ARTICLES

Quick deposition of a fluid on the wall of a tube

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We are interested in the amount of fluid left behind a drop moved inside a capillary tube. Long ago, Taylor showed that for very viscous liquids moved at small velocities, the film thickness is a monotonic increasing function of the capillary number. New data obtained with liquids of low viscosity are reported here and compared with Taylor's law. Two successive effects are observed: above a threshold in capillary number, the film is thicker than a Taylor film; at a very high speed, the deposition law becomes a decreasing function of the drop velocity. Both behaviors are analyzed thanks to scaling arguments and shown to be consequences of inertia. © *2000 American Institute of Physics.* [S1070-6631(00)00610-3]

I. INTRODUCTION

If a drop of a wetting liquid is displaced by air in a tube, the tube remains wet behind the drop (Fig. 1). We note V the deposition velocity, r the tube radius, and h the thickness of the remaining film.

A very classical (and practical) problem is the knowledge of the amount of liquid remaining on the wall of the tube, namely $h(V)$. This problem has been extensively studied in the limit of low velocities of deposition, in particular since Taylor's and Bretherton's first experiments.[1,2] After a brief summary of these regimes, we shall present new data obtained at high velocity with liquids of low viscosity, which allows us to stress the influence of inertia on the film thickness.

Our experiment consisted in blowing air in a tube of millimetric diameter where a drop (of initial length $L=5-10$ cm) was first placed (Fig. 1). The positions x_f and x_r of the front and rear meniscus of the drop were measured as a function of time with a camera, each run being done on about 20 cm. The deposition velocity was deduced from the video recordings ($V=dx_r/dt$) together with the film thickness [$h/r=1-(dx_f/dx_r)^{1/2}$]. The error on the measurement of the thickness is of order 1%. The 50 frames/s of the camera limited the experiment to deposition speeds smaller than 3 m/s.

We used transparent polymeric tubes of radii $r=0.42$, 0.62, 0.78, and 1.46 mm. Liquids of low surface tension γ (about 20 mN/m) were chosen: silicone oils (of viscosity $\eta=0.52$, 2.60, and 531 mPa s), ethanol ($\eta=1.17$ mPa s), and alkanes (heptane: $\eta=0.42$ mPa s and decane: $\eta=0.89$ mPa s).

II. VISCO-CAPILLARY DEPOSITION: THE TAYLOR–BRETHERTON REGIMES

In the limit of slow coating, it was understood early that key roles are played by the interfaces: Because of the liquid viscosity η, the boundary condition at the *solid–liquid interface* causes the deposition of a film; at the same time, the static meniscus at the *liquid–air interface* (the dotted line in Fig. 1), which is hemispherical for small tubes, is deformed by the motion, which the surface tension γ opposes. Since viscous and capillary forces play antagonist roles, the natural parameter to be considered is the ratio of these forces, the so-called *capillary number*:

$$\mathrm{Ca}=\frac{\eta V}{\gamma}. \tag{1}$$

We call a regime where the film thickness (normalized by the tube radius) only depends on Ca *visco-capillary*.

For thin films ($h\ll r$) and negligible inertia, the film thickness was calculated by Bretherton, using planar lubrication equations.[2] The film forms in the *dynamic meniscus*, of thickness of order h and length λ. There, a flow takes place because of the Laplace pressure difference $\Delta p=\gamma/r$ due to the difference in curvature between the film and the static meniscus. Balancing the viscous force with the pressure gradient along the dynamic meniscus yields

$$\frac{\eta V}{h^2}\sim\frac{1}{\lambda}\frac{\gamma}{r}. \tag{2}$$

In the latter equation, λ is unknown. It is calculated by matching the static and the dynamic meniscus, which is written by balancing the Laplace pressures:

$$-\frac{\gamma}{r}-\frac{\gamma h}{\lambda^2}\sim-\frac{2\gamma}{r}. \tag{3}$$

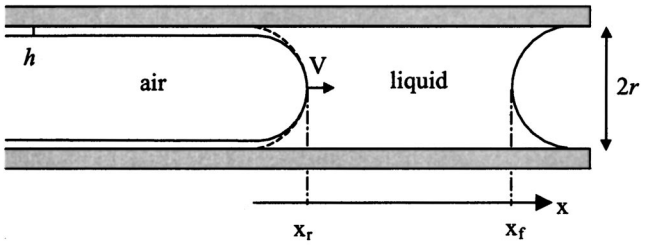

FIG. 1. A drop of a wetting liquid moved in capillary tube leaves behind a film. The thickness h of this film generally depends on the drop velocity V. x_f and x_r are the time-dependent front and rear positions of the drop and the tube radius is noted r.

Thus we have $\lambda \sim \sqrt{hr}$, from which the classical *Bretherton law* can be deduced:[2]

$$h/r \sim \mathrm{Ca}^{2/3}. \tag{4}$$

This equation is set for small capillary number ($h \ll r$ implies $\mathrm{Ca} \ll 1$). As Ca increases, r must be replaced by $(r-h)$ in Eqs. (2) and (3), so that the deposition scaling law becomes

$$h/r \sim \frac{\mathrm{Ca}^{2/3}}{1+\mathrm{Ca}^{2/3}}. \tag{5}$$

Equation (5) implies: (i) a dependence of the normalized film thickness on one single parameter, namely the capillary number; and (ii) a convergence of the thickness at large Ca, obviously related to the confinement which imposes $h<r$.

Both these results agree with the data obtained by Taylor with different viscous oils (Fig. 2).[1] A fit of form (5) can even be found (maybe coincidentally) to describe quite precisely the data: the full line in Fig. 2 is $h/r = 1.34\mathrm{Ca}^{2/3}/(1+1.34*2.5\mathrm{Ca}^{2/3})$, where the coefficient 1.34 was indeed derived by Bretherton; the coefficient 2.5 is empirical. We call *Taylor's law* this convenient empirical equation. Excellent numerical solutions of the problem, which remarkably fit Taylor's data, have been proposed since, in particular by Reinelt and Saffman,[3] Martinez and Udell,[4] or Giavedoni and Saita.[5,6]

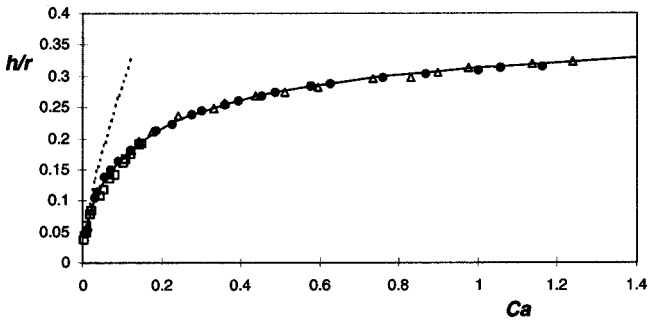

FIG. 2. Normalized film thickness as a function of the capillary number for viscous liquids. The full circles are Taylor's data, which are compared with data we obtained with silicone oil of viscosity 2.6 mPa s (open squares) and 531 mPa s (open triangles). The broken line is Bretherton's law (which is observed to be valid only when the film thickness is negligible compared with the tube radius). The straight line is an empirical fit we call Taylor's law: $h/r = 1.34\,\mathrm{Ca}^{2/3}/(1+1.34*2.5\,\mathrm{Ca}^{2/3})$.

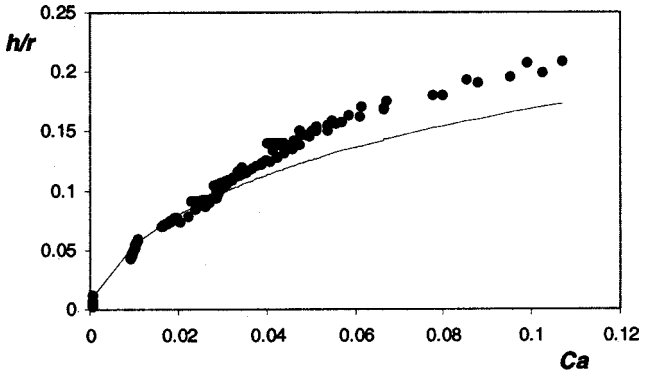

FIG. 3. Normalized thickness of the film deposited by drops of ethanol (of initial length 5–10 cm) moved in a capillary tube of radius $r=0.78$ mm, as a function of the capillary number ($\mathrm{Ca}=\eta V/\gamma$, where η and γ are the liquid viscosity and surface tension). The drop velocities lie between 1 cm/s and 2 m/s. It is observed that the film thickness leaves Taylor's law (drawn in full line) above a threshold in capillary number denoted Ca*. When dividing the film thickness by the Taylor thickness, this quantity is found to pass by a maximum, for a capillary number denoted Ca**.

We performed a first series of experiments with the most viscous oils of the series ($\eta = 2.6$ and 531 mPa s). The data we obtained are also displayed in Fig. 2 (open symbols) and found to obey Taylor's law. They correspond to moderate deposition speed, and all lie in the range of capillary numbers $\mathrm{Ca}=0.01-1$. In the same interval, using liquids of low viscosity (of order 1 mPa s, the water viscosity) implies high speed deposition, which is the object of this study.

III. THE VISCO-INERTIAL REGIME

A. Thickening effect of inertia

The results of the same experiment done at high speed (up to 2 m/s) with ethanol are displayed in Fig. 3 and compared with Taylor's law (drawn in a full line).

It can be observed that the film thickness only obeys Taylor's law at small capillary number (here, $\mathrm{Ca}<0.03$). Then, the film becomes thicker. The difference with Taylor's law first increases with Ca (at its maximum, this difference is of order 30%). Then, it slowly decreases because of the geometric effects at large thickness which impose a convergence regime. We note [Ca*,Ca**] the interval in capillary number where the film thickness increases faster with Ca than Taylor's law.

B. Scaling laws

We try to define the regime observed between Ca* and Ca**. Because of the corresponding high speed, inertia can be suspected to be the cause of this effect. In previous work on fiber coating, we have found that indeed inertia could thicken the film entrained by a fiber.[7] We also showed that this effect could be understood by modifying the scaling laws (2) and (3) in a very simple way. Taking into account both inertia and the fact that h is not necessarily negligible if compared with r leads to the scaling Navier–Stokes equation:

$$\frac{\eta V}{h^2} \sim \frac{1}{\lambda}\left(\frac{\gamma}{r-h}\right) - \frac{1}{\lambda}\rho V^2, \tag{6}$$

with ρ the liquid density. Similarly, the matching between both static and dynamic meniscus writes, instead of Eq. (3),

$$-\frac{\gamma}{r-h} - \frac{\gamma h}{\lambda^2} \sim -\frac{2\gamma}{r-h} + \rho V^2. \tag{7}$$

By eliminating λ in Eq. (6), it finally yields

$$h/r \sim \frac{\mathrm{Ca}^{2/3}}{1+\mathrm{Ca}^{2/3}-\mathrm{We}}, \tag{8}$$

which shows how inertia modifies Eq. (5). The dimensionless number quantifying the importance of inertia appears to be the *Weber number* We, which compares inertia with the capillary force:

$$\mathrm{We} = \frac{\rho V^2 (r-h)}{\gamma}. \tag{9}$$

Equation (8) indicates that the corrections to the Bretherton law [Eq. (4)] at intermediate or high velocities are antagonist: geometry (term in $\mathrm{Ca}^{2/3}$, in the denominator) favors a convergence of the film thickness (Taylor's behavior), while inertia thickens the film. This is different from fiber coating, where both corrections were favoring a thick film: Then, the geometric term was found to be of opposite sign (writing $-\mathrm{Ca}^{2/3}$), which explains why the effect of inertia was observed to be much more spectacular (divergence of the film thickness).[7]

C. Discussion

Even if this scaling approach cannot provide a quantitative description of the data, it is helpful for identifying the role played by the different parameters. It shows for example that the tube radius should be a control parameter for organizing the different effects into a hierarchy: In Taylor's law, the tube radius only appears as a scale for the thickness, while changing r affects the Weber number without modifying the capillary number. Thus, the inertial effect reported in Fig. 3 should depend on the value of r.

Figure 4 shows the film thickness versus Ca for a light silicone oil ($\eta = 0.52$ mPa s), in tubes of different radii (all smaller than the capillary length, for allowing us to neglect the influence of gravity).

It is observed that the tube radius does affect the inertial thickening: the larger the tube, the smaller the threshold Ca*, and the higher the deviation toward Taylor's law (with the largest tube, the film thickness can be twice the Taylor thickness). In the same spirit, Figs. 3 and 4 can be compared. For a given radius ($r = 0.78$ mm), the viscosity is divided by a factor of order 2: it is found that the lower the viscosity, the smaller Ca*, and the steeper the effect. The latter behavior was confirmed by using heptane of still smaller viscosity ($\eta = 0.42$ mPa s) and decane of intermediate one ($\eta = 0.89$ mPa s).

Equation (8) finally allows us to draw a scaling form for the different thresholds. Since inertia and geometry play antagonist roles, the thickening effect should only be observed

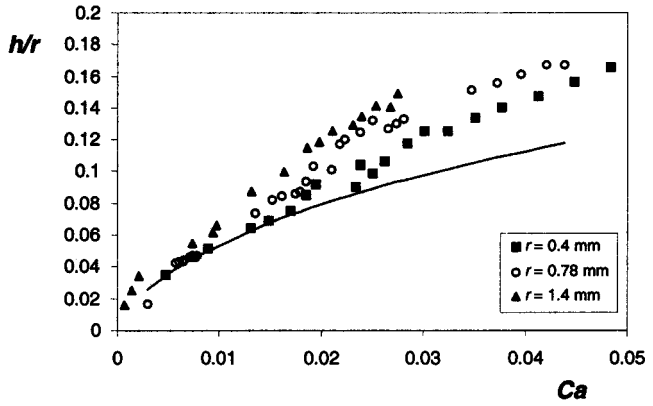

FIG. 4. Normalized film thickness deposited by a light silicone oil ($\eta = 0.52$ mPa s) on the wall of different tubes [$r = 0.42$ mm (closed square), $r = 0.78$ mm (open circle), $r = 1.46$ mm (closed triangle)] as a function of the capillary number.

in situations where We can become of order $\mathrm{Ca}^{2/3}$, which yields a velocity which indeed depends on the tube radius r. This velocity is written: $V^* \sim \gamma^{1/4}\eta^{1/2}\rho^{-3/4}r^{-3/4}$, from which the threshold Ca* in capillary number is deduced:

$$\mathrm{Ca}^* \sim \left(\frac{\eta^2}{\gamma\rho r}\right)^{3/4}. \tag{10}$$

All our results can be summarized in Fig. 5 by plotting the threshold in capillary number Ca* above which the visco-inertial regime is observed, as a function of the dimensionless number $\eta^2/\rho\gamma r$ which appears in Eq. (10). It is observed that the scaling law (10) (the slope $\frac{3}{4}$ is drawn in a full line) is quite well obeyed by the data.

Similarly, a simple criterion can be proposed for the limit in capillary number Ca** above which the thickened visco-inertial regime is dominated by the convergence effect due to the geometry. As soon as Ca is larger than Ca*, the Weber term dominates the capillary term in the denominator of Eq. (8). We simply write that the geometry imposes $h < r$, which prevents the thickness from diverging as it would

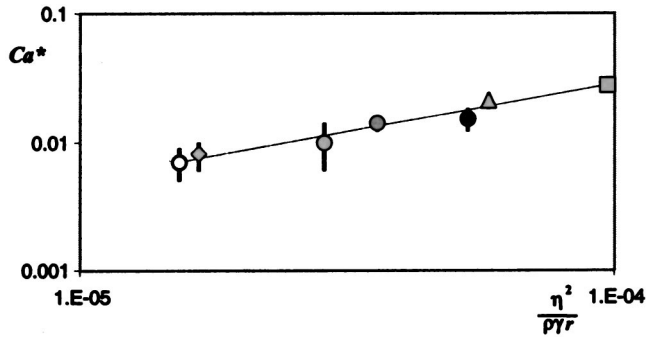

FIG. 5. Threshold in capillary number Ca* above which a visco-inertial regime is observed. Ca* is plotted vs $\eta^2/\rho\gamma r$, a dimensionless number which is function of both the liquid and the tube radius. The data, which correspond to a series of experiments similar to the ones reported in Figs. 3 and 4, were obtained with different tubes ($r = 1.46$, 0.78, 0.62, and 0.42 mm: the darker the symbol, the smaller the tube) and various liquids [heptane (open diamond), silicone oil $\eta = 0.52$ mPa s (open circle), decane (open triangle), ethanol (open square)].

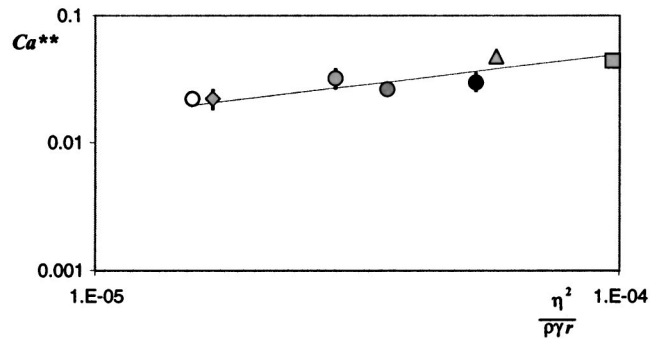

FIG. 6. Threshold in capillary number Ca** above which convergence effects, due to the tube confinement, dominate visco-inertial effect. The symbols are the same as in Fig. 5.

in Eq. (8) if this condition were not specified. Since We $=(r\rho\gamma/\eta^2)\mathrm{Ca}^2$, this condition imposes a capillary number smaller than Ca**, given by the scaling law

$$\mathrm{Ca}^{**}\sim\left(\frac{\eta^2}{\gamma\rho r}\right)^{1/2}, \tag{11}$$

which is larger than Ca*, for Ca<1. Incidentally, the visco-inertial regime should be observed if it takes place before the Taylor convergence regime (i.e., before Ca$=1$), which imposes: $\eta^2/r\rho\gamma\ll 1$. This regime is indeed reported here for liquids of low viscosity in rather large tubes.

The upper limit in capillary number for the visco-inertial regime can also be deduced from the data. Dividing the film thickness by the Taylor thickness provides curves increasing from 1 at low capillary number to some maximum (typically of order 1.5), before decreasing because of the geometric convergence imposed by the tube. Ca** is defined as the value of the capillary number at this maximum. We report in Fig. 6 the values of Ca** drawn from the data as a function of the dimensionless number $\eta^2/\rho\gamma r$, in the same logarithmic scales as in Fig. 5.

The observed behavior is in fair agreement with Eq. (11) (the full line indicates a slope $\frac{1}{2}$), in spite of a relatively poor precision on these data (stressed by the error bars). Ca** is rather delicate to measure, because the transition between the visco-inertial and the saturation regime is not defined as well as the one between the (well-known) Taylor and visco-inertial regime. However, this simple analysis allows us to define the interval in capillary number where an anomalous behavior due to inertia is observed.

IV. THE VISCOUS BOUNDARY LAYER REGIME

Another natural limit must finally be stressed. Viscous and visco-inertial regimes implicitly suppose that the viscous boundary layer is well developed in the dynamic meniscus. As shown in fiber coating, this assumption is not necessarily satisfied at a very high deposition velocity.[7] In this limit, the thickness of the deposited film can be limited by the viscous boundary layer, which develops in the drop as it is moved. At the rear of the drop, its thickness δ is simply given by balancing inertia and viscosity, which yields the classical Prandtl law:

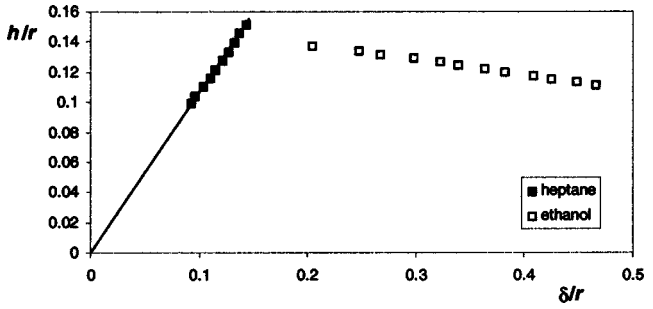

FIG. 7. Film thickness h at high speed for two series of experiments, as a function of the thickness of the viscous boundary layer δ given by Eq. (12). For a liquid such as ethanol at intermediate velocities, the data form a decreasing line in this plot, because the entrainment law (either visco-capillary or visco-inertial) is an increasing function of the velocity. It is no more the case for a centimetric drop of heptane (of very low viscosity) moved at a high speed (up to 1.2 m/s for these data). Then, the deposition law is indeed found to obey Eq. (12) (deposition of only a viscous boundary layer): the full line is the curve $h=\delta$.

$$\delta\sim\left(\frac{\eta L}{\rho V}\right)^{1/2}, \tag{12}$$

where L is the drop length (defined as x_f-x_r, in Fig. 1). If δ is larger than the thickness discussed in Eqs. (4), (5), or (8) (i.e., for a small velocity, a viscous fluid, or a long drop), then the latter laws should be valid. Otherwise, the formation of the boundary layer should limit the fluid deposition, and Eq. (12) should be the law for the film thickness. Since it corresponds to a high speed regime, the Weber number will be (most generally) much larger than one, which justifies that capillary effects (i.e., the basis of Bretherton–Taylor discussion) no longer have to be considered.

To get a chance to observe this regime, we performed a series of experiments with heptane, a liquid of very low viscosity ($\eta=0.4$ mPa s), moved in a large tube ($r=1{,}4$ mm). Then, the drop length was measured as a function of time, from which the film thickness h could be measured and compared with the calculated thickness δ of the viscous boundary layer. In Fig. 7, h is plotted versus δ. The data obtained with heptane (closed squares) are compared with the ones previously obtained in the visco-inertial regime with ethanol (open squares). In addition, the line $h=\delta$ is drawn as a full line.

In the visco-inertial regime (open squares), such a plot generates a decreasing variation, since a higher velocity makes both the film thickness increase (see Figs. 3 and 4) and δ decrease [Eq. (12)]. Moreover, the data clearly lie below the line $h=\delta$, which was stressed to be a condition for this regime to be valid. On the other hand, data with heptane are observed to obey the equation $h=\delta$. Thus, the thickness of a film deposited at a very high velocity is indeed found to be limited by the development of the viscous boundary layer. Moreover, the numerical coefficient deduced from the data (Fig. 7) is found to be close to 1, which remains to be quantitatively understood.

V. CONCLUSION

Thanks to a series of experiments at high speed with liquids of low viscosity, we could provide new data on the old problem of the amount of liquid left behind a wetting drop moved in a tube. Above a threshold in velocity (typically of order 1 m/s), the film was found to be thicker than a Taylor film, for which the thickness is fixed by a balance between capillarity and viscosity. This deviation toward Taylor's law was attributed to inertia, but the analysis was complicated by the fact that this visco-inertial (thickening) effect is superposed with geometric effect, which finally makes the film thickness converge to a finite fraction of the tube radius, because of the confinement due to the geometry. Thanks to scaling arguments, we could propose a first-order analysis, allowing us to describe the interval in capillary number where the visco-inertial regime develops. A numerical solution remains to be achieved, in order to provide a fully quantitative description of the data. At a still higher velocity V, the finite length L of the drop was found to be responsible for a new regime: then, the drop only leaves behind the viscous boundary layer which can develop on the time scale L/V.

ACKNOWLEDGMENT

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