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On the formation and propagation of hydrothermal waves in liquid layers with phase change

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ABSTRACT

This paper reports on a numerical model expressly developed to inquire about the role of solidification in determining the properties of the emerging surface-tension-driven flow in typical models of oxide crystal growth. Following earlier efforts in the literature, we consider substances which have already enjoyed a widespread consideration for such a kind of studies, i.e. sodium nitrate ($NaNO_3$, Pr = 8) and succinonitrile (SCN, Pr = 23). Specific numerical examples are expressly elaborated and presented to provide inputs for an increased understanding of the main cause-and-effect relationships driving fluid flow and determining its properties. It is shown that, by interfering with the hydrothermal mechanism, namely the preferred mode of instability of Marangoni flow over a wide range of substances and conditions, solidification contributes to the chaoticity of the system by increasing the complexity of the emerging patterns and enriching the spectral content of the flow.

1. Introduction

Over recent years, the progressive refinement of manufacturing techniques in the main field of crystal growth has enabled the production of transparent materials with "controlled" microscopic structures suitable for a variety of advanced applications. Despite such improvements, however, relevant industrial production methods are still adversely affected by *undesired fluid-dynamic instabilities* that develop when the material is in a liquid state ([1–8] and references therein). Indeed, the "purity" and "perfection" of the resulting crystalline structures are in general *crucial* factors for the exploitation of such materials and the success of related applications. In turn, these factors (both) are strongly dependent on the (fluid-dynamic) phenomena that occur when the transition from an initial liquid state (melt) to the final solid phase takes place.

There is general consensus that the temperature field in the melt can give rise to gradients of density, or surface tension, or both, and that fluid motion resulting from such gradients can substantially interfere with the growth process. These thermally-induced fluid flows can indeed interact with dynamic boundary conditions such as *an advancing solidification front*. The resultant fluid motion is often complex, but always critical to crystal quality [9,10].

Examples of oxide materials of current interest are $MgAl_2O_4$ (spinel), Al_2O_3 (sapphire), $Y_3Al_5O_{12}$ (yttrium aluminium), $YAlO_3$ (yttrium orthoaluminate), $Gd_3Ga_5O_{12}$ (gadolinium gallium garnet), $LiTaO_3$ (lithium tantalate). The Prandtl number of these materials when they are in the liquid state is larger than one (Pr > 1), which can be regarded as a clear distinguishing mark with respect to standard semiconductors or liquid metals. Related applications include solid state lasers, magnetic bubble device substrates, insulating substrates for semiconductors, and monolithic crystal filters. Another category of similar substances is represented by the so-called transparent conductive oxides (TCOs). These oxides are widely used as transparent "metallic" electrodes or solar cells and flat panels including liquid crystal displays (LCDs) and organic light emitting diodes (OLEDs). Because of their unique properties of coexistence of "optical transparency in visible region" and "controllability of electronic conduction from insulator to metal" [11], such materials have enjoyed a widespread use in a variety of power saving opto-electrical circuitries and technological applications [12].

Control of convective instabilities occurring when the considered material is in a liquid state, in general, is regarded as an essential topic from a "product quality" perspective. Although a plethora of studies have been appearing over recent years motivated, completely or in part, by the need to elaborate new means to mitigate such instabilities (and hence to produce single crystals of higher quality), unfortunately, most of these studies were focusing on idealized geometries with limited translational applicability to effective production methods. A large cross section of fundamental research on these topics has been reviewed, e.g., by Lappa [13,14].

For the case of thermogravitational (buoyancy) convection, a first step in this direction was undertaken by Coriell et al. [15],

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who considered the stability of the vertical parallel flow between a crystal-melt interface and a wall held at a temperature above the melting point of the crystal for Prandtl numbers ranging from 10^{-2} to 10^{2}. Three modes of instability were identified accordingly: (a) a buoyant mode, (b) a shear mode, and (c) *a coupled crystal-melt mode*. The buoyant and shear modes were found to be similar to those that occur for flow between two vertical rigid walls held at different temperatures. For Prandtl numbers greater than approximately two, the coupled crystal-melt mode was observed at a lower Grashof number than the other two modes. Detailed results were presented for succinonitrile (SCN, $Pr = 22.8$) and lead ($Pr = 0.0225$).

For the other limiting situation in which the parallel flow is horizontal instead of vertical, Lan et al. [16] conducted a stability and bifurcation analysis with relevance to the classical (open-boat) Bridgman method. Their model accounted for two-dimensional heat transfer and fluid flow in a molten semiconductor material ($Pr << 1$), related morphological evolution of the growth interface, and heat diffusion in the crystal. The stability of various families of (flow) solutions was examined in the framework of a linear stability approach. The bifurcation diagrams were found to be slightly changed due to interface deformation (with respect to the idealized case with fixed geometry). Nevertheless, no changes were detected in the qualitative characteristics of the instability, i.e. its physical nature.

This research framework was further expanded by Bertrand et al. [17], Tönhardt and Amberg [18], El Ganaoui [19], Semma et al. [20], Stickland et al. [21] and Wang et al. [22] on the basis of different numerical methods (each with its own advantages and bottlenecks) able to address the solution of the Navier–Stokes and energy equation in their complete (non-linear and multiphase) formulation.

Some studies have been also appearing where the interaction between the solidification process and the other dominant kind of convection in typical crystal production processes, namely the so-called Marangoni flow (or thermocapillary convection), was considered. In such a context, as an example, Shyy and Chen [23] and Lan and Kou [24] were among the first investigators to carry out combined simulations for the so-called floating zone technique. The latter authors compared the typical dynamics emerging for high Prandtl-number materials, e.g., $NaNO_3$, and low Prandtl-number materials, such as silicon (Si). The unknown shapes of the melt-gas, melt-crystal and melt-feed interfaces were calculated for different situations: (a) conduction only, (b) natural convection and (c) hybrid Marangoni and buoyancy flow. Marangoni convection was found to increase the convexity of the melt-crystal interface and reduce the maximum surface temperature, this effect being more pronounced in the high-Pr melt case ($NaNO_3$) than in the low-Pr melt case (Si) (though it should be mentioned Marangoni convection was observed to be far stronger in the Si melt).

Using succinonitrile (SCN) as work fluid (a paradigmatic substances for the study of such dynamics owing to its relatively low melting temperature, reduced reactivity and transparency to visible light), Cerimele et al. [25] concentrated on the horizontal Bridgman solidification process where the Marangoni effect emerges at the open top of the crucible. Additional existing fundamental research on this topic has been reviewed by Amberg and Shiomi [26] and Tsukada et al. [27]. Along these lines, it is also worth citing the very interesting experimental study by Matsunaga and Kawamura [28], who, yet for the case of succinonitrile, focused on the influence of convection on crystal shape. Similarly, Hong et al. [29] visualized Marangoni convection in a BaB_2O_4 (77 wt%)–Na_2O (23 wt%) mixture melt.

Unfortunately, analyses of such a kind are rather sparse and rare as most of past studies examined either crystal growth or thermocapillary (Marangoni) convection as disjoint subjects (typically on the basis of one-way coupling approaches, i.e. crystals with fixed shape and fixed boundary conditions used to study the response of the fluid system to the application of well-defined stimuli in terms of imposed temperature or concentration gradients).

For the specific case of two-way coupling methods (in which fluid-dynamic and phase-change aspects are fully coupled), it is worth citing Lan and Chian [30] and Lappa and Savino [31] who performed some numerical studies for circumstances in which the Marangoni flow is in *supercritical conditions*. However, these authors limited to the case of semiconductor melts ($Pr << 1$) in floating zones. It is a well-known fact (see, e.g., [32]) that for these values of the Prandtl number, the typical instabilities of Marangoni flow display notable differences with respect to those expected for high-Pr materials such as oxide melts or SCN.

In the case of liquid layers or shallow rectangular cavities, as an example, the typical supercritical convective modes of two-dimensional parallel Marangoni flow (the so-called *return flow* solution, originally considered by Smith and Davis [33]) are represented by *hydrothermal waves* (HTW). It is known that these waves have a different direction of propagation according to the Prandtl number. They are nearly perpendicular to the basic state for low-Pr materials, i.e. the disturbance propagates almost exactly in the spanwise direction (the wave has a longitudinal wavefront in such a case); and nearly parallel to the basic flow for high-Pr materials, i.e. the disturbance propagates almost exactly in the *upstream direction* with respect to the surface flow (i.e. from the cold side to the hot side, the waveform in such a case being transverse). In the latter case, two-dimensional (2D) simulations can be used to simulate the real phenomenon with an acceptable approximation. There are several numerical efforts along these lines, see, e.g., Peltier and Biringen [34], Xu and Zebib [35], Shevtsova et al. [36], Tang and Wu [37], Lappa [38]. Simulations expressly dedicated to SCN or other convenient fluids (such as $NaNO_3$ or silicone oils used to mimic the typical properties of oxide materials) are also available (Bucchignani and Mansutti [39]). Important extensions to the original work by Smith and Davis [33] and the later weakly nonlinear analysis by Smith [40] were provided by Priede and Gerbeth [41], who analysed the effect of various thermal boundary conditions on the linear stability of the parallel-flow solution. In particular, they concentrated on the more realistic case in which the adiabatic bottom is replaced by a conducting boundary (to mimic an external metallic container) with the free liquid–gas surface retaining an adiabatic behavior. Some other interesting results were presented recently by Lappa [42], who focused on the emergence of hydrothermal waves in shallow cavities with an obstruction located on the bottom (a forward-facing step, a backward-facing step or a combination of both geometrical features).

None of these studies, however, examined the interplay of these modes of convection with solidification. Given the absence of numerical studies addressing such problems for high-Pr liquids (the molten form of transparent oxide materials), in the present work we concentrate expressly on the case of parallel Marangoni flow and related supercritical states represented by HTWs.

Following earlier efforts in the literature, we consider the typical substances which have already been widely used as “work” fluids for such a line of inquiry, that is sodium nitrate ($NaNO_3$, $Pr = 8$) and succinonitrile (SCN, $Pr = 23$). In particular, we provide (original) numerical examples specifically conceived to cover heretofore unseen dynamics. Indeed, the present work has been carried out under the idea or belief that advanced simulations may provide insights into still poorly known mechanisms and help expedite companion experimental studies (leaving aside for a while the experimental counterparts or the effective industrial applications, it is clear that conducting numerical simulations can become worth in

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