Journal of Colloid and Interface Science **243,** 262–264 (2001)
doi:10.1006/jcis.2001.7891, available online at http://www.idealibrary.com on IDEAL®

NOTE

Falling Slugs

The fall of viscous slugs in vertical capillary tubes is described. Deviations toward Poiseuille law are analyzed by taking into account the dissipation in menisci, together with the existence of a film behind the slug. Slugs are found to fall slower in dry tubes than in prewetted ones, which is quantitatively discussed in term of viscous friction. A criterion for the minimal length of the slug obeying the Poiseuille solution is finally derived. © 2001 Academic Press

Key Words: **Poiseuille law; dynamic contact angle.**

INTRODUCTION

A very simple way to measure the viscosity of a liquid consists of determining the velocity with which a liquid slug moves downward in a capillary tube. If the slug is long enough, the velocity simply results from a balance between viscous force and gravity, which leads to the Poiseuille law,

$$V_o = \frac{\rho g R^2}{8\eta}, \qquad [1]$$

where ρ and η are, respectively, the liquid density and viscosity, and R the tube radius. Equation [1] is independent of the slug length L, since the latter quantity fixes both the weight and the viscous force. If it is obeyed, the liquid viscosity can be simply deduced from the slug velocity. We also supposed that the liquid wets the tube material. In a partial-wetting situation, the slug can stay at rest, because of the contact-angle hysteresis: if the capillary force associated with the latter is larger than the weight (i.e., for short slugs), the slug sticks to the tube.

DRY VERSUS WET

Experiments were made using a glass tube of inner radius $R = 127\ \mu$m and a wetting silicone oil of surface tension $\gamma = 20.6$ mN/m, density 0.95, and viscosity $\eta = 16.7$ mPa.s (measured with a classical Ostwald viscosimeter). The fall velocity is expected from Eq. [1] to be 1.12 mm/s. The different experiments lead to a fall velocity significantly lower than this value, as shown in Fig. 1, where the velocity V is plotted as a function of the slug length L (the horizontal dashed line illustrates Eq. [1]). Because of a film left behind, the drop gets shorter during the fall—but we checked that this variation remained always smaller than 5%, which allowed us to consider the slug velocity a constant during the whole motion.

The shorter the slug, the smaller the velocity, and the effect is more pronounced if the tube is dry than if it is prewetted with a thin film of the same liquid. Such a film was obtained by first moving a slug at a constant velocity along the tube, producing the deposition of a film of constant thickness $h = 1.5\ \mu$m. In both cases, the velocity tends toward the value given by Eq. [1] for very long slugs (at least 5 cm), much larger than the tube radius.

ZOOM ON BOTH INTERFACES

It is also observed that the front meniscus is flatter than the rear one. Two effects contribute to this effect, as shown in Fig. 2: (i) a dynamic (advancing) contact angle sets up at the front, which flattens the meniscus; (ii) the film left behind reduces the radius of the rear meniscus. This kind of situation has been theoretically investigated by Jensen (1), and we restrict ourselves here to the simplified case where the variation of the slug length during its motion can be neglected, along with the flows inside the film.

The radii of curvature R_a ($>R$) and R_r ($<R$) of the menisci generate Laplace pressures which do not compensate and thus are a force opposing the motion. The stationary regime is found by balancing the pressure drop due to viscosity along the slug (the Poiseuille term) with gravity lowered by this Laplace pressure difference. This balance is written as

$$\frac{8\eta V}{R^2} L = \rho g L + \frac{2\gamma \cos\theta}{R} - \frac{2\gamma}{R-h}, \qquad [2]$$

where γ is the liquid surface tension, θ the dynamic contact angle, and h the difference between the tube radius and the actual meniscus radius. As shown by Bretherton (2), h is proportional to h_∞, the thickness of the film deposited behind the rear meniscus. When both the angle and the film thickness are small ($\theta \ll 1$ and $h \ll R$), which physically corresponds to slow motions, Eq. [2] becomes

$$\frac{8\eta V}{R^2} L = \rho g L - \frac{2\gamma}{R}\left(\frac{\theta^2}{2} + \frac{h}{R}\right). \qquad [3]$$

The dynamic angle θ and the thickness h are generated by viscosity, and are limited by surface tension. Thus, they are both expected to be a function of the capillary number Ca, which compares these forces ($Ca = \eta V/\gamma$). In the same limit as previously ($Ca \ll 1$) and for wetting liquids, it is indeed the case: h obeys Bretherton's law ($h/R = 2.9\, h_\infty/R = 3.88\, Ca^{2/3}$) (2), while θ is given by Hoffman–Tanner's law ($\theta = \alpha Ca^{1/3}$, with α a numerical constant of the order of 4–5, for a dry tube) (3, 4).

One way to understand Tanner's law is to derive the viscous force f_η in the liquid wedge (5–7), which can be written, per unit length of the contact line

$$f_\eta = 3\eta \int \frac{V}{\theta x}\, dx, \qquad [4]$$

where x is the coordinate along the tube ($x = 0$ defines the position of the contact line) and the local thickness y in the meniscus is linearized in the vicinity of the contact line ($y = \theta x$). The coefficient 3 comes out from the detailed

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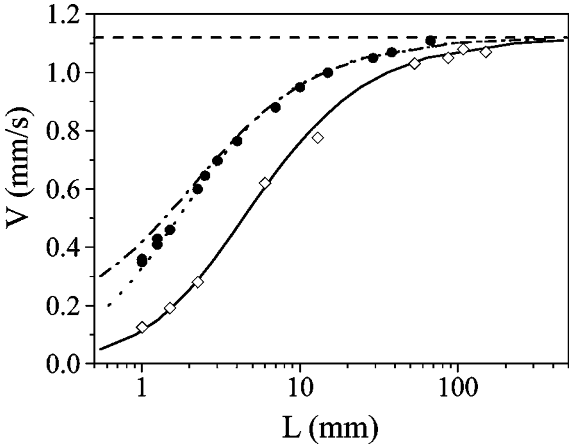

FIG. 1. Velocity V of a liquid slug falling in a vertical tube as a function of its length L. Experiments were done in a glass tube of inner radius 127 μm, using silicone oil (which totally wets the tube wall) of density 0.95, viscosity 16.7 mPa.s, and surface tension 20.6 mN/m. Open symbols, dry tube; closed symbols, tube prewetted with a film of thickness 1.5 μm. The data are compared with Eq. [1] (horizontal dashed line) and Eq. [7]. Solid line, $\Gamma = 15.3$ (dry tube); dotted line, $\Gamma = 5.1$ (prewetted tube); dashed line, correction of Eq. [7] by Jensen's theory for a prewetted tube.

calculation (7). Introducing natural cutoff lengths (a molecular size a and the tube radius R) allows a treatment of the logarithmic divergence of the integral, which finally gives

$$f_\eta = \frac{3\eta V}{\theta}\ln(R/a). \qquad [5]$$

The logarithmic factor $\Gamma = \ln(R/a)$ can be considered a constant, of the order of 13 for a millimetric tube. The stationary shape of the dynamic meniscus can finally be expressed by balancing the viscous force f_η with the capillary one ($\gamma\theta^2/2$), which yields $\theta = (6\Gamma Ca)^{1/3}$. For $\Gamma = 13$, the numerical coefficient in the latter law is 4.3, in close agreement with Hoffman's data (3).

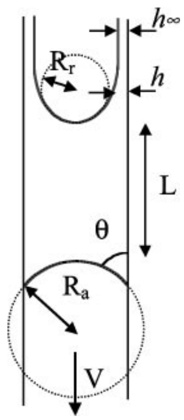

FIG. 2. Effects of the liquid viscosity on the shapes of the front and rear interfaces. The dynamic contact angle at the front flattens the interface and a film is deposited behind, which increases the curvature of the rear meniscus. Both effects reduce the speed of the falling slug.

Introducing Hoffman–Tanner's and Bretherton's laws in Eq. [3] leads to the equation for the motion

$$\frac{8\eta V}{R^2}L = \rho g L - \frac{2\gamma}{R}\beta Ca^{2/3}, \qquad [6]$$

with β a numerical coefficient ($\beta = (6\Gamma)^{2/3}/2 + 3.88$) of the order of 10, for a dry tube. Thus, the deviation toward Poiseuille law expressed by Eq. [6] is mainly due to the dynamical angle at the front, since the correction due to the deposited film is 30% of the previous one. Equation [6] can finally be simplified, using the maximum velocity V_o (Eq. [1]) and a length L_o defined by $L_o = \beta(\gamma R/8\rho g)^{1/3}$. We find

$$L = L_o\frac{(V/V_o)^{2/3}}{1 - V/V_o}, \qquad [7]$$

where V_o clearly appears to be the limit for the velocity at large slug lengths L. For a slug length equal to the characteristic length L_o, V is only 43% of V_o. L_o only depends on the static parameters of the problem (γ, ρ, and R), and is typically of about several millimeters.

COMPARISON WITH THE DATA

Figure 1 shows that Eq. [7] fits quite well the experimental data obtained with a dry tube (open diamonds). The figure also stresses the (logical) increase of the slug velocity due to the prewetting film. This can be understood phenomenologically as a reduction of the coefficient Γ in Tanner's law, and consequently of L_o in Eq. [7]: since the slug is lubricated by the film, the minimal cutoff length in the derivation of Eq. [5] is no longer a molecular size, but can be taken as the thickness h_0 of the prewetting film. Γ is written as $\ln(R/h_0)$, which gives $\Gamma = 4.5$ for $h_0 = 1.5$ μm, in good agreement with the fit displayed in Fig. 1 (in dotted line). Another solution to the problem was recently proposed by Jensen, who carefully derived the Stokes equation for the same geometry and boundary conditions (1). In the case when h_0/R and Ca are of the same order, he found that the presence of a film induces an additive correction to Tanner's law: $\theta^2 = (13.5Ca)^{2/3} - 2.55h_0/R$. Then, Eq. [3] is corrected as well as Eqs. [6] and [7] which can finally be compared with the data (dash-dotted line). A good agreement is found once again between the experimental data and the argument presented above. More systematic experiments remain to be done to discriminate between the models and to more precisely determine the friction, in particular as a function of the thickness of the prewetting film.

The characteristic lengths L_o are respectively found to be 3.8 mm for the dry tube and 1.6 mm for the prewetted one. This experiment finally provides an experimental framework for simple measurements of the viscosity using falling slugs. It is found that slugs much longer than L_o must be used ($L \gg L_o$), which in practicality means slugs of several centimeters. Then, corrections due to the dynamic angle and to the presence of a film at the rear of the slug can be ignored, and Eq. [1] simply used.

Note finally that this analysis is restricted to the use of small capillary and Reynolds numbers. At large Ca and for a forced slug, both the dynamic angle (which reaches 180°) and the film thickness converge toward a limit independent of Ca (8). But a slug always falls at a capillary number smaller than 1: Equation [1] shows that the capillary number is at most $R^2\kappa^2/8$, noting κ^{-1} the capillary length. Since we have $R < \kappa^{-1}$, we immediately get $Ca < 1$. On the other hand, the Reynolds number $\rho RV/\eta$ can become of the order of 1 or larger, using liquids of small viscosity. Then, the problem formally becomes much more complicated, because of (small) corrections on both the dynamic angle (9) and the thickness of the deposited film (10).

ACKNOWLEDGMENT

It is our pleasure to thank Oliver Jensen for a critical reading of the manuscript and for very useful suggestions.

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José Bico
David Quéré[1]

Laboratoire de Physique de la Matière Condensée
URA 792 du CNRS, Collège de France
75231 Paris Cedex 05, France

Received February 2, 2001; accepted July 30, 2001

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[1] To whom correspondence should be addressed.