Supplemental Material

Scaling of mixing time for droplets of different size traveling through a serpentine microchannel

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This PDF file includes

- 1) Additional data
- 2) Additional methods
- 3) Movies



Figure S1. The Dependence of dimensionless droplet size (*a*) on the flow rate ratio $Q_d/Q_c = 0.5$ (circles) and 2.0 (triangles) for various total flow rates Q_{total} . Geometry: cross section microfluidics (with geometrical dimensions as height $h = 100 \mu m$, width $w = 100 \mu m$, arm length $L_{arm} = 150 \mu m$, inner and outer radius of bend = 50, 150 μm and angle bend= 160 degrees).

2. Data Analysis:

From the video feed captured, it is required to identify the droplets, tracking them while the droplets were moving through the field of view and meanwhile calculating the extent of mixing. The droplets moving through the serpentine are imaged using a high-speed camera (Vision Research Phantom 9.1). A flow scheme with additional explanation on the operations performed by the MATLAB® code is shown in Figure S2.



Figure S2. Flow chart of the data processing algorithm.

2.1. Fluorescent and color mapped images

In actual measurements for studying the mixing, images of droplets were captured in the fluorescence mode. A typical picture captured with fluorescence mode is shown in Figure S3 (a). From this picture, it is evident that only the aqueous phase containing dye has contrast from the rest of the system. Moreover, there is a contrast inside a single droplet, indicating concentration distribution of the fluorescent dye inside the droplet. Thus the region of the droplet that is with a higher concentration of the fluorescent dye is brighter than the region with a lower concentration of the dye. This is further justified with a color-mapped version of the same picture as presented in Figure S3 (b).



Figure S3. (a) Fluorescent image of droplets with a=3 (formed at $Q_d/Q_c = 0.5$) moving through the serpentine geometry. (b) Color mapped image of droplets from (a), showing concentration distribution. The droplets are traveling at $Q_{total} = 10 \ \mu l/min$.

2.2 Identification of a droplet

The very first step in the image processing, after loading the image-file into MATLAB® code, involves subtraction of the dark field image from the image being processed to remove the standard noise of the camera sensor. After the removal of the camera sensor noise, the image is cropped to a part directly after the cross-junction.

2.3 Droplet tracking

The algorithm from this point onwards contains a tracking mechanism, followed by the operations performed as discussed above (identification of the droplet and calculating the extent of mixing). The tracking mechanism involves loading the image following the previous image, which would be the just pinched of droplet image. For this next image, the identification steps as discussed before are executed again. If the droplet is still fully inside the rectangular box for which the image is cropped, the homogeneity factor and displacement are calculated as discussed before. If the droplet is not fully inside, the rectangular box for the cropping is shifted a single pixel in the horizontal direction and the identification steps will be performed, until the droplet is once more fully inside the rectangular cropped binary image. Once the droplet has been tracked and identified inside the next image, the homogeneity

factor and displacement are calculated for this new frame. Crucial for the tracking mechanism is that the new location of the rectangular cropping box is stored and used as a starting position for images following. This prevents other droplets being picked instead of the droplet of interest, as the rectangular box is following the direction of the droplets motion, which is the horizontal direction. The tracking mechanism is repetitive for any image following: based on the position of the rectangular cropping box of the previous image, the box is shifted inside the image following until the same droplet is inside the box once more. This tracking algorithm is executed until the droplet has move out of the (user defined) field of view. Once this has happened, the rectangular cropping box is set to the initial coordinates and awaits a new droplet pinching off.

2.4. Aura removal process

At this point, all the data of the droplet is identified and contained. But before calculating the extent of mixing, another operation was performed. This operation concerns the removal of part of the outer pixels of the droplet (as shown in Figure S4). The removal of the aura is illustrated based on the binary identified droplet, as outer pixels are removed maintaining the droplet shape. This aura removal process is illustrated in Figure S4(a-d) and the resulting color mapped shrunk droplet in Figure S4(e).



Figure S4. Aura removal process demonstrated for the binary identified droplet, with (a) the original binary droplet, and (b) the pixels contributing to the aura, which is removed from the binary droplet to produce (c) binary identified without the aura. The Color-mapped normalized intensity inside the identified droplet (d) before and (e) after removal of the aura.

2.5 Effect of Aura removal on homogeneity factor

Figure S5 shows that the homogeneity factor is not affected by removing 10% or 25% of the outer part of the droplet, when the droplet has just pinched off. On the other hand, the homogeneity factor of the fully mixed droplet is reduced significantly by a factor of three and reaches to 0.10. Although not shown here, more percentages of aura artefact removal were tested. We found that 25% removal of the outer part of the droplet was sufficient to yield asymptotic value of the homogeneity factor. It should be noted here that further cropping of the droplet area did not yield a significant change in the homogeneity factor of the fully mixed droplet while leading to information loss before the droplet reached fully-mixed condition. All results on the extent of mixing were therefore calculated based on 25% removal of the outer part of the droplet.



Figure S5. Color-map intensity distribution of a droplet at three different locations inside microfluidic device with different percentages of aura artifact removal. Three different locations are: just at pinch off, at partially mixed location, and at fully mixed condition. Homogeneity factor for each case are shown.



Figure S6. Schematic of the three micro-fluidic channels with different curvature ratios. Top and middle: A flow-focusing device with serpentine channel and geometrical dimensions as $w = 100 \,\mu m$, $L_{arm} = 150 \,\mu m$, angle bend = 160 degrees, and (a) radius ratio of the inner and outer wall of bend $R_{in}/R_{out} = 100/200$, and (b) $R_{in}/R_{out} = 50/150$. Bottom: A T-junction microfluidic with the serpentine and geometrical dimensions as, $w = 100 \,\mu m$, $L_{arm} = 200 \,\mu m$, angle bend = 80 degrees, and (c) radius ratio of the inner and outer wall of bend $R_{in}/R_{out} \sim 0/100$.

Supplementary Movies Legends

Supplementary Movie S1. Fluid mixing in droplets with a = 3 (formed at $Q_d/Q_c = 0.5$). The droplets are traveling at $Q_{total} = 10 \mu l/min$. The speed of the movie is slowed down 100 times.

Supplementary Movie S2. Fluid mixing in droplets with a = 8 (formed at $Q_d/Q_c = 2.0$). The droplets are traveling at $Q_{total} = 10 \mu l/min$. The speed of the movie is slowed down 100 times.