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Breakup of O/W/O double emulsion droplets with low viscosity shell in capillary single-step emulsification

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Abstract

Based on micro high-speed photography experiment, the influence of flow rate on the breakup mode of double emulsion droplets in coaxial capillary is investigated， and the dynamic characteristics of the downstream endpoint in the droplet generation is analyzed. Result show that with the increase of middle phase flow rate, the periodicity is presented within the generation of droplet. Under the periodicity of droplet generation, the outer interface is broken and forms six different breakup model through two mechanisms of PFD and PBD. The distribution of phase diagram of breakup modes varying with flow rate is setup by expanding the scope of the condition.

1 Introduction

Due to the unique core-shell structure, double emulsion droplets have great potential application value in such fields as controllable drug release, cell culture, carbon dioxide adsorption and energy storage. In these applications, the accurate control of size and structure of double emulsion droplets is the key to determine its performance. Compared with traditional preparation methods, the generation method of double emulsion droplets by microfluidics has attracted much attention due to its advantages of good monodispersity and controllability of size and structure of droplets. In capillary, the generation of double emulsion droplets is essentially a phenomenon of coaxial jets break up. The size and structure of double emulsion droplets are significantly affected by the coupling effect of inner and outer interfaces. Therefore, understanding the dynamic behavior of interface under coupling during the generation of double emulsion droplet will be the key to realize accurate generation of double emulsion droplets.
Although there are obvious advantages on microfluid-based generation of double emulsion droplets, the research on the coupling effect between the inner and outer interfaces is not comprehensive, especially the influence of the periodic generation of inner droplet on the outer interface needs to be further explored. This paper will explore the periodic generation characteristics of inner droplets by adjusting the flow rate, and investigate the influence of periodic breakup of the inner layer interface on the outer layer interface.

2 Experimental Section

In order to realize the periodic generation of inner droplets, the experiment model in this paper is designed based on the coaxial capillary device adopted by Utada et al. (2005) and combined with the flow-focusing structure used by Garstecki et al. (2004). The injection tube is tapered using a micropipette puller (P-2000, Sutter Instrument, Inc), the collection tube is heated using an alcohol lamp. To generate double emulsion droplets more easily, the surface of square tube and collection tube is modified to hydrophobicity. Finally, the injection tube and collection tube are inserted into the square tube, and fixed with epoxy resin.

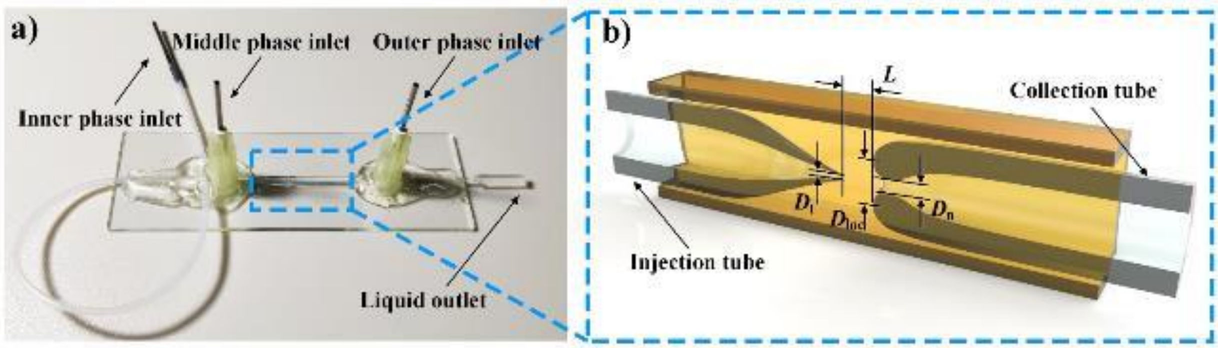

Figure 1: The experimental device. (a) The capillary device. (b) Schematic of capillary device.

Liquid paraffin (Sinopharmaceutical group chemical reagent co, Inc) is used for inner and outer phase fluids, and glycerine aqueous (Sinopharmaceutical group chemical reagent, Inc) solution with mass fraction of 4.5% is applied for middle phase fluids. Sudan 3 (Sigma, Inc) is added to the inner phase to improve the visibility of inner phase. Tween 20 (J&K Chemicals, Inc) with a mass fraction of 0.50% is added to the middle phase, and Span 80 (J&K Chemicals, Inc) with a mass fraction of 0.75% is added to the outer phase to stabilize the generation of droplets.

3 Breakup Modes of Interfaces

It is found that the double emulsion droplets generation are determined by the different breakup modes of the inner and outer interfaces. Six different breakup modes are observed, as shown in Fig. 2 S (Single), U (Ultra-thin), D (Double) and D2 (double-2) are defined to represent single emulsion droplets, ultra-thin shell double emulsion droplets, thick shell double emulsion droplets, and double emulsion droplets with double cores separately. According to the generation sequence of droplets in one cycle, the breakup modes of are defined to D, US, UUS, UD, USUS and D2 respectively.

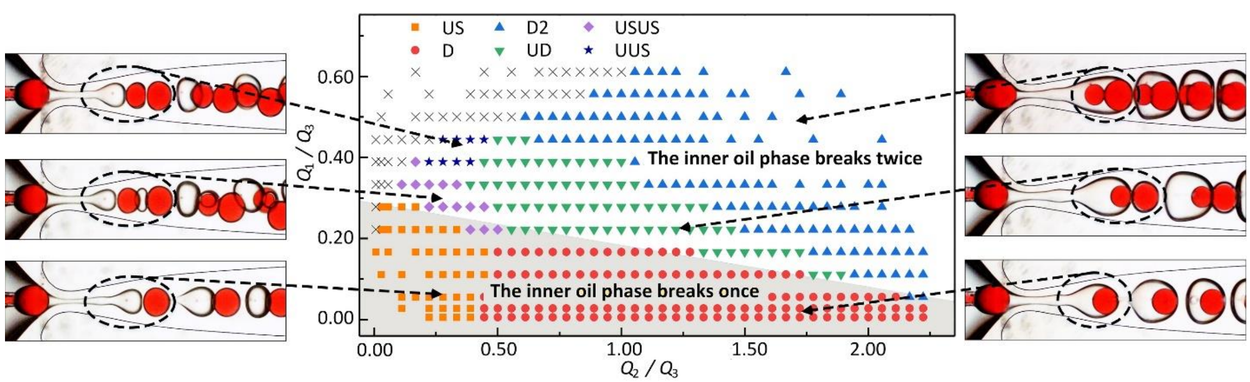

Figure 2: Phase diagram of the breakup modes on double emulsion droplets

In order to further explore the conversion between breakup modes, mark the breakup modes on different conditions, the phase diagram of the breakup modes on double emulsion droplets is setup, as shown in Fig. 2 It can be observed clearly from the image that the distribution of breakup modes on different flow rate and the transition relationship between them.

The phase diagram can also be divided into two regions, inner phase breaks up once and inner phase breaks up twice, according to the different number of inner layer interface break up within the same cycle, as shown in Fig. 2

4 Periodic Generation of Inner Droplets

To quantitatively analyze the dynamic behavior of inner droplets generation, the distance from the end surface of the collecting tube to the point F is defined as L_f, as shown in Fig. 3(b). The velocity V_f and the distance L_f of point F are extracted as shown in Fig. 3(a), the flow rate of inner and outer phase is kept in Q_1 = 120 μL/h and Q_3 = 1080 μL/h.

The modes US, D, UD and D2 are displayed successively as the flow rate of middle phase increasing from Q_2 = 300 μL/h to 2580 μL/h, and the inner interface is broken up once in modes US and D in one cycle, and twice in modes UD and D2.

As shown in Fig. 3(a), it can be found that the velocity of point F does not change with the increase of the flow rate of the middle phase in the growth stage. In the deformation stage, as the flow rate of the middle phase increasing, the speed and distance of point F gradually increases.

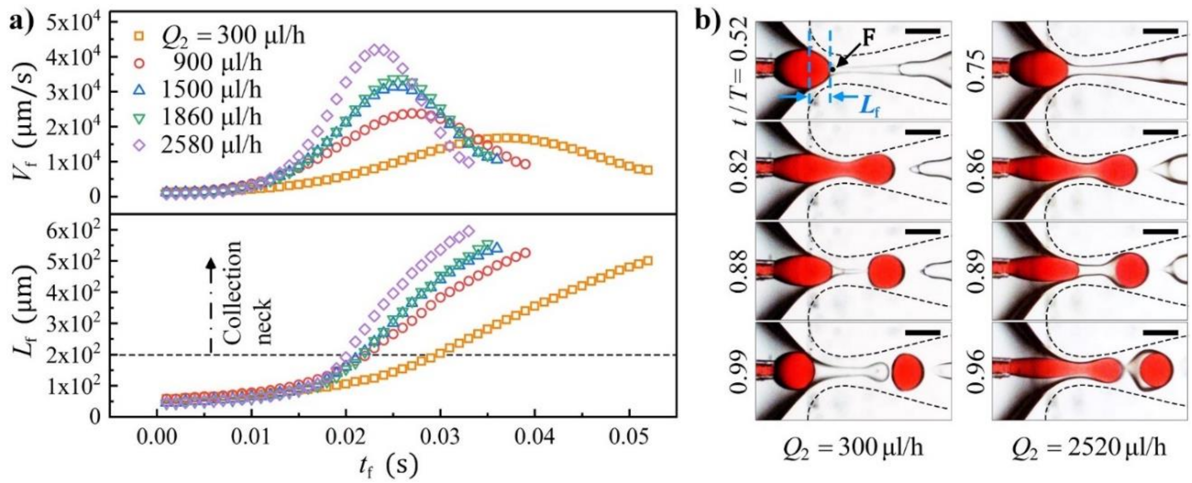

Figure 3: Influence of Q_2 on dynamic characteristics of point F. (a) variation rules of speed V_f and distance L_f of point F with respect to time under different Q_2. (b) breakup process of inner phase when Q_2 = 300 μl/h and Q_2 = 1500 μl/h.

The breakup process of the inner interface in each period is shown in Fig. 3(b), as Q_2 = 300 μL/h and Q_2 = 1500 μL/h. It can be found that the inner interfaces start to accelerate downstream at the beginning stage under both conditions, and then contract at the neck, exacerbating the Plateau-Rayleigh instability, and rupture under the influence of pressure difference. As t/T is 0.99 and 0.96, respectively, the remaining part of the inner phase directly retracts after the first breakup under Q_2 = 300 μL/h. The remaining part of the inner phase continues to move downstream to occur the second breakup under Q_2 = 1500 μL/h.

5 Influence of inner droplets on breakup of outer layer interface

Contrasting the breakup of the middle phase under different modes, it is found that breakup of outer interface can be summarized as two basic of mechanism: Pinch-off behind a droplet (PBD) and Pinch-off in front of a droplet (PFD).

In order to explore the characteristic of PBD, the process of US and D modes is compared. The difference between the two modes is only whether PBD occurs, as shown in Fig. 4(a). As Q_1 = 120 μL/h, Q_3 = 1080 μL/h are maintained, when middle phase flow is adjusted to Q_2 = 240μL/h, the average diameter of the liquid bridge W_b is only 23 μm, and the liquid bridge breaks up rapidly due to large curvature. When the middle phase flow is increased to Q_2 = 600 μL/h, the W_b is 61 μm. According to Laplace-Young's equation, the differential pressure on both sides of the interface decreases with the decrease of interface curvature, so the Pinch-off position is necked without breakup.

Similarly, the dynamic process of mode UD and D2 are compared. The difference between the mode UD and D2 is only whether PFD occurs, as shown in Fig. 4(b). As Q_1 = 300μL/h and Q_3 = 1080μL/h are maintained, when Q_2 = 1080μL/h, it is found that the liquid bridge breaks up rapidly. For Q_2 = 1620 μL/h, the liquid bridge is necked, but when liquid bridge is not broken up, the second droplets have been generated and quickly entered the first phase droplets in the middle of the shell, double emulsion droplet with double cores is forming.

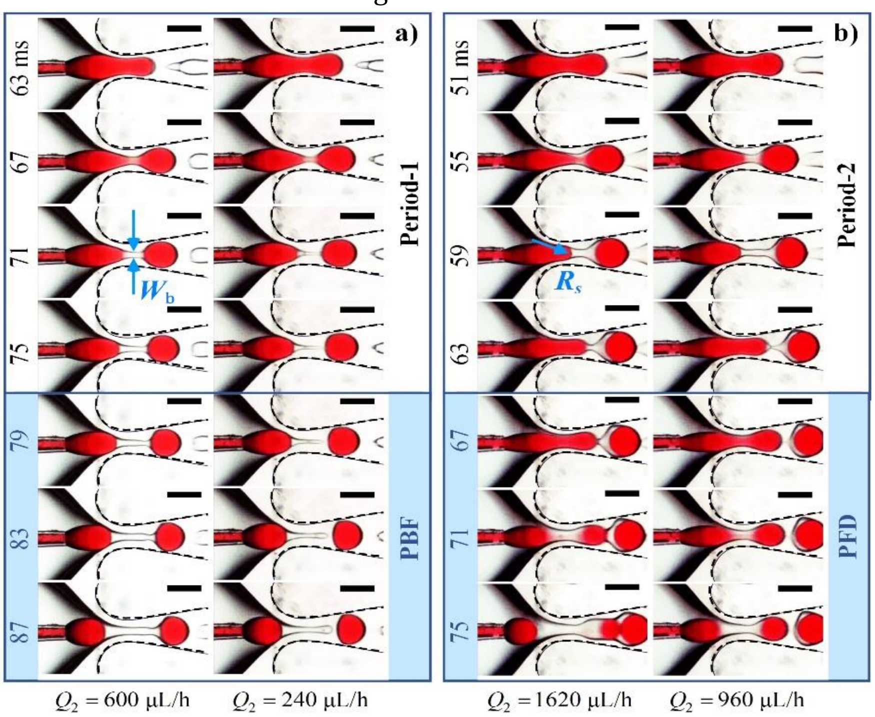

Figure 4: The breakup process. (a) Under PBD. (b) Under PFD.

In order to identify the factors that can possibly affect the pinch-off, the flow field during the middle phase pinch-off is visualized through micro-PIV.

The flow field of middle phase under the mode PBD is shown in Fig. 5. It is found that as the inner droplet moving downstream, the velocity of the middle phase becoming maximized at the neck position, and the neck position is broken by the law of mass continuity. The flow field of the middle phase fluid under the mode PFD is shown in Fig. 6. It can be found that the neck of the middle phase occurs in front of the inner droplet during the inner droplet movement downstream. At the narrowest position of middle phase, the velocity of middle phase flow field reaches the maximum.

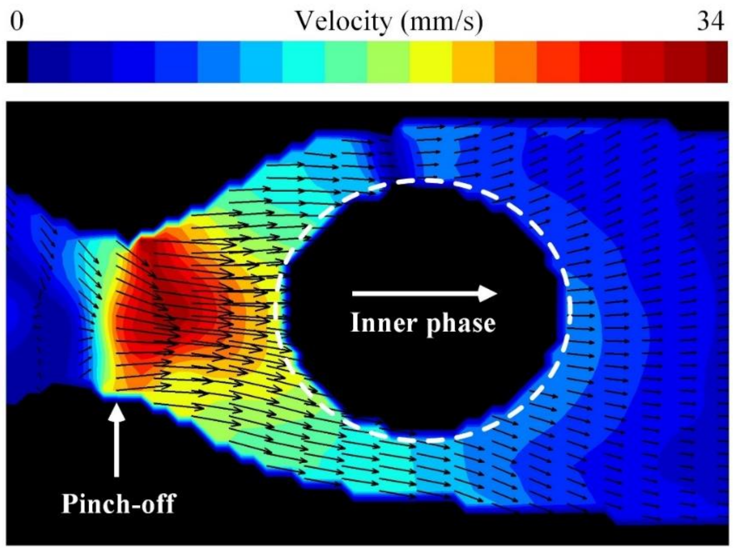

Figure 5: The flow field of the mode PBD.

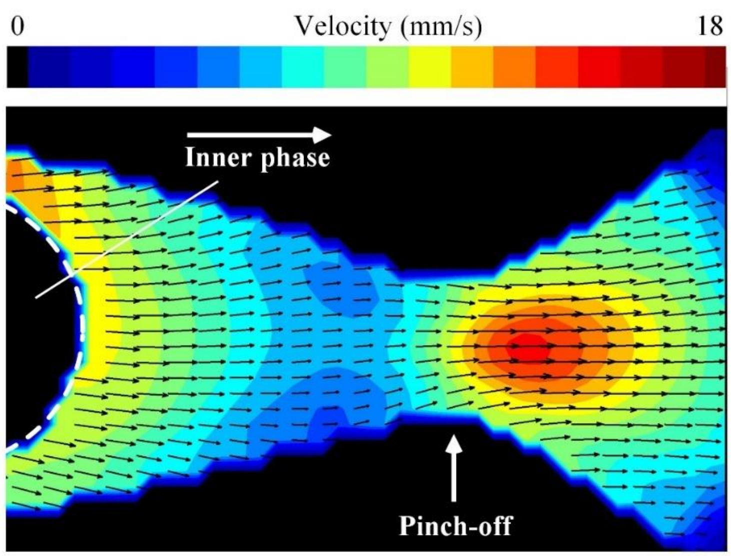

Figure 6: The flow field of the mode PFD.

6 Conclusion

In this paper, the breaking behavior of double emulsion droplets in capillary tube is studied. The influences of flow rate of inner and middle phase on the periodic generation of inner droplets are considered. It is found that the increase of inner and middle phase flow rate makes the inner interface break up transform from primary to secondary in the same cycle, and induces the breakup of the outer interface through the mechanisms of mode PBD and mode PFD. In addition, the breakup of the middle phase is caused by fluid mass conservation in both mode PBD and mode PFD. The size and structure of double emulsion droplets can be controlled by adjusting the inner droplets movement.

Acknowledgements

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