JASMAC Conference of the Japan Society of Microgravity Appllication

PS16

アスペクト比が HZ 内の温度差と濃度差に起因するマランゴニ対流の対流構造に及ぼす影響

Effect of Aspect Ratio on Thermal and Solutal Marangoni Convection Structure in Half-Zone Melts

岸凪沙 [1], 島袋泰成 [2], 水口尚 [1]
Nagisa KISHI[1], Taisei SHIMABUKURO[2], and Hisashi MINAKUCHI[1]
[1] 琉球大学工学部, Faculty of Engineering, University of the Ryukyus, Okinawa, Japan,
[2] 琉球大学大学院理工学研究科,Graduate School of Engineering and Science, University of the Ryukyus, Okinawa, Japan
* Correspondence: mhisa522@cs.u-ryukyu.ac.jp

Abstract: In Si/Ge alloy crystal growth using the Floating Zone (FZ) method under microgravity conditions, Marangoni convection significantly influences crystal quality. This study analyzed this convection under fixed aspect ratios of 0.4 and 0.5. Results confirmed that wave numbers appear in the convection as the solutal Marangoni number increases, and these wave numbers increase as both thermal and solutal Marangoni convections strengthen. A comparison of aspect ratios 0.4 and 0.5 showed that a similar zoning of the flow regimes was possible.These findings contribute to the control of Marangoni convection for the high-quality crystal growth of Si/Ge alloys.

Keywords: numerical simulation, crystal growth, Marangoni convection

1. Introduction

SiGe alloys, which support the performance enhancement of electronic products, are difficult to produce as uniform and high-quality single crystals on Earth due to gravity-induced segregation. For this reason, the Floating Zone (FZ) method in a microgravity environment, where the influence of gravity is suppressed, has been proposed. In the FZ method, a polycrystalline feedstock is heated to form a molten zone, which is then cooled with a seed crystal to obtain a single crystal. However, the FZ method is prone to strong flows caused by surface tension, known as Marangoni convection, which hinders the growth of high-quality crystals. When temperature and concentration differences exist simultaneously, a complex convection structure is formed. Elucidating its generation mechanism and controlling the convection structure are extremely important for improving the quality of SiGe bulk single crystals.

Previous studies have primarily focused on Marangoni convection caused by temperature differences[1-2]. In previous research, it was recognized that when the aspect ratio is changed, the flow pattern of thermal Marangoni convection, the wavenumbers (m), changes[3-4]. In this study, we use the frequently employed half-zone model and also consider solutal Marangoni convection, which is caused by the surface tension difference of alloy components[5-6].

2. Numerical Methods

This study employs a half-zone model, assuming the upper part of a Si/Ge melt in the FZ method. Figure 1 illustrates the model used, where arrows indicate the directions of thermal and solutal Marangoni numbers. The analysis region is confined to the melt section, with radius a and height L. The As, defined as a/L, is set to 0.4 and 0.5 in this research. The side surface acts as a free interface, with the upper disk heated and the lower disk cooled. It is assumed that the system is under microgravity conditions, and the gas-liquid and free interface shapes are flat. Additionally, the melt is assumed to be an incompressible Newtonian fluid. The fundamental governing equations are the continuity equation (1), Navier-Stokes equation (2), energy equation (3), and diffusion equation (4). These equations are shown below.

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

$$\frac{\partial u}{\partial t} + u \cdot \nabla \boldsymbol{u} = -\frac{1}{\rho}\nabla p + \nu \nabla^2 \boldsymbol{u} \tag{2}$$

$$\frac{\partial T}{\partial t} + \boldsymbol{u} \cdot \nabla T = a \nabla^2 T \tag{3}$$

$$\frac{\partial C}{\partial t} + u \cdot \nabla C = D \nabla^2 C \tag{4}$$

The boundary conditions for the free interface, top surface, and bottom surface are presented below. :
Free surface (r=a):

$$u_r = 0 \tag{5}$$

$$\mu\left\{r\frac{\partial}{\partial r}\left(\frac{u_\theta}{r}\right)\right\} = \frac{1}{r}\left(\frac{\partial \sigma}{\partial T}\right)\frac{\partial T}{\partial \theta} + \frac{1}{r}\left(\frac{\partial \sigma}{\partial C}\right)\frac{\partial C}{\partial \theta} \tag{6}$$

$$\mu\frac{\partial u_z}{\partial r} = \left(\frac{\partial \sigma}{\partial T}\right)\frac{\partial T}{\partial z} + \left(\frac{\partial \sigma}{\partial C}\right)\frac{\partial C}{\partial z} \tag{7}$$

$$\frac{\partial T}{\partial r} = 0 \tag{8}$$

$$\frac{\partial C}{\partial r} = 0 \tag{9}$$

Upper cold disc (z=L):

$$u_r = 0, u_\theta = 0, u_z = 0, T = T_c, C = C_{si} \qquad (10)\sim(14)$$

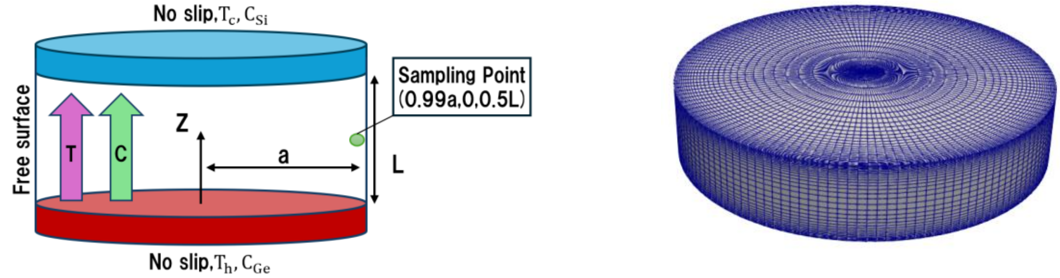
Lower hot disc (z=0):

$$u_r = 0, u_\theta = 0, u_z = 0, T = T_h, C = C_{Ge} \qquad (15)\sim(19)$$

where, u (u_r, u_θ, u_z) is the velocity vector, t is the time, p is the pressure, T is the temperature, and C is the silicon concentration. The thermal Marangoni number and solutal Marangoni number, are presented below.

$$Ma_T = -\frac{\partial \sigma}{\partial T}\frac{\Delta T L}{\mu \upsilon}, Ma_C = \frac{\partial \sigma}{\partial C}\frac{\Delta C L}{\mu \upsilon} \qquad (20), (21)$$

The physical properties of Si/Ge (Si_xGe_{1-x}) used in this study are listed in Table 1. The open-source CFD software OpenFOAM was utilized. The governing equations (Equations 1-4) were discretized using the Finite Volume Method (FVM), and the PISO algorithm was employed as the unsteady algorithm.



(a) (b)

Figure 1. Geometry of the analytical liquid bridge(a)
and mesh geometry of the analytical model at As=0.4(b).

Table 1. Physical properties of Si_xGe_{1-x}.

Kinematic viscosity $\boldsymbol{\upsilon}$	1.4×10^{-7} [m²/s]
Thermal Diffusion Coefficient $\boldsymbol{\alpha}$	2.2×10^{-5} [m²/s]
Diffusion coefficient $\boldsymbol{D}$	1.0×10^{-8} [m²/s]
Prandtl number $\boldsymbol{Pr}$	6.37×10^{-3} [-]
Schmidt number $\boldsymbol{Sc}$	14.0 [-]

3. Results and discussion

3.1. Description of Wavenumbers(m)

From the analysis results, eight different wavenumbers were observed at an aspect ratio of 0.4, as shown in Figure 2. The classification of wavenumbers is divided by wavenumbers and whether it is steady or unsteady. As shown in Figure 3, unsteady flows are oscillatory flows, and the rotation and contraction of wavenumbers are observed.

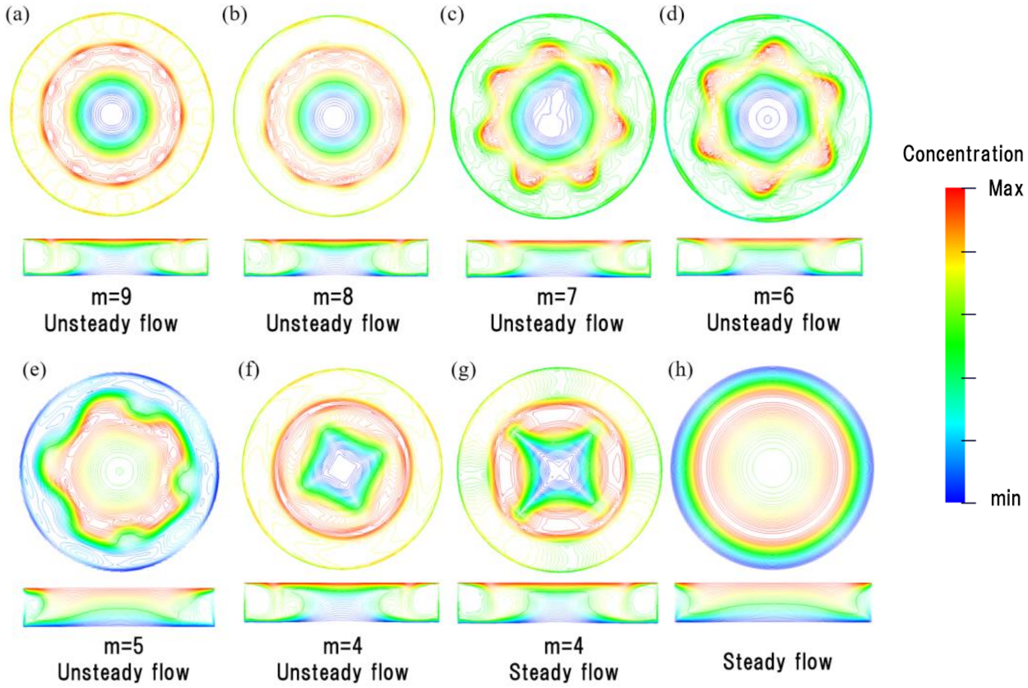
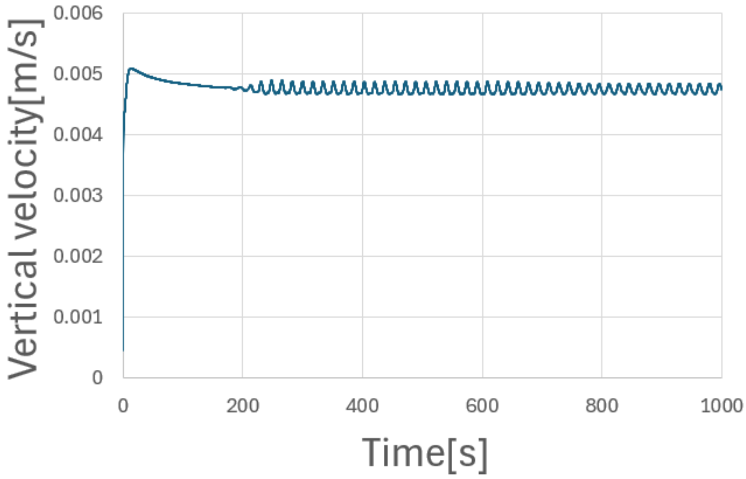
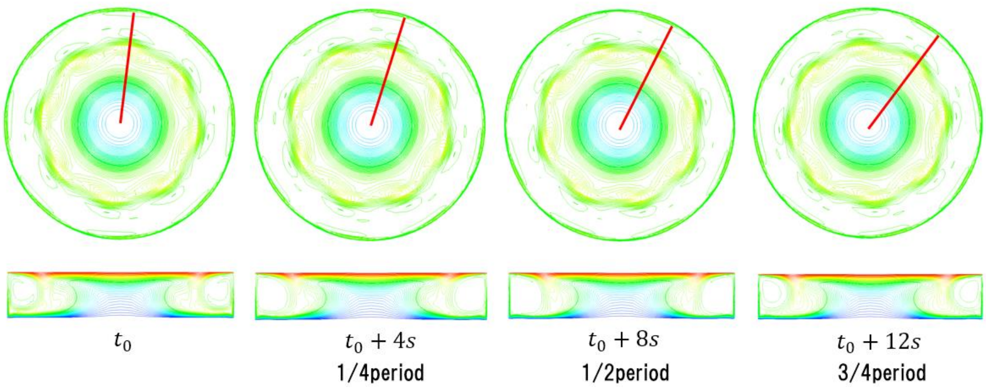


Figure 2. Horizontal Concentration Distribution:
(Ma_C ,Ma_T)=(a)(1607,3571),(b)(1607,3124),(c)(1607,1071),(d)(1607,714),(e)(1607,0),(f)(357,3571),(g)(179,3571)and(h)(179,0).



(a)



(b)

Figure 3. Oscillatory flow represented by the vertical velocity (a) and rotation of convection (b) at (Ma_C=1786, Ma_T =2500).

The observed wavenumbers are represented as a convection structure map, with the horizontal axis indicating the solutal Marangoni number (Ma_C) and the vertical axis indicating the temperature Marangoni number (Ma_T), as shown in Figure 4. The convection structure map for an aspect ratio of 0.4 is shown with its zones.

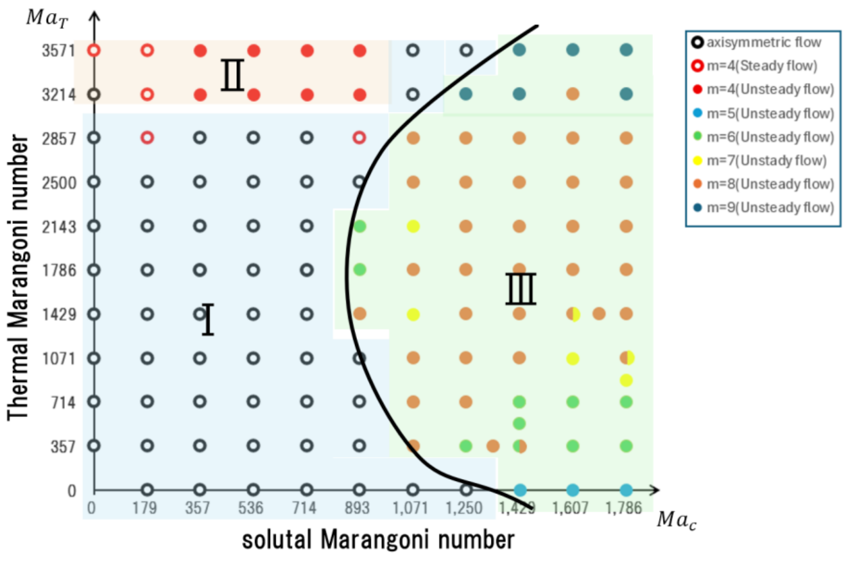


Figure 4. Flow map,(Ma_T, Ma_C) at As=0.4.

In Figure4, Zone I is defined as axisymmetric steady flow, Zone II as steady flow with wavenumbers, and Zone III as oscillatory flow. Specifically, Zone II is a region that includes both steady flows with wavenumbers and oscillatory flows with the same wavenumber. In Zone III, the wavenumber showed a tendency to increase as both the solutal Marangoni number and the thermal Marangoni number increased. This phenomenon is considered to be due to the superposition of the two flows, leading to an increase in wavenumber. For instance, in the case of solutal Marangoni convection alone (on the horizontal axis), a wavenumber 5 oscillation was observed at *Mac*=1429, but with co-directional coexistence, a wavenumber 8 oscillation occurred at a lower *Mac*=893.

Zone I is a region of steady flow where the influence of both thermal Marangoni convection and solutal Marangoni convection is limited. As the flow transitions from Zone I to Zone II, the influence of the thermal Marangoni convection begins to emerge, leading to the appearance of wavenumbers. As the solutal Marangoni number increases, a transition from steady flow to oscillatory flow occurs within Zone II.

From Zone II to Zone III, the influence of the solutal Marangoni number becomes even greater, and the wavenumber increases when a critical value is exceeded. Similarly, a transition from steady flow to oscillatory flow occurs when a critical value of the solutal Marangoni number (*Mac*) is exceeded, also from Zone I to Zone III.

At an aspect ratio of 0.4, in regions with a high solutal Marangoni number and a low thermal Marangoni number (*m*=5), irregular flows that did not involve rotation or contraction were observed. The points indicated in two colors in Figure 4 show that two types of wavenumbers coexist. This suggests that the coexistence of solutal and thermal Marangoni convections becomes unstable wavenumber.

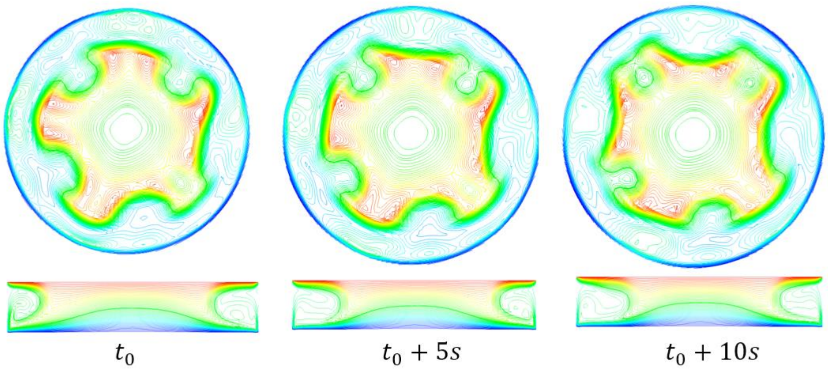


Figure 5. Temporal Evolution of Wavenumber at *m*=5(*Mac*=1786, Ma_T =0).

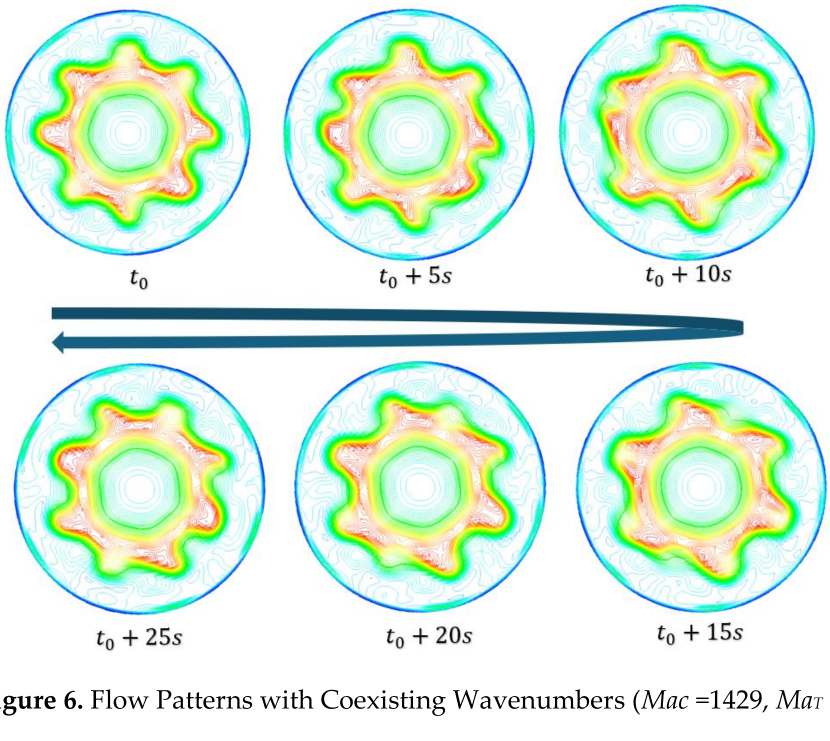


Figure 6. Flow Patterns with Coexisting Wavenumbers (*Mac* =1429, Ma_T =357).

Figure 7 shows a convection structure map for an aspect ratio of 0.5, which is zoned in the same manner as Figure 4. A higher wavenumber was observed at an aspect ratio of 0.4 than at 0.5. The "superposition phenomenon" was confirmed for both aspect ratios, where the maximum wavenumber from solutal Marangoni number alone plus the maximum wavenumber from thermal Marangoni number alone matches the maximum wavenumber observed in the map. This suggests that solutal Marangoni flow and thermal Marangoni flow coexist and mutually strengthen the flow, leading to the generation of more complex flow patterns.

The black lines in the flow maps (Figure 3 and Figure 4) indicate the critical values at which the flow transitions to unsteady state. Both maps show a bulging shape at the critical boundaries, but for *As*=0.4, the bulge is in the Ma_T range of 1429~2143, while for *As*=0.5, it is in the Ma_T range of 357~1429. This suggests that as the aspect ratio decreases, the circulation space within the liquid column becomes more restricted, causing the bulge in critical values to appear at higher Ma_T values. Observing Mac =893 in Figure 7, initially, the flow is axisymmetric and steady as it is not significantly affected by either the solutal Marangoni number or the thermal Marangoni number. Subsequently, as the thermal Marangoni number increases, it crosses a critical point and transitions to oscillatory flow. Furthermore, when the thermal Marangoni number increases and becomes dominant, the flow temporarily returns to a steady state and then transitions again to an oscillatory flow influenced by the thermal Marangoni number. The black line is bulging because the flow becomes unstable when solutal and thermal Marangoni convections coexist.

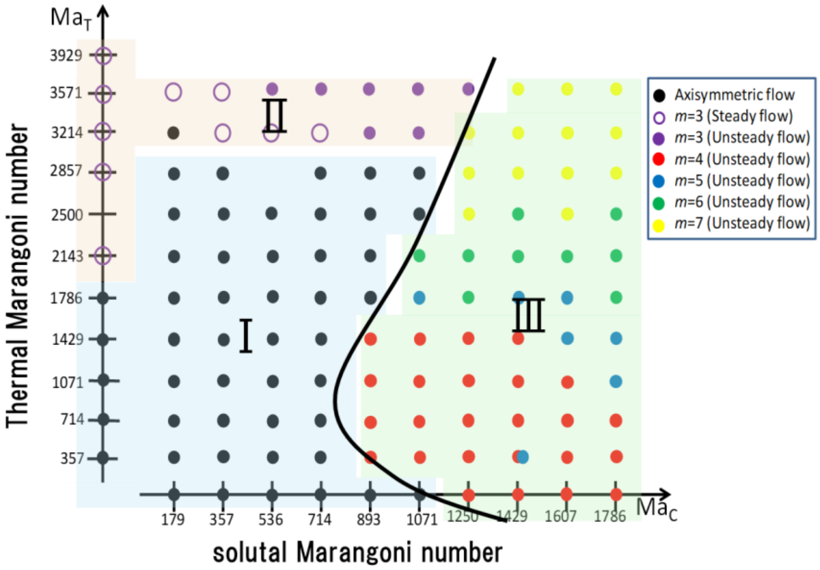


Figure 7. Flow map,(Ma_T, Mac) at *As*=0.5.

4. Conclusions

In this study, we analyzed the convection structures at aspect ratios of 0.4 and 0.5 for thermal and solutal Marangoni convections in the half-zone liquid bridge using OpenFOAM, leading to the following conclusions.

・It was demonstrated that the convection maps for aspect ratios of 0.4 and 0.5 can each be classified into three main zones: axisymmetric steady flow, steady flow with wavenumbers, and oscillatory flow.

・Changes in the aspect ratio were confirmed to influence the distribution of oscillatory flow in the convection map.

・In specific regions at an aspect ratio of 0.4, unstable flows accompanied by aperiodic wavenumber fluctuations and complex transition behaviors were observed.

References

1) C.W.Lan , J.H.Chian : Three-dimensional simulation of Marangoni flow and interfaces in floating-zone silicon crystal growth, J. Cryst. Growth, 230 (2001) 172.
2) Z.Zeng, H.Mizuseki, K.Simamura, T.Fukuda, K.Higashino, Y.Kawazoe: Three-dimensional oscillatory thermocapillary convection in liquid bridge under microgravity, Int. J. Heat Mass Transf., 44 (2001) 3765.
3) Z.Zeng, H.Mizuseki, K.Higashino,Y.Kawazoe:Direct numerical simulation of oscillatory Marangoni convection in cylindrical liquid bridges, J. Cryst. Growth, 204(1999) 395.
4) M.Lappa, R.Savino, R.Monti:Three-dimensional numerical simulation of Marangoni instabilities in liquid bridges: influence of geometrical aspect ratio, Int. J. Numer. Meth. Fuids 36(2001) 53.
5) H.Minakuchi, Y.Okano, S.Dost: Effect of thermo-solutal Marangoni convection on the azimuthal wave number in a liquid bridge, J. Cryst. Growth, 468 (2017) 502.
6) H.Minakuchi, Y.Takagi, Y.Okano, S.Gima, S.Dost: The relative contributions of thermo-solutal Marangoni convections on flow patterns in a liquid bridge, J. Cryst. Growth, 385(2014) 61.