Velocity measurements of individual droplets in liquid-liquid Taylor flows in circular capillaries

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Abstract. This study is focused on the effect of droplet length on droplet velocity in liquid-liquid Taylor flows for microfluidic applications. An experimental set up was designed to measure droplet velocity over a wide range of droplet lengths and flow velocities while also varying viscosity ratio. Five different fluid combinations were examined by employing AR20, FC40, HFE7500 and water. Results indicate the complexity of predicting droplet velocity in such flow regimes and also show a strong influence of viscosity ratio and Bond number.

1. Introduction
The use of liquid-liquid two-phase flows within mini/micro channels has been shown to offer much higher heat transfer coefficients compared to traditional single-phase flows [1]. Liquid-liquid two phase flows consist of two immiscible fluids flowing in a channel. The dispersed phase forms droplets within the carrier phase and does not wet the wall as a thin film (film thickness) of the carrier phase is present (See figure 1). As the fluids move within the channel internal circulations result in an enhanced mixing rate and increased heat and mass transfer compared to single phase flow [2]. Chemical processing, DNA analysis and nitration reactions are all examples of applications that take advantage of two-phase flows in microreactor technology [3,4,5]. However, the successful operation of all applications incorporating such flows depends on droplet transportation in confined channels. Therefore, a comprehensive understanding of how such droplets behave is key to achieving optimum performance. It has been shown by various studies that several parameters affect droplet mobility in liquid-liquid flows including channel geometry, droplet size, presence or absence of surfactants, flow velocity, viscosity of liquids and interfacial tension [6]. Over the past decade a number of studies, mostly focusing on liquid-gas flows, have been carried out to characterize the dispersed phase within milli and micro scale channels [7,8]. Despite this, the relationship between the governing dimensionless groups and the dispersed phase is still unclear and not fully understood. In this regard, the current experimental study is focused on droplet characteristics in liquid-liquid flows with the objective of determining the effect of droplet size on droplet velocity. An automated system is designed to monitor the speeds of individual droplets at different flow velocities with a high accuracy. Measurements were obtained over a wide range of carrier to dispersed viscosity ratios (0.06 < μc/μd < 23.2), droplet length to capillary diameter ratios (0.8 < Ld/D < 16), Bond numbers (0.05 < Bo < 3.2) and flow velocities (7.3 < U < 53.6mm/s). Five different flow combinations were generated within round capillaries with inner diameters of 1.6mm and 0.55mm.

2. Experimentation
The experimental configuration used to measure slug length and velocity is shown in figure 1. Droplets were generated in 1.6 and 0.55mm ID, 5.7m length FEP Teflon tubes (Cole Parmer™) using a custom designed traverse stage and reservoir. The tube inner diameter was confirmed using the procedure outlined in the supplementary material (S1). To generate droplets, the tube was firstly primed with the carrier liquid. Following this, both liquids were drawn into the tube by running a Harvard Pico Plus Elite syringe pump, located at one end of the tube, in withdraw mode and simultaneously vertically moving the traverse/dipping stage into a reservoir containing both immiscible fluids. The traverse system and syringe are controlled and synced using a G-Code in order to generate individual droplets.
with different lengths \( (L_D) \). As the tip of the tubing enters the dispersed liquid, the pump draws in the desired volume of that liquid. 10ml Hamilton glass syringes were used during testing to minimize any uncertainty due to deformation of the barrel and piston during the experiments. In each experiment, only a single droplet was formed and then monitored in the test section.

Droplet length \( (L_D) \) and speed \( (U_D) \) were determined using two photodiodes and two LEDs located along tube as shown in figure 1. Photodiodes with integrated amplifiers (OPT301M), provided by Burr-Brown were used. As a droplet passed each photodiode the light intensity, emitted by white-LEDs, and recorded by each sensor, changes due to reflection and the different refractive indices of the carrier and dispersed liquids. This change in light intensity results in a change in the voltage output of the photodiode and is recorded as a step change and used to determine \( L_D \) along with droplet velocity. Further details and results from this sub-system are presented in the supplementary material (S2). A CCD camera with maximum frame rate of 30 fps was employed for visualization purposes. Five different flow combinations were used in this study; in three flow combinations FC40, HFE7500 and AR20 were chosen as carrier fluids while water was the dispersed phase and in two other flows AR20 was dispersed in FC40, HFE7500. The viscosity ratio \( (\mu_C/\mu_D) \) changes from 0.059 (HFE7500-AR20) to 23.2 (AR20-Water) to allow for an analysis of effect of this parameter on the hydrodynamics of the flows. Measurements were obtained over a wide range of Reynolds \( (0.7 \text{ to } 10^4) \), Capillary \( (5 \times 10^{-8} \text{ to } 3.7 \times 10^{-6}) \) and Weber \( (0.003 \text{ to } 3.7) \) numbers. Further details on the various liquids (fluid properties) and range in dimensionless groups are presented in the supplementary material (S3). The interfacial tension of all liquids was measured using a commercial CAM 2000 Pendant Drop system detailed in the supplementary material (S4).

Figure 1.

Experimental configuration for droplet length and velocity measurements and illustration of two-phase flow structure highlighting droplet length \( L_D \), film thickness \( \delta \) and internal circulations within the flow.

3. Results

Figure 2 presents droplet velocity measurements for all fluid combinations. Results from AR20-Water, FC40-Water and HFE7500-Water with carrier to dispersed viscosity ratios greater than unity are presented in Figure 2(a). Figure 2(b) presents results from FC40-AR20 and HFE7500-AR20 where the viscosity ratio is less than one. In these graphs, the X-axis represents the droplet length normalized by the capillary diameter and the Y-axis is the ratio of droplet velocity to mean flow velocity. Measurements were performed over four different flow velocities while droplet length ranged from a very low value \( (L_D/D \approx 0.7) \) up to the maximum possible value in each flow combination. During measurements, it was observed that from a certain length, droplets began to split and separate with the likelihood of this occurrence increasing at higher droplet velocities. A first indication of droplet break-
up might be given by the Weber number, which links inertia forces with surface tension. In this study, experiments with HFE7500-AR20 at the maximum Weber number range (0.1 < \(\text{We} < 3.7\)), presents the minimum droplet length ratio (0.8 < \(L_D/D\) < 7).

From Figure 2(a) it can be observed that by increasing the flow velocity, U, the droplet velocity ratio also increases. This increment is associated with the parabolic velocity profile across the capillary cross-section. It has been shown [9] that the thickness of the film, \(h\), separating the droplet from the capillary wall increases with velocity, hence causing the dispersed phase (droplet) velocity to also increase. In experiments with AR20-Water, for long droplets (\(L_D/D \geq 2.5\)), droplet velocity is almost constant and not affected by droplet length. A similar result is also seen for experiments with FC40-Water and HFE7500-Water at \(L_D/D \geq 2\). On AR20-Water, as \(L_D/D\) reduces below 2.5 droplet velocity decreases significantly. This reduction in droplet velocity is believed to be associated with the flow topology inside the droplet and the absence of well-formed internal circulations causes higher hydrodynamic resistance of the droplet [6]. This behaviour is not observed in experiments with FC40-Water and HFE7500-Water. In these cases, viscosity ratios are low, \(\mu_c/\mu_d = 4.3\) for FC40-Water and \(\mu_c/\mu_d = 1.3\) for HFE7500-Water, in comparison to the AR20-Water with \(\mu_c/\mu_d = 23\) and it has been shown that by decreasing viscosity ratio, internal circulations within droplets are suppressed and a more uniform flow pattern is exists within the more viscous droplets [10]. Therefore, in cases with a low viscosity ratio, the effect of internal flow topology on the variation of droplet velocity with droplet length has been diminished due to the weak internal circulations within the droplet. In AR20-Water a further reduction in droplet length below \(L_D/D \sim 1.2\), causes the droplet velocity to increase considerably. This is due to the fact that droplet cross-sectional area is much smaller than the capillary cross-sectional area and the droplet can move with a much higher velocity. In these cases, Taylor flow is said to no longer exist with the channel. For experiments with FC40-Water and HFE7500-Water, this increase only occurred at the maximum flow velocity of the experiments (49.3mm/s) and at lower flow velocities further decreasing of droplet length resulted in significant reduction in droplet velocity. This reduction in droplet velocity is caused by increased Bond number and the influence of buoyancy. Results from FC40-AR20 and HFE7500-AR20 with viscosity ratios of 0.18 and 0.6 respectively, presented in Figure 2(b), show a similar trend where very small droplets move with a lower velocity and in some cases slower than the mean flow velocity.

Figure 3 presents images captured by a high-speed camera from FC40-Water experiments. These images show a side view of a water droplet engulfed by FC40 as the carrier phase within the capillary at two different velocities but constant droplet volume. It is clear that, at a flow velocity (8.2mm/s) the droplet is not moving in the centre of the channel but is closer to the top surface. At a higher flow velocity (49.3mm/s), this vertical displacement of the droplet is not evident as seen in Figure 3 (b). The Bond number (ratio of buoyancy to interfacial tension forces, \(\Delta \rho g l^2/\gamma\)) for AR20-Water is 0.1 but in cases with FC40-Water and HFE7500-Water Bond numbers are 0.3 and 0.47 respectively and buoyancy
force presents a greater impact. By increasing the flow velocity, a thicker film separates the droplet from the top surface and the droplet is moved to align symmetrically with the capillary centre line. Figure 3(c) shows the results of FC40-water experiments with a smaller capillary diameter (0.55mm) and a lower Bond number (Bo=0.05). In this case, the effect of buoyancy force on droplet velocity has been clearly eliminated. In FC40-AR20 and HFE7500-AR20 with the high Bond number values of 3.2 and 1.7 respectively, buoyancy force affects the droplet velocity even at the maximum flow velocity of the experiments.

Figure 3. Images of water dispersed in FC40 with a constant droplet volume over velocities of (a) 8.2mm/s and (b) 49.3mm/s while the Bond number is 0.3 and (c) variation in droplet velocity ratio with dimensionless droplet length for experiments with FC40-Water while the Bond number is 0.05.

4. Conclusions
Droplet velocity in liquid-liquid Taylor flows was investigated experimentally by means of five different fluid combinations over a wide range of viscosity ratios and flow velocities. The results indicate a complex dependency of droplet velocity on various parameters in particular, an effect of viscosity ratio and Bond number on droplet mobility. As the Bond number increases, buoyancy forces dominate the interfacial forces and cause the droplet to move asymmetrical with the respect channel center. This phenomenon along with the parabolic velocity profile in the carrier phase, contributed to a sharp reduction in velocity ratio at lower LD/D values.

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References
[1] Eain M M G, Egan V and Punch J 2015 Local Nusselt number enhancements in liquid–liquid Taylor flows *International Journal of Heat and Mass Transfer* **80** 85-97
[2] Malisch D, Kielpinski M, Merthan R, Alber, J, Mayer G, Köhler J M, Sülle H, Stahl M and Henkel T 2008 μPIV-analysis of Taylor flow in micro channels *Chemical Engineering Journal* **135** S166-S172
[3] Khan W, Chandra A K, Kishor K, Sachan S and Alam M , 2017 Hydrodynamics and simulation studies of liquid-liquid slug flow in micro-capillaries *International Conference on Advances in Mechanical, Industrial, Automation and Management Systems (AMIAMS)* 281-284 IEEE
[4] Burns M A, Johnson B N, Brahmasandra S N, Handique K, Webster J R, Krishman M, Sammarco T S, Man P M, Jones D, Heldsinger D and Mastrangelo C H 1998 An integrated nanoliter DNA analysis device *Science* **282** 484-487
[5] Fidalgo L M, Whyte G, Bratton D, Kaminski C F, Abell C and Huck W T 2008 From microdroplets to microfluidics: selective emulsion separation in microfluidic devices *Angewandte Chemie* **120** 2072-2075
[6] Jakiela S, Makulska S, Korczyk P M and Garstecki P 2011 Speed of flow of individual droplets in microfluidic channels as a function of the capillary number, volume of droplets and contrast of viscosities Lab on a Chip 11 3603-3608

[7] Fuerstman M J, Lai A, Thurlow M E, Shevkoplyas S S, Stone H A and Whitesides G M 2007 The pressure drop along rectangular microchannels containing bubbles Lab on a Chip 7 1479-1489

[8] Hodges S R, Jensen O E and Rallison J M 2004 The motion of a viscous drop through a cylindrical tube Journal of fluid mechanics 501 279

[9] Eain M M G, Egan V and Punch J 2013 Film thickness measurements in liquid–liquid slug flow regimes International journal of heat and fluid flow 44 515-523

[10] Ma S, Sherwood J M, Huck W T and Balabani S 2014 On the flow topology inside droplets moving in rectangular microchannels Lab on a Chip 14 3611-3620