



Sensitivity of lateral heat transfer on the convection onset in a transient Rayleigh-Bénard-Marangoni flow

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ABSTRACT

The convection onset in an evaporation thermal model of a solute/solvent solution is numerically studied. The transient nature of the diffusion temperature profile requires to control the perturbations which will lead to the transition to a convective flow. Contrary to previous papers, we adopted here a perturbation mode, which is physical and experimentally reproducible, by accounting for a heat transfer at the lateral walls. Based on the Péclet number evaluated in the core flow field only, the critical viscosities and the corresponding times for the transition between a diffusion and convection regimes were calculated as a function of the fluid layer thickness, the aspect ratio and the convective heat transfer coefficient. The study of the spatio-temporal diagrams allowed us to characterize the way the convection propagates in the fluid domain as well as the evolution of the mean cell size and wavenumber.

1. Introduction

Many industrial processes such as painting, inking or packaging involve the drying of binary liquid films with a least one volatile component. For a solute/solvent solution brought in contact with a gas environment at standard conditions, the solvent vaporization at the liquid/gas interface leads to an unsteady heat and mass transfer into the whole liquid layer. The solvent evaporation at the liquid/gas interface creates, at the same time, a decrease in the solvent concentration, a cooling due to the phase change endothermic phenomenon, and a reduction in the solution layer thickness. These thermal and solutal variations at the interface then diffuse into the fluid layer. Under specific conditions, this transient diffusive regime can turn out to be unstable because of the buoyancy and/or surface tension forces induced by the thermal and/or solutal gradients. The slow decrease in the fluid layer thickness, combined with the viscosity increasing produced by the enriched solution in solute, implies the convective flow must always fades away over the long time, so that the transfer evolution ends up being driven by the diffusion, again.

The mechanisms leading to natural convective flows have been widely studied. Amongst the earlier works upon convective instabilities, we can mention the theoretical contribution by Pearson [17] for surface tension driven flows (Bénard-Marangoni instabilities) and the experiments by Blair and Quinn [4] for the buoyancy-driven flows (Rayleigh-Bénard instabilities). A recent numerical, experimental and

theoretical review involving surface tension instabilities has been published by Wang et al. [22] to elucidate the mechanisms of the Marangoni effect.

The models used to study the film or droplet evaporation introduce different levels of descriptions. For example, the evaporation of a pure fluid involves the temperature variable, while the concentration of the components must also be taken into account for mixtures. The transfers at the interface are also the subject of numerous models. They can result from the full simulation of the heat and fluid flow in the gaseous phase, or be determined by assuming a diffusion regime in the environment, or more simply be modelled via Newton's laws and global transfer coefficients. The choice for the model of the interface deformation depends greatly on the studied problem, too. It can be rigid and motionless if the evaporated liquid volume is negligible, moveable but non-deformable to account for volume loss, or completely deformable when its shape results from the stress jumps and the continuity of the velocity. The dynamic of solutions may converge toward a steady or quasi-steady flow, or evolve continuously as a function of time, depending on the applied boundary conditions or the temporal range of interest. The consequences upon numerical methods for the search of the convection onset are important since the classical linear stability analysis is *a priori* not applicable when the diffusive solution is unsteady.

In the evaporation case of pure volatile liquids, the experimental works by Colinet et al. [6] and Mancini and Maza [15] focused on the coupling between the solution and the surrounding gas phase, while

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numerical studies were conducted by Merkt and Bestehorn [16]. They showed that, for moderate evaporation rates, the one-sided layer model where only the liquid film is simulated is a good model for real problems. In that case, a generalized heat transfer coefficient accounting for both the convective heat transfer and the thermal evaporation flux is introduced at the liquid/gas interface. In this scope, Machrafi et al. [14] developed a one-sided model to study the thermal Marangoni instability produced by a liquid evaporation in an inert gas. The use of a linear stability analysis of the transient base state using the frozen time approach showed the conditions for which the introduction of a generalized heat transfer coefficient, function of the perturbation wave-number, is particularly relevant at the interface.

In the case of evaporation of a solute/solvent solution, both thermal and solutal contributions are able to destabilize the diffusive solution. In experimental studies by Bassou and Rharbi [1], the authors highlighted the correlation between the solutal Bénard-Marangoni convection cell size and the size of the surface corrugations visible on the deposit at the end of the drying process. For a water/ethanol solution, Machrafi et al. [12] have numerically studied the buoyancy-surface tension coupling. In a quasi-steady regime, the authors developed linear stability analysis to obtain the neutral stability curves as a function of the solutal/thermal Marangoni/Rayleigh numbers. Later on, they carried on their works with transient solutions and the frozen-time approach [13], what allowed them to compute the neutral stability curves in the parameter plane spanned by the liquid layer thickness and the elapsed time after the exposure of the aqueous solution to the surrounding gas.

The present contribution is a part of a more general study devoted to the understanding of the fluid flow and heat and mass transfer in a specific solution of Polyisobutylene and Toluene in which the viscosity strongly depends on the solute concentration. The first work was conducted by Toussaint et al. [19] with experiments. They interested in the convection onset for wide ranges of initial liquid layer thicknesses and solute concentrations (viscosities). They also studied the lifetime of convection cells and the time from which the viscous skin is created at the fluid surface. Based on these experimental results and considering the very large difference in the thermal and solutal diffusion time scales, two evaporation models have been worked out. One is valid at the first beginning of the drying and rests on a uniform concentration field, while the second model is valid later and assumes a thermal equilibrium in the whole fluid layer. The study of the solutal model was carried out by Trouette et al. [21]. The transition from the diffusive state to a convective flow was studied with transient numerical simulations, and the preponderance of the solutal Marangoni convection over the buoyancy one was shown. Recently, Yiantsios et al. [23] performed numerical simulations of the solutal Bénard-Marangoni convection, considering both the strong dependence of viscosity with the solute concentration and the free surface deformation. They showed that the wrinkles observed by Bassou and Rharbi [1] at the end of the drying may be viewed as the signature of the convection cells. The transient thermal model was studied successively by Touazi et al. [18] and Trouette et al. [20], who carried out non-linear numerical simulations in two or three-dimensional geometries, applying random perturbations on some initial fields (velocity [18] and temperature [20]) in order to control the convection onset. The viscosity thresholds were obtained for a wide range of fluid layer thicknesses and these results turned out to be in qualitative agreement with the experimental data by Toussaint et al. [19]. The linear stability analysis by Doumenc et al. [7], performed with the non normal approach, provided threshold values in the same order of magnitude than those obtained with the non-linear simulations and the experimental works [18–20]: for transient problems and a given layer thickness, the diffusive and convective transition viscosity is not clearly defined, as for steady base flows, but by a blurred interval depending essentially on the amplitude of the initial perturbation and the used criterion to delimit the diffusion and convection regimes. An overview upon some of these recent works,

including the thermal and solutal models, can be found in Ref. [8].

The former works on Polyisobutylene/Toluene solutions rely on a control of the convection onset through random disturbances applied at the initial time. One difficulty raised by this approach is how to choose physically the magnitude, and in a lesser extend, the shape of the initial perturbations. Furthermore, the introduction of uncertainties in the numerical model makes the reproducibility of the simulation results tricky, except if the perturbation field is fully known on a given mesh, and if the same numerical code is used; otherwise, the solution risks to be modified. As a consequence, this approach may raise the issue of the convergence of the numerical solutions as a function the mesh size, when the disturbance is mesh dependent.

In this contribution, we consider a thermal model for the evaporation of a Polyisobutylene/Toluene solution. The perturbations of the diffusion transient solution, which give rise to the convection onset, are physically caused by the convective heat transfer with the surrounding gas modelled at the lateral boundaries. The rest of the paper is organized into three sections. The first one describes the physical, mathematical and numerical models. The next section is devoted to the result presentation. After a short introduction and discussion upon the transient solutions and the used criterion to define the transition between the diffusion and convection regimes, the critical viscosities are given as a function of the layer thickness. The accurate description of the convective cell appearance, evolution and their fade-out is then proposed on the basis of spatio-temporal diagrams. Some comparisons are also provided using simulations with disturbances at the initial conditions. The last section is devoted to the concluding remarks and a placement in perspective of our threshold viscosities and those of the literature.

2. Physical, mathematical and numerical models

A homogeneous Polyisobutylene/Toluene (PIB/Toluene) solution at rest is contained into an open container topped by air at standard atmospheric conditions. In contact with the surrounding gas, the liquid solvent evaporates. As evaporation is an endothermic process, some amount of latent heat is taken away from the solution at the gas/liquid interface. Subsequently, the cooling and the solvent deficit are diffusing into the fluid layer thickness. Under specific dimensionless parameters (to be determined), the transient diffusive solution becomes unstable and a fluid motion occurs. This instability may be driven by surface tension and/or density variations, each of them induced by thermal or solutal inhomogeneities. As the physical approximations have been widely discussed in previous papers [7,8,18–20], only the main assumptions are here reminded for sake of brevity. A one-sided model is used and the heat transfer with the surrounding gas is taken into account by a constant and uniform convective coefficient. Since the mass diffusivity of the PIB/toluene solution is about three order of magnitude smaller than its thermal diffusivity, it is possible to identify a thermal regime for short times where the solvent fraction is assumed uniform in the entire liquid film [3,8]. From this assumption, it results that both the physical properties and the fluid thickness do not evolve substantially: they are kept constant at their initial values. Lastly and in accordance with the previous works [18,20], a two-dimensional approximation is used since the transitions from diffusion solutions to convection flows do not depend significantly on the space dimension. Therefore, the fluid layer consists of a rectangular domain of aspect ratio $A = L/e$, where L is the horizontal length and e the thickness of the layer. Thus, the heat transfer and fluid flow are governed by the Navier-Stokes and energy equations, expressed with the Boussinesq approximation.

Based on the fluid thickness e , the thermal diffusivity α and the density ρ , the velocity, time and pressure scales are respectively defined by α/e , e^2/α and $\rho\alpha^2/e^2$. Let us first note $H = h + L_{\text{vap}}(d\Phi_s/dT)|_{T_0}$, with h the convective heat transfer coefficient at the free surface, Φ_s the mass flux density, L_{vap} the specific latent heat and T_0 the temperature of the surrounding gas. The reduced temperature difference then writes

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