# Free-surface flow of confined volatile simple fluids driven by a horizontal temperature gradient: From a comprehensive numerical model to a simplified analytical description

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## Abstract

We study the flow in a confined layer of volatile simple liquid subjected to a horizontal temperature gradient. The somewhat unusual feature of this problem is that the gas layer, which contains a mixture of vapor and air, plays a very important role. Due to phase change occurring at the free surface, the mean flow in the liquid layer and its stability are controlled almost entirely by the mass and heat transport in the gas phase. To explain why this is the case, we use numerical simulations based on a comprehensive two-sided transport model to motivate a simplified analytical description of the problem, which allows us to compute the interfacial temperature and hence the thermocapillary stresses at the free surface which control the flow. The analytical solutions are found to agree quite well with the results of our numerical simulations as well as the results of relevant previous studies.

*Keywords:* Marangoni convection, two-phase flows, interfacial flows, free surface flows, phase change, surface tension effects, thermocapillarity, heat pipes

## 1. Introduction

The problem of convection in volatile fluids with a free surface subject to horizontal temperature gradient has received a lot of attention due to the increasing demand for more efficient and compact cooling technologies. In particular, two-phase evaporative cooling technologies achieve high heat fluxes by exploiting large latent heat associated with phase change at the liquid-gas interface. Thermal management devices such as heat pipes, thermosyphons, and heat spreaders are typically sealed (to avoid loss of working fluid due to evaporation), with a layer of liquid in contact with a layer of gas mixture containing the vapor and the noncondensable gases (such as air) [1]. Under terrestrial conditions, horizontal temperature gradient generates a convective two-phase flow which is driven primarily by a combination of capillary force, thermocapillary stresses, and – for larger devices – buoyancy.

While noncondensable gases are well-known to impede phase change and reduce heat transfer coefficient associated with condensation [2], it is usually infeasible to remove them completely from the gas phase, as air tends to dissolve in liquids and be absorbed in solids. Therefore, a fundamental understanding of two-phase flows and heat and mass transfer with varying levels of noncondensables is essential for better design of evaporative cooling devices. However, our current understanding of this problem remains incomplete (a detailed review of the state-of-art in the field and various open questions can be found in Ref. [3]). A key observation is that the design of thermal management devices to this day is based on experimental studies of buoyancy-thermocapillary convection, the vast majority of which have been performed at ambient (atmospheric) conditions, while the ideal operating conditions correspond to

the gas phase dominated by the vapor. For instance, many theoretical studies [4, 5, 6, 7, 8, 9, 10, 11, 12, 13] focus on the liquid layer and ignore transport in the gas layer, which is only justified at atmospheric conditions, so their results are of limited value where they matter the most. Few theoretical studies have used comprehensive models which described momentum, heat, and mass flux in both liquid and gas phase, as well as a detailed description of phase change at their interface [14, 15, 16, 17]. Yet, without such detailed models it is impossible to correctly identify the limiting factors affecting the performance of evaporative cooling devices or construct simplified models that could be used to build intuition about the key design principles.

We have previously developed variations of detailed two-sided transport models for two-phase flow of volatile fluids driven by horizontal temperature gradients in the presence of noncondensables, which include all four basic components of this problem: (1) the fluid flow and the heat transport in the liquid phase, (2) the fluid flow, and the heat and mass transport in the gas phase, (3) the dynamics at the interface between the two phases (*e.g.* the heat and mass transport across the interface), and (4) the heat conduction within the solid walls of the cavity containing the working fluid. While these models provide a comprehensive description of two-phase flows, they are only valid in the limiting cases when the gas phase is dominated by either air [14, 16] or vapor [17, 18] and cannot accurately describe intermediate compositions. This study will first present a more general transport model valid for arbitrary composition of the gas phase, which can only be solved numerically. Next, to gain the insight into the numerical solutions, we derive a greatly simplified transport model, which can be solved analytically.

The outline of the present study is as follows: The comprehensive model is described in detail in Section 2. The results of
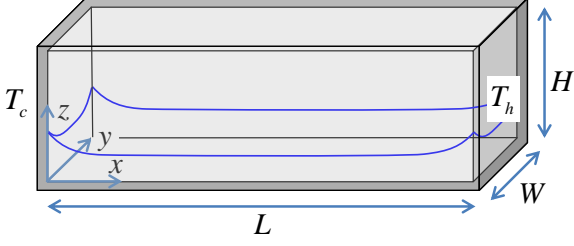
Figure 1: The test cell containing the liquid and gas mixture. Gravity is pointing in the negative $z$ direction. The shape of the contact line reflects the curvature of the free surface.

the numerical investigation of this model are analyzed in Section 3. The simplified model is derived and its predictions are compared with those of the comprehensive numerical model in Section 4. Finally, our summary and conclusions are presented in Section 5.

## 2. Comprehensive Transport Model

### 2.1. Governing Equations

The generalized two-sided model of two-phase flow of non-isothermal volatile fluids presented below is loosely based on the previous models valid in the limiting cases where the gas phase is dominated by either air [16] or vapor [19, 17]. In those models, mass transport was described using the advection-diffusion equation for the mass density of the dilute component. The present model describes mass transport in terms of the molar fraction, which makes it applicable over the entire range of composition. Specifically, local mass conservation for the vapor can be written in terms of its number density $n_v$

$$\partial_t n_v + \nabla \cdot \mathbf{j}_v = 0, \tag{1}$$

where the number density of vapor $n_v$ can be related to the total number density $n_g$ through the vapor concentration (molar fraction) $c_v$

$$n_v = c_v n_g, \tag{2}$$

and $\mathbf{j}_v$ is the total number flux of vapor

$$\mathbf{j}_v = n_v \mathbf{u} - \mathbf{j}'_v, \tag{3}$$

which consists of two components: the first term on the right-hand-side represents the advection due to the gas mixture flowing with the average speed $\mathbf{u}$, and the second term – the diffusion component. The diffusion number flux $\mathbf{j}'_v$ is driven by the concentration gradient, which can be described using the Maxwell-Stefan relation [20]. For a binary mixture, this relation essentially reduces to Fick's law

$$\mathbf{j}'_v = -n_g D \nabla c_v, \tag{4}$$

where the binary mass diffusivity $D$ is a function of pressure and temperature

$$D = D_0 \frac{p_g^0}{p_g} \left( \frac{T}{T_0} \right)^{3/2}, \tag{5}$$

and $D_0$ is the mass diffusivity at the reference temperature $T_0$ and pressure $p_g^0$. With the help of (2), (3), and (4) we find

$$\partial_t(c_v n_g) + \nabla \cdot (c_v n_g \mathbf{u}) = \nabla \cdot (n_g D \nabla c_v). \tag{6}$$

Note that, in this study, we are ignoring the thermodiffusion (Soret) and diffusion-thermo (Dufour) effects, which are typically negligible and trivial to take into account.

Both the vapor and air are assumed ideal, hence the total number density $n_g$ can be computed using the ideal gas law

$$n_g = \frac{p_g}{k_B T}, \tag{7}$$

where $p_g$ is the total pressure in the gas phase and $k_B$ is the Boltzman constant. The corresponding chemical potential is

$$\mu_g = \mu_g^0 + RT \ln \frac{p_g}{p_g^0}, \tag{8}$$

where $R$ is the universal gas constant. The constancy of the chemical potential implies that the gas pressure is constant (even taking viscosity into account this remains an excellent approximation [19]). Since the gas is non-isothermal, the flow cannot be considered incompressible. The conservation law for the total number density is

$$\partial_t n_g + \nabla \cdot (n_g \mathbf{u}) = 0. \tag{9}$$

Combining (6) and (9) we find

$$\partial_t c_v + \mathbf{u} \cdot \nabla c_v = n_g^{-1} \nabla \cdot (n_g D \nabla c_v). \tag{10}$$

Since the product $n_g D \propto T^{1/2}$ depends rather weakly on the temperature (and hence on the position), we consider it constant, yielding an advection-diffusion equation for the concentration of vapor

$$\partial_t c_v + \mathbf{u} \cdot \nabla c_v = D \nabla^2 c_v. \tag{11}$$

The transport equation (11) is valid for the entire range of gas composition. Although a similar equation can be written for air, its concentration $c_a$ can instead be found from

$$c_v + c_a = 1. \tag{12}$$

The conservation equations for the mass, momentum and temperature are standard and remain the same as in the previous models [16, 17]. The liquid phase is considered incompressible, since the corresponding thermal expansion coefficient is small

$$\nabla \cdot \mathbf{u} = 0. \tag{13}$$

For the gas phase, in steady state, we find from (7) and (9)

$$\nabla \cdot \mathbf{u} = -n_g^{-1} \mathbf{u} \cdot \nabla n_g = T^{-1} \mathbf{u} \cdot \nabla T, \tag{14}$$

where the right-hand-side can also be neglected for small temperature gradients considered here (e.g., an applied temperature difference of $\Delta T = 10$ K corresponds to about 1% variation in

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the temperature along the cell). The momentum transport in the bulk is described by the Navier-Stokes equation

$$\rho \left( \partial_t \mathbf{u} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \mu \nabla^2 \mathbf{u} + \rho(T, c_v)\mathbf{g}, \qquad (15)$$

where $p$ is the fluid pressure, $\mathbf{u}$ is the fluid velocity, $\rho$ and $\mu$ are the fluid's density and viscosity, respectively, $c_v$ is the concentration of vapor, and $\mathbf{g}$ is the gravitational acceleration. Following standard practice, the Boussinesq approximation is used, where the density variation due to temperature and concentration is only considered in the last term on the right-hand-side, which represents the buoyancy force. In the liquid phase

$$\rho_l = \rho_l^0 [1 - \beta_l (T - T_0)], \qquad (16)$$

where $\rho_l^0$ is the reference value for density at the reference temperature $T_0$, and $\beta_l = -(\partial \rho_l / \partial T)/\rho_l$ is the coefficient of thermal expansion. In the gas phase

$$\rho_g = \rho_a + \rho_v = (m_a^1 c_a + m_v^1 c_v) n_g, \qquad (17)$$

where $m_a^1$ and $m_v^1$ are the masses of the air and vapor molecules. On the left-hand-side of (15) the density is considered constant for each phase, and is set to be equal to the spatial average of $\rho(T, c_v)$. Finally, the heat transport in the bulk is described by the advection-diffusion equation

$$\partial_t T + \mathbf{u} \cdot \nabla T = \alpha \nabla^2 T, \qquad (18)$$

where $\alpha = k/\rho C_p$ is the thermal diffusivity, $k$ is the thermal conductivity, and $c_p$ is the specific heat capacity, of the fluid.

For a volatile fluid in a sealed cavity, the external temperature gradient causes both evaporation and condensation, which leads to movement of the liquid-gas interface, and the change of the volumes of the two phases. In a numerical implementation of the model, the change in the liquid volume does not satisfy the mass flux balance at the interface exactly due to numerical errors (e.g., discretization). These errors eventually accumulate, leading to a loss off mass conservation for the liquid phase, and resulting, due to the large ratio of the densities, in a large relative error for the mass of vapor, unless corrected. The approximations made in deriving the mass transport equation (11) will also contribute to the error. To ensure global mass/number conservation for each component (vapor and air) we will require

$$\int_{\text{liquid}} n_l \, dV + \int_{\text{gas}} c_v n_g \, dV = N_v = \frac{m_v}{m_v^1} \qquad (19)$$

and

$$\int_{\text{gas}} (1 - c_v) \, n_g \, dV = N_a = \frac{m_a}{m_a^1}, \qquad (20)$$

where $N_v$ and $N_a$ are the total number of the vapor and air, and $m_v$ and $m_a$ are the initial net mass of vapor and air. A (spatially uniform) pressure offset $p_o$ and vapor concentration correction $\Delta c_v$ are introduced to enforce these conservation laws. They are updated at each time step by solving the constrains (19) and (20) with

$$c_v = c_v' + \Delta c_v, \qquad (21)$$
$$p_g = p + p_o,$$

where $c_v'$ is the (uncorrected) numerical solution of the transport equation (11), $p_g$ is the absolute pressure, and $p$ is the dynamic pressure in the gas phase obtained by solving the momentum conservation (15) and incompressibility equation (13).

## 2.2. Boundary Conditions

The system of coupled evolution equations (13), (15), (18), and (11) for the velocity, pressure, temperature, and concentration fields should be solved in a self-consistent manner, subject to the boundary conditions describing the balance of momentum, heat, and mass/number fluxes. In the comprehensive model, we will describe phase equilibrium using the Antoine equation, which is valid over a wide range of conditions,

$$\ln p_v = A_v - \frac{B_v}{C_v + T}, \qquad (22)$$

where $A_v$, $B_v$, and $C_v$ are empirical coefficients. In the simplified description, it will be more convenient to use the more popular Clausius-Clapeyron equation

$$\ln \frac{p_v}{p_v^0} = -\frac{\mathcal{L}}{\bar{R}_v} \left( \frac{1}{T} - \frac{1}{T_0} \right), \qquad (23)$$

where $p_v^0$ is the reference value of the vapor pressure at the reference temperature $T_0$ and $\bar{R}_v$ is the specific gas constant for vapor. The Clausius-Clapeyron and the Antoine equation are equivalent for small deviations from $T_0$ when the latent heat is defined according to

$$\mathcal{L} = \frac{B_v \bar{R}_v T_0^2}{(C_v + T_0)^2}, \qquad (24)$$

as can be verified by evaluating the relations (22) and (23) and their partial derivatives with respect to $T$ at $T_0$. In the following, these relations will be used to define the saturation temperature $T_s$ in terms of the partial pressure $p_v$.

We will rely on the kinetic theory of gases [21] which assumes that the chemical potential and the temperature are continuous across the liquid-gas interface

$$T_l = T_i = T_g, \qquad (25)$$

yielding the following expression for the number density flux across the interface due to phase change

$$j_v = \frac{2\lambda}{2 - \lambda} n_g c_v \sqrt{\frac{R_v T_i}{2\pi}} \left[ \frac{p_l - p_g}{\rho_l R_v T_i} + \frac{\mathcal{L}}{R_v T_i} \frac{T_i - T_s}{T_s} \right], \qquad (26)$$

where $\lambda$ is the accommodation coefficient and $T_s$ is computed from (22). For nonpolar liquids, the accommodation coefficient is found to be equal (or very close) to unity [22, 23], so we set $\lambda = 1$ in this study.

The mass/number flux balance for the vapor on the gas side of the interface is given by

$$j_v = n_g c_v \hat{\mathbf{n}} \cdot (\mathbf{u}_g - \mathbf{u}_i) - n_g D \, \partial_n c_v, \qquad (27)$$

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where $\mathbf{u}_i$ is the velocity of the interface and $\hat{\mathbf{n}}$ is the surface normal vector. Since air is noncondensable, its flux across the interface is zero:

$$0 = n_g c_a \hat{\mathbf{n}} \cdot (\mathbf{u}_g - \mathbf{u}_i) - n_g D \, \partial_n c_a. \tag{28}$$

Using (12), (27), and (28) we can find a pair of boundary conditions for $\mathbf{u}_g$ and $c_v$ at the interface:

$$\hat{\mathbf{n}} \cdot (\mathbf{u}_g - \mathbf{u}_i) = \frac{j_v}{n_g}, \tag{29}$$

and

$$\partial_n c_v = -\frac{1 - c_v}{D} \frac{j_v}{n_g}. \tag{30}$$

The mass/number flux balance on the liquid side of the interface is given by

$$\hat{\mathbf{n}} \cdot (\mathbf{u}_l - \mathbf{u}_i) = \frac{j_v}{n_l}, \tag{31}$$

where the right-hand-side can be effectively set to zero, since $n_l \gg n_g$. While the normal component of velocity is discontinuous at the interface in the presence of phase change, the tangential components of velocity are continuous:

$$(\mathbb{I} - \hat{\mathbf{n}}\hat{\mathbf{n}}) \cdot (\mathbf{u}_l - \mathbf{u}_g) = 0. \tag{32}$$

The normal component of the stress balance at the interface

$$\hat{\mathbf{n}} \cdot (\mathbf{\Sigma}_l - \mathbf{\Sigma}_g) \cdot \hat{\mathbf{n}} = \sigma \kappa + J_v^2(\rho_l^{-1} - \rho_v^{-1}) \tag{33}$$

incorporates the capillary pressure and vapor recoil [24], where

$$\mathbf{\Sigma} = \mu \left[ \nabla \mathbf{u} + (\nabla \mathbf{u})^T \right] - p\mathbb{I} \tag{34}$$

is the stress tensor and $J_v = m_v^1 j_v$ is the mass flux of vapor. The effect of vapor recoil is found to be negligible in this study. The tangential component of the stress balance

$$(\mathbb{I} - \hat{\mathbf{n}}\hat{\mathbf{n}}) \cdot (\mathbf{\Sigma}_l - \mathbf{\Sigma}_g) \cdot \hat{\mathbf{n}} = \nabla_s \sigma \tag{35}$$

accounts for the thermocapillary effect, where

$$\nabla_s \sigma = -\gamma \nabla_s T_i, \tag{36}$$

$\gamma = -\partial \sigma / \partial T$ is the temperature coefficient of surface tension and

$$\nabla_s = (\mathbb{I} - \hat{\mathbf{n}}\hat{\mathbf{n}}) \cdot \nabla \tag{37}$$

is the surface gradient. The heat flux balance across the interface is given by

$$\mathcal{L} J_v = k_g \hat{\mathbf{n}} \cdot \nabla T_g - k_l \hat{\mathbf{n}} \cdot \nabla T_l. \tag{38}$$

Finally let us discuss the boundary conditions at the inner surfaces of the cavity confining the fluid. Following the previous experimental [25] and numerical studies [16, 17], we assume the fluid is contained in a rectangular test cell with inner dimensions $L \times W \times H$ (cf. Fig. 1) and thin walls of thickness $\delta_w$ and conductivity $k_w$. The left wall is cooled with constant temperature $T_c$ imposed on the outer surface, while the right

wall is heated with constant temperature $T_h > T_c$ imposed on the outer surface. Since the walls are thin, one-dimensional conduction (along the $x$-direction) is assumed through all the walls, yielding the following mixed boundary conditions on the inner surfaces of the side walls:

$$T|_{x=0} = T_c + k_\iota \frac{\delta_w}{k_w} \hat{\mathbf{n}} \cdot \nabla T, \tag{39}$$

$$T|_{x=L} = T_h + k_\iota \frac{\delta_w}{k_w} \hat{\mathbf{n}} \cdot \nabla T,$$

where $\hat{\mathbf{n}}$ is the wall-normal vector, and $\iota = g$ ($\iota = l$) above (below) the contact line. The remaining walls of the cell are assumed to be in contact with air, which is a poor thermal conductor, and hence can be considered adiabatic:

$$\partial_n T = 0. \tag{40}$$

Standard no-slip boundary conditions $\mathbf{u} = 0$ for the velocity and no-flux boundary conditions for the concentration of vapor

$$\hat{\mathbf{n}} \cdot \nabla c_v = 0 \tag{41}$$

are imposed on all the inner surfaces of the walls.

## 3. Results

To validate the generalized two-sided transport model described here, we implemented it numerically and reproduced several key results obtained previously for the well-studied problem [26, 16, 17, 3] of buoyancy-thermocapillary-driven flow of volatile silicone oil (hexamethyldisiloxane) confined in a sealed rectangular test cell made of quartz (fused silica). The inner dimensions of the test cell are $L \times H \times W$ = 48.5 mm $\times$10 mm $\times$10 mm and all material parameters of the fluid and its vapor can be found in Ref. [17].

A horizontal temperature gradient is applied by keeping the outer surfaces of the end walls at the temperatures $T_c = T_0 - \Delta T/2$ and $T_h = T_0 + \Delta T/2$. For comparison with the previous numerical studies, the same average thickness of the liquid layer $d_l$ = 2.5 mm (which corresponds to the dynamic Bond number $Bo_D = O(1)$) and contact angle of 50° were used. While the numerical implementation of the model can describe flows in either 2D or 3D geometry, the results presented here were obtained exclusively for 2D flows (where the variation as well as the flow in the $y$-direction are neglected).

### 3.1. Dilute Limits

Previous numerical studies [16, 17] used transport models that were only justified in the limiting cases where the gas phase is dominated by either vapor or air. In these limits the predictions of the present model should agree with those older models, and in fact this is what we find. Fig. 2 shows the asymptotic states of the flow (after initial transients have died down) for $T_0$ = 293 K and $\Delta T$ = 4 K at the average air concentration $\bar{c}_a$ = 0.01 (when the vapor is dominant) and $\bar{c}_a$ = 0.96 (atmospheric conditions, when the air is dominant). This temperature difference was chosen since it corresponds to steady flows for
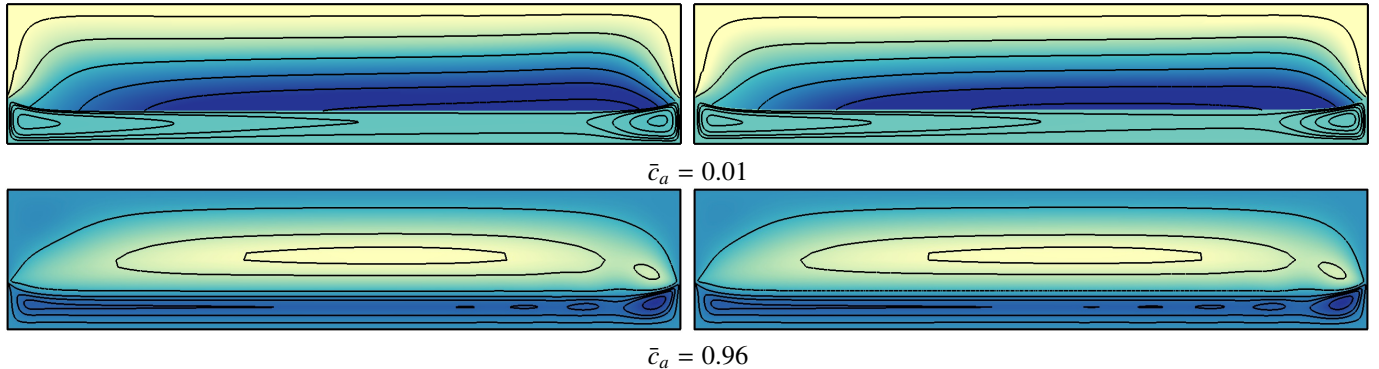
$\bar{c}_a = 0.01$

$\bar{c}_a = 0.96$

Figure 2: Numerical results for the flow field at $\Delta T = 4$ K with different average concentrations of air $\bar{c}_a$ using the new (general) model (left column), and the previous (limiting-case) models [16, 17] (right column). The cold end wall is on the left. Solid lines represent the streamlines of the flow. Here and below, the background represents the value of the stream function $\psi$, where darker (lighter) shade indicates higher (lower) values of $\psi$.

the entire range of $\bar{c}_a$, as we show next (cf. Fig. 5). Although the shape of the streamlines depends on the (arbitrary) choice of the streamfunction values, the flow patterns produced using different transport models are found to be essentially indistinguishable in both cases.

A more quantitative comparison in terms of the interfacial mass flux $J_v$ describing phase change, interfacial temperature profile $T_i$ which controls the thermocapilllary stresses, and the resulting interfacial flow velocity $u_i$ is provided in Fig. 3. Again, we find the predictions of the present model to be quite similar to those of the previous studies. The slight discrepancy in $u_i$ observed near the cold end of the test cell is due to the difference in the treatment of the gradient of the number density at the interface between the two models. The previous models [16, 17], which rely on local mass conservation of the dilute component, lead to the following relation between the mass fractions of vapor and air

$$\frac{\partial_n \rho_v}{M_v} + \frac{\partial_n \rho_a}{M_a} = -\frac{p_g}{RT_i^2}\partial_n T. \qquad (42)$$

The current formulation uses molar fractions instead, where (12) yields instead

$$\partial_n c_v + \partial_n c_a = 0. \qquad (43)$$

The two relations become formally equivalent only when the gradient of $T$ (or $n_g$) normal to the interface vanishes. When the gradient is not too large, the difference is quite small, so both models can be considered to provide a comparably accurate description of transport.

### 3.2. Flow Regimes

Having validated the generalized transport model, we next investigate the effect of air on the convective flow over the entire range of $\bar{c}_a$, which was previously inaccessible to numerical simulations. Most of the results presented correspond to the imposed temperature difference of $\Delta T = 10$ K, to enable comparison with previous studies. Another advantage of this choice is that, as $\bar{c}_a$ varies between the two limits, the flow exhibits all the qualitatively different regimes observed previously

at $Bo_D = O(1)$, with the corresponding flow states shown in Fig. 4. At atmospheric conditions (cf. Fig. 4(a)), we find an oscillatory multicellular flow (OMC) with the convection rolls covering the entire liquid layer. As $\bar{c}_a$ is decreased, the flow becomes steady and a steady multicellular (SMC) convection pattern is found (cf. Fig. 4(b)) with convection rolls still covering the entire liquid layer but becoming noticeably weaker. As $\bar{c}_a$ is decreased further (cf. Fig. 4(c-d)), the convection rolls in the liquid layer further weaken and start to disappear near the cold end, but persist near the hot end; the resulting state is referred to as the partial multicellular (PMC) pattern. At even lower $\bar{c}_a$ (cf. Fig. 4(e-f)), convection rolls in the bulk of the liquid layer disappear completely. This pattern is known as a steady unicellular flow (SUF) and features only a pair of convection rolls driven by buoyancy, one near each end wall.

The presence of air also strongly affects the flow in the gas phase. At relatively high $\bar{c}_a$, phase change is greatly suppressed and the flow in the gas phase is similar to that in the liquid (cf. Fig. 4(a)), with convection rolls appearing at almost the same positions along the interface, but rotating in the opposite direction. As $\bar{c}_a$ decreases, the convection rolls gradually weaken and disappear, also starting near the cold end (cf. Fig. 4(b-c)). At $\bar{c}_a \lesssim 0.3$, phase change becomes stronger, as indicated by the streamlines that originate and terminate at the interface (cf. Fig. 4(d-e)). For ever lower $\bar{c}_a$ (cf. Fig. 4(f)) all recirculation zones in the gas phase disappear, and the flow becomes unidirectional, from the hot side where the liquid evaporates to the cold side where the vapor condenses.

The different regimes are summarized in the flow regime map presented in Fig. 5 as a function of the average air concentration $\bar{c}_a$ and the interfacial Marangoni number

$$Ma_i = \frac{\gamma d_l^2}{\mu_l \alpha_l}\bar{\tau}. \qquad (44)$$

Here $\bar{\tau}$ is the spatial average of the interfacial temperature gradient $\tau = \nabla_s T_i$, which reflects the imposed temperature difference. As the Figure illustrates, the flow regimes found in the simulations based on the generalized transport model are found to be in good agreement with both the experimental observations [25], and the predictions of the linear stability analysis
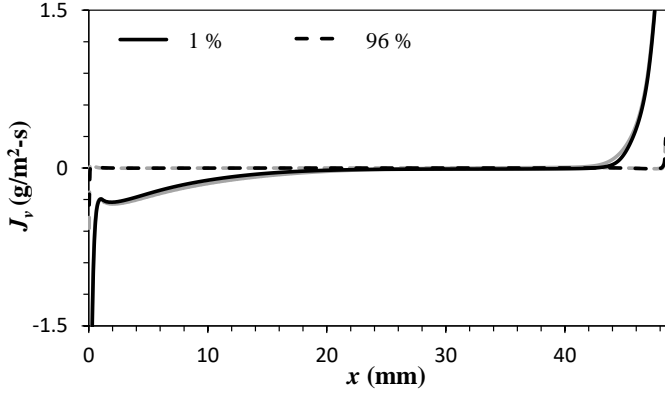
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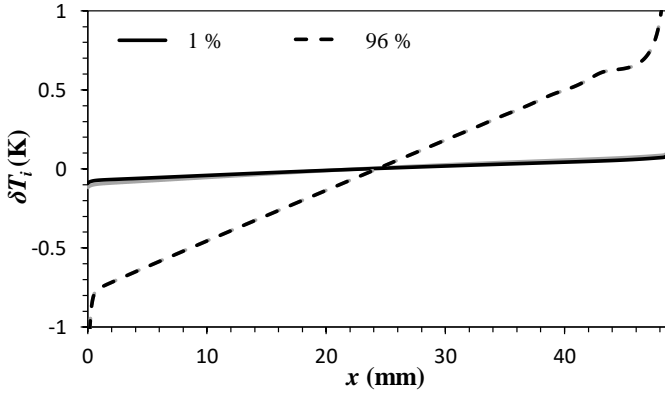


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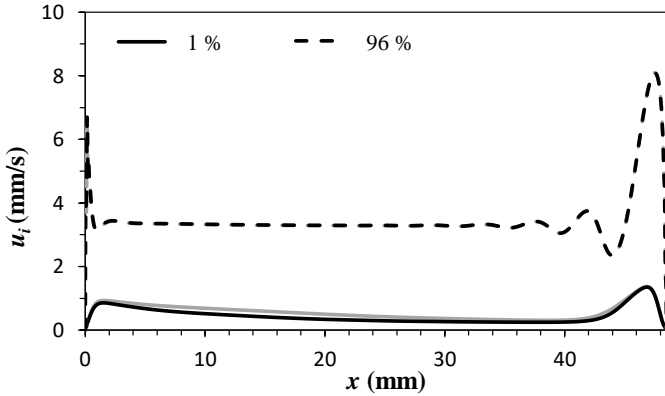


(c)

Figure 3: Numerical results for the mass flux due to phase change (a), interfacial temperature profile (b), and the magnitude of the interfacial velocity (c) at $\Delta T = 4$ K with different average concentrations of air $\bar{c}_a$. To highlight the variation of the mass flux $J_v$ and the interfacial temperatude $T_i$ in the central region of the cavity, the $y$-axis is truncated in (a) and (b), respectively. In addition, the deviation $\delta T_i = T_i - \langle T_i \rangle_x$ from the average is plotted in (b). Black lines represent the results based on the current model, grey lines – the results based on the previous models [16, 17].
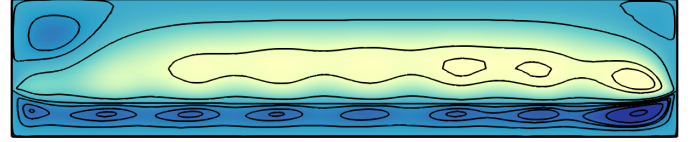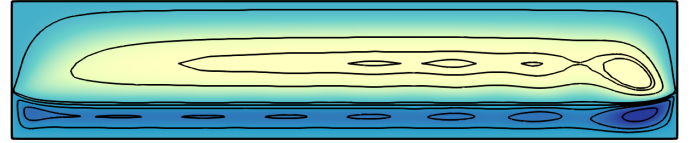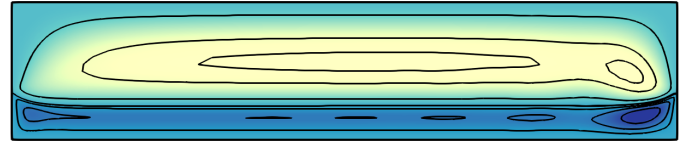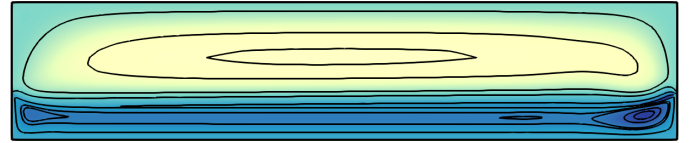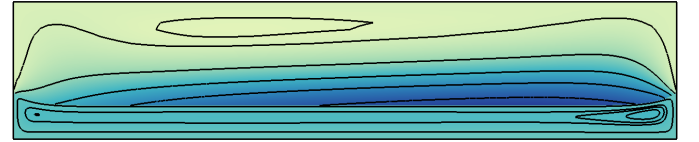


(a) $\bar{c}_a = 0.96$

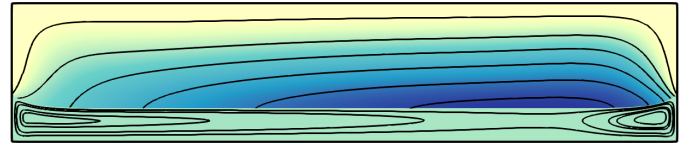(b) $\bar{c}_a = 0.70$

(c) $\bar{c}_a = 0.50$

(d) $\bar{c}_a = 0.30$

(e) $\bar{c}_a = 0.08$

(f) $\bar{c}_a = 0.01$

Figure 4: Numerical results for the flow field at $\Delta T = 10$ K. The cold end wall is on the left. Solid lines represent the streamlines of the flow; color corresponds to the values of stream function $\psi$, where darker (lighter) indicates higher (lower) values of $\psi$.
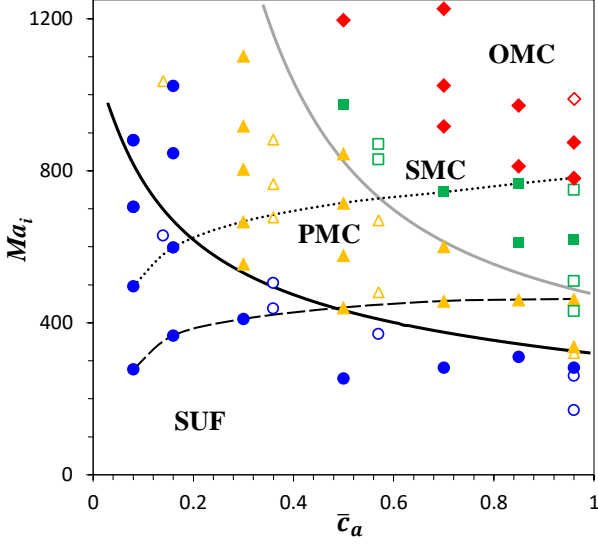
Figure 5: Flow regime diagram in the $\bar{c}_a - Ma_i$ parameter space: SUF ($\bigcirc$), PMC ($\triangle$), SMC ($\square$), and OMC ($\diamond$). Open symbols correspond to experimental results of Li *et al.* [25] and filled symbols – numerical results obtained in this study. Thick solid lines represent the critical Marangoni number for onset of the PMC (black) and SMC (dark grey) predicted by the linear stability analysis [27]. Thin dotted (dashed) black line corresponds to $\Delta T = 10$ K ($\Delta T = 4$ K).

[27], except at $\bar{c}_a \lesssim 0.2$ (the reasons for the discrepancy will be discussed later).

## 4. Simplified Transport Model

In this section we will use the insight provided by the numerical simulations to derive a simplified transport model capable of providing a quantitative description of the flow in the SUF regime. The previous analysis of the $\bar{c}_a \to 0$ limit [17] has shown that it is the concentration field in the gas phase that ultimately plays the dominant role in this problem: it determines the interfacial temperature, surface stresses, and hence the flow field. As we will show below, a pretty accurate analytical description of the concentration field can be obtained for the base flow over the entire range of $\bar{c}_a$ in the central region of the flow, where the interface is nearly flat. The original analysis assumed that phase change and advection in this region are negligible to compute the concentration field $c_v = c_v(x)$. Why, and under which conditions, this result is valid will be discussed below.

Following Ref. [28], we will introduce the rescaled coordinates $\chi = x/d_g$ and $\zeta = z/d_g$, where $d_g = H - d_l$ is the thickness of the gas layer in the central region. If we assume the interface coincides with the plane $z = 0$, then the gas phase corresponds to $0 < \zeta < 1$ and $0 < \chi < \Gamma_g$, where $\Gamma_g = L/d_g$ is the aspect ratio of the gas layer. Since the flow field is constrained to the $\chi - \zeta$ plane and is incompressible, it can be written in terms of the stream function $\psi(\chi, \zeta)$,

$$\mathbf{u}_g = \hat{\mathbf{x}}\partial_\zeta\psi - \hat{\mathbf{z}}\partial_\chi\psi. \tag{45}$$

As Fig. 3(a) illustrates, phase change in the central region is indeed negligible in the SUF regime. Hence, for a sufficiently

large $\Gamma_g$ (in this study $\Gamma_g \approx 6.5$), the flow of gas in this region can be assumed horizontal

$$\mathbf{u}_g = u_x\hat{\mathbf{x}} + u_z\hat{\mathbf{z}} \approx u_x\hat{\mathbf{x}}, \tag{46}$$

where $u_z/u_x = O(\Gamma_g^{-1})$. We can therefore simplify the mass transport equation (11) to

$$d_g u_x \partial_\chi c_v = D(\partial_\chi^2 c_v + \partial_\zeta^2 c_v), \tag{47}$$

with the vertical component $u_z$ of the velocity yielding a higher order correction. Furthermore, the horizontal component of velocity $u_x$ can be decomposed into two contributions:

$$u_x = \bar{u}_x + \tilde{u}_x(\chi, \zeta), \tag{48}$$

where $\bar{u}_x = \text{const} < 0$ represents the mean flow (the vapor flows in the direction opposite to the applied temperature gradient) and $\tilde{u}_x$ represents the zero-mean recirculation flow

$$\int_0^1 \tilde{u}_x d\zeta = 0. \tag{49}$$

Correspondingly, we can write

$$\psi = \bar{u}_x\zeta + \tilde{\psi}(\zeta) + O(\Gamma_g^{-1}), \tag{50}$$

where $\tilde{u}_x = \partial_\zeta\tilde{\psi}$ and

$$\int_0^1 \partial_\zeta\tilde{\psi}d\zeta = \tilde{\psi}(1) - \tilde{\psi}(0) = 0. \tag{51}$$

Since the phase change at the liquid-gas interface is negligible in the central region, the mass/number flux vanishes, i.e., $\partial_\zeta c_v = 0$, at both $\zeta = 0$ and $\zeta = 1$. The solution obtained in Ref. [17] can be reproduced by setting $u_x = \bar{u}_x$ (i.e., $\tilde{u}_x = 0$) and solving (47) subject to the no-flux boundary conditions on $c_v$, which yields

$$c_v = C_0 + C_1 e^{-Pe_m\chi}, \tag{52}$$

where

$$Pe_m = \frac{|\bar{u}_x|d_g}{D} \tag{53}$$

is the Péclet number which corresponds to the mean flow and $C_0$ and $C_1$ are the constants which can be determined based on the net amount of vapor and the net mass flux associated with phase change (to be defined later). In the general case (i.e., $\tilde{u}_x \neq 0$), the solution to (47) is

$$c_v = C_0 + C_1 e^{-Pe_m\chi}[1 + g(\zeta)], \tag{54}$$

where the function $g(\zeta)$ describing the vertical concentration profile satisfies the differential equation

$$g''(\zeta) = \frac{\bar{u}_x\tilde{u}_x(\zeta)d_g^2}{D^2}[1 + g(\zeta)]. \tag{55}$$

The right-hand-side of (55), and hence $g(\zeta)$ itself, is of order $\epsilon = Pe_m Pe_r$, where

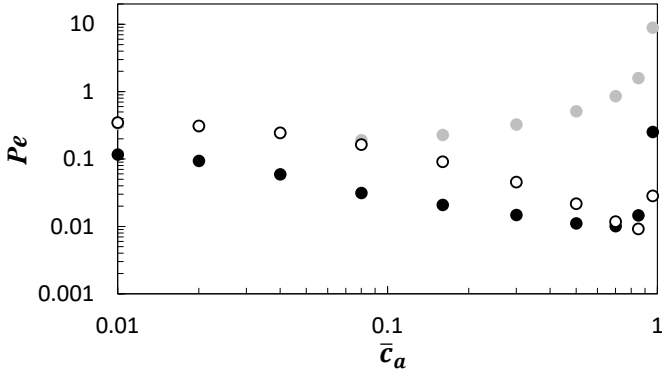$$Pe_r = \max_\zeta \frac{|\tilde{u}_x(\zeta)|d_g}{D} \tag{56}$$

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Figure 6: The numerical values of $Pe_m$ (open symbols), $Pe_r$ (gray symbols), and their product $\epsilon$ (black symbols) at $\Delta T = 10$ K.

is the Péclet number describing the strength of the recirculation flow $\tilde{u}_x$. To leading order in $\epsilon$, we find

$$g(\zeta) = \frac{\bar{u}_x d_g^2}{D^2} \int \tilde{\psi}(\zeta) d\zeta + O(\epsilon^2). \tag{57}$$

The no-flux boundary conditions for $c_v$ require $g'(0) = g'(1) = 0$. This is only consistent with (57) when (51) is satisfied, which justifies the separation of the flow field into the two components, $\bar{u}_x$ and $\tilde{u}_x$.

As long as $\epsilon \ll 1$, the variation of $c_v$ in the vertical direction is negligible and the general solution (54) reduces to the special case (52). The crucial observation is that although $Pe_r$ can become quite large for $\bar{c}_a \to 1$, $\epsilon$ remains small regardless of the average concentration $\bar{c}_a$ of air for sufficiently low $\Delta T$, as Fig. 6 illustrates. Indeed, numerically computed solutions for all $\bar{c}_a$ are characterized by concentration field (cf. Fig. 7) whose gradient in the vertical direction is small compared with that in the horizontal direction in the central region of the test cell.

In order to compare the analytical predictions with numerical results quantitatively, we need to determine the mean flow velocity in the gas. Since mass transport is essentially one-dimensional in the central region, the fluxes of vapor and air satisfy

$$J = m_v^1 n_g (D\partial_x c_v - \bar{u}_x c_v),$$
$$0 = m_a^1 n_g (D\partial_x c_a - \bar{u}_x c_a), \tag{58}$$

where $J$ is the mass flux of vapor. Adding these two equations (notice that $c_v + c_a = 1$, $\partial_x c_v + \partial_x c_a = 0$), we find

$$\bar{u}_x = -\frac{J}{m_v^1 n_g}. \tag{59}$$

Using mass conservation, the mean mass flux at location $x$, $J_g(x)$, can be approximated by integrating the local mass flux $J_v$ describing phase change at the interface, from $x$ to $L$:

$$J_g(x) = \frac{1}{d_g} \int_x^L J_v(x') \, dx'. \tag{60}$$

Since evaporation (condensation) takes place mainly near the hot (cold) wall, mean flux is a function of $x$, however, as suggested by the numerical results, it becomes essentially constant

$J_g(x) \approx J_0$ in the central region of the test cell (cf. Fig. 8), where we have defined the characteristic mean mass flux

$$J_0 = \max_x J(x). \tag{61}$$

We will therefore use $J_0$ in place of $J$ in (59) for computing the mean flow velocity in the gas.

Let us next consider the dependence of the mean flux $J_0$ on the average concentration of air $\bar{c}_a$. For sufficiently low $\Delta T$ and highly volatile fluids, the overall heat transport between the end walls is dominated by the latent heat associated with phase change, and the heat flux balance gives

$$J_0 \mathcal{L} = \frac{\Delta T}{Z_T}, \tag{62}$$

where

$$Z_T = Z_o + Z_d, \tag{63}$$

is the total net thermal resistance, $Z_o$ is the thermal resistance due to conduction through the end walls and the liquid wedges between the wall and the interface, and $Z_d$ is the diffusive resistance of the gas layer [28]. While $Z_o$ can be considered independent of the air concentration, $Z_d$ depends sensitively on $\bar{c}_a$.

Since heat flows though the end walls and the liquid wedges between the wall and the interface sequentially, $Z_o$ is a sum of the conduction resistances of the end walls $Z_w = \delta_w/k_w$ and the conduction resistances of the liquid wedges $Z_l \approx 0.5 d_l/k_l$, yielding

$$Z_o = 2(Z_w + Z_l) \approx 2\frac{\delta_w}{k_w} + \frac{d_l}{k_l}. \tag{64}$$

To determine $Z_d$, we solve the system of equations (58) together with (59), which yields

$$J_0 = \frac{m_v^1 n_g D}{c_a} \partial_x c_v. \tag{65}$$

For volatile fluids, $T_i$ is very close to $T_s$ [19], so the concentration and temperature profiles are related. The phase equilibrium condition (23) can be expressed in terms of the concentrations using the relation

$$\frac{p_v}{p_v^0} = \frac{c_v}{\bar{c}_v}, \tag{66}$$

where $\bar{c}_v = 1 - \bar{c}_a$, such that

$$\frac{1}{T_i} = \frac{1}{T_0} - \frac{\bar{R}_v}{\mathcal{L}} \ln \frac{c_v}{\bar{c}_v} \tag{67}$$

and consequently

$$\partial_x c_v = -\partial_x c_a = \frac{\mathcal{L} c_v}{\bar{R}_v T_i^2} \tau, \tag{68}$$

where $\tau = \partial_x T_i$. Substituting the mean values for the concentrations and interfacial temperatture into (68) and (65) yields

$$J_0 = \frac{1 - \bar{c}_a}{\bar{c}_a} \frac{m_v^1 n_g \mathcal{L} D}{\bar{R}_v T_0^2} \bar{\tau}, \tag{69}$$

$\bar{c}_a = 0.96, \Delta T = 4$ K, $\Delta c_a = 5.14 \times 10^{-3}$      $\bar{c}_a = 0.96, \Delta T = 10$ K, $\Delta c_a = 1.13 \times 10^{-2}$

$\bar{c}_a = 0.70, \Delta T = 4$ K, $\Delta c_a = 3.70 \times 10^{-2}$      $\bar{c}_a = 0.70, \Delta T = 10$ K, $\Delta c_a = 7.18 \times 10^{-2}$

$\bar{c}_a = 0.50, \Delta T = 4$ K, $\Delta c_a = 5.5 \times 10^{-2}$      $\bar{c}_a = 0.50, \Delta T = 10$ K, $\Delta c_a = 1.05 \times 10^{-1}$

$\bar{c}_a = 0.30, \Delta T = 4$ K, $\Delta c_a = 6.48 \times 10^{-2}$      $\bar{c}_a = 0.30, \Delta T = 10$ K, $\Delta c_a = 1.22 \times 10^{-1}$

$\bar{c}_a = 0.08, \Delta T = 4$ K, $\Delta c_a = 4.68 \times 10^{-2}$      $\bar{c}_a = 0.08, \Delta T = 10$ K, $\Delta c_a = 9.14 \times 10^{-1}$

$\bar{c}_a = 0.01, \Delta T = 4$ K, $\Delta c_a = 1.03 \times 10^{-2}$      $\bar{c}_a = 0.01, \Delta T = 10$ K, $\Delta c_a = 2.08 \times 10^{-2}$
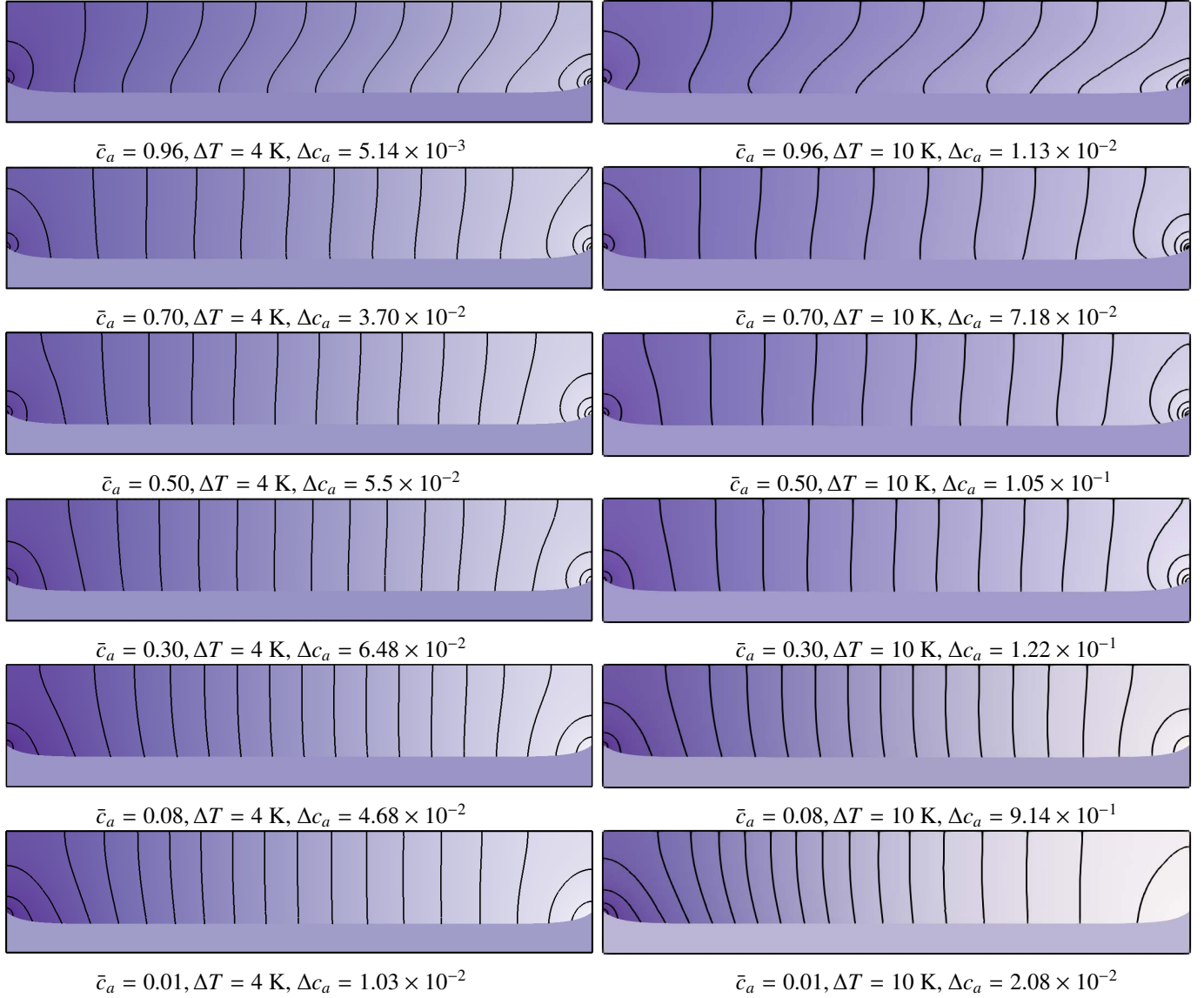
Figure 7: Numerical results for air concentration $c_a = 1 - c_v$ in the gas phase at $\Delta T = 4$ K (left column) and 10 K (right column), with different $\bar{c}_a$. Solid lines represent the twenty equally spaced level sets of the concentration fields. In the gas phase darker background indicates higher air concentration, while in the liquid phase the concentration field is undefined.
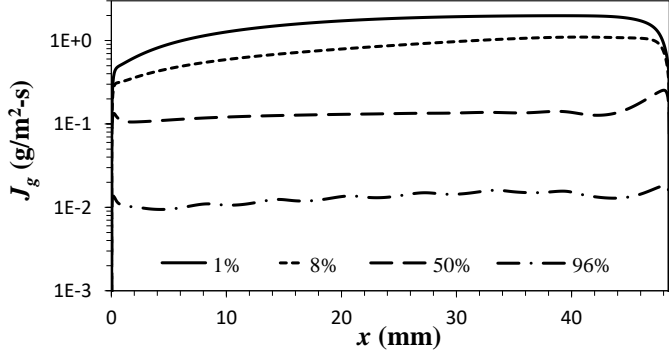
Figure 8: Mean mass flux $J_g(x)$ in the gas phase (60) across the vertical cross-section of the cavity at location $x$. The value is computed numerically at $\Delta T = 10$ K with different average concentrations of air $\bar{c}_a$.
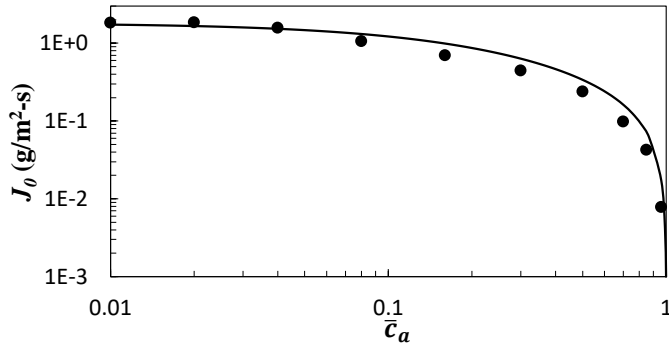


Figure 9: Characteristic mass flux $J_0$ as a function of the average concentration of air $\bar{c}_a$ at $\Delta T = 10$ K. Solid line represents the analytical estimate based on (62), symbols – numerical results obtained using (60) and (61).



(a)



(b)

Figure 10: Normalized concentration profiles in the gas phase at $\Delta T = 10$ K for vapor (a) and (b) air with different $\bar{c}_a$. Numerical and analytical results are represented by gray and black lines, respectively. Numerical results correspond to the mid-height of the gas layer.

where $\bar{\tau} = \Delta T'/L$ and $\Delta T' = T|_{x=L} - T|_{x=0}$ is the temperature difference between the inner surfaces of the hot and cold end walls. The heat flux balance at the interface near either of the end walls yields a relation similar to (62) which only includes the diffusive resistance
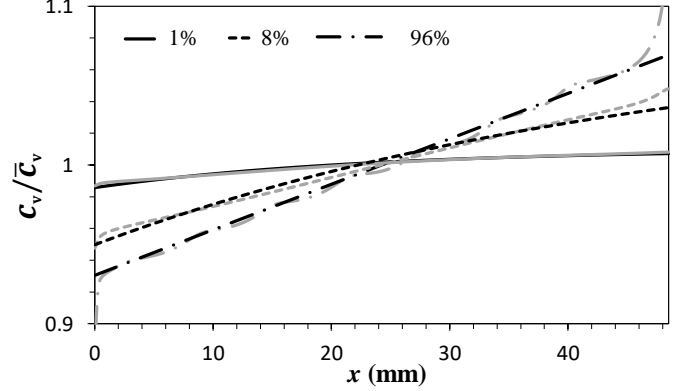
$$J_0 \mathcal{L} \approx \frac{\Delta T'}{Z_d}. \tag{70}$$
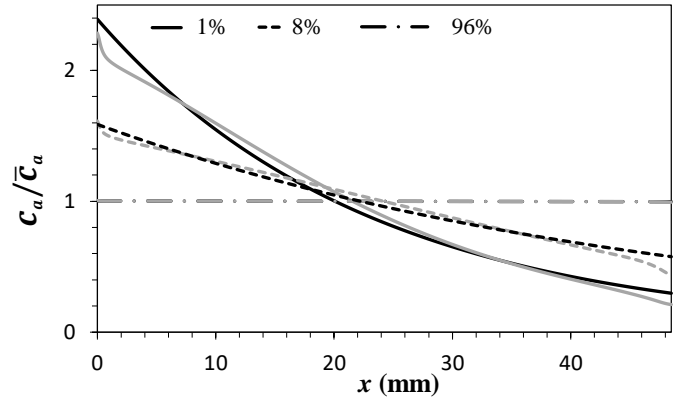
Solving (7), (69), and (70) together, we find

$$Z_d \approx \frac{\bar{c}_a}{1 - \bar{c}_a} \frac{\bar{R}_v^2 T_0^3 L}{D p_g \mathcal{L}^2}, \tag{71}$$

where $D p_g = D_0 p_0$ is independent of $\bar{c}_a$ according to (5). This result corresponds to the effective condensation thermal conductivity $k_c = L/Z_d$ obtained by Peterson et al. [29].

With the solution for $Z_o$ and $Z_d$, we can finally obtain the analytical estimate for $J_0$ using (62) and compare it with the numerical result for $J_0$, which is obtained using the numerical solution of local mass flux given by (60) and (61). Fig. 9 shows that the analytical and numerical results are in good agreement over the entire range of $\bar{c}_a$, suggesting that the one-dimensional description of transport in the gas phase is reasonably accurate. Note that at $\Delta T = 10$ K the flow is in the OMC regime at $\bar{c}_a = 0.96$ (cf. Fig. 5), so the predictions of our simplified transport model can be trusted far outside the SUF regime. As

expected, the vapor flux is the largest when the air is removed entirely from the cell. An increase in $\bar{c}_a$ leads to an increase in the diffusive resistance and, correspondingly, a decrease in the vapor flux. The minor discrepancy between the numerical and analytical results is likely due to the contribution of heat conduction and advection in the liquid layer that have been ignored in our analysis of heat flux balance (62). These contributions are negligible for low $\bar{c}_a$, but would become progressively more important at higher $\bar{c}_a$, when phase change is suppressed, and our estimates of $J_0$ are expected to overestimate the numerical results, consistent with Fig. 9. In practical applications (e.g., for heat pipes) the liquid layer will be substantially thinner, so convective heat flux would be negligible and the prediction of our simplified transport model would be even more accurate.

To compare the analytical prediction (52) for the concentration profiles with the numerical results, we need to determine the constants $C_0$ and $C_1$ in (52) or (54). When the concentration profile can be considered one-dimensional (i.e., for $\epsilon \ll 1$), the solution (52) can be obtained directly from the second of the two equations in (58) which yields

$$\partial_x c_a = -\frac{Pe_m}{d_g} c_a. \tag{72}$$

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The average value of air concentration satisfies

$$\frac{1}{L}\int_0^L c_a dx = \bar{c}_a, \tag{73}$$

so that

$$c_a = 1 - c_v = \bar{c}_a \frac{Pe_m \Gamma_g e^{-Pe_m \chi}}{1 - e^{-Pe_m \Gamma_g}}. \tag{74}$$

Note that $Pe_m$ can be computed from (53), (59), and (69) as long as the relation between $\bar{\tau}$ and $\Delta T$ is known.

As Fig. 10 illustrates, we find rather good agreement between the analytical predictions and the numerical results in the central region of the cavity. In the general case, the concentrations of both components in the gas mixture have an exponential profile. The concentration profiles become approximately linear only when $Pe_m \ll \Gamma_g^{-1}$. This limit corresponds to low values of the mean flow velocity $\bar{u}_x$ and hence low values of $\Delta T$ and/or high values of $\bar{c}_a$. In particular, for $\Delta T = 10$ K, we have $Pe_m < \Gamma_g^{-1}$ for $\bar{c}_a > 0.08$, and the concentration profiles indeed become essentially linear for $\bar{c}_a \gtrsim 0.08$ as shown in Fig. 10. The deviations noticeable at low $\bar{c}_a$ are mainly due to the breakdown of our assumption that phase change at the interface is negligible.

Substituting (72) into (68) and replacing $T_i$ with $T_0$ we find

$$\tau = \frac{\bar{R}_v T_0^2}{d_g \mathcal{L}} \frac{Pe_m c_a}{1 - \bar{c}_a}. \tag{75}$$

This expression shows that the interfacial temperature profile is linear, quadratic, or exponential when the air concentration profile is constant, linear, or exponential, respectively. This is illustrated in Fig. 11, which shows that the analytical estimates for the interfacial temperature agree well with numerics. These results are in stark contrast with the very common assumption of linear temperature profile made in modeling heat pipes. While the temperature profile may indeed be linear in experiments with relatively small $\Gamma_g$ and conducted at atmospheric pressure, the condition $Pe_m \ll \Gamma_g^{-1}$ will most certainly not be satisfied for high-aspect-ratio heat pipes operating at low $\bar{c}_a$, where the temperature profile will be exponential. Similarly, these results show the limitations of linear stability analysis of this flow [27] which assumed $\tau$ to be constant. In fact, the discrepancy between the numerical results and predictions of the linear stability analysis in this limit found in Fig. 5 are likely due in part to the deviation of $\tau$ from a constant. Another reason for the discrepancy is that the linear stability analysis breaks down at low $\bar{c}_a$ due to the wavelength of the instability becoming comparable to the system size $L$.

Whether the temperature profile is linear or not, the mean value of the interfacial temperature gradient can be found by substituting $c_a = \bar{c}_a$ into (75), which yields

$$\bar{\tau} \propto \frac{\bar{c}_a}{1 - \bar{c}_a} Pe_m = \frac{\bar{c}_a}{1 - \bar{c}_a} \frac{J_0 d_g}{m_v^1 n_g D} \propto \frac{\bar{c}_a}{1 - \bar{c}_a} J_0, \tag{76}$$

where $n_g D = p_g D / (\bar{R} T_0)$ is independent of $\bar{c}_a$ and $J_0$ can be obtained using the expression (62). At low air concentration,
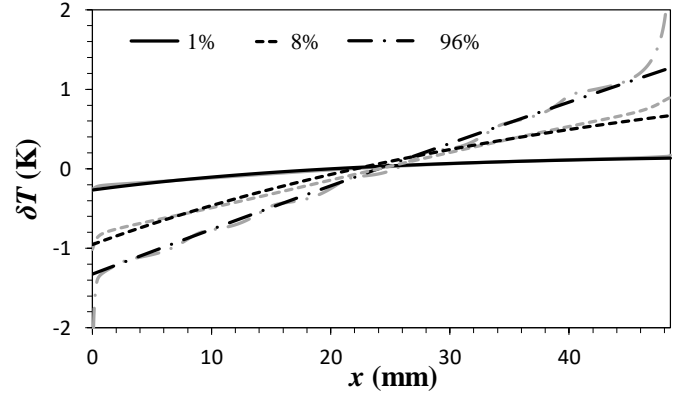


Figure 11: Interfacial temperature at $\Delta T = 10$ K with different $\bar{c}_a$. The variation $\delta T_i = T_i - T_0$ about the mean is plotted. Numerical and analytical results are represented by gray and black lines, respectively.

thermal resistance is dominated by the contribution $Z_o$, which is independent of $\bar{c}_a$. Hence $J_0$ is also independent of $\bar{c}_a$ and $\bar{\tau} \propto \bar{c}_a \Delta T / L$. In particular, in the limit $\bar{c}_a \to 0$ we also have $\bar{\tau} \to 0$, consistent with our previous finding [19]. At high air concentration, thermal resistance is dominated by the contribution $Z_d$ given by (71). Hence, $J_0 \propto (\Delta T / L)(1 - \bar{c}_a)/\bar{c}_a$, such that $\bar{\tau} \propto \Delta T / L$ is independent of $\bar{c}_a$. This is consistent with the results of both numerical simulations [17] and experiments [25] which found that the interfacial velocity (and hence thermocapillary stresses) remain essentially constant for $\bar{c}_a \gtrsim 0.14$.

Fig. 12 shows that our theoretical predictions match the numerical results for the interfacial temperature gradient at low $\bar{c}_a$, but overestimate them at high $\bar{c}_a$. The reason is the breakdown of our assumption that conductive and convective heat transfer in the liquid layer is negligible. Numerical simulations show that $\bar{\tau} = \Delta T / L$ only at sufficiently low $\Delta T$. At higher $\Delta T$ we have [16]

$$\frac{\Delta T}{L} \approx \frac{1 + a\bar{\tau}^2}{1 + b\bar{\tau}^2} \bar{\tau} \geq \bar{\tau}, \tag{77}$$

where $a$ and $b \leq a$ are parameters that dependent on $\bar{c}_a$ and the geometry of the liquid layer. In particular, the data in Fig. 12 corresponds to numerical simulations for $\Delta T = 10$ K, where strong convection in the (relatively thick) layer makes the relationship between $\bar{\tau}$ and $\Delta T / L$ nonlinear. For thin liquid layers characteristic of, e.g., heat pipes, both conductive and convective heat transfer in the liquid phase can be ignored, so our theoretical description should be quite accurate over the entire range of $\bar{c}_a$.

In conclusion of this section, let us return to the issue of validity of the one-dimensional solution (52) for the concentration profile. The value of $\epsilon$ can be easily estimated in the practically important case $\bar{c}_a \to 0$. In this limit, the gas velocity is much larger than the liquid velocity, so the flow profile in the gas layer in the central region is nearly parabolic and symmetric about its mid-height, so $\max_\zeta |\bar{u}_x| = |\bar{u}_x|$. Furthermore, diffusive resistance $Z_d$ is negligible and, since the wall material (fused quartz) is a far better thermal conductor than the liquid (silicone oil), $Z_g \approx Z_o \sim d_l/k_l$. Substituting (59) and (62) into (53)
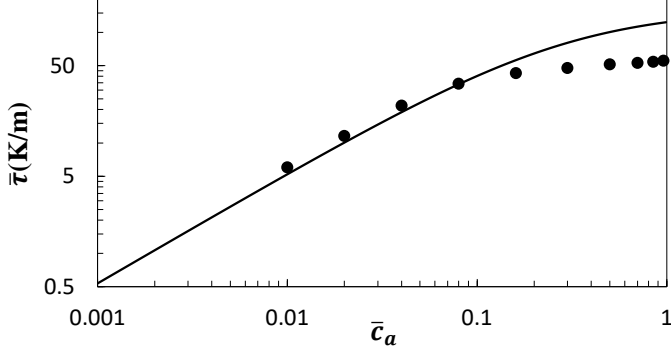
Figure 12: The mean value of the interfacial temperature gradient $\bar{\tau}$ as a function of the average air concentration $\bar{c}_a$ at $\Delta T = 10$ K. Numerical and analytical results are represented by solid line and symbols, respectively.

and (56) we find

$$Pe_r = Pe_m = \frac{\Delta T d_g}{m_v^1 n_g D \mathcal{L} Z_g} \sim \frac{d_g}{d_l} \frac{\mu_l}{\mu_v} Sc_v E, \qquad (78)$$

where $Sc_v = \nu_v/D$ is the Schmidt number for the vapor and

$$E = \frac{k_l \Delta T}{\mu_l \mathcal{L}} \qquad (79)$$

is the evaporation number [30]. The analytical solution (52) provides a good approximation when $\epsilon \ll 1$, which is equivalent to

$$\Delta T \ll Sc_v^{-1} \frac{d_l}{d_g} \frac{\mu_v \mathcal{L}}{k_l} \approx 27 \text{ K}. \qquad (80)$$

In particular, for $\Delta T = 10$ K, Eq. (78) yields $Pe_r = Pe_m = 0.37$, which is very close to the value (0.4) obtained numerically (cf. Fig. 6).

In the opposite limit (e.g., at atmospheric conditions), diffusive resistance dominates, $Z_g \approx Z_d$, so with the help of (71) we find

$$Pe_m \approx \frac{\Delta T d_g}{\mathcal{L} Z_g} = \frac{1 - \bar{c}_a}{\bar{c}_a} \frac{\mathcal{L}}{\bar{R}_v T_0} \frac{d_g}{L} \frac{\Delta T}{T_0}, \qquad (81)$$

which, as Fig. 6 illustrates, vanishes as $\bar{c}_a \to 1$. The recirculation component of the flow in this limit can be estimated by balancing the viscous and thermocapillary stresses [17]

$$\max_\zeta \tilde{u}_x \approx \frac{1}{4} \frac{\gamma d_l}{\mu_l} \bar{\tau} \sim \frac{1}{4} \frac{\gamma d_l \Delta T}{\mu_l L}, \qquad (82)$$

which yields

$$Pe_r \sim \frac{1}{4} \frac{\gamma d_l d_g \Delta T}{\mu_l L D} = \frac{1}{4} \frac{1}{1 - \bar{c}_a} \frac{\gamma d_l d_g \Delta T p_v^0}{\mu_l L D_0 p_0}. \qquad (83)$$

As our estimate shows and numerical data presented in Fig. 6 confirms, $Pe_r$ diverges at $\bar{c}_a \to 1$, however the product of the two Peclet numbers approaches a constant

$$\epsilon \sim \frac{1}{4} \frac{\gamma d_l \mathcal{L}}{\mu_l \bar{R}_v D_0} \frac{p_v^0}{p_0} \frac{d_g^2}{L^2} \frac{\Delta T^2}{T_0^2}. \qquad (84)$$

In particular, we find that $\epsilon \ll 1$ when

$$\Delta T \ll 2 \frac{L}{d_g} \left[ \frac{\mu_l \bar{R}_v D_0}{\gamma d_l \mathcal{L}} \frac{p_0}{p_v^0} \right]^{1/2} T_0 \approx 34 \text{ K}. \qquad (85)$$

Note that the estimate (82) and hence (83) and (85) is conservative since, for $\Delta T \gtrsim 10$ K, the interfacial temperature gradient $\bar{\tau}$ is only a fraction of $\Delta T/L$ (a third to a quarter for $\bar{c}_a = 0.96$ [16]).

To sum up, for the silicone oil studied here, our estimates suggest that the one-dimensional approximation (52) should hold for any $\bar{c}_a$ as long as $\Delta T$ is around 10 K or less; the more general expression (54) has to be used for larger temperature gradients. Indeed, our numerical simulations show that the concentration profile develops noticeable variation in the vertical direction only for $\Delta T \gtrsim 10$ K. For comparison, the onset of convection pattern (SMC regime) at atmospheric conditions corresponds to $\Delta T \approx 4$ K in this geometry [16, 25]. It should be noted that some of our assumptions (most notably, that phase change at the interface is negligible) start to break down when convection pattern develops, which leads to spatial modulation developing on top of the (nearly linear) interfacial temperature profile (see, e.g., Fig. 5 in Ref. [16]).

## 5. Summary

We have formulated and implemented numerically a generalized two-sided transport model for two-phase flow of non-isothermal volatile fluids which accounts for momentum, mass, and heat transport in both phases, as well as phase change at the liquid-gas interface. The model is valid for arbitrary composition of the gas phase, described in terms of the average concentration of air $\bar{c}_a$ here. It has been thoroughly validated by comparing its predictions both against other numerical models in the limiting cases $\bar{c}_a \to 1$ and $\bar{c}_a \to 0$ and against experimental observations [25] and linear stability analysis [27] over the entire range of $\bar{c}_a$, . The model has been used to study the flow of low viscosity silicone oil confined in a sealed cavity and driven by a horizontal temperature gradient. In particular, we have investigated how the flow changes when the pressure inside the cavity, and with it the composition of the gas layer, changes. Numerical simulations reported here and elsewhere [16, 19, 17] show that the flow in the liquid layer is very different between the limits $\bar{c}_a \to 0$ and $\bar{c}_a \to 1$.

In order to understand these differences, we have developed a simplified transport model which showed that, somewhat surprisingly, for $\bar{c}_a > 0$, it is the gas layer that controls the flow in the liquid layer. More specifically, mass transport in the gas layer determines its local composition, the interfacial temperature, and hence the thermocapillary stresses which drive the flow in the liquid layer. Quite interestingly, we found that, in high-aspect ratio geometries (such as the interior of a heat pipe), the mass transport in the gas layer remains diffusion-dominated and hence independent of the flow, even in the limit when the traditionally defined Peclet number, which corresponds to our $Pe_r$, becomes large compared to unity. We have shown that

mass transport is in fact described by two different Peclet numbers – $Pe_r$ which describes recirculation, and $Pe_m$ which describes the mean flow – and that it is the product $\epsilon = Pe_r Pe_m$ that controls whether mass transport in the gas phase is dominated by diffusion or advection. We have also shown that, for small to moderate imposed temperature gradients, $\epsilon$ remains small regardless of the composition of the gas phase.

Our simplified transport model has not just qualitatively explained a range of numerical and experimental results, it was also found to be capable of producing quantitatively accurate predictions. For instance, it correctly predicted the concentration profile in the gas phase and the interfacial temperature profile over the entire range of $\bar{c}_a$. In particular, we found that the interfacial temperature profile is generally exponential, not linear as assumed by many one-sided transport models of two-phase flows. The assumption of linearity can be justified in the limit $\bar{c}_a \rightarrow 1$ (*e.g.*, at atmospheric conditions), but not in the practically important limit $\bar{c}_a \rightarrow 0$. Furthermore, the model correctly predicted the dependence of the net vapor flux (and hence the net heat flux) and the average interfacial temperature gradient (and hence thermocapillary stress) on $\bar{c}_a$. Finally, it is worth pointing out that although the simplified transport model is formally valid only in the steady unicellular flow regime, it was found to give reasonably accurate predictions even when convection rolls appears.

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