

Some exploratory aspects of two-liquid flows in microgeometries

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Abstract: *An experimental study concerning two-liquid channel flows and the corresponding flow regimes at the millimetric scale is presented for a wide range of the flow rate ratio. The injection of two pairs of miscible liquids leads to the development of wave-like instabilities and to the formation of droplet-like diffusive slugs, that we characterize by means of their oscillation and production frequency. We extend our research to the microscale by testing the droplet formation capabilities of a divergent-convergent microfluidic chamber. A method of deflecting satellite droplets from a layer of monodisperse droplets is observed when an obstacle is present in the microfluidic chamber.*

Key Words: *flow regimes, channel flow, divergent-convergent cell.*

1. INTRODUCTION

Millimetric and micrometric devices are actively investigated due to their potential of offering a framework to study the interaction between miscible and immiscible fluids at small scales. Depending on a variety of control parameters and physical mechanisms, a wide range of hydrodynamic instabilities may be observed.

Numerous studies on microscale flows have been conducted due to their potential impact on many scientific and industrial developments, such as biomaterial synthesis [1], micropower generation [2], and colloidal particle production [3]. Multiphase flows such as droplets or parallel flows are a potential cost-effective alternative to study fast, time-dependent biochemical reactions [4]. In a millimetric channel, one can periodically produce slug-like droplets in a precise manner if the injected fluids are immiscible, as reported in our previous work [5], by manipulating the inlet flow rates. At the microscale, several experimental and computational studies have been carried out on bubble and droplet formation and describe the manner in which these regimes related to the fluid properties, flow parameters, and geometry [6-9].

Other fundamental studies have tackled the identification and classification of the encountered hydrodynamic instabilities such as threading, folding, dripping, jetting, tubing, and displacement [10-14]. Atypical instabilities and flow regimes have been observed if

surfactant is added to the aqueous liquid phase, and thus substantially reducing the interfacial tension between the injected fluids [15, 16].

The possibility to control the hydrodynamic instabilities that may arise at small scales could significantly enhance the mixing and emulsification capabilities of millimetric and micrometric devices. Therefore, in the field of chemical engineering, these platforms facilitate the production of soft materials, such as foams [17, 18] and emulsions [19, 20].

The paper presents an exploratory study of two-liquid flows in microgeometries. We find that the injection of miscible fluid pairs lead to the development of (a) wave-like instabilities, that we characterize by means of their oscillation frequency, and of (b) a production process of droplet-like diffusive slugs. At the microscale, we make observations on a method of producing droplets in a divergent-convergent microfluidic chamber.

2. EXPERIMENTAL DETAILS

We experimentally investigate the flow regimes in a millimeter-sized channel, consisting of three fixed PMMA (polymethyl methacrylate) plates. The device has three inlets, two of which are positioned at a 30° angle to the median line of the central channel, as schematically depicted in figure 1.

The lateral channels have an aspect ratio of $w_2/h = 2/4$ (width per height), while the central channel has an aspect ratio of $w_1/h = 1/4$. We focus on the flow regimes that arise in the main channel, where $w = h = 4$ mm. The millimetric channel is positioned horizontally, with a high-speed camera (FASTCAM Mini UX100) recording the investigated phenomena at over 250 frames per second from above. The experimental data was acquired using the ImageJ software package.

We inject pairs of miscible fluids, either glycerol through the central channel and water through the lateral channels or vice versa. The material properties of the working fluids are presented in Table 1. We denote by η the viscosity and by ρ the density of the fluids, while the subscripts “1” and “2” refer to the dispersed fluid phase, which is injected through the central channel, and to the continuous fluid phase, which is injected through the lateral channels, respectively.

The flow rates of the fluid pairs are controlled by syringe pumps (Harvard Pump 33). For the first pair of miscible fluids, Q_1 is equal to 5, 7, 9, 11, or 13 mL/min while Q_2 is fixed at 7 mL/min, and for the second pair, Q_1 is equal to 2, 4, 6, 8, or 10 mL/min and Q_2 is fixed at 4 and 6 mL/min, respectively.

We consider the characteristic length scale of this millimetric channel to be the hydraulic diameter, which we compute for each inlet channel as $D_h = 4wh/(2w + 2h)$. For the main channel, $D_h = h$, as it has a square cross section. The flow regimes we describe in this paper are discussed in terms of the Reynolds number, $Re = \rho V D_h / \eta$, and the Strouhal number, $Sh = f D_h / V$, where $V = Q/wh$ is the fluid's average velocity at each inlet and f is a frequency that characterizes the periodic oscillations of the fluid. For each fluid pair, we calculate the flow rate ratio as $\varphi = Q_1/Q_2$.

Table 1 – Fluid properties of the investigated fluid pairs at the millimetric scale

Dispersed phase	η_1 [mPa·s]	ρ_1 [kg/m ³]	Continuous phase	η_2 [mPa·s]	ρ_2 [kg/m ³]
Glycerol	1000	1214	Water	1	998
Water	1	998	Glycerol	1000	1214

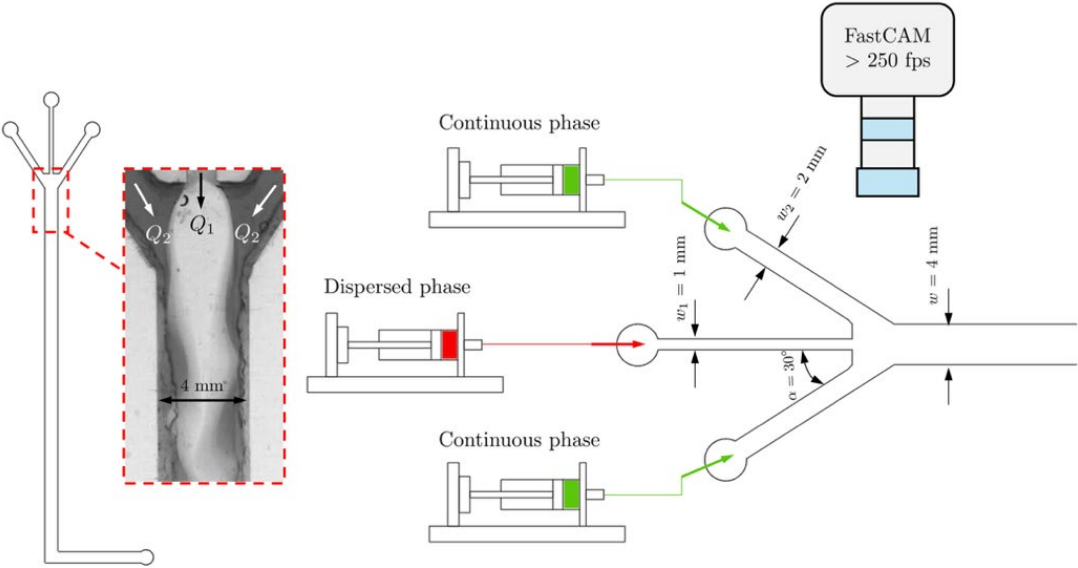


Fig. 1 – Schematic representation of the experimental setup for the millimetric channel

At the micrometric scale, we use a microfluidic platform manufactured using photolithography on a 4 inches silicon wafer. The fabrication method is described thoroughly in our previous study [13].

The microfluidic device consists of (a) three square inlets ($w = h = 100\text{ }\mu\text{m}$) that merge into a single, long microchannel at a right angle flow focusing section, (2) a diamond-shaped diverging-converging microfluidic chamber, and (3) an outlet microchannel (see Figure 2). We place the microfluidic module under an inverted microscope equipped with a high-speed camera working at over 4000 frames per second.

Droplets are produced by continuously injecting a water-glycerol (67% w/w) solution of viscosity $\eta_1 = 20\text{ mPa}\cdot\text{s}$ through the middle inlet microchannel with a volumetric flow rate Q_1 , and silicone oil (PDMS) having the same viscosity $\eta_2 = 20\text{ mPa}\cdot\text{s}$ through the lateral microchannels at a volumetric flow rate Q_2 . The liquids are injected through microfluidic tubing with the aid of two syringe pumps.

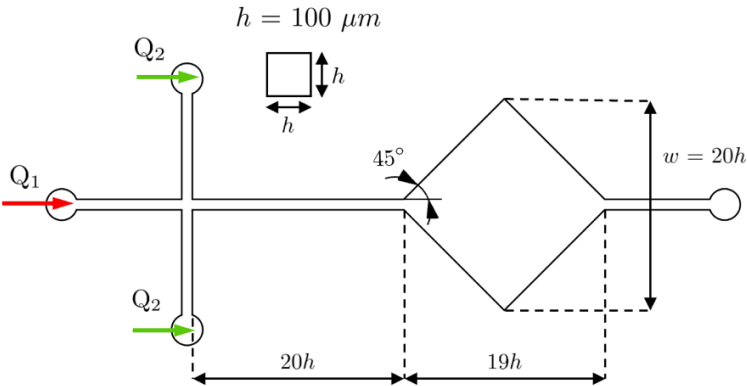


Fig. 2 – Schematics of the divergent-convergent microchannel

3. INSTABILITIES OF MISCIBLE FLUIDS

At the millimetric scale, we explore hydrodynamic instabilities that develop after the Y-junction of the channel. For the first studied case, we inject glycerol through the middle channel and water through the lateral sides.

The glycerol fluid stream flows through the centre and destabilizes once the lateral water streams narrow the space at the junction. The glycerol starts flowing in a wave-like pattern as it advances in the main channel.

This flow regime ($Re_1 \approx 10^{-2}$ and Re_2 between 10 and 10^2) is defined by highly unstable periodic oscillations of the glycerol fluid stream. After approximately 1 cm from the junction, the instability takes the shape of sharper corners and it becomes less visible, as diffusive processes gain importance. Figure 3 shows the dynamics of this wave-like hydrodynamic instability over time.

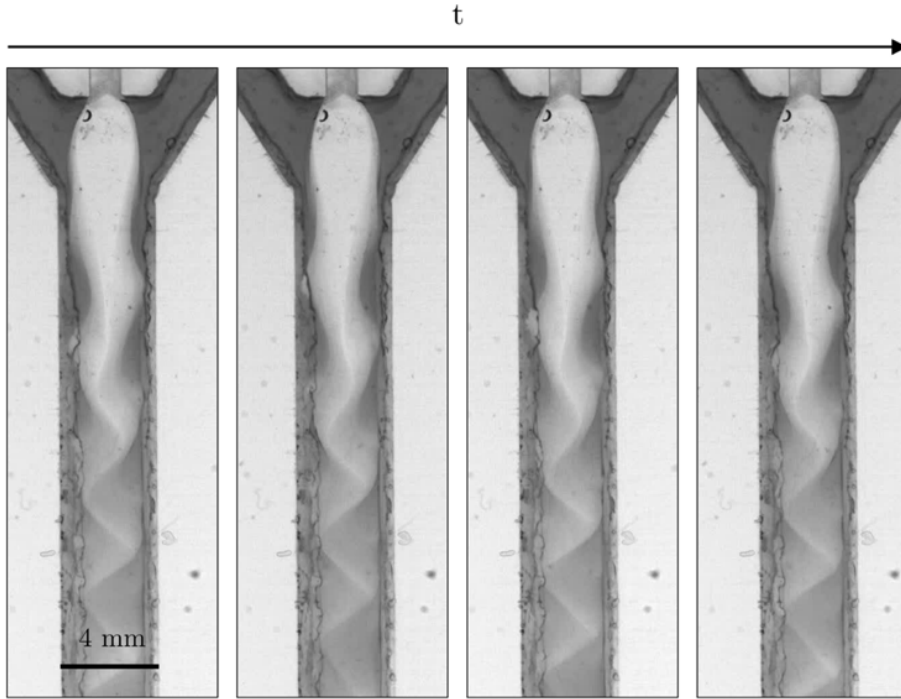


Fig. 3 – Wave-like hydrodynamic instability of miscible fluids in the millimetric channel

We measure the oscillation frequency of the glycerol waves and compute the Strouhal numbers for both fluid phases. We find that the Strouhal number associated with the dispersed phase tends to remain constant, $Sh_1 = 0.25$, when representing it as a function of the flow rate ratio $\varphi = Q_1/Q_2$, as shown in figure 4.

Although the inlet flow rate of the central fluid phase Q_1 is increased gradually, while keeping Q_2 fixed, the Strouhal number does not vary significantly, which indicates that the ratio between the oscillation frequency f and the velocity of the glycerol stream V_1 stays approximately constant (the hydraulic diameter is the same).

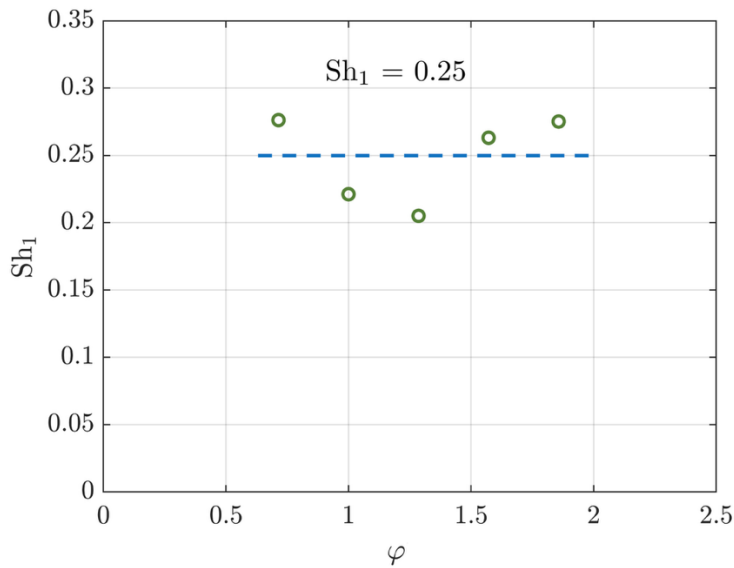


Fig. 4 – Strouhal number for the dispersed phase as a function of the flow rate ratio

In contrast, the Strouhal number associated with the continuous phase Sh_2 increases with the flow rate ratio φ , as seen in figure 5. This dimensionless representation highlights that the oscillation frequency of the waves f increases as the flow rate of the glycerol Q_1 is increased.

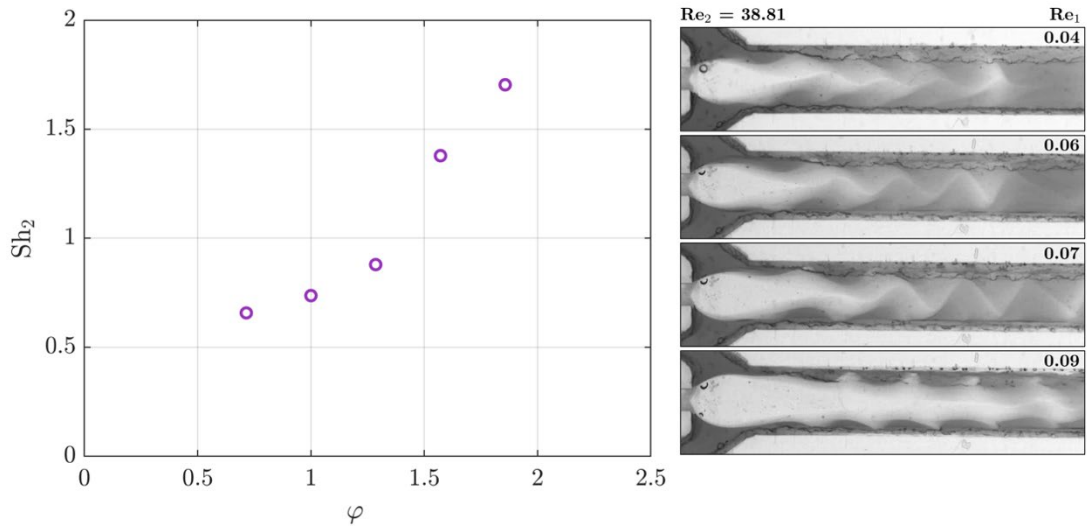


Fig. 5 – Strouhal number for the continuous phase as a function of the flow rate ratio

When injecting water through the central channel and glycerol through the lateral side channels, we report the formation of droplet-like diffusive slugs in the main channel. These diffusive slugs are periodically generated at the Y-junction by keeping the glycerol flow rate constant and varying the flow rate of water.

As the Reynolds number of the central phase increases, the slug’s length tends to increase, as depicted in figure 6. Additionally, they become more pronounced, likely due to an increase in the velocity of the water and thus, the phenomenon becomes dominated by convection rather than diffusion.

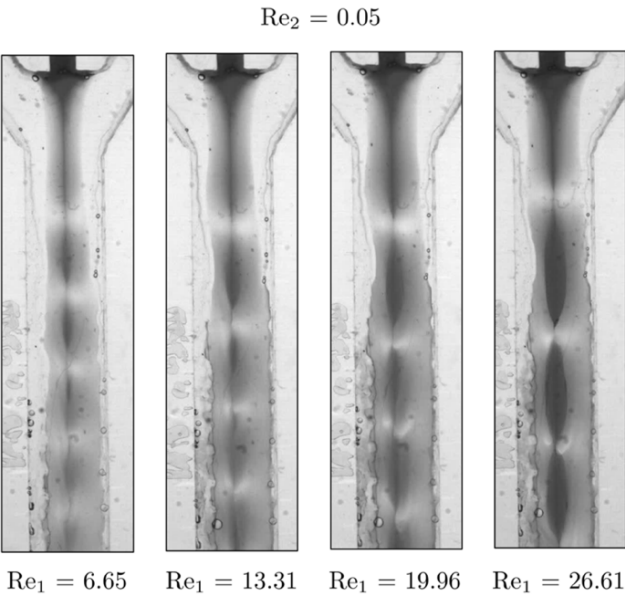


Fig. 6 – The formation of droplet-like diffusive slugs

We measure the first and second lengths (L_1 and L_2) of the diffusive slugs. Figure 7 shows the dependence of the dimensionless lengths L/h on the flow rate ratio φ . We find that the length increases as the flow rate of water increases.

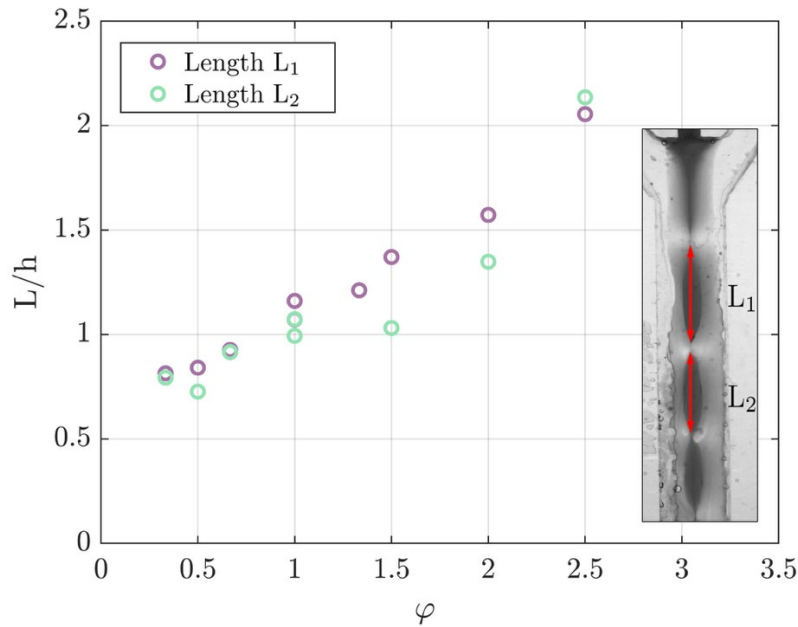


Fig. 7 – The lengths of the first and second droplet-like diffusive slugs in dimensionless form as a function of the flow rate ratio

These apparent droplets periodically “detach” from the central water stream as they flow through the channel, while keeping an approximately equal distance between them. We measure the frequency at which these fluid structures are produced and find that the Strouhal

number calculated for the central phase Sh_1 decreases as the flow rate ratio φ is increased, following a power law, as shown in figure 8-a.

This emphasizes that the frequency decreases as the flow rate of the water is increased. Conversely, the Strouhal number for the continuous phase is constant $Sh_2 = 2.15$ regardless of the flow rate ratio (see Figure 8-b).

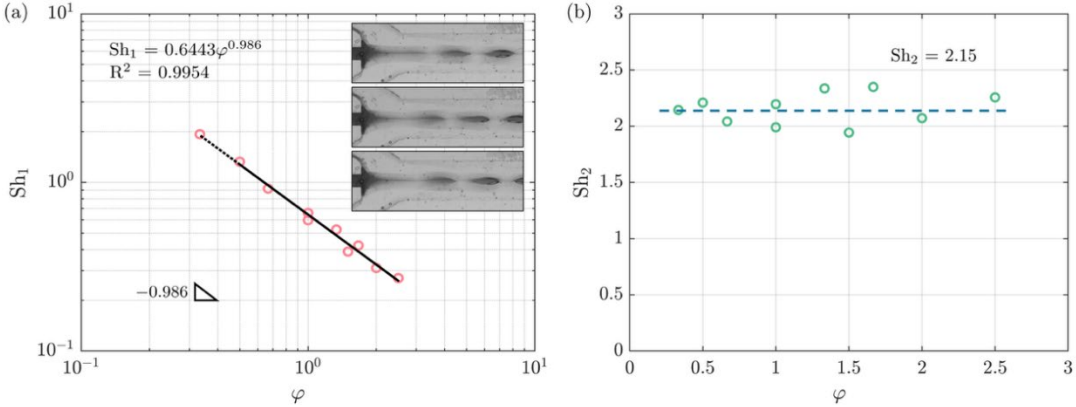


Fig. 8 – Strouhal number for the dispersed phase (left) and continuous phase (right) as functions of the flow rate ratio

4. DROPLET GENERATING PROCESSES AT THE MICROSCALE

The continuous injection of an immiscible fluid pair into a diverging-converging microchannel generates regular trains of monodisperse droplets at the flow focusing section. The droplets are made of a water-glycerol mixture (the dispersed phase), while the continuous phase is composed of a silicone oil. In this section, we experimentally observe the flow behaviour of these trains of droplets as they flow through the microfluidic chamber.

The droplets form at the junction of the device, travel through the main microchannel, and enter the diverging-converging chamber. During the breakup of the droplets from the central fluid stream, the generation of satellite droplets is observed. However, their size is negligible compared to the dimensions of the main droplets. In the microfluidic chamber, the droplets flow sequentially, while the satellite droplets form strings that resemble arcades at the top and bottom of the central droplet train.

For flow rate ratios $0.01 < \varphi < 0.04$, a single train of monodisperse droplets is formed, as depicted in figure 9-a. The distance between the droplets shrinks towards the centre of the microfluidic chamber, as their velocity decreases under the influence of the microchannel's geometry. We noticed that the droplet generation frequency increases with the flow rate ratio $\varphi = Q_1/Q_2$, or as the flow rate of the central phase is increased, and thus, the distance between the droplets becomes even smaller, resembling a chain of pearls (see Figure 9-b).

If we increase the flow rate ratio φ to 0.05 or 0.06, multiple layers of droplets are produced, as shown in figures 9-c and d. As the flow rate of the central phase is progressively increased, we observe ordered arrangements of two or three layers of droplets, while the arcades of satellite droplets flow above and below at a higher distance from the main flow. However, droplet coalescence phenomena appear at a threshold of $\varphi = 0.1$. In that case, one can observe the formation of a bigger droplet towards the end of the microfluidic chamber due to the coalescence of smaller droplets advancing in the microfluidic chamber. If the flow rate ratio exceeds 0.7, droplets are no longer produced.

We experimentally report a method of deflecting the arcades of satellite droplets by designing an “obstacle” in the divergent-convergent microchannel, which in our case is an accidentally pinned larger droplet.

We noticed that such an obstacle can change the direction of the satellite droplets train which could be evacuated from the device if an additional outlet channel is included. Figure 10 shows micrographs of stable droplet generation and deviation of the satellite droplets arcade in the presence of a bigger pinned droplet.

Not only can the satellite droplets be redirected, but also the main layers of droplets, which can have a significant impact on applications where stable droplet formation is essential, such as microreactors for encapsulating cells and chemicals [21, 22].

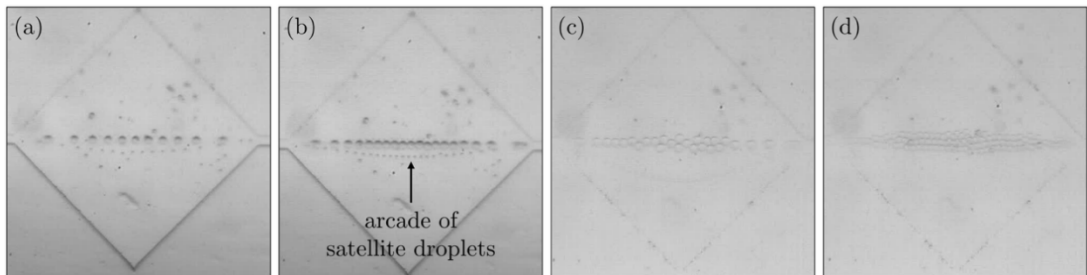


Fig. 9 – Flow regimes for the first fluid pair: (a) $\varphi = 0.01$; (b) $\varphi = 0.02$; (c) $\varphi = 0.05$; (d) $\varphi = 0.06$

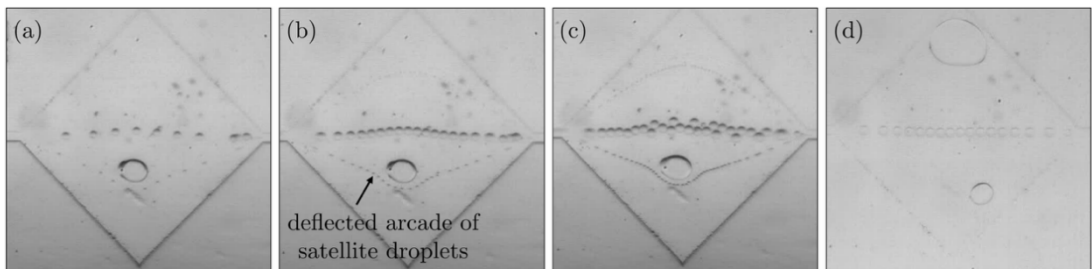


Fig. 10 – Deflection of the lower satellite droplets arcade: (a) $\varphi = 0.01$; (b) $\varphi = 0.02$; (c) $\varphi = 0.07$; (d) $\varphi = 0.2$

5. CONCLUSIONS

In this paper, we study an instability that occurs in the millimetric channel when the working fluids are miscible. This hydrodynamic instability consists of wave-like movements of the dispersed phase as it advances in the millimetric channel. We find that the Strouhal number for the dispersed phase is constant when represented as a function of the flow rate ratio. The Strouhal number for the continuous phase increases with the flow rate ratio. When the miscible fluids are inverted, we report the formation of droplet-like diffusive slugs and find that their dimensionless characteristic length increases with the flow rate ratio. The Strouhal number for the dispersed phase decreases with the flow rate ratio, while the Strouhal number for the continuous phase is constant when represented as a function of the flow rate ratio.

Immiscible fluid flow regimes are studied in the divergent-convergent microchannel. We describe a droplet generating method, emphasizing critical flow rate ratios for the onset of coalescence in the microfluidic chamber. Furthermore, we show that the presence of an obstacle in the divergent-convergent microchannel has the potential of deflecting the satellite droplets from the main flow of droplets. Such experimental observation is of great importance due to its potential to generate a single layer of monodisperse droplets, without satellite

droplets potentially influencing the system. Stable droplet production is generally necessary in the encapsulation of biomaterials or microreactors.

The study explores hydrodynamic instabilities and introduces approaches for generating periodic droplets across both microscale and millimeter-scale systems. The findings are essential in fields including biomedical, chemical, and petroleum engineering, where they may improve mixing of miscible fluids and facilitate emulsification in immiscible fluid systems.

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