TRANSIENT FLOW OF GRAVITY-DRIVEN VISCOS FILMS USED FOR COATING SUBSTRATES WITH VARIABLE TOPOGRAPHY

N.K. Lampropoulos, J. Tsamopoulos

Laboratory of Fluid Mechanics and Rheology, Department of Chemical Engineering, University of Patras, Patras 26500, Greece

We study the two dimensional flow over solid substrates with variable topography, a flow that has practical applications in microelectronics and microfluidics. The problem we address here is the advancing of a thin liquid film over a square-shaped trench with variable depth and width, under the influence of the gravitational force. Completely different wetting patterns result depending on the characteristics of the topography, the capillary number and the dynamic contact angle. On one hand, continuous coating can be achieved in which a thin film of fluid is developed throughout the trench, while a steady flow is established upstream and downstream the topography. This is the desirable pattern in coating of microelectronic devices. The other extreme possibility is that the film completely bypasses the trench, entrapping air inside it. The coating is clearly poor in this case, but this pattern reduces the drag coefficient on the flowing film and, therefore, it is desirable in the operation of super-hydrophobic surfaces for microfluidic applications. Between these two extremes a large variety of patterns exists in which the film partially wets the trench forming an air inclusion all along its bottom or its upstream or downstream inner corners or the film may break up periodically. We produce a comprehensive map covering a wide range of depths and widths of trenches for relevant values of the capillary number and the contact angle.

INTRODUCTION

Our point of interest is the dynamics of flow of a thin coating film of a Newtonian fluid with moving contact line over a solid 2D orthogonal topography. Although the driving force is the gravity establishing a continuous fluid flow of nearly constant thickness down the wall, the method is readily applicable to centrifugally driven flows aiming at coating electronic devices. Several methods have been reported in the literature in order to determine the transient motion of the fluid contact line over plates, trenches, wedges and mounds. Kalliadasis et al. [1] used the lubrication approximation, so that the equations of motion simplify to a single nonlinear partial differential equation for the evolution of the free surface in time and space. Mazouchi et al. [2] studied the time dependent, gravitationally driven fluid flow over flat plates with specific contact angle by means of the vorticity - stream function formulation. The same method was extended to wedges and trenches by Gramlich et al. [3] in which they studied film coating over orthogonal trenches for a few Ca and contact angle (φ) combinations. A map of coating patterns was sketched identifying areas of successful or incomplete coating or dripping failure, while at the same time a wide area in parameter space was left ambiguous. This may be attributed to the limited number of simulations but, more importantly, to the inability of the method they used to simulate discontinuities, such as film rupture and rejoining or highly irregular film configurations. All these problems are circumvented here by examining the two-phase flow of two immiscible fluids, namely air and a viscous liquid, of high density and viscosity ratio. This is accomplished by the Volume of Fluid (VOF) method, first presented by Hirt and Nichols [4] and subsequently advanced by many research groups and applied by our group in flows with axial symmetry [5, 6]. More recently it has been incorporated in the framework of the OpenFOAM open source CFD software package [7]. In OpenFOAM the full transient Navier-Stokes equations are discretized on structured or unstructured computational grids, while they are solved in time through an Euler implicit time-marching method by which the time step is adjusted automatically in order to guarantee numerical stability. The computational cost is reduced by the application of parallel execution on a cluster of interconnected CPUs using Message Passing Interface (MPI).

PROBLEM DEFINITION AND NUMERICAL METHOD

The physical phenomenon studied herein is the gravity-driven flow of a liquid film down a plane as it encounters the rectangular trench to be coated. At the inlet we assume fully developed film flow with the substrate aligned with gravity and we set as boundary conditions the velocity profiles resulting from the solution to the problem described in Figure 1 (left). A film of water of width \( h_1 \) runs down a plate while stationary air fills the much larger gap \( h_2 \) to the second plate. The Stokes equation written for water and air reads as in (1) and (2) respectively:
\[ \mu_1 \frac{\partial^2 u_{x,1}}{\partial y^2} = -\rho_1 g^+ \frac{dP_1}{dx} \]  \tag{1}

\[ 0 = -\rho_2 g^+ \frac{dP_2}{dx} \]  \tag{2}

The boundary conditions are: The no-slip condition at the walls, equality of fluid velocities and a force balance on it assuming that it is flat. The resulting velocity profiles are a parabolic one for water, namely:

\[ u_{water} = \frac{\rho_1 - \rho_2}{2 \mu_1} gh_i \left( \frac{2 y}{h_1} - \frac{y^2}{h_1^2} \right) \]  \tag{3}

and a linear one for air:

\[ u_{air} = \frac{\rho_2 - \rho_1}{2 \mu_1} g \frac{h_1}{h_2} \left[ y h_1 - (h_1 + h_2) h_1 \right] \]  \tag{4}

**Figure 1.** Developed flow of a film of water of width \( h_1 \) down a plane. Air of width \( h_2 \) fills the rest of the gap (left). Outline of the computational grid and detail in the vicinity of the right-upper corner (right).

We also assume that the outlet is occupied by air at atmospheric conditions. In terms of the computational domains used in the simulations, they are Cartesian ones having a width of 5 length units (1 length unit is the width of the liquid film at the entrance, while the remaining 4 units are occupied by air). The domain upstream the trench is set equal to 20 units so that at the front of the running film forms a fully developed profile descending at constant velocity well before it reaches the trench to be coated. The outlet lies 5 units downstream the trench and the upstream and downstream corners of the trench are rounded, having a radius of 0.1 units (Figure 1, right). All calculations are dimensional, where the liquid and air physical properties are tabulated in table 1. The capillary number (\( Ca \)) is initially set 0.1, leading to the film width \( b=0.553 \times 10^{-3} \) m through the definition of \( Ca \)

\[ Ca = \frac{\rho g b^2}{\sigma} \]  \tag{5}

The mean value of the parabolic velocity profile of the fluid at the inlet is
\[ \bar{u} = \frac{\rho gb^2 \sin(\alpha)}{3\mu} = 10^{-3} \text{m/s} \]  

(6)

where the inclination angle, \( \alpha \), of the substrate is taken to be 90° and the resulting dimensionless Reynolds number is \( Re = 10^4 \). The fluid flow is simulated by making use of the \textit{interFoam} solver existing in the OpenFOAM version 2.3.1 software package which uses the Volume of Fluid (VOF method) in order to compute the incompressible laminar flow and the interface between two immiscible fluids, namely the liquid and air. The main advantages of this tool are that it guarantees boundedness of the phase fraction variable (this variable represents the ratio of the volume fraction of one of the phases in each cell) through the use of a multidimensional limiter (MULES), it allows a higher interface resolution extending to only a couple of computational cells and enhances convergence through temporal sub-cycling. Since the convergence and stability of VOF-methods are very sensitive with respect to the equation for phase fraction [7] OpenFOAM proposes that it is beneficial to relax this equation several times within a single time step. Thus the numerical method is enhanced, allowing the use of greater time steps for the transport equations.

**VALIDATION**

This CFD code, as applied with the above mentioned boundary conditions and a computational grid as in Figure 1, is validated by comparing its predictions with the results published in [2]. Here the evolution of a falling film was examined for \( Ca = 0.1 \) and two contact angles, namely \( \phi = 90^\circ \) and \( \phi = 135^\circ \). In both cases, initially the static thin film is assumed to have a uniform thickness, contact angle of 90° and occupy part of the substrate. When it starts moving, it contracts, since the surface tension first minimizes its front by properly adjusting the contact angle, moving the contact line in the direction opposite to gravity and giving it a more rounded shape. The latter results from both the upward motion of the contact line and the downward moving liquid. After a while the upward movement of the contact line stops and the thin film reverses its flow direction, running downward and tending to form at its front a bulge of constant shape. The computed steady state shapes of the film for two contact angles are compared to the results in [2] in Figure 2. In both cases, the interfaces, computed by the two different methods, nearly coincide. Although transient results are available in [2], a quantitative comparison with the present work is not feasible, since the position of the inflow boundary is not explicitly defined in [2].

<table>
<thead>
<tr>
<th>Viscosity</th>
<th>Liquid</th>
<th>Air</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \mu_{\text{liquid}} = 1 \frac{Ns}{m^2} )</td>
<td>( \mu_{\text{air}} = 1.48 \times 10^{-5} \frac{Ns}{m^2} )</td>
<td></td>
</tr>
<tr>
<td>Density</td>
<td>( \rho = 1000 \frac{Kg}{m^3} )</td>
<td>( \rho = 1 \frac{Kg}{m^3} )</td>
</tr>
<tr>
<td>Surface tension</td>
<td>( \sigma = 0.03 \frac{Kg}{s^2} )</td>
<td></td>
</tr>
</tbody>
</table>

Table 1. Physical properties of liquid and air.
Figure 2. Comparison of developed free surface profiles of two viscous thin films running down a plate either calculated by the present code or as presented in [2]. The capillary number is common (Ca = 0.1), while the contact angle $\phi$ is set at $\phi=90^\circ$ (left) and $\phi=135^\circ$ (right). All dimensions are in meters.

RESULTS

Next we study the gravity-driven viscous film flow, when it encounters an orthogonal trench. Here we discuss only the case with Ca=0.1 and contact angle $\phi=30^\circ$ and present the possible film configurations with respect to the size of the trench. Firstly, we have the full or complete coating when the contact point runs throughout the upstream wall, the floor of the trench and the downstream wall, while the bulge formed at the front of the film does not encounter any of the downstream walls prematurely. In this case, no air is entrapped inside the trench as depicted in Figure 3 and occurs in not very deep trenches.

On the other hand, if the trench is not wide enough the bulge in the film front encounters the downstream wall while the contact point of the interface is still on the upstream wall, thus leaving entrapped air in the trench (the so-called capping failure) [3], presented in Figure 4 (the three snapshots in the left). The same capping failure can happen in deeper topographies, where the bulge expands downward due to gravity, while the contact point is still in the upstream wall and finally hits the downstream wall, see Figure 4 (the three snapshots in the right).

Figure 3. Complete coating.

Figure 4. Capping failure arises in narrow trenches (left). The same phenomenon in deeper trenches (right).
In wider trenches, the film may also reach the so-called runout length (this is the maximum depth of the trench that can be coated) [3], before hitting the downstream wall. Beyond this limiting value the front bulge expands due to gravitational force, the film may dewet the upstream wall and finally drips off, if the trench is wide enough, or develops a capping failure as in Figure 4 (right) in narrower trenches. Air can be entrapped also, if the bulge hits the downstream wall, while the contact point runs the floor of the trench, leading to a Bubble in Downstream Corner (BDC) type of failure (Figure 5, snapshots in the left). In even wider trenches the bulge hits the wall, while the contact point is on the downstream wall forming an air bubble on this wall. In this case we have the Bubble in Downstream Wall (BDW) type of failure (Figure 5, snapshots in the right).

Another possible pattern that also depends on the trench width may result in the formation of a Bubble in the Upstream Corner (BUC_1) when the bulge hits the wall forming two contact lines, one moving in the trench floor and the other one in the upstream wall (Figure 6, left snapshot). Then a big bubble is formed and a continuous coating takes place except for this area in the corner. In wider trenches the formed bubble breaks up leading to a collapse of the film as in Figure 6 (right snapshot). Since the upstream wall is continuously wetted by the incoming flow a new front bulge is subsequently formed after the breaking. Depending on the trench depth the incoming film may join the remaining fluid in the trench so that a continuous flow is established again with a Bubble in the Upstream Corner (BUC_2). For certain trench depths marginally bigger than the run out length full coating can also be achieved. This happens as the contact line, while approaching the trench floor, leaves a small amount of fluid near the upstream corner. Once the contact line, after repeated merging and breaking events of the film, meets this existing fluid, the film advances over the floor bed leading to full coating (the so called Drip – rejoin full coating). Finally for even wider trenches we have continuous dripping, where no continuous film flow can be achieved.

In general a contact angle of $\phi = 30^\circ$ is small enough (this value indicates a hydrophilic surface) enabling the film to enter relatively narrow trenches. The bulge expands slower, while coating of the upstream wall takes place, reaching a runout length. For the present parameter values the runout length equals 11.1 length units. Beyond this limiting length the trench cannot be fully coated. The contact point upon reaching the runout length reverses direction, namely dewetting of the wall occurs, while at the same time the bulge expands driven by gravity. The above mentioned patterns are depicted in a complete coating map (Figure 7). Complete coating is achieved above a certain trench width which is a monotonic function of depth. This applies until a certain limiting depth (the so called runout length) beyond which the trench cannot be coated completely without entrapped air bubbles. There is no upper limit in the width of the trenches that can be fully coated, namely for very wide trenches (width =13 or even bigger) a continuous flow of a film that fully coats the trench can be established. In Figure 7, the left map is an enlargement for small trench dimensions. Here we identify that for increasing width capping is followed by BDC failure and subsequently BDW failure until full coating is achieved. The entrapment of bubbles within the trench stops at a depth equal to 5, beyond which the trench either is capped by the film or gets fully coated. Upon reaching the run out length continuous full coating can be achieved only within the area depicted as (Drip – rejoin full coating). By the BUC_1 and BUC_2 failure we
have continuous coating with a big air bubble in the upstream corner, whereas by wider trenches the continuous flow is interrupted and continuous dripping is established.

Figure 7. Coating map for \( \text{Ca}=0.1, \varphi=30^\circ \) (Middle). Expanded view for shallower trenches (left) and deeper ones (right).

CONCLUSIONS

We have investigated the time dependent flow of a gravity driven viscous fluid film over rectangular trenches of variables sizes. This study was implemented by solving the Navier-Stokes equations with the VOF method in order to simulate the incompressible, immiscible, two phase flow at very low Reynolds numbers by making use of the OpenFOAM package. This method, in contrast to other approaches, such as the lubrication approximation [1], or boundary integral methods [2], [3], enables the prediction of the film snap off and its rejoin by coating deep and wide trenches. This is the first time that such coating patterns are predicted. In the future, we will investigate flows and fluids characterized by different contact angles and capillary and Reynolds numbers and examine the coating patterns of trenches of various depths and widths.

Acknowledgments

This work has been supported financially by the General Secretariat of Research and Technology of Greece through the program “Excellence” (Grant No. 1918, entitled “FilCoMicrA”) in the framework “Education and Lifelong Learning” co-funded by the European Social Fund and National Resources.

References