

Recent Developments in Oscillatory Marangoni Convection

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Abstract: A Marangoni Convection Modeling Research group was formed in Japan in order to investigate oscillatory thermocapillary flow systematically over a wide range of Prandtl number (Pr). The research by the group represents the current status of the subject. The present article reports the work done by the group members. The work is divided into three Pr ranges (low, medium and high) because the cause of oscillations is different in each range. For the low-Pr case, the transition to oscillatory flow is preceded by a steady bifurcation to three-dimensional convection. For the first time an experimental proof of this first transition is provided by means of very precise measurements of temperature at different azimuthal positions. Then it is discussed that the causes of oscillations in low and medium Pr ranges are well understood. However, the oscillatory flow of high Pr fluid is not yet fully understood. The critical conditions for high Pr fluid are extremely sensitive to the free surface heat transfer in some situations but very insensitive in other situations.

Keyword: Marangoni convection, liquid bridge, oscillations

1 Introduction

Surface tension exists on the interface of two different and immiscible fluids. Generally this surface tension is a function of temperature, concentration or electrical potential depending on the nature of the fluid. When a temperature gradient exists on the interface, the surface tension varies along the interface, resulting in bulk fluid motion, called thermocapillary flow. Thermocapillary flow tends to be overwhelmed by buoyancy-driven flow in normal gravity, but in the reduced gravity environment of space it could become an important driving force [Ostrach (1982)]. Also, it becomes possible in microgravity to perform containerless materials processing with less restrictions on the size. It was thought that since buoyancy generated transport is considered to be detrimental to growth of some high quality crystals on earth, a microgravity environment offers the opportunity to grow large and high quality crystals. For this reason, the subject of thermocapillary flow has been given much attention in the last three decades as more opportunities became available to perform experiments in microgravity.

Since one of the main motivations to go to space was to perform containerless processing of materials, a zone-melting technique called float-zone process was considered to be a promising technique in microgravity. Because of this history, thermocapillary convection in a simulated float-zone configuration, called the half-zone or liquid bridge configuration, has been investigated extensively in the past [see, for example, Lappa (2004)]. In this configuration a liquid column is suspended between two differentially heated cylindrical walls, as illustrated in Fig. 1. The present paper focuses on this configuration. The coordinate system employed herein is also shown in Fig. 1.

When we discuss the importance of thermocapillary flow in various applications, it is convenient to use a dimensionless parameter called Prandtl number (Pr). The Prandtl number is defines as Pr = ν/α, where ν is the fluid kinemtaic viscosity and α is the thermal diffusivity. In such applications as crystal growth from melts, we are dealing mainly with liquids with Pr much less than unity. For example, Pr of Ge and Si are on the order of 0.01. Many experiments have been per-

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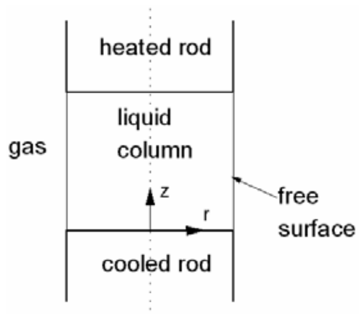

Figure 1: Half-zone (or liquid bridge) configuration

formed with high Pr fluids, mainly silicone oils with Pr = 20 – 100, because it is easier to perform experiments with these fluids. Therefore, we must consider this wide range of Pr when we discuss thermocapillary flow.

Because of past work, steady thermocapillary convection is well understood under various configurations and over a wide range of Pr. It is known that thermocapillary flow becomes three-dimensional and time-dependent (usually oscillatory) under certain conditions. Although the subject of oscillatory thermocapillary convection has important implications to various applications, our understanding of it is not as complete as with steady convection. For this reason NASDA (National Space Development Agency of Japan) assembled a team of scientists in order to perform a comprehensive investigation of oscillatory thermocapillary flow. The project was called 'Marangoni Convection Modeling Research'. Much was accomplished by the group before NASDA was reorganized as JAXA (Japan Aerospace Exploration Agency) in 2003. The group's main focus now is to perform microgravity experiments on oscillatory thermocapillary flows aboard the International Space Station (ISS). The project members, at various times, are: N. Imaishi, Y. Kamotani, M. Kawaji, H. Kawamura, H.C. Kuhlmann, S. Matusumoto, K. Nishino, and S. Yoda. IHI Aerospace Co. is involved in the design and fabrication of the experimental cells. As the lead scientist, the first author (Kamotani) is supervising the group effort. The results obtained by the group members have been reported annually in NASDA Technical Memoranda and JAXA Research and Development Report. The present paper is based mainly on the results obtained so far in this project.

2 Important dimensionless parameters for thermocapillary flow

As in other fluid flow and heat transfer applications, it is useful to discuss the results in terms of dimensionless parameters. As illustrated in Fig. 1, a liquid bridge of diameter D and length L is considered. We are interested mainly in cylindrical bridge (straight free surface) in the present paper. Temperature difference $\Delta T = T_H - T_C$ is imposed across the length of the bridge, where T_H and T_C are the hot and cold wall temperatures, respectively. The important dimensionless parameters for steady thermocapillary flow in the liquid bridge configuration, in the absence of gravity, are known to be: Marangoni number (Ma= $\sigma_T \Delta T L/\mu\alpha$), Pr= ν/α, and bridge aspect ratio (Ar= L/D), where σ_T is the temperature coefficient of surface tension (absolute value) and μ is the dynamic viscosity. Ma represents the ratio of thermocapillary convection to conduction heat transfer. Instead of Ma, surface tension Reynolds number (Re) is sometimes used. Re is equal to Ma/Pr and represents the ratio of inertia to viscous forces of thermocapillary flow.

Another important parameter in the present paper is related to the heat transfer between the liquid and the surroundings through the free surface. As for a dimensionless parameter representing the total free surface heat transfer rate, Kamotani et al (2003) uses a parameter based the total heat transfer rate Q, which is called (modified) Biot number, Bi. Bi is defined as

$$Bi = \frac{Q}{2\pi k L \Delta T} \tag{1}$$

where k is the thermal conductivity of the liquid. The total heat transfer rate can be expressed as $Q = \overline{h}\Delta T_g(\pi DL)$, where $\overline{h}$ is the average heat transfer coefficient and ΔT_g is the characteristic temperature difference between the liquid and the surrounding gas. A conventional Biot number is usually based on $\overline{h}$ (e.g. $\overline{h}L/k$). If we adopt a conventional Biot number, we must also specify ΔT_g

to compute Q. Instead of specifying two parameters, we represent the heat transfer rate by the above modified Biot number for simplicity. Bi is defined such that it is positive when heat is lost (or positive when gained) at the free surface. There are other parameters which are related to buoyancy, liquid column shape, and onset of oscillations. They will be introduced in the following discussion as needed.

From the beginning, our efforts have been split into three groups covering three different Prandtl number (Pr) ranges: low Pr range (Pr is of order 0.01), medium Pr range (Pr of order unity), and high Pr range (Pr above about 10), since the flow in each Pr range has some unique features. Therefore, the following discussions are also divided into three different Pr ranges.

3 Development in low Prandtl range

Since Pr is much less than unity in this case, Re is much larger than Ma. Under the experimental conditions where the onsets of oscillatory flows were detected in float-zone melts, Ma and Re are on the order of 10^2 and 10^4, respectively. Therefore, inertia effects are very important in the oscillation phenomenon, and thus Re is the more appropriate parameter to use than Ma. However, since Ma is more often used in literature, Ma is used herein to make comparisons of various results easier.

Eyer et al. (1984) showed, in their microgravity experiment with silicon melt, that time-dependent thermocapillary flow causes microstriations in the crystal. Since then several experiments were performed on time-dependent thermocapillary flows in various melts both in microgravity and normal gravity, including Jurisch (1990) with Mo and Nb, Levenstam et al. (1996) with Si, Nakamura et al. (1998) with Si, and Cröll et al. (1998) with GaSb. There are some studies in the half-zone configuration in relatively low temperature [Yang and Kou (2001) with Sn, and Han et al. (1996) with Hg]. According to these investigations the critical Ma for a transition from steady to oscillatory state ranges from 200 to 900 approximately.

According to linear stability analysis [Wanschura et al. (1995)] as well as three-dimensional numerical simulations of the flow for low Pr fluids [Imaishi et al. (2001); Lappa, Savino and Monti (2001); Leypoldt et al. (2005); Gelfgat et al. (2005); Lan and Yeh (2005); Lappa (2005)], the originally axisymmetric steady flow in the liquid-bridge configuration becomes nonsymmetric, three-dimensional steady flow beyond a critical Ma (or Re). This critical Ma is called the first critical Ma or Ma_{c1}. This is a result of hydrodynamic instability associated with inertia effects [Wanschura et al. (1995)].

With further increase in the temperature difference, the flow undergoes a second transition: the steady three-dimensional flow becomes time-oscillatory. This transition occurs at the second critical Ma, Ma_{c2}. This is the transition observed in the aforementioned experiments. The transition has also been investigated in numerical simulations [e.g. Imaishi et al. (2001); Leypoldt et al. (2005)]. The numerically predicted Ma_{c2} is somewhat smaller than the aforementioned experimental values.

Now that numerical simulation results have become available, one can compare experimental and numerical results in detail in order to better understand the transition process. This was the motivation of the low Pr research work by our group. In order to obtain more accurate and detailed results than the past experiments, JAXA built a high-vacuum chamber to conduct a liquid bridge experiment with molten tin (Pr = 0.009). Tin (Sn) was chosen over Si because it has a lower melting point (= 505K), which makes it easier to melt the sample. One important problem with the Sn experiment was that the melt surface became covered by tin oxides (SnO and SnO_2) easily in the test temperature range (below 800K). After much work this problem has successfully been solved. The setup is illustrated in Figs. 2 and 3. It is described in detail in Takagi et al. (2001).

A liquid bridge of molten tin is formed in high vacuum ($\sim 10^{-5}$ Pa). The zone diameter is variable from 3 to 7 mm and the range of Ar covered is from 0.4 to 1.1. The top and bottom walls are made of iron. E-type thermocouples that are inserted into the bottom cold wall and radiation

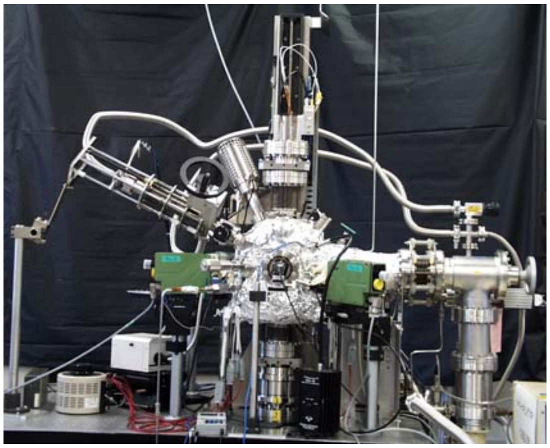

Figure 2: Low Prandtl number experimental facility at JAXA

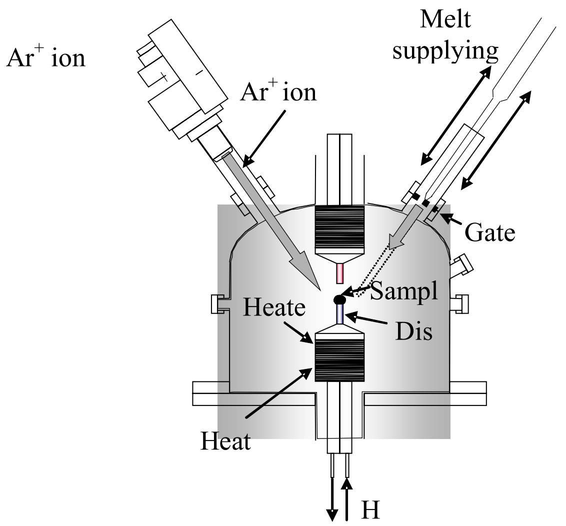

Figure 3: Details of test section of low Prandtl number facility

thermometers are used to monitor the fluid temperature changes. Imaishi and his group are performing numerical simulations and the staff scientists at JAXA are conducting the experimental work.

According to Imaishi et al. (2001), Ma_{c1} is about 20 for Pr = 0.01 and Ar = 0.5. However, no experimental proof of this first transition is available because it is difficult to detect this critical point experimentally for two reasons. One reason is that the critical temperature difference between the walls (ΔT_{c1}) is small. For the experimental system with 5-mm diameter tin, ΔT_{c1} is equal to 0.3°C. The second reason is that since conduction heat transfer is dominant at the time of instability, it is difficult to detect the temperature variation in the azimuthal direction after the instability. Nevertheless, JAXA succeeded in detecting the transition [Matsumoto et al. (2005)]. The three thermocouples placed on one side of the liquid bridge have nearly the same readings at first but they show sudden increases beyond a certain ΔT (see Fig. 4).

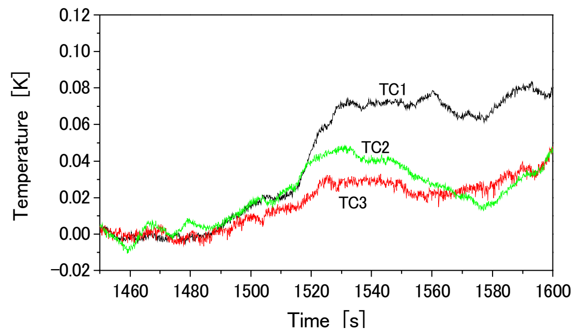

Figure 4: Typical thermocouple outputs near Ma_{c1} (Ar = 0.9, ΔT_{cr} = 1.7°C)

After the change the three thermocouples have different but nearly steady outputs, within the experimental noises. This clearly suggests that the flow structure undergoes a transition from axisymmetric to non-axisymmetric steady flow. The values of Ma_{c1} determined for various Ar are compared with the predictions in Fig. 5. The agreement is reasonable knowing the difficulty in determining ΔT_{c1} experimentally.

According to the energy balance analysis by Wanschura et al. (1995), the flow becomes unstable due to shear instability. As in other shear flows, there exists a large velocity gradient normal to the main flow direction (the radial direction in the present flow) so that the disturbance velocity in the normal (radial) direction destabilizes the flow when the inertia effects become sufficiently strong.

With further increase in the temperature difference, the flow undergoes a second instability: the steady three-dimensional flow becomes time-oscillatory. This transition occurs at the second critical Ma (Ma_{c2}). For Pr = 0.01, Ma_{c2} is about 80

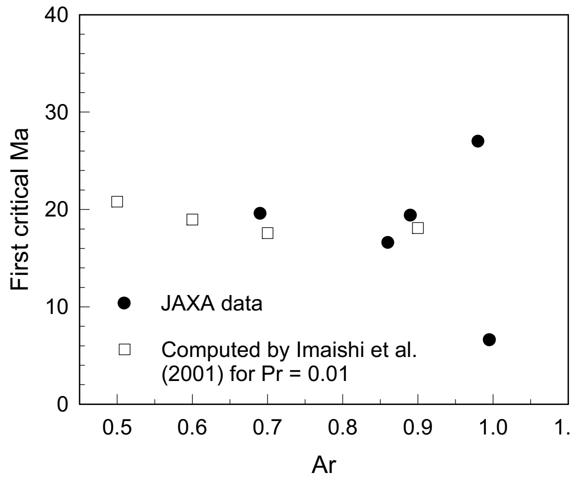

Figure 5: Measured and computed first critical Ma

depending on Ar. Since Ma is still small for convection transfer to be important, the second transition is also caused mainly by inertia effects. Since the resultant time-dependent convection has important implications to crystal growth, it has been investigated numerically and experimentally by several investigators in the past, as discussed above. The present work is intended to obtain accurate experimental data on the oscillatory flow under controlled conditions.

In the experiments at JAXA, ΔT is increased slowly, at a rate of about 0.3°C/min, and the thermocouple outputs are monitored to detect the onset of oscillations. Since the thermocouple outputs are amplified (5×10^4 times) to increase the sensitivity, there are inherent noises, so the outputs have to be analyzed carefully to identify the onset of oscillations. As illustrated in Fig. 6 for Ar = 0.6, relatively high frequency ($\sim$ 1.2 Hz) oscillations are detected soon after Ma_{c2}. They are followed, with increasing ΔT, by low ($\sim$ 0.02 Hz) and medium ($\sim$ 0.3 Hz) frequency oscillations. The simulation by Imaishi [discussed in Matsumoto et al. (2005)] shows the same sequence with the corresponding frequencies of 2.5, 0.04 and 0.25 Hz, so they are in reasonable agreement.

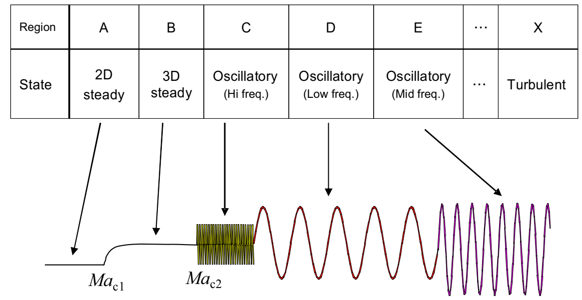

Figure 6: Oscillation patterns after Ma_{c2}

The experimental values of Ma_{c2} are compared with those predicted by simulation for various values of Ar in Fig. 7. The experimental data scatter but they are near the predicted values. The prediction shows a peak around Ar = 0.6, but it is difficult to confirm it within the experimental scatters.

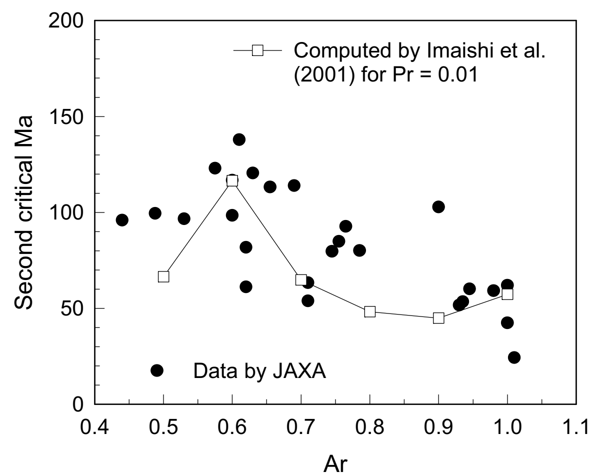

Figure 7: Measured and computed Ma_{c2}

In summary, the present work has accurately identified the two transitions predicted by stability analyses and simulations. So far, all the information has been gathered by investigating the temperature fields by the thermocouples. It is very useful to obtain additional information regarding the velocity fields, since the velocity field undergoes a larger change than the thermal field after the instabilities in the ranges of Ma and $R\sigma$ of present interest. Kakimoto et al. (1989) has used an X-ray radiography technique to visualize fluid motion in silicon melt. Since this technique requires relatively large tracers (about 1 mm in diameter), its spatial resolution is not sufficient to detect the onset of oscillations accurately in the present work. Therefore, a novel three-dimensional ultrasonic visualization (3D-UV) technique is being developed at JAXA [Matsumoto et al. (2005)]. $LiNbO_3$ (LN) transducers are used to transmit ultrasonic beams and also to receive echo beams. The tracers are Fe- and Ni-plated Shirasu-balloons and they are about 100 μm in diameter.

4 Development in medium Prandtl range

When Pr is of order unity, Ma $\approx$ Re so that the inertia and convection effects are on the same order. According to the linear stability analysis by Wanschura et al. (1995) for Pr = 4 and the numerical simulations work by Leypoldt, Kuhlmann and Rath (2000) for Pr = 4 and 7, steady flow becomes oscillatory beyond a certain Ma_{cr} with the appearance of hydro-thermal waves. According to the energy balance analysis by Wanschura et al. (1995) for Pr = 4, the flow becomes unstable due to convection effects. There exists a relatively large temperature gradient in the radial direction due to strong convection. When the hydro and thermal waves propagating in the azimuthal direction have a certain phase difference, the convection in the radial direction by the disturbance flow amplifies the waves if it is sufficiently strong. In relation to the high Pr case to be discussed later, it is important to note that the flow in the medium Pr range becomes unstable in the bulk region, away from the regions near the hot and cold walls.

Schwabe et al. [e.g. Schwabe et al. (1978), Preisser et al. (1983), Velten et al. (1989)] obtained extensive data with $NaNO_3$ (Pr = 0.7) and KCl (Pr = 1) melts. However, since the melting points of these materials are much above room temperature (307˚C and 770˚C for $NaNO_3$ and KCl, respectively), the heat loss from the liquid bridge by radiation (both internal and surface) and convection is considerable but it is difficult to estimate it accurately. Chun and Wuest (1979) used alcohol (Pr $\sim$ 1) but due to its large evaporation rate it was not possible to obtain steady results.

Therefore, we decided to perform experiments and analysis in this Pr range under controlled conditions. Kawaji (experiment) and Kuhlmann (linear stability analysis) of our group are involved in this work. The experimental work is described by Kawaji et al. (2000, 2001). Acetone (Pr = 4.3) and methanol (Pr = 6.8) are used as the test fluids. Liquid bridges of 5 and 10 mm in diameter are studied. To reduce the evaporation rates and the shape changes, the liquid bridges are covered by a quartz tube with a gap of 2 $\sim$ 3 mm. However, the liquids still evaporate slowly. The evaporation rates are measured: about 0.15 and 0.05 % relative volume change per second for 5 mm diameter acetone and methanol bridges, respectively. The experiment shows that the bridge shape does not have strong effects on the critical condition, unlike in high Pr range to be discussed later. Therefore, the main effect is the evaporation heat loss at the free surface, which can be calculated from the evaporation rate. It turns out that this heat loss is significant, equivalent to Bi of about 5. The values of ΔT_{cr} were relatively small (less than about 2˚C for acetone and about 3˚C for methanol), so ΔT must be measured accurately. The experimental results with acetone can also be found in Simic, Kawaji and Yoda (2006).

The measured values of Ma_{cr} are generally much larger than the predicted values by linear stability analysis with a thermally insulated free surface (a factor of three to five). However, the heat loss due to evaporation is included in the analysis, its prediction agrees well with the experimental data [Nienhuser et al. (2000)]. Since the heat loss is large, the basic flow is altered significantly. In particular, the radial temperature gradient is reduced by the free surface heat loss. From the instability mechanism discussed above, this reduction in the radial temperature gradient reduces the convection effects and thus stabilizes the flow.

5 Development in high Prandtl range

From the beginning of oscillatory thermocapillary flow investigations many experiments have been performed in liquid bridges of high Pr fluids, such as silicone oils and alkanes, because they are much less susceptible to free surface contamination than low Pr fluids, so the experimental setup is much simpler. A review of earlier studies is given by Preisser et al. (1983). Experiments with high Pr fluids are often conducted in room air. Some experiments have been conducted in microgravity where larger liquid bridges than normal gravity can be formed [e.g. Monti et al. (1995)]. Despite all these investigations, our understanding of the cause of oscillatory thermocapillary flow is not complete. This is in contrast to the situations for low and medium Pr ranges discussed above, where the causes of oscillations are well understood based on stability analyses and simulations.

Kamotani et al. (1998a, 2000) performed microgravity experiments on oscillatory thermocapillary flow with high Pr fluid (Pr $\approx$ 25) in open cylindrical containers (see Fig. 8).

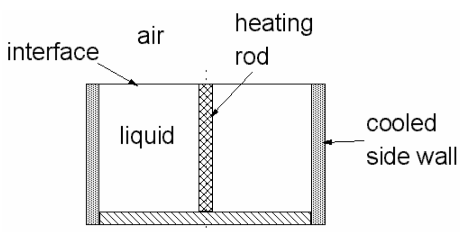

Figure 8: Thermocapillary flow experiment in open cylindrical container

There are many similarities between the oscillatory flow in the half-zone configuration and that in the cylindrical container configuration, so the results from the latter configuration are also referred to herein.

The prevailing idea about the cause of oscillations is that it is a result of hydrothermal-wave type instability, as in the medium Pr case [e.g. Wanschura et al. (1995)]. If this were the case, the critical condition will be specified by Ma_{cr}. However, available data taken in normal- and micro-gravity show that Ma_{cr} increases substantially with increasing liquid diameter, for a given Ar and Pr. Some available data of Ma_{cr} are plotted against the liquid bridge length in Fig. 9. The figure shows that Ma_{cr} varies by an order of magnitude. This should not occur if Ma, Ar, and Pr are the only important parameters. Since it takes a longer time to get a steady state with increasing bridge size, one can argue that the data for large bridges, which are taken in microgravity, may not be steady-state values, because the experimental time is limited in microgravity. However, the data taken by Kamotani et al. (1998a, 2000) in open cylindrical containers show the same trend, namely Ma_{cr} increases with container size. In their microgravity experiments, care was taken to determine Ma_{cr} under steady conditions. Apparently, some additional features must be considered to explain the oscillation phenomenon in high Pr fluids. Whether this additional feature just modifies the linear stability concept or introduces a new oscillation mechanism is still being debated.

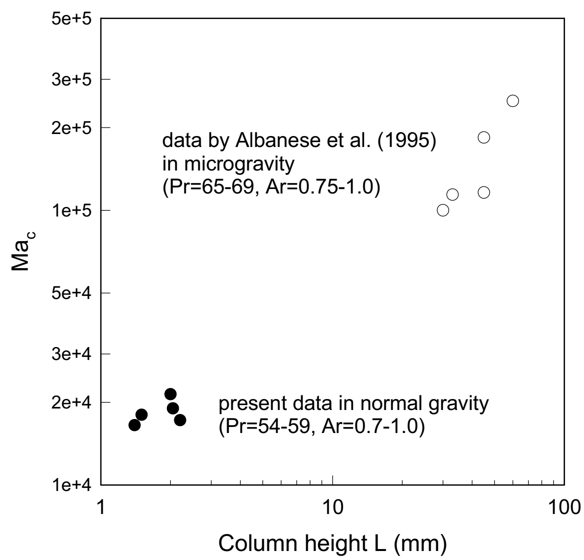

Figure 9: Critical Ma as a function of liquid column height

One important aspect of thermocapillary flow of high Pr fluid is that convection is strong when Ma is in the range where the flow is found to become oscillatory. Due to strong convection, the temperature distribution along the free surface, which is directly related to the thermocapillary driving force, is modified by the flow. Some numerically computed free surface and temperature distributions at various Ma (discussed in Kamotani and Ostrach (1998b)) are shown in Fig. 10. As Ma increases, the temperature gradient near the hot wall (near $z/L = 1$) increases, and eventually the driving force is confined to a relatively small region next to the hot wall. This region is called hot corner. There is also a cold corner (near z/L = 0) where the surface temperature gradient is also large (see Fig.10), but since the flow is driven right to the cold wall, the driving force in the cold corner is not effective in driving the whole flow. Therefore, the flow is driven mainly in the relatively small hot corner. One aspect of hot-corner driven flow is that the flow remains viscous-dominated even when Re is large [Kamotani and Ostrach (1998b)].

This appearance of hot corner near the critical condition is unique for high Pr fluid. To show this, the computed surface velocity and temperature distributions are shown in Fig. 11 for three cases corresponding to the conditions near the onset of oscillations for three different Pr fluids. As seen in

Fig. 11, except for the high Pr case (Pr = 30), one does not see concentrated driving force near the hot wall.

As discussed earlier, the flow in the low and medium Pr range becomes unstable in the bulk region. In contrast, available experimental evidence suggests that the flow in high Pr range becomes unstable starting from the hot corner. Apparently, the observation that the cause of oscillations for high Pr fluid is more complex than that for low and medium Pr fluid is related to this fact.

Because of the hot corner and for other reasons it is difficult to perform linear stability analysis or conduct direct numerical simulations of oscillations accurately at high Pr. The linear stability work by Wanschura et al. (1995) covers up to Pr = 4 and the simulation work by Leypoldt et al. (2000) goes up to Pr = 7. In the simulation work by Sim and Zebib (2004), the oscillations are investigated for Pr = 27. However, their predicted Ma_{cr}, 5.7×10^3 for Ar = 0.7, is much smaller than available experimental data. In our group, Kawamura and his co-workers are conducting simulations at high Pr . Their simulation work is performed at Pr = 28 [Hashimoto et al. (2003, 2005)]. The work includes dynamic free surface deformation. The predicted Ma_{cr} for Ar = 0.5 is 3.03×10^4

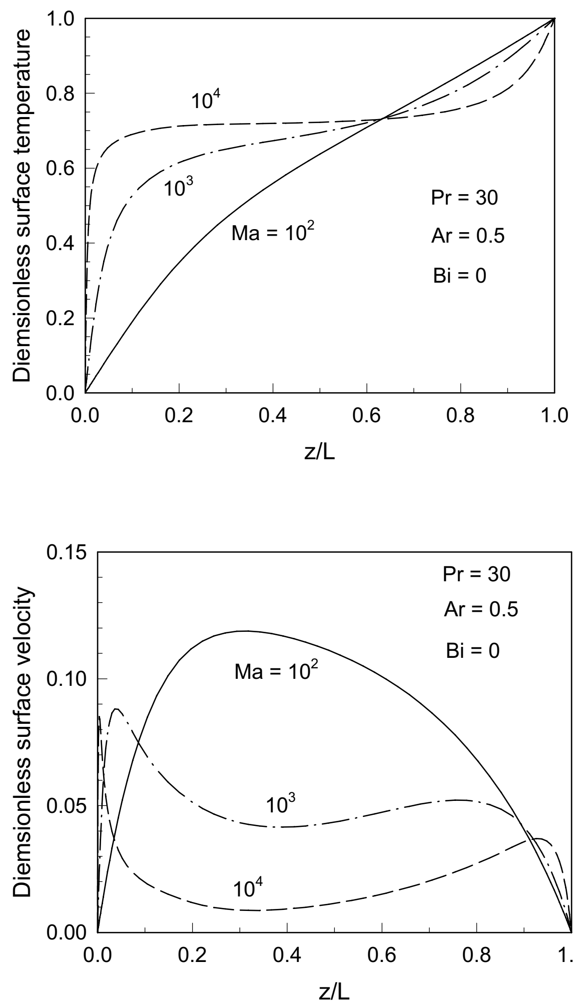

Figure 10: Computed temperature and velocity distribution along free surface for large Pr fluid

and 3.24×10^4, with and without dynamic free surface deformation, respectively [Hashimoto et al. (2005)]. The accuracy of this prediction will be discussed later.

Kamotani et al. (1984) and Masud et al. (1997) have discussed various contradictions when Ma_{cr} is used to specify the critical conditions in the liquid-bridge configuration. They also found similar contradictions for oscillatory thermocapillary flow in open cylindrical containers [Kamotani et al. (1992,1998a, 2000)]. These studies suggested a need for a different oscillation mechanism from the hydrothermal-wave instability. Based on the analyses of various experimental

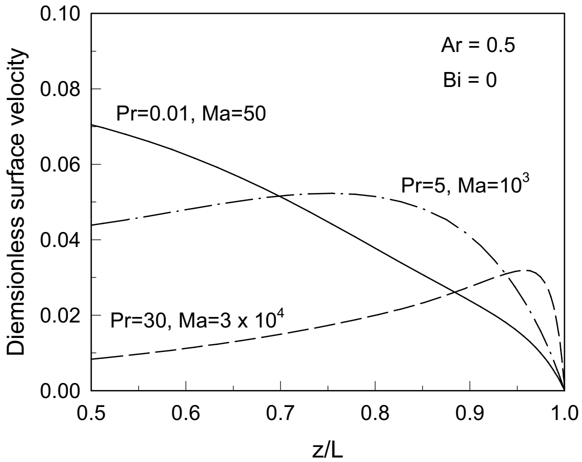

Figure 11: Velocity distributions near critical conditions for various Pr fluids

data as well as theoretical studies, Kamotani and Ostrach (1998b) hypothesized the importance of dynamic free surface deformation in the oscillation mechanism. The dynamic free surface deformation is relatively small during oscillations, so its effect is neglected by other researchers. However, since the main driving force is confined to a small hot corner, as discussed above, it is possible to affect the region significantly even by small free surface deformations. The basic concept is that the dynamic free surface deformation affects the thermal boundary layer development in the hot corner in a transient situation, which changes the driving force and the flow field. The flow field changes in such a way that the surface deformation is amplified, resulting eventually in oscillations. This three-way non-linear coupling among the deformation, velocity and temperature fields is considered to be the cause of oscillations. Since it is very difficult to vary the dynamic free surface deformation in a controlled manner experimentally and study its effect on the onset of oscillations, a direct proof may not be possible. However, there are various ways, albeit indirect, to validate this hypothesis, as discussed below.

Based on the three-way coupling hypothesis, Kamotani and Ostrach (1998b) derived a parameter called S-parameter. It is defined as

$$S = \frac{\sigma_T \Delta T}{\sigma}\frac{1}{\mathrm{Pr}}\mathrm{Ma}^{3/14} \tag{2}$$

The S-parameter represents the dynamic free surface deformation relative to the thermal boundary layer thickness in the hot corner in a transient thermocapillary flow. They also derived a similar parameter for the cylindrical container configuration [Kamotani, Ostrach and Masud (2000)]. The S-parameters seem to correlate various data on the onset of oscillations reasonably well, both in the liquid bridge and circular container configurations.

Another way to check the validity of the three-way coupling is to check their phase relations. In order for this coupling to cause oscillations, three components must maintain certain phase relations during oscillations in the hot corner. Those phase relations have been investigated by Nishino et al. (2000, 2001, 2005) in normal gravity. However, as discussed later, experiments at room temperature are very much affected by the heat transfer at the free surface. How the three-way coupling is affected by the heat transfer is not well understood. The free surface heat transfer is mainly caused by the natural convection air around the liquid bridge. Therefore, microgravity tests are needed to investigate this phase relationship accurately since the free surface heat transfer is much suppressed in microgravity.

The work by Kamotani and Ostrach (1998b) predicts how the amplitude of surface deformation just after the onset of oscillations changes with the liquid bridge diameter. This aspect is investigated also by Nishino et al. Since one can perform experiments in normal gravity only with small bridges, they can cover a limited range of diameter. Although the data by Nishino et al. appears to support the scaling law of Kamotani and Ostrach (1998b), we need to cover a wider range of diameters, in microgravity, to validate the scaling law accurately.

It should be mentioned that the importance of dynamic free surface deformation is still being debated [e.g. Kuhlmann and Nienhuser (2002)]. Therefore, more work is needed to validate, or disprove, the three-way coupling hypothesis. Microgravity tests are needed to do this. For this reason our group is scheduled to perform high Pr experiments aboard the ISS, as will be discussed later.

It has been found recently that the onset of oscillations is very sensitive to the heat transfer at the free surface [Kamotani et al. (2001a, 2003)]. In a typical test at room temperature, the average temperature of the liquid is higher than the surrounding air temperature so that heat is lost from the free surface. The heating and cooling arrangement of the experiment causes natural convection around the liquid bridge. Since this convection is relatively weak, its effect had been neglected earlier. However, in the experiments in which the free surface heat transfer is altered in various ways, Kamotani et al. (2003) found that the critical conditions change substantially. It is shown that the oscillatory flow is greatly affected by simply placing a thin disk around the liquid bridge (without touching it). Since the viscosity ratio between the surrounding air and the liquid is very small (about 0.005) but the thermal conductivity ratio is not so small (about 0.23), the effect of air motion change observed experimentally is not a hydrodynamic one but a thermal effect. Clearly, the oscillation mechanism for high Pr fluid is extremely sensitive to the free surface heat transfer.

It is very difficult to measure the free surface heat transfer rate accurately with small liquid bridges, so Kamotani et al. (2003) computed the heat transfer rate by simulating the airflow at the experimental conditions numerically. Bi is determined based on this simulation.

Typical results are presented in Fig. 12, where Ma_{cr} is plotted against Bi. As the figure shows, Ma_{cr} decreases with increasing Bi (or with increasing heat loss). Ma_{cr} becomes as small as about 6,000 when Bi is about 0.7 and remains at this value when Bi is increased further. It can be shown that even at Bi = 0.7, the total heat loss is only 30% of the overall heat transfer rate through the liquid. Fig. 12 shows that in a relatively small range between Bi = 0.3 and 0.7, Ma_{cr} changes by several factors. The oscillation mechanism must explain this high sensitivity to the heat loss.

Bi is around 0.5 in a typical room temperature test, which means that many past experiments were significantly affected by the heat loss. When the heat loss is negligible (Bi less than about 0.3),

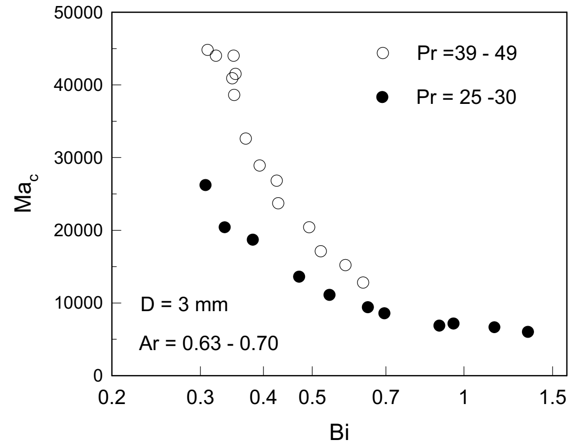

Figure 12: Critical Ma as a function of Bi

Ma_{cr}seems to approach a constant value. However, Fig. 12 suggests that the asymptotic value of Ma_{cr} depends on the diameter (suggesting that Ma is not the proper critical parameter) and Pr. For Pr = 24-28, Ma_{cr} is above 2×10^4 but below 3×10^4 depending on the diameter. The aforementioned prediction by Kawamura's group is near this range.

Why is the critical condition affected by the heat loss? In the case of medium Pr discussed earlier, Ma_{cr} increases with increasing Bi (increasing heat loss) as the heat loss reduces the radial temperature gradient in the bulk region. In the numerical simulation work by Sim and Zebib (2004), Ma_{cr} is shown to increase with increasing Bi. However, the experimental results exhibit an opposite trend. As discussed above, the hot corner is more important than the bulk region for the oscillations in high Pr fluid, so the heat loss effect is not straightforward. The opposite trend may be explained by the fact that the heat loss increases the overall free surface temperature gradient, which increases the surface velocity. The work by Kawamura's group has shown that Ma_{cr} indeed decreases with increasing heat loss when Ma_{cr} is around 2×10^4. This trend at relatively high Ma is caused by increased inertia forces due to increased velocity. However, Ma_{cr} in Fig. 12 becomes much smaller so that the inertia forces are less important. It can also be shown that the surface velocity near the critical condition at Bi = 0.7 is actually slower than that at Bi = 0.2. Therefore, increased surface velocity alone cannot explain the experimental trend of Fig. 12.

The fact that the oscillation mechanism is highly sensitive to a small change in the free surface heat transfer may be explained by the oscillation mechanism based on dynamic free surface deformation. If the dynamic free surface deformation, which is also small, is important, it is possible that the heat transfer affects the free surface deformation significantly, and consequently the critical condition is very sensitive to the heat transfer. Based on this concept, the data in Fig. 12 are replotted in terms of S-parameter in Fig. 13. In the figure, Bi is modified by $Pr^{1/2}$.

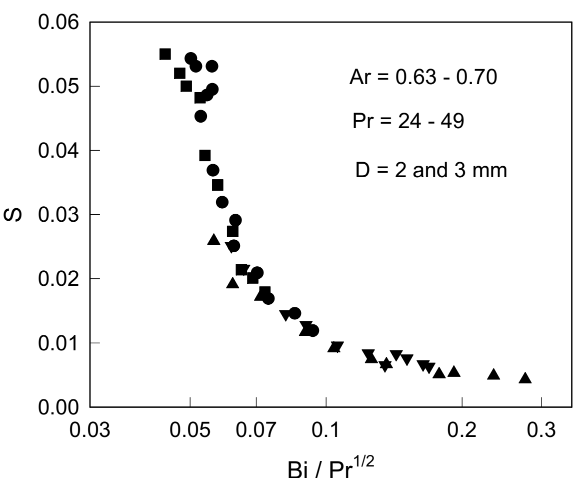

Figure 13: Critical S-parameter as a function of heat loss parameter

According to Kamotani et al. (2003), in a transient situation, the free surface heat transfer and the dynamic deformation interact as the latter affects the thermal boundary layer development along the free surface in the hot corner. The ratio of the deformation to the thermal boundary layer development is represented by $Pr^{1/2}$. Therefore, as $Pr^{1/2}$ increases, the deformation proceeds faster so that the thermal boundary layer, or the heat loss effect, is confined to a smaller layer and thus its effect is less. This is why Bi is modified by $Pr^{1/2}$ in Fig. 13. The S-parameter seems to correlate all of the experimental data well.

Although the various trends of the critical data may be explained by the free surface heat transfer and the dynamic free surface deformation, it is found that the oscillatory thermocapillary flow in open cylindrical dishes, where small dynamic free surface deformations seem to play an important role, is much less sensitive to the free surface heat loss. It is known that the critical condition is sensitive to the liquid bridge shape, called the shape effect [Masud et al. (1997)]. By decreasing the volume of the liquid, the bridge shape becomes concave. There are two branches: when the surface is neatly straight, Ma_{cr} increases with decreasing volume (increasing concavity), called the hot branch, and when the volume is below a certain value, Ma_{cr} is reduced to a relatively small value, called the slender branch. It has been shown that the above sensitivity to the free surface heat loss occurs only in the fat branch [Kamotani et al. (2001b)]. The critical condition in the slender branch is much less sensitive to the heat transfer. So, under certain situations, the critical condition is very sensitive to the free surface heat transfer but in other situations, the critical condition is hardly affected by it. At present we do not have a satisfactory explanation to this conflicting effect. Clearly more work is needed to understand this free surface heat transfer effect under various conditions. Otherwise, the cause of oscillations in high Pr fluids will remain unsolved.

As discussed above, there are some unanswered questions concerning the cause of oscillations for high Pr fluid. For this reason this group is scheduled to perform experiments aboard the ISS in near future. It will use the Fluid Physics Experiment Facility (FPEF) in the Japanese Experiment Module (JEM). The main objectives of the experiments are: (i) to determine the critical conditions under various conditions (variable diameter, Ar and liquid bridge shape), (ii) to investigate the nature and extent of dynamic free surface deformation and its role in the oscillation mechanism, and (iii) to study the effect of free surface heat loss.

In this regard, scientists from Europe and Japan are working together to formulate and design some experiments on thermocaipllary flows using the Fluid Science Laboratory (FSL) in the Columbus Module and FPEF of ISS. So, thermocapillary flow remains to be an interesting subject.

6 Conclusions

Recent developments in oscillatory thermocapillary flows in liquid bridges are reviewed. A group of investigators worked together to study the cause of oscillatory thermocapillary flows over a wide range of Pr. The cause of oscillations can be explained except in high Pr range, where it seems we need to consider some new features to explain the trends of available experimental data.

References

Chun, C.-H.; Wuest, W. (1979): Experiments on the transition from steady to the oscillatory Marangoni convection of a floating zone under reduced gravity effect. Acta Astronautca, vol. 6, pp. 1073-1082.

Croll, A.; Kaiser, Th.; Schweizer, M.; Danilewski, A.N.; Lauer, S.; Tegetmeier, A.; Benz, K.W. (1998): Floating-zone and floating-solution-zone growth of GaSb under microgravity. J. Crystal Growth, vol. 191, pp. 365-376.

Eyer, A.; Leiste, H.; Nitsche, R. (1984): Crystal growth of silicon in Spacelab-1 – Experiment ES 321. Proc. 5th European Symposium on Material Sciences under Microgravity, ESA SP-222, pp. 173-182.

Gelfgat, A.Yu; Rubinov, A.; Bar-Joseph, P.Z.; Solan, A. (2005): On the three-dimensional instability of thermocapillary flow in arbitrarily heated floating zones in microgravity environment. *Fluid Dynamics & Materials Processing*, vol. 1, no. 1, pp. 21-32.

Han, J. H.; Sun, Z. W.; Dai, L. R.; Xie, J. C.; Hu, W. R. (1996): Experiment on the thermocapillary convection of a mercury liquid bridge in a floating half zone. J. Crystal Growth, vol. 169, pp. 129-135.

Hashimoto, T.; Ueno, I.; Kawamura, H.; Yoda, S. (2003): Numerical simulation of Marangoni convection in consideration of free surface displacement (part 6). In Technical Memorandum NASDA-TMR-0300004E, NASDA, pp. 67-98.

Hashimoto, T.; Kousaka, Y.; Ueno, I.; Kawamura, H.; Yoda, S. (2005): Numerical simulation of Marangoni convection in consideration of free surface displacement. In JAXA Research and Development Report JAXA-RR-04-027E, pp. 49-75.

Imaishi, N.; Yasuhiro, S.; Akiyama, Y.; Yoda, S. (2001): Numerical simulation of oscillatory Marangoni flow in half-zone liquid bridge of low Prandtl number. J. Crystal Growth, vol. 230, pp. 164-171.

Jurisch, M. (1990): Surface temperature oscillations of a floating zone resulting from oscillatory thermocapillary convection. Journal of Crystal Growth, vol. 102, pp. 223-232.

Kakimoto, K.; Eguchi, M.; Watanabe, H.; Hibiya, T. (1989): Natural and forced convection of molten silicon during Czochralski single crystal growth. J. Crystal Growth, vol. 94, pp. 412-420.

Kamotani, Y.; Ostrach, S.; Vargas, M. (1984): Oscillatory thermocapillary convection in a simulated floating zone configuration. Journal of Crystal Growth, vol. 66, no. 1, pp. 83-90.

Kamotani, Y.; Lee J. H.; Ostrach, S.; Pline, A. (1992): An experimental study of oscillatory thermocapillary convection in cylindrical containers. Physics of Fluids A, vol. 4, pp. 955-962.

Kamotani, Y.; Ostrach, S.; Masud, J. (1998a): Oscillatory thermocapillary flows in open cylindrical containers induced by CO_2 laser heating. International Journal of Heat and Mass Transfer, vol. 42, pp. 555-564.

Kamotani, Y.; Ostrach, S. (1998b): Theoretical analysis of thermocapillary flow in cylindrical columns of high Prandtl number fluids. J. Heat Transfer, vol. 120, pp. 758-764.

Kamotani, Y.; Ostrach, S.; Masud. J. (2000): Microgravity experiments and analysis of oscillatory thermocapillary flows in cylindrical containers. J. Fluid Mech., vol. 410, pp. 211-233.

Kamotani, Y.; Wang, L.; Hatta, S., Selver, R.; Yoda, S. (2001a): Effect of free surface heat transfer on onset of oscillatory thermocapillary flow of high Prandtl fluid. J. Japan Society of Microgravity Application, vol. 18, pp. 283-288.

Kamotani, Y.; Wang, L.; Hatta, S., Bhu-

nia, P.; Yoda, S. (2001b): Study of oscillatory thermocapillary flow of high Prandtl number fluid. In Technical Memorandum NASDA-TMR-010015E, NASDA, pp. 25-42.

Kamotani, Y.; Wang, L.; Hatta, S.; Wang, A.; Yoda, S. (2003): Free surface heat loss effect on oscillatory thermocapillary flow in liquid bridges of high Prandtl number fluids. International Journal of Heat and Mass Transfer, vol. 46, pp. 3211-3220.

Kawaji, M.; Otsubo, F.; Simic, S.; Yoda, S. (2000): Transition to oscillatory Marangoni convection in liquid bridges of intermediate Prandtl number. In Technical Memorandum NASDA-TMR-000006E, NASDA, pp. 75-114.

Kawaji, M.; Simic, S.; Psofogiannakis, G.; Yoda, S. (2001): Transition to oscillatory Marangoni convection in liquid bridges of intermediate Prandtl number. In Technical Memorandum NASDA-TMR-010015E, NASDA, pp. 107-132.

Kuhlmann, H. C.; Nienhuser, Ch. (2002): Dynamic free-surface deformations in thermocapillary liquid bridge. Fluid Dynamic Research, vol. 31, pp. 103-127.

Lan C. W., Yeh B.C., (2005), Effects of rotation on heat flow, segregation, and zone shape in a small-scale floating-zone silicon growth under axial and transversal magnetic fields, *FDMP: Fluid Dynamics & Materials Processing*, Vol. 1, No. 1, pp. 33-44.

Lappa, M. (2004): *Fluids, Materials & Microgravity : Numerical Techniques and Insigths into Physics*. Elsevier.

Lappa M., (2005), Review: Possible strategies for the control and stabilization of Marangoni flow in laterally heated floating zones, *FDMP: Fluid Dynamics & Materials Processing*, Vol. 1, No. 2, pp. 171-188.

Lappa, M.; Savino, R.; Monti, R. (2001): Three-dimensional numerical simulation of Marangoni instabilities in non-cylindrical liquid bridges in microgravity. International Journal of Heat and Mass Transfer, vol. 44, pp. 1983-2003.

Leypoldt, J.; Kuhlmann, H. C.; Rath, H. J. (2000): Three-dimensional numerical simulation of thermocapillary flows in cylindrical liquid bridges. J. Fluid Mech., vol. 414, pp. 285-314.

Masud, J.; Kamotani, Y.; Ostrach, S. (1997): Oscillatory thermocapillary flow in cylindrical columns of high Prandtl fluids. J. Thermophysics and Heat Transfer, vol. 11, no. 1, pp. 105-111.

Matsumoto, S.; Hayashida, H.; Komiya, A.; Natsui, H.; Yoda, S. (2005): Experimental study of thermocapillary flow in the half-zone liquid bridge of low Prandtl number fluid. In JAXA Research and Development Report JAXA-RR-04-027E, pp. 112-128.

Monti, R.; Albanese, C.; Cartenuto, L.; Castagnolo, D.; Evangelista, G. (1995): An investigation on the onset of oscillatory Marangoni flow. Adv. Space Res., vol.16, no. 7, pp. 87-94.

Nakamura, S; Hibiya, T.; Kakimotao, K.; Imaishi, N.; Nishizawa, S.; Hirata, A.; Mukai, K.; Yoda, S.; Morita, T. (1998): Temperature fluctuations of the Marangoni flow in a liquid bridge of molten silicon under microgravity on board the TR-IA-4 rocket. Journal of Crystal Growth, vol. 186, 85-94.

Nienhuser, Ch.; Kuhlmann, H. C.; Rath, H. J.; Yoda, S. (2000): Linear stability of the two-dimensional flow in half-zones: the influence of free surface heat transfer, aspect ratio, and volume of liquid. In Technical Memorandum NASDA-TMR-000006E, NASDA, pp. 215-246.

Nishino, K.; Yoda, S. (2000): The role of dynamic surface deformation in oscillatory Marangoni convection in liquid bridge of high Prandtl number fluid. In Technical Memorandum NASDA-TMR-000006E, NASDA, pp. 43-71.

Nishino, K.; Matida, E.; Yoda, S. (2001): Flow and heat transfer characteristics of thermocapillary convection of high Prandtl number fluid in cylindrical bridge. In Technical Memorandum NASDA-TMR-010015E, NASDA, pp. 45-72.

Nishino, K.; Li, X.; Kanashima, Y.; Yoda, S. (2005): Dynamic surface deformation of cylindrical bridge of high Prandtl number fluid in oscillatory thermocapillary convection- TSPI measurement and g-jitter effect. In JAXA Research and Development Report JAXA-RR-04-027E, pp. 25-48.

Ostrach, S. (1982): Low-gravity fluid flows. Ann. Rev. Fluid. Mech., vol. 14, pp. 313-345.

Preisser, F.; Schwabe, D.; Scharmann, A. (1983): Steady and oscillatory thermocapillary convection in liquid columns with free cylindrical surface. J. Fluid Mech., vol.126, pp. 545-567.

Schwabe, D.; Scharmann, A.; Preisser, F.; Oder, R. (1978): Experiments on surface tension driven flow in floating zone melting. Journal of Crystal Growth, vol. 43, pp. 305-312.

Sim, B.-C.; Zebib, A. (2004): Thermocapillary convection in cylindrical liquid bridges and annuli. Comptes Rendus Mechanique, vol. 332, pp. 474-486.

Simic, S.; Kawaji, M.; Yoda, S. (2006): Onset of oscillatory thermocapillary convection in acetone liquid bridges: the effect of evaporation. International Journal of Heat and Mass Transfer, vol. 49, pp. 3167-3179.

Takagi, K.; Otaka, M.; Natsui, H.; Arai, T.; Yoda, S.; Yuan, Z.; Mukai, K.; Yasuhiro, S.; Imaishi, N. (2001): Experimental study on transition to oscillatory thermocapillary flow in a low Prandtl number liquid bridge. J. of Crystal Growth, vol. 233, pp. 399-407.

Velten, R.; Schwabe, D.; Scharmann, A. (1989): The periodic instability of thermocapillary convection in cylindrical liquid bridges. Physics of Fluids A, vol. 3, pp. 729-736.

Wanschura, M.; Shevtsova, V. M.; Kuhlmann, H. C.; Rath, H. J. (1995): Convective instability mechanisms in thermocapillary liquid bridges. Physics of Fluids, vol. 7, pp. 912-925.

Yang, Y.; Kou, S. (2001): Temperature oscillation in a tin liquid bridge and critical Marangoni number dependency on Prandtl number. Journal of Crystal Growth, vol. 222, pp. 135-143.