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ABSTRACT

Steady motion of long, non-wetting droplets carried by a surrounding liquid in a circular capillary has been the subject of many experimental, theoretical, and numerical simulation studies. Theoretical approaches, even after the application of lubrication approximation in hydrodynamic equations and after neglecting inertia and gravity effects, still lead to a numerical procedure to determine the speed of a droplet or the thickness of the film between a droplet and the wall of the capillary. Here, we develop the lubrication approximation further to introduce an analytical formula for the speed of droplets as a function of the capillary number and of the ratio of the viscosity coefficients of the two immiscible phases. We achieve this by identification of a scaling function within the lubrication approximation. The equations that we propose here corroborate well with the results of numerical simulations of droplet flow in circular capillaries.

INTRODUCTION

We consider the stationary motion of a long, non-wetting viscous droplet in a tube of circular cross section. The practical motivation for the interest in the speed of droplets is fueled by the dynamic development and use of the techniques of droplet microfluidics. Droplets are used in biological experiments to cultivate microorganisms[1] and in chemical experiments as reactors.[2] The ability to link the mobility (or speed) of droplets with the material parameters of their content could allow the development of label free methods to infer properties of droplets from their speeds, as, for example, to judge the density of a bacterial culture inside the droplet from the measurement of its speed.[3]

In this article, we describe an intermediate step to achieve this goal by introducing an algebraic formula for the thickness of the film between a droplet and a capillary and for the speed of a long, non-wetting droplet in a channel of circular cross section.

Fairbrother and Stubbs were one of the first who raised the question whether the speed of the flow of a gas bubble moving in a tube filled with a liquid may serve as a proxy for the speed of the liquid itself.[4] The speed of a long, non-wetting droplet which flows in a steady motion in a tube also differs from the average speed of the surrounding (continuous) liquid that wets the walls of the tubing.[5] Between the droplet and the wall, there is a thin wetting film of the continuous liquid. As a result, the cross section swept by the droplet is smaller than the lumen of the tube (Fig. 1). We study the mobility β of the droplet defined by the ratio of the speed of the droplet U to the average speed of the continuous phase

$$\beta = \frac{U}{V}. \tag{1}$$

Fairbrother and Stubbs observed that the length of a bubble l does not affect its mobility, when the length is larger than three tube radii, $l > 3r$. A theoretical expression of the mobility of a bubble has been introduced by Bretherton.[6] To find the mobility of a bubble, Bretherton used Stokes equations with surface tension on the liquid's interface. In the region where the profile of the interface is almost parallel to the channel's wall, Bretherton neglected perpendicular components of the velocity (lubrication approximation). He also considered no-slip boundary conditions on both the interfaces—between the droplet and the continuous liquid and between the continuous phase and the walls of the channel. This leads to an ordinary differential equation for the profile of the film—i.e., the thickness of the film along the length of the droplet. Bretherton also used the fact that at small values of the capillary number, the front of the bubble has a hemi-spherical shape. By matching the curvature of the profile of the film with the curvature of a semi-spherical cap in a small-slope region, he was able to calculate the film thickness, $\epsilon \equiv b/r$,

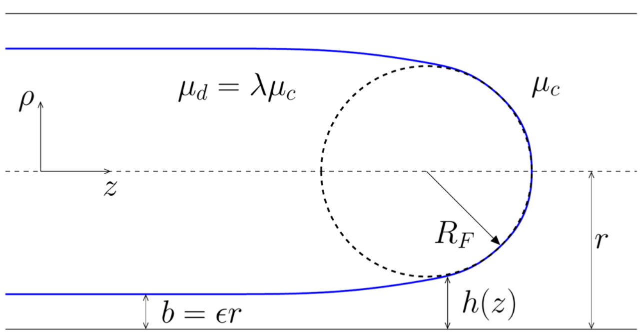

FIG. 1. Schematic sketch of the droplet moving in a circular tube.

as a function of the capillary number

$$\epsilon = 0.643(3Ca)^{\frac{2}{3}}. \tag{2}$$

Bretherton also derived the formula for the mobility which we represent by

$$\beta = 1 + 1.29(3Ca)^{\frac{2}{3}}. \tag{3}$$

The film thickness and the mobility of a bubble are determined solely by the capillary number $Ca = (U\mu_c)/\sigma$, which contains the viscosity of the continuous phase, μ_c, and the surface tension, σ. Bretherton obtained the above formula for a droplet with zero viscosity, $\mu_d = 0$, in a tube in the limit of small capillary numbers by neglecting the effects of inertia in hydrodynamic equations, neglecting also gravity effects, and assuming uniform distribution of the surface tension.

Bretherton's approach has later been extended to the case of a viscous droplet. Finite viscosity of the liquid inside the droplet requires taking into account viscous forces exerted by the droplet phase on the surrounding continuous liquid. The procedure can be found, e.g., in the papers of Schwartz *et al.*[7] and Teletzke *et al.*,[8] and it is also described in detail in the recent paper of Balestra *et al.*[9] Within the lubrication approximation, Bretherton neglected the components of the velocity and pressure field in Stokes equations which are perpendicular to the tube axis. He used this assumption in the layer between the droplet and the wall of the channel. In the case of a viscous droplet, lubrication approximation is extended for the velocity field inside the droplet.[7–9] For a viscous droplet, formula (2) generalizes to the following equation:

$$\epsilon = (3Ca)^{\frac{2}{3}} P(\lambda\epsilon) \tag{4}$$

[cf. Eq. (3.18) in Ref. 7], with function $P(m)$ which Schwartz *et al.*[7] determined numerically. For a given viscosity ratio λ and the capillary number Ca, one must solve numerically Eq. (4) to calculate the film thickness, ϵ. Knowing the film thickness, one can determine the mobility of a long viscous droplet from the formula of Goldsmith and Mason[5]

$$\beta = \frac{1 + \left(2\epsilon - \epsilon^2\right)(-1 + 2\lambda)}{1 + (4\epsilon - 6\epsilon^2 + 4\epsilon^3 - \epsilon^4)(-1 + \lambda)}. \tag{5}$$

According to our knowledge, the above procedure is the state of the art, i.e., the simplest method to calculate the film thickness around a long non-wetting droplet and its speeds in the limit of small capillary numbers within the lubrication approximation. Equation (4) cannot be solved analytically, as it requires iterative numerical approximations. Here, we show that further analysis within the lubrication approximation for viscous droplets is possible.

First, we identify a scaling equation for the film thickness. Second, we derive algebraic formulas to determine the mobility of a long, non-wetting droplet in terms of the capillary number and the contrast of viscosity coefficients between the droplet and continuous liquids.

RESULTS

The starting point for the derivation is formula (4). It has three independent parameters ϵ, λ, and Ca. When used in such a form, it must be solved numerically for each pair of values of Ca and λ. Here, we show that the notation can be simplified to avoid the need for numerical solutions. We multiply both sides of Eq. (4) by the viscosity ratio λ, obtaining $\lambda\epsilon = \lambda(3Ca)^{\frac{2}{3}} P(\lambda\epsilon)$. The resulting equation can be expressed in terms of only two independent parameters

$$m \equiv \lambda\epsilon \tag{6}$$

and

$$g \equiv \lambda(3Ca)^{\frac{2}{3}}. \tag{7}$$

This substitution simplifies formula (4) to the following equation $m = gP(m)$. Solution of this equation defines m as a function of g, which we represent in the following form:

$$m = gM(g). \tag{8}$$

We determine $M(g)$ numerically and introduce the following fitting formula for our numerical solution:

$$M(g) \approx M_{fit}(g) = b_0 + \frac{g + b_4 g^2 + \left(b_0 2^{2/3} - b_0\right)g^3}{b_1 + b_2 g + b_3 g^2 + g^3}, \tag{9}$$

$$b_0 = 0.643,\ b_1 = 4.109,\ b_2 = 8.906,\ b_3 = 10.144,\ b_4 = 3.575.$$

Figure 2 shows the function $M_{fit}(g)$. The relative error of the fit in comparison with the numerically exact result, $|M(g) - M_{fit}(g)|/M(g)$, is less than 4×10^{-3} for the whole range of $g \in (0, \infty)$, as shown in Fig. 3.

Combination of Eqs. (6)–(8) leads to the following expression:

$$\epsilon = (3\text{Ca})^{\frac{2}{3}} M\left(\lambda(3Ca)^{\frac{2}{3}}\right), \tag{10}$$

and with the fit given by formula (9), it determines the film thickness. Along with formula (5), the above equation also determines the mobility of droplets. For the case of inviscid droplets, $\lambda = 0$, we recover Bretherton's results for the film thickness and mobility given by Eqs. (2) and (3), respectively.

Importantly, the film thickness in Eq. (10) is represented by a single parameter function, $M(g)$. It follows that the combination of the film thickness and capillary number $\epsilon/(3\text{Ca})^{\frac{2}{3}}$ collapses into a single curve, $\epsilon/(3Ca)^{\frac{2}{3}} = M\left(\lambda(3\text{Ca})^{\frac{2}{3}}\right)$ described by the $M(g)$ function. For this reason, we call $M(g)$ the scaling function.

Algebraic formulas (5), (9), and (10) for the film thickness and mobility of droplets contain the capillary number, $Ca = U\mu_c/\sigma$, based on the speed of droplets. It is also interesting to know the form of those formulas in terms of capillary numbers based on the speed

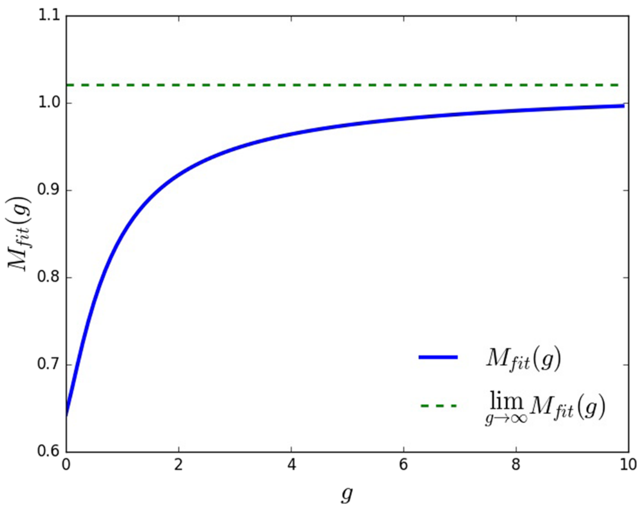

FIG. 2. Scaling function $M_{fit}(g)$ from Eq. (9).

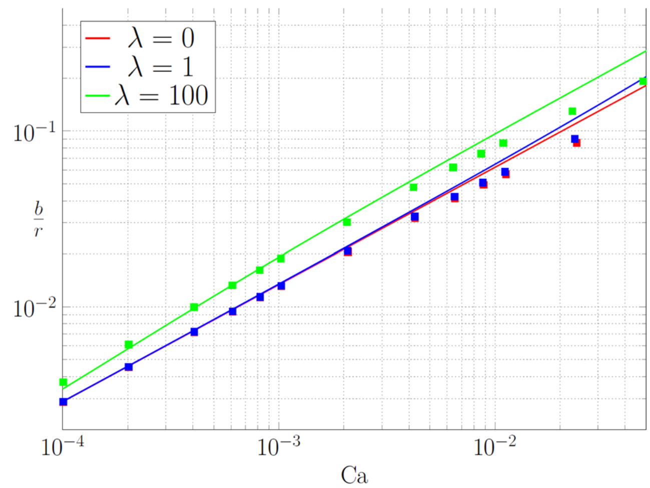

FIG. 4. Film thickness between the wall and the droplet as a function of the capillary number *Ca* for different viscosity ratios λ = 0, 1, and 100. The lines represent lubrication approximation given by Eqs. (9) and (10). The squares represent the numerical simulations from Balestra *et al.*[9]

of the continuous phase, $Ca_c = V\mu_c/\sigma$. Both capillary numbers are related by the mobility, $Ca = Ca_c\beta$. Using this relation in Eqs. (5), (9), and (10), one can obtain expressions for the film thickness and mobility in terms of Ca_c. To the leading order for small capillary numbers, those expressions are the same as Eqs. (5), (9), and (10), but with Ca_c instead of Ca. The difference between using capillary numbers based on the speed of droplets and the speed of the continuous phase in Eqs. (5), (9), and (10) is of the order of ϵ^2 for the film thickness and the mobility.

Figure 4 compares the film thickness obtained by our procedure with the results of numerical simulations. Among different available results,[10–15] we use the results by Balestra *et al.*[9] who performed numerical simulations for a wide range of capillary numbers including the careful study of capillary numbers around 10^{-3}. In the limit of small capillary numbers, we expect a perfect agreement between the numerical and analytical results because the approximations used to derive our formulas are expected to work in this regime. Consequently, for higher capillary numbers, the analytical results should start deviating from numerical results. This picture is confirmed by the comparison presented in Fig. 4. The comparison for viscosity ratios λ = 0, 1, 100 shows that the lubrication approximation described by Eqs. (9) and (10) follows numerical simulations. In both cases, the film thickness increases with the capillary number and decreases with the viscosity ratio. The difference between the lubrication approximation and the numerical simulations increases with the capillary number, but it would be possible to recognize the

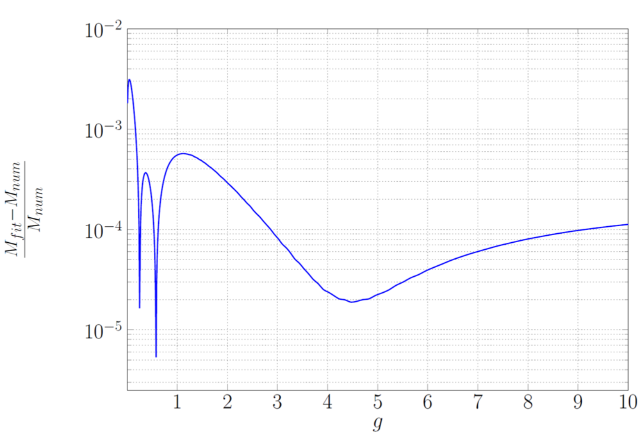

FIG. 3. Difference between the scaling function determined numerically using the method described by Schwartz *et al.*[7] denoted by $M_{num}(g)$ and its fit $M_{fit}(g)$ from Eq. (9).

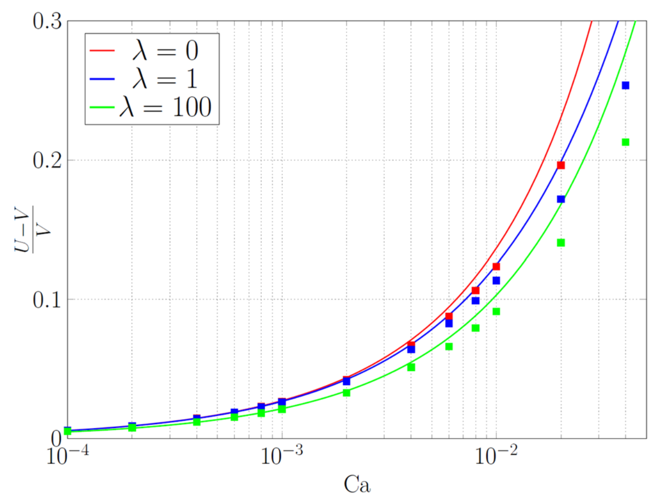

FIG. 5. Droplet's mobility as a function of the capillary number *Ca* for different viscosity ratios λ = 0, 1, and 100. The lines represent lubrication approximation given by Eqs. (5), (9), and (10). The squares represent the numerical simulations from Balestra *et al.*[9]

difference between droplets with $\lambda = 0$ and $\lambda = 100$ for capillary numbers up to $Ca \approx 10^{-2}$. For example, in case of water droplets in hexadecane, this value corresponds to the speed 0.8 m/s. We found similar agreement between the numerical simulations and the lubrication approximation for the case of speeds of viscous droplets. The comparison is presented in Fig. 5. Up to $Ca \approx 10^{-2}$, the difference between numerical and our theoretical values, $|\beta - \beta_{sim}|/(\beta_{sim} - 1)$, is less than 10%.

DISCUSSION

The key point of this article is identification of the scaling function for the film thickness in the lubrication approximation for long, non-wetting droplets. This is expressed by passing from Eq. (4) to expression (10), and it allows the introduction of algebraic formulas (5), (9), and (10) for the film thickness and mobility of droplets. In perspective, it can be used to infer properties of a droplet from its speed in microfluidic experiments, such as recent very precise measurements of the film thickness[16] and speed of droplets for capillary numbers below 10^{-3}.[3,17] The speed of the droplet requires measuring distance and time, which can be done very precisely. For example, Sklodowska *et al.*[3] measured time of passage between two sensors with the relative accuracy about 10^{-4} for capillary number $Ca \approx 10^{-4}$. Answering the question how precisely the viscosity (or surface tension) can be inferred from the lubrication approximation with our algebraic formulas demands further study. There is also an open question whether the above concept of scaling function can be applied in the case of channels of different cross sections, in approaches going beyond lubrication approximations such as Hodges *et al.*,[18] in extensions for higher capillary numbers,[19–21] or in determination of pressure drop induced by the droplet.[9,22]

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