Space experiment on the instability of Marangoni convection in large liquid bridge - MEIS-4: Effect of Prandtl number -

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Abstract. Microgravity experiments on the thermocapillary convection in liquid bridge, called Marangoni Experiment in Space (MEIS), are carried out in “KIBO” of ISS. Three series of experiments, MEIS-1, 2, and 4, have been conducted so far. This paper reports the results obtained from MEIS-4, in which 20cSt silicone oil (Pr=207) is used to generate large liquid bridges. They are suspended between coaxial disks that are 50mm in diameter, with their maximum length equal to 62.5mm. MEIS-4 aims at (1) determining the critical temperature difference for the onset of oscillatory flow; (2) realizing high Marangoni number conditions for high Pr fluid; (3) clarifying the effects of volume ratio, heating rate, hysteresis, and cooled disk temperature; and (4) observing whether the hydrothermal wave with azimuthal mode number m=0 appears or not. The main results are presented and compared with those obtained in MEIS-1 and 2, which utilized liquid bridges of 5cSt silicone oil (Pr=67).

1. Introduction
Thermocapillary convection is the flow that is driven by the surface tension-gradient along a liquid-gas interface. In general, the surface tension, σ, negatively depends on the temperature of the working fluid. Along the liquid-gas interface with a non-uniform temperature field, the surface tension difference drives the flow from the higher temperature region toward the lower temperature region. The resultant convection is called thermocapillary convection or Marangoni convection. Thermocapillary convection becomes important in liquid films, droplets, bubbles, and liquid bridges.

The liquid bridge (LB) is the geometry that is originally seen in the floating-zone method for crystal growth, as illustrated in Fig. 1a. This method is for manufacturing high-quality single crystals of high purity. The feed material is heated by a ring heater and the melt is suspended between the feed rod at one end and the already grown crystal at the other end. The temperature difference between the melting region and the solidified region is the driving force for the thermocapillary convection. This flow configuration is modeled to a LB suspended between two coaxial disks with different
temperatures, as illustrated in Fig. 1b. This simplified geometry is called a half-zone model. The temperature difference between the disks, $\Delta T$, drives a flow that is directed from the heated disk toward the cooled disk, along the surface, and returns inside the LB. This geometry is now recognized as one of the basic flow fields for the study of thermocapillary convection, along with its instability mechanisms.

It is known that the thermocapillary convection in a LB changes from a steady, axisymmetric state to an oscillatory, non-axisymmetric one, if $\Delta T$ exceeds a certain level. The appearance of such a flow causes the deterioration of the quality in the grown crystal. This instability is related to many factors, such as, the aspect ratio of the LB, the physical properties of the working fluid, the ambient gas condition, and so on. Therefore, a large number of studies have been conducted for the instability mechanisms in half-zone LBs.

A series of microgravity experiments, called Marangoni Experiment in Space (MEIS), have been conducted as the first science experiment on the Japanese Experimental Module “KIBO” of the International Space Station (ISS). Three series of experiments, MEIS-1, MEIS-2, and MEIS-4 were carried out in 2008, 2009, and 2010, respectively. The overview of MEIS-1 and 2 was reported by Kawamura et al. [1] in 2010. The present paper reports the main results of MEIS-4. This experiment was conducted in the period from October 26 to December 22, 2010. Its purpose is to collect data on the following phenomena, in large LBs (50mm in diameter and up to 62.5mm in length), which can form only in a long-period microgravity environment on ISS:

1. to determine the critical temperature difference for the onset of oscillatory flow,
2. to realize high Marangoni number conditions for high $Pr$ fluid,
3. to clarify the effects of volume ratio, heating rate, hysteresis, and cooled disk temperature, and
4. to observe if the hydrothermal wave with the azimuthal mode number $m=0$ appears.

Figure 1. Thermocapillary convection (a) in the floating-zone model and (b) in the half-zone model.

Figure 2. Schematic image of experimental apparatuses installed in the Fluid Physics Experiment Facility (FPEF).

2. Method

2.1. Fluid Physics Experiment Facility (FPEF)
All the experiments were performed in the Fluid Physics Experiment Facility (FPEF), which was especially designed for the present microgravity experiment. It was remote-controlled by the operation staffs and the science team members from Tsukuba Space Center (TKSC) of JAXA in Japan. Each experimental run started from 6:00 o’clock and ended at 15:00 o’clock in Japanese Standard Time (JST). This period is corresponding to 21:00 and 6:00 in Greenwich Mean Time (GMT), during which the astronauts are expected to sleep. Therefore, the g-jitters that might be caused by the astronaut activities can be minimized during experiment.

As shown in Fig. 2, several measurement apparatuses are installed in FPEF. They are (1) three top-view cameras for three-dimensional particle tracking velocimetry, (2) a side-view camera for
observing the shape of the LB and the overall flow pattern, (3) an IR camera for measuring surface temperature and observing hydrothermal waves, and (4) thermocouple sensors for measuring temperature distribution and detecting the onset of oscillation. Three finger-sized B/W CCD cameras are mounted near the heated disk. Their image is 768×494 pixel at a frame rate of 30fps. The heated disk is made of sapphire so that the top-view cameras can observe the flow in the LB. An IR camera having a wavelength sensitivity of 8-14\( \mu \)m is used to visualize surface temperature from the same viewing direction as the side-view camera. A traversable thermocouple sensor is adopted in MEIS-4. It is to detect the onset of oscillation and can be traversed in axial and radial directions.

2.2. Experimental conditions

A LB of silicone oil is formed between two coaxial disks heated differently. The chamber is filled with argon gas at 94kPa and about 21\(^{\circ}\)C. The geometry of the LB is illustrated in Fig. 1b. The cooled disk is made of aluminum. The disk diameter is 50mm in MEIS-4 and 30mm in MEIS-1 and 2. Each disk has 45-degree edge to prevent liquid leakage. The cooled disk is traversed in the axial direction so that a wide range of aspect ratio, \( Ar = H/D \), where \( H \) and \( D \) are the LB length and the disk diameter, respectively, can be realized. The values of the maximum LB length, with the 50mm and 30mm disks, are respectively 62.5mm and 60mm, for which \( Ar=1.25 \) and 2.0, respectively. Note that the maximum length of the LB in the 1g environment is about 3mm, and the longer LB can be generated only in a microgravity environment.

Figure 3 shows one such LB with \( Ar=1.25 \); it is generated in MEIS-4. The volume ratio, \( Vr = V/V_0 \), defines the ratio of the LB volume, \( V \), to the gap volume between the disks, \( V_0 = D^2H/4 \). The \( Vr=1.0 \) condition gives a straight cylindrical shape. The volume ratio is basically fixed at 0.95 to stabilize the LB; and, thus, to prevent fluid leakage from the edge of the disks. In some experiments, the volume ratio was changed in the range from 0.91 to 1.05 to study the effect of \( Vr \) on the onset of the oscillation.

Silicone oil with the kinematic viscosity of 20cSt is used as the working fluid in MEIS-4, while silicone oil with the kinematic viscosity of 5cSt is used in MEIS-1 and 2. The Prandtl number, \( Pr \), of the former liquid is 207, while that of the latter one is 67, at 25\(^{\circ}\)C (Shin-Etsu Chemical Co., LTD, KF-96L). The effect of \( Pr \) is, thus, examined. One of the important purposes in MEIS-4 is to observe whether hydrothermal waves with \( m=0 \) appear or not. This is motivated by the linear stability analysis of Xu and Davis [2], who analyzed thermocapillary instabilities in an infinitely long LB. They reported that the azimuthal mode number of the hydrothermal waves depends on \( Pr \) of the working fluid and that \( m=0 \) appears when \( Pr \) exceeds the value of 62.2. More recently, however, Ryzhkov [3] carried out a linear stability analysis which is similar to that reported by Xu and Davis [2] but he reported that the critical mode with \( m=0 \) for \( Pr \) higher than 62.2 is not confirmed in his analysis. The \( Pr \) in MEIS-4 is 207, which is high enough to check if the critical mode with \( m=0 \) appears for long liquid bridges.

![Figure 3. A liquid bridge for \( Ar=1.25 \) in MEIS-4.](image)

| Table 1. Basic physical properties of working fluid. |
|---------|-----------------|-----------------|
| Working fluid | 20cSt silicon oil at 25\(^{\circ}\)C | 5cSt silicon oil at 25\(^{\circ}\)C |
| Prandtl number, \( Pr \) | 207 | 67 |
| density, \( \rho \) (kg m\(^{-3}\)) | 950 | 915 |
| kinematic viscosity, \( \nu \) (m\(^{2}\) s\(^{-1}\)) | 2.0×10\(^{-6}\) | 5.0×10\(^{-6}\) |
| thermal diffusivity, \( \alpha \) (m\(^{2}\) s\(^{-1}\)) | 9.67×10\(^{-8}\) | 7.46×10\(^{-8}\) |
| surface tension, \( \sigma \) (N m\(^{-1}\)) | 20.6×10\(^{-3}\) | 19.7×10\(^{-3}\) |
| temp. conf. of \( \sigma \), \( \sigma_f \) (N m\(^{-1}\) K\(^{-1}\)) | -6.24×10\(^{-5}\) | -6.58×10\(^{-5}\) |

| \(^{a}\) MEIS-1, \(^{b}\) MEIS-2 | | |
The physical properties of each of the silicone oil are given in Table 1. The working fluid is seeded with the tracer particles for the flow visualization. These particles are gold-nickel-coated polymer particles, and their average diameter and density are 180 μm and 1300 kg/m³, respectively. The density ratio between the tracer particle and the silicone oil is close to unity, and the fidelity of the particle to the fluid motion is checked numerically.

The critical temperature difference, \( \Delta T_c \), required for the onset of the oscillation of the flow and temperature fields, is determined by a stepwise adjustment of \( \Delta T (= T_h - T_c) \). The cooled disk temperature, \( T_c \), is kept constant at 20°C, except for a few cases where \( T_c \) is varied to especially examine its effect. On the other hand, the \( T_h \) is always changed in a stepwise manner. The lower and higher temperature limits of the cooled and the heated disks, respectively, are 10°C and 85°C. The maximum temperature difference is about 75°C. A sufficiently long waiting time (more than 60 min) is given after each of the stepwise temperature changes, \( \Delta T \). The onset of the oscillation is detected by the traversable thermocouple sensor and the motion of tracer particles in the LB.

3. Result and discussion

3.1. Instability of thermocapillary convection

In MEIS-4, \( \Delta T_c \) was measured for \( Ar=0.225 \sim 1.25 \). Figure 4 shows the measured \( \Delta T_c \) plot as a function of \( Ar \). From here, it is evident that the \( \Delta T_c \) decreases with \( Ar \), showing local peaks at \( Ar=0.87 \). The results of MEIS-1 and 2 are in good agreement with each other, while the results of MEIS-4 show appreciably larger values. These results are perhaps caused by the differences in \( Pr \) and in the size of the LB (30 mm vs. 50 mm in diameter).

For better understanding, the \( \Delta T_c \) is non-dimensionalized as the critical Marangoni number, \( Ma_c \), as follows:

\[
Ma = \frac{\sigma_T \Delta TH}{\rho \nabla \alpha}
\]  

(1)

where \( \rho \), \( \nu \), \( \alpha \), and \( \sigma_T \) are the density, kinematic viscosity, thermal diffusivity and temperature coefficient of surface tension, respectively. The values of \( \rho \), \( \alpha \), and \( \sigma_T \) for the 20 cSt and 5 cSt silicone oils are given in Table 1 where \( \nabla \) is the mean of \( \nu(T_h) \) and \( \nu(T_c) \), and dependent on the disk temperature. The \( Ma \) represents the strength of thermocapillary convection, by which the onset of oscillatory flow can be specified. In microgravity, other dimensionless parameters (such as, Grashof number and Bond number), which could have potentially affected the onset of the oscillation, become zero. This is advantageous for the study of the instability mechanisms of thermocapillary convection.

The obtained \( Ma \) is plotted as a function of \( Ar \), in Fig. 5. The results from MEIS-1 and 2 can be seen to be in good agreement with each other. Moreover, the results from MEIS-2 and 4 show a local peak at \( Ar=0.87 \).

Figure 6 shows the oscillation frequency, \( f \), plotted as a function of \( Ar \). It shows that \( f \) decreases with \( Ar \), for MEIS-1, 2, and 4. An interesting behavior observed in MEIS-4 is the presence of the fundamental (or 1st) frequency and its 2nd harmonics, for \( Ar>0.63 \). This behavior is observed at a temperature difference that is only slightly higher than \( \Delta T_c \). Note the stepwise increase of \( \Delta T \) in the determination of \( \Delta T_c \). Interestingly, the 2nd harmonics disappears when the \( \Delta T \) is increased further slightly. The appearance of the 2nd harmonics at nearly \( \Delta T_c \) was not observed in MEIS-1 or 2. In Fig. 7, the dimensionless oscillation frequency, \( \tilde{f} \), is plotted as a function of \( Ar \). This non-dimensionalization was proposed by Preisser et al. [4], as follows:

\[
\tilde{f} = \frac{H^2}{\alpha \sqrt{Ma}} f
\]  

(2)
It is seen that the dimensionless fundamental frequency from MEIS-4 is in agreement with the dimensionless oscillation frequencies from MEIS-1 and 2, for \( Ar = 0.67 - 1.25 \). This may indicate that the non-dimensionalization of Eq. (2) is appropriate for the LBs of different, high \( Pr \) fluids. The data from MEIS-2 exhibit a jump of \( \tilde{f} \) at \( Ar = 1.25 - 1.50 \), suggesting that the oscillation mode changes from azimuthal mode into axial mode (Schwabe [5], Yano et al. [6]). This behaviour is not confirmed in MEIS-4 due to the \( Ar \leq 1.25 \) condition.

![Figure 4. Plot of \( \Delta T_c \) as function of \( Ar \).](image)

![Figure 5. Plot of \( Ma_c \) as function of \( Ar \).](image)

![Figure 6. Plot of \( f \) as function of \( Ar \).](image)

![Figure 7. Plot of \( \tilde{f} \) as function of \( Ar \).](image)

### Table 2. Critical values and oscillation modes obtained from MEIS-4.

| \( Ar \) (-) | \( Vr \) (-) | \( T_h \) (°C) | \( T_c \) (°C) | \( Ma_c \) (-) | \( \tilde{f} \) 1st/2nd (Hz) | Oscillation mode | \( m \) (-) |
|---|---|---|---|---|---|---|---|
| 0.225 | 0.95 | 76.9 | 20.0 | 2.96 \times 10^4 | 0.44/- | Rotating | 3 |
| 0.25 | 0.95 | 72.6 | 20.0 | 2.88 \times 10^4 | 0.48/- | - | - |
| 0.30 | 0.95 | 61.3 | 20.0 | 2.57 \times 10^4 | 0.49/- | Standing | 2 |
| 0.35 | 0.95 | 54.0 | 20.0 | 2.24 \times 10^4 | 0.54/- | Standing | 2 |
| 0.40 | 0.95 | 48.7 | 20.0 | 2.22 \times 10^4 | 0.60/- | Standing | 2 |
| 0.45 | 0.95 | 42.4 | 20.0 | 1.84 \times 10^4 | 0.54/- | Rotating | 2 |
| 0.50 | 0.95 | 36.7 | 20.0 | 1.48 \times 10^4 | 0.57/- | Rotating | 1 |
| 0.63 | 0.95 | 32.8 | 20.0 | 1.36 \times 10^4 | 0.71/1.42 | Standing | 1 |
| 0.75 | 0.95 | 33.6 | 20.0 | 1.90 \times 10^4 | 1.03/1.87 | Standing | 1 |
| 0.81 | 0.95 | 35.3 | 20.0 | 2.46 \times 10^4 | 1.04/2.09 | Standing | 1 |
| 0.87 | 0.98 | 40.6 | 20.0 | 3.22 \times 10^4 | 1.32/2.65 | Standing | 1 |
| 0.93 | 0.98 | 35.8 | 20.0 | 2.60 \times 10^4 | 1.90/- | - | - |
| 1.00 | 0.99 | 31.9 | 20.0 | 2.04 \times 10^4 | 1.76/3.52 | Standing | 1 |
| 1.12 | 0.99 | 28.9 | 20.0 | 1.66 \times 10^4 | 1.98/3.95 | - | - |
| 1.25 | 0.99 | 29.9 | 20.0 | 2.07 \times 10^4 | 2.21/4.42 | Standing | 1 |
The modes of oscillation observed in MEIS-4 are given in Table 2, together with the value of $f$ and $T$ \( (=f^{-1}) \). The azimuthal mode, $m$, was determined from the motion of the tracer particles, and the oscillation with $m=0$ was not observed in the present experiment. This observation is in agreement with the result reported by Ryzhkov [3] who found a new branch of neutral mode with $m=1$ for high $Pr$ fluids (say $Pr\geq 19.95$) for thermally insulated boundary conditions. Although not shown in this paper, the IR images acquired for $Ar=1.00$ and 1.25 indicate that the hydrothermal waves propagate along the LB surface in the direction from the heated disk towards the cooled disk (cf. Yano et al. [6]). This propagation direction is in opposite to that reported by Ryzhkov. The reason for this discrepancy is not clear yet and, as conjectured by Ryzhkov, the direct comparison between experiment for LB with finite length and linear stability analysis for LB with infinite length may not be possible. From the data obtained in MEIS-1 and 2, Yano et al. [6] pointed out several fundamental differences in the characteristics of oscillatory flow and temperature fields between an infinitely long LB (considered by Xu and Davis [2]) and the finitely long LBs that are experimented in MEIS. This issue will be studied in MEIS-3, which is to be conducted soon by using a 20cSt silicone oil LB with $Ar=2.0$.

3.2. High Marangoni number conditions

Several experiments for high $Ma$ conditions were also carried out in MEIS-4, by exploiting the hardware capability, which allowed us to increase $T_h$ up to as high as 85°C. As a result, $\Delta T=64.8^\circ$C and 61.9°C was achieved for $Ar=1.0$ and 1.25, respectively. These conditions correspond to $Ma/Ma_c=7.1$ and 8.3, respectively. Note that such high $Ma$ conditions, relative to $Ma_c$, for high $Pr$ fluids, are extremely difficult in the ground experiments where $\Delta T_c$ becomes high for short liquid bridges. Figure 8 presents the IR images of the surface temperature fluctuations at $Ma/Ma_c=2.1$, 5.2, and 8.3, for $Ar=1.25$. The mean brightness levels are subtracted to enhance the brightness levels corresponding to the surface temperature fluctuations. In these figures, higher and lower temperature regions appeared as the white and black regions, respectively. It can be observed that the lower temperature regions propagate along the axial direction, from the heated disk towards the cooled disk. Yano et al. [6] reported that such a propagation of the temperature field is associated with the propagation of the velocity field; and, therefore, this propagation is a hydrothermal wave. Initially the oscillation mode of the hydrothermal wave is a standing wave (see Fig. 8a), which latter changes into a rotating wave (see Fig. 8b). At $Ma/Ma_c=8.3$, in Fig. 8c, the propagation starts to exhibit chaotic features, consisting of fine and fluctuating structures in the surface temperature distributions.

![Image of IR camera visuals with temperatures labeled: (a) $Ma/Ma_c=2.1$, (b) $Ma/Ma_c=5.2$, (c) $Ma/Ma_c=8.3$]

**Figure 8.** Surface temperature fluctuation visualized by the IR camera.

4. Conclusions

This paper reports the basic characteristics of the instability of thermocapillary convection observed in large liquid bridges that are formed in long microgravity environment on the ISS. The working fluid is 20cSt silicone oil, whose Prandtl number ($Pr=207$) is nearly three times higher than that of the 5cSt silicone oil ($Pr=67$), which was used in our previous space experiments. The effect of the Prandtl number is taken into account in the comparison of these space experiments.
The critical temperature difference, $\Delta T_c$, and the resultant $Ma_c$, which is required for the onset of oscillatory flow, are determined in a wide range of $Ar$ ($Ar=0.225\sim1.25$). As expected, the measured $\Delta T_c$ for the $Pr=207$ is substantially higher than that for the $Pr=67$, for all the $Ar$ cases. However, the plots of $Ma_c$ obtained for these Prandtl numbers, agree well with each other, with both showing a local $Ma_c$ peak at $Ar=0.87$. The oscillation frequency is also determined for the same $Ar$ range. Non-dimensional oscillation frequencies for $Pr=207$ and 67 agree well with each other, but those for $Pr=207$ reveal the presence of a fundamental (or 1st) frequency and its 2nd harmonics for $Ar>0.67$.

The mode of oscillation for each $Ar$ is obtained. The hydrothermal wave with $m=0$ is not observed in the present experiment. This may indicate that the linear stability analysis, assuming an infinitely long liquid bridge, does not provide direct comparison with the experiments, whose liquid bridges have inevitably a finite length. The IR camera reveals that the surface temperature fluctuations propagate in the axial direction, from the heated disk towards the cooled disk. The oscillation mode of hydrothermal wave for, $Ar=1.25$, is standing wave, while it changes into a rotating wave, or chaotic state, with increasing $Ma$.

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