

## **OSCILLATORY THERMOCAPILLARY FLOWS IN SIMULATED FLOATING-ZONES WITH TIME-DEPENDENT TEMPERATURE BOUNDARY CONDITIONS**

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### Abstract

This study deals with numerical simulations of the Maxus sounding rocket experiment on oscillatory Marangoni convection in liquid bridges. The problem is investigated through direct numerical solution of the non-linear, time-dependent, three-dimensional Navier-Stokes equations. In particular a liquid bridge of silicon oil 2[cs] with a length  $L = 20$  [mm] and a diameter  $D = 20$  [mm] is considered. A temperature difference  $\Delta T = 30$  [K] is imposed between the supporting disks, by heating the top disk and cooling the bottom one with different rates of ramping.

The results show that the oscillatory flow starts as an "axially running wave" but after a transient time the instability is described by the dynamic model of a "standing wave", with an azimuthal spatial distribution corresponding to  $m=1$  (where  $m$  is the critical wave number).

After the transition, the disturbances become larger and the azimuthal velocity plays a more important role and the oscillatory field is characterized by a travelling wave.

The characteristic times for the onset of the different flow regimes are computed for different rates of ramping.

### 1. Introduction

Oscillatory thermocapillary flows in liquid bridges have been investigated, experimentally, numerically and by the hydrodynamic stability theory, as model problem to study surface tension-driven effects in the containerless method of float-zone crystal growth. The comparisons of results obtained in microgravity experiments with those obtained by linear stability analyses, based on idealized mathematical models are complicated, among other reasons, by the fact that the experiments typically involve time-dependent boundary conditions, while the usual stability analysis assumes steady basic states. To shed some light on this issue a numerical code has been developed to give a direct three-dimensional and time-dependent simulation of the standard (zero surface-deformation) half-zone model with time-dependent end-wall temperatures. The field equations have been solved using finite-differences methods explicit in time and a staggered spatial mesh in cylindrical co-ordinates. The numerical model has been applied to the study of high Prandtl number liquids<sup>1</sup> and semiconductor melts<sup>2</sup>, respectively.

The computations have confirmed some previous theoretical and numerical results obtained by linear stability studies, e.g. that for low Prandtl numbers the instability breaks the spatial axisymmetry prior to the onset of time dependence (stationary bifurcation), whereas for high Prandtl numbers the first bifurcation is oscillatory (Hopf bifurcation).

In particular for high Prandtl numbers the stability theory predicts that the three dimensional supercritical state after the Hopf bifurcation point can be interpreted as a superposition of two counter-propagating waves, characterized by an axial and an azimuthal component, so that the resulting disturbance exhibits an azimuthal component running under a certain angle against the axis of the bridge<sup>3-4</sup>.

The superposition of two waves with equal amplitude results in a "standing wave", with the minimum and maximum disturbances at fixed azimuthal positions; while the superposition of waves with different amplitude gives rise to a "travelling wave", with the minimum and maximum disturbances travelling in azimuthal direction. Unfortunately, nothing can be predicted by the linear stability analyses about the amplitude of these disturbances.

The standing wave or the travelling wave modes were observed by different investigators<sup>5,6</sup> during experimental ground-based or microgravity research. However, the relationship between the two flow instabilities mechanisms is unknown. In recent on ground experiments Velten, Schwabe and Scharmann<sup>7</sup> observed an oscillatory instability with zero azimuthal phase shift of the temperatures measured by thermocouples located in different azimuthal positions, identifying a third spatio-temporal model of oscillatory

convection. This model, characterized by certain waves running with zero azimuthal component, is referred to as "axially running wave" (in this case the azimuthal wave number is  $m = 0$ ).

This experimental result seems to be in disagreement with the linear stability analysis performed by Kuhlmann and Rath<sup>3</sup> and Shevtsova et al.<sup>4</sup> who predict always critical wave numbers  $m \geq 1$ , i.e. instability waves with non-zero azimuthal component.

To investigate these questions an experiment has been proposed by the authors and selected by the European Space Agency for a Maxus sounding rocket mission. Aim of the experiment is the study of the different flow and temperature distributions of the oscillatory Marangoni convection in the presence of time-dependent boundary conditions on the supporting disks.

In this paper we report the results of numerical simulations for the definition of the experiment. In particular, a silicon oil floating-zone, suspended between cylindrical disks, is investigated when a time-dependent difference across the liquid bridge is established with symmetrical temperature ramps at the top and bottom supports.

## 2. Mathematical model

The problem under investigation consists of a standard half-zone model with rigid surface and time-dependent temperature profiles on the disks ( $T = T_0 \pm \mathfrak{R} t$ ). The final temperature difference at the end of the ramp will be denoted by  $\Delta T = T_{\text{hot}} - T_{\text{cold}} = 2 \mathfrak{R} t_{\text{ramp}}$ , where  $\mathfrak{R}$  is the rate of the ramping and  $t_{\text{ramp}}$  is the duration of the ramp. The bridge is characterized by length  $L$ , diameter  $D$  and aspect ratio  $A = L/D$  (Fig 1).

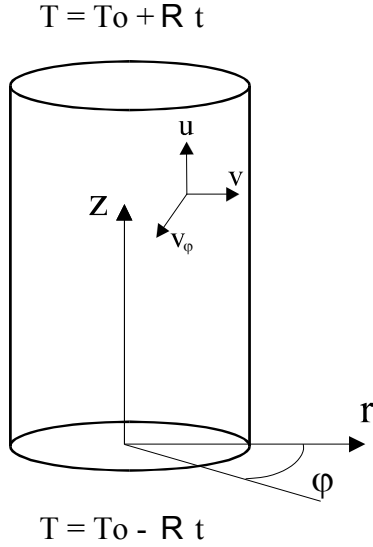


Fig 1: Scheme of the liquid bridge

The flow in the volume phase is governed by the continuity, Navier-Stokes and energy equations :

$$\nabla \cdot \mathbf{V} = 0 \quad (1a)$$

$$\frac{\partial \mathbf{V}}{\partial t} = -\frac{1}{\rho} \nabla p - \nabla \cdot [\mathbf{V}\mathbf{V}] + \nu \nabla^2 \mathbf{V} \quad (1b)$$

$$\frac{\partial T}{\partial t} = -\nabla \cdot [\mathbf{V}T] + \alpha \nabla^2 T \quad (1c)$$

where  $\nu$  and  $\alpha$  are the kinematic viscosity and the thermal diffusivity. The initial and boundary conditions read:

$$t=0: \mathbf{V}(z, r, \varphi) = T(z, r, \varphi) = 0 \quad (2)$$

$$\text{cold disk} \quad 0 \leq r \leq R ; 0 \leq \varphi \leq 2\pi$$

$$\mathbf{V}(z=0, r, \varphi, t) = 0; T(z=0, r, \varphi, t) = -\mathfrak{R} t \quad (3a, b)$$

$$\text{hot disk} \quad 0 \leq r \leq R ; 0 \leq \varphi \leq 2\pi$$

$$\mathbf{V}(z=L, r, \varphi, t) = 0; T(z=L, r, \varphi, t) = \mathfrak{R} t \quad (4a, b)$$

cylindrical free surface:

$$0 \leq z \leq L; 0 \leq \varphi \leq 2\pi$$

$$v(z, r=R, \varphi, t) = 0 \quad (5)$$

$$\mu \frac{\partial u}{\partial r}(z, r=R, \varphi, t) = -\sigma_T \frac{\partial T}{\partial z}(z, r=R, \varphi, t) \quad (6)$$

$$r \frac{\partial V_\varphi}{\partial r}(z, r=R, \varphi, t) - V_\varphi(z, r=R, \varphi, t) =$$

$$-\frac{1}{\mu} \sigma_T \frac{\partial T}{\partial \varphi}(z, r=R, \varphi, t) \quad (7)$$

$$\frac{\partial T}{\partial r}(z, r=R, \varphi, t) = 0 \quad (8)$$

The solution method has been described in Ref<sup>1</sup> where an independent validation of the computational model has been carried out comparing the numerical results with other three-dimensional numerical solutions available in literature and with experimental data obtained in microgravity experiments.

### 3. Results and discussion

#### 3.1 Onset of instability and influence of time-dependent boundary conditions

For the numerical computations we consider a liquid bridge of silicon oil 2 [cs], with a length  $L = 20$  [mm] and a diameter  $D = 20$  [mm]. The temperature difference between the supporting disks was increased from zero to  $\Delta T = 30$  [K]. When the critical temperature difference between the supporting disks is exceeded, a transient oscillatory phase develops until a stable supercritical oscillatory three-dimensional flow is reached.

All the points of the liquid bridge show an oscillatory behaviour of the temperature and velocity similar to that one reported in Fig. 2. and Fig. 3.

For the liquid bridge under study, the thermocapillary convection becomes oscillatory for  $\Delta T > 7$  K. When the imposed temperature difference is further increased one observes periodic oscillations of the temperature in the liquid. The linear stability theory cannot predict the amplitude of the disturbances after the onset of the instability conditions.

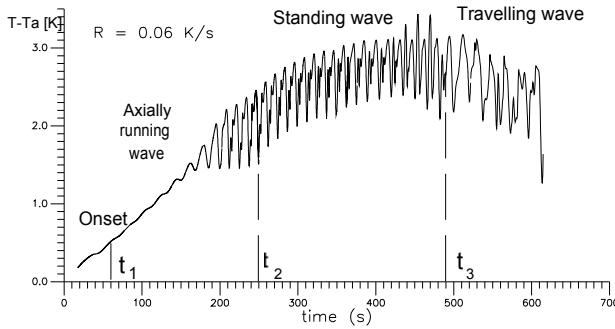


Fig 2: Variation in time of the temperature at  $z = 0.75 L$ ,  $r = 0.9R$ ,  $\phi = \pi$

The numerical results show that for  $\Delta T > \Delta T_c$  the oscillations amplitude is an increasing function of  $\Delta T$ . This result agrees with previous experimental analysis. Preisser, Schwabe and Scharmann<sup>5</sup> found that the amplitude  $B$  of the oscillations increases as a power function of  $\Delta T$  (or of the instantaneous Marangoni number):

$$B \cong \Delta T (Ma - Ma_c)^{1/2} \quad (9)$$

According to Velten, Schabe and Scharmann<sup>7</sup> the functional dependence for  $B$  can be generalized as

$$B \cong \Delta T \left[ \frac{\Delta T - \Delta T_c}{\Delta T_c} \right]^y \quad (10)$$

where  $y$  is an exponent that depends on the geometry of the bridge and on the properties of the fluid.

The present results agree well with the equation (10) assuming for the exponent the value  $y = 0.6$ . The experimental results<sup>5-7</sup> show that also the frequency of the oscillations increases with  $\Delta T$ . The numerical computation show that, for  $\Delta T = 30$  [K] the frequency is twice the frequency computed for  $\Delta T = 10$  [K].

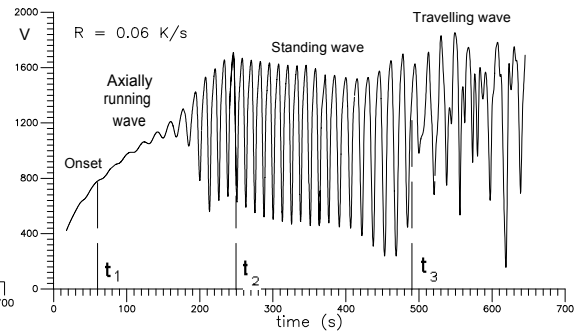


Fig 3: Variation in time of the nondimensional velocity at  $z = 0.75 L$ ,  $r = 0.9R$ ,  $\phi = \pi$

In Ref<sup>8</sup> a characteristic frequency has been identified that well correlates the available experimental data obtained on ground and in microgravity, for  $8 \leq Pr \leq 74$  and aspect ratio  $0.1 \leq A \leq 1.6$ . If  $Ma_c$  denotes the critical Marangoni number at the onset of oscillatory instability, the characteristic frequency can be expressed as:

$$f = \frac{1}{2\pi} \alpha Ma_c^{2/3} L^{-3/2} D^{-1/2} \quad (11)$$

The results obtained with the present method show that the equation (11) can be extended also to the supercritical conditions corresponding to  $\Delta T > \Delta T_c$  considering, instead of  $Ma_c$ , the instantaneous Marangoni number  $Ma$  during the temperature ramp.

Similarly to the experimental finding by Chun and West<sup>6</sup> and Velten, Schwabe and Scharmann<sup>7</sup>, the computations show an interesting dynamic

development of the frequency spectra. For large enough  $\Delta T$  ( $\Delta T = 25$  [K] in the case of Figs. 2-3) the structure of the oscillations becomes more complicated and the oscillatory functions result from the superposition of a second order harmonic function (with frequency  $f_1$ ) to the basic fundamental frequency ( $f_0$ ). This behaviour can be interpreted on the basis of the Hopf bifurcations theory that predicts, for a non-linear system, an oscillatory state characterized by a single fundamental frequency, in the neighbourhood of the onset of instability, and more "turbulent" Fourier spectra, due to the presence of other harmonics, when a supercritical state is reached.

$\Delta T$	$f_0$ (mhz)
10 K	37
15 K	50
20 K	59
25 K	77
30 K	83

Table 1 - Fundamental frequency  $f_0$  versus  $\Delta T$

In Table 1 the fundamental frequency  $f_0$  has been reported for different values of  $\Delta T$ . The second harmonics has a characteristic frequency  $f_1 = 2f_0$ . Figs. 2-3 show that the amplitude of the secondary harmonics is less than the fundamental one. When  $\Delta T$  increases, the amplitude of the fundamental harmonics increases, as discussed above, together with the amplitude of the additional harmonics. For  $t > t_{\text{ramp}}$  ( $t_{\text{ramp}}=240$  sec), i.e. when the temperature difference is held constant, very regular oscillations are observed with amplitude and frequency constant with time.

### 3.2 Different instabilities and oscillatory flow regimes

The oscillatory behaviour of the temperature and velocity is only one aspect of the oscillatory regime in the liquid bridge. The onset of time-dependence is characterized by relevant changes of the spatial velocity and temperature distributions within the cylinder.

The numerical results show that the flow becomes oscillatory for  $\Delta T = 7$  K.

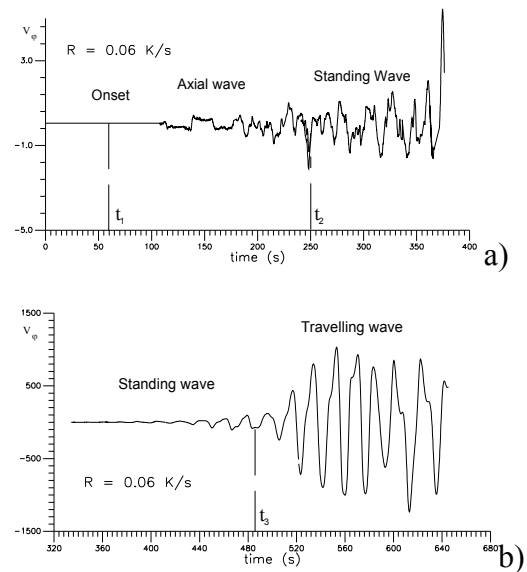


Fig 4: Nondimensional azimuthal velocity at  $z = 0.75 L$ ,  $r = 0.9R$ ,  $\varphi = \pi$

It is very interesting to observe, however, that when the oscillations of the temperature begin, the azimuthal component is still very small, so that the flow field is still axisymmetric. In a certain neighborhood of the onset there is in fact no phase shift of the temperature in the azimuthal direction, and the azimuthal velocity is negligible (Fig 4a).

We found, on the contrary, a large phase shift of the temperature oscillations in the axial direction (Fig 5), in agreement with the model of the running wave with zero azimuthal component, observed by Velten, Schwabe and Scharmann<sup>7</sup>.

On the surface of the bridge an axisymmetrical temperature spot appears near the hot disk and propagates towards the cold one (Fig. 6).

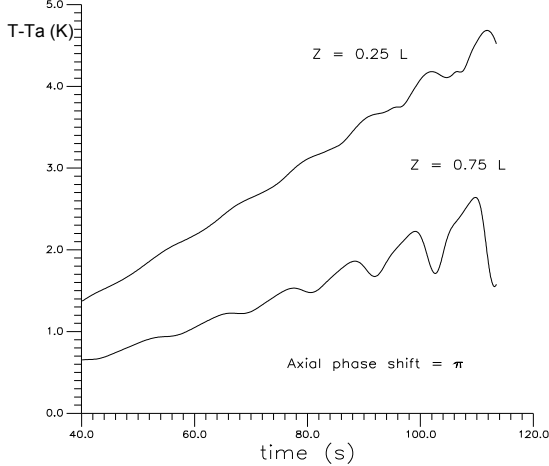


Fig 5a: Variation in time of the temperature computed by "numerical" thermocouples located at  $r = 0.9 R$ ,  $\varphi = \pi$ , at different axial coordinates

This behaviour can be interpreted as the effect of a disturbance travelling in the axial direction only.

The instability starts as an "axially running wave", i.e. following the onset the disturbances travel mainly in the axial direction, in the form of a wave characterized by an axisymmetric front and hence characterized by an azimuthal wave number  $m = 0$ .

The amplitude of the oscillations is larger inside the liquid bridge (Fig 5 b). This result gives an important indication for the experimental procedures, aimed at determining the oscillations of the temperature using thermocouples.

When the time increases, the azimuthal disturbances become larger and the azimuthal velocity plays a more important role.

To study the three-dimensional disturbance in the neighborhood of the bifurcation point, the axisymmetric basic flow has been subtracted to the numerically computed three-dimensional time-dependent solution.

Fig 7 shows that the flow, after a relatively short transient time, may be properly described by the dynamic model of a "standing wave".

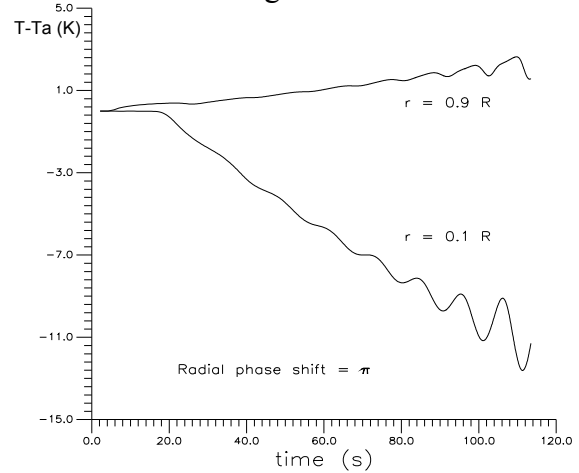


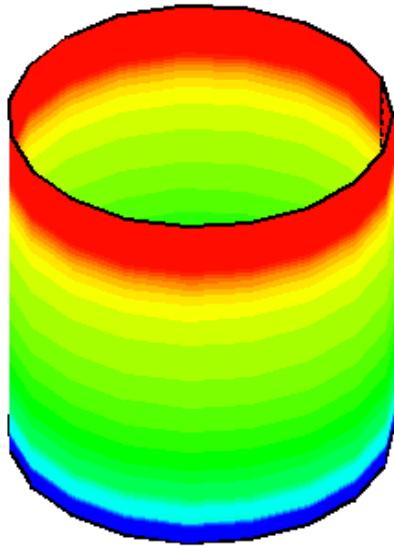
Fig 5b: Variation in time of the temperature computed by "numerical" thermocouples located at  $z = 0.5 L$ ,  $\varphi = \pi$ , at different radial coordinates

The three-dimensional temperature disturbance consists of a pair of spots (hot and cold) pulsating at the same azimuthal positions along the interface. As discussed in Ref<sup>1</sup> these spots are responsible for thermocapillary effects in the azimuthal direction, causing a pair of counter-rotating vortex cells in the transversal section.

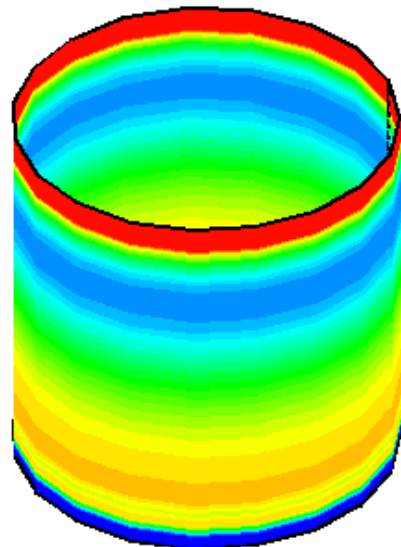
At the same time strong interior temperature extrema appear, conductively heating or cooling them.

As originally proposed by Smith and Davis<sup>9</sup> thermocapillary forces support this mechanism, so that the spots become pulsating (the hot spot replaces the cold one and viceversa).

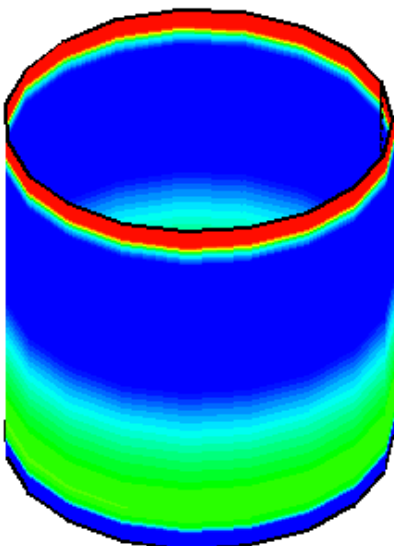
The presence of an axially running wave ( $m=0$ ) together with a standing wave ( $m=1$ ) is in contrast with the results of the linear stability analysis obtained by Kuhlmann and Rath<sup>3</sup> and Shevtsova et al.<sup>4</sup>, who predicted a critical wave number  $m=1$ , but agrees with those obtained by Xu and Davis<sup>10</sup>, who predicted the possible coexistence of modes  $m = 1$  and  $m = 0$  at high Prandtl numbers.



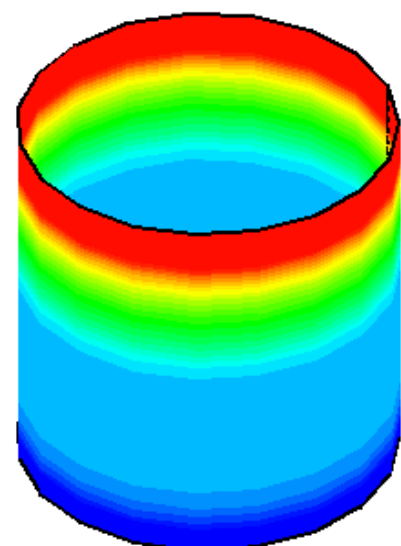
6a)



6b)

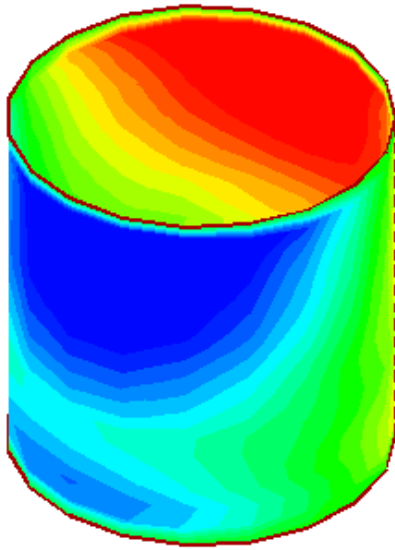


6c)

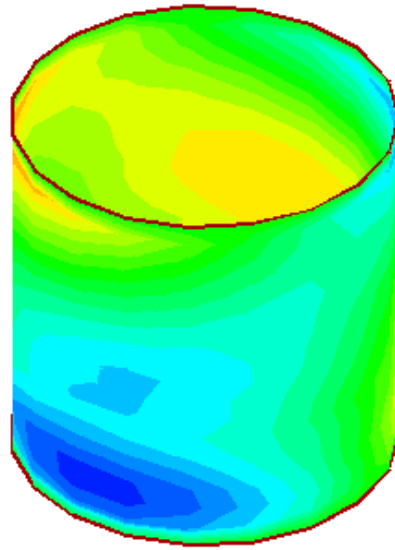


6d)

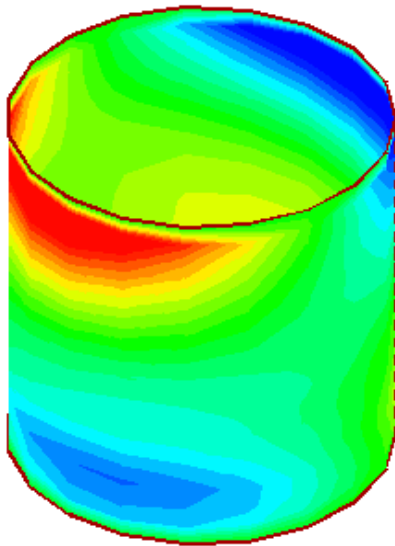
Fig 6: Temperature distribution at different times during the oscillation period, for the axially running wave , at  $t = 60$  (s) in the case  $R = 0.06$  [K/s].



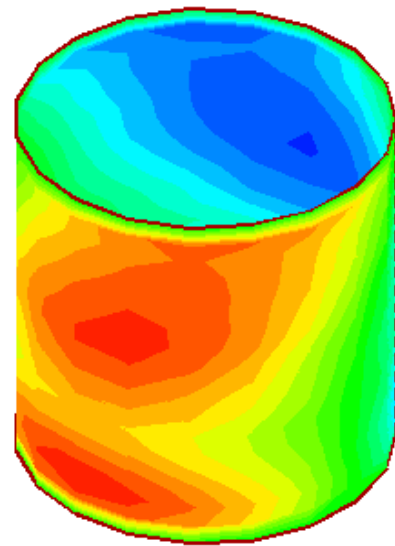
7a)



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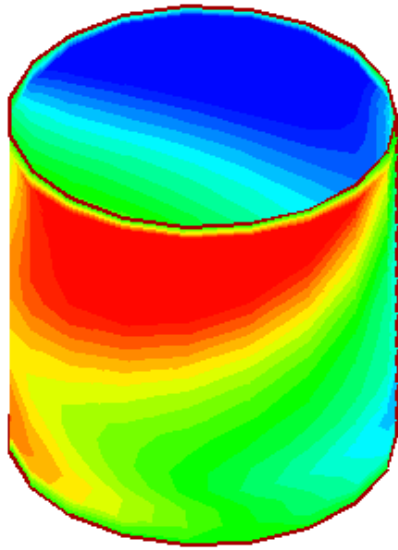
7c)



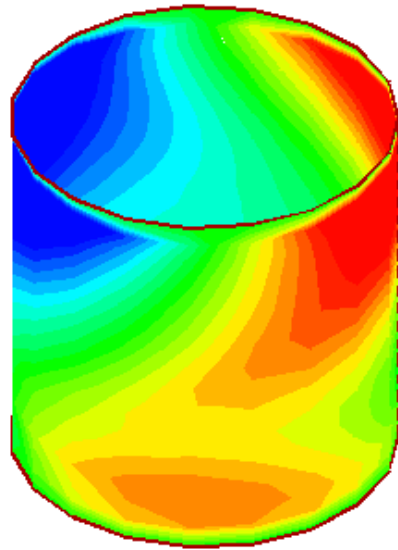
7d)

Fig 7: Temperature distribution at different times during the oscillation period, for the azimuthal standing wave, at  $t=250$  (s) in the case  $R = 0.06$  [K/s]

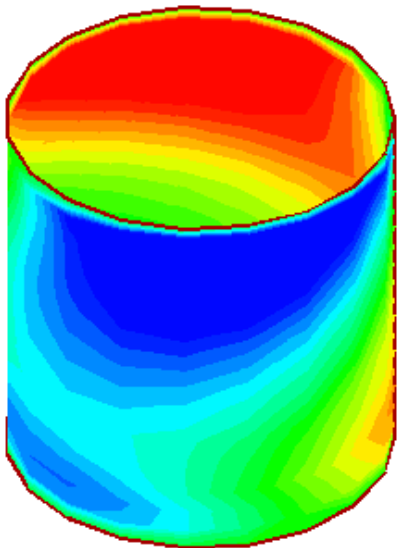




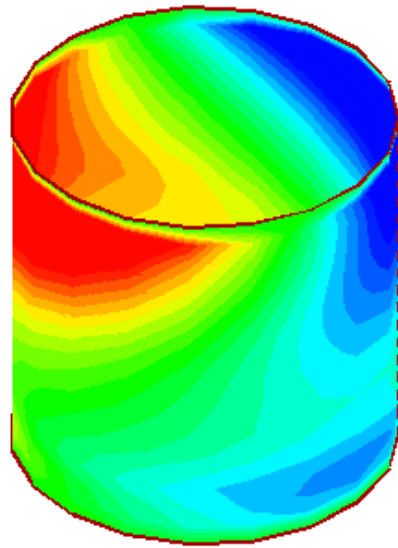
8a)



8b)



8c)



8d)

Fig 8: Temperature distribution at different times during the oscillation period, for the azimuthal travelling wave, at  $t = 488$  (s) in the case  $R = 0.06$  [K/s]

When the azimuthal velocity increases, a travelling wave appears, characterized by rotating temperature spots along the free surface of the liquid bridge (Figs. 8).

By looking at the liquid surface the time dependence is described by hot and cold spots rotating around the axis of the floating zone. A similar behaviour has been observed by thermocamera images during the Spacelab D2 mission<sup>8</sup>. The variation in time of the surface temperature distribution, in the oscillatory regime, shows a quite isothermal surface with hot and cold zones rotating around the bridge.

In this case the temperature disturbance is a periodic function of the time and of the azimuthal co-ordinate.

To illustrate these behaviours, four "numerical" thermocouples have been located along the free surface of the bridge (Fig 9a) at different azimuthal positions (with a shift of 90 degree). In Fig. 9b the differences between the computed temperatures of the two thermocouples T3 and T1, and T4 and T2, are shown for the standing wave regime ( $t = 253$  s). Fig 9c shows the results obtained for the travelling wave regime ( $t=488$  s).

For the standing wave model, the temperature difference between the numerical thermocouples T3 and T1 (corresponding to the pulsating hot and cold spots) is larger than the difference between T2 and T4.

In the travelling wave model the time temperature profiles show a phase displacement depending on the azimuthal co-ordinate. Since the critical wave number is  $m = 1$ , the oscillations show a phase displacement of  $\pi/2$  between two numerical thermocouples located at angular distance of 90 degrees.

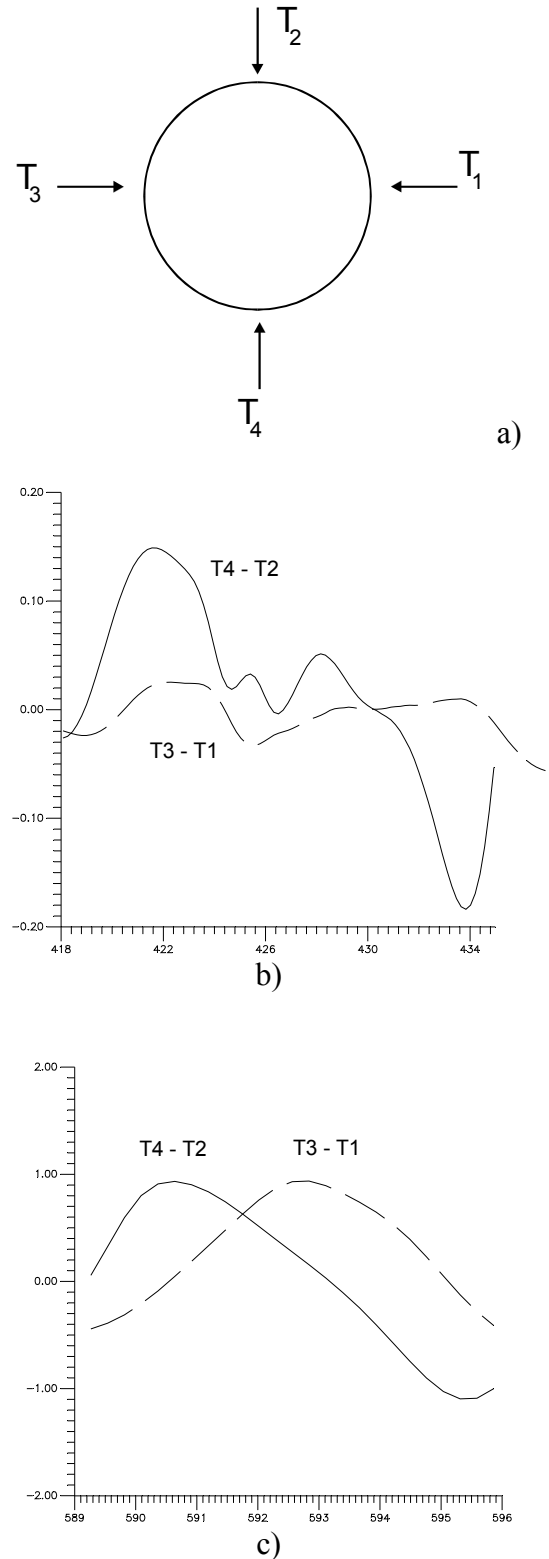


Fig 9: Variation in time of the temperature differences measured by numerical thermocouples azimuthally located along the free surface in the section  $z = 0.75 L$ . a) scheme of the thermocouples; b) standing wave; c) travelling wave

When the spots are located in correspondence of the thermocouples T2 and T4 the temperature difference is maximum, but the difference between T1 and T3 is negligible.

Conversely, since the spots are not azimuthally fixed but rotate, after a quarter of the period the temperature difference between T2 and T4 is negligible and the difference between T1 and T3 is maximum.

This result agrees with the experimental observations by Presser and Schwabe<sup>5</sup> who measured the temperature oscillations by two thermocouples located at the same axial and radial coordinate, but at different azimuthal coordinates, and found that the temperatures oscillate synchronously with the same frequency and a phase shift  $\Delta\theta$  depending on the azimuthal distance  $\Delta\varphi$ . The critical wave number was experimentally determined as  $m = \Delta\theta / \Delta\varphi = 1$

Moreover, the experimental results indicate that the distortion of the thermo-fluid-dynamic field, with respect to the basic axi-symmetric state, can be described by sinusoidal rotating disturbances in the form  $\hat{f}(z,r) \sin(\omega t \pm m\varphi)$ . This agrees with the present numerical results if the three-dimensional perturbations are analysed by subtracting the axisymmetric basic flow  $[f_0(z,r)]$  to the numerically computed three-dimensional time-dependent solution (basic flow + perturbation).

Since the surface flow in the azimuthal direction is coupled with the axial flow in the z-direction, if a vertical section is sketched for a fixed time, the three-dimensional flow is the result of both these effects. In Fig. 10 this situation is illustrated for a fixed time.

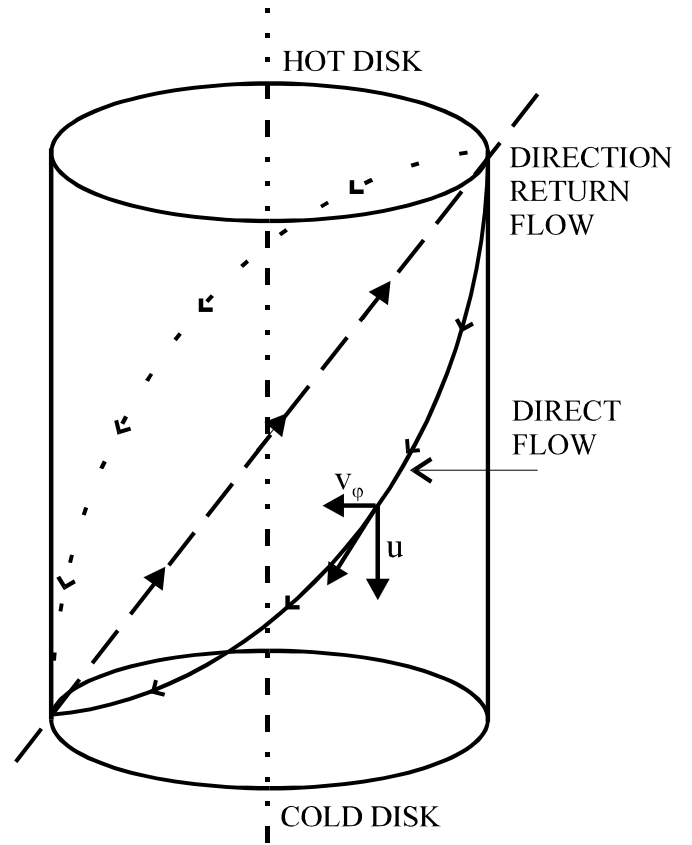
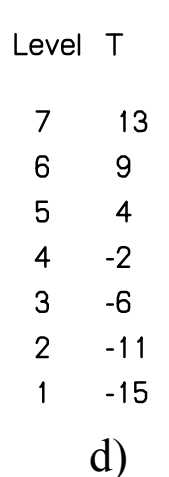
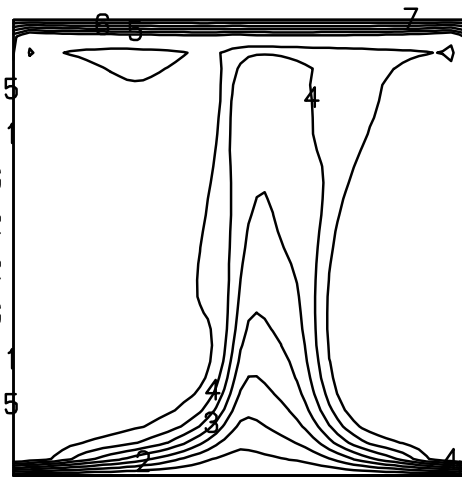
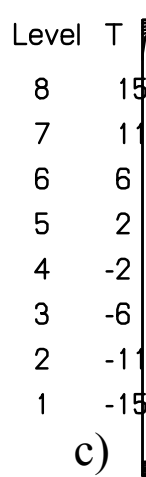
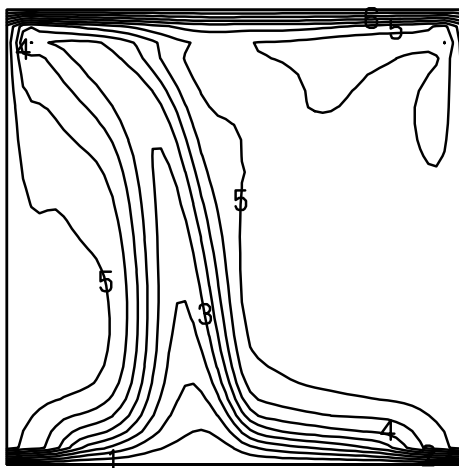
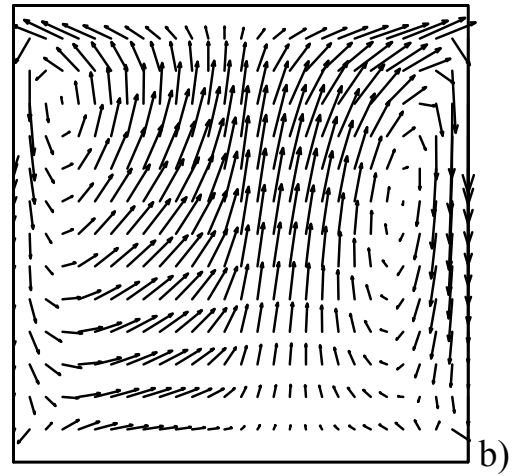
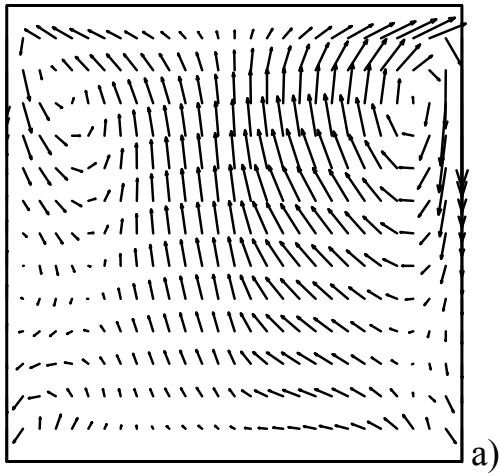


Fig 10: Scheme of the 3D flow in the bridge at a fixed time

Due to the oscillatory behaviour of the azimuthal and radial velocity components, the flow field is characterized by a full rotation of the deformed torous around the axis of the cylinder. This is shown in the Figs.11, where the vector plots (a,b) and the isotherms (c,d) in the meridian section  $\varphi = 0$ , are illustrated for the two times corresponding to the maximum and minimum of the oscillation in the period.



Figs.11: vector plots (a,b) and isotherms (c,d) in the meridian section  $\varphi = 0$ , are illustrated for the two times corresponding to the maximum and minimum of the oscillation in the period

### 3.3 Effect of the rate of ramping

In real experiments the "basic" axisymmetric field that becomes unstable is unsteady due to the time-dependent temperature boundary-conditions; therefore, different ramp degrees correspond to different critical temperature differences.

In particular, the experiments on sounding rockets, where only six minutes are available and fast rampings are needed, give higher values of the critical Marangoni numbers, compared to the ones obtained by linear stability analysis (steady basic states).

To analyze the effect of the rate of ramping we have performed different numerical simulations assuming three values of the constant  $R$  (Fig 12).

The results show that the critical  $\Delta T$  for  $R = 0.24$  K/s is almost 150% greater of that one obtained considering a ramp degree of 0.06 K/s .

$R$ [K/s]	$\Delta T_c$ [K]
0.06	7
0.12	11
0.24	19

Table 2 -  $\Delta T_c$  versus ramping rate

Moreover, we observed that when the rate of ramping increases and the onset is shifted to higher Marangoni numbers, the characteristic times for the onset of the axially running wave ( $t_1$ ) and of the azimuthal standing wave ( $t_2$ ) decrease . The characteristic time for the onset of the azimuthally travelling wave ( $t_3$ ) shows instead a different behaviour, and when  $R$  increases  $t_3$  increases.

	$R=0.06$ [K/s]	$R=0.24$ [K/s]
$t_1$ [s]	60	40
$t_2$ [s]	253	162
$t_3$ [s]	488	798

Table 3 - characteristic times versus ramping rate

This behaviour show that if the aim of the experiment is the evaluation of the critical Marangoni corresponding to a steady basic state (similarly to the one computed by linear stability analysis), a slow temperature ramp should be used. On the contrary, if the aim of the experiment is the study of the different modes of three-dimensional oscillatory convection, the optimization of the low microgravity time available, requires a rather fast temperature ramp.

The ramp, however, must not be too large because the time  $t_3$  considerably increases when the rate of ramping is very large.

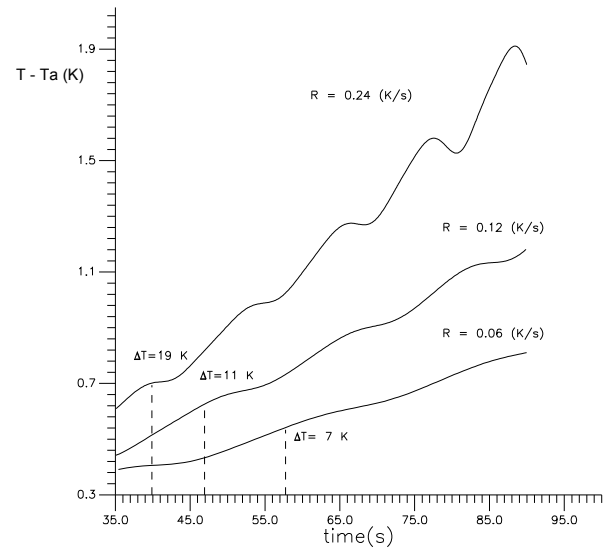


Fig 12: Temperature evolution measured by thermocouples located at  $z = 0.75 L$ ,  $r = 0.9R$ ,  $\varphi = \pi$

## 4 Conclusions

The results discussed in this paper have shown that three possible oscillatory flow configurations are possible: axisymmetric, pulsating, rotating.

The instability starts as an "axially running wave". In this first case the temperature and the velocity are periodic in time, but still axisymmetric, and the oscillatory mode is produced by

waves disturbances travelling in the axial direction only.

After a certain time two spots, one hot and the other cold, appear on the surface of the bridge and the resulting flow is the superposition of two counter-propagating azimuthal waves with equal amplitudes (azimuthally standing wave). In this configuration the spots pulsate non axisymmetric (the cold spot grows in time at the expense of the hot spot and vice versa). Finally, a third configuration appear, resulting from the superposition of two counter-propagating azimuthal waves with different amplitudes (wave travelling in the azimuthal direction).

The numerical calculation have been performed in preparation of a Maxus Sounding rocket experiment. The results pointed out that large temperature maxima (minima) occur in the interior of the liquid bridge, so that during the experiment, different thermocouples should be introduced in the liquid and located azimuthally in the transversal section near the hot disk. An other important result found by the present computation is that the temperature ramp must be not too large, in order to analyze the evolution of the instability from one model to the other, but at the same time not too small, in order to optimize the microgravity time available for the experiment.

## 5. References

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