Solutal Marangoni instability of binary mixtures evaporating into air: an analytical model describing highly unstable cases

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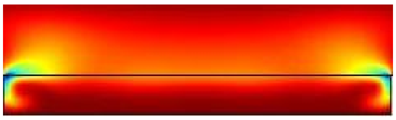
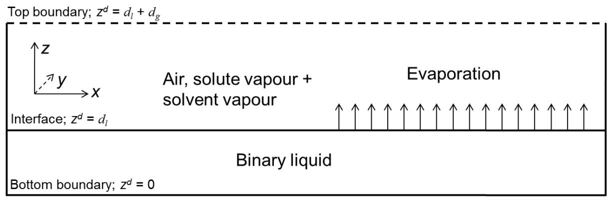
INTRODUCTION

Let us consider a horizontal binary mixture that evaporates into air. The evaporation process gives rise to the development of transient solute mass fraction profiles in both the liquid and gas phases. Here the reference solution is defined as the corresponding horizontally homogeneous base state where the liquid is at rest. After the beginning of evaporation, mass fraction or temperature gradients can trigger instabilities when a certain time threshold is surpassed.



The purpose here is to find this critical time that marks the onset of instability as a function of the liquid thickness. Systems having a gas layer much larger than the liquid one are of primary concern in this work. In this case, the critical time for the onset of Marangoni instability proves to correspond to the moment when the mass fraction boundary layers are still far from the respective bottom liquid and top gas boundaries. The mass fraction reference solutions can then be approximated by an analytical auto-similar description, considering semi-infinite liquid and gas layers. As far as the perturbations are concerned, a special situation occurs. It appears that at the onset of instability, contrary to the reference boundary layers, the perturbations in the liquid reach the bottom of the liquid. The perturbations in the gas, though, are still far from the top of the gas. This gives rise to a new analytical formulation for the instability onset. Hereby, the assumptions made in the Pearson-like model (described in [1]) are followed, albeit with a solutal Biot-like number that depends on the wavenumber:

$$\tilde{Bis}(\tilde{k}) \equiv Bis(\tilde{k},\tilde{H})\|_{\tilde{H}\to\infty} = \rho D \frac{(1-c_{i,ref,l})K_e + c_{i,ref,l}\delta_D\delta_M p_{sat1}}{\left[1+c_{i,ref,l}(\delta_M-1)\right]^2 \delta_M' p_t^d}\tilde{k}.$$



HYPOTHESES AND MODEL DESCRIPTION

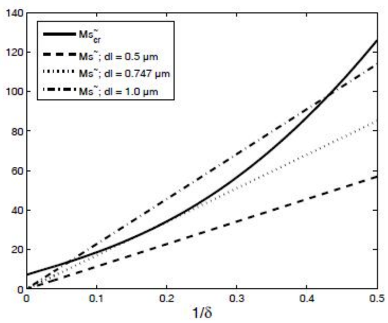
- Boussinesq approximation
- Air absorption is neglected
- Local equilibrium at the interface
- Rigid bottom plate
- A transfer distance is assumed in the gas phase
- Atmospheric pressure in the gas phase
- Binary liquid: 10 wt% of ethanol in water
- External fixed temperature of 300 K
- Semi-infinite gas and liquid layers for the reference solutions
- Semi-infinite gas layer for the perturbations
- *Finite liquid layer for the perturbations*

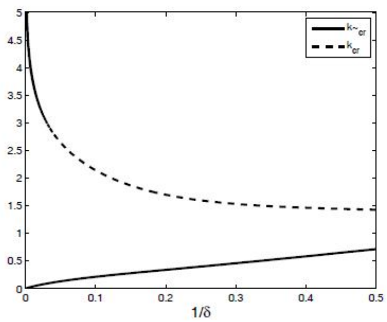
OUTLINE

- Transient reference profiles are used for all the profiles (fully transient)
- The frozen-time approach for the transient profiles
- Perturbations are added to the reference equations
- Linearization
- Using the boundary layer as the length scale
- The main control parameter that is kept is d_l
- It is the purpose to calculate the instants of t at which the system passes through the marginal condition at a certain wavenumber k, that is the critical times.
- A closed-form of the critical solutal Marangoni number is obtained for this purpose:

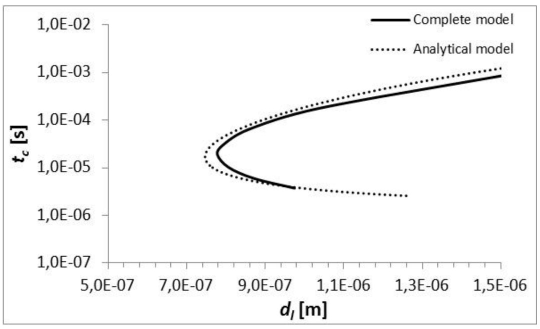
$$\tilde{M}s(\tilde{k},\delta) = \left(\left(1+\frac{\tilde{Bis}(\tilde{k})}{\tilde{k}} - e^{-2\tilde{k}\delta} + \frac{\tilde{Bis}(\tilde{k})}{\tilde{k}}e^{-2\tilde{k}\delta}\right)\sqrt{\pi}\left(1-e^{-4\tilde{k}\delta}-4e^{-2\tilde{k}\delta}\tilde{k}\delta\right)\right)\times$$
$$\Big(1-2e^{4\tilde{k}^2}\tilde{k}\sqrt{\pi}\mathrm{erfc}[2\tilde{k}]+e^{-6\tilde{k}\delta}-e^{-4\tilde{k}\delta}-e^{-2\tilde{k}\delta}+4e^{4\tilde{k}^2-6\tilde{k}\delta}\tilde{k}\sqrt{\pi}-4e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}+4e^{-4\tilde{k}\delta-\frac{\delta^2}{4}}\tilde{k}\delta$$
$$-4e^{-2\tilde{k}\delta-\frac{\delta^2}{4}}\tilde{k}\delta+8e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}^2\sqrt{\pi}\delta+e^{-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2-e^{-2\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2-2e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2-2e^{4\tilde{k}^2-6\tilde{k}\delta}\tilde{k}\sqrt{\pi}\mathrm{erfc}[2\tilde{k}]$$
$$+2e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\mathrm{erfc}[2\tilde{k}]+2e^{4\tilde{k}^2-2\tilde{k}\delta}\tilde{k}\sqrt{\pi}\mathrm{erfc}[2\tilde{k}]-4e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}^2\sqrt{\pi}\delta\mathrm{erfc}[2\tilde{k}]+4e^{4\tilde{k}^2-2\tilde{k}\delta}\tilde{k}^2\sqrt{\pi}\delta\mathrm{erfc}[2\tilde{k}]$$
$$+e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2\mathrm{erfc}[2\tilde{k}]+e^{4\tilde{k}^2-2\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2\mathrm{erfc}[2\tilde{k}]-2e^{4\tilde{k}^2-6\tilde{k}\delta}\tilde{k}\sqrt{\pi}\mathrm{erfc}\left[-2\tilde{k}+\frac{\delta}{2}\right]$$
$$+2e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\mathrm{erfc}\left[-2\tilde{k}+\frac{\delta}{2}\right]-4e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}^2\sqrt{\pi}\delta\mathrm{erfc}\left[-2\tilde{k}+\frac{\delta}{2}\right]+e^{4\tilde{k}^2-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2\mathrm{erfc}\left[-2\tilde{k}+\frac{\delta}{2}\right]$$
$$+2e^{4\tilde{k}^2}\tilde{k}\sqrt{\pi}\mathrm{erfc}\left[2\tilde{k}+\frac{\delta}{2}\right]-2e^{4\tilde{k}^2-2\tilde{k}\delta}\tilde{k}\sqrt{\pi}\mathrm{erfc}\left[2\tilde{k}+\frac{\delta}{2}\right]-4e^{4\tilde{k}^2-2\tilde{k}\delta}\tilde{k}^2\sqrt{\pi}\delta\mathrm{erfc}\left[2\tilde{k}+\frac{\delta}{2}\right]$$
$$-e^{4\tilde{k}^2-2\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2\mathrm{erfc}\left[2\tilde{k}+\frac{\delta}{2}\right]-e^{-4\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2\mathrm{erfc}\left[\frac{\delta}{2}\right]+e^{-2\tilde{k}\delta}\tilde{k}\sqrt{\pi}\delta^2\mathrm{erfc}\left[\frac{\delta}{2}\right]\Big)^{-1}.$$

Comparison with a complete model [2]





$$\tilde{M}s = \frac{\gamma_C\sqrt{D_l t^d}(c_{i,ref,l}-c_b)}{D_l\mu_l} = \frac{\gamma_C(c_{i,ref,l}-c_b)}{D_l\mu_l}\frac{d_l}{\delta}$$



RESULTS AND DISCUSSION

- For very small d_l the system is always stable
- As time increases after the beginning of evaporation, the thickness of the boundary layer developing from the top of the liquid layer also increases (δ^{-1} ↑)
- Consequently, the lower limit of the liquid layer gets closer and closer to the bottom of this boundary layer
- For this reason, the stabilizing influence of the boundary conditions at the bottom of the liquid becomes more important for the thickening boundary layer, which makes the critical Marangoni curve increase with δ^{-1}. The Marangoni number (*Ms*), proportional to $\sqrt{D_l t_d}$, also increases with δ^{-1}, but for small d_l, this (linear) increase is not fast enough to compensate for the simultaneous increase of the critical Marangoni number and the system remains always stable
- As dl increases the Marangoni number increases until it reaches the critical value
- This critical liquid thickness is found to be 0.747 μm

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[1] H. Machrafi, A. Rednikov, P. Colinet, P.C. Dauby, Bénard instabilities in a binary-liquid layer evaporating into an inert gas, J. Colloid & Interface Sc. 349 (2010), 331-353

[2] H. Machrafi, A. Rednikov, P. Colinet, P.C. Dauby, Very unstable systems: analysis of an evaporating binary liquid, to be submitted in Physcial Review E (2012)