

Study on “interface – shrinkage – driven” breakup of droplets in co-flowing microfluidic devices



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ABSTRACT

In co-flowing microfluidic device, it is usually thought the droplet size is significantly influenced by the capillary number in dripping flow. However, it was found in this study that when the contact angle of the dispersed fluid with the dispersed fluid channel was sufficiently large, the droplet size would be little affected by the capillary number. To explain this phenomenon, an “interface – shrinkage – driven” breakup mechanism was proposed. Three different systems and six different microfluidic devices were used in the experiment to prove the universality of the mechanism. The critical contact angle of the dispersed fluid with the dispersed fluid channel was finally discussed.

1. Introduction

In the past decade, microfluidic devices have been the focus of numerous studies because of their high efficiency, safety, repeatability and facile controllability (Ehrfeld et al., 2000). The multiphase flow in these microfluidic devices has received priority for investigation, as it is significant for enhancing and extending the performance of microfluidic systems. At present significant advances have been made in the use of microfluidic devices for controlling multiphase flow patterns and numerous studies on stable microfluidic flow patterns such as drop flow, slug flow and co-laminar flow (Thorsen et al., 2001; Günther and Jensen, 2006; Xu et al., 2006; Zhao and Middelberg, 2011; Hu et al., 2013; Nunes et al., 2013) have been done. These studies have utilized various microstructure geometries including shear-flowing microchannels, cross flowing microchannels, hydrodynamic focusing microchannels, and co-flowing microchannels in an attempt to better understand and control emulsion and flow types in microfluidic devices. The drop and slug flow patterns have been widely used for chemical reaction (Wang et al., 2006), liquid-liquid extraction (Dittrich et al., 2006; Li et al., 2012), biological analysis (Zhang et al., 2015; Tao et al., 2015), crystallization (Zheng et al., 2004), nanoparticle synthesis (Yen et al., 2005; Li et al., 2008), and structural material preparation (Dendukuri and Doyle, 2009; Lan et al., 2011; Nunes et al., 2013; Martino et al., 2016). For these applications, the droplet size, size distribution, flow behavior need to be precisely controlled. Microfluidic technology offers unique characteristics to meet these crucial demands. Monodispersed droplets can be produced in microfluidic devices, and the droplet size can be adjusted in a wide range.

To control the droplet size, the droplet breakup mechanism is an important issue. As a usual phenomenon in the nature and industry, the droplet breakup has been researched for more than one hundred years. Many researchers studied the droplet breakup behavior in different conditions such as turbulent pipe flow (Hughmark, 1971; Rozentsvaig, 1981; Rozentsvaig and Strashinskii, 2016), stir mixing (Shinnar, 1961; Rozentsvaig, 1985), pulsation (Liu et al., 2016), shear flow (Karam and Bellinger, 1968; Verhulst et al. 2009; Xu et al., 2006), etc. Based on these researches, three dimensionless numbers, the Weber number, the Bond number, and the Capillary number were found to be of great significance in describing the droplet behavior. The Weber number ($We = \rho u^2 d / \gamma$) characterizes the relative importance of the inertia compared to the interfacial tension. The Bond number ($Bo = \Delta \rho g d^2 / \gamma$) is used to measure the importance of body forces compared to interfacial tension forces. The Capillary number ($Ca = \mu u / \gamma$) represents the relative effect of viscous forces versus interfacial tension. A great number of research works have been reported to discover the relationship between the droplet breakup behavior and the dimensionless numbers. Especially in microchannels, the Capillary number is usually much larger than the other two dimensionless numbers. Therefore, it is usually thought that the Capillary number dominates the droplet breakup in microchannels (Xu et al., 2006; Lan et al., 2015).

To research deep into the droplet breakup in co-flowing microfluidic devices, it was found that droplets breakup in co-flowing microfluidic devices can be separated into two distinct breakup regimes: dripping in which droplets pinch off near the capillary tip,

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and jetting in which droplets pinch off from an extended thread downstream of the capillary tip (Christopher and Anna, 2007). For dripping regime, droplet breakup is determined by the balance of the shear force and the interfacial force, and the mechanism is called shear-driven breakup (Thorsen et al., 2001). The Capillary number, as described above, is usually used to express this balance. Umbanhowar et al. (Umbanhowar et al., 2000) predicted the droplet size by finding the roots of the following equation

$$\bar{d}^3 - (1 + \frac{1}{3Ca})\bar{d}^2 - \frac{\varphi}{\alpha}\bar{d} + \frac{\varphi}{\alpha} = 0 \quad (1)$$

where $\bar{d}$ is the dimensionless droplet size, α is the parameter of the device geometry and φ is the flow rate ratio. As long as the internal flow rate is slow enough, the above equation reduces to

$$\bar{d} \approx 1 + \frac{1}{Ca} \quad (2)$$

For jetting regime, the droplet size scales with the flow rate ratio as (van Hoeve et al., 2010; Castro-Hernandez et al., 2012; Lan et al., 2015)

$$d \sim \varphi^{1/2} \quad (3)$$

In general, droplets are smaller when the continuous phase velocity is faster in both regimes, due to larger shear stresses exerted on the interface. And the droplet diameter decreases with decreasing interfacial tension in dripping regime due to decreased resistance to breakup (Cramer et al., 2004; Christopher and Anna, 2007; Nunes et al., 2013; Lan et al., 2015; Anna, 2016).

In this study, we found a new droplet breakup mechanism in co-axial microchannel for dripping regime. We called it “interface-shrinkage-driven” breakup. When the droplet breakup is dominated by this mechanism, the capillary number has little influence on the droplet size, which is quite different from “shear-driven” breakup. Besides, the “interface-shrinkage-driven” breakup lead to much smaller droplet size compared to the “shear-driven” breakup.

2. Experiments

2.1. Materials

We chose three different working systems in our experiments. The liquids used in the three systems were listed in Table 1. For all the three systems, the former liquid was served as the dispersed phase and the latter as the continuous phase. The interfacial tensions between them were measured by pendant drop method (OCAH200, DataPhysics Instruments GmbH). The viscosities of the continuous phases were measured using the spinning digital viscometer (NDJ-5S, Shanghai Jinglian Electronic Machine Co. Ltd.). The values were listed in Table 1. All chemicals were analytically pure and purchased from Beijing Chemicals Factory (Beijing, China).

2.2. Experimental set-up

The microfluidic device used is shown in Fig. 1. The device was fabricated on a 50 mm×20 mm×3 mm PMMA plate using micromachining technology. A Teflon tube was inserted as the main channel for the multiphase flow, while a capillary was inserted into the tube co-axially for dispersed phase flow. The microfluidic device was sealed using another 50 mm×20 mm×2 mm PMMA plate with ultrasonic assisted sealing technique (Li et al., 2009). Three different materials were selected as the dispersed fluid channel (DFC) – polypropylene (PP), Teflon and glass. PP capillary was self-pulled to change the inner diameter. Six microfluidic devices were used in this study. Their geometry parameters were listed in Table 2. Three micro-syringe pumps and three gastight micro-syringes were used to pump the fluids into the microfluidic device. A microscope coupled with a high-speed

Table 1
Physical properties of the three systems.

No.	System	Viscosity of continuous fluid (mPa s)	Interfacial tension (mN/m)	Contact angles (°)		
				PP	Teflon	Glass
(O1)	Water/1-octane	0.51	48.09	160	148	70
(O2)	Water/0.3 m ³ /m ³ Tributyl phosphate (TBP) in 1-octane	0.77	11.70	165	150	77
(O3)	Water/1-octanol	9.42	8.72	135	131	86

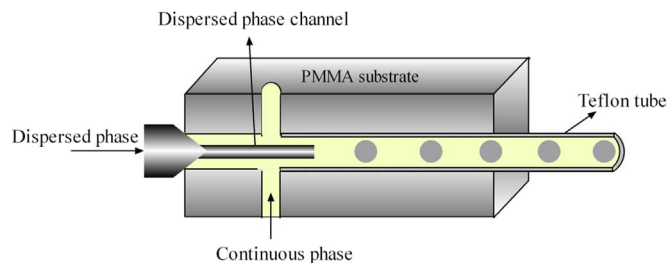


Fig. 1. The co-axial microdevice.

Table 2
The parameters of the microfluidic devices.

Device no.	Materials of DFC	Inner diameter of main channel D_i (μm)	Inner diameter of DFC d_i (μm)
(D1)	PP	760	267
(D2)	PP	1000	270
(D3)	PP	2000	258
(D4)	PP	2000	400
(D5)	Teflon	760	250
(D6)	Glass	760	200

CCD video camera was used to observe the flow pattern in the microfluidic device. Images were recorded at speed of 200 frames per second. The droplet diameter was measured from recorded images using custom-made image-analysis software. Immersed in continuous fluid, the contact angel of water with the three different materials (PP, Teflon and glass) was measured. The results were listed in Table 1.

3. Results and discussion

3.1. Droplet generation process

Previous studies usually focus on the effect of wettability of the main channel, which determines the emulsion type. In our experiment, we found that the wettability of the DFC has significant influence on the flow behavior. We investigated the flow pattern of system O2 in device D1, D5 and D6. The three devices had similar dimensions but different DFC materials. The droplet generation process is shown in Fig. 2. When using Teflon or glass as DFC, the DFC is fully filled by the dispersed fluid in the droplet generation process. When the droplet grows big enough, the neck connecting the droplet and the dispersing channel starts to shrink, and finally break up. Here the neck forms outside the DFC. This phenomenon is similar with those reported in previous studies (Cramer et al., 2004; Christopher and Anna, 2007; Nunes et al., 2013; Lan et al., 2015; Anna, 2016). However, when using PP as the DFC, the phenomenon is different. When the droplet grows to a specific size, the continuous fluid flows along the wall into the DFC. Then the neck forms, shrinks and breaks up inside the DFC. After the

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