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## Modulation of Marangoni convection in liquid films

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## ABSTRACT

Non-isothermal liquid films are subject to short- and long-wave modes of Marangoni instability. The short-wave instability leads to the development of convection cells, whereas long-wave instability is one of the primary causes of the film rupture.

In this paper different methods for modulation of Marangoni convection and Marangoni-induced interface deformation in non-isotherm liquid films are reviewed. These methods include modification of substrates through topographical features, using substrates with non-uniform thermal properties, non-uniform radiative heating of the liquid–gas interface and non-uniform heating of substrates. All these approaches aim at promotion of temperature gradients along the liquid–gas interface, which leads to emergence of thermocapillary stresses, to the development of vortices and to the interface deformation.

Finally, Marangoni convection in a liquid film supported by a substrate with periodic temperature distribution is modeled by solution of steady state creeping flow equations. This approach is justified for low Reynolds numbers and for Marangoni convection in liquids with high Prandtl numbers. The model predicts interaction between Marangoni convection induced by non-uniform wall heating and the Marangoni short-wave instability.

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## 1. Introduction

Hydrodynamics and heat and mass transport processes in two-phase flows and at the phase interfaces are encountered in numerous natural phenomena and technical applications in energy conversion, process technology, material processing and microfluidics. The typical two-phase flow configurations include films or rivulet, droplets

and bubbles. The capillary forces which are determined by surface or interfacial tension play an important role in hydrodynamics of films, droplets and bubbles, especially if the ratio between the liquid–gas interface area and the volume of liquid is high (e.g. for thin films, small droplets and bubbles).

The surface or interfacial tension is generally a function of the fluid composition and temperature. The surface tension of the common liquids decreases with increasing temperature. In the presence of concentration or temperature gradients along the interface between two phases the surface tension gradients induce interfacial stresses and,

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therefore, bring the liquid into motion [1,2]. This phenomenon is known as Marangoni effect.

Thermocapillary flows are Marangoni flows induced by interfacial temperature gradients. They inevitably arise in configurations, in which the temperature gradients are intentionally imposed. One of the examples is a classical configuration of a horizontal planar liquid layer with a free upper surface, which is bounded from two sides by walls having different temperatures [1,3–5]. In this case the nearly constant interface temperature gradient leads to the development of a return flow pattern in a film with a maximal velocity at the liquid–gas interface. This velocity is directed from the hot wall to the cold wall in the vicinity of interface and in the opposite direction within the film, so that at the steady state the total flow rate in each cross-section of the film is equal to zero. Further frequently studied configurations are motivated by the crystal growth techniques. Liquid bridges [4,6,7] are axisymmetric columns of liquid with a free lateral surface and top and bottom in contact with walls of different temperatures. The temperature gradient in liquid bridges is established along the lateral surface in an axial direction. The basic thermocapillary flow in a liquid bridge is characterized by toroidal streamlines. Another configuration is a layer of liquid between two concentric cylindrical walls (annular geometry) kept at different temperatures, where the top side of the annular region is a free surface subject to radial temperature gradient [4,8]. In this configuration the streamlines of the basic flow have a toroidal shape.

In all the above configurations the basic flow pattern is unstable starting from a certain critical value of the Marangoni number defined as:

$$Ma = \frac{\sigma_T \Delta T l}{\mu a}, \quad (1)$$

where  $\mu$  is the dynamic viscosity,  $a$  is the thermal diffusivity of the liquid,  $l$  is the characteristic size,  $\Delta T$  is the characteristic temperature difference in the system and  $\sigma_T = \left| \frac{d\sigma}{dT} \right|$  is the temperature coefficient of surface tension. The instability of the basic flow leads to development of new flow patterns, including convection cells, waves and oscillatory structures.

The Marangoni effect is also important in configurations, in which the basic state is characterized by a constant interface temperature and zero velocity at each point within the liquid. A paradigmatic system for investigation of Marangoni convection is a horizontal liquid film of infinite horizontal extent and a thickness  $h$  covering a hot wall of a temperature  $T_w$  and exposed to a cool gas of a temperature  $T_g$ . In theoretical investigations it is normally assumed that the heat transfer between the liquid and the gas can be described as thermal boundary condition of the third kind with a constant heat transfer coefficient  $\alpha$ . If the experiment is performed in an open air, then the heat transfer coefficient is determined by the free convection. The free convection in the gas can be suppressed by using an isothermal cover placed at a small distance from the liquid–gas interface [4,9]. In this case  $\alpha$  is determined by the heat conduction in a gas layer. The evaporation of liquid can also be taken into account by an appropriate definition of  $\alpha$ . Heat transport in a film in undisturbed state takes place due to a one-dimensional conduction, and the temperature at the liquid–gas interface is given by

$$T_i = T_w - (T_w - T_g) \frac{Bi}{1 + Bi}, \quad (2)$$

$$Bi = \frac{\alpha h}{k}, \quad (3)$$

where  $Bi$  denotes the Biot number and  $k$  is the thermal conductivity of the liquid.

The basic state of the quiescent liquid layer is subject to two kinds of thermocapillary instability [3,4]. The first of them is called short-wave instability and is induced by the following mechanism. If a certain point at the liquid–gas interface becomes hotter than the environment due to a temperature fluctuation, the local surface tension decreases. The temperature non-uniformity leads to development of thermocapillary stresses which pull the liquid from the hot spot to the colder regions. This local flow leads to the flow of the hot liquid from the wall to the interface, to compensate the Marangoni-induced outflow of the liquid. As a result, the local temperature further increases, the local surface tension decreases and the thermocapillary stresses become stronger. This process is opposed by the viscosity which reduces the flow velocity and by heat conduction which tends to level out the temperature differences. The random fluctuation leads to the development of persisting flow patterns if the Marangoni effect prevails over viscosity and heat conduction, or if the Marangoni number based on the film thickness as the characteristic length and  $\Delta T = T_w - T_i$  exceeds a certain critical value. The critical Marangoni number has been first determined theoretically by Pearson [10] by a linear stability analysis under an assumption of a non-deformable liquid–gas interface. The resulting Marangoni number corresponding to neutral stability depends on the dimensionless wave number of disturbance  $K = 2\pi h/\lambda$  (where  $\lambda$  is the wavelength of the disturbance) and the Biot number:

$$Ma_{cr} = \frac{16K(K \cosh K + Bi \sinh K)[2K - \sinh(2K)]}{4K^3 \cosh K + 3 \sinh K - \sinh(3K)}. \quad (4)$$

For each Biot number the  $Ma_{cr}(K)$  curve is characterized by a single minimum,  $Ma_{min}$ . Below this value the quiescent film is stable. At  $Ma = Ma_{min}$  the disturbance can grow for one certain wave number. For  $Ma > Ma_{min}$  the film is unstable for disturbances in a certain range of wave numbers. For a constant wave number the critical Marangoni number increases with increasing  $Bi$ . For  $Bi = 0$  the minimum of the  $Ma_{cr}(K)$  curve corresponds to  $Ma_{min} = 79.6$  reached for  $K = 1.99 \approx 2$ . This means that the wavelength of disturbance which starts to grow as soon as the Marangoni number reaches 79.6 is  $\lambda = \pi h$ , or has the same order of magnitude as the film thickness. The growth of disturbance leads to development of regular convective cells first observed and reported by Bénard [11]. The size and shape of these cells, as well as onset of instationary convection are determined by the Marangoni number [2,12,13].

The second limiting mode of instability is the so called long-wave instability, which is dominated by the deformability of the liquid–gas interface. A local fluctuation of the interface temperature (hot spot) leads, as discussed above, to the appearance of thermocapillary stresses pulling the liquid in the near-interface region from the hot spot outwards. This outflow leads to the local decrease of the film thickness. From Eqs. (2)–(3) it is seen that the interface temperature decreases with increasing the film thickness. Therefore the local film thinning leads to the further increase of the hot spot temperature and to increasing thermocapillary stresses. The self-sustaining film thinning may lead to the local film rupture. This process is opposed by the hydrostatic pressure which tends to level off the film thickness gradients towards a flat film. The local film thinning is also opposed by the surface tension which tends to reduce the gradients of the interface curvature. The stability of thin film in the presence of Marangoni effect has been studied theoretically in the framework of the long-wave theory [3,14,15]. This theory is applicable for liquid films with low inclination angle of the liquid–gas interface relative to the horizontal, in which the inertial forces and heat convection do not play an important role [16]. Application of the long-wave theory leads to an evolution equation for the film thickness:

$$\mu h_t + \frac{1}{3} \nabla \left[ h^3 \nabla (\sigma \nabla^2 h - \rho g h) \right] - \frac{\sigma_T}{2} \nabla (h^2 \nabla T_i) = 0, \quad (5)$$

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