Prediction Models of Droplet Characteristics Based on the Locally Convergent Configurations in PMMA Microchips

Zhongxin Liu¹, Zhiliang Wang², Chao Wang¹ and Jinsong Zhang*¹

¹ School of Mechatronic Engineering and Automation, Shanghai University, Shanghai City, 200444, China
² School of Mechanics and Engineering Science, Shanghai University, Shanghai City, 200444, China
Email: zhangjs@shu.edu.cn

Abstract. This paper novel designed the local convergence configuration in the coaxial channels to study the two-phase flow (lubricating oil (continuous phase, flow rate \( Q_c \)) / deionized water (dispersed phase, flow rate \( Q_d \))). Two geometric control variables, the relative position (x) and tapering characteristics (\( \alpha \)), had the different effects on the droplet formation. The increase of relative position x caused the higher frequency and finer droplets, and the increase of convergence angle \( \alpha \) took the opposite effects. The results indicated that the equivalent dimensionless droplet length \( L_d/W_{out} \) and the flow rate ratio \( Q/Q_c \) had an exponential relationship of about 1/2. Similarly, it was found that the dispersed droplets generating frequency and the two-phase capillary number, \( \text{Ca}_{TP} = u_{ref}d/f \sigma \), had an exponential relationship. The advantage of the convergent configurations in micro-channel was the size and efficiency of droplet generation was very favorable to be controlled by \( \alpha \) and x.

Keywords. Prediction model, Two-phase flows, Droplet generation, Convergent coaxial flow

1. Introduction

Lots of literatures have shown that there is a power function relationship between then droplet size and the two-phase flow rate ratio, \( L_d/W_{out} \propto (Q/d)^{0.5} \), called the ‘flow-rate-controlled-flow’ diagram [1–7] in certain flow parameter zones, where the range of \( k \) is \( -0.5 \sim 1 \). There are relatively less studies on droplet frequency \( f \). It was proposed that there are many different relationships between the droplet frequency and other parameters \( f \propto (Q/d)^{0.5} \) [8], \( f \propto u_dL_d/W_{out} \) [9], and \( f \propto L_d/W_{out} \) [10, 11], where \( L_d \) is the characteristic capillary time. It can be seen that the prediction models of droplet frequency are not consistent or physically conflicting, and need further in-depth studies.

Prior to those, we put forward a convergent geometry of coaxial micro-channel in our early work [12], which is characterized by the tapering cone-shaped outer tube and the inner needle in a round configuration. Recently, we took the experimental studies of micro-channel tapering on droplet forming acceleration in liquid paraffin/ethanol coaxial flows [13]. Two convergence angles (\( \alpha = 0^\circ \) and \( \alpha = 2.8^\circ \)), limited to the constrains of making perfect conical inner wall by stretching glass tube while heating, were considered to show the big differences of flow charts and droplet characters on slight tapering in round co-axial digital micro-flows. Based on the previous theoretical and numerical results, in this paper we intend to take a more detailed discussion of the effects about tapering geometry on droplet forming through experiments. PMMA microchips of rectangle cross-section channels, instead of round millimeter tubes, are adopted, and serials of samples are observed and analyzed.
2. Materials and Micro-Channel
The materials used are 5W-20 lubricating oil (continuous phase, density $\rho_c=826 \text{ kg/m}^3$, viscosity $\mu_c=40.96 \text{ mPa}\cdot\text{s}$, interfacial tension $\sigma=20.08 \text{ mN/m}$) and deionized water (dispersed phase, density $\rho_d=986.2 \text{ kg/m}^3$, viscosity $\mu_d=1.23 \text{ mPa}\cdot\text{s}$, interfacial tension $\sigma=20.08 \text{ mN/m}$) separately.

![Figure 1. The co-flowing in the tapering micro-channel geometry.](image)

**Table 1.** Two parameters to define tapering geometries of PMMA microchip samples and four typical flow patterns.

| Samples | Convergence Angle $\alpha$ (°) | Nozzle Stretch $x$ (μm) | Flow pattern |
|---------|--------------------------------|-------------------------|--------------|
| A3      | 5                              | 1170                    | ![Slag](image) |
| B3      | 11                             | 1170                    | ![Dripping](image) |
| C3      | 17                             | 1170                    | ![Jetting](image) |
| D3      | 29                             | 1170                    | ![Sausages](image) |
| C1      | 17                             | 0                      |              |
| C2      | 17                             | 620                     |              |
| C4      | 17                             | 1620                    |              |
| C5      | 17                             | 2160                    |              |
| E       | 0                              | -                       |              |

Figure 1 shows the coaxial flow configuration and geometric parameters of convergent micro-channel on a PMMA microchip. The dispersed phase and continuous phase flow in from the left side, and flow out toward the right side. Both phases mix at the convergence area, whose length is $X_L = 2680 \mu m$ and convergence angle $\alpha$. The syringe needle, whose inner diameter is $d = 200 \mu m$ and outer diameter $D = 400 \mu m$, of dispersed phase is inserted into this tapering area to form two-phase flow (region 1). The downstream micro-channel is a straight tunnel (region 2), with rectangular cross-section of width $W_{out} = 400 \mu m$ and height $h = 600 \mu m$. $L_d$ is the axial size (length) of the droplet generated.

Many samples of PMMA microchips are prepared. The combinations of the two geometrical parameters, convergence angle $\alpha$ and needle displacement $x$, are shown in table 1, labelled as sample A3, B3, C3, D3, C1, C2, C4, C5 and E.
3. Result and Discussion

3.1. Scaling up Droplet Length in Slug and Dripping Regimes

Figure 2(a) shows the variation of \( \frac{L_d}{W_{\text{out}}} \) corresponding to the increase of the flow rate ratio \( \frac{Q_d}{Q_c} \) at different needle displacements \( x \). The dashed line is an empirical slug-dripping line at \( \frac{L_d}{W_{\text{out}}} \approx 1.2 \) to divide these two regimes. Above which, such as \( \frac{L_d}{W_{\text{out}}} > 1.2 \), is the slug regime for much bigger droplets. We find the linear correlation between \( \frac{L_d}{W_{\text{out}}} \) and \( \frac{Q_d}{Q_c} \) in double logarithm coordinates, which can be described as

\[
L_d / W_{\text{out}} = A \left( \frac{Q_d}{Q_c} \right)^k,
\]

where \( A \) represents fitting coefficients, and exponent \( k \) is \( 0.49 \pm 0.05 \) under variation of the needle displacement \( x \).

![Figure 2. Droplet length \( \frac{L_d}{W_{\text{out}}} \) varies with \( \frac{Q_d}{Q_c} \). (a) Samples C1–C5 for \( \alpha = 17^\circ \) with \( x \) changing from 0, 620, 1170, 1620, to 2160 \( \mu \text{m} \); (b) samples A3, B3, C3 and D3 for \( x = 1170 \mu \text{m} \) with \( \alpha \) changing from 5°, 11°, 17° to 29°.](image)

Similar to figure 2(a), figure 2(b) gives the variation of \( \frac{L_d}{W_{\text{out}}} \) corresponding to the increase of the flow rate ratio \( \frac{Q_d}{Q_c} \) at different convergence angles \( \alpha \). The same fitting formula equation (1) is derived even for the scaling exponent \( k \).

These are very interesting results, and it is not only for the same scaling law of droplet length for both slug and dripping regimes. It shows that the increase of inner needle displacement \( x \) can produce finer droplets, which is consistent with our previous results obtained in round tube experiments and simulations. While increasing the convergence angle \( \alpha \), contrarily, takes opposite effects. Hence, there must exist an intrinsic variable to reflect these phenomena. Analyzing the essential physical or geometrical characteristics, we find that in the tapering configuration, both the increase of needle displacement \( x \) and the decrease of convergence angle \( \alpha \), cause that the outer flow (continuous oil phase) meets the inner flow at a relative small channel width, which means that the outer flow is accelerated. Therefore, the local micro-channel width right at the inner needle exit, \( W_{\text{local}} \), can be the intrinsic variable, and is defined by

\[
W_{\text{local}} = W_{\text{out}} + 2(X_\text{c} - x) \tan(\alpha / 2) .
\]

Hence, we rearrange the data in figure 2(a) and figure 2(b) by multivariate nonlinear regression analysis and replot, as shown in figure 3. Here, the formula can be derived as

\[
L_d / W_{\text{local}} = 1.32 \left( \frac{Q_d}{Q_c} \right)^{0.69} .
\]
In equation (3), all the lines in Figure 2(a) and Figure 2(b) collapse to form a single line, and the exponent and coefficient are given by constants. Therefore, the influence of the needle displacement $x$ and convergence angle $\alpha$ is unified.

Figure 3. Droplet length $L_d/W_{local}$ varies with $Q_d/Q_c$ for samples C1–C5, A3, B3 and D3.

3.2. Scaling up Droplet Generating Frequency in Slug and Dripping Regimes

In the present tapering micro-channel, the influence of the needle displacement $x$ and the convergence angle $\alpha$ on the droplet length $L_d$ is exponentially related to the water/oil flow rate ratio $Q_d/Q_c$, while the droplet-generating frequency $f$ is exponentially related to the total water and oil flow rates $Q_{total} = Q_d + Q_c$, as shown in figure 4(a) and figure 4(b).

Figure 4. Droplet forming frequency $f$ varies with $Q_{total}$. (a) Samples C1–C5 for $\alpha = 17^\circ$ with $x$ changing from 0, 620, 1170, 1620, to 2160$\mu$m; (b) samples A3, B3, C3 and D3 for $x = 1170\mu$m with $\alpha$ changing from 5$, 11^\circ$, 17$^\circ$ to 29$^\circ$.

When the needle displacement $x$ and convergence angle $\alpha$ are fixed, the droplet frequency $f$ shows an increasing trend with the increase of the two-phase flow rate $Q_{total}$. When the convergence angle $\alpha$ is constant, the droplet frequency $f$ increases with the increase of the nozzle needle displacement $x$, as shown in figure 4(a). When the nozzle needle displacement $x$ is constant, the droplet frequency $f$ decreases as the convergence angle $\alpha$ increases, as shown in figure 4(b). In both figure 4(a) and figure 4(b), we gain relation for the droplet frequency $f$ with $Q_{total}$ formulated as

$$f = A Q_{total}^k,$$  (4)
where $A$ represents experimental coefficients separately, and $k = 1.36 \pm 0.05$ is the corresponding exponent. For the same reasons of deriving equation (3), we can combine the influence of the needle displacement $x$ and convergence angle $\alpha$ on the droplet frequency $f$ into a degenerated form. By introduce the capillary time $t_{cap} = \sqrt{\rho d^3 / \sigma}$, we nondimensionalize equation (4), and the dimensionless droplet forming frequency $f \cdot t_{cap}$ can be expressed as

$$f \cdot t_{cap} = 1.82 Ca_{TP}^{1.36}$$

(5)

where $Ca_{TP} = \mu \mu_c / \sigma$ represents the two-phase capillary number defined on the downward velocity and oil viscosity, and $k = 1.36$ is the exponent. As seen in figure 5, the error between equation (5) and experimental data is less than 30%.

3.3. Extend the Present Models to Non-Tapering Geometries, Test and Compare

Sample E, listed in table 1, is a configuration without tapering (convergence angle $\alpha = 0^\circ$). We use equation (3) to predict the dimensionless droplet length $L_d / W_{local}$, compare the predicted values ($L_d / W_{local}$)$_{pre}$ with their experiment values ($L_d / W_{local}$)$_{exp}$ directly from sample E, and plot in figure 6(a). Also, We use equation (5) to predict the dimensionless droplet-generating frequency $f \cdot t_{cap}$, compare the predicted values ($f \cdot t_{cap}$)$_{pre}$ with their experiment values ($f \cdot t_{cap}$)$_{exp}$ from sample E, and plot in figure 6(b). It is found that the errors are both less than 20%. From figure 6(a) and figure 6(b), we can find that the predictive models are proved to fit usage of non-tapering geometries.

Figure 5. Dimensionless droplet forming frequency $f \cdot t_{cap}$ varies with $Ca_{TP}$ for samples C1–C5, A3, B3 and D3.

Figure 6. Scaling laws extend to predict for and compare with the experiment data from sample E. (a) ($L_d / W_{local}$)$_{pre}$ vs. ($L_d / W_{local}$)$_{exp}$, (b) ($f \cdot t_{cap}$)$_{pre}$ vs. ($f \cdot t_{cap}$)$_{exp}$.
To end our discussion, we would like to compare our scaling models with those derived for straight (non-tapering) micro-channels [14–16], as listed in Table 2. The original formula is translated into the normalized models, as in equation (6)–(9), for comparing on the same bases. We use all these models as in Table 2 on a non-tapering geometry sample E and on a tapering geometry sample C3 for comparing. Only the droplet length is considered here, owing to lack of literature work on droplet forming frequency. The correlation between the predictive model values \((L_d/W_{out})_{pre}\) and the experimental data \((L_d/W_{out})_{exp}\) is plotted in Figure 7.

Table 2. Predictive model in this paper vs. those derived only for non-tapering structure.

| Literature | Original predictive model | Normalized model | Equation |
|------------|---------------------------|-----------------|----------|
| Present    | \(L_d/W_{local} = 1.32(Q_d/Q_c)^{0.49}\) | \(L_d/W_{out} = 1.32(Q_d/Q_c)^{0.49} (W_{local}/W_{out})\) | (6) |
| Ref.[14]   | \(d/W = 1.277(Q_d/Q_c)^{0.031} C_d^{-0.18}\) | \(L_d/W_{out} = 1.277(Q_d/Q_c)^{0.031} C_d^{-0.18}\) | (7) |
| Ref.[15]   | \(L/W = 1 + Q_d/Q_c\) | \(L_d/W_{out} = 1 + Q_d/Q_c\) | (8) |
| Ref.[16]   | \(L_d/D_t = 2.2(Q_d/Q_c)^{0.7} C_d^{0.1}\) | \(L_d/W_{out} = 2.2(Q_d/Q_c)^{0.7} C_d^{0.1}\) | (9) |

In Figure 7, the diagonal means the perfect coincidence between the predicted values \((L_d/W_{out})_{pre}\) and the experimental values \((L_d/W_{out})_{exp}\) for the upper left \((L_d/W_{out})_{pre} > (L_d/W_{out})_{exp}\), or reversely, \((L_d/W_{out})_{pre} < (L_d/W_{out})_{exp}\). It is shown that the present model is solid for sample C3 and sample E within 20% error limit lines, which means its validation both for non-tapering and tapering geometries. Lin’s model [16] shows well consistency for non-tapering sample E, but cannot extent its prediction to tapering sample C3, where its predicted value \((L_d/W_{out})_{pre}\) is lower than the corresponding experimental value \((L_d/W_{out})_{exp}\). Moreover, both Tostado’s [14] and Xiong’s [15] prediction models overestimate and give high deviation from the related experimental value both on non-tapering sample E and tapering sample C3.

4. Conclusion
We find unified scaling laws valid for both of slug and dripping regimes, not only on the droplet length, but also on their forming frequency. The droplet length is exponentially proportional to the water/oil flow rate ratio, and the droplet forming frequency is exponentially proportional to the two-phase capillary number. These laws, though derived from tapering geometries, can be extended to non-tapering cases. Therefore, we can simulate straight channel flow results in tapering ones and realize real-time and effective manipulations of droplet forming characteristics. The comparisons with the literatures on non-tapering cases also show the validation and universality of the present work.
Acknowledgements
This article is funded by the National Key Research and Development Program “8.5 Generation Printing OLED Display Industrial Demonstration Project” (2017YFB0404503).

References
[1] Tice J D, Song H, Lyon A D and Ismagilov R F 2003 Formation of droplets and mixing in multiphase microfluidics at low values of the reynolds and the capillary numbers Langmuir 19 9127–33
[2] Ward T, Faivre M, Abkarian M and Stone H A 2005 Microfluidic flow focusing: Drop size and scaling in pressure versus flow-rate-driven pumping Electrophoresis 26 3716–24.
[3] Xu J H, Luo G S, Li S W and Chen G G 2005 Shear force induced monodisperse droplet formation in a microfluidic device by controlling wetting properties Lab on A Chip 6 131–136.
[4] Garstecki P, Fuerstman M J, Stone H A and Whitesides G M 2006 Formation of droplets and bubbles in a microfluidic t-junction scaling and mechanism of break-up Lab on a Chip 6 437–446
[5] Cubaud T and Mason T G 2008 Capillary threads and viscous droplets in square microchannels Physics of Fluids 20 501–123.
[6] Xu J, Li S, Tan J and Luo G 2008 Correlations of droplet formation in t-junction microfluidic devices: from squeezing to dripping Microfluidics and Nanofluidics 5 711–717.
[7] Sheu T S, Chen Y T, Lih F L and Miao J M 2010 Ferrofluid-in-oil two-phase flow patterns in a flow-focusing microchannel Physics Procedia 9 147–151.
[8] Derzsi L, Kasprzyk M, Plog J P and Garstecki P 2013 Flow focusing with viscoelastic liquids Physics of Fluids 25 368–141.
[9] Lian J, Luo X, Huang X, Wang Y, Xu Z and Ruan X 2019 Investigation of microfluidic co-flow effects on step emulsification: Interfacial tension and flow velocities Colloids and Surfaces A: Physicochemical and Engineering Aspects 568 381–390.
[10] Liu C, Zhang Q, Zhu C, Fu T, Ma Y and Li H Z 2018 Formation of droplet and string of sausages for water-ionic liquid [bmim][pf6] two-phase flow in a flow focusing device Chemical Engineering and Processing - Process Intensification 125 8–17.
[11] Qian J, Li X, Gao Z and Jin Z 2009 Mixing efficiency and pressure drop analysis of liquid-liquid two phases flow in serpentine microchannels Journal of Flow Chemistry 1–11.
[12] Wang Z L 2015 Speed up bubbling in a tapered co-flow geometry Chemical Engineering Journal 263 346–355.
[13] Zhang J, Wang C, Liu X, Yi C and Wang Z L 2020 Experimental studies of microchannel tapering on droplet forming acceleration in liquid paraffin/ethanol coaxial flows Materials 13 1–16.
[14] Tostado C, Xu J, Du A and Luo G 2012 Experimental study on dynamic interfacial tension with mixture of sdspeg as surfactants in a coflowing microfluidic device Langmuir 28 3120–28.
[15] Xiong R, Bai M and Chung J N 2007 Formation of bubbles in a simple co-flowing micro-channel Journal of Micromechanics & Microengineering 17 1002.
[16] Lin X, Bao F, Tu C, Yin Z, Gao X and Lin J 2019 Dynamics of bubble formation in highly viscous liquid in co-flowing microfluidic device Microfluidics and Nanofluidics 23