A numerical study of buoyancy-Marangoni convection of volatile binary fluids in confined geometries

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ABSTRACT

A horizontal temperature gradient can cause a flow in a layer of liquid with a free surface via several different mechanisms. The most universal one is due to thermocapillary stresses that arise due to the temperature dependence of surface tension. For binary liquids, the flow can also be driven by solutocapillary stresses that arise due to the dependence of surface tension on the composition of the liquid. For some binary liquids, such as water-alcohol mixtures, solutocapillary stresses are primarily due to phase change (e.g., differential evaporation or condensation of the two components), and these two mechanisms can counteract each other. A recent experimental study (Li and Yoda, 2016) has demonstrated that the flow direction can be reversed by changing the amount of air present inside the experimental apparatus. To understand how the presence of air affects the interfacial stresses, we have developed and implemented numerically a comprehensive two-sided transport model, which accounts for transport of heat, mass, and momentum in both phases and phase change across the interface and is able to reproduce the experimental results. The detailed analysis of these results shows that air tends to suppress phase change and hence solutocapillary stresses. Removing the air enhances phase change, instead suppressing the variation in the interfacial temperature and hence thermocapillary stresses.

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1. Introduction

Various types of convective flows can arise in layers of binary liquid during evaporation [1–5] or condensation [6–10] driven by the gradient in either vapor concentration or temperature normal to the free surface. Perhaps the most famous example is "wine tears" that form when a mixture of water and ethanol is allowed to evaporate. This mixture is an example of a positive binary fluid [11], where the more volatile component (ethanol) has lower surface tension compared with the less volatile component (water). Preferential evaporation of ethanol from a thin layer near a side wall reduces surface tension of the mixture there and generates solutocapillary forces that drive the liquid towards, and in some instances up, the wall, which is a key physical mechanism behind the formation of wine tears [12,13]. More recently it has been discovered that thermocapillary stresses generated via evaporative cooling of the liquid surface also play a role in this phenomenon [14]. In this specific case, evaporation causes the temperature near

the edge of the film to decrease, further increasing surface tension, so thermocapillary stresses *enhance* solutocapillary ones.

Under certain conditions, however, thermocapillary and solutocapillary stresses can oppose each other. This property can be usefully exploited when thermocapillarity has an adverse effect, e.g., in thermal management devices, such as heat pipes, which rely on evaporative cooling. Heat pipes are effectively sealed cavities partially filled with a volatile liquid, and it is the temperature gradient tangential to the free surface that drives the system out of equilibrium and generates the flow. For pure fluids, thermocapillary stresses drive the flow away from the hot end of the heat pipe, which can cause dry-out leading to a complete loss of evaporative cooling and a dramatic increase in the temperature of the hot end. The adverse effect of thermocapillarity can be ameliorated by using a positive binary coolant [15], where the differential evaporation of the two components causes solutocapillary stresses towards, rather than away from, the hot end. Indeed, experimental studies have shown that the direction of the flow can be reversed by using a mixture of water with ethanol [16] or methanol [17]. Beneficial effects of using a binary mixture on the performance of a heat pipe in microgravity have also been demonstrated [18].

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There is a vast literature on convection in binary fluids driven by a vertical [19–22] temperature gradient, but almost all of it is devoted to nonvolatile liquids, where solutocapillary stresses arise due to the Soret effect [23] rather than differential phase change. We should also mention studies of the role of solutocapillary stresses in nucleate boiling [24], droplet evaporation [25], and thin film evaporation [26]. However, other than an earlier work [27], there are no theoretical (either numerical or analytical) studies of convection in volatile binary mixtures subjected to a horizontal temperature gradient; the single relevant numerical study [28] did not consider the effects of phase change. Consequently, there is a lack of understanding of the effects of transport (of heat or mass) in the gas phase or the effects of noncondensable gases such as air. The present study addresses this gap in our understanding by introducing a comprehensive two-sided model that provides a quantitative description of transport of heat, mass, and momentum in both the liquid and the gas phase as well as across the liquid-gas interface.

The model is described in detail in Section 2. The results of the numerical investigations of this model are presented, analyzed, and compared with experimental observations reported by Li and Yoda [17] in Section 3. Our summary and conclusions are presented in Section 4.

2. Mathematical model

2.1. Governing equations

When the liquid is a binary mixture of two volatile components (in this study we will focus on the special case of water-methanol mixtures), the gas phase above the liquid layer is generally a multi-component mixture of the vapors of the two components of the liquid and various noncondensable gases (e.g., air) that tend to be dissolved in the liquid. Under typical experimental conditions, one tends to find a ternary mixture containing air whose concentration depends on whether the liquid has been degassed and whether the cavity (cf. Fig. 1) has been evacuated before being filled with the binary liquid. The transport model describing a layer of binary liquid in local thermodynamic equilibrium with the ternary gas mixture can be constructed as a generalization of the two-sided transport models [29–32] describing single-component liquids. Both phases (liquid and gas) will be considered incompressible

$$\nabla \cdot \mathbf{u} = 0 \tag{1}$$

with momentum transport in the bulk described by the Navier-Stokes equation in the Boussinesq approximation

$$\rho(\partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u}) = -\nabla p + \mu \nabla^2 \mathbf{u} + \rho \mathbf{g}, \tag{2}$$

where $\mathbf{u}$ is the velocity, p is the pressure, ρ and μ are the density and dynamic viscosity of the fluid, respectively, and $\mathbf{g}$ is the gravi-

Fig. 1. A sealed test cell containing the liquid and air/vapor mixture. Gravity is pointing in the negative z direction.

tational acceleration. Heat transport in the bulk is described by the advection-diffusion equation

$$\partial_t T + \mathbf{u} \cdot \nabla T = \alpha \nabla^2 T, \tag{3}$$

where T is the temperature and $\alpha = k/\rho C_p$ is the thermal diffusivity of the fluid.

The density of the liquid mixture is

$$\rho_l = \rho_{l,m} + \rho_{l,w}, \tag{4}$$

where $\rho_{l,b}$ is the density of component b in the mixture. Here and below the subscript denotes the phase (l for the liquid, g for the gas), and/or the component in the mixture (m for methanol, w for water, a for air). We will use the subscript i to denote the values at the liquid-gas interface. A linear dependence of the density of each component on the temperature is assumed,

$$\rho_{l,b} = \rho_{l,b}^0 [1 - \beta_{l,b}(T - T_0)], \tag{5}$$

where $\beta_{l,b} = -\rho_{l,b}^{-1} \partial \rho_{l,b}/\partial T$ at $T = T_0$ is the coefficient of thermal expansion, $\rho_{l,b}^0$ is the density of component b in the mixture at the reference temperature T_0, which is given by

$$\rho_{l,b}^0 = n_l Y_b m_b^1, \tag{6}$$

where n_l is the total number density in the liquid, m_b^1 is the mass of one molecule, and $Y_b = n_{l,b}/n_l$ is the concentration (molar fraction) of component b in the liquid phase.

The density and pressure of the gas mixture are

$$\rho_g = \rho_{g,m} + \rho_{g,w} + \rho_{g,a},$$
$$p_g = p_{g,m} + p_{g,w} + p_{g,a}, \tag{7}$$

where all components are assumed to be ideal,

$$\rho_{g,b} = \frac{X_b p_g}{R_b T} = n_g X_b m_b^1,$$
$$p_{g,b} = X_b p_g = n_g X_b k_B T, \tag{8}$$

$X_b = n_{g,b}/n_g$ is the concentration, $R_b = R/M_b$ is the specific gas constant, $M_b = m_b^1 N_A$ is the molar mass of component b, and $R = k_B N_a$ is the universal gas constant. According to the Boussinesq approximation, the spatial average of ρ_l and ρ_g is used on the left-hand-side (but not the right-hand-side) of the Navier-Stokes Eq. (2) for the liquid and the gas phase.

To avoid the assumption of dilute mixtures used in formulating the transport models for simple fluids [29,32], we will describe mass transport in both phases using molar fractions rather than mass densities. The local mass/number conservation for component b (in either the liquid or the gas phase) can be described in terms of the corresponding number density n_b

$$\partial_t n_b + \mathbf{u} \cdot \nabla n_b = -\nabla \cdot \mathbf{j}_b, \tag{9}$$

where $\mathbf{j}_b$ is the diffusive number flux of component b with respect to the bulk mixture that moves with velocity $\mathbf{u}$. The liquid phase is a binary mixture, so we can use Fick's law

$$\mathbf{j}_b = -n_l D_l \nabla Y_b, \tag{10}$$

where D_l is the conventional binary mass diffusivity of the two components. With the assumptions of incompressible flow and constant total number density n_l, the local mass/number conservation Eq. (9) can be rewritten as an advection-diffusion equation for, say, the water concentration in the liquid

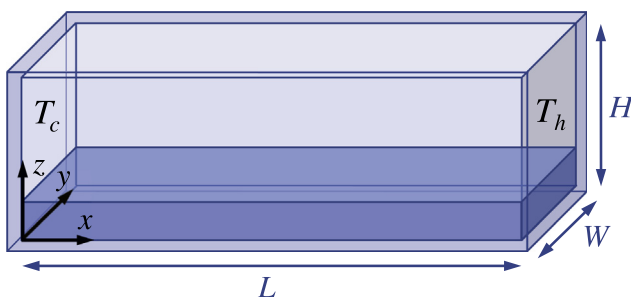$$\partial_t Y_w + \mathbf{u} \cdot \nabla Y_w = \nabla \cdot (D_l \nabla Y_w) \tag{11}$$