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Bénard instabilities in a binary-liquid layer evaporating into an inert gas

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ABSTRACT

A linear stability analysis is performed for a horizontal layer of a binary liquid of which solely the solute evaporates into an inert gas, the latter being assumed to be insoluble in the liquid. In particular, a water-ethanol system in contact with air is considered, with the evaporation of water being neglected (which can be justified for a certain humidity of the air). External constraints on the system are introduced by imposing fixed "ambient" mass fraction and temperature values at a certain effective distance above the free liquid–gas interface. The temperature is the same as at the bottom of the liquid layer, where, besides, a fixed mass fraction of the solute is presumed to be maintained. Proceeding from a (quasi-)stationary reference solution, neutral (monotonic) stability curves are calculated in terms of solutal/thermal Marangoni/Rayleigh numbers as functions of the wavenumber for different values of the ratio of the gas and liquid layer thicknesses. The results are also presented in terms of the critical values of the liquid layer thickness as a function of the thickness of the gas layer. The solutal and thermal Rayleigh and Marangoni effects are compared to one another. For a water–ethanol mixture of 10 wt.% ethanol, it appears that the solutal Marangoni effect is by far the most important instability mechanism. Furthermore, its global action can be described within a Pearson-like model, with an appropriately defined Biot number depending on the wavenumber. On the other hand, it is also shown that, if taken into account, water evaporation has only minor quantitative consequences upon the results for this predominant, solutal Marangoni mechanism.

1. Introduction

In standard studies [1–8] of convective instabilities of horizontal liquid layers, the destabilizing gradient across the layer is directly controlled from the outside of the system, through boundary conditions below and above the layer. The instability due to gravity and density variations is usually called Rayleigh–Bénard instability, while Marangoni–Bénard instability refers to the case where the surface tension variations are the driving factor behind it. When both effects couple, the term "Rayleigh–Marangoni–Bénard instability" is used. If it is the concentration dependence of the density and surface tension that causes the onset of convection, the corresponding instabilities can be called "solutal" Rayleigh–Marangoni–Bénard instabilities. Likewise, "thermal" instabilities result from the similar role of the temperature.

When evaporation takes place at the upper surface of the liquid layer, the situation becomes even more intricate. Indeed, evaporation is an endothermic process, resulting in the cooling of the liquid surface. In the case of binary mixtures, it is also accompanied by concentration gradients across the layer. Since the density and surface tension depend on both the temperature and the concentration, evaporation can thus indirectly destabilize the liquid layer.

Convection due to evaporation is an important phenomenon that occurs in many applications, such as during the drying of paint films, coatings, heat exchangers and process engineering installations. It also occurs in nature when for instance a salty lake dries out due to the evaporation of water, leaving behind structures on the soil. Several theoretical works have already been published on evaporation-driven Bénard instability of a one-component liquid layer [9–15], with the liquid evaporating into either an inert gas [9–13] or into its own vapor [6,14,15]. To our knowledge, the studies of two-component systems have been rather limited in this context. One can mention a scaling analysis [16] or experiments [17]. From the theoretical viewpoint, quite a comprehensive study has been carried out in the case of a spherical geometry, when the Marangoni (both thermal and solutal) instability has been considered for an evaporating binary-liquid droplet [18]. Let us also mention experiments in a Hele-Shaw cell configuration with evaporating water–alcohol solutions [19,20], where density-fingering (plume-like) patterns have been observed after a certain time had elapsed since the exposure of the solution to the air, which is clearly a manifestation of a buoyancy-driven (Rayleigh) mechanism. Cellular Marangoni patterns have also been observed [19], which are then suppressed by adding a surfactant, and the

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theoretical part of [19] concerns just the buoyancy-driven instability.

In the present paper, the Rayleigh–Marangoni–Bénard instability induced by evaporation is studied by means of a linear stability analysis in the case of a binary-liquid layer, when both the solutal and the thermal factors are involved. The particular model used here assumes a dilute solution of which only the solute evaporates (even though the latter limitation is relaxed at a later stage). In this case, the gas layer consists of an inert gas and the vapor of the evaporating liquid. The aim of the paper is to study the different instability mechanisms and to assess the degree of their mutual importance using a configuration as simple as possible. A concrete example followed throughout the paper is a 10/90 wt.% ethanol/water mixture at normal conditions.

The paper is organized as follows. In Section 2, the studied configuration is described, and a mathematical formulation of the problem is provided assuming that it is only the solute (ethanol) that evaporates from the binary mixture. The reference state is considered in Section 3. Section 4 is concerned with the formulation of the linear stability problem. The results of the linear stability analysis are presented in Section 5, and the relative importance of various instability mechanisms is assessed. An approximate analytical treatment of the reference profile and of the marginal stability conditions (by means of a Pearson-like model) is carried out in Section 6, making use of various simplifications possible within the full model. In Section 7, the model is generalized to account for solvent (water) volatility, although the subsequent analysis is limited to the approximate approach framework of Section 6. Finally, the conclusions are summarized in Section 8.

2. Description of the problem

The system the instability of which is studied in this paper is presented in Fig. 1. It consists of a binary-liquid layer (thickness d_l, also denoted h^d, see the distinction between the two below) in contact with a gas layer (thickness d_g, also denoted $(H^d - h^d)$, so that H^d is the total thickness of the two-layer system). The liquid layer rests on a horizontal solid surface with a fixed temperature. The liquid–gas interface is assumed to be undeformable. The liquid layer is made up of a solute in dilute concentration and a solvent. The gas layer consists of air (the absorption of which in the liquid is neglected) and the vapors of the solute and the solvent.

The treatment of the gas layer adopted here follows Haut and Colinet [10]. The thickness d_g in such an approach is just viewed as a semi-heuristic quantity describing the typical equivalent (effective) diffusion length in the gas phase as determined by external air currents which may be naturally present or deliberately created (ventilation) therein: d_g is the distance at which the diffusion is formally of the same magnitude as the convective transport in a real setup (a transfer distance). In this sense, the gas phase above this layer is considered as perfectly mixed while ensuring given "ambient" values of temperature (the same as at the bottom of the liquid layer in the case considered here) and concentration at the effective upper boundary of our gas layer. In this respect, the approach is actually rather similar to the so-called "stagnant film" approach, often used in chemical engineering [21]. The shear-induced influence of such currents on the liquid layer is neglected however, and thus no net horizontal flow in the gas phase is explicitly included into the model. Besides, the model is formulated assuming no externally imposed horizontal non-uniformities upon the system, implying that the scale of any horizontal non-uniformity that may exist in a real setup (as opposed to the present idealized configuration) is much greater than the scale of the phenomena to be studied here (evaporation-induced Bénard instability). As for the hydrodynamic conditions at the effective upper boundary of the gas layer, we shall use the "soft" ("stress-free") ones: no tangential stress and a given uniform pressure/normal stress. We note that, overall, despite its heuristic character, the proposed approach is more detailed and general than the frequently used one based upon describing the transport processes in the gas by means of simply a transfer coefficient (Biot number): the former permits to assess an active role of the gas phase in the studied phenomena, whereas the latter (being essentially a one-sided model of the liquid layer) does not.

The solvent is considered to be much less volatile than the solute. Actually, in the main body of the paper (Sections 2–6), the solvent is formally treated as non-volatile. For a dilute solution of ethanol (solute) in water (solvent), which is a concrete example followed throughout the paper, such a treatment is expected to be approximately valid at some ambient humidity of the air, when the water vapor is nearly saturated relative to the solution (otherwise, even though water is indeed much less volatile than ethanol, its greater amount in the solution may make the effects of its evaporation nonetheless noticeable). At the end of the paper, however, we shall come back to the question of how the results change if water evaporation is incorporated into the model for an arbitrary humidity of the air.

Here we shall also assume that it is not only the temperature that is fixed at the bottom of the liquid layer, but also the concentration. While the latter assumption seems to be rather artificial, it will permit to study in a simple way (i.e. for a quasi-stationary reference state) the Bénard instability mechanisms pertinent to an evaporating binary-liquid layer and to assess their mutual importance, which is the main goal of the present paper.

Now a few words about the notations d_l, d_g, h^d and H^d as used here. Due to evaporation, the liquid thickness changes with time t^d, which we describe here by introducing the function $h^d = h^d(t^d)$. We can then define a constant quantity d_l as the thickness of the liquid layer at a certain reference, or initial, time $t^d = t_0^d$, i.e. $d_l \equiv h^d(t_0^d)$. This thickness d_l will then be chosen as the unit length for non-dimensionalization. In dimensionless form, the bottom plate is located at $z = 0$, and the interface then corresponds to $z = h(t)$, with $h(t_0) = 1$. Note that the superscript "d" stands for the dimensional character of a particular quantity, whenever it is used elsewhere in dimensionless form. Similarly, in view of the meaning attributed to the gas layer thickness here, H^d (defined as the total

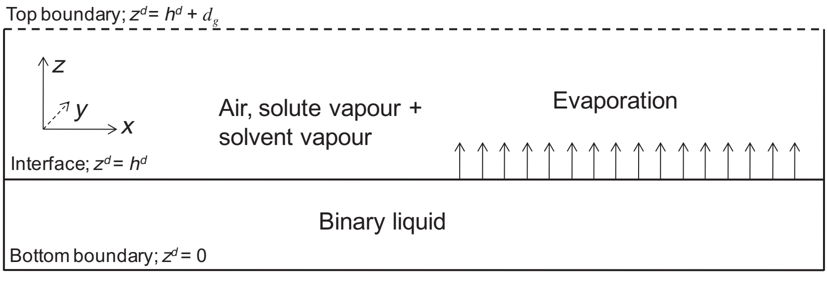

Fig. 1. Sketch of the studied system.