Self-patterning induced by evaporation and a solutal Marangoni effect

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In this study, we explore through numerical simulations the impact of a solutal Marangoni effect on the deposit obtained by drying a polymer solution. A hydrodynamic model based on the lubrication approximation is used to describe the liquid phase in a dip-coating–like configuration. We focus on the regime of small capillary numbers where the liquid flow is driven by evaporation, in contrast to the well-known Landau-Levich regime dominated by viscous forces [1]. We consider a receding meniscus on a moving substrate, fed by a rectangular inlet duct taking some polymer solution from an infinite and perfectly mixed reservoir at constant polymer volume fraction. The present study follows a previous work dedicated to a dip-coating-like configuration [2]. A hydrodynamic model taking into account varying viscosity and Marangoni effect was developed and succeeded in capturing a self-patterning phenomenon for some values of the substrate velocity [2, 3].

The model examines a drying binary solution (one volatile solvent and one non-volatile solute) in a 2D meniscus. The geometry is represented in Fig. 1. The solution is in contact with a moving, flat substrate with a no-slip boundary condition. Gravity is neglected, and a uniform concentration over the thickness (z direction) is assumed in the liquid phase. The free surface is in contact with air at atmospheric pressure and we assume a local thermodynamic equilibrium at the interface. The whole system is supposed to be at a constant temperature (isothermal problem).



Fig.1: Geometry of the dip-coating like configuration.

The studied case considers evaporation in stagnant air (diffusion-limited evaporation), which results in a coupling between liquid and gas phases. Self-patterning due to the solutal Marangoni effect is observed for some ranges of the control parameters. Viscosity, surface tension, and saturated vapor pressure depend on solute concentration. When surface tension increases with polymer concentration, numerical simulations show that a periodic regime appears (Fig. 2) for some values of the control parameters (substrate velocity and initial concentration).



Fig.2: Periodic time evolution of the liquid height, substrate velocity = 30μ m/s, solute bulk volume fraction = 0.01, and period τ =8.69 s. *The dotted vertical line is the position of the drying front* [2].

Under these conditions, when the solvent evaporates in ambient air, the evaporation flux close to a contact line or a meniscus exhibits strong spatial variations induced by 2D diffusion of the vapor in the gas phase. Due to the important role played by evaporation as a driving force for the flow of the polymer solution, we first investigate the effect of evaporation rate on the deposit morphology. Then we address the role of the spatial heterogeneity in the occurrence of the periodic regime by comparing a 2D diffusion model with a simplified 1D model. Also, we discuss the relative influence of advection and diffusion on polymer transport and show the importance of the inlet boundary condition at very low substrate velocity, when backward diffusion of polymer flow must be taken into consideration.

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