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Surface deformation and film corrugation during drying of polymer solutions induced by Marangoni phenomena

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ABSTRACT

Solutal Marangoni phenomena occurring during drying of polymer solutions are investigated through numerical simulations. Bénard–Marangoni convection and free surface deformation are considered, taking into account the strong dependence of viscosity with solute concentration. Simulations show that free surface deformation which is resisted by capillary pressure reaches an overall magnitude that is proportional to the capillary number. Decreasing *Ca* also inhibits the interface deformation by favoring the formation of plumes that enhance mixing of the solute. For the range of capillary numbers corresponding to experiments, free surface deformation is not significant. However Bénard–Marangoni convection induces non uniformity of solute concentration and the varying viscosity leads to accumulation of solute at the periphery of convective cells that is observed up to the end of drying. Though simulations have been performed with dimensionless numbers significantly milder than the experimental ones, they are in agreement with experimental observations presented here and in the literature, suggesting that wrinkles may be viewed as the signature of Bénard–Marangoni convection.

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1. Introduction

Marangoni phenomena have a wide range of manifestations, i.e. in crystal growth [1], heat or mass transfer enhancement [2], biological systems [3], thin film coatings and paints, etc. [4–8]. They have also been proposed as a possible cause for the development of organized structures in thin films with potentially promising properties for organic photovoltaics and photodiodes [9]. The most common factors inducing changes in surface tension, and thus Marangoni phenomena, are gradients of temperature or concentration of dissolved species and surfactants [10]. Theoretically, the mechanisms leading to hydrodynamic instabilities have been elucidated first by Pearson [11] for the surface-tension-driven flow (Bénard–Marangoni convection). Another instability mechanism due to surface deformation can also be present: it is known as the long-wave instability [12–14]. Earlier theoretical and experimental studies of thermocapillary phenomena are reviewed in [1,15]. Aspects particular to thin-film flows can be found in the reviews of Oron et al. [16] and Craster and Matar [17].

In the specific case of evaporating thin films, thermocapillary phenomena arise spontaneously because temperature gradients develop due to the energy transported through the film that is required to provide the latent heat for evaporation [18,19]. In multicomponent evaporating films surface tension gradients develop not only due to the effects of temperature but of species concentration as well. Several experimental studies confirm the influence of Marangoni phenomena, with stagnant or spin-coated films of various solutions where the volatile solvent invariably had lower surface tension than the solute [5,20–22]. Theoretical and numerical studies have also been devoted to the study of such configurations [6–8,23–27].

The present study is based on two previous and complementary approaches concerning solutal isothermal problems. On the one hand, theoretical analyses including linear stability and nonlinear simulations are presented in Serpetsi and Yiantsios' work [8]. They account for a deformable free surface but assume constant viscosity. Distinct modes of instability similar to those of Pearson [11] and Scriven and Sternling [12] were found. The former mode leads to cell development with minimal interfacial deformation, whereas the latter is oscillatory and induces significant interfacial deformation. Moreover, it was found that systems where the solute decreases the surface tension are also susceptible to instability in the form of a long-wavelength, non-oscillatory deformational mode. A related phenomenon of solutocapillary motion arises in the leveling of evaporating paints where a characteristic oscillatory

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Nomenclature

a_i	coefficient in experimental viscosity correlation [-]
Bo	Bond number [-]
$\boldsymbol{C}$	convection matrix for finite element diffusion equation [-]
Ca	capillary number [-]
D	mutual diffusion coefficient [m^2/s]
E	normalized evaporation velocity [-]
E'	volumetric evaporation rate [m/s]
$\boldsymbol{e_y}$	normalized unit vector in y-direction [-]
$\boldsymbol{G}$	matrix for pressure gradient terms of finite element momentum equation [-]
h	dimensionless local and instantaneous film thickness [-]
H_0	initial mean liquid film thickness [m]
$\boldsymbol{L}$	Laplacian matrix for finite element diffusion equation [-]
$\boldsymbol{L_2}$	matrix for viscous terms of finite element momentum equation [-]
M	Marangoni number [-]
$\boldsymbol{M}$	mass matrix for finite element diffusion equation [-]
$\boldsymbol{n}$	unit vector normal to the interface [-]
P	normalized liquid pressure [-]
P_0	pressure scale [Pa]
$\boldsymbol{S_1}$	boundary condition matrix for finite element diffusion equation [-]
$\boldsymbol{S_2}$	boundary condition matrix for finite element momentum equation [-]
Sc	Schmidt number [-]
t	time [s]
$\boldsymbol{u}$	normalized velocity [-]
U_0	Velocity scale [m/s]
$\boldsymbol{v_G}$	normalized grid velocity [-]
x	spatial coordinate [m]
y	spatial coordinate [m]

Greek symbols

Γ	normalized stress in scaling analysis [-]
$\Delta\sigma$	difference of solute and solvent surface tensions [N/m]
l	normalized wavelength of film thickness perturbations [-]
μ	normalized solution viscosity [-]
μ_0	initial solution viscosity [kg/m s]
ρ	solution density [kg/m^3]
σ_0	solvent surface tension [N/m]
$\boldsymbol{\tau}$	normalized viscous stress tensor [-]
ω	solute volume fraction [-]
$\boldsymbol{\omega}$	numerical vector of solute volume fraction nodal values [-]

Superscripts and subscripts

0	initial value
n	instantaneous value at nth time step
x	x-derivative
y	y-derivative
t	time derivative

behavior is observed. This has been explained by Overdiep [28] as a result of the synergistic action of capillary pressure and the concentration induced surface tension gradients.

On the other hand, linear stability analysis and direct numerical simulations have been performed by Doumenc et al. [26,27], taking into account the variation of viscosity with solute concentration, but disregarding the deformation of the interface. They focus on the onset of Bénard–Marangoni convection and point out the effect of varying viscosity. The aim of the present study is to bring together the two approaches and to get a view of the evolution of the interface deformation and concentration field during the drying, considering variable viscosity.

Indeed, one aspect particularly relevant to evaporating polymer solutions is the patterning of the final dried films, in the form of corrugations, wrinkles, pinholes, etc. [4]. Theoretical studies are virtually non-existent, except from a few that are limited to the long-wavelength approximation and the lubrication regime for the fluid flow and scalar transport [29,30]. The development of patterns in thin films encompasses a complex and multifaceted array of phenomena attributed to various factors. For some authors, a notable factor is the formation of an elastic skin and the incompatibility of its mechanical properties with those of the substrate or the lateral boundaries. Considerations and analyses along this line have been presented in the literature to explain various manifestations of wrinkling pattern development [31–33]. On a different length scale well known are phenomena of phase separation in polymer blends and in evaporating solutions [34–36]. However, Marangoni instabilities are also considered a potent factor that may lead to patterned dry films [5,21,35,37]. Notably, experiments performed by Bassou and Rharbi [5] with polystyrene/toluene solutions show a strong correlation between the convection cell morphology and the final corrugation of the dried film. From these observations, these authors infer that the final state of the film is the signature of solutal convection which redistributes the solute along the thin film, inducing lateral concentration non-uniformities, which might be finally frozen when most of the volatile solvent is removed by evaporation. Following this track, focus is placed in this paper on solutal Marangoni phenomena that could potentially be responsible for the wrinkling behavior observed in the experiments. Through numerical simulations, we explore the onset and development of solutal Bénard–Marangoni convection and consequently the development of concentration non-uniformities. We also investigate the deformation of the interface that could also results in corrugations if it is significant and continues until the end of the drying. The question addressed in this paper is therefore the relative importance of these two phenomena, according to values of capillary and Marangoni numbers, and the role played by viscosity variation with concentration.

The paper is organized as follows. First an experimental section presents some drying experiments performed with polyisobutylene/toluene solution, showing the drying and corrugation kinetics. Model assumptions, governing equations and numerical approach are developed in Section 3. The last section is devoted to the analysis of simulation results, focusing first on the free surface deformation and then on the solute distribution along the film.

2. Experimental

Drying experiments were performed as follows: polymer solution was poured in a glass dish (diameter about 0.11 m) and put on a balance located in an extracting hood at room temperature. The polymer/solvent solution is polyisobutylene/toluene. Polyisobutylene (PIB) was supplied by Sigma–Aldrich with $M_w = 5\ 10^2$ kg/mol and polydispersity = 2.5. As is well known in polymer solutions, the viscosity strongly depends on solvent content. Fig. 1 displays the viscosity measurements for the PIB/toluene solution used in the experiments. An empirical law interpolated from measurements is used in the simulations of the present study, to describe the variation of viscosity with polymer concentration [27]: $log_{10}(\mu) = \sum_{i=0;4} \alpha_i Y^i$, with $Y = \log_{10}(\varphi_{PIB})$,

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