

Motivation

This study was motivated by *the controversy on the origin of the film-thickening effect* in the surfactant-laden Landau-Levich (dip-coating) problem compared to the prediction in the clean interface case [1]:

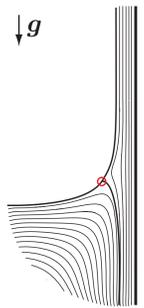


Figure 1: Classical flow topology: film is deposited on a plate moving up.

- ▶ Starting with Groenvelt [2], Marangoni phenomena were deemed responsible for the effect, but no rigorous justification was given.
- ▶ Asymptotic studies [3, 4] suggested film thickening, but the underlying lubrication approximation leads to the flow structure with a stagnation point at the interface, cf. Figure 1.
- ▶ Careful experiments and numerical simulation [5, 6] showed inability of the above views to explain the phenomena under common coating conditions.
- ▶ This controversy has generated a number of speculations on the film thickening and possible flow topologies [7, 8], but none of them proves to provide an adequate understanding [9].

Therefore, the idea was to study the behavior of the stagnation point with the help of experimental flow visualization.

Experimental set-up and materials

▶ Experimental set-up:

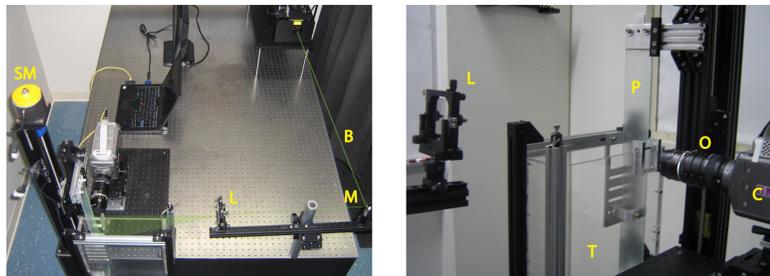


Figure 2: Experimental set-up: (B) Argon laser beam of up to 150 mW at 488 nm, (L) a cylindrical lens (Newport), (M) flat mirrors (Thorlabs), (P) glass plate 50 x 6 x 0.3 cm, (T) plexiglass tank, (SM) stepper motor (Velmex BiSlide), (O) Nikon 55mm lens with extension tubes (could be replaced with ULWD objectives), (C) high-speed camera Phantom v 5.2.

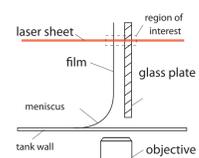


Figure 3: Flow visualization.

We brought the plate close to the tank wall so that capillary rise is high enough to allow one to observe the region of interest far from the wall, ~ 3 cm, cf. Figure 3 (as we learned later, this idea was also used in the flow visualization near a moving contact line [10]). Particles were added to the SDS surfactant solutions and scatter light from the laser sheet. Vibrations of the glass plate were damped with a special set of bearings.

- ▶ **Materials:** hollow polystyrene particles (Polysciences, Inc.) of $1 \mu\text{m}$ size and density $\geq \sim 1.00 \text{ g/cm}^3$, sodium dodecyl sulfate (SDS) from Fisher Science with 99 % purity and MW 288.38, DI water of $\sim 18 \text{ M}\Omega\text{-cm}$ resistivity.

Experimental results

▶ Steady-state flow topology at different withdrawal speeds:

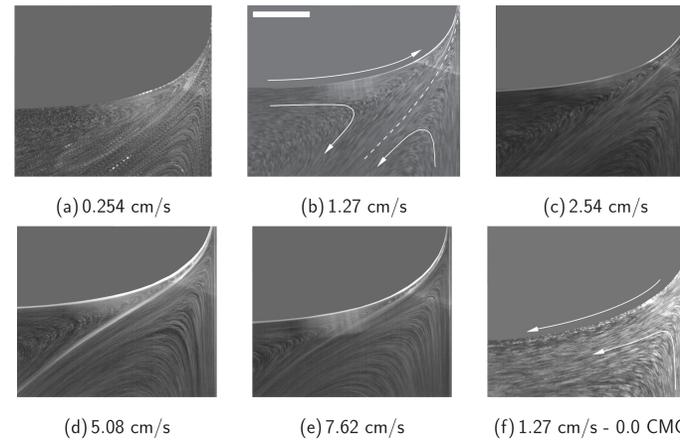


Figure 4: Streaklines at 0.5 CMC (4.1 mol/m^3); scale-bar is 2 mm.

The observed flow topology does not depend on the surfactant concentration, i.e. the ‘two-wedge’ flow pattern is observed for 0.25 - 5.0 CMC. Particles close to the separating streamline (SSL) decelerate when approaching the stagnation point and then accelerate. Flow topology with surfactants is radically different from the clean interface case, cf. Figure 4f.

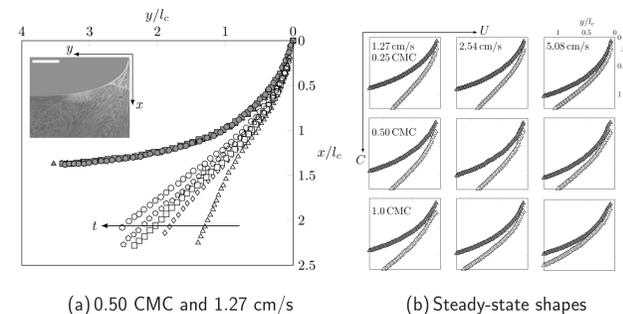


Figure 5: Changes in shape of both the surface (gray) and separating streamline (white) with respect to time, withdrawal speed, and surfactant concentration.

▶ Transient effects:

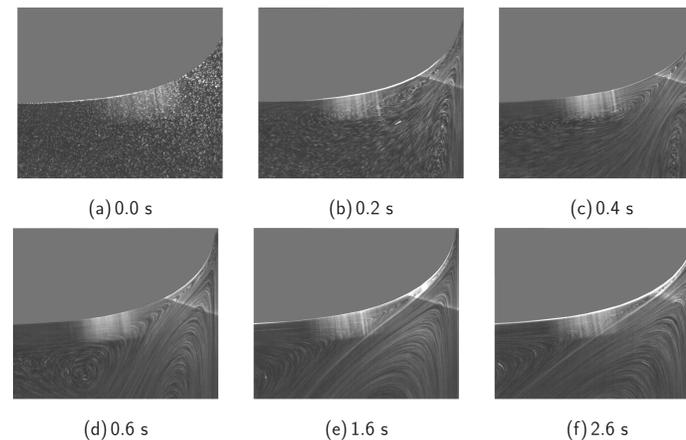


Figure 6: Particle paths at 0.5 CMC and 7.62 cm/s withdrawal speed

These are the true hydrodynamic transient effects, i.e. not due to the kinematics of the stepper motor, of which acceleration and deceleration are $\sim 20 \text{ cm/s}^2$ and happen on a shorter scale than the run-time.

Theoretical reconciliations

The above experiments bring the following important points:

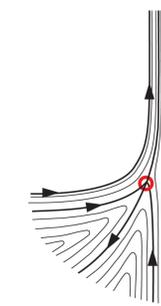


Figure 7: True flow topology.

- ▶ Steady flow structure setup time is determined by the characteristic penetration length-scale $\sim \sqrt{\nu t}$ of the first Stokes problem, which must be of the order of the capillary length $l_c = 1 - 3 \text{ mm}$ to achieve a steady state. In all the considered experiments, the run-time indeed allows one to reach a steady state on the meniscus scale.
- ▶ Flow topology in Figure 7 proves to be not what is classically assumed in Figure 1 – it is also kinematically feasible and consistent with the air phase motion. Notably, the lubrication analysis [1, 3, 4], which is bound to a quadratic velocity profile, does predict the classical flow topology with a stagnation point at the interface, but fails to capture the observed flow structure due to the unidirectional flow assumption in the lubrication approach [9]. Notably, the observed flow pattern in Figure 7 is qualitatively consistent with an idealized analysis of the flow near a moving contact line [11] for certain contact angles and phase viscosities.

- ▶ While one would assume that the flow type for this problem should be simply determined by the Reynolds number based on the capillary length, $Re = \sqrt{La}/Ca = O(10)$, it is dictated by [5]:

$$\sqrt{La}(\partial_t + u\partial_x + v\partial_y)\mathbf{v} = -\nabla p + \mathbf{g} + Ca\Delta\mathbf{v}, \quad Ca = \frac{\mu U}{\sigma}, \quad La = \frac{\rho U^4}{\sigma g},$$

which proves to be in the Stokes flow regime since $La \sim 10^{-5}$.

- ▶ Absence of a stagnation point at the interface enables Marangoni stresses to pump fluid into the film thus explaining the film thickening effect.

Conclusions

- ▶ A true flow topology in the Landau-Levich problem is determined experimentally, which resolves the film thickening controversy.
- ▶ Given these new insights, there may exist physical conditions (surface tension, phase viscosities), when flow topology in Figure 1 is observable and thus enables Marangoni film-thinning effects.

Acknowledgements. R.K. is grateful to Professor Bud Homsy for donating equipment to our laboratory.

Bibliography

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