Stability analysis of the evaporation of a binary liquid into an inert gas, considering solute/thermal and gravity/surface tension effects

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Summary

A stability analysis is performed for a binary liquid of which solely the solute evaporates into an inert gas. After having determined a stationary reference solution, stability curves are calculated for the solutal Marangoni number. The critical liquid thickness and the corresponding mass fraction are calculated as a function of the thickness of the gas layer. Also, a comparison is made with a purely solutal Pearson's model, using a suitably defined Biot number.

Purpose

The evaporation of a horizontal liquid layer is an endothermic process, resulting in the cooling of its surface. This gives rise to a vertical temperature gradient across the liquid layer. Evaporation of mixtures also induces concentration gradients across the layer. Since the density and surface tension of the liquid depend on temperature and concentration, evaporation can thus destabilize the liquid layer [1-3]. The instability due to gravity and density variations is usually called the Rayleigh-Bénard instability, while Marangoni-Bénard instability refers to the case where the surface tension variations induce motion in the fluid. When both effects couple, the name "Rayleigh-Bénard-Marangoni instabilities" is used. If the concentration dependence of the density and surface tension causes the onset of convection, the corresponding instabilities can be called "solutal" Rayleigh-Bénard-Marangoni instabilities. Similarly, "thermal" instabilities result from the influence of temperature. In this work, the system consists of a binary liquid of which only the solute evaporates. The gas above the layer is a mixture of the evaporated solute and an inert gas which is not absorbed in the liquid phase. The aim of the work is to perform a linear stability analysis of this system. This will be done by adding small perturbations to a reference state. In the gas phase, the reference concentration and temperature profiles are non-linear and stationary, while in the liquid phase the reference profiles of the temperature and the concentration are linear and stationary.

Methodology

The system comprises a binary liquid layer in contact with a binary gas layer. The liquid layer consists of a solvent and a solute, with mass fraction C, in dilute concentration. The Soret effect is considered in the liquid phase [4]. The gas layer consists of air and the vapour of the solute component. The bottom of the liquid layer is a rigid plate with a fixed temperature and the mass fraction is also assumed to be fixed at that boundary. The latter condition can be approached by a thin porous layer under which a fluid with fixed mass fraction is circulated, maintaining thus the mass fraction roughly constant at the bottom boundary of the system. At the top of the gas layer, the temperature has the same value as at the bottom of the system, while the mass fraction of the solute compound is assumed to vanish. No net horizontal flow is considered in this system. The Boussinesq approximation is used in both the liquid and the gas. In particular, the density variations with temperature and mass fraction are taken into account only in the buoyancy terms of the momentum equations. The usual equations for the momentum, energy and species balances are used for both the liquid and gas phases. The conditions for evaporation are defined by local thermodynamic equilibrium and expressed by Henry's law [5]. The reference quasi-steady-state solution [6] is calculated and small perturbations with respect to this solution are considered. The corresponding (linear) equations are obtained. Then, a standard normal mode technique is introduced (with a decomposition of the perturbations into Fourier modes characterized by their growth rate σ and wave number k) and the final equations, which are solved by using a Tau-Chebyshev spectral method, are the following:

$$\left(D_z{}^4 - \left(2k^2 + Pr_l{}^{-1}\sigma\right)D_z{}^2 + \left(k^4 + k^2 Pr_l{}^{-1}\sigma\right)\right)w_l = Ra_l k^2 \theta_l + Rs_l Le_l k^2 c_l, \tag{1}$$

$$(D_z{}^2 - k^2 - \sigma)\theta_l = (T_{i,ref} - T_b)w_l, \tag{2}$$

$$(Le_l D_z{}^2 - Le_l k^2 - \sigma)c_l = (C_{i,ref,l} - C_b)w_l - Le_l \psi_s(D_z{}^2 - k^2)\theta_l, \tag{3}$$

$$\left(\nu D_z{}^4 - \left(2\nu k^2 + Pr_l{}^{-1}\sigma\right)D_z{}^2 + (\nu k^4 + k^2 Pr_l{}^{-1}\sigma) - \frac{J_{ref}}{\rho E}(D_z{}^3 - k^2 D_z)\right)w_g = \alpha Ra_l k^2 \theta_g + \varepsilon Rs_l Le_l k^2 c_g, \tag{4}$$

$$\left(\sigma + \frac{J_{ref}}{\rho E}D_z - \kappa\left(D_z{}^2 - k^2\right)\right)\theta_g = -\frac{\partial T_{g,ref}}{\partial z}w_g,$$ (5)

$$\left(\sigma + \frac{J_{ref}}{\rho E}D_z - DLe_l\left(D_z{}^2 - k^2\right)\right)c_g = -\frac{\partial C_{g,ref}}{\partial z}w_g$$ (6)

D_z means the derivative in the z-direction. J_{ref} is the evaporation flux in the reference state. D is the ratio of the gas diffusion coefficient to the liquid diffusion coefficient. The symbols $\rho, \kappa, \upsilon, \alpha, \varepsilon, \lambda, \mu$ denote the same ratios for respectively the density, the thermal diffusivity, the kinematic viscosity, the thermal expansion coefficient, the solutal expansion coefficient, the thermal conductivity and the dynamic viscosity. The symbol ψ_S quantifies the Soret effect. The perturbations in the liquid (subscript l) and in the gas (subscript g) for the vertical velocity, the temperature and the mass fraction of the solute are indicated by respectively w, θ and c. The symbol E ($=L/\theta c_{p,l}$) is a non-dimensional number that enters through the non-dimensionalisation of the evaporation flux. Setting this quantity to one, amounts to use the temperature scale equal to $\theta = L/c_{p,l}$. The boundary conditions used are the following:

At z = 0 (the bottom of the liquid phase):

$$D_z w_l = 0, \qquad w_l = 0, \qquad \theta_l = 0, \qquad c_l = 0.$$ (7)

At z = H (the top of the gas phase):

$$D_z w_g = 0, \qquad w_g = 0, \qquad \theta_g = 0, \qquad c_g = 0.$$ (8)

At z = 1 (the liquid-gas interface):

$$\theta_g = \theta_l, \quad -D_z\theta_l + \lambda D_z\theta_g = \frac{\rho}{1-\rho}(w_g - w_l), \quad D_z w_l = D_z w_g, \quad w_l = \rho w_g,$$ (9)

$$-\mu\left(k^2 w_g - D_z^2 w_g\right) + \left(k^2 w_l - D_z^2 w_l\right) + Ma\left(k^2\theta_l\right) + MsLe\left(k^2 c_l\right) = 0,$$ (10)

$$\frac{1}{1-\rho}(w_g - w_l) = -\frac{Le_l D\theta_C}{1-C_g}D_z c_g,$$ (11)

$$\frac{\rho}{1-\rho}(w_g - w_l) = -\frac{Le_l\theta_C}{1-C_l}\{D_z c_l + \psi_S D_z\theta_l\},$$ (12)

$$\frac{C_{i,g}}{M_g + C_{i,g}(1-M_g)} = \frac{C_{i,l}}{M_l + C_{i,l}(1-M_l)}\frac{K_e}{P_{tot}},$$ (13)

The physical meaning of conditions (7) and (8) is straightforward. Let us only stress that the last of equations (7) expresses that the mass fraction at the bottom is kept constant. At the interface, different equations in (9) express the temperature continuity, the energy conservation, the no slip condition and the mass-flux conservation. Equation (10) expresses the tangential stress balance. Equations (11) and (12) express respectively the non-absorption of air in the liquid and the non-evaporation of water. Finally, equation (13) is Henry's law expressed in terms of mass fractions. The following dimensionless numbers are used: the Prandtl number ($Pr_l = \nu_l/\kappa_l$), the thermal Rayleigh number ($Ra_l = \alpha_l g\theta d_l^3/\kappa_l\nu_l$), the solutal Rayleigh number ($Rs_l = \varepsilon_l g\theta_C d_l^3/D_l\nu_l$), the Lewis number ($Le_l = D_l/\kappa_l$), the thermal Marangoni number ($Ma = \gamma_T\theta d_l/\kappa_l\mu_l$) and the solutal Marangoni number ($Ms = \gamma_C\theta_C d_l/D_l\mu_l$). The symbols θ_C, M_g, M_l and H denote respectively the scaling factor of the mass fraction (=1), the ratio of the molar masses of the solute to the air, the ratio of the molar masses of the solute to the solvent and the ratio of the total height of the system to the liquid height. The symbols T_b and T_t express the temperatures at the bottom plate and at the top plate. The symbols C_b and C_t denote the same for the mass fractions. The symbols $T_{i,ref}$ and $C_{i,ref}$ are the values of the temperature and the mass fraction at the interface. For the mass fraction different values exist for the liquid phase and the gas phase (denoted by subscripts l and g). In equations 5 and 6, the derivatives of the reference solutions for respectively the temperature and the mass fraction are written. The reference solutions for the temperature and mass fractions in both the liquid and gas phase can be calculated from the energy and species balance considering a quasi-steady-state [6]. The equations in the liquid phase are straightforward and linear, while those in the gas phase are expressed by:

$$T_{g,ref} = \frac{(T_{i,ref}-T_t)e^{\left\{\frac{J_{ref}}{\kappa\rho E}(z-1)\right\}} + T_t - T_{i,ref}e^{\left\{\frac{J_{ref}}{\kappa\rho E}(H-1)\right\}}}{1 - e^{\left\{\frac{J_{ref}}{\kappa\rho E}(H-1)\right\}}}$$ (14)

$$C_{g,ref} = \frac{(C_{i,ref,g}-C_t)e^{\left\{\frac{J_{ref}}{DLe_l\rho E}(z-1)\right\}} + C_t - C_{i,ref,g}e^{\left\{\frac{J_{ref}}{DLe_l\rho E}(H-1)\right\}}}{1 - e^{\left\{\frac{J_{ref}}{DLe_l\rho E}(H-1)\right\}}}$$ (15)

By using the equivalents of equations (9), (11) to (13), but for the references state, the reference solutions for the temperature ($T_{i,ref}$), mass fraction ($C_{i,ref,l}$ and $C_{i,ref,g}$) and evaporation flux (J_{ref}) at the interface can be calculated.

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Results

The results presented hereafter concern a linear stability analysis and a comparison between the model used in this work and the one-sided model corresponding to the Pearson analysis [3].

Linear stability analysis

Let us first introduce the "actual" solutal Marangoni number, which is denoted by Ms* and which is defined by: $Ms^* = \frac{\gamma_C d_l (C_b - C_{i,ref,l})}{D_l \mu_l}$. Consider now the results of the analysis in the case of a binary mixture with a bottom mass fraction of ethanol in water equal to 0.1.
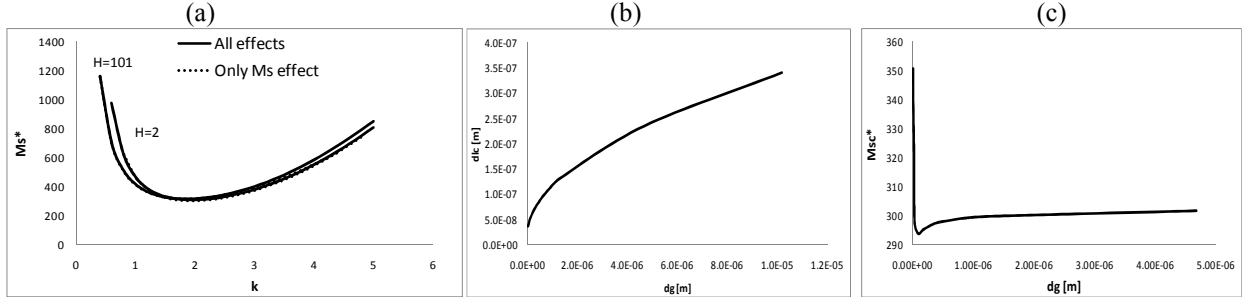
| (a) | (b) | (c) |

Figure 1: (a) Critical solutal Marangoni number as a function of the wave number for H=2 and H=101, considering either all effects or only the solutal Marangoni effect (dotted lines), (b) Critical liquid height as function of the gas thickness, (c) Critical Marangoni number as a function of the gas thickness (10% of ethanol in water)

Figure 1(a) presents the critical Marangoni number Ms* as a function of the wave number for H=2 and 101. In these calculations two cases are considered: the case where all effects are considered and the case where only the solutal Marangoni effect is considered. It can be seen from figure 1(a) that omitting the Rayleigh effect, the Soret effect and the thermal Marangoni effect has clearly no influence on the Ms* curve. This suggests that the solutal Marangoni effect is the most important one. This conclusion is valid for both the H values of 2 and 101.

For H=2 (considering all the effects), the critical Ms* number equals 304.2, to which corresponds a mass fraction difference of 0.074, a critical liquid height of 38.1 nm and a temperature difference of 0.15 K. The following entities can also be calculated at H=2: $Ma^*_{T,c} = 5.12 * 10^{-3}$, $Ra^*_{T,c} = 1.69 * 10^{-13}$ and $Ra^*_{C,c} = 4.71 * 10^{-9}$. Doing the same for H=101 gives a critical Ms* number of 314.5, with a mass fraction difference of 0.0027, a critical liquid height of 1.07μm and a temperature difference of 0.0063 K. For H=101 one can also calculate: $Ma^*_{T,c} = 5.93 * 10^{-3}$, $Ra^*_{T,c} = 1.53 * 10^{-10}$ and $Ra^*_{C,c} = 3.80 * 10^{-6}$. The gas heights that correspond to the critical liquid height are 38.1 nm for H=2 and 0.107 mm for H=101. Doing this for several values of H allows plotting the critical liquid height as a function of the gas height. This is presented in figure 1(b). Figure 1(b) shows that when the gas layer thickness increases, the critical liquid height increases as well. This suggests that for higher gas layer thicknesses, the reference state can be considered as more stable. It is interesting to link this finding with the results that are presented in figure 1(c). This figure shows that the critical solutal Marangoni number shows two tendencies: first it decreases and then it increases when the gas thickness is increased. The second tendency confirms that the system becomes more stable when the gas thickness is increased. The steep increase of the critical Marangoni number for very small gas thickness is however a bit astonishing and could hide a different instability mechanism that will be studied in more detail in the future.

Comparison of the full approach with a Pearson-like model

When looking at figure 1(a) it appears moreover that for both the values H=2 and 101, the critical wave number is about 2. This and also the order of magnitude of the critical Ms* numbers (~300) are not far from the case considered by Pearson [3]. Therefore, this is scrutinized further. A comparison of these calculations with the case considered by Pearson has been made. For this purpose, the model of the present work can be simplified as follows. Convection and temperature diffusion in the gas phase can be neglected if the dynamic viscosity and the thermal conductivity in the gas phase are much smaller than in the liquid phase, while the opposite should be the case for the diffusion coefficient. This is actually satisfied in our work. This results into an equation for the perturbation of the solute mass fraction, converted in normal modes, at zero growth rate:

$$D_z^2 C_g - k^2 C_g = 0 \tag{16}$$

A solutal Biot number can be defined by solving for equation 16, with appropriate boundary conditions. It then turns out that a solutal equivalent of the Biot number that can be found in Pearson's model, can be deduced, which is expressed as:

$$Bi(k) = \frac{1-C_{i,l}}{1-C_{i,g}} \frac{M_l}{M_g} \left\{ \frac{C_{i,g}+M_g(1-C_{i,g})}{C_{i,l}+M_l(1-C_{i,l})} \right\}^2 \frac{K_e}{P_i} \rho D k \coth[k(H-1)] \tag{17}$$

The solutal Marangoni number as a function of the wavenumber is then given by:

$$Ms(k) = 8k \frac{-\cosh^3(k)Bi(k)-\cosh^2(k)\sinh(k)+k^2\cosh(k)+\cosh(k)Bi(k)+k\sinh(k)Bi(k)}{-\cosh^2(k)\sinh(k)+k^3\cosh(k)+\sinh(k)} \tag{18}$$

Figure 2(a) shows the comparison of the solutal Marangoni number calculated in this work with that using equation 18 for H=2. Figure 2(b) presents the same for H=101.
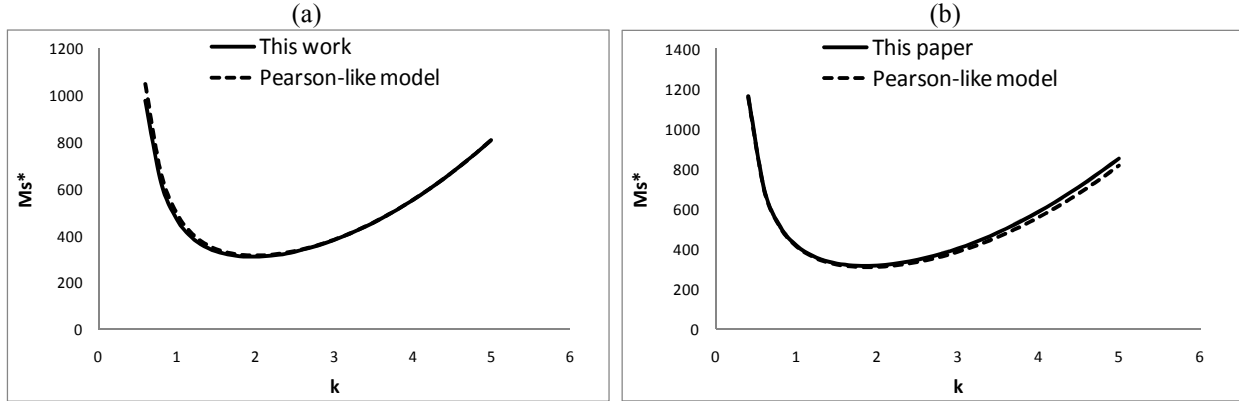

Figure 2: Comparison of the solutal Marangoni number from this work and that from the case of Pearson for (a) H=2 and (b) H=101 (10% of ethanol in water)

Figures 2(a) and 2(b) show that the solutal Marangoni number calculated in this work corresponds both qualitatively and quantitatively well with that obtained by the Pearson-like model, with Biot number defined by equation (17). This suggests that convection and temperature diffusion in the gas phase play a minor role with respect to mass diffusion for values of H equal or higher than 2.

Conclusions

It appeared that higher critical liquid heights are obtained for higher gas thicknesses. The neutral stability curves of the solutal Marangoni number showed that the solutal Marangoni effect dominates the other effects (including the thermal Marangoni effect and the Rayleigh effect). This curve showed furthermore, that some similarities are found with respect to the much simpler problem studied by Pearson. When comparing the solutal Marangoni numbers from this work with those from the case of Pearson, it appeared that both qualitatively and quantitatively the comparison is good. The difference is not more than 1%. This suggests that according to the model describing the system of this work, the convection plays a minor role in the gas phase. Finally it is observed that the critical liquid heights are rather small. The consequence is that the kind of system used in this present work is quite instable for higher liquid thicknesses.

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