MARANGONI CONVECTION IN A LIQUID BRIDGE UNDER MICROGRAVITY CONDITIONS DURING PARABOLIC FLIGHT

AKIRA HIRATA*, SHIN-ICHI NISHIZAWA,
MOTOHARU NOGUCHI AND MASATO SAKURAI
Department of Chemical Engineering, Waseda University, Tokyo, 169
SHOUICHI YASUHIRO AND NOBUYUKI IMAISHI
Institute of Advanced Material Study, Kyushu University, Kasuga, 816

Key Words: Marangoni Convection, Interfacial Tension Gradient, Heat Transfer, Fluid Mechanics, Liquid Bridge, Microgravity, Floating Zone, Interfacial Velocity, Silicone Oil, Parabolic Flight

Marangoni convection in a silicone oil liquid bridge under microgravity conditions during parabolic flight was observed. The developing process of axis-symmetric laminar Marangoni convection was studied experimentally and numerically. To reach a steady state of the temperature field, much more time was needed than to reach a steady state of the velocity field.

The effects of initial temperature conditions inside a liquid bridge, heat transfer across the free surface, and temperature dependence of viscosity on both velocity and temperature field were studied numerically.

Introduction

Single-crystal semiconductors, compound semiconductors, and oxides are very important as device materials in the electronics industry. When single crystals are grown from the melt, Marangoni convection induced by the interfacial tension gradient on the free surface and buoyancy convection induced by density difference occur. These convections affect the quality of the single crystals. The usefulness of microgravity for making homogeneous, exceptionally pure crystals has been demonstrated a number of times. Its major advantage in growing crystals is the absence of buoyancy convection. To obtain high-quality single crystals under microgravity condition it is necessary to clarify and control the Marangoni convection. With this objective in mind, many studies have been carried out related to Marangoni convection, both numerically¹⁾ and experimentally^{2, 9, 10, 16)}. However, many points remain unclear.

The authors have carried out fundamental studies of Marangoni convection³⁾, the quantitative evaluation of Marangoni convection for the melt inside a two-dimensional rectangular open boat^{5, 11)}, in a float zone^{5, 12)}, and during Czochralski growth^{6, 7, 13, 14)}. This paper examines the developing process of laminar Marangoni convection in a silicone oil liquid bridge under microgravity conditions supported by parabolic flight.

1. Experiment

Figure 1 shows a schematic diagram of liquid bridge formation. The liquid bridge was suspended between two coaxial aluminum disks of 15 mm diam-

eter. The length of the liquid bridge was varied from 5 to 20 mm. Silicone oil (KF-96: 5, 10, 20, 50 cSt, and KF-56: 15 cSt, manufactured by Shin-etsu Chemical Co. Ltd.) was used as the test liquid. Spherical aluminum particles with a diameter of 50 μm were used as a tracer to observe the velocity distribution. Thermally sensitive liquid crystals (W-40: cholesteric type, manufactured by Nippon Capsule Products Co. Ltd.) which change their color from red at 37.5°C, green at 40.0°C and blue at 42.5°C were used as a tracer to observe the temperature distribution inside the liquid bridge. Marangoni convection was visualized by the light cut sheet technique.

Figure 2 shows the sequence of the parabolic flight experiment. After preparation during level flight (1 g), (i) to obtain the maximum velocity, the airplane noses down (1.2 - 0.6 g, 45 sec.); (ii) to proceed to parabolic flight, the airplane noses up (2.5 g, 15 sec.); (iii) during parabolic flight, a short-term microgravity condition (10^{-2} g, 20 sec.) was achieved; (iv) to recover the flight path, the air plane noses up again (2 g, 10-15 sec.). In this experiment, during the preparation time a liquid bridge of 1 mm length with some temperature difference was formed. After the microgravity condition was achieved, firstly the lower disk moved downward, silicone oil was injected and liquid bridge was extended; then the developing process of laminar Marangoni convection was observed under a constant temperature difference between the two disks; and finally the lower disk moved upward and liquid bridge was closed.

2. Analysis

The theoretical model is shown in Fig. 3. Velocity and temperature distributions in a liquid bridge were

* Received July 8, 1993. Correspondence concerning this article should be addressed to A. Hirata.

Fig. 1 Schematic diagram of liquid bridge formation

Fig. 2 Sequence of parabolic flight

- ① Liquid bridge extension
- ② Observation of Marangoni convection
- ③ Liquid bridge closing

assumed to be two-dimensional and axis-symmetric. The basic equations and boundary conditions are described by the following equations.

The equation of continuity:

$$\rho \left(\frac{\partial U}{\partial R} + \frac{U}{R} + \frac{\partial W}{\partial Z} \right) = 0 \quad (1)$$

The equation of motion:

$$\rho \left(\frac{\partial U}{\partial t} + U \frac{\partial U}{\partial R} + W \frac{\partial U}{\partial Z} \right) = -\frac{\partial P}{\partial R} + \frac{1}{R} \frac{\partial}{\partial R} (R \tau_{RR}) + \frac{\tau_{\theta\theta}}{R} + \frac{\partial \tau_{RZ}}{\partial Z} \quad (2)$$

$$\rho \left(\frac{\partial W}{\partial t} + U \frac{\partial W}{\partial R} + W \frac{\partial W}{\partial Z} \right) = -\frac{\partial P}{\partial Z} + \frac{1}{R} \frac{\partial}{\partial R} (R \tau_{RZ}) + \frac{\partial \tau_{ZZ}}{\partial Z} + \rho g \quad (3)$$

where

$$\tau_{RR} = -2\mu \frac{\partial U}{\partial R}, \quad \tau_{\theta\theta} = -2\mu \frac{U}{R}$$

$$\tau_{ZZ} = -2\mu \frac{\partial W}{\partial Z}, \quad \tau_{RZ} = -\mu \left(\frac{\partial W}{\partial R} + \frac{\partial U}{\partial Z} \right)$$

The equation of energy:

$$\rho C_p \left(\frac{\partial T}{\partial t} + U \frac{\partial T}{\partial R} + W \frac{\partial T}{\partial Z} \right) = \lambda \left\{ \frac{1}{R} \frac{\partial}{\partial R} \left(R \frac{\partial T}{\partial R} \right) + \frac{\partial^2 T}{\partial Z^2} \right\} \quad (4)$$

Boundary conditions:

$R = 0$ (along axial axis);

$$U = 0, \quad \frac{\partial W}{\partial R} = 0, \quad \frac{\partial T}{\partial R} = 0 \quad (5)$$

Fig. 3 Theoretical model

Table 1. Physical properties of silicone oil

KF-96: 10 cSt* KF56: 15 cSt**				
ρ	density	[kg/m ³]	9.276×10^2	9.84×10^2
μ	viscosity	[kg/m/s]	8.974×10^{-3}	1.29×10^{-2}
ν	kinematic viscosity	[m ² /s]	9.674×10^{-6}	1.31×10^{-5}
λ	thermal conductivity	[J/m/s/°C]	7.4×10^{-3}	7.9×10^{-3}
C_p	specific heat	[J/kg/°C]	9.5×10	9.3×10
β	thermal expansion coefficient	[1/°C]	1.08×10^{-3}	1.07×10^{-3}
$\partial\sigma/\partial T$	temperature dependence of interfacial tension	[N/m/°C]	6.8×10^{-5}	7.02×10^{-5}
			* at 27°C	** at 30°C

$R = 1$ (free surface);

$$U = 0, \quad \mu \frac{\partial W}{\partial R} = \frac{\partial \sigma}{\partial T} \frac{\partial T}{\partial Z}, \quad \lambda \frac{\partial T}{\partial R} = h_{air} (T - T_{air}) \quad (6)$$

$Z = 0$ (bottom interface);

$$U = 0, \quad W = 0, \quad T = T_H \quad (7)$$

$Z = 1$ (top interface);

$$U = 0, \quad W = 0, \quad T = T_C \quad (8)$$

Eqs. (1)-(8) are discretized by the control volume method and solved by the SIMPLER method¹⁵⁾. Physical properties used in numerical simulations are listed in Table 1. Since the initial temperature distribution in a liquid bridge was unknown in the experiment, its initial temperature profile was studied for several cases of numerical simulation. The effect of free surface heat flux conditions and the temperature dependence of viscosity are considered in sections 3.3 and 3.4.

3. Results and Discussion

3.1 Comparison with experimental results

Forced convection induced during the liquid bridge extension stopped immediately after the liquid bridge was completely formed, and then laminar Marangoni convection was developed by the temperature difference between the two ends of the liquid bridge. In experi-

Fig. 4 Comparison between experimental results and numerical simulation results (KF-56, Gr = 15, Ma = 98, Pr = 152)
(a) streak lines (b) velocity (c) stream lines
(d) temperature (e) temperature, taking account of distortion (f) Photo in experiment

ments, the temperature difference was much higher than the critical temperature difference where the interface was kept rigid below⁸⁾. **Figure 4** shows a comparison of experimental results with numerical simulation results. In Fig. 4 (a), plots show the particle path observed in experiments, taking account of distortion by optical refraction, and arrows show the calculated streak lines. The calculated velocity, stream function and temperature distributions are shown in Figs. 4 (b), (c) and (d). The developing process of the temperature field can be seen in Fig. 4 (f) from the color of thermally sensitive liquid crystals. To compare the experimental results with numerical results directly, the calculated temperature distributions were re-made in Fig. 4 (e), taking account of distortion by optical refraction. In this experimental system, hot silicone oil was injected and then cooled from the cold disk side. In this simulation, the initial temperature condition is assumed as

$$T(t=0) = 1.0, \quad \text{for } 0.0 \leq Z \leq 0.5$$

$$T(t=0) = -2.0(Z - 0.5) + 1.0 \quad \text{for } 0.5 \leq Z \leq 1.0$$

$t = 0$ was when the liquid bridge extension was finished. There was good agreement between numerical and experimental results. It was suggested that the initial temperature distribution in the experiment is somehow similar to the assumed profile. **Figure 5** shows the time evolution of the interfacial velocity at the center and

Fig. 5 Time evolution of interfacial velocity (KF-56, Gr = 15, Ma = 98, Pr = 152)

Fig. 6 Relationship between Marangoni number and Reynolds number based on interfacial velocity

near the cold disk. After 10 seconds, the interfacial velocity was near steady state. In Figs.4 and 5, velocity distributions, especially along the free surface, developed quickly, but temperature distributions in bulk changed gradually. It was found that to obtain the steady state of the temperature field, a much longer time was needed than to obtain the steady state of the velocity field. **Figure 6** shows the relationship between the Marangoni number and the Reynolds number based on the interfacial velocity at the center of the free surface. In Fig. 6, plots show the experimental results and lines show the previous theoretical results¹²⁾. In the previous study, it was found by an order-of-magnitude analysis that in the range of $Re \ll 1$, the Reynolds number is directly proportional to the Marangoni number, and that in the range of $Re \gg 1$, the Reynolds number is proportional to the $2/3$ power of the Marangoni number and the $-1/3$ power of the Prandtl number. In Fig. 6, experimental results were in good agreement with the previous theoretical study.

3.2 Effect of initial temperature condition

To clarify the effect of the initial temperature condition inside a liquid bridge on the velocity and temperature distribution, three types of initial temperature

Fig. 7 Distribution of streak line, velocity, stream function and temperature
(KF-96: 10 cSt, Gr = 16, Ma = 109, Pr = 121)

condition were considered in numerical simulations;

Case 1: $T(t=0) = T_C$

Case 2: $T(t=0) = (T_H + T_C) / 2$

Case 3: $T(t=0) = T_H$

Figure 7 shows the calculated streak lines, velocity, stream function and temperature distributions in each case. **Figure 8** shows the interfacial velocity distributions at different times. In Fig.8, two peaks appear, one near the cold disk and the other near the hot disk. At the initial stage, the distributions evolved in the liquid bridge are quite dependent on the initial temperature condition. In case 1, the interfacial velocity develops gradually. At the initial stage, the interfacial velocity shows only one peak near the hot disk. But a second velocity peak develops quickly near the cold disk and takes over the first one. At the steady state, the maximum velocity appears near the cold disk. In case 2, the distribution develops very quickly in the liquid bridge. Two velocity peaks appear on the liquid surface from the very initial stage. The largest and second-largest interfacial velocities are established within 30 sec. In case 3, Marangoni convection develops very slowly. The thermocapillary effect is activated only near the cold disk and the induced interfacial flow is obstructed by the cold disk during the initial stage. Thus the liquid near the hot

(a) case 1 : $T(\text{at } t = 0) = T_C$

(b) case 2 : $T(\text{at } t = 0) = (T_H + T_C)/2$

(c) case 3 : $T(\text{at } t = 0) = T_H$

Fig. 8 Interfacial velocity distributions
(KF-96: 10 cSt, $Gr = 16$, $Ma = 109$, $Pr = 121$)

Fig. 9 Time evolution of the largest and second-largest
velocity (KF-96: 10 cSt, $Gr = 16$, $Ma = 109$, $Pr = 121$)

disk remains stagnant for almost 40 sec and then gradually starts its movement.

Figure 9 shows the time evolutions of the largest and second-largest interfacial velocities in above three case. In all cases, it takes 5 to 6 minutes to reach steady state because of the small thermal conductivity of silicone oil.

3.3 Effect of free surface heat flux

Figure 10 shows the calculated velocity, stream function and temperature distributions under different free surface heat flux conditions. Figure 11 shows the maximum interfacial velocity and average Nusselt number. Near the hot disk, with increasing free surface heat flux the temperature gradient increases; then the largest interfacial velocity also increases. On the other

hand, near the cold disk, with increasing free surface heat flux the temperature gradient decreases, then the largest interfacial velocity also decreases.

3.4 Effect of temperature dependence of viscosity

Figure 12 shows the velocity, stream function and temperature distribution at the steady state condition and Fig. 13 shows the time evolutions of the interfacial velocity at $z = 0.5$ cm and $z = 0.8$ cm, taking account of the temperature dependence of viscosity. From Figs. 12 and 13 it is suggested the temperature dependence of viscosity has little effect on the velocity and temperature distribution in a liquid bridge under these conditions.

Conclusions

Marangoni convection in a liquid bridge under microgravity condition during a parabolic flight experi-

Fig. 10 Distribution of velocity, stream function and temperature (KF-96: 10 cSt, $Gr = 16$, $Ma = 109$, $Pr = 121$, $T(t = 0) = T_C$)
(a) $Bi = 0.0$ (b) $Bi = 0.49$

ment was observed. Concerning the developing process of axis-symmetric Marangoni convection, the parabolic flight experimental results were in good agreement with numerical simulation results. The effects of initial temperature condition, free surface heat flux conditions and temperature dependence of viscosity on the velocity and temperature field were clarified.

Acknowledgement

The experiments were performed through Special Coordination Funds of the Science and Technology Agency of the Japanese Government, and through Generic Experiment Technology Developments of the National Space Development Agency of Japan. The authors would like to thank Nissan Motor Co. Ltd. for providing the FTX system. The authors also would like to thank Diamond Air Service Co. Ltd. for its

kind assistance during the experiment.

Nomenclature

a	= Liquid bridge radius	[cm]
Bi	= Biot number = $h_{air}r/\lambda$	[-]
C_p	= Specific heat	[J/g/°C]
D	= Liquid bridge diameter	[cm]
Gr	= Grashof number = $g\beta\Delta TL^3/\nu^2$	[-]
g	= Acceleration due to gravity	[cm/s²]
h	= Heat transfer coefficient	[W/cm²/°C]
L	= Liquid bridge length	[cm]
Ma	= Marangoni number = $-(\partial\sigma/\partial T)\Delta TL/\mu\nu$	[-]
Nu	= Nusselt number = $h_r r/\lambda$	[-]
P	= Pressure	[atm]
Pr	= Prandtl number = ν/α	[-]
R	= Non-dimensional radial position	[-]

Fig. 11 Maximum interfacial velocity and Nusselt number (KF-96: 10 cSt, Gr = 16, Ma = 109, Pr = 121, T (at t = 0) = T_C) (a) Bi = 0.0 (b) Bi = 0.49

(a) $\mu = 1.29 \times 10^{-2}$ [Pa · s]

(b) $\mu = 5.52 \times 10^{-6} \exp \{2334/(T+273)\}$ [Pa · s]

Fig. 12 Distribution of velocity, stream function and temperature (KF-56, Gr = 15, Ma = 98, Pr = 152 at average temperature)

r	= Radial position	[cm]
Re	= Reynolds number = UsL/ν	[-]
T	= Temperature	[°C]
t	= Time	[s]
U	= Velocity	[cm/s]
U_s	= Interfacial velocity at the center	[cm/s]
W	= Velocity	[cm/s]
Z	= Non-dimensional axial position	[-]
z	= Axial position	[cm]
α	= Thermal diffusivity	[cm ² /s]
β	= Thermal expansion coefficient	[1/°C]
λ	= Thermal conductivity	[J/cm/s/°C]

Fig. 13 Time evolution of the interfacial velocity at Z = 0 and Z = 0.8 (KF-56, Gr = 15, Ma = 98, Pr = 152 at average temperature)

μ	= Dynamic viscosity	[Pa·s]
ν	= Kinematic viscosity	[cm ² /s]
ρ	= Density	[g/cm ³]
$\partial\sigma/\partial T$	= Temperature dependence of interfacial tension	[dyne/cm/°C]

<Subscripts>

air	= surrounding gas
C	= cold
H	= hot
l	= liquid

Literature cited

- 1) Chang, C.E. and W.R. Wilcox: *J. Crystal Growth*, **28**, 8-12 (1975)
- 2) Chun, C.H.: *J. Crystal Growth*, **48**, 600-610 (1980)
- 3) Hirata, A., M. Kawakami and Y. Okano: *Can. J. Chem. Eng.*, **67**, 777-786 (1989)
- 4) Hirata, A., S. Nishizawa, N. Imaishi, Y. Okano, M.Z. Saghir and P.J. Paul: IAF-91-407 (1991)
- 5) Hirata, A., M. Tachibana, Y. Okano and T. Fukuda: *J. Chem. Eng. Japan*, **25**, 62-66 (1992)
- 6) Hirata, A., M. Tachibana, Y. Okano and T. Fukuda: *J. Crystal Growth*, **128**, 195-200 (1993)
- 7) Hirata, A., M. Tachibana, T. Sugimoto, Y. Okano and T. Fukuda: *J. Crystal Growth*, **131**, 145-152 (1993)
- 8) Hirata, A., S. Nishizawa, M. Sakurai and N. Imaishi: *J. Chem. Eng. Japan*, **26**, 754-756 (1993)
- 9) McNeil, T.J., R. Cole and R.S. Subramanian: *J. Colloid and Interface Sci.*, **98**, 210-222 (1984)
- 10) McNeil, T.J., R. Cole and R.S. Subramanian: *J. Am. Ceram. Soc.*, **68**, 254-259 (1983)
- 11) Okano, Y., M. Itho and A. Hirata: *J. Chem. Eng. Japan*, **22**, 275-281 (1989)
- 12) Okano, Y., A. Hatano and A. Hirata: *J. Chem. Eng. Japan*, **22**, 385-388 (1989)
- 13) Okano, Y., M. Tachibana, A. Hatano and A. Hirata: *J. Chem. Eng. Japan*, **22**, 389-394 (1989), **23**, 775-776 (1990)
- 14) Okano, Y., T. Fukuda, A. Hirata, N. Takano, T. Tsukada, M. Hozawa and N. Imaishi: *J. Crystal Growth*, **109**, 94-98 (1991)
- 15) Patankar, S.V.: "Numerical Heat Transfer and Fluid Flow", Hemisphere, Washington, D.C. (1980)
- 16) Preisser, F., D. Schwabe and A. Schermann: *J. Fluid Mech.*, **126**, 545-567 (1983)