



# On the critical thickness and wavelength of a condensing thin liquid film in a binary vapor mixture system



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## ABSTRACT

We study the linear stability of a condensing thin liquid film of a binary vapor mixture by solving directly the bulk equations of the gas phase. The boundary layer of a finite thickness is introduced above the liquid film, within which the variables are disturbed. The dynamics of the liquid film is described by the long-wave equation. The neutral stability condition predicts the existence of a critical thickness below which a flat film is stable due to the mass gain effect. However, if we consider the thickening of the liquid film by condensation, the relative neutral stability can be defined such that the growth rate of a disturbance is equal to that of the basic film thickness. The critical thickness and wavenumber obtained from the relative neutral stability condition significantly change from the original ones. Employing the asymptotic analysis for large wavenumbers, the critical thickness and wavelength are numerically calculated for the water–ethanol system. Their dependence on the boundary layer thickness, temperature and ambient vapor concentration is investigated. The critical wavelength obtained from our theory has the same trend in the temperature and concentration as the initial drop distance observed in the experiment.

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## 1. Introduction

Since the first appearance of Marangoni dropwise condensation [1], a number of experimental studies have been devoted to condensation of binary vapor mixtures to observe nonfilmwise condensation [2–5]. In contrast, there are few theoretical works on Marangoni dropwise condensation. Among them, Hijikata et al. [6] investigated the stability of a flat condensate of a binary vapor mixture. Recently, we modeled the dynamics of a condensate liquid film of a binary vapor mixture using the long-wave approximation and the linear stability analysis was carried out based on this model [7]. In both studies the vapor concentration gradient at the liquid–vapor interface in the vertical direction is assumed to be proportional to the concentration difference between the ambient vapor and interface, which is an analogue of well-known Newton's law of cooling for heat transfer. However, if a flat condensate is disturbed the vapor concentration also fluctuates along the liquid–vapor interface. This fluctuation makes the vapor concentration diffuse along the interface. The diffusion of the vapor concentration was disregarded in the above models [6,7].

In the modeling of an evaporating liquid film, Sultan et al. [8,9] took into account static diffusion of the vapor in the long-wave equation describing the spatio-temporal evolution of the film thickness [10,11] by solving the Laplace equation for the vapor concentration. Recently, we proposed a model including advective effects due to the evaporation flow of the gas [12]. In this model, a rather ad hoc model is applied to the vertical diffusion term of the mass transport equation.

In this work, we study the linear stability of a condensing thin liquid film of a binary vapor mixture by solving directly the bulk equations of the gas phase without applying any model to the vapor concentration. As in the previous studies [7,12,13], the boundary layer of a finite thickness is introduced above the liquid layer, within which the variables are disturbed. Then the disturbances in the gas phase are coupled with that of the film thickness of the condensate, which is described by the long-wave equation [7,11]. Here for simplicity the frozen-time approach is adopted, where the linear stability analysis is carried out against the 'hypothetically motionless' interface at each instance or film thickness (of course in reality the interface is moving owing to condensation). As a result, the neutral stability condition is obtained by setting the growth rate of the disturbances to zero. Since the effect of mass gain is stabilizing, there is a critical thickness below which a flat film is stable. However, if the condensate is sufficiently thin

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(about 1  $\mu\text{m}$ ), the temporal evolution of the average film thickness becomes comparable to that of the disturbances. In this case, it is insufficient to address only the growth rate of the disturbances: we have to consider the evolution of the ratio of the amplitude of the disturbance of the film thickness to the average film thickness. If this ratio temporarily increases (decreases), it follows that the disturbance grows (decays) relatively to the average film thickening. Thus the relative neutral stability can be defined such that the time derivative of this ratio vanishes. We will show that this definition of the neutral stability significantly changes the critical thickness and wavenumber compared to the original ones. The dependence of the critical thickness and wavelength on the boundary layer thickness, temperature and ambient vapor concentration is investigated. A comparison is made with the experimental result for the water–ethanol system [5].

## 2. Formulation

We consider a two-dimensional condensate liquid film of a binary vapor mixture on a uniformly-cooled horizontal wall, as depicted in Fig. 1. The  $x$ -axis coincides with the liquid–solid boundary and the  $z$ -axis points vertically upward to the wall. The mass fraction of the surrounding vapor is fixed to  $c_0$  at some distance  $\delta$  from the average height of the liquid–gas interface; here  $\delta$  is assumed to be much larger than the thickness of the film  $h$  so that the former is never affected by the variation of the latter. We scale lengths, time, velocities and pressure of the gas phase by  $d_l$ ,  $d_l^2/\nu_l$ ,  $\nu_l/d_l$  and  $\rho_v \nu_l^2/d_l^2$ , respectively, where  $d_l$ ,  $\nu_l$ , and  $\rho_v$  are the initial film thickness, the kinematic viscosity of the liquid, and the vapor density, respectively. The dimensionless governing equations in the gas phase are the continuity, Navier–Stokes, and mass transport equations:

$$\nabla \cdot \mathbf{v}_v = 0, \quad (1a)$$

$$\partial_t \mathbf{v}_v + \mathbf{v}_v \cdot \nabla \mathbf{v}_v = -\nabla p_v + \nu \nabla^2 \mathbf{v}_v - G \mathbf{e}_z, \quad (1b)$$

$$\Sigma_v (\partial_t c_v + \mathbf{v}_v \cdot \nabla c_v) = \nu \nabla^2 c_v, \quad (1c)$$

where  $\mathbf{v}_v = (u_v, w_v)$ ,  $p_v$ , and  $c_v$  are the velocity, pressure, and mass concentration of the component having a lower boiling point in the gas, respectively. The differential operator is  $\nabla \equiv (\partial_x, \partial_z)$ . The symbols  $\nu = \nu_v/\nu_l$ ,  $G = g d_l^3/\nu_l^2$ , and  $\Sigma_v = \nu_v/D_v$  denote the kinematic viscosity ratio, the Galileo number, and the Schmidt number of the vapor, where  $\nu_v$ ,  $g$ , and  $D_v$  are the kinematic viscosity, the gravitational acceleration, and mass diffusivity of the gas phase, and  $\mathbf{e}_z$  is the unit vector in the  $z$  direction. In Eqs. (1a) and (1b) we have neglected the concentration dependence of  $\rho_v$  and  $\nu_v$  considering a small concentration difference between the ambient vapor and liquid–gas interface or a small condensation rate. In Eq. (1b) buoyancy has been neglected; however, we shall discuss its influence on

the stability of the liquid film in Appendix A. In addition we have neglected also thermo-diffusion and diffusion-thermo effects.

For the liquid phase, the following long-wave equation for the film thickness  $h(x, t)$  and mass transport equation hold [11]:

$$\partial_t h = \partial_x \left\{ \frac{h^3}{3} (G \partial_x h - S \partial_{xxx} h) - \frac{h^2}{2} Ma \partial_x T_l \right\} + \Gamma J, \quad (1d)$$

$$\Sigma_l (\partial_t c_l + \mathbf{v}_l \cdot \nabla c_l) = \nabla^2 c_l, \quad (1e)$$

where the subscripts  $l$  and  $l$  refer to the liquid and interfacial counterparts of the same symbol. The dimensionless numbers are defined by

$$S = \frac{\sigma d_l}{\eta_l \nu_l}, \quad Ma = -\frac{d_l L \rho_v D_v}{\eta_l \nu_l \lambda_l} \left( \frac{\partial \sigma}{\partial T} + \frac{\partial \sigma}{\partial c_l} \frac{\partial c_l}{\partial T} \right), \quad \Gamma = \frac{\rho_v D_v}{\eta_l}, \quad (2)$$

which characterize the effects of surface tension, thermo- and soluto-capillarity, and mass gain, respectively. Here,  $\sigma$  is the surface tension,  $\eta_l$  is the dynamic viscosity of the liquid,  $L$  is the latent heat and  $\lambda_l$  is the thermal conductivity of the liquid. The condensation mass flux  $J(x, t)$  and interface temperature  $T_l(x, t)$  have been nondimensionalized by  $\rho_v D_v/d_l$  and  $L \rho_v D_v/\lambda_l$ . We assume the linear temperature profile across the liquid film, which is a natural consequence of the long-wave approximation (see for example Ref. [11]). Since the heat flux in the liquid balances with the latent heat (the heat flux in the gas is negligible), the interface temperature can be written as

$$T_l = Jh. \quad (3)$$

where the wall temperature has been assumed to be constant and set to 0. For simplicity, we shall neglect the gravitational term in Eq. (1d) by setting  $G = 0$ , which is normally justified for very thin films (say a few micrometers' thickness).

At the top of the gas boundary layer ( $z = 1 + \Delta$ ;  $\Delta = \delta/d_l$ ), we impose the stress-free and constant pressure and concentration boundary conditions,

$$\partial_z u_v + \partial_x w_v = 0, \quad p_v = 0, \quad c_v = c_0, \quad (4a)$$

where the dimensionless pressure has been set to 0 there. At the wall ( $z = 0$ ), the reflecting concentration boundary condition holds

$$\partial_z c_l = 0. \quad (4b)$$

At the liquid–gas interface ( $z = h(x, t)$ ), the conservation laws of the total mass and that of one component and the continuity of the tangential velocity along the interface read

$$-vJ = \Sigma_v \mathbf{v}_v \cdot \mathbf{n}, \quad (4c)$$

$$\nabla c_l \cdot \mathbf{n} + c_l J = \nabla c_v \cdot \mathbf{n} + c_v J. \quad (4d)$$

$$\mathbf{v}_l \cdot \mathbf{t} = \mathbf{v}_v \cdot \mathbf{t}. \quad (4e)$$

Here  $\mathbf{n} = (-\partial_x h, 1)/\sqrt{1 + (\partial_x h)^2}$  and  $\mathbf{t} = (1, \partial_x h)/\sqrt{1 + (\partial_x h)^2}$  are the unit vectors normal and tangent to the interface. In Eq. (4c) we have neglected the interface velocity compared to the vertical vapor velocity. As in our previous study [7], we employ the linearized local thermodynamic equilibrium conditions at the interface:

$$c_{ll} = c_{lr} + c'_l (T_l - T_r), \quad (4f)$$

$$c_{vl} = c_{vr} + c'_v (T_l - T_r), \quad (4g)$$

where  $c_{lr}$  and  $c_{vr}$  are the liquid and vapor concentrations in the thermodynamic equilibrium state at the reference temperature  $T_r$ , and  $c'_l$  and  $c'_v$  are the temperature derivatives of the liquid and vapor concentrations. Although the reference temperature was set to the wall one in our previous study [7], it will be found to be more convenient to fix the initial interface temperature rather than the wall one in the present analysis. For this reason in this study we shall set the reference temperature to the initial interface one.

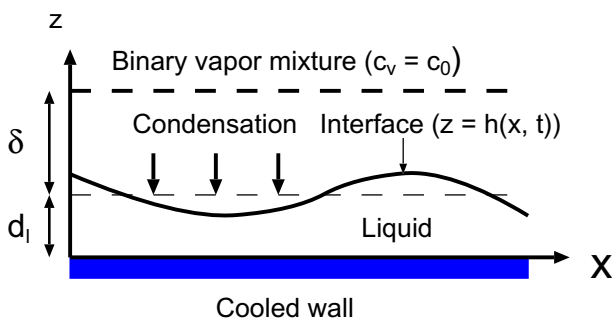


Fig. 1. Sketch of a condensate liquid film of a binary vapor mixture on a cooled horizontal surface.

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