Determination of droplet contours in liquid-liquid flows within microchannels

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Abstract. An experimental analysis of the droplet regime with a silicone oil-water two-phase flow within a micro cross-junction, varying the average velocity of the fluids, has been carried out. The micro cross-junction is made as intersection of two glass microchannels with a stadium-shaped cross-section with height $H = 190$ µm and width $W = 195$ µm within the junction and $W = 390$ µm before and after the junction. The water flow rate is broken in droplets having spherical shape with dimensions and velocity that depend on the average velocity ratio imposed. Different kinds of intermittent droplets have been observed, in the ranges of average velocity (0.0105-0.042) m/s and (0.0004-0.0050) m/s for oil and water, respectively. The droplet velocity has been calculated starting from the detection of the shape of the droplets and then by the evaluation of the displacement of the droplets in the unit of time. The images of the droplets have been obtained from a high-speed camera, connected to an inverted microscope. The procedure of water phase contour detection is based on Matlab Image Toolbox scripts.

1. Introduction
In the last years, several microdevices have been developed for the production of droplets and emulsions, which have a large use in many fields, especially in the field of drug delivery [1-3]. The most common microdevices for droplet generation are based on T-junctions, flow-focusing junctions and co-flowing devices (a classification of these devices can be found in [4]). Devices based on cross-junction are examples of flow-focusing devices widely used for droplet-based microreactors and chemical synthesis [5].

Based on the experimental observations collected in the last years, the geometry of the microfluidic microchannels used in these devices plays an important role in controlling liquid–liquid flows but this is not the only parameter which influences the droplet production. In fact, a complete control over the two-phase flow behavior, by these devices, requires a detailed understanding of the bubble or droplet formation mechanisms. One physical approach is to analyze the forces involved in the droplet formation process in microchannels [6, 7]. The dynamics of droplet formation is mainly governed by some variables, including channel geometry, structure and property (such as channel type, dimension and hydrophobicity), fluid properties (such as density, viscosity, interfacial tension and contact angle), and operating parameters (such as pressure, flow rate ratio, temperature, electric field, etc.). These parameters can be typically expressed by means of several dimensionless parameters, noting that...
small geometric length scales typically lead to low Reynolds numbers (Re<1) in microfluidic devices. Therefore, the effect of the inertia forces can typically be ignored in this kind of flow which is close to a Stokes flow. In this condition, the Capillary number becomes the most important dimensionless parameter.

In micro-junctions, three typical flow regimes have been distinguished: squeezing, dripping [8, 9], and jetting [10, 11]. Corresponding to these flow regimes, two main dynamical models for droplet breakup in a microfluidic T-junction, namely rate-of-flow-controlled breakup [8] and shear-driven breakup [5], have been proposed. At low capillary numbers, typically Ca<0.01, that is, the shear stress is much smaller than the interfacial force; droplet breakup is dominated by the pressure drop across the emerging droplet or bubble. In this squeezing regime, the mechanism is known as rate-of-flow-controlled breakup [8], and it is ubiquitous in the confined T-junction system, and also in flow-focusing geometries. This pressure drop results from the blocking by the droplet or bubble which fills the entire cross-section of the channel. Within this squeezing regime the droplet size becomes a function of the flow ratio of the two fluids [9].

The wetting properties of the fluid–wall interface are extremely important in determining whether ordered or disordered flow patterns occur [12]. When the continuous phase completely wets the microchannels, ordered patterns can be obtained. On the contrary, if the wetting is partial, disordered flow patterns are produced. Typically, water-in-oil (W/O) droplets are produced in hydrophobic devices, and oil-in-water (O/W) emulsions are generated in hydrophilic droplet generators [13]. The hydrophobicity or hydrophilicity property of a solid surface can be obtained by means of specific surface coatings [6].

The aim of this work is to study the water droplet breakup mechanism within a hydrophobic cross-junction in a liquid-liquid (silicone oil-water) two-phase flow (water-in-oil). The micro cross-junction is made as intersection of two glass microchannels with a stadium-shaped cross-section having a restriction within the junction. The water droplet breakup is investigated by varying the average velocity of silicone oil between 0.0105 and 0.042 m/s and water between 0.0004 and 0.0050 m/s.

After the reconstruction of the droplet contour, a droplet average velocity value is computed as well as the main characteristic dimension of the droplet, dimensionless diameter, as a function of the Capillary number and the flow rate ratios (water flow rate/silicone oil flow rate) and a correlation has been proposed.

2. Experimental Setup

The experimental apparatus, shown in figure 1, is based on the use of an inverted microscope (1, Nikon Eclipse TE2000-U) illuminated by means of a Mercury lamp (2a, Nikon C-SHG1) from the bottom and a tungsten lamp (2b, Photon Beard PhotonBeam 1000) from the top of the microdevice.

Figure 1. The schematic diagram of experimental setup
An air immersion lens with a numerical aperture $N_A=0.25$ and magnification $M=10\times$ is used (3, Nikon, CFI DS 10X). The microscope is connected to a high-speed camera (4, Olympus I-speed II) in order to acquire a series of images for reconstructing the evolution of water droplets within the cross-junction (7, Dolomite Junction Chip). Due to the high frequency values of the droplet breakup, in each experimental analysis up to 5000 frames per second have been acquired by means of the Olympus I-speed II software (6). A LCD monitor (5) directly connected to the high-speed camera enables the real-time visualization of the flow conditions through the microchannel. The micro cross-junction is obtained as intersection of two microchannels, one straight and one curved, as shown in figure 2a. In figure 2a, one can see that the Dolomite chip has also a T-junction, but in this paper only the results obtained by the cross-junction are presented. Figure 2b shows also a top view of the junction.

![Figure 2. Sketch of Dolomite chip (a) and sketch of the cross-junction (b)](image)

The microchannels have a stadium-shaped cross-section (see figure 3a), with the width $W=390\ \mu m$ and the height $H=190\ \mu m$ before and after the junction. In the junction, the cross-section has again the shape of a stadium, but the width $W_j=195\ \mu m$ and the height $H=190\ \mu m$ (see figure 3b). Both microchannels are made by hydrofluoric acid (HF) etching on a substrate of glass and then thermally bonded with the same material. The average roughness of the channel inner walls declared by the manufacturer is 5 nm. The inner walls of the cross-junction have been treated with a self-assembled monolayer of silane groups, rendering the surface as water repelling (hydrophobic behavior). Even if it is known that microchannels made by a similar manufacturing process can show a real cross-section different from the geometry declared by the manufacturer [14], after a qualitative optical check of the cross-junction by microscope, it has been observed that the shape of the cross-section of the cross-junction is coherent with that declared in the data-sheet of the manufacturer.

![Figure 3. Cross-section out of the junction (a) and within the junction (b)](image)

The two liquids adopted in these experimental tests are silicone oil (SIGMA-ALDRICH), entering in the two perpendicular and opposite streams, the properties of the silicone oil are shown in table 1, and water, entering in straight direction.

| Table 1. The properties of the silicone oil employed for the experiments |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Viscosity (cSt) | Density (g/ml)  | Vapor pressure (mmHg) | Vapor density   |
| 25 °C           | 20 °C           | 25 °C           | 20 °C           | >1 (vs air)     |
| 20              | 0.95            | <5              | 5               |                 |
The liquids are pumped by means of two syringe pumps (Harvard Apparatus PHD4400 Programmable and Cole-Parmer Version Hills); the operative ranges of the flow rates for the syringe pumps are reported in table 2 as well as their typical uncertainty values. The range of the volumetric flow rate, both for oil and water, tested in this paper are shown in figure 4a; water volumetric flow rates between 0.1 ml/h and 1.2 ml/h have been tested as well as oil volumetric flow rates between 2.5 ml/h and 10 ml/h. In terms of average velocity, values between 0.0105 and 0.042 m/s are imposed for silicone oil and between 0.0004-0.0050 m/s for water (see figure 4b).

| Labels in figure 1 | Instrument                  | Minimum flow rate (μl h⁻¹) | Maximum flow rate (ml min⁻¹) | Uncertainty  |
|--------------------|-----------------------------|----------------------------|-----------------------------|--------------|
| 10                 | Harvard PHD4400             | 0.0001                     | 220.82                      | ± 0.35%      |
| 11                 | Cole-Parmer Hills           | 0.2                        | 8.333                       | ± 0.5%       |

A series of 34 different experimental runs have been made: the complete map of the experimental runs is shown in figure 4a by putting in evidence for each test the imposed values of volumetric flow rates both for oil and water and in figure 4b in terms of imposed average liquid velocity.

![Figure 4. Map of the experiments: volumetric flow rate (a), velocity (b)](image)

3. Processing of the images
A typical sequence of snapshots obtained from the fast camera is shown in figure 5; these snapshots have been obtained by imposing a water volumetric flow rate of 0.6 ml/h and an oil volumetric flow rate equal to 7.5 ml/h.

Figure 5 shows the squeezing of the water liquid phase, followed by the breakup and the formation of a water droplet in oil, in a sequence of 30 snapshots, recorded with a frame rate 5000 fps. Each image has a resolution of 320×240 pixels and the time interval between two images is equal to 200 μs. The snapshots are shown in a sequential order; line by line from top to bottom, in each line from left to right (i.e. the 18th snapshot is in the fourth line, the third element from left). Figure 5 shows the good quality of the acquired images with droplet contours well defined and a level of illumination quite uniform in the whole field-of-view. The images allow to recognize with a good resolution the liquid/liquid interface as well as the solid walls of the cross-junction. The central area of the water droplet is characterized by light gray pixels but there is a dark zone around this region which puts in evidence the curved boundary of the droplet.
3.1. Analysis of the images

Figure 5. Raw images showing the squeezing and the breakup of water in oil (W/O) for an imposed water volumetric flow rate of 0.6 ml/h and an oil flow rate of 7.5 ml/h

Figure 6a shows an image chosen from the sequence reported in figure 5 (12th snapshot).

(a) (b)

Figure 6. Raw image chosen to explain the processing sequence (a), complement of the image of the cross-junction without droplets (b)

The raw images of the sequence of figure 5 are treated as numerical matrices having 320×240 elements in which each pixel is associated to a number in the range [0-256] which correspond to different gray levels of the pixel. In order to individuate the water-oil interface by avoiding any interference between the water-oil interface and the channel walls, the complement of an image of the cross-junction filled with a single phase liquid (oil or water) (figure 6b) is used with the aim to compute a characteristic threshold $t$ for the image, which is a normalized intensity of the image, evaluated by means of the Otsu method [18]. By this method, the weighted sum of the variances of two classes of pixels in the image, the background and the foreground pixels, can be minimized.
The result is a characteristic threshold of the image that can be used for converting an intensity image to a binary image (by using `graythresh` function in Matlab). For the sequence of images shown in figure 5, by means of this method a threshold $t=0.47$ has been evaluated. Then, it is possible to transform the intensity values of the gray scale into new values, in order to filter the gray levels and enhance the contrast between background and foreground pixels (by using `imadjust` function in Matlab). The effect obtained by following this pre-processing procedure on the image shown in figure 6a, is shown in figure 7a. It is evident that the background contribution has been neutralized and in the processed images only the presence of the liquid/liquid interface is emphasized. Finally, the obtained gray level maps can be converted into binary images by means of a new threshold, which for the sequence of images shown in figure 5 is $t=0.7$, by using `im2bw` function in Matlab. The binary images corresponding to the 12th snapshot is shown in figure 7b.

3.2. Detecting the edge

In a previous work [17] it has been demonstrated that among different techniques available for finding the edges of an image the Canny method [19] can offer some advantages in terms of compromise between accuracy and computational time. By this technique, one can find edges by looking for local maximal of the gradient of the binary image. To smooth the image, a Gaussian filter is first applied to convolve with the image. Then, the method uses two thresholds, to detect strong and weak gradients, and includes the weak ones in the output only if they are connected to strong gradients. This method is therefore less likely than the others to be fooled by noise, and more likely to detect true weak edges.

Figure 7. Gray levels image (a) and binary image (b) of the 12th snapshot

Figure 8. Edges extracted with the Canny method by the binary images (a) and reconstruction of the water domain (dark zone) (b)
The edges individuated by applying the Canny method to the binary image shown in figure 7 are shown in figure 8a. Then the domain occupied by the water phase can be reconstructed by filling the edges with the `imfill` function of Matlab: the results are shown in figure 8b.

![Image of binary images showing edges and reconstructed domains.](image)

**Figure 9.** Evolution of the water-phase contours during the squeezing and the breakup of water in oil (W/O) for an imposed water volumetric flow rate of 0.6 ml/h and an oil flow rate of 7.5 ml/h

### 3.3. Post-processing of the image

Now, it becomes possible to obtain, starting from the oil/water contours, the area $A$ occupied by the water-phase, the perimeter of the water-oil interface and the coordinate of the centre of mass of the droplet (by using the `bwboundaries` function of Matlab). In presence of a single droplet, the centre of mass $(x_c, y_c)$ and also the velocity of the droplet can be calculated. Figure 10 shows the position of the coordinate for the centre of the droplet (red lines) obtained for three consecutive snapshots. The yellow lines show the position of the channel walls within the junction. Considering the first two snapshots in the figure, we can obtain the velocity of the droplet by considering the displacement of the centre of mass along the $y$-axis and the time interval between two consecutive images ($v = (y_c(2) - y_c(1))/dt$); here $dt=1/5000$ s.

![Image showing droplet centre for three consecutive snapshots.](image)

**Figure 10.** Determination of the droplet centre for three consecutive snapshots
This evaluation can be repeated by considering different couples of images (i.e. in figure 10, the evaluation of the droplet velocity can be obtained by considering the images 1-2, or 2-3) in order to obtain an average value of velocity for the droplet.

Finally, from the area $A$ obtained from the droplet contour, it is possible to calculate the ideal radius of a circular droplet having the same area $A$ of the real droplet ($R = \sqrt{\frac{A}{\pi}}$). Then, plotting a perfect circle with radius $R$, centred in $(x_c, y_c)$, it is possible to find the eccentricity of the droplet. Figure 11 shows an example of comparison between the real shapes of the droplet (blue line) determined experimentally and the circular contours obtained from the ideal radius associated to the droplet (red line). It is evident that the eccentricity of the droplet is very limited in this specific case.

![Figure 11](image)

**Figure 11.** Comparison between the droplet contour obtained by the experimental images (blue line) and the circular contour drawn from the radius $R$ associated to the droplet (red line)

However, figure 11 shows a deviation from the ideal circular profile especially in the bottom of the droplet; in this case the droplet contour can be assumed to be circular with an error less than 2%. The coordinates of the centre show that the droplet is moving only along the $y$ direction (the variation of $x_c$ between two consecutive images is negligible), with a velocity $v = 8.5$ cm/s for the case depicted in figure 11.

4. Result and discussion

In literature two different dynamical patterns have been recognized for explaining breakup in a microfluidic T-junction or cross-junction: the shear-driven breakup [5], which is derived from the balance of shear and interfacial stresses, and the rate-of-flow-controlled breakup [8], which is proposed for systems operating at low values of the Capillary numbers of the continuous phase ($Ca_c = \mu_c v_c / \sigma$, where $\mu_c$ is the dynamic viscosity, $v_c$ the velocity and $\sigma$ is the interfacial tension between the two fluids) and it is based on the evolution of pressure in the continuous phase upstream of the tip of the dispersed phase.

In shear-driven breakup, the predicted size of a droplet under an external shear force is approximated by balancing the interfacial force with the shear force:

$$\frac{\sigma}{R} = \mu_c \epsilon$$

where $R$ is the final droplet radius and $\epsilon$ is the shear rate. The surface tension ($\sigma$) between silicone oil and water has been evaluated by means of the following correlation suggested in [20]
as \( \sigma = 0.122T + 32.82 \), where \( T \) is the temperature in °C and \( \sigma \) is in mN/m. Casting equation (1) in order to obtain the dimensionless radius, one can easily find that:

\[
R^* = \frac{R}{H} = \frac{1}{Ca_c \varepsilon H v_c}
\]

(2)

where \( H \) is the height of the microchannel. Around the droplet, a shear rate \( \varepsilon \) is established in the continuous phase forced to flow in a restricted cross-section region with area \( S_g \), delimited by the walls at one side and the droplet surface at the other side. The shear rate can be estimated as follows:

\[
\varepsilon = \frac{v_g}{\delta_g}
\]

(3)

where \( \delta_g \) is the distance between the droplet contour and the wall and \( v_g \) is the average velocity of the continuous phase within the restricted section \( S_g \). Substituting equation (3) in equation (2), one can reach to a prediction of the dimensionless droplet radius, as a function of the dimensionless shear rate and of the Capillary number of the continuous phase:

\[
R^* = \frac{R}{H} = \frac{1}{Ca_c \varepsilon H v_c} = \frac{1}{Ca_c \varepsilon_d}
\]

(4)

where \( \varepsilon_d = \frac{v_g}{\delta_g} \) is the dimensionless shear rate.

Figure 12a shows the comparison of the dimensionless diameter of the droplet \( D^* = 2R^* \) calculated by using equation (4), with the experimental results. As pointed by other authors [9, 11, 21], the dimensionless diameter of the droplet decreases with \( Ca_c \) much more slowly than one would expect from the scaling law (equation (4)). De Menech et al. [11] for a T-junction found a good agreement between \( D^* \) and \( (Ca_c \varepsilon_d)^{-0.25} \), while van der Graaf et al. [21] demonstrated that \( D^* \) scales with \( Ca_c^{-0.25} \).

In figure 12a the experimental results obtained in this work have been compared with the correlations proposed by De Menech et al. [11]; the best agreement is observed when \( D^* \) scales with \( (Ca_c \varepsilon_d)^{-0.3} \). In figure 12b the values of the dimensionless diameter of the droplet has been correlated to the flow rate ratios \( \phi \) (defined as the ratio of water flow rate/silicone oil flow rate). As one can see, \( D^* \) for a fixed value of \( Ca_c \) does not change significantly with \( \phi \); the experimental results can be correlated to \( \phi \) and \( Ca_c \) by means of the following relationship: \( D^* = 0.4(\phi^{0.05} \cdot Ca_c^{-0.25}) \).

(a)  
(b)

Figure 12. The dimensionless diameter of the droplet: as a function of \( Ca_c \) compared with the different correlations (a) as a function of the volumetric flow rate ratios \( \phi \) (b)
It has been demonstrated by figure 12 that the dimensionless diameter of the droplets observed experimentally in this work depends to the Capillary number of the continuous phase and very weakly to the flow rate (water/oil) ratio; this observation seems to confirm that the observed droplet breakup can be considered more similar to a *shear-driven* breakup than a *rate-of-flow-controlled* breakup. However, further investigation is needed in order to clarify these aspects.

5. Conclusions

In this work water-in-oil droplet breakup mechanism within a micro cross-junction with hydrophobic walls has been investigated experimentally by means of optical techniques. By the use of a high-speed camera, connected to a microscope, a sequence of snapshots which capture the squeezing of the water phase, followed by breakup and droplet formation, have been recorded and analyzed. Processing of the raw snapshots provides us useful information for the physical description of the evolution of the squeezing mechanism and of the formation of water droplets. In the paper it has been described the procedure by means of which it is possible to estimate the eccentricity, area, average velocity and the dimensionless diameter of the droplet. The experimental data shows a good agreement with correlation when the diameter of the droplet scales with the Capillary number as $(Ca_{c} \epsilon d)^{-0.3}$. The dimensionless diameter has been presented for various flow rate ratios and a correlation to fit the results has been presented.

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