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Unstable thermocapillary flow inside a $NaNO_3$ float zone

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Abstract

In this study, we have numerically investigated the problem of the transition from an axisymmetric state to an unstable i.e. oscillatory state in a liquid float zone which is held suspended between a pair of coaxial disks. The system of governing equations corresponding to a time-dependent-3D model was successfully solved by employing the modified-SIMPLE method. Results have shown that for a low Marangoni number, the flow consists of a steady, axisymmetric and toroidal structure exhibiting a 'purely' axial fluid circulation on the free surface. The vortex center is observed beneath that surface near the hot disk. When the Marangoni number increases beyond the critical value Ma_{CR}, $Ma_{CR} \approx 9750$, the transition to an oscillatory state occurs. The flow and the thermal field become clearly three-dimensional and time-dependent, with the existence of a distorted 'saddle-shape-like' torus which exhibits its 'rotating' and dynamic character around the circumference. The fluid flows following a curved pattern on the free surface. The frequency of the oscillations was evaluated to be $\approx$0.4Hz. These oscillations are believed to be due to the azimuthally travelling thermal disturbances. The influence of the heating ramp-rate on the occurrence of the unstable flow has also been studied. It has been found that Ma_{CR} increases with the ramp-rate, thus indicating that the flow appears to be more stable to a thermal disturbance.

1 Introduction

The float zone technique, because it offers a minimum risk of cross-contamination, has become one of the most interesting techniques to produce

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large and high quality crystals, especially when used under reduced gravity in space. However, the existence of the so-called 'Marangoni flow' or the thermocapillary flow may drastically affect the internal velocity and temperature fields within the liquid zone [1-4]. A review of the previous works on this interesting topic may be found in [5,6]. The temperature fluctuations resulting from this thermocapillary flow may even conduct to an unstable flow. In fact, some experimental observations have eloquently shown that a sufficiently vigorous thermocapillary flow may become unstable i.e. oscillatory [2,3]. The existence of such an oscillatory behaviour has raised many studies both experimentally and numerically, see for example [7-11]. In spite of these tremendous efforts deployed, the real physical mechanism governing the axisymmetric-oscillatory transition has not yet been completely understood. Therefore, more additional investigations will be necessary. In this ultimate objective, a full 3D-time-dependent model has been proposed in order to study the flow dynamic behaviours under the critical i.e. high Marangoni number condition. An extended numerical simulation was carried out for several fluids under μ-g environment. Some significant results obtained for a $NaNO_3$ half-zone will be presented and discussed in the next with an emphasis on the oscillatory flow structure as well as on the influence of the heating ramp-rate on the occurrence of the instability.

2 Mathematical formulation

2.1 Governing equations

The problem studied consists of a cylindrical half-zone of a molten liquid which is suspended between a pair of coaxial and parallel disks (Fig. 1). The dimensions of the zone are R_0 and H. The disks are stationary and have uniform temperatures $t_1=f(\tau)$ and $t_2= t_M$ where $f(\tau)$ is *a priori* known function of time, and t_M is the melting temperature of the material considered. For a proper mathematical formulation of the problem, the fluid is assumed to be Newtonian and incompressible with constant properties (all evaluated at t_M) except for the surface tension σ which is considered to be a linearly decreasing function of the temperature. Furthermore, the viscous dissipation is also assumed negligible.

Under the above conditions, the dimensionless governing equations are :

$$\nabla \cdot \mathbf{V} = 0 \tag{1}$$

$$\partial V_i / \partial \tau^* + \nabla \cdot (\mathbf{V} \cdot V_i) = -\nabla P + (Pr/Ma)\, \nabla^2 V_i + S_i, \quad i = 1,2,3 \tag{2}$$

$$\partial T / \partial \tau^* + \nabla \cdot (\mathbf{V} \cdot T) = (Pr/Ma)\, \nabla^2 T \tag{3}$$

where $\mathbf{V}=(V_R, V_\theta, V_Z)$ is the fluid velocity vector; τ^* is the dimensionless time; S_i are the velocity-related stress terms given as follows :

- <u>for i=1, the R-direction</u>

$$S_1 = (V_\theta^2/R) - (Pr/Ma)\{ V_R/R^2 + (2/R^2)\, \partial V_\theta/\partial\theta \} \tag{4a}$$

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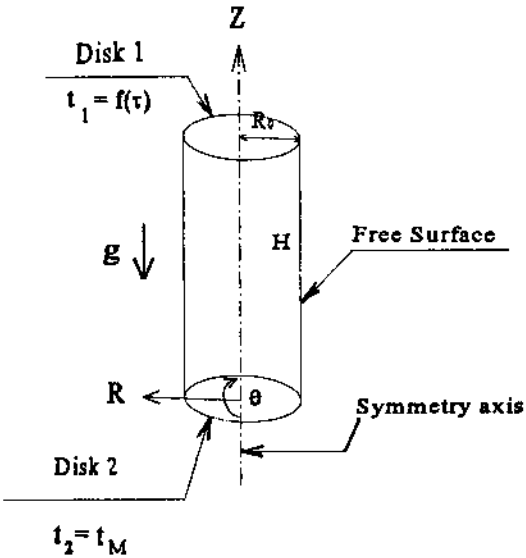

Figure 1: Geometry configuration of the problem studied.

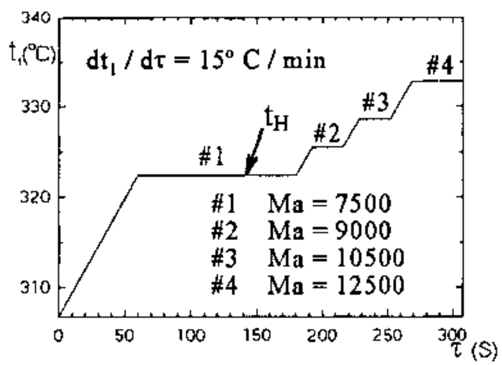

Figure 2: A typical time-evolution of the temperature $t_1(\tau)$ as imposed in the numerical simulations.

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- for i=2, the θ-direction

$$S_2 = (Pr/Ma)\{ (2/R^2)\, \partial V_R/\partial\theta - V_\theta/R^2 \} - V_R V_\theta/R \qquad (4b)$$

- for i=3, the Z-direction

$$S_3 = 0 \qquad (4c)$$

It is important to mention here that the physical quantities H, $\{|\partial\sigma/\partial t|\Delta T/\mu\}$, $\{H\mu/|\partial\sigma/\partial t|\Delta T\}$, $\{\rho[|\partial\sigma/\partial t|\Delta T/\mu]^2\}$ and $\Delta T \equiv t_H - t_M$ have respectively been adopted as the reference length, velocity, time, pressure and the temperature difference (note that t_H refers to the hot disk temperature when it reaches its constant level the end of each step during the heating process). A dimensionless variable is defined by normalising its value with respect to the corresponding reference quantity listed above. In particular, the dimensionless temperature is defined as follows :

$$T = (t - t_M) / \Delta T \qquad (5)$$

2.2 Boundary and initial conditions

The conservation equations (1-3) constitute a set of non-linear and highly coupled PDE. It is subjected to the following boundary and initial conditions :

- on the disks, the usual non-slip and non-penetration conditions prevail. The hot disk i.e. disk no. 1 has an imposed temperature t_1 which is function of time with a fixed ramp-rate $dt_1/d\tau$, for example $dt_1/d\tau$ = 7.5°C/min. (Fig. 2); while the cold disk temperature is set to t_M as mentioned before.
- the liquid free surface is assumed to be cylindrical (i.e. radially non-deformed, V_R=0) and adiabatic. Also, the following equations expressing the balance of the shear stress along the Z and θ directions must be satisfied :

$$\partial V_Z/\partial R = - \partial T/\partial Z \qquad (6a)$$

$$(1/A)\, \partial T/\partial\theta = - \{ \partial V_\theta/\partial R - V_\theta/A \} \qquad (6b)$$

- as initial condition, we consider the quiescent liquid zone having an uniform temperature equal to t_M throughout at the beginning of the heating process i.e. at $\tau=\tau^*=0$.

The above governing equations and their boundary conditions reveal that the problem under study may be characterised by a set of three dimensionless parameters, namely the aspect ratio A, the Prandtl number Pr and the Marangoni number Ma. Their definition is given as follows :

$$A = R_0/H \quad ; \quad Pr = \nu/\alpha \quad ; \quad Ma = |\partial\sigma/\partial t|\, \Delta T \cdot H / \mu\alpha \qquad (7)$$

where α, μ and ν are, respectively, the fluid thermal diffusivity, dynamic and kinematic viscosity.

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3 Numerical method and code validation

In the present study, we have employed the modified-SIMPLE method [13] to successfully solve the system of the governing equations (1-3) subject to the above boundary and initial conditions. The exponential scheme [14] has been used throughout to compute the so-called 'combined-convection-and-diffusion' fluxes of heat and momentum. Several non-uniform and staggered grids were proposed and extensively tested. The $26 \times 26 \times 24$ grid (26, 26 and 24 nodes along the Z, R and θ directions) with highly packed nodes near the disks and the free surface has finally been adopted for the task demanded since it provided an excellent accuracy of results even at high Marangoni number with a reasonable computing time. The time-step $\Delta\tau$ was fixed to be 1/50 S in order to be able to detect any changes in the flow structure. The convergence monitoring was mainly based on the value of the so-called 'residual mass' which is resulting from the integration of the mass-conservation equation (1) over a finite control volume. The converged solution has consistently been obtained with a very low residual mass (usually not exceeding 0.001% on the relative basis). There was no problem related to the convergence observed so far during all of the numerical simulation.

The model has been successfully validated through several test-cases. At first, the numerical results generated for a 2D-axisymmetric half-zone of Silicon-oil (Pr=196.5) under the transient-1g condition have been satisfactorily compared to the corresponding numerical and experimental data [15] (note that for cases under 1-g, the presence of a deformed free surface has been taken into account). A good agreement has also been found while comparing the results with the experimental data [2] for a $NaNO_3$ (Pr=8.9) half-zone as well as with the empirical correlations [16] for the fluid axial velocity on the zone free surface. The complete details regarding the study of the grid independence and the code validation procedure were presented in [17,18].

4 Results and discussion

The numerical simulation was carried out for two different fluids, namely Silicon (Pr=0.016) and $NaNO_3$ (Pr=8.9). In the following, some significant results as obtained for a $NaNO_3$ half-zone will be presented and discussed with an emphasis on the oscillatory flow structure as well as the effect of the heating ramp-rate $dt_l/d\tau$ on the occurrence of the axisymmetric-oscillatory transition.

Figure 2 shows, at first, the typical time-evolution of the hot disk temperature $t_l(\tau)$ as imposed during the heating duration for $NaNO_3$, with $dt_l/d\tau$ fixed to be 7.5°C/min (two other values of $dt_l/d\tau$ were also considered, namely 15 and 40°C/min). It is important to note that at the end of each step when t_l reaches a constant value, a sufficient elapse time has been allowed to the flow to reach its asymptotic state. A typical simulation case required a computing time as much as 170 hr on a Pentium PC. For the results shown in the following, the aspect ratio A was fixed to be 0.732.

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4.1 Structure of the flow and the thermal field

It has been clearly observed that for a low Marangoni number i.e. low ΔT between the disks, the flow remains perfectly steady and axisymmetric. Its structure is characterised by an usual toroidal cell with the vortex center located beneath the liquid free surface on the side of the heated disk. The fluid circulation in the region immediate to the free surface is more pronounced than elsewhere in the zone. It should be noted that the fluid circulation is 'purely' axial on the free surface. Such a features are known as characteristics of a surface-tension-driven flow as the one under study here. We also notice a temperature stratification inside the liquid zone, in particular near the disks where high temperature gradients have been found. For a higher Ma number, the above thermocapillary convection flow is more important and consequently, the temperature stratification becomes more considerable [9,18].

At sufficiently high ΔT between the disks, say for $Ma > Ma_{CR} \approx 9750$ (for $dt_l/d\tau = 7.5$°C/min), it is very interesting to observe that the flow as well as the thermal field clearly become oscillatory in time. Figure 3 shows, for the case no. 3, the time-evolution of the fluid temperature at a specific point located at Z=0.5 on the free surface. The oscillatory character of the case studied is obvious. It is interesting to note that for this case, the flow nearly reaches its fully-developed state at the end of the elapse time shown. The frequency of the oscillations has been estimated to be ≈0.4 Hz by using a FFT technique.

Figure 4 show, for the case no. 3, the instantaneous snap-shot of the velocity field in four different R-Z planes. Although it may be somewhat difficult to visually detect changes between the views shown here, the detailed results have shown that the flow structure clearly becomes three-dimensional and rather complex. Although the usual toroidal form still remains, the latter is now distorted and taking a form of a 'saddle-shape-like' torus where the vortex center exhibits extreme shifts in its position along both the R and Z directions and this, from one angular plane to another. Such a shifts, which produce tremendous effects on the velocity field in the whole zone, have been found to be more pronounced for cases at higher Marangoni number [18]. It should be noted that the views shown in Fig. 4 are instantaneous i.e. at a specific time. In fact, the entire flow and thermal fields exhibit a strikingly 'rotating' character around the zone main axis, thus producing an oscillatory behaviour of dependent variables at a fixed location such as the one shown previously in Fig. 3.

Figure 5 shows, for the case Ma=10500 considered, the instantaneous structure of the isotherms on the free surface as well as in the R-θ plane at Z=0.5. One may notice some distortions on the isotherms in a central region of the liquid free surface, while in the vicinity of the disks, the isotherms remain nearly straight and parallel to the latter. Thus, as expected, the presence of the solid disks plays a certain 'damping' role against any disturbance in the flow. In fact, our numerical results [17,18] have shown that for a particular Ma number, the amplitude of the oscillations is clearly less important in the region near the disks than elsewhere in the zone. Also, it has been found that this amplitude increases appreciably with the increase of the parameter Ma, thus indicating that the flow

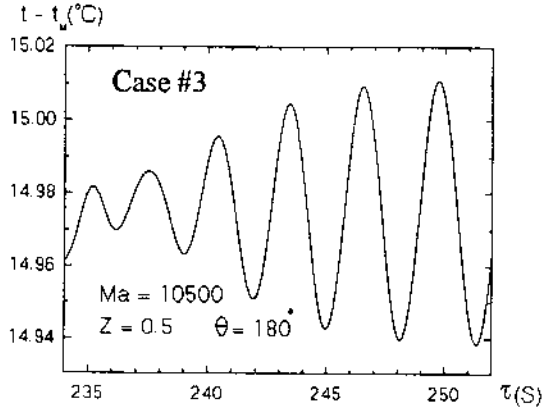

Figure 3: Time-variation of the fluid temperature at a specific point located on the zone free surface.

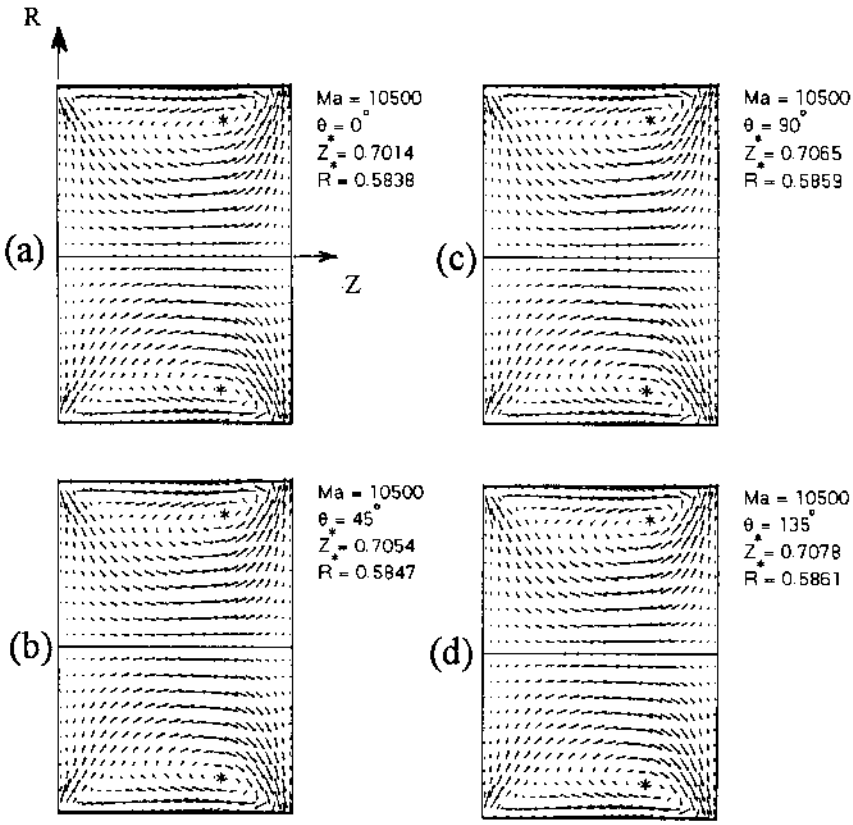

Figure 4: Instantaneous snap-shot of the structure of the velocity field in four different R-Z planes for the case No. 3.

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seems to become more vulnerable to a disturbance under a high temperature difference ΔT between the disks. Such behaviour, which appears to be physically quite realistic, has been found to be consistent with that observed experimentally [2]. From a close examination of the isotherms structure on the liquid free surface while taking into consideration the behaviour of the fluid circulation on that surface, one can see that the instability observed so far on the flow may be resulting from some thermal disturbance waves travelling azimuthally around the circumference [2,11,20].

Figure 6 shows finally an illustrative view (not to scale) of the instantaneous pathway of the vortex center. This pathway, as described earlier, follows a 'saddle-shape-like' curve in space where the extreme positions of the vortex center are represented by the highest and the lowest points of the curve along the Z-direction.

4.2 Comparison with other experimental and numerical data

Table 1 shows the comparison of the values of Ma_{CR} and the oscillations frequency as obtained in this study with other numerical and experimental data by several researchers. The agreement can be regarded as very good. One may notice, as stated in [19], that the critical Marangoni number seems to do not change very significantly with the level of the gravity.

Table 1. Comparison with other experimental and numerical data

	This study	[7] Num	[19] Experimental		[20] Exp.	[2] Exp.
	μ-g	μ-g	μ-g	1-g	μ-g	1-g
Ma_{CR}	9750	11633	9444±250	8907±150	≈9000	≈8000
Frequency (Hz)	0.4	0.56	0.48	0.51	0.5	0.57
$dt_l / d\tau$ °C/min.	7.5	N/A	0.21	0.21	6	0.1

4.3 Effect of $dt_l/d\tau$ on the axisymmetric-oscillatory transition

In order to study the effects of the heating ramp-rate on the transition to an unstable flow, the numerical simulations were also carried out using two other values of $dt_l/d\tau$, namely 15°C/min and 40°C/min. The occurrence of the instability was carefully determined during the very-time-consuming calculation procedure corresponding to the heating duration. Table 2 shows the dependence of Ma_{CR} as well as the amplitude of the oscillations on $dt_l/d\tau$. It is clearly observed that the flow appears to be more stable under higher ramp-rate so that the transition to the oscillatory state is delayed. This result, which appears to be somewhat paradoxical, may be explained by the fact that for a fluid with a

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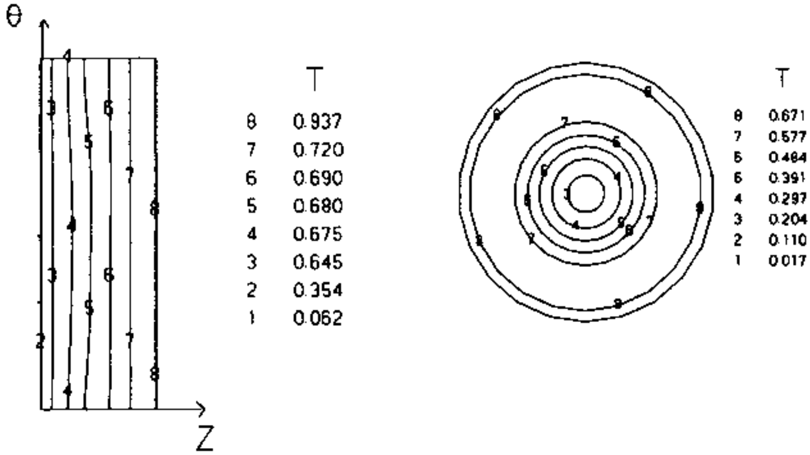

Figure 5: Instantaneous structure of isotherms for the case No. 3.

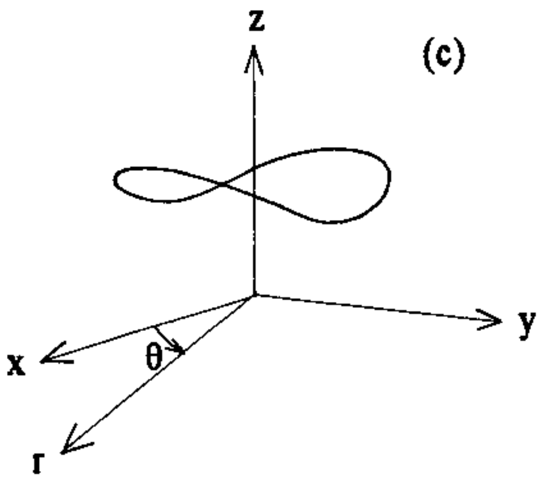

Figure 6: Illustrative view of the instantaneous pathway of the vortex center (not to scale)

Table 2. Influence of $dt_1/d\tau$ on the value of Ma_{CR}.

$dt_1 / d\tau$ (°C / min)	Ma_{CR}	Peak-to-Peak Amp. (°C)
40	12500	0.83
15	10500	0.07
7.5	9750	0.05

relatively high viscosity as $NaNO_3$ liquid, imposing a rapidly and continuously increasing surface-tension-driving-force on the free surface tends to prevent the growth and the propagation of any disturbance within the field, stabilising therefore the flow 'base-state' i.e. the axisymmetric one. As consequence, the latter may persist longer until the thermocapillary convection effects become sufficiently vigorous enough to destabilise the flow, thus permitting a disturbance to set in. It is very interesting to note that a similar behaviour regarding the effect of the heating ramp-rate has also been observed by Savino and Monti [11] for a moderately high Pr number fluid, say Pr = 30 and 74. Finally, from the above discussed trend, one may expect that the value of Ma_{CR} as obtained from a real experimental platform (where high values of $dt_1/d\tau$ were often used due to a lack of time) would be, in general, higher than the corresponding ones obtained from the theoretical models based on the classical linear-stability theory for which a near-zero ramp-rate was usually prescribed.

5 Conclusion

In this work, the problem of the transition to an unstable thermocapillary flow inside a $NaNO_3$ half-zone under μ-g condition has been studied. Results have shown that for a low Ma number, the flow 'base-state' consists of a steady and axisymmetric toroidal cell. For a sufficiently high Ma number, say $Ma > Ma_{CR} \approx 9750$, the transition to an oscillatory state occurs. The 3D and time-dependent flow structure is characterised by the existence of a distorted 'saddle-shape-like' torus which exhibits a 'rotating' behaviour around the zone main axis. The value of Ma_{CR} and the oscillations frequency are in very good agreement with other experimental data. It has also been found that the flow appears to be more stable to a disturbance when a high heating ramp-rate is imposed.

6 Acknowledgements

We sincerely thank the Natural Sciences and Engineering Research Council of Canada, the Ministry of Intergovernmental and Aboriginal Affairs of New Brunswick, the 'Ministère de l'Éducation du Québec' and the 'Université de Moncton' for the grants received for this project. Thanks are also due to the School of Engineering of the 'Université de Moncton' for allowing the computing facilities.

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