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Experimental study of liquid/liquid second-dispersion process in constrictive microchannels

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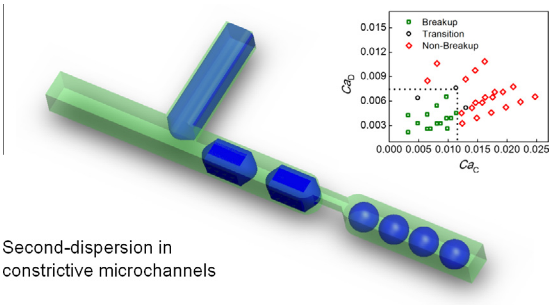
HIGHLIGHTS

- The second-dispersion process in microchannel is systematic studied.
- 3 Behaviors of liquid plugs: breakup, non-breakup and transition are summarized.
- New criterion of plug breakup in orifice is proposed.
- The critical capillary numbers for inducing breakup are provided.
- The generated droplet radius is 2 more times of the orifice hydraulic radius.

GRAPHICAL ABSTRACT



Second-dispersion in constrictive microchannels

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ABSTRACT

Liquid/liquid second-dispersion signifies the process of breaking dispersed droplets, for the purpose of further reducing the droplet size. This study introduces a systemic experimental study on the second-dispersion process in different constricted microchannels and presents the flowing behaviors of liquid plugs passing through them. Both breakup, non-breakup and their transition of plugs were observed at the operating conditions of *Ca* ranging from 0.0002 to 0.025 and *Re* ranging from 0.74 to 27.3 in the orifice. Main factors influencing the plug breakup process, such as surfactant concentration, plug length, flow rate, viscosity and channel geometry, were carefully examined. We found the liquid/liquid second-dispersion process, driven by the capillary pressure criterion, is both a statically confined and dynamical confined process. To realize second-dispersion, the hydraulic radius ratio of downstream microchannel to constrictive orifice should be larger than 1.89 and the operating condition should be lower than the critical capillary numbers ($\sim 10^{-3}$) of continuous phase and dispersed phase. We find these critical capillary numbers reduce as the rising of hydraulic radius of the orifice but changed little with the enlargement of downstream microchannel. The orifice geometry also dominates the daughter droplets, whose radii are at least two times of the orifice hydraulic radius.

1. Introduction

Ever since its emergence, microfluidic technology has developed very fast and it is now widely used in many research fields such as biology, material, chemistry and engineering process [1,2]. Thanks to its controllability, people have skillfully utilized this technology to prepare microdispersed liquid/liquid system, meaning that the production and manipulation of microdroplets can be easily achieved [3–5]. These kinds of liquid/liquid two-phase systems are very important for both scientific research and industrial application, since the enhancements of heat and mass transfer rates are obvious in microstructured chemical device, comparing with common chemical equipment [6]

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To explore controllable dispersion process, many studies have been conducted to display the basic bubble and droplet generation rules in T-junction microchannel [7], co-flowing microchannel [8], flow-focusing microchannel [9] and some other kinds of microstructured devices [10]. The squeezing effect and the shearing effect from continuous phase are two main driving forces in microfluidic dispersion process withstanding the interfacial tension. Two-phase flow ratio (Q_D/Q_C) and capillary number ($Ca = \mu u/\gamma$) are key parameters determining the generated bubble and droplet sizes. Coalescence is another important issue in microfluidic process. Fast bubble or droplet coalescence disfavors the good mixing and mass/heat transfer performance in microstructured chemical device. To further understand the basic rules of coalescence at micrometer scale, experimental and theoretical analyses have been carried out to discover coalescence mechanisms in microchannels [11,12]. Under the effects of dispersion and coalescence, the bubble and droplet sizes change dynamically in operating microstructured chemical device. Keeping the bubble and droplet size in an appropriate range is very important for the effective control of heat/mass transfer process and chemical reactions.

In the purpose of enhancing heat/mass transfer and confronting the adverse impact from coalescence, a good approach is to break big gas slug or liquid plug in situ in microchannels. We call this process the “second-dispersion”. For example, using the T-junction or Y-junction microchannel proposed by Link et al., geometrically mediated droplet breakup has been successfully proceeded [13]. In 2013, Hoang et al. [14] did detailed analysis of the design strategies of a bubble-splitting distributor based on T-junction. With well controlled resistance distribution, small and uniform microdroplets can be easily prepared, while using adjustable microchannel temperature or pneumatic valve, different sizes of daughter droplets can also be prepared [15,16]. Making slug or plug flow against a confined obstacle is another method to realize geometrically mediated breakup. Salkin et al. reported this kind of microfluidic second-dispersion process in 2012 [17].

A simpler and more industrial-adapted second-dispersion structure is the constrictive microchannel, which has a small orifice. It has been tested by Choi et al. [18] with several pneumatic valve controlled microfluidic devices. Droplet breakup was observed by them with the orifice width reduced to a critical size and ten times smaller daughter droplets comparing with the mother droplets were successfully obtained in that study. Compared with the static constrictive microchannel, the breakup control is more complicated in the microchannel with pneumatic valve, since the orifice’s shape is complicated with the working of pressure and it is hard to give a general research on the dispersion rules in this kind of microfluidic device. In preparing static constrictive microchannel, it is also easier to obtain different geometries, which is benefit to understand the breakup laws, as Finke et al. did in 2013 [19], who systematically studied orifice design parameters to understand multiple orifices in customized microsystem high-pressure emulsification. Based on these considerations, the aim of this study is to give a systemic experimental study on the liquid plug breakup laws in different static constrictive microchannels, which has not been generally reported so far as we known. The effects of surfactant concentration, viscosities of continuous phase and dispersed phase, two-phase flow rates as well as the channel geometry on the flow behavior of liquid plugs are carefully examined. Critical second-dispersion conditions and the size law of daughter droplets are obtained from the experimental result and breakup mechanism analysis.

2. Experimental section

2.1. Microchannel device

A schematic figure of microchannel devices we used is shown in Fig. 1. A T-junction was used to generate liquid plugs and the constriction was placed at the T-junction’s downstream. All the upstream channels of the orifice has square cross-sections whose width (W_1) and height (H_1) are both 1 mm. The orifice is 15 mm after the T-junction. 4 Orifice geometries were used as $w \times h \times l$ equals to 0.4 mm × 0.3 mm × 1 mm, 0.2 mm × 0.1 mm × 1 mm, 0.4 mm × 0.6 mm × 1 mm, and 0.6 mm × 0.6 mm × 1 mm. The orifice is placed at the top side of the microchannel as shown in the side view of Fig. 1(b). The downstream channel of orifice had two structures, each has equal width (W_2) and height (H_2) as 1 mm and 1.5 mm, respectively. Geometry details and microchannel numbers are presented in Table 1. The microchannels were fabricated on 60 mm × 30 mm × 4 mm polymethyl methacrylate (PMMA) plate using a mechanical milling device, and were sealed to a chip with another 60 mm × 30 mm × 4 mm PMMA plate in a thermal sealing machine (A274, Techson, China) at 75 °C, 0.4 MPa. Limited by the mechanical fabrication method the microchannel ends have round corners. Before any experiments, the microchannels were immersed in the continuous phase for at least 4 h to avoid the wetting effect of the dispersed phase.

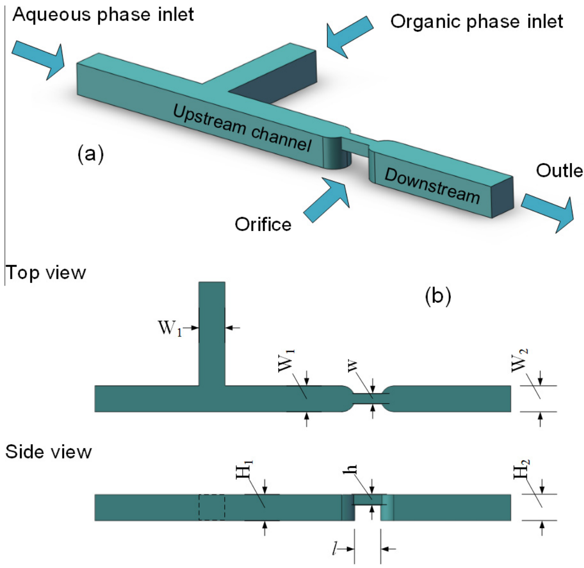

Fig. 1. Structure of microchannel device. (a) 3-D view of the microchannel with a T-junction liquid plug dispersion structure and a constriction microchannel. (b) Top view and side view of microchannel devices.

Table 1
Summary of microchannel geometries.

Microchannel no.	$W_1 \times H_1$	$w \times h \times l$	$W_2 \times H_2$
1	1 mm × 1 mm	0.2 mm × 0.1 mm × 1 mm	1 mm × 1 mm
2	1 mm × 1 mm	0.4 mm × 0.3 mm × 1 mm	1 mm × 1 mm
3	1 mm × 1 mm	0.4 mm × 0.6 mm × 1 mm	1 mm × 1 mm
4	1 mm × 1 mm	0.4 mm × 0.6 mm × 1 mm	1.5 mm × 1.5 mm
5	1 mm × 1 mm	0.6 mm × 0.6 mm × 1 mm	1 mm × 1 mm