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# LANDAU-LEVICH FLOW VISUALIZATION: REVEALING THE FLOW TOPOLOGY RESPONSIBLE FOR THE FILM THICKENING PHENOMENA

Hans Mayer* & Rouslan Krechetnikov*[a)]
*Department of Mechanical Engineering, University of California, Santa Barbara, CA 93106, USA

*Summary* A number of experimental investigations have shown that surfactants may result in a thickening of the film deposited by the withdrawal of a flat substrate from a bath of liquid. We report a set of flow visualization experiments which reveal that in the presence of surfactants the flow field near the meniscus region has a stagnation point residing in the bulk and not at the interface as opposed to the clean interface case, which explains the observed increase in film thickness.

## 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]. Namely, starting with Groenveld [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). However, 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.

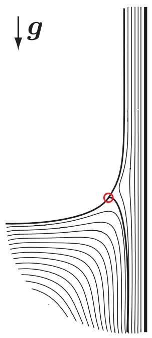
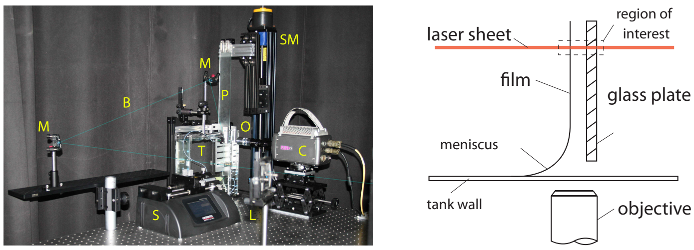



**Figure 1.** Classical flow topology: film is deposited on a plate moving up.

## EXPERIMENTAL SET-UP AND MATERIALS

Experimental set-up shown in Figure 2(a) includes (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) 10x / 50x ultra-long distance objective (Mitutoyo) with a working distance of 33.5 mm, a depth of focus of 3.5 $\mu$m, and a resolving power of 1 $\mu$m, (C) high-speed camera Phantom v 5.2, (S) syringe pump (Harvard Apparatus PhD Ultra). 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, $\sim$ 3 cm, cf. Figure 2(b) (as we learned later, this idea was also used in the flow visualization near a moving contact line [10]). Particles are released locally with a rate controlled by a syringe pump and




(a) Equipment. (b) Flow visualization.

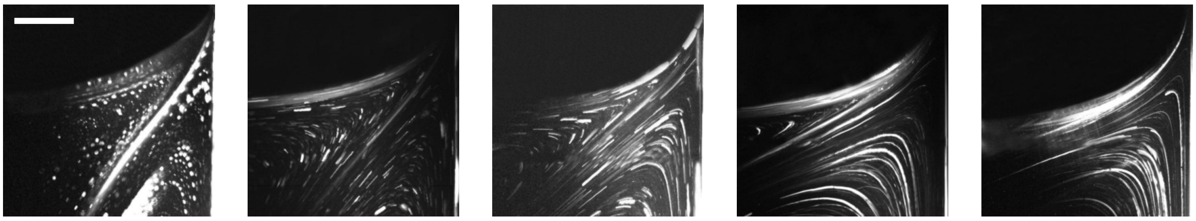
**Figure 2.** Experimental set-up.

excited with a laser sheet. Vibrations of the glass plate were damped with a special set of bearings. Among the materials used are polystyrene particles (Polysciences, Inc.) of 1 and 6 $\mu$m size and density 1.05 g/cm$^3$, sodium dodecyl sulfate (SDS) from Fisher Science with 99 % purity and MW 288.38, DI water of $\sim$ 18 Mohm-cm resistivity.

## EXPERIMENTAL RESULTS

Steady-state flow topology at different withdrawal speeds is shown in Figure 3. The observed flow topology does not depend on (a) surfactant concentration, i.e. the 'two-wedge' flow pattern is observed without surfactants as opposed to the expectation in [7], and (b) particle sizes (1 and 6 $\mu$m). However, smaller particles allow one to get closer to the stagnation point: particles close to the separating streamline decelerate when approach the stagnation point and then accelerate. For both 1 and 0 CMC concentrations we observed a film deposited on a glass plate; in the case of pure DI water we used etching [6], which prevents fast dewetting and thus allows a film deposition. Also, the fact that polystyrene particles have

a) Corresponding author. E-mail: rkrechet@engineering.ucsb.edu.



(a) 0.254 cm/s (b) 1.27 cm/s (c) 2.54 cm/s (d) 5.08 cm/s (e) 7.62 cm/s

**Figure 3.** Streaklines at 1 CMC (8.2 mol/m$^2$); scale-bar is 1 mm.

specific gravity 1.05 prevents them to exhibit any surface active behavior. We do not report higher withdrawal speeds for pure water as the experimental run time is not enough to achieve a steady state.

## THEORETICAL RECONCILIATIONS

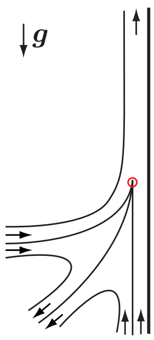



**Figure 4.** True flow topology.

The above experiments bring the following important points:

- Steady flow structure setup time is determined by the characteristic penetration length-scale $\sim \sqrt{\mu\, t}$ of the first Stokes problem, which must be of the order of the capillary length $l_c = 1-3$ 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 4 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 4 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 determined by the Reynolds number based on the capillary length, $Re = \sqrt{La}/Ca = O(10)$, it is dictated by:

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

  which proves to be in the Stokes flow regime [5] since the Landau number $La \sim 10^{-5}$.
- Absence of a stagnation point a 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.

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