Supplementary materials

Three-dimensional visualization and analysis of flowing droplets in microchannels using real-time quantitative phase microscopy

Yingdong Luo[a], Jinwu Yang[a], Xinqi Zheng[a], Jianjun Wang[a], Xin Tu[a], Zhizhao Che[b], Jiakun Fang[c], Lei Xi[d], Nam-Trung Nguyen[e], Chaolong Song[a,*]

[a] School of Mechanical Engineering and Electronic Information, China University of Geosciences, Wuhan, 430074, China, email: songcl@cug.edu.cn

[b] State Key Laboratory of Engines, Tianjin University, Tianjin 300072, China

[c] State Key Laboratory of Advanced Electromagnetic Engineering and Technology, School of Electrical and Electronic Engineering, Huazhong University of Science and Technology, Wuhan, China

[d] Department of Biomedical Engineering, Southern University of Science and Technology, Shenzhen, China

[e] Queensland Micro- and Nanotechnology Centre, Griffith University, 170 Kessels Road QLD 4111, Brisbane, Australia.

Supplementary S1:

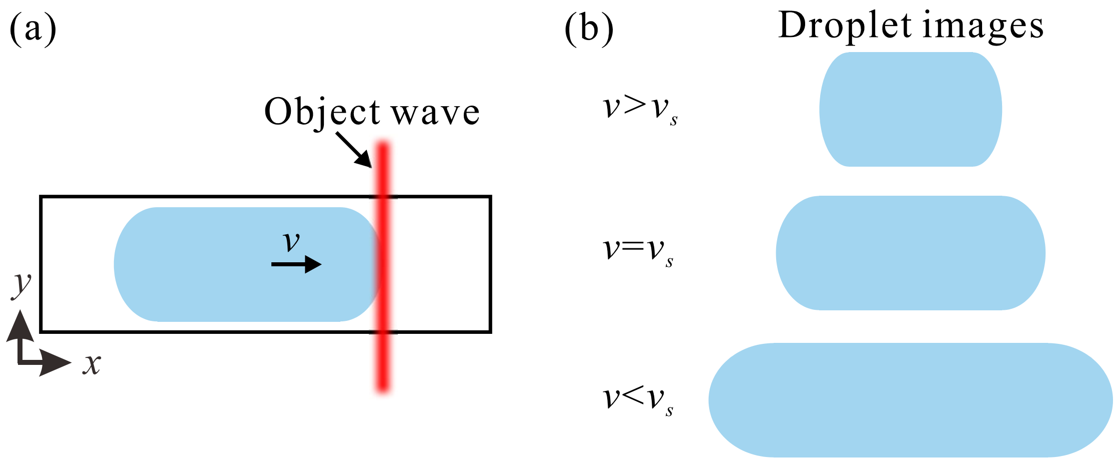


Figure S1: the droplet images recorded by the LACCD under different droplet moving velocities. (a) Illustration of a droplet moving through the laser inspection area; (b) When the droplet moving velocity is higher than the standard velocity, the droplet image would be distorted to be "compressed" due to under-sampling by the LACCD. When the droplet moving velocity is lower than the standard velocity, the droplet image would be distorted to be "stretched" due to over-sampling by the LACCD. When the droplet moving velocity matches the standard velocity, the image of droplet would be remain its fidelity to its true shape.

Herein, the standard droplet velocity (v_s) is defined and calculated as the velocity for acquiring images in their fidelities. Given the microscopic system has a magnification of M, the pitch size of each pixel is p and the frame rate of the LACCD is f, the standard velocity in the system can be defined as

$$v_S = \frac{f \times p}{M} \qquad (1)$$

In this work, the imaging system has a frame rate of f=80kHz, with a magnification of M=48 and a pitch size of 5 μm, and therefore the flow rate in the microchannel can be calculated to be 240 μl/h All the experiments are implemented with this flow rate to achieve images with optimum fidelity. In this case, the resolution along the x-axis direction can be the same as the resolution along y-axis direction, and it can be evaluated using the Rayleigh criterion:

$$R_l = 1.22\frac{\lambda}{NA} \qquad (2)$$

Where R_l is the lateral resolution, λ is the wavelength of the light, NA is the numerical aperture. In our experiment, the wavelength of the light λ is 671nm, the numerical aperture is 0.9, so the lateral resolution can be evaluated as 0.9μm.

Supplementary S2:

The axial resolution can be evaluated as the minimum detected droplet height variation across two adjacent pixels. For example in the wrapped phase map (Fig. 2(d)), each fringe represents a phase variation π. Therefore, the height difference across a single fringe can be described as:

$$\Delta h = \frac{\pi}{\Delta n \times k} \tag{3}$$

Where Δn is the refractive index difference between the dispersed phase and continuous phase, k is the wavenumber. The phase is represented by the gray-level of unwrapped phase map. One fringe in an unwrapped phase map has phase variation π and a corresponding gray-level variation of 2^t. The axial resolution can be expressed as:

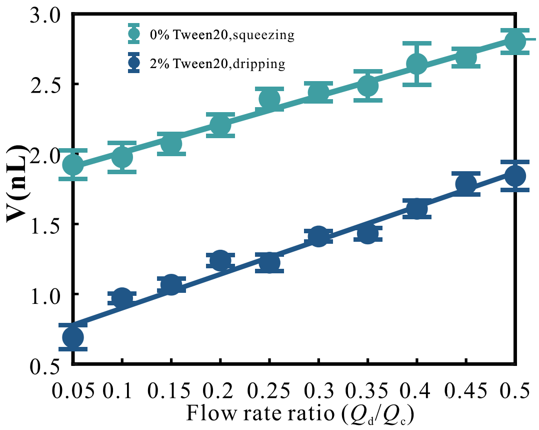
$$R_{\mathrm{a}} = \frac{\Delta h}{2^t} \tag{4}$$

where t is the bit depth of the CCD.

In the experiment, 2.5mol/L calcium chloride solution with 2% mass ratio Tween 20 (RI=1.392) and silicone oil (RI=1.409) worked as the dispersed phase and continuous phase respectively to produce droplets at a T-junction (Fig. 1 (c)). The bit depth in the linear array CCD is 8 bits. According to Eqn.4, we can estimate that the axial resolution is about 77 nm.

Supplementary S3:

(a)



(b)

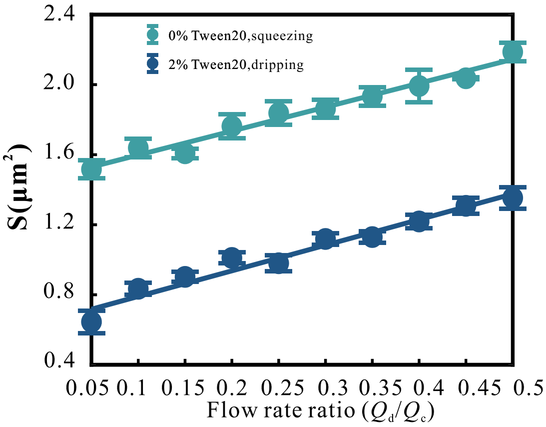


Figure S3: (a) The volume of the droplets as a function of the flow rate ratio (Q_d/Q_c) in squeezing (green points) and dripping regime (blue points). (b) The measured surface area of the droplets in squeezing and dripping regimes.

In the experiment, 2.5 mol/L calcium chloride solution and silicone oil worked as the dispersed phase and continuous phase respectively to produce droplets at a T-junction and the 2% mass ratio Tween 20 was added into the dispersed phase fluid to switch the droplet generation regime from squeezing to dripping. In Fig.S3, the experimental results show that the volume and surface area of the droplet present a linear relationship with the flow rate ratio in both dripping and squeezing regime.

Supplementary S4:

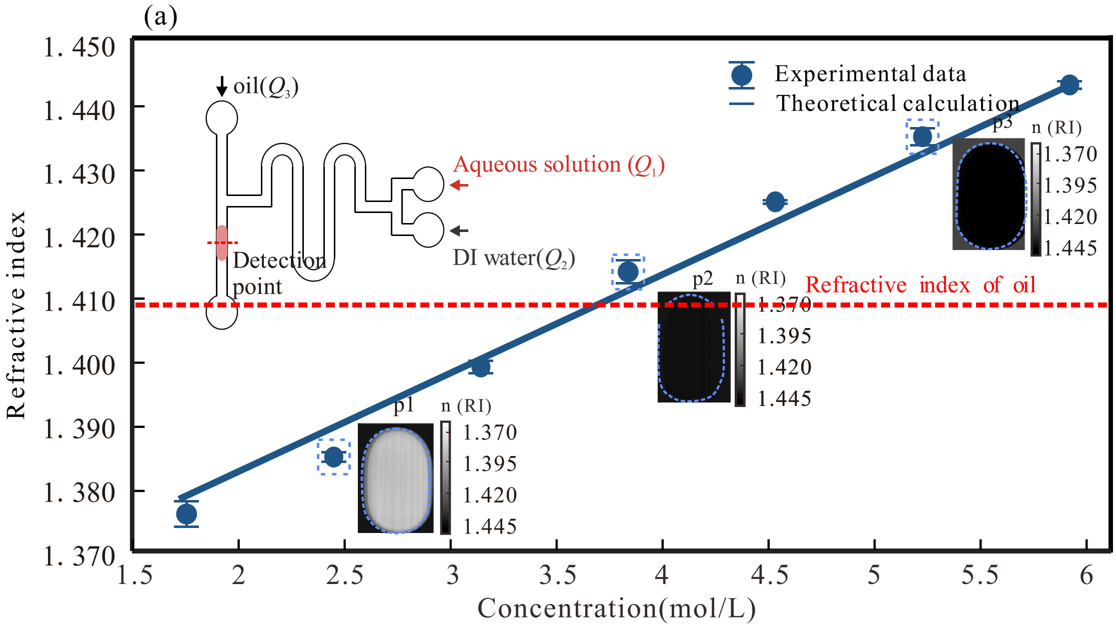


Fig.S4 Quantitative measurement of calcium chloride solution molecular concentrations in droplet-based microfluidics. The measurement is carried out with droplets generated in the squeezing regime that have a constant droplet height. The quantitative evaluations of molecular concentrations can be achieved based on the visualized sample-induced phase variation (illustrated as insets P1-P3).

The silicone oil served as the continuous phase was pumped into the optofluidic chip with flowrate Q_3, and the total flow rate $Q_1+Q_2+Q_3$ was kept at 240μL/h as illustrated in the insets in Fig. S4. The droplets were generated in the squeezing regime, so the height of the droplets can regrade as the height of the microchannel (h=40 μm), and thus the RI mapping can be calculated (Fig.2(g)). The experimental data show a good linear relationship with the theoretical calculations, which have correlation coefficients R^2 of 0.974

Supplementary S5:

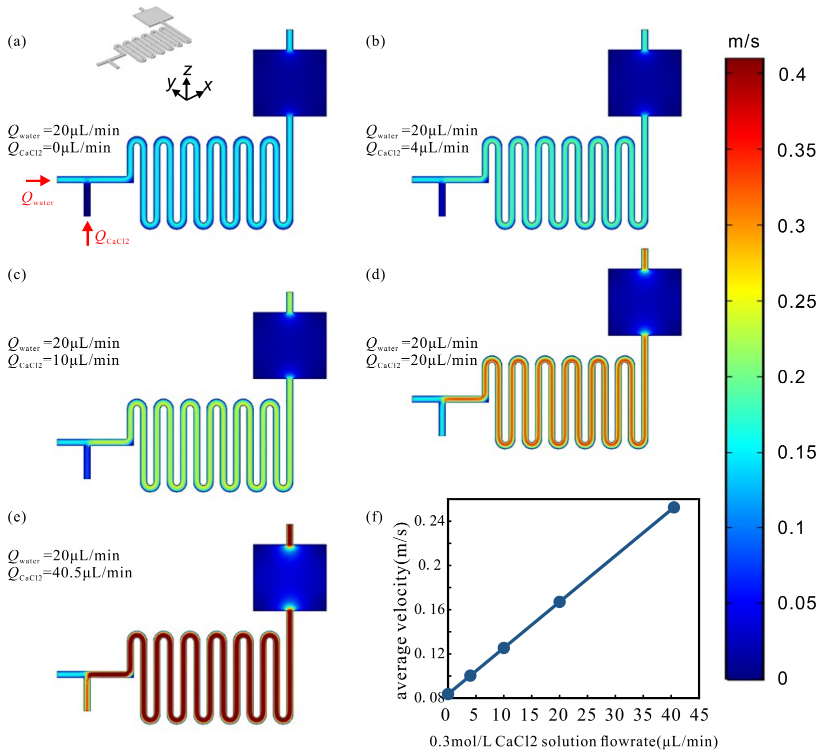


Fig.S5 The numerical simulation of mixing dynamics in the phase shifting element. (a) the flowrate of DI water at 20 μL/min (Q_{water} = 20 μL/min) and the flowrate of 0.3mol/L calcium chloride solution at 0 μL/min (Q_{CaCl2} = 0 μL/min); (b) Q_{water} = 20 μL/min, Q_{CaCl2} = 4 μL/min; (c) Q_{water} = 20 μL/min, Q_{CaCl2} = 10 μL/min; (d) Q_{water} = 20 μL/min, Q_{CaCl2} = 20 μL/min; (e) Q_{water} = 20 μL/min, Q_{CaCl2} = 40.5 μL/min; (f) the average velocity in the cross section of the chamber inlet with different flowrates.

The simulation result of velocity field in the phase shifter (view in *x*-*y* plane) shown in Fig.S5 (a-e). Fig.S5 (f) illustrated the average velocity of the flow at the entrance of the chamber in *x*-*z* plane. The total length of the serpentine channel of the mixer is about 40mm, based on which the time consumed for fluid transport is estimated to be less than one second.

Supplementary videos:

Video S1: The flowing droplets at squeezing regime. The 2.5 mol/L calcium chloride solution and silicone oil worked as the dispersed phase and continuous phase respectively with flow rate ratio between the two phases (Q_d/Q_c) =0.3, total flow rate of Q=240μL/h.

Video S2: The flowing droplets at dripping regime. The 2.5 mol/L calcium chloride solution with 2% mass ratio Tween 20 and silicone oil with flow rate ratio between the two phases (Q_d/Q_c) =0.3, total flow rate of Q=240μL/h.