Spontaneous Capillary Flow: Should a Dynamic Contact Angle be Taken into Account?

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Abstract: Spontaneous capillary flow is an interesting solution to move fluids either in microgravity conditions, i.e. in space, or in microfluidic systems, for biotechnology and biology for example. In both cases, gravity is negligible and capillarity becomes the dominant force. Spontaneous capillary flow onset, as well as the dynamics of capillary flows have been recently documented in the literature for channels of different shapes, confined or open. However, the role of the dynamic contact angle is still unclear. This work aims at bringing a new light on the effect of the dynamic contact angle in the dynamics of capillary flows.

Keywords: Spontaneous capillary flow (SCF), Capillarics, Capillary velocity, Dynamic and static contact angles.

1. Introduction

Capillary actuation of fluids is an interesting solution when the gravitational forces are small: this is the case in space, or in microfluidic systems, for biotechnology and biology for example. In both cases, the ratio between gravity and capillary forces characterized by the Bond number is very small, and capillarity is the dominant force.

The onset of spontaneous capillary flow (SCF) and its dynamics have been widely investigated, first in the years 1920s for cylindrical ducts [1-3], then more recently – with the development of microfluidic systems for biotechnology – for channels of various cross-sectional shapes [4-9].

Most of the time, the interpretation of the dynamics of the flow is performed successfully by using a constant contact angle. This point is largely documented in the literature [1-9]. On the other hand, any moving interface forms a dynamic advancing or receding contact angle with the solid wall. Although the study of the dynamic contact angle on the dynamics of wetting has been the subject of many investigations [10-18], the effect of the dynamic contact angle during a spontaneous capillary flow is still debated [19].

In this work, we propose a correction to the spontaneous capillary velocity and penetration distance laws – a.k.a. the Lucas-Washburn-Rideal laws for cylindrical duct and their generalization to channel of arbitrary cross section, recently proposed by Berthier and coworkers [8] – that takes into account the dynamic contact angle. This correction is based on the calculation of the capillary number and the use of a dynamic contact angle correlation reported in the literature.

Comparison between static and dynamic velocities is presented. It is concluded that the dynamic contact angle has an importance only on the few first millimeters of the channel. The length scale...
of the device is then of utmost importance. A non-dimensional number characterizing the importance of the dynamic contact angle is proposed.

2. Theoretical Approach

Let us consider a closed channel of uniform cross section and arbitrary shape (Fig. 1).

![Fig. 1. Sketch of a rectangular (A), and cylindrical channel (B).](image)

It has been shown that the capillary force writes [6-7]

\[ F_{\text{cap}} = \gamma p_W \cos \theta \], (1)

where \( \gamma \) is the surface tension between the liquid and air, and \( p_W \) and \( \theta \) are the wetted perimeter and the dynamic contact angles in a cross-section of the channel respectively.

From a dynamic standpoint, the velocity of the capillary flow can be determined using a balance between capillary forces and friction with the wall [1-3]. In the case of a uniform cross section, the friction force is

\[ F_{\text{drag}} = \int \tau ds = \int \tau dl \int z(t) = \tau p_W z(t) \], (2)

where \( \tau \) is the local wall friction, \( S \) is the wetted surface between the origin and the front end of the liquid flow, \( \Gamma \) is the wetted contour of the cross-section, \( \tau \) is the averaged wall friction in a cross-section, i.e.

\[ \bar{\tau} = \langle \frac{1}{p_W} \rangle \int \tau dl \] (3)

and \( z \) is the distance of the interface from inlet which depends on the time \( t \).

The force balance on the fluid flow is then

\[ \frac{d(mV)}{dt} = F_{\text{cap}} - F_{\text{drag}} \], (4)

where \( m \) is the increasing mass of the fluid in the channel and \( V \) is the average velocity. The mass of fluid in the channel is proportional to the penetration distance, and can be written as

\[ m = \rho z(t) S_c \], (5)

where \( S_c \) is the cross-section area and \( \rho \) is the volumic mass of the fluid. Remarkably that

\[ V = \frac{dz}{dt} \], (6)

substitution of (5) and (6) in (4) yields the differential equation

\[ \rho S_c \frac{1}{2} \frac{d^2 z^2}{dt^2} = \gamma p_W \cos \theta - \bar{\tau} L z \] (7)

In order to solve this differential equation, one needs the determination of the average friction. The Reynolds number of the fluid being small, the flow is laminar. For a Newtonian fluid, the friction \( \tau \) then depends on the geometry of the channel and on the average velocity \( V \). Locally, the wall friction is

\[ \tau = \mu \frac{\partial v}{\partial n} = \frac{\mu V}{\lambda} \], (8)

where \( \lambda \) is the local friction length, \( \mu \) is the dynamic viscosity and \( n \) is the coordinates perpendicular to the wall (Fig. 2).

![Fig. 2. Definition of the friction factor from the velocity profile.](image)

Conceptually, the friction can be averaged in a whole cross-section

\[ \bar{\tau} = \frac{1}{p_W} \int \tau dl = \frac{1}{p_W} \int \frac{\mu V}{\lambda} dl = \frac{\mu V}{\lambda} \] (9)

where

\[ \frac{1}{\lambda} = \frac{1}{p_W} \int \frac{1}{\lambda} dl \] (10)
is by definition the average friction length. Note that the derivation of (9) assumes a constant value of the viscosity, which is the case of Newtonian fluids. The case of non-Newtonian fluids is more complex and will be the subject of further work.

Substitution of (9) in (7) yields the differential equation

$$\rho \, S_c \, \frac{1}{2} \frac{d^2 z^2}{dt^2} = \gamma \, p_w \, \cos \theta - \mu \, p_w \, \frac{dz^2}{dt}$$  \hspace{1cm} (11)

It can be shown that, most of the time in capillary microsystems, inertia can be neglected because the Reynolds number is small, i.e.

$$Re = \frac{V \, w}{v} \ll O(1),$$ \hspace{1cm} (12)

where \(w\) is the characteristic dimension of the channel, \(v\) the kinematic viscosity, and \(O(1)\) means "order of 1". Another approach for the neglecting of inertia can be followed: Equation (11) is a differential equation in \(z^2\). Noting

$$A = \mu \, p_w / (\lambda \, \rho \, S_c)$$ \hspace{1cm} (13)

and

$$B = 2 \gamma \, p_w \, \cos \theta / (\rho \, S_c),$$ \hspace{1cm} (14)

and using the intermediate variables \(Z = z^2\) and \(U = dZ/dt\), Equation (11) can be integrated twice and we find

$$z^2 = \frac{1}{A} \left[ B \, t + C \left(1 - e^{-At}\right) \right],$$ \hspace{1cm} (15)

where the condition \(C = B/A\) stems from the initial conditions. The second term at the right hand side of (15) corresponds to inertia. Note that the value of \(A\) is usually high, because \(p_w\) and \(\lambda\) are of the same order (\(\lambda\) is a fraction of \(p_w\)), and

$$A = \mu / (\rho \, S_c)$$ \hspace{1cm} (16)

For water flowing in a 100 \(\mu\)m \(\times\) 100 \(\mu\)m channel, one finds \(A > 100\). The second term on the right side of (15) is negligible, after the time \(t = 2/A\). The critical time corresponding to the negligibility of inertia is of the order of 1/100 seconds.

Neglecting inertia, we can either use (11) or (15), and a closed-form expression for the travel distance \(z\) is

$$\frac{dz^2}{dt} = \frac{2 \gamma \lambda}{\mu} \cos \theta$$ \hspace{1cm} (17)

The solution of (17) is

$$z = \sqrt{\frac{\gamma}{\mu} \sqrt{\cos \theta} \sqrt{2 \lambda t}}$$  \hspace{1cm} (18)

The travel distance varies as the square root of the time, in accordance with the Lucas-Washburn-Rideal (LWR) law for capillary flows inside cylinders [1-3]. The liquid velocity can be readily derived from (18)

$$V = \sqrt{\frac{\gamma}{\mu} \sqrt{\cos \theta} \frac{\lambda}{2t}}$$  \hspace{1cm} (19)

Under this form, the fluid velocity is the product of the square root of a “physical” velocity \(\sqrt{\gamma/\mu}\) – related to the physical properties of the materials – by the square root of a “geometrical” velocity \(\sqrt{\lambda/2t}\), and by the cosine of the dynamic contact angle. Note that the friction length \(\lambda\) is purely a geometrical data (in the case of Newtonian fluids). The value of \(\lambda\) can be derived from numerous published tables [8].

On the other hand, because \(dz^2/dt = 2z \, V\) relation (17) immediately produces a relation between the velocity and the travel distance

$$V = \frac{\gamma \lambda}{\mu} \cos \theta \frac{1}{z}$$  \hspace{1cm} (20)

### 3. Dynamic Contact Angle

In the literature, many studies concerning the dynamic contact angle of moving interfaces have been reported. Hoffmann was one of the first to study the advancing contact angle of droplets on solid surfaces [10]. He observed the increase of the dynamic advancing contact angle with the increase of the interface velocity. Soon after, a correlation based on the capillary number was proposed. This correlation is now known as the Hoffmann-Tanner-Voinov law.

The dynamic of the moving leading interface of capillary flows has been studied more recently. Not surprisingly, it has been shown that the dynamic contact angle also depends on the capillary number [13-19].

In our case, the capillary number for the flow can be easily derived from relation (20). Let us recall that the capillary number is the ratio of the viscous forces to the surface tension forces

$$Ca = \frac{\mu \, V}{\gamma}$$ \hspace{1cm} (21)

Substitution of (20) in (21) yields

$$Ca = \cos \theta \frac{\lambda}{z}$$ \hspace{1cm} (22)
Relation (22) determines the capillary number — in the case of SCF — as the product of the ratio $\lambda/z$ by the cosine of the wetting angle. Relation (22) shows that the capillary number is proportional to the non-dimensional ratio $\lambda/z$. We underline here the importance of the average friction length $\lambda$, which is a geometrical property of the cross section related to the topology of the cross section perimeter.

Let us now recall some empirical models for the estimation of the dynamic contact angle [13-19]. Noting $\theta_0$ the static contact angle and $\theta$ the dynamic contact angle, Bracke and colleagues [17] proposed the formula

$$\cos \theta = \cos \theta_0 - 2(1 + \cos \theta_0)C_a z \lambda \theta_0$$

(23)

formula which is very close to that of Seebergh and coworkers [18].

$$\cos \theta = \cos \theta_0 - 2.24(1 + \cos \theta_0)C_a^{0.54}$$

(24)

Relations (23) and (24) show that the dynamic advancing contact angle $\theta$ is always larger than the static contact (Young) angle $\theta_0$.

If we substitute the expression (22) determining the capillary number into relation (23), we find the expression of the dynamic contact angle

$$\cos \theta = \cos \theta_0 - 2(1 + \cos \theta_0)\sqrt{\gamma \mu \lambda} \phi(z)$$

(25)

The cosine of the dynamic contact angle appears on both sides of (25) so that (25) is an implicit relation.

Far from the channel inlet ($\lambda/z << 1$), we can substitute to (25) the explicit formula

$$\cos \theta = \cos \theta_0 - 2(1 + \cos \theta_0)\sqrt{\gamma \mu \lambda} \phi(z)$$

(26)

Note that, in the inlet region ($\lambda/z > 1$), the real expression of the contact angle derived from (25) by the resolution of a second order polynomial is

$$\cos \theta = \cos \theta_0$$

(27)

+ $2(1 + \cos \theta_0)\sqrt{\frac{\lambda}{z}} - \frac{\cos \theta_0}{\sqrt{(1 + \cos \theta_0)^2 - \frac{\lambda}{z}}}$

Relations (26) and (27) are plotted in Fig. 3, for a flat rectangular channel (aspect ratio 7); the explicit formulation is legitimate except near the channel inlet ($z=0$).

Substituting back (26) in the velocity expression (20) we find the expression for the capillary velocity

$$V = \frac{\gamma \lambda}{\mu z} \left[ \cos \theta_0 - 2(1 + \cos \theta_0)\sqrt{\gamma \mu \lambda} \phi(z) \right]$$

(28)

Noting

$$\epsilon = \left( \frac{V_{\text{stat}}}{V} - 1 \right)$$

(29)

the relative error due to the use of the static contact angle instead of the dynamic contact angle, where

$$V_{\text{stat}} = \frac{\gamma \lambda}{\mu \cos \theta_0}$$

(30)

we find

$$\epsilon = \frac{V_{\text{stat}} - V}{V_{\text{stat}}} = 2 \left( 1 + \cos \theta_0 \right) \left( \sqrt{\frac{\lambda}{z}} \phi(z) \right)^{1/2} \left( \frac{\lambda}{z} \right)^{1/2}$$

(31)

In fact, near $z=0$, it is relation (27) that should be used instead of (26). A Taylor expansion of (27) for $z=0$ yields

$$\cos \theta = \cos \theta_0$$

(32)

+ $2(1 + \cos \theta_0)\sqrt{\frac{\lambda}{z}} - \frac{\cos \theta_0}{(1 + \cos \theta_0)^2 \frac{\lambda}{z}}$

showing that the dynamic angle is $\pi/2$ for $z=0$. 

![Fig. 3. Variation of the dynamic contact angle as a function of the travel distance $z$, for seven values of the static contact angle {20, 30, 40, 50, 60, 70, 80} degrees. The dotted lines correspond to the implicit solution (27) while the continuous lines correspond to the explicit relation (26).](image-url)
4. Modeling and Experiments

Relation (31) is plotted in Fig. 4 for $\lambda = d / 6$ – which corresponds to a flattened rectangle cross section with $w >> h$, where $w$ is the channel width and $h$ its height – and a static contact angle of 45°. The relative error is small after a few centimeters in the channel. It is however not negligible in the first few millimeters.

Let us now re-analyze the case of water flowing in a rectangular channel composed of COC walls and a thin plastic cover (aspect ratio 0.3), described in [8]. We find that the best fit with a static contact angle is 49°, while it is approximately 47° using the dynamic formulation (Fig. 5).

5. Discussion and Conclusion

The effect of dynamic contact angle in spontaneous capillary flows (SCFs) is still controversial in the literature. Many papers report that the dynamic contact is not very different from the static contact angle [8, 20-22]; on the opposite some papers point out its importance [23-24].

This work shows that all depends on the value of the capillary number, and defines a new expression for the capillary number based on the channel geometry and the wetting angle

$$Ca = \cos \theta \frac{\lambda}{z}$$  \hspace{1cm} (33)

This expression of the capillary number shows that when the ratio between the average friction length and the penetration distance is small, the effect of the dynamic contact angle can be neglected. For closed channels, using the expression of the friction length derived in [8], one can rewrite (33) as

$$Ca = \cos \theta \frac{8}{f Re} \frac{D_H}{z},$$  \hspace{1cm} (34)

where $D_H$ is the hydraulic diameter and $f Re$ the Fanning coefficient [25-29]. For example, for a cylinder of radius R, the friction length is

$$\lambda = \frac{R}{4}$$  \hspace{1cm} (35)

and the capillary number is

$$Ca = \frac{\cos \theta R}{4} \frac{1}{z}$$  \hspace{1cm} (36)

Assuming that the capillary effect can be neglected as soon as $Ca < 5 \cdot 10^{-3}$, i.e., and assuming a static contact angle of 45°, and an average dynamic contact angle of 67° (median distance between 90° and 45°) one finds that if

$$40 R > z > 20R$$  \hspace{1cm} (37)

the dynamic contact angle can be neglected. This limit is close to that found by Kim and Lee: 5 mm for a cylinder of 200 µm diameter [24].

It is concluded that the importance of the dynamic contract angle strongly depends on the characteristic length of the microfluidic device. The dynamic contact angle is not negligible for very short capillary channels; conversely it is negligible for long channels.

References

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