Two-phase capillary flow in open uniform cross section microchannels and the transport of capillary wagons

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ABSTRACT

Capillary open microsystems are increasingly used in biotechnology, biology, thermics and space science. Theory, numerics and applications of open microfluidics have considerably progressed during the last years. Two-phase capillary flow is however a field that still remains unexplored. In this early work, a theoretical basis for the spontaneous capillary two-phase flow in open channels is presented. A numerical investigation of the morphologies of plugs has been done using the Evolver software. Finally preliminary experimental results are presented.

Keywords: Spontaneous capillary flow (SCF), capillary wetting, two-phase microflow, plug.

1 INTRODUCTION

Capillarics—as defined by Safavieh and Juncker [1] —has seen considerable development recently. This field of science intersects in different domains such as biology, biotechnology, space science and thermics. While capillarity in closed, cylindrical tubes is known since the first studies of Lucas, Washburn, Rideal and Bosanquet [2-3], fundamentals of capillarity in arbitrary channel geometries-including open channels- are very recent [4]. But the physics of two-phase capillary flows is still unexplored.

In this work, a first basis for two-phase capillary flows is presented, in the case of uniform cross section open channels. The morphology of the plugs has been investigated using the Surface Evolver software [5], showing different "modes", i.e. morphologies of the plug and carrier fluid. Finally, preliminary experiments are reported and compared to the numerical approach.

2 THEORETICAL APPROACH

First, the approach to determine SCF for a single liquid is recalled, and then the approach is extended to a two-phase flow.

2.1. Single liquid

Our starting point is the same as that of the single-phase spontaneous capillary flow: it is the Gibbs thermodynamic equation (Gibbs 1873) [6] which is

$$dG = \sum_i \gamma_i \, dA_i - p\, dV - S\, dT , \tag{1}$$

where G is the Gibbs free energy, A_i the (multiple) liquid surface areas, γ_i the surface tensions at the interfaces A_i, V the liquid volume, p the liquid pressure, S the entropy and T the temperature. Assuming a constant temperature, we consider the two following cases: first, the liquid volume is constant, as for a drop with negligible or very slow evaporation, and second an increasing volume of liquid associated to a capillary flow. In the first case, (1) reduces to

$$dG = \sum_i \gamma_i \, dA_i \; . \tag{2}$$

The equilibrium shape and position of the droplet is obtained by finding the minimum of the Gibbs free energy. In this case the minimization of the Gibbs energy is equivalent to the minimization of the liquid surface area. This approach is widely documented in the literature [4,7]. The second case—that of a SCF—is different: there is not a constant volume of liquid in the system ($dV \neq 0$), and the system evolves in the direction of lower energy. Hence

$$dG = \sum_i \gamma_i \, dA_i - p \, dV < 0 . \quad (3)$$

Then

$$\sum_i \gamma_i \, dA_i < p . \quad (4)$$

In the case of a spontaneous capillary flow (SCF), the inlet pressure is zero, and the pressure is then less than zero everywhere. Relation (4) becomes

$$\sum_i \gamma_i \frac{dA_i}{dV} < 0 , \quad (5)$$

where the index i defines each one of the neighboring media. A_i, γ_i denote the surface area, surface tension of the liquid with medium i (figure 1).

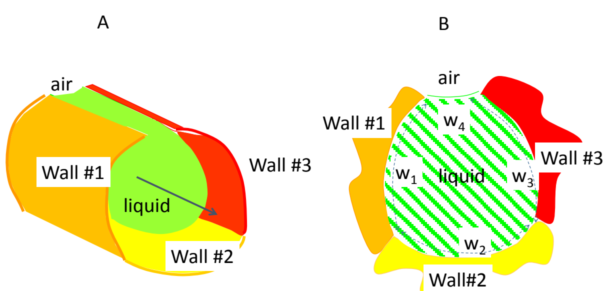

Fig.1. Cross-section of a partly open composite microchannel. The areal fractions are $f_i = w_i / L$ where $L = \sum_i w_i$.

In the case of a single liquid in contact with different composite walls and air, after some geometrical and algebraic considerations [4,8], it has been shown that (5) can be cast in the form of a condition on the generalized Cassie angle θ*

$$\cos\theta^* = \sum_i \left(f_i \cos\theta_i \right) > 0 . \quad (6)$$

For a single flow liquid, the SCF condition is that the generalized Cassie angle is less than 90°.

2.2. Two-phase microflow

Let us now consider a two-phase flow where a plug of liquid is located in front of another immiscible liquid that would experience SCF if alone in the channel.

Let us denote with the index 1 the SCF primary or carrier liquid (and a summation index i), and index 2 the plug (with a summation, index k). Hence, from the Gibbs law, the surface energy for liquid 2, which is of constant volume (dV_2=0), satisfies

$$\sum_k \gamma_{k,2} \, dA_{k,2} = 0 , \quad (7)$$

Let us write now the Gibbs condition for both liquids

$$\sum_i \gamma_{i,1} \, dA_{i,1} - p_1 dV_1 + \sum_k \gamma_{k,2} \, dA_{k,2} - p_2 dV_2 < 0 \quad (8)$$

Using (7), and because dV_2=0, we find that the condition for SCF is identical to that of liquid 1 alone, i.e.

$$\sum_i \gamma_{i,1} \frac{dA_{i,1}}{dV_1} < 0 . \quad (9)$$

In other words, the presence of the plug does not alter the SCF condition for liquid 1. <u>Hence an immiscible plug never stops the flow (in a uniform cross section channel).</u>

In the following section, this condition is verified numerically using the software Surface Evolver.

3. NUMERICAL APPROACH

The software Surface Evolver [5] can be used to predict the SCF condition [8]. It is emphasized that Evolver cannot treat the dynamics of the flow, but it iteratively relocates the interface to lower the energy. In the case of SCF, no equilibrium location exists, but we can still use Evolver to predict if the fluid moves and the direction of motion.

An Evolver data file for this type of problem requires the specification of the different surface tensions (γ_{L1A}, γ_{L2A}, γ_{L1L2}) and the two contact angles θ_1 and θ_2. For this study, many different values of these data have been used. In all cases a two-phase SCF is observed as soon as the main liquid satisfies the SCF condition.

More specifically, taking the example of a rectangular U-groove, three different flow "modes" have been found: (1) a shift mode where the plug is horizontally displaced and translates in front of the flow; (2) a lift mode where the plug is lifted up and the main liquid flows underneath; (3) a bridge mode where the plug sticks to the floor of the channel and the capillary flow of the main liquid flows above, like over a bridge.

Shift mode

By definition the shift mode corresponds to the translation motion of the plug by the primary fluid. Figure 2.A illustrates the shift mode. Both liquids flow at the same velocity, and SCF continues theoretically endlessly.

Lift mode

In this case the primary liquid flows below the plug and progressively lifts the plug. The liquid 1 SCF continues with the plug of liquid 2 on top of it (figure 2.B). As Evolver only handles the statics of the interfaces, it is not possible to know if the plug is pinned and immobile or carried by the SCF.

Bridge mode

In the bridge mode, the primary liquid flows over the plug, which stays in contact with the channel bottom. The SCF continues theoretically endlessly (figure 2.C).

Capillary wagons

A particular case of shift mode is that of "capillary wagons". In such a case, a succession of plugs does not stop the SCF, as shown in figure 2.D. The different plugs are moving together with the carrier fluid.

4. EXPERIMENTS

Experiments have been performed in rectangular or half-cylindrical channels of 1 mm width and 1.6 mm depth, milled in PMMA. The primary fluids are either pentanol, mineral oil, or baby oil. The aqueous plug is made of DI water with or without addition of IPA (5%). Oil is dyed blue using "blue oil' from Sigma Aldrich, while the aqueous phase is tinted red, green or yellow with food coloring.

A bridge mode is observed with baby oil and 5% IPA aqueous solution, as shown in figure 3. Each time the flow passes on top of a plug, the velocity is considerably reduced, but is partly regained afterwards.

Figure 4 shows the travel distance (distance between the inlet and front end of the capillary flow) as a function of time. The three passages over the plugs are clearly seen at t=20, 25 and 45 seconds. One observes a stepwise progression of the main fluid. The progression is very slow when passing over the bridge: the depth of the channel being smaller due to the presence of the bridge, the air boundary is proportionally larger and the capillary force decreases. Hence the capillary flow advances slowly when passing over the bridges.

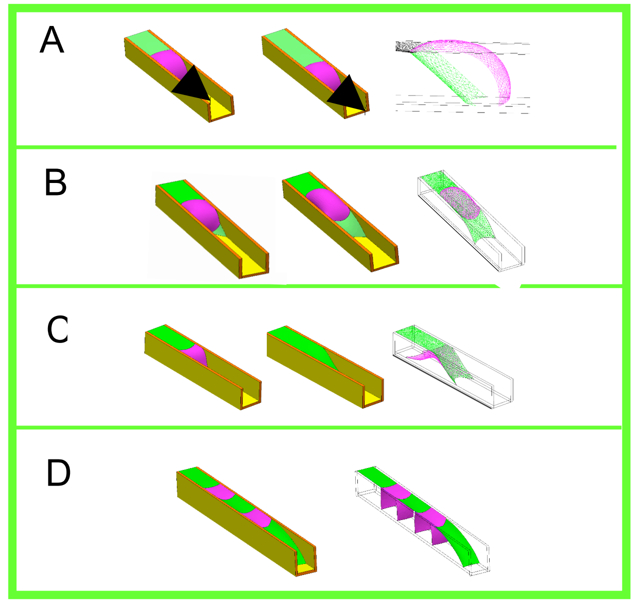

Fig.2 A, shift mode: left and middle, shift motion of the plug; right, detail of the shape of the plug. (γ_{L1A}= 30 mN/m, γ_{L2A} = 72 mN/m, γ_{L1L2}=30 mN/m, θ_1 =50°, θ_2=70°). B, lift mode: left and middle, lift up of the plug by primary fluid; right, details of the shape of the plug (γ_{L1A}= 72 mN/m, γ_{L2A} = 36 mN/m, γ_{L1L2}=60 mN/m, θ_1= 50°, θ_2=110°). C, bridge mode: left and middle, the primary fluid covers the plug; right, details of the shape of the plug (γ_{L1A}= 30 mN/m, γ_{L2A} = 42 mN/m, γ_{L1L2}=16 mN/m, θ_1 =40°, θ_2=55°). D, shift mode for capillary wagons (γ_{L1A}= 42 mN/m, γ_{L2A} = 36 mN/m, γ_{L1L2}=60 mN/m, θ_1= 50°, θ_2=60°).

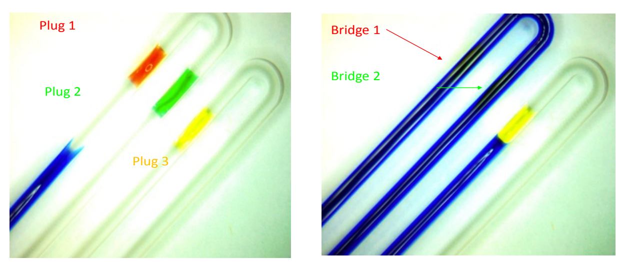

Fig. 3. Bridge mode: a baby oil capillary flow bridges over three successive plugs of aqueous liquid (5% IPA solution).

A mixed mode "shift-lift" is observed in rectangular channels when capillary filaments are present [9]. In such a case the capillary filaments flowing ahead of the bulk flow move the plug. This is the case of figure 5, where a pentanol capillary flow moves a 5% IPA aqueous solution. Interestingly, observing the change of color of the plugs, we deduce that the mixing of the plugs is very fast.

A somewhat similar phenomenon occurs in the round channel (figure 6). In this case, a film of primary liquid (pentanol) "lifts" and displace the plug (5% IPA solution). This mode could be called a "raft"mode.

In view of establishing the existence of capillary wagons, droplets have been deposited on top of a SCF. Figure 7 shows the transport of these capillary wagons.

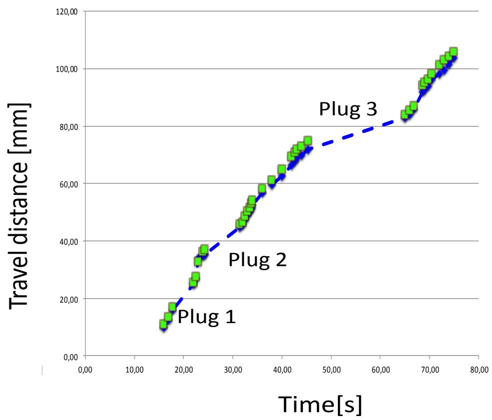

Fig.4. Travel distance vs. time: the three interruptions correspond to the SCF bridging over the three successive plugs. The interruptions are progressively longer.

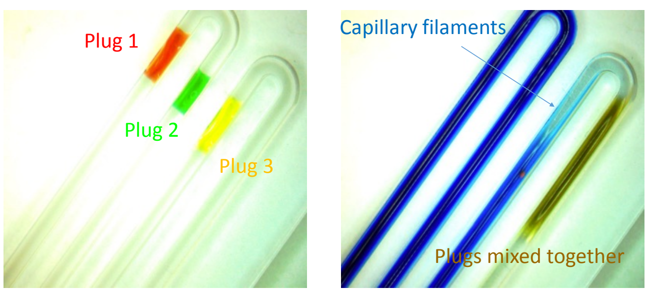

Fig.5. Pentanol SCF successively moves three aqueous plugs in a rectangular channel. The mixing of the plugs is very fast. One can see the depletion of primary fluid at the backend of the plug.

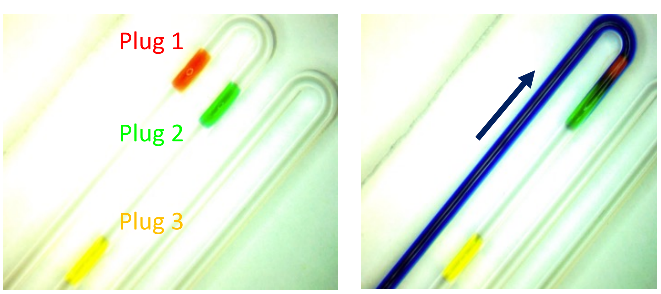

Fig.6. Pentanol SCF successively moves three aqueous plugs in a round channel. The mixing of the plugs is very fast.

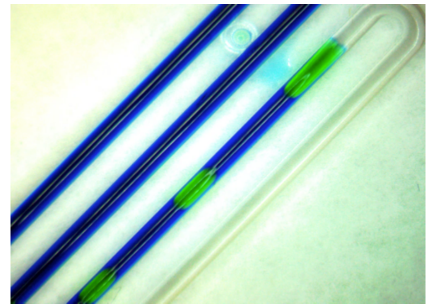

Fig.7. Transport of aqueous plugs (green) by the main capillary flow (pentanol flow, blue).

5. DISCUSSION AND CONCLUSION

In this work, a first approach to the understanding of capillary two-phase flows is performed. Theoretical arguments show that, in a uniform cross section channel, a plug can never stop a SCF. A numerical approach with Evolver confirms this conclusion, and shows that three modes, i.e. three configurations of the flow, can be observed: shift, lift, and bridge modes. It has also shown that a SCF is not interrupted by a succession of multiple plugs (capillary wagons). Finally, the first experiments performed have revealed the existence of modes resembling to that predicted by the numerical approach. Research is presently ongoing to establish all the configurations for capillary wagons in the shift mode.

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