= thermal diffusivity	[m ² /s]
β	= thermal expansion coefficient	[1/K]
δ	= thickness of boundary layer along a free interface	[m]
μ	= viscosity	[Pa · s]
ν	= kinematic viscosity	[m ² /s]
ρ	= density	[kg/m ³]
σ	= interfacial tension	[N/m]

<Subscripts>

av	= average
C	= cold
H	= hot
r	= radial
s	= free interface
z	= axial

Literature Cited

- 1) Change, C. E. and W. R. Wilcox: *Int. J. Heat Mass Transfer*, **19**, 355 (1976).
- 2) Chun, Ch.-H.: *J. Crystal Growth*, **48**, 600 (1980).
- 3) Kamotani, Y. and J. Kim: *J. Crystal Growth*, **87**, 62 (1988).
- 4) Kobayashi, N.: *J. Crystal Growth*, **66**, 63 (1984).
- 5) Kobayashi, N.: *Japanese J. Appl. Phys.*, **27**, 20 (1988).
- 6) Kozhoukharova, Zh. and S. Slavchev: *J. Crystal Growth*, **74**, 236 (1986).
- 7) McNeil, T. J., R. Cole and R. S. Subramanian: *J. Colloid and Interface Sci.*, **98**, 210 (1984).
- 8) McNeil, T. J., R. Cole and R. S. Subramanian: *J. Am. Ceram. Soc.*, **68**, 254 (1985).
- 9) Okano, Y., M. Itoh and A. Hirata: *J. Chem. Eng. Japan*, **22**, 275 (1989).
- 10) Preisser, F., D. Schwabe and A. Scharmann: *J. Fluid Mech.*, **126**, 545 (1983).
- 11) Ruckenstein, E.: *AIChE J.*, **26**, 850 (1980).