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Thermocapillary flow in double-layer fluid structures: An effective single-layer model

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Abstract

Thermocapillary flows are of considerable technological importance in materials processing applications such as crystal growth from the melt, particularly under microgravity conditions where the influence of buoyancy-driven convection is minimized. In this study, thermally driven convection within a differentially heated rectangular cavity containing two immiscible liquid layers is considered in the absence of gravity. The introduction of a more viscous encapsulant layer leads to a significant reduction in the intensity of the thermocapillary flow within the encapsulated layer. Interface deformations are small when the contact line of the interface is pinned on the solid boundaries. The higher viscosity of the encapsulant layer gives rise to a larger pressure gradient in that layer, thereby resulting in interface deformations that are qualitatively different from those observed at the free surface in the absence of the encapsulant layer. The flow pattern in the encapsulated layer and the resulting interface deformations are strongly dependent on both the thickness and the viscosity of the encapsulant layer. It is shown that the flow within the encapsulated layer may be closely approximated by simply considering the single-layer problem with a modified stress condition at the interface. The modified tangential stress balance for the effective single-layer model is derived based on asymptotic results for small-aspect-ratio double-layer systems and the insight gained from double-layer computations for finite-aspect-ratio systems. It is shown that the single-layer model accurately predicts the flow in the double-layer system even for large aspect-ratios.

Keywords: Thermocapillary flow; Marangoni stress; Interface deformation

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1. Introduction

Space-based processing of electronic materials has received much attention because a number of undesirable features of semiconductor crystals grown on earth have been attributed to the effect of gravity [1,2]. Production of III–V semiconductor crystals, such as GaAs and InP, involves additional complications which are not present in the growth of single element crystals like silicon or germanium, namely the existence of volatile components such as arsenic (in GaAs) and phosphorous (in InP). In growing crystals from a melt containing volatile components, stringent control of the stoichiometry is crucial in order to avoid crystallographic defects and degradation of electronic properties of the resulting product. One approach to minimize evaporation of the volatile component from the melt during processing of these materials has been the encapsulation of the melt in a low melting point amorphous molten glass phase, such as boron oxide or pyrolytic boron nitride. The main distinction between the unencapsulated and the liquid-encapsulated crystal (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 significantly influence crystal quality.

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In comparison with the unencapsulated process, modeling of LEC growth has received considerably less attention. One of the most widely studied model problems associated with the crystal growth process has been thermocapillary convection in a single fluid layer confined within a differentially heated rectangular cavity, representing an idealization of the open-boat crystal growth technique (cf. review by Kuhlmann [3]). These studies have been extended to the case of immiscible double liquid layers with the imposed temperature gradient parallel to the free surface [4–10]. Villers and Platten [4] performed a one-dimensional analysis assuming a constant temperature gradient across the open cavity, while others obtained solutions of the two-dimensional problem for small-aspect-ratio liquid layers. The results of these studies show that under microgravity conditions, the strength of the thermocapillary convection in the layer in contact with the solid boundary can be significantly reduced, suggesting that liquid encapsulation 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., boron oxide), 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 [11], who showed that a solid encapsulation technique can eliminate striations in silicon crystals. Although the results of these studies suggest that encapsulation may be used to reduce the strength of steady thermocapillary convection, the effect of encapsulation on the stability of this flow remains an open question.

Prakash and Koster [12,13] 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 [4] and an asymptotic analysis for shallow rectangular cavities similar to the single layer analysis of Sen and Davis [14]. The results of their analysis indicate that for equal layer heights and encapsulant viscosities, the flow in the middle (encapsulated) layer is qualitatively the same, but weaker than that obtained by Sen and Davis for a single layer. However, when the middle layer is much thicker than the encapsulant, interface deformations can be quite different from those for a single layer. Hence, for small-aspect-ratio cavities, the flow pattern in the encapsulated layer seems to be strongly dependent on the encapsulation thickness. We will show that it is a combination of the viscosity and thickness of the encapsulant layer which determines the flow pattern and interfacial deformations of the encapsulated layer. This insight allows for the effective single-layer modeling of flow within the pinned-interface multi-layer system; we will elaborate on this simplification later in this article.

Aside from the recent computations of Liu et al. [10] for a B_2O_3/GaAs two-layer system, previous studies of thermocapillary convection in multi-layered fluid structures have focused on flow in small-aspect-ratio liquid layers. Moreover, most of these studies have neglected deformations of the fluid–fluid interfaces in order to avoid the complexity associated with the application of interfacial boundary conditions on the unknown interface shapes. The flight experiment of Koster [15] underscores the need for a more detailed investigation of thermocapillary convection in multi-layered systems accounting for finite interface deformations in conjunction with a suitable dynamic contact line condition. In this paper, we perform a numerical study of thermocapillary convection in a rectangular cavity containing two horizontal immiscible fluid layers differentially heated from the side in the absence of gravity. We use domain mapping in conjunction with a finite-difference scheme on a staggered grid to solve for the temperature and flow fields while allowing the interface to deform. We use the insight gained from these computations in conjunction with the asymptotic results for low-aspect-ratio systems to develop an effective single-layer model that can be used to closely approximate the flow within the encapsulated layer for any layer aspect-ratio.

2. Problem formulation

In this section, the dimensionless equations and boundary conditions governing the momentum and energy transport processes in a differentially heated rectangular cavity in the absence of gravity are presented. The two-dimensional cavity of height H and length L contains two immiscible incompressible liquid layers denoted by D_1 and D_2, with initial aspect-ratios $\phi_1 = H_1/L$ and $\phi_2 = H_2/L$, respectively, as shown in Fig. 1.

In the following presentation, subscripts 1 and 2 are used to denote the quantities associated with phases D_1 (melt) and D_2 (encapsulant), respectively, and the subscript s is used to represent an interface-specific property. All lengths are made dimensionless with the cavity length L, velocities with a characteristic velocity U obtained through a tangential stress balance at the interface (cf. Ostrach [16]), stresses with $\mu_1 U/L$ (where μ_1 denotes the viscosity of the bottom phase), time with L/U, and temperature with

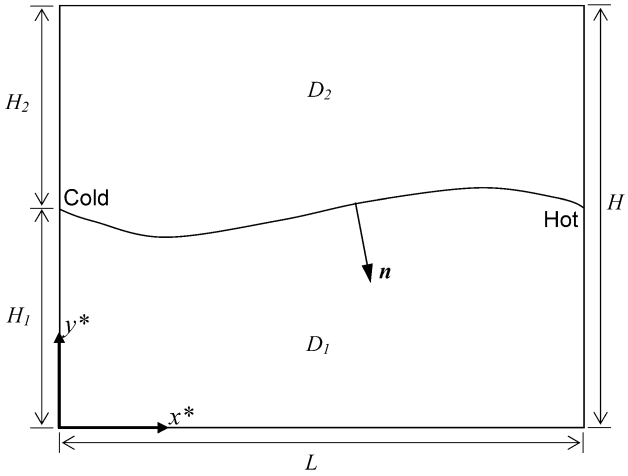

Fig. 1. Schematic of a double layer of immiscible fluids in a differentially heated rectangular cavity.