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New features in the fluid-dynamics of Floating zones under Marangoni effect

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Summary

The article presents a discussion of the floating-zone (FZ) problem and of the related three-dimensional flow instability in terms of numerical simulations and prototype applications. The difference between the "half-zone" and the "full-zone" and the related possibility to use one or the other configuration to capture the FZ fundamental hydrodynamics are elucidated. The first bifurcation of the flow is investigated in the case of semiconductor melts for various environments (microgravity and normal gravity) and geometrical parameters (wide ranges of aspect ratio and volume). In the case of Silicon the axi-symmetric flow loses its stability to a steady three-dimensional flow. The model (half or full column) used for the simulations, however, plays a critical role in determining the related azimuthal structure and critical threshold. For $Pr=0.02$ (Gallium) and on-ground conditions, a surprising behaviour is observed. The flow may become unstable directly to an oscillatory disturbance.

Introduction

In the last years increasing interest has been directed towards crystals of semiconductors and towards the crystallisation process of special doped metals and alloys. In fact, single crystals with superior purity and structural quality are needed to support evolutionary progress in material science and computer technology. Current experimentation along these lines is enabling the production of limited quantities of high-quality electronic materials and of materials that exhibit unique properties for use as benchmarks. Within this context, of particular relevance is the Floating Zone (FZ) technique that seems to be one of the best means for obtaining such advanced crystals.

This method has been often used in the terrestrial environment. Additional purity and quality of the specimens however has been obtained in the weightlessness conditions provided by sounding rockets and other orbiting platforms where buoyancy driven convection (which is an important cause of crystal imperfections) is suppressed. Also in absence of gravity however the melt can be

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affected by the presence of another type of convection induced by surface tension forces (the so-called Marangoni flow). This convection is dominant in space and even under normal gravity conditions in situations where the Marangoni flow is emphasised with respect to buoyancy forces (microscale processing). Further improvements in this field require in principle an exhaustive knowledge of the structure of the flow within the melt. For these reasons various models of the FZ technique have been introduced over the last years for capturing the underlying relevant fluid-dynamics in terms of basic flow, related transitions and bifurcations (see Fig.1).

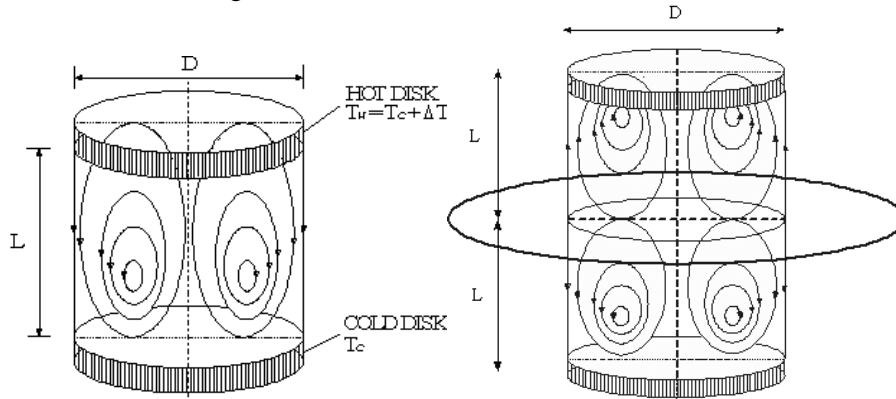


Fig. 1: a) The half-zone and its typical boundary conditions b) The full-zone heated by an equatorial ring heater

The half-zone model simulates half of a real floating zone (the liquid between one of the ends of the domain and the equatorial plane); it consists of a pair of coaxial, differentially heated cylindrical disks with a bridge of liquid material suspended between them (heat flow is neglected through the free surface).

The ends of the full-zone are plane and isothermal as in the case of the half-zone, but the supporting disks are posed at the same temperature (Fig.1b). The presence of a ring heater around the equatorial plane of the zone is simulated imposing a specified heat flux distribution on the free surface with a maximum of the flux in correspondence of the equatorial plane.

The geometrical aspect ratio of the liquid column of full-zone extent (A_F) is defined as $A_F = 2L/D$; the aspect ratio of the corresponding half-zone is defined as $A_H = L/D = A_F/2$ (D is the diameter of the supporting disks). The so-called volume or shape factor is defined as ratio of the volume held between the supporting disks and the volume of the corresponding straight configuration with cylindrical liquid/air interface. The Marangoni number reads $Ma = Re \cdot Pr = \frac{\sigma_r \Delta T L}{\mu \alpha}$ where ΔT is the temperature gradient on the free surface.

Cylindrical interface and microgravity conditions

The major outcome of computations carried out for the full-zone under the constraint of cylindrical interface and zero-g conditions is that for both even and odd critical azimuthal wave-numbers the mirror symmetry with respect to the mid-plane is broken (see e.g. Fig. 2 showing the surface azimuthal velocity).

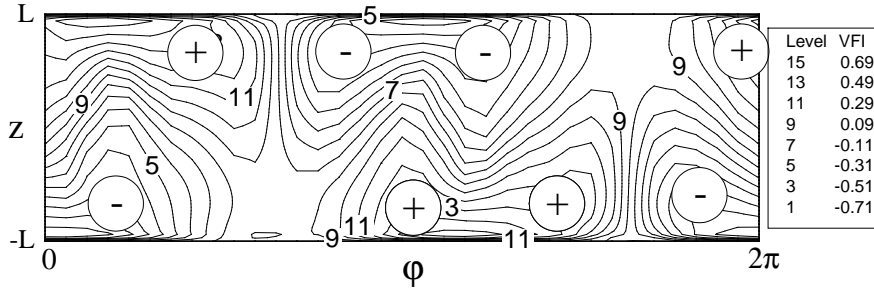


Fig. 2: Surface azimuthal velocity distribution at the 3D stationary state ($A_F=1.3$, $m=1$, $Ma \approx 30$, zero-g).

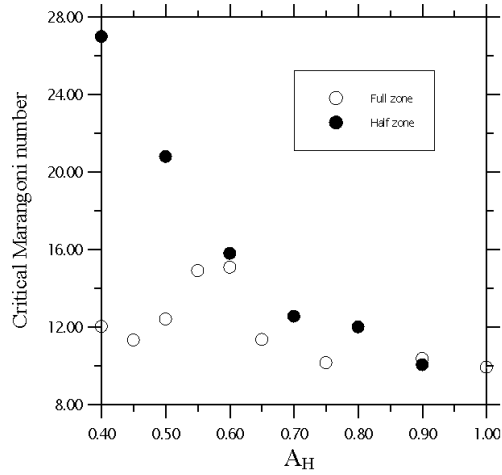


Fig. 3: Comparison between the critical Marangoni number (Ma_{c1} , first bifurcation of the axi-symmetric flow) for the full-zone configuration (Lappa [3]) and available results (Chen et al. [1] and Imaishi et al. [2]) dealing with the half-zone configuration ($Pr=0.01$).

This finding is of paramount importance if one considers that, in the light of this result, the half-zone (supposed to model the flow in a half of the real floating zone under the constraint that this flow is symmetric by reflection about the plane at mid-height between the rods) should not be used to obtain quantitative data about the FZ technique.

The aforementioned symmetry breaking is the main reason for which the Ma_{c1} values computed for the cylindrical half-zone model by several investigators (e.g. Chen, Hu, Prasad [1]; Imaishi et al. [2]) are two times higher than the values recently calculated for the case of full-zone model (see Fig. 3). The flow symmetry with respect to the mid-plane is an additional and relevant part of the problem. It indeed introduces a new degree of freedom. The differences between the two configurations however are not limited to this feature (see below).

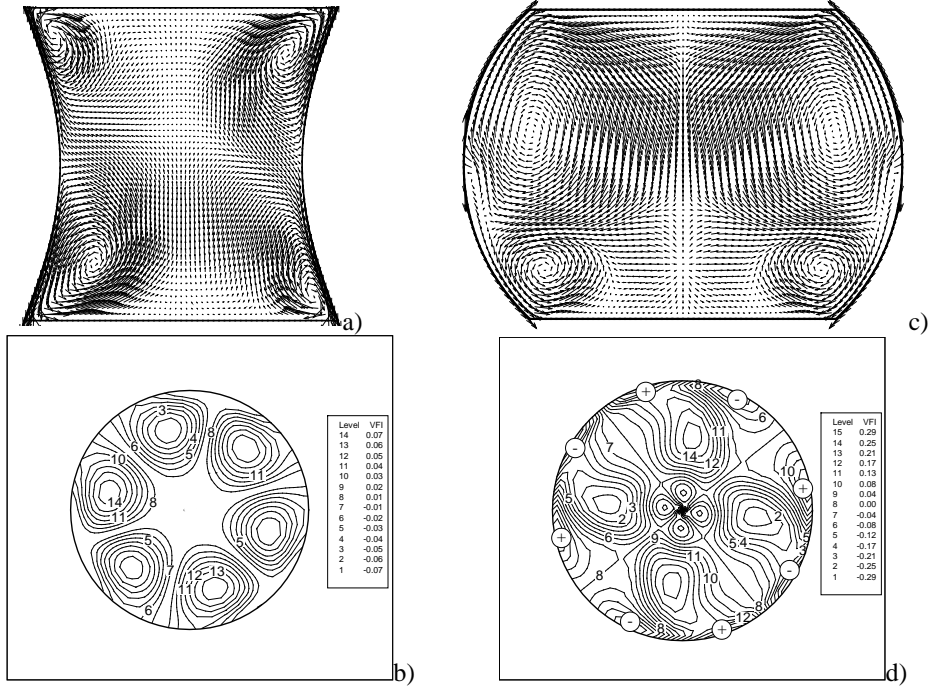
Concave and convex volumes in microgravity

It is known (see e.g. Lappa et al. [4]) that in the case of liquid metals and half-zone, for a fixed aspect ratio the critical azimuthal wave-number can be shifted to higher values by increasing S (convex shape) or to lower values by decreasing S (concave shape). The full-zone exhibits a quite different behaviour since the azimuthal wave-number increases in case of deviation from a volume equal to the cylindrical one for both cases $S < 1$ and $S > 1$ (Tables I and II).

Table I: Critical azimuthal wave-number and critical Marangoni number versus the volume of the liquid zone ($Pr=0.01$, full-zone, microgravity conditions).

A_F	L [cm]	$S=V/V_o$	m	Ma_{c1}
1.0	0.5	0.80	4	48.08
1.0	0.5	0.85	3	34.59
1.0	0.5	1	2	12.413
1.0	0.5	1.2	2	6.85
1.0	0.5	1.3	2	10.87
1.5	0.75	1	1	10.15
1.5	0.75	1.2	1	1.454
1.5	0.75	1.4	2	11.53

The new degree of freedom plays a crucial role also with regard to these aspects: In principle each convection roll is bounded by a wall from one side and it is free to interact in non linear way with the opposite convection roll from the other side. It can puff up crossing the mid-plane and protruding in the opposite side of the liquid zone; however this degree of freedom is somehow reduced in the case $S < 1$. In case in fact, due to the concave shape of the free surface, the convection rolls are confined each to the respective half of the full zone configuration (Fig. 3a; this prevents mutual interference and strengthens the constraints for the flow field leading to a larger value of the critical Marangoni number). On the contrary, for $S > 1$ each toroidal roll is free to interact with the opposite roll and to become pervasive throughout the system (Fig. 3b) lowering the stability threshold. Such an interaction is also responsible for the "apparent" doubling of the azimuthal wave number in the mid-plane shown in Fig. 3d.



Figs. 3: On the left, structure of 3D Marangoni flow with $m=3$, $A_F=1.0$, $S=0.85$, $Ma=50$, zero-g; (a) velocity field in the generic meridian plane; (b) azimuthal velocity in the equatorial plane. On the right, structure of 3D Marangoni flow with $m=2$, $A_F=1.0$, $S=1.3$, $Ma=25$, zero-g; (c) velocity field in the generic meridian plane; (d) azimuthal velocity in the equatorial plane (the wave number therein seems to be $m=4$).

On ground conditions

Table II: Critical azimuthal wave-number and critical Marangoni number versus the volume of the liquid zone ($Pr=0.01$, full-zone, $L=0.5$ [cm], $A_F=1.0$, on ground conditions, after Lappa [5])

$S=V/V_0$	m	Ma_{c1}
0.80	3	33.24
0.85	3	27.99
0.90	2	12.39
1	1	8.40
1.1	1	3.31
1.2	2	9.54

Comparison between Tables I and II for the case of full-zone gives insights into the effect of gravity on the features of the instability of Marangoni flow. For a

volume equal to the cylindrical one the on ground Marangoni flow is destabilised with respect to microgravity conditions and the azimuthal wave-number is shifted to a lower value. The same trend occurs in the case of $S < 1$; gravity acts destabilising the Marangoni flow. Vice versa stabilisation occurs in the case $S > 1$. For the half-zone, the scenario completely changes. For instance for $S=1$, if the on-ground deformation of the shape is taken into account, gravity always stabilises the Marangoni flow regardless of its direction (parallel or anti-parallel to the axis).

The three-dimensional flow structure is different according to the heating direction (from above or from below). In the latter case, the critical Marangoni number is larger and the critical wave-number is smaller, compared with the opposite condition.

Table III: Critical Marangoni number for the half-zone under normal gravity conditions ($L=1$ [cm], $A_H=1$, $S=1$, Silicon, $Pr=0.01$) after Lappa, Yasuhiro and Imaishi [6].

	Cylinder 1g Heating from above	Cylinder Zero g	Cylinder 1g Heating from below	1g shape Heating from above	1g shape Heating from below
Ma_{c1}	15.36	15.24	14.92	16.62	18.78

The effect of the shape and of the heating condition can be even more dramatic in the case of particular melts for which the deformation is more pronounced than the case of Silicon (for instance Gallium since its surface tension is lower). For the case of Gallium ($A_H=1$, $S=1$ and heating from below condition) in fact the steady bifurcation is suppressed and instability occurs directly in the form of an oscillatory disturbance (Lappa, Yasuhiro and Imaishi [6]).

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