Time-dependent thermally-driven interfacial flows in multilayered fluid structures

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Abstract

A computational study of thermally-driven convection in multilayered fluid structures will be performed in order to examine the effect of interactions among deformable fluid-fluid interfaces on the structure of time-dependent flow in these systems. Multilayered fluid structures in two model configurations will be considered: the differentially heated rectangular cavity with a free surface, and the encapsulated cylindrical liquid bridge. An extension of a numerical method developed as part of our recent NASA Fluid Physics grant will be used to account for finite deformations of fluid-fluid interfaces.

1 Introduction

In growing crystals from a melt containing volatile components (such as GaAs or InP), stringent control of the stoichiometry is crucial in order to avoid crystallographic defects and degradation of electronic properties of the resulting product. Two approaches have been devised to minimize evaporation of the volatile component from the melt during processing of these materials: pressurization of the growth chamber with the volatile component's vapor, and encapsulation of the melt in an amorphous liquid glass phase. The former approach often requires precise temperature control to avoid deposition of the volatile material on surfaces of the components inside the growth chamber. Furthermore, the pressurized vapor is often reactive (e.g., arsenic) and poses a safety hazard which represents a major drawback of this approach, particularly in connection with space-based processing. The second approach wherein the melt is encapsulated in a low melting point amorphous molten glass, such as boron oxide (B_2O_3) or pyrolitic boron nitride (PBN), holds more promise for implementation in space. The viability of this technique was originally demonstrated by Metz et al. [25] who encapsulated germanium, PbTe, and PbSe single crystals with molten B_2O_3. Liquid-encapsulated crystal growth (abbreviated LEC growth) has since been used in the Bridgman and Czochralski configurations by researchers. Integration of this approach into the float zone process on earth has been impractical due to severe limitations on the maximum diameter of a stable molten zone (including the encapsulant) that can be supported by interfacial tension forces in the presence of destabilizing buoyancy forces. In addition, any electromagnetic levitation of the molten zone provided by RF heating coils would be lost upon encapsulating the floating zone since the encapsulant is typically nonconductive (e.g., B_2O_3). Johnson [19] used a liquid-encapsulated float-zone (LEFZ) technique to produce 10 mm diameter ingots of polycrystalline GaAs by pulling a GaAs rod through B_2O_3 contained in a quartz tube. He estimated the maximum attainable crystal diameter in that process to be about 15 mm as compared with commercially useful diameters of 50 mm or more. Under microgravity conditions, however, the molten zone (including the encapsulant) will be affected predominantly by interfacial tension forces, and the limitations imposed on the maximum crystal diameter by *capillary* instabilities are not as severe as those dictated by buoyancy-driven instabilities on earth. In addition, there have been a number of interesting attempts aimed at controling interfacial instabilities through encapsulation of the bridge in an annular liquid column (cf. Metz et al. [25], and Barocela and Jalilevand [1]), and the introduction of a coflow in the encapsulating layer (cf. Lowry and Steen [23]). Hence, the LEFZ technique represents a promising approach for space-based production of monocrystalline semiconductors with a volatile component.

The main distinction between the unencapsulated and LEC growth techniques is the presence of the additional liquid-liquid interface between the melt and the encapsulant in LEC growth. The addition of this new interface in a highly nonisothermal environment can be a source of new and complex dynamics that may critically affect crystal quality. It is well-known that nonuniform distributions of temperature on the interface between two fluids can cause variations in interfacial tension across the interface. Such interfacial tension gradients act like tangential stresses on the interface and drive a surface flow from regions of low interfacial tension to those of high interfacial tension. The resulting surface flow can penetrate into the bulk phases on the two sides of the interface through the action of viscosity and, in turn, induce bulk fluid motions which are called *Marangoni* or *thermocapillary* convection. In general, thermocapillary convection will be significant when buoyancy effects are negligible, as in systems involving small scales or under microgravity conditions.

Two of the most widely studied model problems in the analysis of thermocapillary convection have been the differentially-heated rectangular cavity with one free surface, and the cylindrical liquid bridge consisting of a liquid drop fixed between two coaxial circular disks (cf. review by Kuhlmann [21]). The former represents an idealization of the open-boat crystal growth technique while the latter is a model for the upper or lower half of the liquid domain in the float-zone process (cf. Ostrach [27]). For thermocapillary convection in a single fluid layer, the structure of the steady, linear and nonlinear, two-dimensional and axisymmetric flow and temperature distributions are well understood. Though significant progress has been made in understanding the nature of time-dependent flows, the role of interface deformations as well as the nonlinear dynamics of time-dependent and three-dimensional thermocapillary flows still provide challenges for future numerical investigations.

In comparison with the unencapsulated process, modeling of LEC growth has received considerably less attention. Recent numerical simulations of steady thermocapillary convection in liquid encapsulated Czochralski growth [37, 11, 34] have predicted multicellular flow patterns in the two liquid phases. There have also been a few studies of *steady* thermocapillary convection in double layers of immiscible liquids within differentially heated rectangular cavities, with the imposed temperature gradient parallel to the free surface. Villers and Platten [38] performed a one-dimensional analysis assuming a constant temperature gradient across the cavity, while others [32, 12, 7, 22] obtained numerical solutions of the two-dimensional problem for low to moderate aspect ratios. The results of these studies show that under microgravity conditions, the strength of the thermocapillary flow in the layer in contact with the solid boundary can be significantly reduced, suggesting that the liquid encapsulation method can be used as a means of suppressing thermocapillary convection in the melt. Since the encapsulant in LEC growth is typically very viscous (e.g., B_2O_3), a reduction in the strength of the thermocapillary convection in the melt by the encapsulant is consistent with the experimental results of Eyer and Leiste [10], who showed that a solid encapsulation technique can eliminate striations in silicon crystals. Most recently, Prakash and Koster [30, 31] extended the above analyses to include a third immiscible liquid layer. They performed both a one-dimensional analysis similar to that of Villers and Platten [38] and an asymptotic analysis for shallow rectangular cavities similar to the single layer analysis of Sen and Davis [36]. The results of their analysis indicate that for equal layer heights and encapsulant viscosities, the flow in the middle (encapsulated) layer is qualitatively similar to, but weaker than, that obtained by Sen and Davis for a single layer. However, when the middle layer is much thicker than the encapsulant, the interface deformations are quite different from those for the single layer. Hence, the flow pattern in the encapsulated layer strongly depends on the encapsulation thickness.

All of the above studies of thermocapillary convection in multilayered fluid structures have been limited to steady convection. Furthermore, aside from a few exceptions [37, 12, 31], previous studies have neglected deformations of the fluid-fluid interfaces in order to avoid the complexity associated with the determination of the unknown interface shapes. Recent numerical simulations of thermocapillary flows accounting for finite deformations of the free surface indicate that the shape of the interface may strongly influence the flow and temperature fields [24, 3, 4, 6, 20, 5, 2]. Some studies [28, 3, 20] have even suggested that oscillatory flows may be established as a consequence

of a coupling between interface deformation and thermocapillary convection. Another example of the importance of interface deformability in determining the dynamics of thermocapillary flows in LEC growth is the recent flight experiment of Koster [35] involving a three-layer fluid structure in a container differentially heated parallel to the free surface. The goal of the experiment was to identify the criteria for reducing thermocapillary convection in the middle liquid layer which is presumed to be representative of the encapsulated melt of an electronic material in LEC growth. Upon initial operation of the experiment, when two curtains separating the three fluid layers were drawn, the layers collapsed. Although the cause is not fully understood, the growth of capillary instabilities present at the fluid-fluid interfaces —as well as the contact-line region— can provide a plausible explanation for the failure of the experiment.

Based on the limited available results, it is reasonable to expect the presence of multiple deformable fluid-fluid interfaces in LEC growth to lead to interesting new dynamics which can play an important role in determining the quality of the resulting crystals grown under microgravity conditions. Thus, the effect of interactions among deformable fluid-fluid interfaces on the structure of time-dependent thermocapillary convection in these systems remains an open question that warrants further investigation. It is the purpose of this study to address this issue for thermocapillary convection in multilayered fluid structures in the cavity and bridge configurations. We intend to remove the limitations associated with the assumption of nondeformability of interfaces in previous studies of this problem by using an extension of a numerical method developed as part of our recent NASA Fluid Physics grant [15, 16] to allow interface shapes to evolve in accordance with the physics of the problem. The results will help extend our knowledge of the dynamics and stability of multilayered fluid systems, and will provide insight into the governing physics of the LEC growth process.

2   Formulation

In what follows, the subscript 's' denotes an interface-specific quantity, while a plain symbol or one with a caret references a property of the bulk phase on one or the other side of the interface. The interfacial tension, σ, is taken to depend on the temperature, T, as well as the surface concentration of surface-active contaminants, Γ. The temperature dependence of other fluid properties can also be easily included. Using the velocity scale $U \equiv (GL/\mu)\sigma_{,T}$, obtained through a tangential stress balance at an interface, the following well-known parameters arise in the equations governing the various transport processes in each phase:

$$Re = UL/\nu, \qquad Ma = UL/\alpha, \qquad Ca = \mu U/\sigma, \qquad Bi = hL/k, \qquad Br = (\mu U^2/kGL),$$

where G denotes the imposed temperature gradient and h is the heat-transfer coefficient at the liquid-vapor interface. In addition to the above dynamic and thermal parameters, there are also a number of geometric and physical property ratios for the various fluid phases. The vector-invariant forms of the governing equations in each fluid phase are given by

$$\mathbf{u}_{,t} + \nabla \cdot (\mathbf{uu}) = \frac{1}{Re}\nabla \cdot \mathbf{\Pi}, \qquad \nabla \cdot \mathbf{u} = 0, \qquad T_{,t} + \nabla \cdot (\mathbf{u}T) = \frac{1}{Ma}\left[\nabla^2 T + Br\tau : \nabla \mathbf{u}\right],$$

where $\mathbf{\Pi} = -p\mathbf{I} + \tau$ is the Newtonian stress tensor with $\tau = \nabla \mathbf{u} + (\nabla \mathbf{u})^\dagger$.

The interfacial boundary conditions on $\mathbf{x} = \mathbf{x}_s(t)$ include

$$\hat{\mathbf{n}} \cdot (\mathbf{\Pi} - \lambda\hat{\mathbf{\Pi}}) + \nabla_s \sigma = \sigma(\nabla_s \cdot \hat{\mathbf{n}})\hat{\mathbf{n}}, \qquad \mathbf{u} = \hat{\mathbf{u}}, \qquad d\mathbf{x}_s/dt = \mathbf{u}, \qquad T = \hat{T}, \qquad \hat{\mathbf{n}} \cdot \nabla T = \beta\hat{\mathbf{n}} \cdot \nabla\hat{T},$$

where $\hat{\mathbf{n}}$ denotes the unit normal vector at the interface. The thermal boundary condition at the liquid-vapor interface is $-\hat{\mathbf{n}} \cdot \nabla T = Bi(T - T_o)$ for some ambient temperature T_o. The solid-liquid boundary conditions are standard, except in the vicinity of the contact line, where a multi-valued

velocity gives rise to a stress singularity (cf. Dussan V. and Davis [8]). This singularity will be regularized through the introduction of a slip coefficient, a, according to (Huh and Scriven [18])

$$\hat{\mathbf{b}} \cdot \mathbf{\Pi} \cdot \hat{\mathbf{e}} = a\mathbf{u} \cdot \hat{\mathbf{e}}, \qquad (a \ll 1),$$

where $\hat{\mathbf{b}}$ and $\hat{\mathbf{t}}$ are the unit vectors normal to the solid-liquid boundary and tangent to the contact line, respectively, and $\hat{\mathbf{e}} = \hat{\mathbf{t}} \wedge \hat{\mathbf{b}}$.

In the presence of bulk–insoluble surface contamination, the concentration Γ of the surface-active impurity on a fluid-fluid interface is governed by

$$\frac{\partial \Gamma}{\partial t} + \nabla_s \cdot (\mathbf{u}_s \Gamma) + (\mathbf{u} \cdot \hat{\mathbf{n}})(\nabla_s \cdot \hat{\mathbf{n}})\Gamma = \frac{1}{Pe_s}\nabla_s^2 \Gamma,$$

where Pe_s denotes a surface Peclet number. This surface convective-diffusion equation will be converted into a volumetric one and treated as such (cf. [17]).

3 Method

As part of a previous MSAD-funded effort, a computer code for the simulation of flows driven, or affected, by interfacial forces was developed; its main features are:

* Solution of the three-dimensional, incompressible Navier-Stokes equation,
* Temperature-dependent properties,
* Automatic adaptive grid refinement near fluid-fluid interfaces,
* Determination of surface topology (e.g. $\hat{\mathbf{n}}$, curvature, etc.), and normal distance to the interface via the solution of the level-set[1] equations (cf. Osher and Sethian [26]),
* Smearing of the interfacial forces into a body force occupying a thin band about the original crisp interface, via a mollified delta function (cf. Peskin [29]) in the interface-normal direction.

This code has been well calibrated and tested, and is being parallelized in order to achieve significantly faster speeds. The adaptive local refinement of the underlying grid in the vicinity of fluid-fluid interfaces is an essential feature of the code (cf. Haj-Hariri et al. [14]), and has been achieved using an oct-tree data structure.

Briefly, the computational approach involves the treatment of interfacial forces as body forces, and smearing of all fluid properties across the interface, resulting in an effectively single-phase flow problem. For example, the momentum equation involves a body-force concentration, $\mathbf{f}$, accounting for the effect of an interface:

$$\mathbf{f} = [(\mathbf{I} - \hat{\mathbf{n}}\hat{\mathbf{n}}) \cdot \nabla \sigma - (\nabla \cdot \hat{\mathbf{n}})\sigma \hat{\mathbf{n}}]\delta(\phi).$$

In the above expression, δ denotes a mollified Dirac delta function (cf. Peskin [29]) and ϕ is the 'distance' from the interface, obtained via the level-set construction of Osher and Sethian [26]:

$$|\nabla \phi| = 1, \qquad \phi_s = 0.$$

The gaussian curvature of the interface ,$\nabla \cdot \hat{\mathbf{n}}$, is then obtained using $\hat{\mathbf{n}} = \nabla \phi$. The treatment of the surface convective-diffusion equation (arising when an interface is contaminated with surface–active impurities) is more challenging, as the physical quantity Γ is genuinely a surface quantity and its treatment as a volume quantity generates ambiguity for its behavior in the normal direction. To avert the ambiguity, it is shown that Γ should be extended away from the interface using

$$\hat{\mathbf{n}} \cdot \nabla \Gamma = 0,$$

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[1]The concept of the level sets is not new; it is akin to the constant-phase surfaces which are obtained via the solution of the Eikonal equation in monochromatic wave-propagation problems

so that the surface convective-diffusion equation is then equivalent to a volumetric one.

Another point requiring special treatment is the contact-line area. The slip-coefficient formulation is used to regularize the force singularities. However, one needs to further implement a model accounting for the contact-angle hysteresis as well as the general dependence of the advancing and receding contact angle on the in-plane normal speed of the contact line (e.g. power-law dependence $U = \kappa(\theta - \theta_{eq})^m$; cf. Ehrhard and Davis [9]). This information is incorporated into the formulation through the imposition of the interface normal on the interface at the contact line, i.e. $\hat{n}$ is determined such that $\hat{n} \cdot \hat{b} = \cos\theta$ (while $\hat{n} \cdot \hat{t} = 0$). This specification supercedes the definition $\hat{n} = \nabla\phi$, which is applied at all other points within the computational domain.

While the stability problem will be addressed via a complete time-dependent solution, a linear stability analysis in the spirit of Ramanan and Homsy [33] will also be performed in order to illuminate the energetics of the flow. Namely, any physical quantity, q, will be expanded about it's value at time t:

$$q(t + \delta t) = q(t) + \delta q,$$

and a linearized equation for δq, as well as an evolution equation for an energy-like quadratic, will be determined. The latter equation will provide further insight into the nature of the otherwise purely-numerical solution by indicating the main contributions to the production and dissipation of energy; this information will be helpful in understanding and controlling the behavior of the solution.

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