International Journal of Heat and Mass Transfer 58 (2013) 413–419

Contents lists available at SciVerse ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Stability of a condensing liquid film in a binary vapor mixture system

Kentaro Kanatani *

Faculty of Engineering, Yokohama National University, 79-5 Tokiwadai, Hodogaya, Yokohama 240-8501, Japan

ARTICLE INFO

Article history:
Received 9 April 2012
Received in revised form 6 August 2012
Accepted 15 November 2012
Available online 12 December 2012

Keywords:
Marangoni dropwise condensation
Binary mixture
Stability analysis
Thin liquid film

ABSTRACT

We study stability of a condensing liquid film of a binary vapor mixture. When a binary vapor mixture of some kind is cooled on a substrate, a condensing liquid film emerges to take an inhomogeneous form such as a droplet one due to the solutal Marangoni effect. In order to analyze this phenomenon, we apply the long-wave approximation to the condensing liquid film and derive a nonlinear partial differential equation describing the spatio-temporal evolution of the film thickness. An interfacial boundary condition taking account of an effect of mass gain of the liquid film is adopted. Based on this model, we perform a linear stability analysis around a flat-film solution. We obtain an evolution equation of the amplitude of a disturbance, from which the cutoff and fastest growth wavenumbers are deduced. The maximum value of the cutoff wavenumber relative to the film thickness and its film thickness are estimated for water–ethanol mixture at atmospheric pressure. We numerically verify the long-wave nature of the instability of the condensate liquid film in this system. A significant difference in their values is found for low-ethanol fractions of the ambient vapor whether or not the temperature dependence of the mass transfer coefficient is considered. The wavenumber of a pattern of the liquid film observed in the experiment has the same parameter dependence as that of the fastest growth wavenumber.

1. Introduction

When a binary vapor mixture of miscible liquids of some kind is condensed on a cooled substrate, the condensate liquid film often takes an inhomogeneous state such as a dropwise form. This phenomenon was first presented by Mirkovich and Missen [1]. Up to the present, a number of experiments on binary vapor condensation have been carried out and demonstrated nonfilmwise condensation (see Table 1 of Ref. [2] or Ref. [3]).

Ford and Missen [4] expressed the instability criterion of the condensate liquid film as

$$\frac{\partial \sigma}{\partial h} > 0, \tag{1}$$

where σ and h represent the surface tension and film thickness. Eq. (1) corresponds to the case that the surface tension is stronger on a thicker part of the condensate than on a thinner one. In such a case, a stronger surface tension on a thicker portion pulls the liquid away from a thinner one and the deviation from the flat-film state is amplified. Since the surface tension is a function of temperature and concentration,

$$\frac{\partial \sigma}{\partial h} = \left\{ \frac{\partial \sigma}{\partial T_I} + \frac{\partial \sigma}{\partial c} \left(\frac{\partial c}{\partial T_I} \right)_{\mathrm{sat}} \right\} \frac{\partial T_I}{\partial h}, \tag{2}$$

where T_I and c are the interface temperature and mass fraction of the component having a lower boiling point. The local thermodynamic equilibrium state is assumed at the liquid–vapor interface in evaluating $(\partial c/\partial T_I)_{\mathrm{sat}}$ in Eq. (2), which is negative by the definition, unless the surface tension has a minimum or maximum point at an intermediate concentration. Since the substrate is cooled, the condition $\partial T_I/\partial h > 0$ always holds. Furthermore, in most cases the dependence of surface tension on temperature is negligible compared to that on concentration. Therefore, the necessary condition for the condensate liquid film to be destabilized by the solutal Marangoni effect is

$$\frac{\partial \sigma}{\partial c} < 0, \tag{3}$$

which indicates that the surface tension of the high-boiling-point component is larger than that of the low-boiling-point component.

Stability of a flat-film state of the condensing liquid film in such a system was investigated by Hijikata et al. [5] However, they disregarded two interfacial boundary conditions necessary for condensation of binary mixtures: mass balance of the total mixture and that of a one component of the mixture. The first boundary condition entails mass gain of the liquid during the condensation process. It follows that Ref. [5] did not consider this effect. However, the experiments [4,6] confirmed that there always exists a very thin layer of the liquid (1 μm order) between the condensate droplets, preventing the liquid film from rupturing. This observation is different from the theoretical consequence that an unstable

---

* Tel./fax: +81 45 339 3897.
E-mail address: kentaro@ynu.ac.jp

0017-9310/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijheatmasstransfer.2012.11.060

liquid film subjected to only the destabilizing (thermal) Marangoni effect and the stabilizing gravity effect inevitably tends to rupture [7]. Moreover, since the length scale at which intermolecular forces between the liquid film and the solid surface influence the dynamics of the liquid–gas interface is about 10 to 100 nm, it is found that the aforementioned very thin liquid layer between the droplets is not induced by any intermolecular forces, or is essentially irrelevant to the wettability of the solid surface. The above consideration implies that the effect of mass gain by condensation balances with that of solutocapillarity in such a length scale.

The second boundary condition which Hijikata et al. [5] ignored yields a relationship between the condensation mass flux and the concentration field. Since the liquid and vapor concentrations at the interface are determined by the interface temperature via the phase equilibrium condition, the mass flux becomes a function of the interface temperature. In Ref. [5] a similar relationship between the mass flux and the interface temperature is also formulated, assuming that the mass transfer coefficient is independent of temperature. However, if we allow for the second boundary condition, it can be shown that the mass transfer coefficient depends on temperature. Therefore, when the interface is irregular the value of the mass transfer coefficient locally varies owing to the variation of the interface temperature.

In this work, we first model the dynamics of a condensing liquid film of a binary vapor mixture. Applying the long-wave approximation [8] to the condensate liquid film, a nonlinear partial differential equation describing the spatio-temporal evolution of the film thickness is derived. The advantages of the use of such a low-dimensional approximate model are twofold: (i) to obtain analytical expressions for the dispersion relation of disturbances and for the stability criterion; and (ii) to reduce the computational amount in seeking time-dependent solutions at the strongly nonlinear regime. In the present paper, we focus on the linear stability of the flat-film state of the condensate, pertaining to the former advantage, and nonlinear calculations of the model, pertaining to the latter, shall be done subsequently. We incorporate the two boundary conditions mentioned above in deriving the model. The first boundary condition makes the basic state with a flat liquid–vapor interface time-dependent. Most studies on the stability of evaporating or condensing liquid layers (e.g. Ref. [9]) have treated the basic state as a steady state, assuming that the variation of the basic state is sufficiently slow compared to the growth rate of disturbances (the quasi-steady hypothesis). However, in the framework of the long-wave model the evolution equation of the amplitude of disturbances is easily integrable even when the average thickness of the liquid layer is varying by evaporation or condensation [10]. Reference [10] distinguished an algebraic behavior of the amplitude, arising from the mass loss or gain, from an exponential one and neglected the former. Thus the linear stability property provided by the long-wave model is more global in time. We also examine the influence of the dependence of the mass transfer coefficient on the interface temperature resulting from the second boundary condition.

2. Formulation

2.1. Governing equations

We consider a two-dimensional condensate liquid film of a binary vapor mixture on a horizontal wall uniformly cooled at a temperature T_w, 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 $z = H$; here we assume that H is much larger than the thickness of the condensate and never affected by the variation of the latter. The governing equations of the liquid phase are the continuity, Navier–Stokes, energy and mass transport equations:

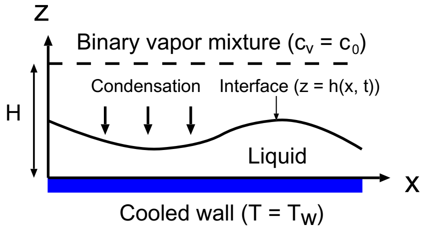

Fig. 1. Sketch of a condensate liquid film of a binary vapor mixture on a cooled horizontal surface.

$$\nabla \cdot \boldsymbol{v} = 0, \tag{4}$$

$$\rho(\boldsymbol{v}_t + \boldsymbol{v} \cdot \nabla \boldsymbol{v}) = -\nabla p + \eta \nabla^2 \boldsymbol{v} - \rho g \boldsymbol{e}_z, \tag{5}$$

$$T_t + \boldsymbol{v} \cdot \nabla T = \kappa \nabla^2 T, \tag{6}$$

$$c_t + \boldsymbol{v} \cdot \nabla c = D \nabla^2 c, \tag{7}$$

where $\boldsymbol{v}$, p, T and c are the velocity, pressure, temperature and mass concentration fields, respectively, inside the condensate. The differential operator is $\nabla \equiv (\partial_x, \partial_z)$. The symbols ρ, η, κ and D denote the density, dynamic viscosity, thermal diffusivity and mass diffusivity of the liquid, respectively, all assumed to be constant, g the gravitational acceleration, and $\boldsymbol{e}_z$ the unit vector in the z direction. Here we have neglected buoyancy, thermo-diffusion and diffusion-thermo effects.

We impose the no-slip, no-penetration, constant temperature and reflecting concentration boundary conditions at the wall,

$$\boldsymbol{v} = \boldsymbol{0}, \quad T = T_w, \quad c_z = 0 \quad \text{at} \quad z = 0. \tag{8}$$

At the liquid–vapor interface ($z = h(x,t)$), the mass balance condition is expressed as

$$J = \rho(\boldsymbol{v}_I \cdot \boldsymbol{n} - \boldsymbol{v} \cdot \boldsymbol{n}), \tag{9}$$

where J is the condensation mass flux, $\boldsymbol{n}$ the upward-directed unit vector normal to the interface,

$$\boldsymbol{n} = \frac{(-h_x, 1)}{\sqrt{1 + (h_x)^2}}, \tag{10}$$

and $\boldsymbol{v}_I$ the interface velocity, which satisfies the kinematic condition,

$$\boldsymbol{v}_I \cdot \boldsymbol{n} = \frac{h_t}{\sqrt{1 + (h_x)^2}}. \tag{11}$$

Eq. (9) was not taken into account in Ref. [5]. We assume the continuity of the interfacial stress and energy:

$$(p - p_0)\boldsymbol{n} - 2\eta \boldsymbol{E} \cdot \boldsymbol{n} + 2\sigma K \boldsymbol{n} + (\boldsymbol{t} \cdot \nabla \sigma)\boldsymbol{t} = \boldsymbol{0}, \tag{12}$$

$$JL = \lambda \nabla T \cdot \boldsymbol{n}, \tag{13}$$

where p_0 is the ambient vapor pressure, $\boldsymbol{E}$ the rate-of-strain tensor in the liquid phase, σ the surface tension, K the mean curvature of the interface,

$$2K = \frac{h_{xx}}{\{1 + (h_x)^2\}^{3/2}}, \tag{14}$$

$\boldsymbol{t}$ the unit tangent vector to the interface,

$$\boldsymbol{t} = \frac{(1, h_x)}{\sqrt{1 + (h_x)^2}}, \tag{15}$$

L the latent heat and λ the thermal conductivity of the liquid. Note that the effects of vapor recoil, stress induced by the vapor motion,