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Review: Possible strategies for the control and stabilization of Marangoni flow in laterally heated floating zones

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Abstract: The paper presents a comparative and critical analysis of some theoretical/experimental/numerical arguments concerning the possible stabilization of the surface-tension-driven (Marangoni) flow in the Floating Zone technique and in various related fluid-dynamic models. It is conceived as a natural extension of the focused overview published in Cryst. Res. Tech. 40(6), 531, (2005) where much room was devoted to discuss the intrinsic physical mechanisms responsible for three-dimensional and oscillatory flows in a variety of technological processes. Here, a significant effort is provided to illustrate the genesis of possible control strategies (many of which are still in a very embryonic condition), the underlying ideas, the governing nondimensional parameters, the scaling properties. Particular attention is devoted to their range of applicability that is still the subject of controversies in the literature. The discussion is supported by some novel numerical results. These simulations are used to provide additional insights into the physics of problems where experimental data are not available.

keyword: Floating Zone Technique, Marangoni flow, convective instabilities, thermal feedback control, Magnetic fields, Forced high-frequency vibrations, Thermovibrational effects.

1 The FZ technique

During the Floating-Zone (FZ) process a melt zone is established between a lower seed material and an upper feed material by applying localized heating (see, e.g. Fig. 1). This floating zone is moved along the rod (by means of relative motion of the heating device) in such a way that the crystal grows on the seed (which is below the melt) and simultaneously melting the feed material above the floating zone. The seed material as well as the feed rod is supported but no container is in contact with the growing crystal or the melt, which is held in place only by surface tension. Therefore, the key characteristic of this method is that the molten zone does not need to be in contact with a foreign solid (crucible) that, besides being awkward to realize in the practice (the working temperature of the crucible must be well above the 1690 K of the melting temperature of silicon, e.g.), would introduce impurities unacceptable for the applications envisaged (molten silicon is a very reactive material).

Of course, containerless processing on massive samples can only be done in the microgravity environment of space where the forces used for suspending and manipulating the specimens are not overwhelmed by gravity. Microgravity requires much smaller forces to control the position of containerless samples, so the materials being studied are not disturbed as much as they would be if they were levitated on Earth.

Under Earth conditions the zone height is limited because the liquid tends to run down when the molten zone becomes too big; this fact limits the possible diameter of crystals that are grown in Earth's gravity. In space, the maximum zone height is given by the circumference of the crystal; therefore, floating-zone experiments with higher zone heights and larger diameters become feasible. Also, in such environment, buoyancy forces, that are the most important cause of crystal imperfections on Earth, are absent or very weak.

Along these lines, microgravity experimentation is enabling the production of limited quantities of high-quality large-size samples that exhibit unique properties for use as benchmarks. This pioneering research is leading to next-generation commercial crystal products (for instance, most of the very pure silicon produced today is processed by the Floating-Zone technique).

Crystal growth by the FZ technique in space was pioneered by Eyer et al. (1984). Despite the advantages offered in terms of size and purity by this environment (see also Muller, 1988 and Benz, 1990), however it was

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observed that, even in the absence of gravity the presence of Marangoni convection can be responsible for the presence of defects in the crystals (the characteristic number of this type of convection is defined as Ma=$\sigma_T \Delta TL/\mu\alpha$ where L is a reference length, α the melt thermal diffusivity, μ the dynamic viscosity, ΔT the temperature difference along the free interface and σ_T the derivative of the surface tension with respect to the temperature; also, Ma=Re·Pr where Re=$\sigma_T \Delta TL/\rho\nu^2$, ν being the kinematic viscosity and Pr=ν/α being the Prandtl number).

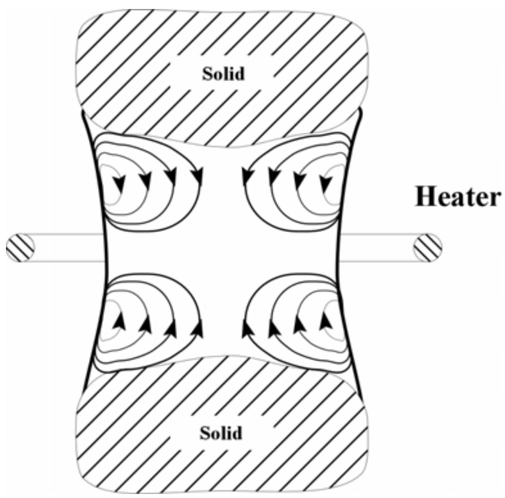

Figure 1 : The floating zone (FZ) technique.

Subsequent theoretical and numerical studies disclosed that this type of convection can undergo a primary bifurcation from axisymmetric motion to steady three-dimensional flow (Ma_{c1}) and a second transition to time-dependent convection (Ma_{c2}), the first being responsible for the presence of macroscopic defects (radial segregation with a non-axisymmetric distribution), the latter for microscopic striations (microscopic variations of the dopant concentration); these striations are mostly caused by fluctuations of the microscopic growth rate or the mixing of the melt; the fluctuations in turn result from the aforementioned time-dependent flow in the melt, caused by oscillatory pure Marangoni convection under zero-g or by oscillatory mixed buoyant-Marangoni convection under normal gravity conditions.

2 Possible models

One of the major difficulties in the experimental/theoretical analysis of Marangoni flow in real floating zones is that, due to phase change related to melting and solidification of the material, the geometry of the boundary of the liquid volume is not known a priori. Irregular melting and freezing interfaces and thermal conditions associated with latent-heat effects make the analysis of the features of the Marangoni flow very difficult from an experimental point of view. These difficulties led the investigators, in the past, to create a new configuration, the so-called half-zone (usually referred to as liquid bridge). This model of a float zone melt was introduced, in fact, in the mid-1970s as a vehicle for performing experiments in well-controlled conditions. It simulates half of a real floating zone (the liquid between one of the ends of the domain and the equatorial plane) and consists of a pair of coaxial, solid cylindrical disks (one hot and the other cold) with a bridge of liquid material suspended between them.

Although it is a very crude simplification, one may distinguish between materials-science-oriented research and fluid-science-oriented research, according to whether the interest is centered in the microstructure of the final product (grown silicon crystal, e.g.) or on understanding the molten zone behavior during the processing.

Both the floating zone and the liquid bridge are held by surface tension forces (capillarity), spanning between two sharply-edged coaxial solids against the natural tendency of liquids to adopt a spherical shape in the absence of other forces, and the tendency to creep down the rod in a gravity field. The real FZ, however, is not a static configuration; rather, as explained before, it is a dynamic process governed by temperature gradients that force the tip of the feeding rod to melt and the tip of the grown material to freeze. Nevertheless, the mechanical model of a quasi-steady series of liquid bridges has already shown to be relevant to some key aspects of the problem. For instance, by liquid-bridge-based investigations, it was disclosed that in the case of transparent model liquids (high Prandtl number liquids, Pr>1) the instability is of hydrothermal nature (related to the onset and propagation of opposite azimuthal hydrothermal waves, see e.g., Wanschura et al., 1995 and Lappa et al., 2001) whereas in the case of semiconductor melts (Pr $\ll$ 1) it is of hydrodynamic nature (i.e. it is strictly related to an instability of the shear flow below the free surface, see, e.g., Levenstam and Amberg, 1995). More recently, however, it has been proven that the "full-zone" (a pair of coaxial, solid cylindrical disks with a column of liquid material suspended between them and laterally heated by an axisymmetrical energy source), even though a static configuration as the half-zone, seems to be a more relevant model for the investigation of many crucial aspects of the FZ process (see Lappa, 2003, 2004a, 2004b, 2005a, Gelfgat et al., 2005).

3 Control of Marangoni convection

As emphasized in the introduction, during crystal growth from the melt, the flow regime in the liquid is the dominating factor for the heat and mass transport during the growth process and finally for the quality and yield of the crystal.

For this reason the possibility to somewhat control the intensity of these flows as well as their bifurcations has become a topic of great interest over the years.

The three-dimensional (3D) flow instabilities discussed before can appear even if the temperature gradient along the free surface is very small (the critical Marangoni number for the first flow bifurcation in the case of semiconductor melts and liquid metals $Ma_{c1}$is of O(10) and, in practice, this corresponds to $\Delta T=O(1)$ [K] for a typical length L=O(1) [cm]). Along these lines, control of the flow is usually regarded by the investigators as an essential and growing part of the problem and accordingly different approaches with increasing complexity have been proposed during the last decade (many of which are still in an embryonic condition).

3.1 Suppression of hydrothermal waves

Since, as mentioned before, in the case of high Prandtl number liquids the instability in liquid bridges is hydrothermal and the related mechanism involves a communication between free-surface temperature perturbations and bulk-liquid temperature (see, e.g., Zeng et al., 2004), it was speculated (e.g., Benz et al., 1998) that by eliminating the free-surface temperature oscillations caused by hydrothermal waves this coupling could be broken and the oscillations would cease. Accordingly, some experiments have been carried out over recent years.

In all these experiments, the input to the control law was usually a measurement of the local surface temperature, and the corresponding output was "heat flux" added (subtracted) by heaters (coolers) at the surface.

In the pioneering experiments of Benz et al. (1998) this strategy was tested for the case of shallow layers of transparent liquids (Pr»1): free-surface temperature oscillations of a hydrothermal wave state were sensed at two locations using an infrared camera, allowing the phase speed of each wave to be determined. At a location further downstream (in the sense of the wave-propagation direction), a CO2 laser used the actual temperature signature of each hydrothermal wave to supply heat to troughs of disturbance temperature. When done successfully, hydrothermal waves disappeared downstream of the periodic-heating location.

Experiments by Petrov et al. (1996, 1998) demonstrated suppression of oscillatory convection in liquid bridges by means of a similar technique (the sensing of surface-temperature variations and the application of surface heating using externally placed elements).

Ideas from non-linear dynamics were applied to effect control, with the control law constructed from observations of the bridge response to randomly applied perturbations. Attempts to effect control of a helical traveling-wave state using a single sensor/heater pair were unsuccessful: It changed the mode into a standing wave with a node at the sensor location. This was resolved with the addition of a second sensor/heater pair allowing the algorithm to distinguish between clockwise- and counterclockwise-propagating waves.

Within this context it is also worth mentioning the landmark studies on " active feedback control " of Shiomi et al. (2001, 2002, 2003 and 2005), Amberg and Shiomi (2005).

The active control was realized by locally modifying the surface temperature of a half-zone of a transparent liquid using the local temperature measured at different locations fed back through a simple control law. The performance of the control process was quantified by analyzing local temperature signals, and the flow structure was simultaneously identified by flow visualization. With optimal placement of sensors and heaters, and proportional control, these researchers were able to raise the critical Marangoni number by more than 40%. The amplitude of the oscillation was suppressed to less than 30% of the initial value for a wide range of Marangoni number, up to 90% of the critical value.

In practice, it was proven that, with a proper choice of actuators, it is possible to modify the properties of the three-dimensional flow with linear and weakly nonlinear control. Simple "cancellation" schemes were constructed with only a few controllers by strategically placing sensor/actuator pairs (controllers).

These methods can be regarded as an effective means to control the flow stability in the case of liquid layers, shallow annular configurations (e.g., the Czochralski Method) for both cases Pr<1 and Pr>1 and liquid bridges of organic liquids (Pr>1). In all these cases, in fact, it is known (e.g., Lappa, 2005b) that waves of a hydrothermal nature can arise as the most dangerous disturbances.

The possible application of these procedures to the real FZ technique for low Prandtl number fluids, however, has still to be proven. In this case, in fact, the instability, being hydrodynamic, is not driven by surface azimuthal temperature gradients and/or waves (Lappa, 2005b).

3.2 Magnetic fields

Since molten semiconductors are excellent electrical conductors, magnetic fields are widely used as a reliable and useful flow-control strategy (Series and Hurle, 1991).

This effect was introduced for the first time in material science to suppress temperature fluctuations in the melt during the horizontal Bridgman growth process and is currently used by commercial crystal growers around the world to grow more homogeneous Si, GaAs and InP crystals with various techniques, e.g., Bridgman (horizontal or vertical) and Czochralski methods.

Its action can lead to the braking of the flow (i.e. the reduction of the rate of convective transport) or to the damping of possible oscillatory convective instabilities, the first effect being important with respect to the macroscopic homogeneity, the latter with respect to the formation of striations.

3.2.1 Physical principles and governing equations

The motion of the electrically conducting melt under a magnetic field induces electric currents. Lorentz forces, resulting from the interaction between the electric currents and the magnetic field, affect the flow. The complete theoretical description is rather complex; it is comprehensively treated in the monograph of Chandrasekhar (1981) on magnetohydrodynamics. Different possible degrees of approximation for the magnetohydrodynamic equations have been discussed by Baumgartl and Muller (1992).

In the following the "physics" of the problem and the introduction of the corresponding model equations are considered for the simple and representative case of constant (static and uniform) magnetic fields.

As outlined before, a uniform magnetic field Bo (magnetic flux density) generates a damping Lorentz force through the electric currents induced by the motion across the magnetic field. Like the case of other body forces (e.g. the buoyancy forces), this force can be added to the melt momentum balance equation.

Scaling the magnetic flux density with Bo and the electric current density with $\sigma_e V_{ref}$Bo, this equation including the Lorentz force, can be written in nondimensional form ($\underline{V}$ is the velocity, p the pressure and the reference quantities are based on the thermal diffusion velocity $V_{ref} = \alpha/L$) and in the absence of phase transitions as:

$$\frac{\partial \underline{V}}{\partial t} = -\underline{\nabla} p - \underline{\nabla} \cdot [\underline{VV}] + Pr\, \nabla^2 \underline{V} +$$

$$Pr\, Ra(T - T_{ref})\underline{i}_g + P_r H_a^2 (\underline{J} \wedge \underline{i}_{Bo}) \quad (1)$$

where $Ra = g\beta_T \Delta T L^3/\nu\alpha$ (T is the non-dimensional temperature and β_T is the thermal expansion coefficient) and Ha is the Hartmann number

$$Ha = BoL \left(\frac{\sigma_e}{\rho\nu}\right)^{1/2} \quad (2)$$

In these relationships, σ_e is the electrical conductivity and $\underline{i}_{Bo}$ the unit vector in the direction of $\underline{B}$o.

In practice, in the derivation of eq.(1) the magnetic flux density vector $\underline{B}$, that can be generally expressed as $\underline{B}=\underline{B}o+\underline{b}$ (where $\underline{B}o=\mu_o\underline{H}$, H being the imposed magnetic field strength, μ_o the permeability of vacuum and $\underline{b}$ the melt's magnetic response), is set equal to $\underline{B}$o (the applied magnetic field) since the induced field $\underline{b}$ is small for $Re_m \ll 1$, where Re_m is the magnetic Reynolds number:

$$Re_m = \mu_p \sigma_e V_{ref} L \quad (3a)$$

(μ_p is the melt's magnetic permeability); this parameter represents the characteristic ratio of the induced to imposed fields; it is known (see, e.g., Baumgartl et al.,

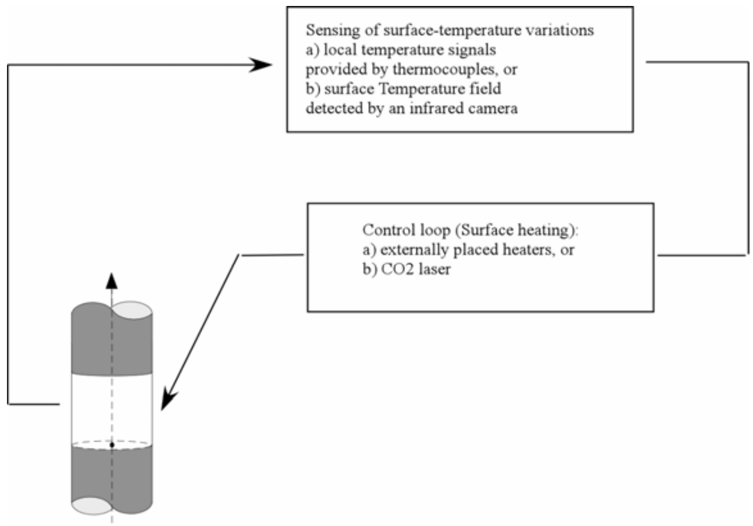

Figure 2 : Conceptual sketch of the control loop for the suppression of hydrothermal waves.

1990) that, in practice, Re_m can be regarded as a measure of the "bending" of field lines of the original applied undisturbed magnetic field $\underline{B}o$ by the fluid flow with velocity $\underline{V}$ and that:

$$|\underline{B}| = |\underline{B}o| + O(Re_m) \tag{3b}$$

thus, the deviation of the disturbed field $\underline{B}$ from the original applied field $\underline{B}o$, as mentioned above, can be estimated to be very small since in the case of semiconductor melts and related processing Re_m=O(10^{-3}).

Additional insights into this approximation can be provided by the following arguments: the magnetic diffusion coefficient $(\mu_p \sigma_e)^{-1}$ is about 4 orders of magnitude greater than the thermal diffusivity α which in turn is 10^2 times higher than the kinematic viscosity ν (values given for liquid silicon); so on a time-scale defined by α or ν which gives a natural time-scale for transport processes in the fluid (if the flow velocity is not too high) the magnetic field $\underline{B}$ retains always its steady-state value.

Accordingly, the nondimensional electric current density $\underline{J}$ is given by Ohm's law for a moving fluid:

$$\underline{J} = \underline{E} + \underline{V} \wedge \underline{i}_{Bo} \tag{4}$$

where $\underline{E}$ is the electric field normalized by V_{ref}·Bo. Since, as mentioned before, the unsteady induced field $\underline{b}$ is negligible, in particular, the electric field can be written as the gradient of an electric potential:

$$\underline{E} = -\underline{\nabla}\Phi_e \tag{5}$$

The conservation of the electric current density gives:

$$\underline{\nabla} \cdot \underline{J} = 0 \tag{6}$$

which combined with eq. (4) gives a Poisson equation for the electric potential:

$$\nabla^2 \Phi_e = \underline{\nabla} \cdot (\underline{V} \wedge \underline{i}_{Bo}) = \underline{i}_{Bo} \cdot \underline{\nabla} \wedge \underline{V} \tag{7}$$

Finally, the momentum equation can be rewritten as:

$$\frac{\partial \underline{V}}{\partial t} = -\underline{\nabla}p - \underline{\nabla} \cdot [\underline{VV}] + Pr\, \nabla^2 \underline{V} + Pr\, Ra_r (T - T_{ref}) \underline{i}_g + P_r H_a^2 (-\underline{\nabla}\Phi_e \wedge \underline{i}_{Bo} + \underline{V} \wedge \underline{i}_{Bo} \wedge \underline{i}_{Bo}) \tag{8}$$

3.2.2 *Historical developments and recent contributions*

A number of theoretical studies have appeared during recent years concerning the effect of static uniform magnetic fields on the "basic" flow motion in several geometrical models of widespread semiconductor growth techniques (the term "basic" is used here to indicate the various types of steady or unsteady, two-dimensional or three-dimensional buoyant, Marangoni or "mixed" convection without the presence of magnetic fields that can occur in these configurations, see Lappa (2005b,c).

For instance, the effect of a constant magnetic field on the electrically conducting liquid-metal flows in heated cavities has been mainly studied for the lateral heating corresponding to horizontal Bridgman crystal growth configurations. Ben Hadid et al. (1997) and Ben Hadid and

Henry (1997) investigated numerically the case of parallelepipedic or cylindrical cavities, respectively. Different magnetic field orientations were considered, as well as different situations in the parallelepipedic case, namely buoyancy driven convection in a closed cavity or in an open cavity with a stress-free surface at the top boundary, and Marangoni convection in cavities with the upper boundary subjected to surface-tension variation. In the case of shallow layers, the magnetic damping was found to be more effective with a vertical field.

Similar results were obtained by Gelfgat and Bar-Yoseph (2001); the influence of a uniform magnetic field with different magnitudes and orientations on the stability of the two distinct two-dimensional (2D) possible flow patterns in a rectangular container with aspect ratio A=length/height=4 (velocity field with a single vortex or two vortices) was investigated (see Lappa, 2005b for additional discussion about the possible existence of multiple states of steady buoyancy convection in laterally heated containers). It was shown that a vertical magnetic field provides the strongest stabilization, and also that multiplicity of steady states is suppressed by the electromagnetic effect, so that at a certain field level only the single-cell flow remains stable.

The effect of a vertical magnetic field in the case of a horizontal differentially-heated cylinder was considered by Davoust et al. (1999).

Similar studies concerning the effect of static magnetic fields in laterally heated cylindrical vessels introduced as models of the vertical Bridgman technique were carried out by Baumgartl et al. (1990) and more recently by Gelfgat et al. (2001) in the case of a axial direction of the fields. In particular, in Baumgartl et al. (1990) the use of magnetic fields was proposed as a possible alternative to the expensive microgravity and a critical comparison of the disadvantages and advantages provided by these two different approaches were discussed in a quite exhaustive way.

Owing to the experimental evidence that neither magnetic fields nor microgravity alone have produced perfect ideal crystal, Ma and Walker (1996, 1997) also considered the use of magnetic fields as a possible means to damp the typical disturbances of the microgravity environment (g-jitters and spikes of residual acceleration) in laterally heated cylinders.

The corresponding Rayleigh-Bènard problem for the case of cylinders heated from below on the ground has been considered recently by Touihri et al. (1999) for axial and horizontal magnetic fields and by Grants and Gerbeth (2004) in the case of both rotating and static fields.

Three-dimensional natural convection in parallelepipedic closed cavities driven by vertical temperature gradients and undergoing stationary magnetic fields of arbitrary direction was investigated by Möβner and Muller (1999).

For the influence of vertical magnetic fields on convection arising in a fluid layer heated from below with a free upper surface (the canonical Marangoni-Bènard problem), the reader may consider the book of Chandrasekhar (1981).

Concerning the pure Marangoni flow, Priede et al. (1995) were the first to elucidate the effect exerted by a vertical magnetic field on the hydrothermal wave instability of thermocapillary driven shear flow in a horizontal three-dimensional planar layer of a liquid metal. The linear stability analysis was limited to the disturbances traveling crosswise the basic flow and it was shown that the critical Reynolds number increases with the square of the strength of the applied magnetic field, while the wavelength of the most unstable mode is inversely proportional to the field strength.

Some interesting numerical results for similar effects on buoyancy convection and Marangoni flow in Czochralski configurations/models have been reported in Khine and Walker (1994) and Khine and Walker (1995), respectively; the combined effect of buoyancy forces, Marangoni forces and static magnetic fields has been analyzed by Jing et al. (2000).

3.2.3 Extension to the FZ technique

The use of magnetic fields has been extended over the years to the FZ technique (see, e.g., Kimura et al., 1983). Baumgartl et al. (1990) and Prange et al. (1999), demonstrated by numerical investigation that the application of axial static magnetic fields can lead to a significant increase of the critical Marangoni number for both full- and half-zones.

In agreement with their findings, Figs. 3 show for the typical reference case (a cylindrical full-zone with aspect ratio A_F=length/diameter=1, Length = 1[cm]) that, for a fixed value of the input power Q supplied to the heating source (see Lappa (2003) for additional details about the surface heat flux distribution used for the simulations),

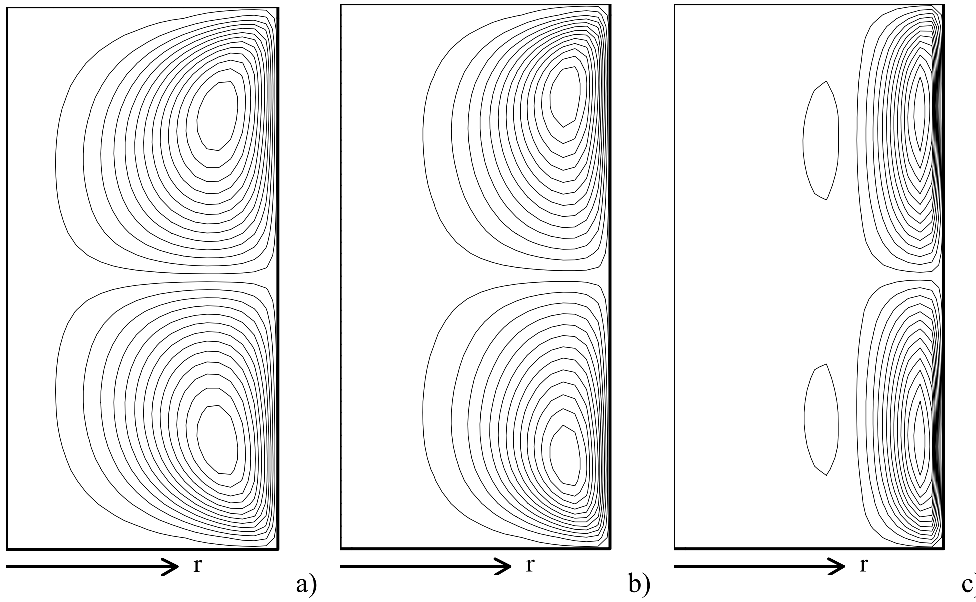

Figure 3 : Streamlines of axisymmetric convection in a silicon floating zone (Pr=0.01, A_F=1, Q=3.3[W], half meridian plane is shown, r being the radial coordinate): (a) Pure Marangoni convection (Ma≅9), (b) Ha=10, (c) Ha=50.

Table 1 : Critical azimuthal wave number and critical Marangoni number versus the Hartmann number (Pr=0.01, full-zone, A_F=1, microgravity conditions, surface heat flux distribution given in Lappa, 2003)

Ha	m	Ma_{c1}
0	2	12.41
10	2	25.42

axial constant magnetic fields lead to a concentration of the Marangoni flow close to the free surface (see also the theoretical analysis of Chen and Roux, 1991) as well as to the damping of the flow velocity (see Fig. 4).

Owing to this effect, the diameter of the toroidal convection rolls (located above and below the equatorial plane) is increased and correspondingly their effective aspect ratio is decreased. As a consequence of this behavior, axial magnetic fields tend to favor modes with larger wave numbers (Prange et al., 1999). Table 1 and Fig. 5 show that Ma_{c1} is doubled for Ha=10.

Fig. 6 shows the axial velocity distribution in a cross section.

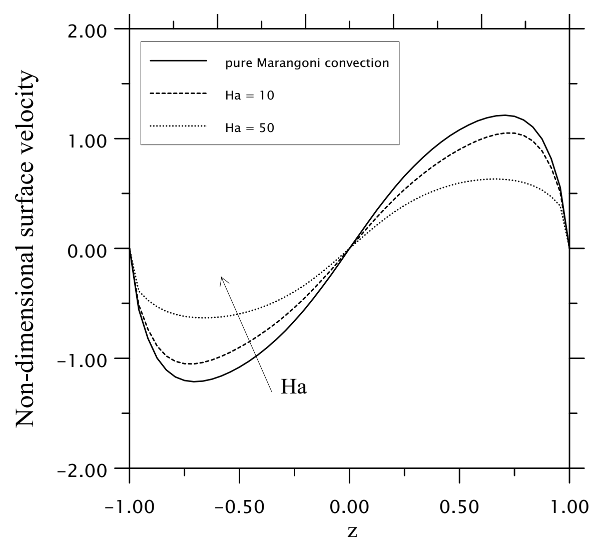

Figure 4 : Surface-velocity distribution (Pr=0.01, A_F=1) versus the Hartmann number (axial static and uniform magnetic field), for the same conditions considered in Fig. 3.

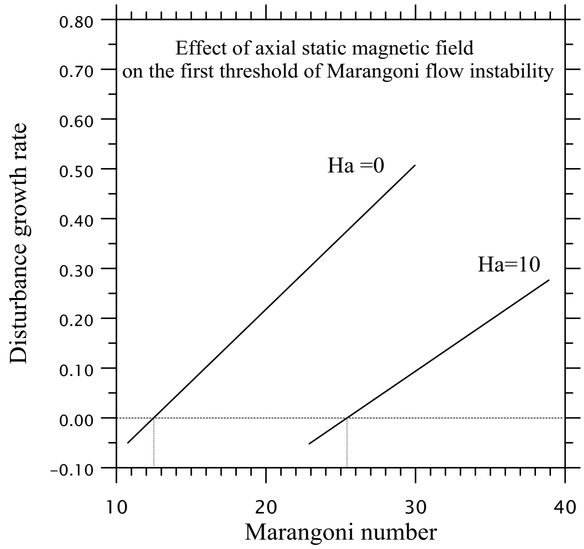

Figure 5 : Disturbance growth rate versus the Marangoni number for two different values of the Hartmann number (Pr=0.01, A_F=1).

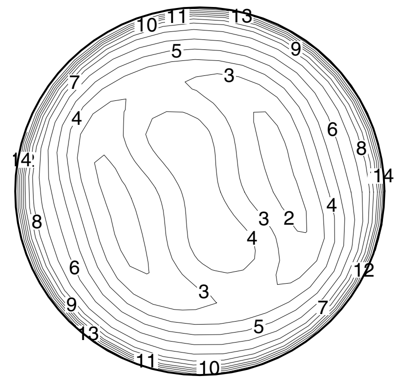

Figure 6 : Non-dimensional axial velocity component distribution in a cross section (Pr=0.01, A_F=1, Ha=10, Ma≅40, level 1→-0.95, level 15→2.83).

It is worthwhile to stress that possible stabilization is not restricted to the use of static fields. It has been shown, in fact, by means of experimental investigation that it is possible to reduce or suppress Marangoni convection during the FZ process, and thus the formation of dopant striations, through the use of either static (Dold et al., 1998; Cröll et al., 1998) or dynamic (Dold et al., 2001) magnetic fields. The transition from a time-dependent to a laminar flow regime in these cases is usually coupled with a resymmetrization of the flow geometry.

Over recent years, the first choice has nearly always been the application of static magnetic fields to suppress time-dependent flows. The possible application of transversal static fields has been also considered; for instance, the effects of both axial and transversal magnetic fields were investigated numerically through a three-dimensional selfconsistent model in a recent study by Lan and Yeh (2004). They illustrated that a transversal field is more efficient in suppressing the unsteady Marangoni flow. The required magnetic strength for getting a steady flow by means of the transversal field is lower than that required by the use of the axial one. Static transversal magnetic fields, however, are featured by some disadvantages: the flow suppression/damping tends to be effective only in planes parallel to the magnetic field; the flow in planes perpendicular to the direction of the magnetic field is not suppressed due to the induced electric potential (and this leads to a highly asymmetric molten zone and growth interface).

Transverse rotating magnetic fields, follow a different approach: it is not intended to damp the fluid motion, but to dominate the irregular flow structure by overlaying it with fast, azimuthal, axisymmetric flows. For this latter case relevant theoretical background is reported in the analyses of Fischer et al. (1999) and Walker et al. (2003), who approached the problem in terms of time integration of the full three-dimensional governing equations and linear stability analysis, respectively.

A number of other strategies exist to control melt flow; for example, by rotating or accelerating the crystal/crucible (see, e.g., Chun and Wuest (1982) and Roux et al., 1991). It is known that the centrifugal force due to the azimuthal velocity can significantly alter thermocapillary convection. For the FZ process, however, the angular velocity is limited by the fact that the associated centrifugal force can overwhelm the surface tension, breaking the liquid column.

The possible combination of static or rotating axial magnetic fields and of rotation of the solid boundaries (supporting the liquid column) in the opposite direction was considered by Lan and Yeh (2005) and Ma et al. (2004), respectively; this combination, however, was found to be not beneficial in the case of a rotating field.

3.3 "Ambient" effects

A different novel possible control strategy based on a chemical approach to the problem rather than on hydrodynamic bases, has been recently introduced by Azami et al. (2001) and Hibiya et al. (2003). They tried to control the features of Marangoni convection by changing the oxygen partial pressure in the ambient gas mixture surrounding the floating column of silicon melt. In practice, these authors disclosed by experimental investigation that both surface tension and its derivative with respect to temperature exhibit a marked dependence on the ambient oxygen partial pressure ($\partial\sigma/\partial T$ decreases with increasing oxygen adsorption at the surface of the molten silicon); accordingly, both the magnitude of flow velocity and the onset of 3D instability can be somewhat controlled by regulation of this parameter.

A non-contaminating method based on the possible influence of the external "conditions" was also proposed by Dressler and Sivakumaran (1988). They showed that a vertical jet of air blown tangentially over the free surface of a silicone oil liquid bridge, can be used to produce a viscous shear drag opposing the Marangoni (surface-tension) shear at the surface. An average reduction of 66% in Marangoni velocities was obtained during their experiments. This principle was somewhat confirmed by the experiments of Velten et al. (1991) who revealed that the air motion around the liquid column, mainly caused by buoyancy due to the heating and cooling arrangement of the experiment, has a strong effect on the onset of oscillatory flow.

The effective usefulness of this approach for the case of semiconductor melts needs still to be demonstrated. Moreover, its applicability to a real floating zone is questionable as the air jet blown tangentially to damp the intensity of one of the two counter-rotating toroidal rolls (shown, e.g., in Fig. 3a), would lead to strengthen the other one.

3.4 Forced high-frequency vibrations

Another method, more recently suggested, is to use axial vibrations. In some cases Marangoni and thermovibrational mechanisms produce flow motions of opposite direction, so that their competition is expected to be useful for controlling the intensity of the flow. Moreover, vibrational fields can change the Marangoni flow structure and are capable of exerting a strong influence on the flow stability.

3.4.1 Past history and current status

In reality, this possible strategy was already suggested by many authors as a possible means for the dynamic control of flow instabilities of gravitational origin. The stability of terrestrial fluid systems undergoing time-periodic forcing was initially considered by Davis (1976) and Ostrach (1982). Most of the subsequent analyses focused on Rayleigh-Bènard convection subjected to gravity modulation or boundary temperature modulation. The case of sinusoidal gravity modulation was studied by Gresho and Sani (1970) and the more general situation was treated extensively in the book on convective instability by Gershuni and Zhukhovitskii (1976). More recently there have been a number of studies employing both linear and non-linear theories to investigate the effect of modulation on the onset and stability of gravitational convection during Bridgman growth, using numerical or a combination of numerical and asymptotic solution techniques (Wadih and Roux, 1988; Biringen and Danabasoglu, 1990; Wheeler et al., 1991; Naumann, 2000; Fedoseyev and Alexander, 2000).

This approach was considered for the control of Marangoni convection for the first time by Birikh et al., (1993). The analysis of the joint action of vibrational and thermal Marangoni convection mechanisms demonstrated the possibilities for providing effective control of flow in an infinite fluid layer (it was found that longitudinal vibrations stabilize the flow by deforming the velocity profiles). A theoretical approach to model similar effects in a liquid column supported by two end rods with one rod vibrating, was proposed by Lee et al., (1996) and Lee (1998). It was based on a one-dimensional nonlinear model of a viscous liquid. The effect on the hydrothermal instability in liquid bridges was demonstrated by Tang et al. (1996) in the case of silicone oils. The solidification process and the resulting microstructure during float zone processing of sodium nitrate ($NaNO_3$) with and without vibrations were considered by Anilkumar et al. (1993) and Shen et al. (1996). The possible utilization of such a strategy for the FZ problem with liquid metals has been suggested by Gershuni et al. (1994), Lyubimova et al. (1994), and Lyubimova et al. (2003). Like the case of magnetic fields, vibrations allow a contactless control of the melt flow. It is a rather new and a yet less investigated technique that can be used more universally because it is not restricted to the electrically conductive melts as is the case for magnetic fields or to high Prandtl number liquids as is the case for active temperature control.

3.4.2 The thermovibrational theory

Disturbances induced in a fluid domain by a forced sinusoidal displacement

$$\underline{s}(t) = \text{b} \sin(\omega t)\underline{\hat{n}} \quad (9)$$

where b is the amplitude and ω=2πf (f is the frequency) induce an acceleration:

$$\underline{g}(t) = \underline{g}_{\omega} \sin(\omega t) \quad (10)$$

where $\underline{g}_{\omega} = \text{ b } \omega^2\underline{\hat{n}}$.

Using the Boussinesq approximation, the related body force can be added to the momentum equation, that in nondimensional form reads

$$\frac{\partial \underline{V}}{\partial t} = -\underline{\nabla} p - \underline{\nabla} \cdot [\underline{VV}] + Pr\, \nabla^2 \underline{V} + Pr \cdot \frac{b\omega^2 \beta_T \Delta T L^3}{\nu\alpha} T \sin(\frac{L^2\omega}{\alpha} t)\underline{\hat{n}} \quad (11)$$

The above equation has been the subject of intensive research in the last decade (many scaling and order-of-magnitude analyses). Many theoretical and numerical studies have been devoted to this topic (see, e.g., Monti et al., 1987; Schneider and Straub, 1989; Alexander, 1990; Lizèe and Alexander, 1997; Savino and Lappa, 2003).

It is well known that relevant nondimensional parameters for this problem are the nondimensional frequency (Ω) and displacement (Λ):

$$\Omega = \frac{\omega L^2}{\alpha} \quad (12a)$$

$$\Lambda = b\frac{\beta_T \Delta T}{L} \quad (12b)$$

The nondimensional momentum equation, in fact, can be re-written as:

$$\frac{\partial \underline{V}}{\partial t} = -\underline{\nabla} p - \underline{\nabla} \cdot [\underline{VV}] + P_r \nabla^2 \underline{V} + \Lambda\Omega^2 T \sin(\Omega t)\underline{\hat{n}} \quad (13)$$

Numerical solution of eq. (13) with the additional one for energy disclosed that when soliciting the fluid by periodic accelerations, the velocity field $\underline{V}$ is made up by an average value $\overline{\underline{V}}$ plus a periodic oscillation of amplitude $\underline{V}'$ ($\underline{V} = \overline{\underline{V}} + \underline{V}'$) at the forced vibration frequency f or at frequencies that are multiple of f.

As a result of such a convective field, the scalar quantities (temperature and/or species concentration) are also distorted.

These distortions in turn are also made up by a steady plus an oscillatory contribution (T=$\overline{T}$+T').

It is well known (see, e.g., Savino and Lappa, 2003) that the amplitude of the periodic temperature disturbances tends quickly to decrease with frequency; conversely the average disturbances are less dependent on the frequency so that one expects the steady disturbances to prevail over the unsteady ones at high frequencies (and vice versa).

This suggested the investigators to introduce strong simplifications in the analysis of the disturbances computation for the case of high-frequency vibrations (the so-called Gershuni formulation or thermovibrational theory). According to this formulation (Gershuni et al., 1982; Gershuni and Zhukhovitskii, 1986) the time-averaged distortions can be simply computed (i.e. with much less computation time) by a simplified set of equations in terms of quantities averaged over the oscillation period.

Under the assumptions of small amplitudes ($\Lambda \ll 1$) and large frequencies of the oscillatory accelerations ($\Omega \gg 1$), Gershuni and Lyubimov (1998) showed that, for a given Prandtl number, the steady (streaming) convection depends only on one relevant dimensionless parameter, the vibrational Rayleigh number:

$$Ra_V = \frac{(b\omega\beta_T \Delta T L)^2}{2\nu\alpha} = \frac{(\beta_T \Delta T L)^2}{2\nu\alpha}\left(\frac{g}{\omega}\right)^2 = \frac{\Omega^2\Lambda^2}{2\,\text{Pr}} \quad (14)$$

this formulation leads to a closed set of equations for the time-averaged quantities. The time-averaged continuity, and energy equations remain unchanged; the time-averaged momentum equation must be re-written as:

$$\frac{\partial \overline{\underline{V}}}{\partial t} = -\underline{\nabla}\overline{p} - \underline{\nabla} \cdot \left[\overline{\underline{VV}}\right] + P_r \nabla^2 \overline{\underline{V}} + \text{Pr} \cdot Ra_V \left[(\underline{w} \cdot \underline{\nabla}\overline{T})\underline{\hat{n}} - \underline{w} \cdot \underline{\nabla w}\right] \quad (15)$$

where $\underline{w}$ is an auxiliary potential function satisfying the equations:

$$\underline{\nabla} \cdot \underline{w} = 0 \quad (16)$$

$$\underline{\nabla} \wedge \underline{w} = \underline{\nabla T} \wedge \underline{\hat{n}} \rightarrow \nabla^2 \underline{w} = -\underline{\nabla} \wedge \left(\underline{\nabla T} \wedge \underline{\hat{n}} \right) \quad (17)$$

The effect of high-frequency vibrations, like that of a steady acceleration, strongly depends on the direction of the vibration $\underline{\hat{n}}$ relative to the temperature gradient. In particular, vibrations parallel to the temperature gradient tend to maintain initial diffusive conditions. In fact, as pointed out by Birikh at al. (1993), the mean vibration force is a bulk driving action induced by temperature gradients normal to the vibration axis (i.e. thermovibrational convection arises when the isotherms are not perpendicular to the vibration axis). A local feature of this force is that, if temperature distortions, with respect to the purely diffusive case, are induced by another type of convection, average vibrational flows arise in such a way as to permit the isotherms to turn and again become perpendicular to the vibration direction.

The above considerations suggest that undesired Marangoni convection can be weakened by appropriate orientation of the liquid zone with the imposed temperature gradient along the imposed forced vibrations.

3.4.3 Results

Like the case of magnetic fields treated in section 3.2.3, a full-zone of silicon with A_F=1 (zero-g) heated by an equatorial ring heater is considered as a reference case (Ma=O(10)). The entire system is supposed to oscillate in the axial direction with displacement amplitude b and angular frequency ω.

The time-averaged formulation (the Gershuni formulation already illustrated before) is used for the treatment of thermovibrational convection since it considerably simplifies the initial problem in the case of small amplitude and high frequency of the periodic disturbances and can be applied with considerable saving of computing time.

Figures 7a and 8a show the classical Marangoni vortex cells generated in the upper and lower parts of the liquid zone in the case of vibrations being absent (Ra_v=0). On the surface the flow is directed from the equatorial plane towards the supporting disks. For the considered conditions convection is axisymmetric since the Marangoni number is Ma≅9<Ma_{c1}.

If the case of pure thermovibrational convection is considered (surface-tension effects "switched off"), Figs. 7b and 8b show that the flow has the form of two vortices with the direction of circulation opposite to that of Marangoni flow. The location of these vortices is almost coincident with that of Marangoni vortices.

These features suggest that if the vibrational Rayleigh number is tuned in such a way that the magnitude of the surface velocities is comparable to that related to the Marangoni flow, it should be possible to considerably damp Marangoni convection.

Both types of convection structures, in fact, (as shown in Figs. 7a and 7b) are incompatible in the sense that their respective transport mechanisms exclude each other.

Fig. 9 shows that the vibrational Rayleigh number providing such a condition is $Ra_v \cong 150$. The velocity field resulting from the combined action of surface tension and vibrations is shown in Figs. 7c and 8c. Figure 8c shows that two small thermocapillary rolls are still present; they are confined close to the disks. Figure 7c, however, illustrates that these cells are very weak. Other counter-rotating cells driven by thermovibrational convection are located in a zone close to the ring heater. This simple example makes it possible to conclude that the vibrational mechanism can be effectively used for the suppression of the Marangoni flow. The vibration effects reduce the intensity of the circulation cell, the local temperature deformations and the shear stresses below the free surface.

Like the case of magnetic fields, the application of axial vibrations does not alter the fact that the first instability of Marangoni flow is axisymmetry-breaking. Table 2 shows the significant increase of the critical threshold for the onset of 3D convection as Ra_v is increased (see also Fig. 10).

The axial velocity distribution in a cross section for Ma≅40 and Ra_v=100 is reported in Fig. 11.

The case of vibrations perpendicular to the free surface is not considered herein. For the sake of completeness, however, it should be pointed out that the application of such vibrations to the case of high Prandtl number liquid bridges results in a strong stabilization of the so-called standing-wave regime with hot and cold surface-temperature disturbances pulsating along the direction of the imposed vibrations (it is known that free Marangoni convection in liquid bridges can undergo a first bifurcation to a pulsating instability, i.e. the aforementioned standing-wave regime, and a second spontaneous transition to a subsequent rotating regime, also known as traveling -wave, see Lappa et al., 2001).

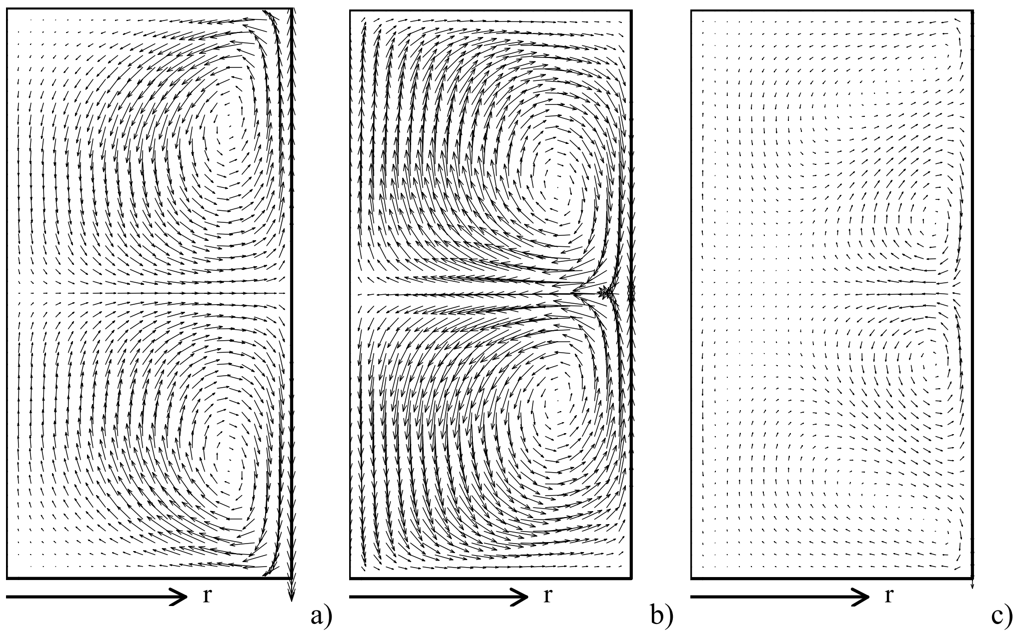

Figure 7 : Velocity field in a silicon floating zone (Pr=0.01, A_F=1), Q=3.3[W]: (a) Pure Marangoni convection (Ma≅9), (b) Pure thermovibrational convection (Ra_v=150), (c) Combined convection.

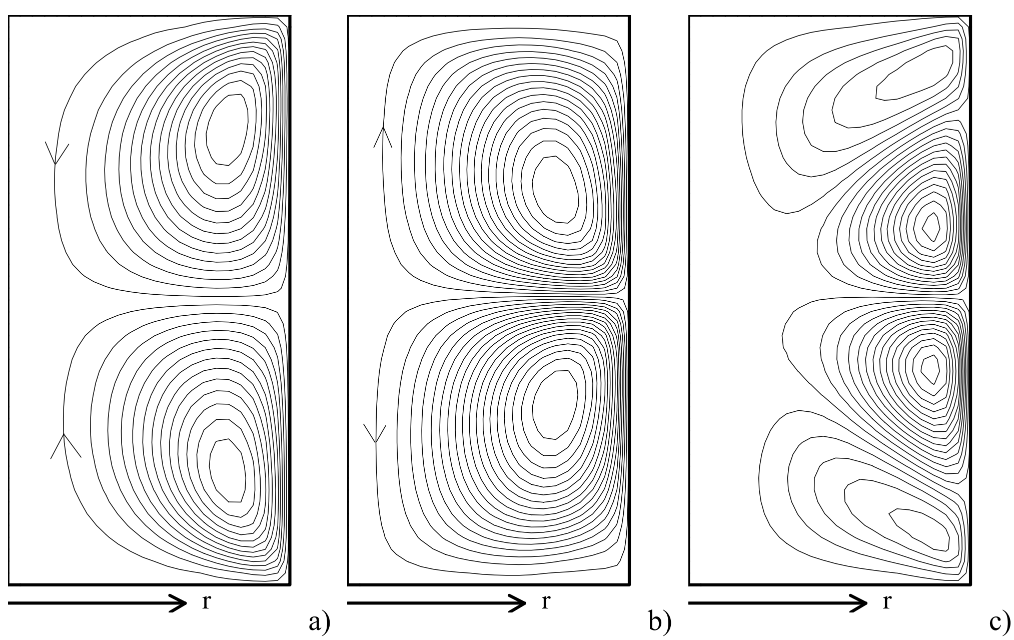

Figure 8 : Streamlines of axisymmetric convection in a silicon floating zone (Pr=0.01, A_F=1, Q=3.3[W]): (a) Pure Marangoni convection (Ma≅9), (b) Pure thermovibrational convection (Ra_v ≅150), (c) Combined convection.

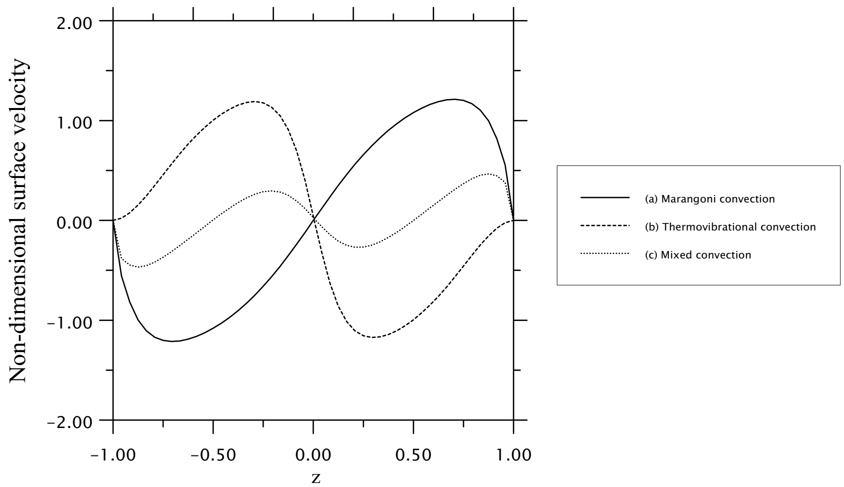

Figure 9 : Surface-velocity distribution (Pr=0.01, A_F=1), Q=3.3[W]: (a) Pure Marangoni convection (Ma≅9), (b) Pure thermovibrational convection (Ra_v=150), (c) Combined convection.

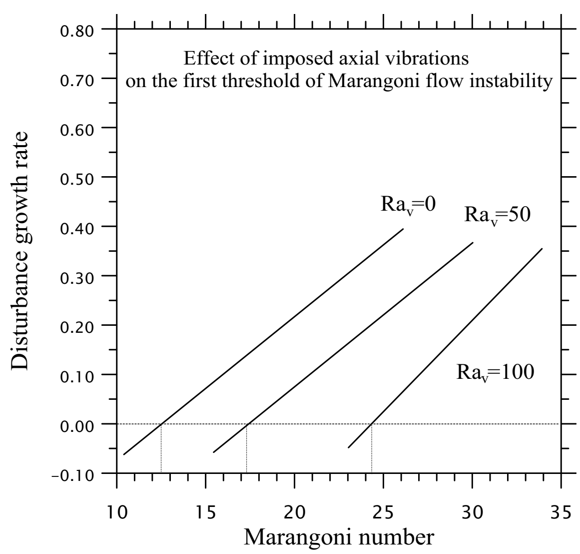

Figure 10 : Disturbance growth rate versus the Marangoni number for different values of the vibrational Rayleigh number (Pr=0.01, A_F=1).

Table 2 : Critical azimuthal wave number and critical Marangoni number versus the vibrational Rayleigh number (Pr=0.01, cylindrical full-zone, A_F=1, microgravity conditions)

Ra_v	m	Ma_{c1}
0	2	12.41
50	2	17.26
100	2	24.17

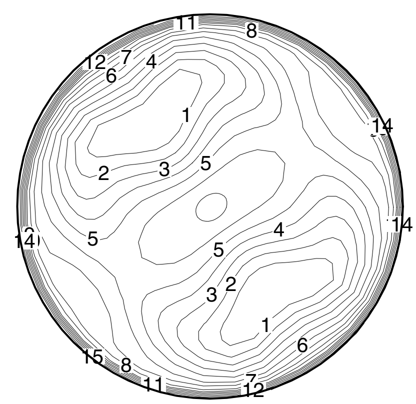

Figure 11 : Non-dimensional axial velocity component distribution in a cross section (Pr=0.01, A_F=1,Ra_v=100, Ma≅40, level 1→-0.98, level 15→2.17).

4 Conclusions

Possible strategies for the damping/stabilization of typical Marangoni flow in canonical geometrical models of the Floating Zone technique have been discussed. The discussion has been also supported by "ad hoc" numerical simulations specially used to get insights into phenomena for which experimental data are limited or still not available.

The application of "thermal feedback control", though fascinating from both theoretical and technological points of view, requires further investigation for effective application to the suppression of the hydrodynamic instabilities in real floating zones and thus for the control of the related process. Vice versa, magnetic fields, the most commonly used tool for the control/damping of Marangoni flow and related instabilities, are limited to the case $Pr \ll 1$.

The application of forced high-frequency vibrations has been illustrated as a possible and not expensive alternative to the use of the aforementioned methods, potentially useful for both cases of hydrothermal and hydrodynamic instabilities in liquid bridges and floating zones.

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