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# Stability boundaries for transient evaporative Rayleigh-Benard-Marangoni convection

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## Summary

Thermally driven flow is studied for an horizontal liquid layer suddenly submitted to evaporation. The determination of the onset of convection in this unsteady setting is the main focus of the present work, taking into account buoyancy-driven and surface tension-driven convection. Two approaches are used and compared to previous experimental visualizations performed during the drying of polymer solutions: a numerical 2D simulation of Navier-Stokes equations and a linear stability analysis based on a non-normal approach.

**Keywords:** free convection, evaporation, stability, non-normal approach, heat transfer

## 1. Introduction

Thermally driven flow is studied for an horizontal liquid layer suddenly submitted to free surface evaporation. The solvent evaporation induces a decrease of the temperature at the free surface due to the vaporization latent heat. The increase of the thermal gradients may induce a convective motion driven by buoyancy and/or surface tension [1, 2]. The determination of the onset of convection in this unsteady setting is the main focus of the present work. Two approaches are used and compared to previous experimental visualizations performed during the drying of polymer solutions [3]. First a numerical 2D simulation of Navier-Stokes equations has been developed. Then a linear stability analysis suitable for transient problems and based on a non-normal approach is presented, which is an improvement of previous models as the frozen time approach or the amplification theory (cf. for example [4, 5]). To the best of our knowledge, this approach is the only one capable to provide clear-cut answers on instability problems for truly unsteady basic flows. Since the transition between stable and unstable regime depends on somehow "arbitrary" parameters (chosen criteria, perturbation definition and amplitude for the non linear simulations), it is more suitable to view the results as the estimation of a transition region between a domain exhibiting strong convection and a domain where initial perturbations are damped or have no times to significantly develop during the transient regime. All the results presented here for the two approaches and for different criteria show that this transition region is thin compared to the large domain of Marangoni and Rayleigh covered by the experiments, so that the notion of stability threshold is still valid for this transient problem.

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## 2. Simplifying assumptions and governing equations

Drying experiments that underlie the simulations presented in this paper have been performed on the system Polyisobutylene (PIB)/Toluene [3]. The two control parameters used in the experiments are the initial thickness ( $0.3\text{mm} \leq e \leq 14.3\text{mm}$ ) and the initial polymer mass fraction ( $0 \leq \omega_P \leq 15\%$ , corresponding to an initial viscosity  $5.5 \times 10^{-4}\text{Pa.s} \leq \mu \leq 2.4\text{Pa.s}$ ). When evaporation begins, convective patterns have been observed at the very beginning of the experiment. The Lewis number being very small (about  $10^{-3}$ ), it can be assumed that convective patterns observed in the first minutes are mainly driven by thermal effects. Moreover, the rates of the thickness decrease and change in the polymer mass fraction and in the viscosity are small. It is then possible to use the following simplifying assumptions, valid at the beginning of the drying only: Solutal convection is not taken into account, the physical properties of the solution are assumed constant as well as the solution thickness. The free surface is assumed flat (see [6, 7] for more details).

Given these assumptions and with the Boussinesq approximation, the dimensionless mass conservation, momentum conservation and energy balance equations read:

$$\vec{\nabla} \cdot \vec{v} = 0 \quad ; \quad \frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \vec{\nabla}) \vec{v} = -\vec{\nabla} p + Ra Pr \theta \vec{e}_y + Pr \Delta \vec{v} \quad ; \quad \frac{\partial \theta}{\partial t} + (\vec{v} \cdot \vec{\nabla}) \theta = \Delta \theta \quad (1)$$

where scales for length, velocity, time and pressure are respectively  $e$ ,  $\alpha/e$ ,  $e^2/\alpha$  and  $\rho\alpha^2/e^2$ , with  $e$  the fluid thickness,  $\alpha$  the thermal diffusivity, and  $\rho$  the density. The temperature scale is  $\theta = \frac{T-T_0}{\Delta T}$  where  $T_0$  is the initial temperature and  $\Delta T$  is defined in the following.

$Pr = \nu/\alpha$  is the Prandtl number,  $Ra = \beta_T g \Delta T e^3 / (\nu \alpha)$  is the Rayleigh number with  $\nu$  the kinematic viscosity and  $\beta_T$  the thermal expansion coefficient.

The boundary conditions are the following: apart from the free surface, the velocity satisfies the no-slip condition and the wall is assumed adiabatic. At the free surface a shear stress boundary condition is imposed, given by the balance of surface tension forces with the viscous stresses in the fluid. The thermal boundary condition is derived from the conservation of energy flux at the surface and simplifying assumptions resulting from the small amplitude of temperature variations (cf. [7] for more details). The analysis is limited to a one layer model and transfers with the ambient air are described by global heat and mass transfer coefficients.

$$-\left(\frac{\partial \theta}{\partial y}\right)_{y=1} = Bi (\theta(x, y=1, t) + 1) \quad \text{and} \quad \left(\frac{\partial v_x}{\partial y}\right)_{y=1} = -Ma \left(\frac{\partial \theta}{\partial x}\right)_{y=1} \quad (2)$$

with  $Ma = -\frac{e \Delta T}{\mu \alpha} \left(\frac{\partial \gamma}{\partial T}\right)$  the Marangoni number and  $Bi = \frac{H_{th} e}{k}$  the Biot number.  $\gamma$ , the surface tension, is a linearly decreasing function of temperature,  $\mu$  is the dynamic viscosity,  $k$  is the thermal conductivity and  $H_{th}$  is given by the following equation:  $H_{th} = h_{th} + L \frac{\partial \Phi_{ev}}{\partial T} |_{T_0}$ , where  $h_{th}$  is the heat transfer coefficient,  $L$  the latent heat of vaporization and  $\Phi_{ev}$  the evaporative flux that can be assumed independent of the solvent concentration at the beginning of the drying. The temperature difference  $\Delta T$  used in the temperature scaling is the difference between the initial temperature  $T_0$  and the steady temperature obtained at the end of the transient regime, when the temperature is uniform in the solution. From the dimensional equations we get  $\Delta T = \frac{L \Phi_{ev}(T_0)}{H_{th}}$ .

## 3. Non linear model

To characterize the presence of observable convection, a criterion based on the thermal Péclet number was chosen. The thermal Péclet number  $Pe = e v / \alpha$  compares the relative importance of advection and diffusion, and convection will be considered significant if, when the system is submitted to an initial perturbation, there is a time  $t$  where the perturbation is significantly amplified, i.e. such as  $dPe(t)/dt > 0$  and  $Pe(t) > 1$ . The velocity used in the estimation of the Péclet number is the maximal value of the velocity norm. Simulations have been performed for a large aspect ratio (20) to avoid boundary effects.

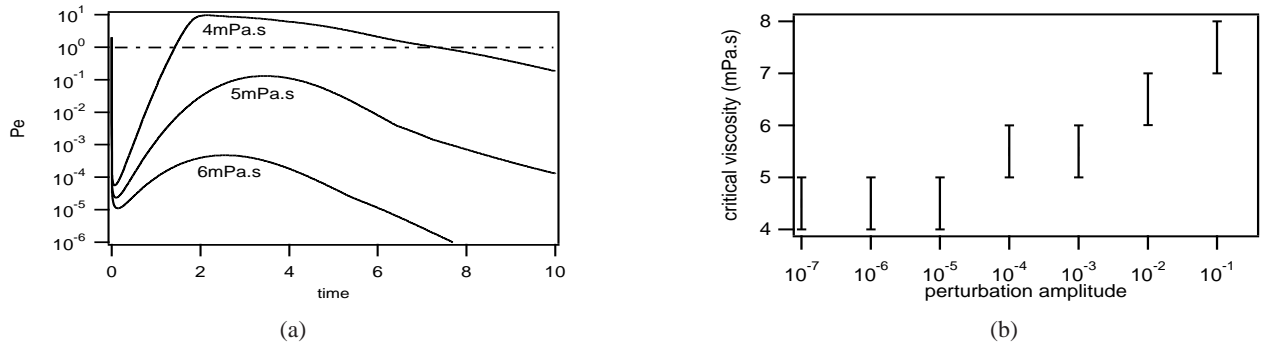


Figure 1: (a) Péclet number for  $e=1\text{mm}$  and various dynamic viscosities, initial perturbation = random velocity perturbation (uniform law with mean=0 and amplitude=4) - (b) Critical viscosity for different amplitudes of the initial temperature perturbation (uniform law with mean=0)

An example of results is given in Figure 1(a) where the time evolution of the Péclet number is drawn for  $e = 1\text{mm}$  and for different values of the dynamic viscosity  $\mu$ . For this example, the critical viscosity corresponding to the chosen criterion lies between  $4\text{mPa.s}$  and  $5\text{mPa.s}$ . As the problem is sensitive to initial conditions, it is important to analyze the sensitivity of the critical viscosity to the amplitude of the initial disturbance. As can be seen on Figure 1(b), only a factor two on the critical viscosity is found when the initial disturbance amplitude changes over six orders of magnitude. Conclusions are the same for initial velocity perturbations and for different form of the initial perturbations.

#### 4. Linear transient stability analysis

As already said, classical linear stability approach is not relevant for this transient problem and a specific method was used, based on the non-normal approach. It is presented in detail in [6]. First, as usually done, linear perturbation equations are derived from the model described above. Two amplification gain are defined. The first one  $G_V(t)$  is based on the kinetic energy of velocity perturbations and the second one  $G_T(t)$  is a quadratic term based on temperature perturbation. Then for each wavenumber  $k$  (spatial development of the perturbation in the infinite horizontal direction) and each time  $t$  an optimization problem is solved in order to get the vertical profile ( $0 < y < e$ ) of the initial optimal perturbation which leads to the maximization of  $G_V(t, k)$  (velocity perturbation) or  $G_T(t, k)$  (temperature perturbation). For a given set of non dimensional parameters ( $Ra$ ,  $Ma$ ,  $Pr$  and  $Bi$  numbers) we then define  $G_V^* = \text{Max}_{t,k} G_V$ , the larger amplification for any time and wavenumber when the initial perturbation is imposed on the velocity, and  $G_T^* = \text{Max}_{t,k} G_T$  that is the larger amplification for any time and wavenumber when the initial perturbation is imposed on the temperature.  $G_V$  and  $G_T$  are normalized with the initial values of the kinetic energy or temperature norm, so that  $G^* < 1$  means that the initial perturbation is never amplified [6].

#### 5. Comparison

The thresholds values obtained with the two approaches (linear analysis and non linear simulations), for different values of the criteria are given in Figure 2(a) for the Marangoni problems ( $Ra = 0$ ) in the plane  $Bi/Ma$  (conclusions are the same for the Rayleigh-Bénard configuration, cf. [6, 7]). As can be seen, all the results are close. That means that even if a precise threshold value has no meaning in such a problem, a thin transition zone between stable and unstable configurations can be defined. The critical Marangoni number is shown to depend very slightly on the Prandtl number [6, 7] and to depend non monotonically on the Biot number, with a minimum value around  $Bi = 2$ .

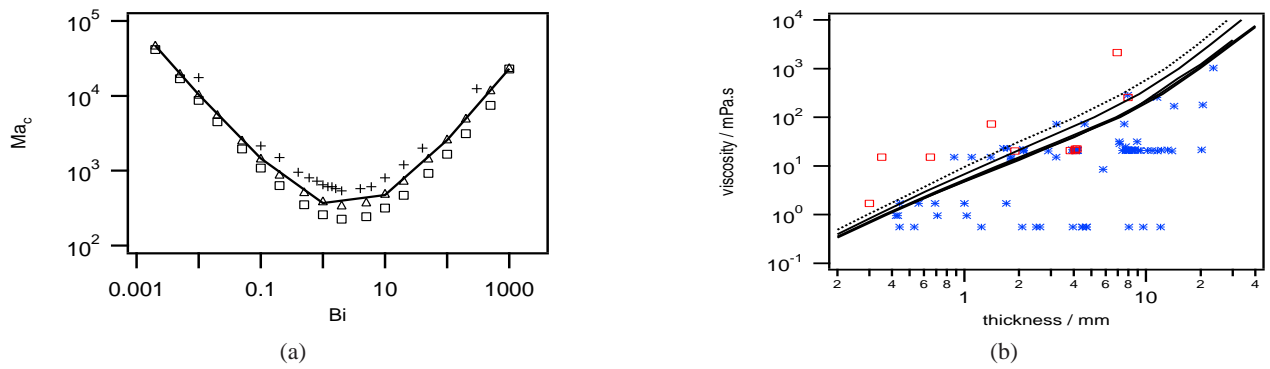


Figure 2: (a) Critical Marangoni as a function of Biot for different criteria: non linear simulation, velocity perturbation,  $Pe = 1, Pr = 1$  (cross) - linear analysis  $G_T^* = 1, Pr = \infty$  (square) -  $G_T^* = 100, Pr = \infty$  (triangle)-  $G_V^* = 1, Pr = 1$  (continuous line) (b) Comparison of theoretical and experimental results in the plane "thickness-viscosity" for different criteria (from top to bottom: linear analysis  $G_T^* = 1, G_T^* = 100$ , non linear simulation with velocity perturbation and  $Pe = 1$ , linear analysis  $G_V^* = 100$  - Experimental symbols: no convection (red squares), convective patterns (blue stars)

Results for the Rayleigh-Bénard-Marangoni configuration are presented in the plane thickness/viscosity for direct comparison with experimental observations. As can be seen in Figure 2(b), the transition zone dividing stable and unstable configurations is consistent with experimental points. Here again the bandwidth of uncertainty due to the choice of threshold and perturbation types is not very broad and does not modify the order of magnitude of the critical thickness.

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