

Effect of mixture velocity on flow topology inside Taylor plugs in a microchannel: Experiments and numerical simulations

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We use microparticle image velocimetry and numerical simulations to study the vortex evolution in liquid-liquid plug flows in a 400 μm T-junction microchannel. Experiments are conducted using the [C4mim][T₂fN] ionic liquid as the continuous phase and a glycerol-water blend as the dispersed phase, giving a dispersed to continuous phase viscosity ratio, λ , of 0.117. The range of mixture velocities studied is 0.002 – 0.03 m/s, which corresponds to a capillary number range of $\text{Ca}_p \sim 0.0075 - 0.16$. We characterize the vortical structures based on geometry, vorticity, and circulation times within the plugs. Good agreement is found between the experimental data and the numerical predictions in terms of plug length, film thickness, and circulation patterns. The plug length is found to decrease while the film thickness increases with Ca_p . Three pairs of vortices are formed within the plug in a reference frame moving with the steady plug speed. The pairs comprise a main pair that occupies the central region of the plug and two secondary pairs located at the plug front and rear. The secondary vortices diminishment with Ca_p is quantified based on their geometry and vorticity, subsequently correlated to the critical film thickness for vortex loss based on the predictions of Balestra *et al.* [*Microfluid. Nanofluid.* **22**, 67 (2018)]. It is found that the front and rear secondary vortex pairs diminish at different rates depending on Ca_p . With increasing Ca_p , based on their size, the rear vortex pair is lost before the front one, while, based on vorticity, only the front vortex pair is lost.

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I. INTRODUCTION

Two-phase flows in small and microchannels have been extensively investigated as a means of process intensification [1]. This intensification is predominantly driven by high surface area to volume ratios and short diffusion lengths that enhance heat [2–4] and mass [5,6] transfer rates, often to reach orders of magnitude higher than those of conventional systems [7–9]. One of the primary applications of small channels is in intensified liquid-liquid solvent extraction [10,11]. Driven by

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applications such as secondary resource processing [12] and requirements for increased efficiencies, solvent adaptations are being made. Among the solvents that are being developed, ionic liquids (ILs) have played a prominent role, as they are safer compared to organic solvents and offer improved extraction efficiencies [5,13]. However, they are not widely used because of their cost. The use of IL within micro and small channels therefore offers a promising solution in which channel confinement offers increased efficiencies with reduced solvent demand [11].

However, it is important to note that ILs generally have high viscosities, which can pose challenges in handling and utilization in small channels, such as large pressure drops. In addition, Scheiff *et al.* [14] and Balestra *et al.* [15] have discussed that a more viscous continuous phase in a small channel may be detrimental to mass transfer due to the effect on flow topology and hydrodynamics (e.g., recirculation time, film thickness, and flow pattern characteristics), leading to reduced mass transfer rates as documented by Van Stee *et al.* [16], Vereycken *et al.* [17], and Pheasey and Angeli [18]. To support their use in micro/small channel applications, studies are needed on the effects of the properties of ILs, and particularly their viscosity, on hydrodynamics and mass transfer.

Mass transfer in small channels is influenced by both the flow pattern (hydrodynamics) and the flow topology (mixing). Hydrodynamic studies have focused primarily on the plug (or segmented, the so-called Taylor) flow regime [19], formed when surface tension dominates. In this regime, elongated drops of one phase, with size larger than the channel diameter, are separated by continuous phase slugs, while a thin film exists between them and the wall, resulting in large surface areas for mass transfer. The hydrodynamic properties of this regime are well documented, quantifying plug dimensions in relation to channel geometry, flow conditions, and fluid properties in experimental and numerical investigations.

In contrast, flow topology or internal plug circulation patterns, due to their complexity, are poorly understood. Circulation in plug flow is established as a result of shear forces as the continuous phase comes into contact with the channel wall and the plug interface [20]. This shear stress creates a velocity difference across the width of the plug, with internal circulations initiated in the plug and in the continuous phase slug to compensate [7]. The circulations consist of two central primary axisymmetric counter-rotating vortices with additional complex secondary vortices at the front and rear of the plug [15]. These complex vortices improve mass transfer by frequent renewal of the boundary layer material close to the interface and by reducing the diffusion lengths [7]. Additionally, they enhance heat transfer, with the recirculations creating efficient convective mixing, significantly increasing heat transfer compared to unidirectional Poiseuille flow [2]. To quantitatively describe the vortices, characteristics such as plug and slug velocity fields, which can be used to derive recirculation times, and the vortex characteristics are needed.

The primary central vortices within the plug are known to change with the fluid properties and flow constraints; however, investigations have yielded contrasting findings [11]. It is generally agreed that the normalized stagnation locations of the symmetric vortex pair remains relatively constant in the radial direction with increasing mixture velocity and decreasing plug length for $Ca < 0.001$ [21–24]. However, their axial locations can vary with the velocity of the mixture and the length of the plug [11]. For long plugs, increased wall shear leads to the circulation areas moving to the rear of the plug [22]. Afkhami *et al.* [23] found a similar behavior with an increase in the ratio of dispersed to continuous phase viscosities, λ , from 0.01 to 1. In contrast, Scheiff *et al.* [14] showed that the primary circulations in IL slugs surrounding a heptane and paraffin oil plug (characterized by $\lambda = 212.3$ and $\lambda = 3.2$), respectively, covered the entire plug length; no axial shift was observed in the location of the stagnation points with increasing viscosity ratio.

Despite the work on primary vortices, there are limited studies on the secondary vortices forming at the front and rear of the plugs because interactions with the curved interfaces are complex, and the small space they occupy makes measurements difficult. The formation and behavior of these secondary vortices has been found to depend on both the viscosity ratio, λ , and the capillary number, Ca . Hodges *et al.* [22] showed that for low viscosity ratios ($\lambda \ll 1$), secondary vortices

exist with stagnation points on the plug interface, one at the plug axial center (plug front and rear) and a second radially and axially displaced from the plug axis. As λ increases, these second stagnation points migrate along the interface towards the axial center points at the front and rear of the plug, where eventually they merge, leading to the disappearance of the secondary vortices at higher viscosity ratios. In contrast, Afkhami *et al.* [23] demonstrated that increasing λ from 0.01 to 1 increased the velocities of the secondary vortices, while increasing Ca to 0.27 led to the loss of the rear vortices and a decrease in size and velocity of the front vortices. Balestra *et al.* [15] further investigated this phenomenon numerically, quantifying secondary vortices as a function of both λ and the normalized film thickness, δ/R . They identified a critical normalized film thickness $\delta/R = 1 - 1/\sqrt{2} \approx 0.29$, below which secondary vortices exist because external recirculation patterns in the continuous phase are sufficiently strong to induce secondary recirculation inside the plugs regardless of λ , but above which they disappear depending on λ . The mechanism of secondary vortex loss followed the findings of Hodges *et al.* [22], with convergence of the stagnation points of the secondary vortices at the plug interface, as Ca and δ increase. Afkhami *et al.* [23] and Balestra *et al.* [15] found that the rear vortices reduce in size and velocity faster than the front vortices, leading to loss of the former before the latter with an increase in Ca . In contrast to these numerical investigations, Dore *et al.* [25] showed experimentally that for $Ca < 0.004$, no secondary vortices were observed, for $0.005 < Ca < 0.015$ vortices were detected at the rear and front of the plug, and for $0.02 < Ca < 0.06$ these vortices disappeared. Yao *et al.* [11] highlighted that the discrepancy between the theoretical and experimental results remains poorly understood and requires further investigation.

The intensity of a vortex can also be quantified using the normalized circulation time τ_0 [21], which is defined as the time it takes for an element of fluid to move from one end of the plug to the other, over the characteristic time T_s needed by the plug to move a distance equal to its own length. In a review by Yao *et al.* [11], the analytical, numerical, and experimental results of τ_0 were plotted against T_s highlighting a linear relationship (apart from the cases where $\lambda > 1$ and for sufficiently small Ca), indicating that shorter plugs/higher mixture velocities produce shorter mixing times inside them.

Understanding the vortex structure and intensity within the plugs in liquid-liquid flows is imperative for heat and mass transfer optimization; however, detailed data are lacking with clear discrepancies between numerical and experimental investigations and little specific detail regarding secondary vortex development. The studies of Balestra *et al.* [15] and Afkhami *et al.* [23] highlighted the critical capillary numbers at which secondary vortices are lost; however, experimental quantification has not yet been performed to validate these findings. Given the influence of the secondary vortices on mass transfer and the findings of Pheasey and Angeli [18] and Vereycken *et al.* [17], which highlighted reduced mass transfer when using an ionic liquid continuous phase at high capillary numbers, an experimental study of these phenomena is needed. Here, we investigate systematically plug flow hydrodynamics with changes in mixture velocity, covering the vortex topology and plug geometric characteristics (film thickness and plug length) experimentally and computationally. In particular, the evolution of the vortex is analyzed in terms of both geometry and vorticity, highlighting the contrasting findings regarding secondary vortex loss. We chose an ionic liquid as the continuous phase to understand the reduction in mass transfer rates at low viscosity ratios, λ , ($\lambda = \mu_d/\mu_c$) observed in previous studies [16–18]. The remainder of this paper is organized as follows. In Sec. II, we present details of the experimental setup and the laser-based microparticle image velocimetry system used to measure the plug characteristics and the velocity profiles. In this section, we also provide details of the numerical methodology for the plug flow. In Sec. III, we discuss the experimental and numerical results, focusing on the influence of the mixture velocity on the vortex characteristics within the plug; validation of the numerical predictions and comparisons with previous works are also provided. Finally, concluding remarks are made in Sec. IV.

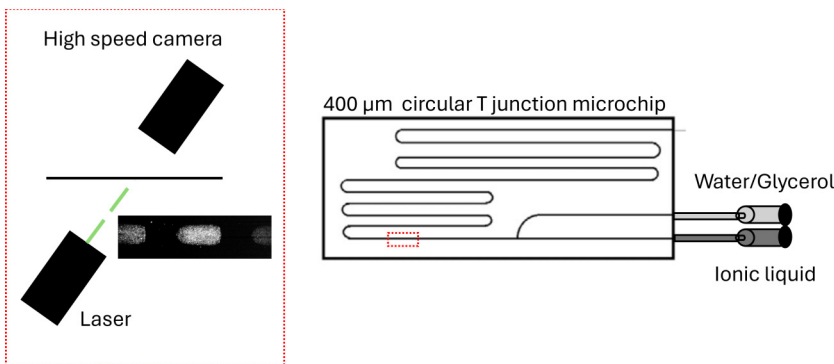


FIG. 1. Experimental setup of the high-speed camera (μ PIV) technique.

II. METHODOLOGY

A. Experimental methods and materials

1. Experimental setup

Experiments were performed in a circular T-junction glass microchannel (Dolomite). The channel has a diameter $D_c = 400 \mu\text{m}$ while the glass wall is coated with a silane monolayer to become hydrophobic. Two high-precision glass syringe pumps (KD Scientific) fed the two phases into the T junction. The aqueous phase entered perpendicularly to the main channel forming the dispersed phase and the organic phase entered tangentially, as shown in Fig. 1. We define the mixture velocity as $U_{\text{mix}} = (Q_1 + Q_2)/A$, where Q_1 and Q_2 are the inlet flow rates of the dispersed and continuous phases, respectively, and A is the cross-sectional area of the main channel. Four mixture velocities were investigated, $U_{\text{mix}} = 0.002, 0.01, 0.02$, and 0.03 m/s . The channel was prewetted with the continuous phase at each experimental run prior to the addition of the dispersed phase.

To match the refractive index of the aqueous (dispersed) and organic (continuous) phases and improve the imaging of the interface, the dispersed phase was a blend of 40%w/w glycerol and 60%w/w deionized water, this was premixed before use to form a homogeneous solution. The IL [C4mim][T₂fN] (Sigma Aldrich) was used as the continuous solvent phase. Due to its hygroscopic nature, the IL was presaturated with the dispersed phase by magnetically stirring equal quantities of each phase for 1 h and then separating them in a funnel prior to use.

The density and viscosity of the dispersed and continuous phases (after presaturation) were 1145 kg/m^3 and 0.0048 Pa s , and 1390 kg/m^3 and 0.041 Pa s , respectively, giving a viscosity ratio of $\lambda = 0.117$. The viscosity was measured using a digital DV-111 Ultra Rheometer (Brookfield) ($\pm 1\%$). The interfacial tension of 4.43 mN/m ($\pm 0.3 \text{ mN/m}$) was measured with the pendant drop method using a DSA100E drop shape analyzer (KRÜSS Scientific). The viscosity and interfacial tension measurements were taken at room temperature (22°C), which is the temperature at which all subsequent experiments were performed.

2. Plug velocity field

Microparticle image velocimetry (μ PIV) experiments were performed to obtain the velocity fields inside the plug. A continuous laser (Laserglow, 532 nm, 3000 mW) was used which allows observation in time of the plug displacement. The dispersed aqueous phase was seeded with $3.2 \mu\text{m}$ carboxylate-modified microspheres (FluoSpheres), with absorbance and emission wavelengths of 532 nm and 612 nm, respectively. The PIV image pairs were taken using a 16-bit high-speed camera (Phantom, v1310) with a resolution of 1260×800 pixels equipped with a Navitar $24\times$ magnification lens. The lens was mounted with a high-pass filter ($>565 \text{ nm}$) to eliminate reflections from the channel walls.

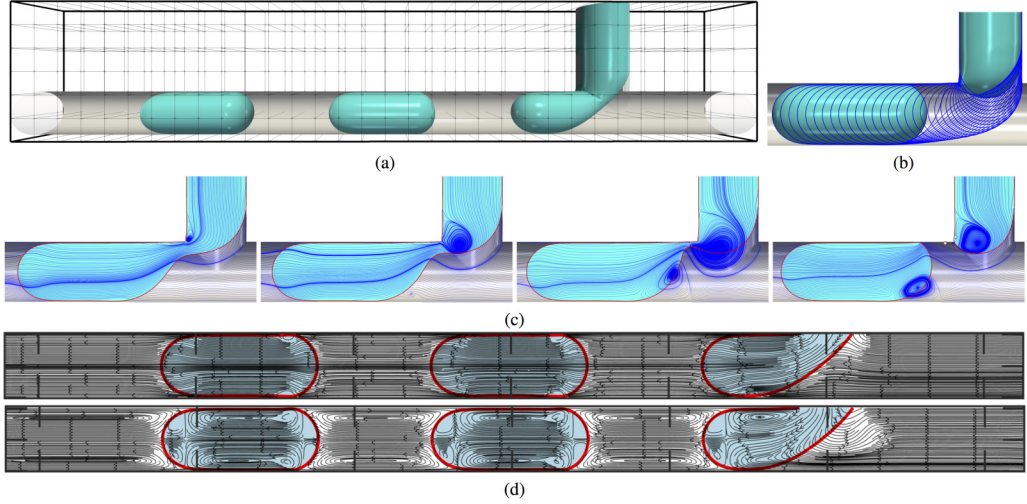


FIG. 2. (a) Three-dimensional numerical configuration highlighting the computational domain of $6.464 \times 0.404 \times 1.212 \text{ mm}^3$ and divided into $192 = 32 \times 1 \times 6$ subdomains. (b) Temporal evolution of the first plug formation with constant temporal snapshots of $25 \mu\text{s}$. (c) Vortical structure evolving during pinching mechanism. (d) Streamlines represented in a fixed laboratory frame (top panel) and its representation in moving plug-frame.

For each measurement, image pairs were acquired at a frequency of 2000 Hz on a plane covering the entire channel width ($r/D_c \in [-1, 1]$, with r being the radial direction). Measurements were recorded away from the T-junction inlet ($x/D_c = 10$, with x being the streamwise direction). All velocity fields were computed with the free PIV software CPIV-IMFT developed at the Institute of Multiphase Flows in Toulouse (IMFT) which has been used in our previous studies [26,27]. It is based on an FFT cross-correlation method with peak-locking reduction schemes and parallelization. In the processing, 16×16 pixel interrogation boxes and 50% overlap were used, resulting in a grid resolution of $14.5 \mu\text{m}$.

3. Plug characteristics

To obtain the geometric parameters of the plug including the film thickness and the plug length, at each flow condition 20 plugs were averaged. The plug shape was determined by the clear white line between the two phases in the grayscale inversed images. The film thickness was obtained by subtracting the width of the plug from the internal diameter of the channel, D_c . Where a film of nonuniform thickness was present, only the thickness of the locally uniformly thick region was measured [15]. The film thickness, δ , is nondimensionalized using the channel radius to obtain a dimensionless thickness, δ/R . The error is calculated as the standard deviation of the sample. The error ranges for the film thickness and plug length were 1.1% – 1.6% and 0.9% – 5.6%, respectively. The uncertainty in measuring the plug length and film thickness corresponds to 1 pixel, which is equal to $1.82 \mu\text{m}$.

B. Numerical studies

The computational approach used to predict the flow outcome involves numerically solving the Navier-Stokes equations in a three-dimensional (3D) domain of size $6.464 \times 0.404 \times 1.212 \text{ mm}^3$, which has been divided into $192 = 32 \times 1 \times 6$ subdomains, as shown in Fig. 2(a). The configuration setup of the T junction is constructed using a module for the fluid-structure interaction, which is based on an immersed boundary method, as used in Refs. [28–31]. A 3D global static distance

function, $\Phi(x, r, z)$, was generated, which is positive and negative in the fluid and solid regions, respectively. The shape of the T junction used in the simulation and represented in Fig. 2(a) corresponds to the zero isovalue of this distance function, $\Phi(x, r, z) = 0$.

The governing equations correspond to those of continuity and momentum transport, respectively, given by

$$\nabla \cdot \mathbf{u} = 0, \quad (1)$$

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \nabla \cdot \mu (\nabla \mathbf{u} + \nabla \mathbf{u}^T) + \mathbf{F}_{\text{int}}. \quad (2)$$

Here, $\mathbf{u}$ represents the velocity vector, t represents time, p denotes the pressure, and ρ and μ are the local density and viscosity, respectively; $\mathbf{F}_{\text{int}}$ corresponds to the local surface tension force at the interface:

$$\mathbf{F}_{\text{int}} = \int_{\mathcal{A}} \gamma \kappa \delta(\mathbf{x} - \mathbf{x}_p) d\mathcal{A}, \quad (3)$$

where γ stands for the interfacial tension, κ the curvature, and $\delta(\mathbf{x} - \mathbf{x}_p)$ the 3D Dirac function in which $\mathbf{x}$ is the position vector and $\mathbf{x}_p$ the parametrization of the interface $\mathcal{A}$.

The physical properties of both phases, including density and viscosity, are defined across the entire domain using a spatiotemporal function $\mathcal{H}(\mathbf{x}, t)$. This function is set to zero in the continuous phase and unity in the dispersed phase:

$$\begin{aligned} \rho(\mathbf{x}, t) &= \rho_c + (\rho_d - \rho_c) \mathcal{H}(\mathbf{x}, t), \\ \mu(\mathbf{x}, t) &= \mu_c + (\mu_d - \mu_c) \mathcal{H}(\mathbf{x}, t), \end{aligned} \quad (4)$$

where the subscripts c and d designate the continuous and dispersed phase quantities, respectively.

The simulations are initialized with a quiescent velocity field in the entire domain. Regarding the boundary conditions, a Hagen-Poiseuille laminar profile is imposed at the two inlets of the phases with a constant and equal average velocity, and associated with zero flux for the pressure, $\partial p / \partial n = 0$. Here, n refers to the normal direction to each inlet, which corresponds to the x and z directions for the continuous and dispersed inlet phases, respectively. As for the outlet boundary condition, a constant value for the pressure $p = 0$, associated with a zero-flux for the velocity field, is imposed. Moreover, no-slip and no-penetration conditions are imposed on the microchannel walls. The spatial resolution is fixed to 64 structured grid cells per channel diameter, and the time step is chosen at each iteration as the minimum value between the viscous and capillary time steps and that set by the Courant-Friedrichs-Lewy condition. Details of the numerical procedure can be found in the works of Kahouadji *et al.* [28] and Kalli *et al.* [31].

III. RESULTS AND DISCUSSION

The vortical structures and the velocity fields obtained from the high-speed μPIV experiments and CFD simulations are discussed below. The effect of increasing the mixture velocity on the evolution of the plug geometry, relative velocity, and vorticity of the primary (labeled V_2) and secondary vortices (labeled V_1, V_3) (see Fig. 3) within the plug frame of reference, is examined. In addition, the circulation time of the primary vortices V_2 will be quantified from the experimental results as a means of characterizing the mixing intensity [21].

A. Plug formation dynamics

In Fig. 2, we show the spatiotemporal evolution of the interface leading up to the formation of the plug. It is clearly seen that as the plug begins to develop and travel downstream following its exit from the T junction, its trailing end, which is connected to the dispersed phase liquid in the vertical section of the microchannel, begins to thin. This is accompanied by the formation of a vortex at

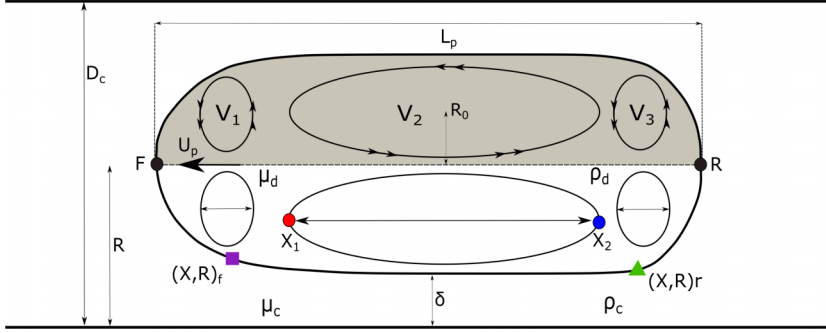


FIG. 3. Schematic illustration of the domain highlighting the main features of the plug, and defining the flow geometry, the plug length (L_p), the axial locations of its leading (F) and trailing (R) edges, and the film thickness (δ). In a reference frame moving with the plug, V_1 and V_3 represent the secondary pairs of counter-rotating vortices while the primary pair of vortices is labeled V_2 . The axial locations (X_1 , X_2) of the vertices of the primary counter-rotating vortex pair V_2 are also shown. R_0 denotes the radial distance of the stagnation point of V_2 from the microchannel axis, and $(X, R)_f$ and $(X, R)_r$ represent the interface stagnation points of the front and rear secondary vortices, respectively.

the corner joining the vertical and horizontal sections of the microchannel, which grows in size as the trailing edge thins further. It can also be seen that separatrices appear that separate regions of vortical activity within the plug. As the interface at the plug trailing end approaches rupture, a vortex forms in this region, which grows after rupture and plug formation. These phenomena have been observed experimentally by Chinaud *et al.* [32] using two complementary μ PIV techniques.

As the plug travels downstream, its speed reaches a constant value, U_p , and the vortical structures within the plug evolve, leading to the formation of two pairs of counter-rotating vortices at the leading and trailing ends of the plug, which we label V_1 and V_3 (see Fig. 3), respectively. These vortices are separated by a larger pair of counter-rotating vortices, labeled V_2 in Fig. 3, which occupy a large fraction of the plug. A discussion of these vortices, and other plug characteristics, will be provided below.

B. Plug speed, film thickness, and plug length

We examine the characteristics of a fully developed plug and their dependence on system parameters. In Fig. 4, we show the plug velocity U_p in relation to the mixture velocity, U_{mix} . There is good agreement between the experimental and computational results with a discrepancy in the range of 0–7%. Furthermore, the results are in good agreement with the experimental investigations of Li and Angeli [10] who used the same IL as in the present work and a 1M HNO_3 aqueous phase leading to $\lambda = 34.12$, and the experiment-based correlation of Kashid *et al.* [33] who studied a water-paraffin oil system. In all cases, the plug velocity is greater than the mixture velocity because of the surrounding film. Given this deviation, in all subsequent analyses, the capillary number is defined as $\text{Ca}_p = \mu_c U_p / \gamma$.

In Fig. 5, we show the dependence of the normalized thickness, δ/R , on the capillary number, in which we have included our experimental data with an error of 1.1–1.6% and numerical predictions with an error of 0%. It is clearly seen that the film thickness across the uniform region increases monotonically, but at a rate that decreases with increasing Ca_p . Close inspection of Fig. 5 reveals excellent agreement between the experimental data and the results of the numerical simulations, which improves for the larger Ca_p values examined; the discrepancy is 3% for $\text{Ca}_p \sim 0.15$. This inspires confidence in the reliability of the numerical procedure used to perform the computations.

The results are also compared with the predictions by Balestra *et al.* [15], who considered a plug in isolation in the channel compared with the combined inlet and downstream approach modeled

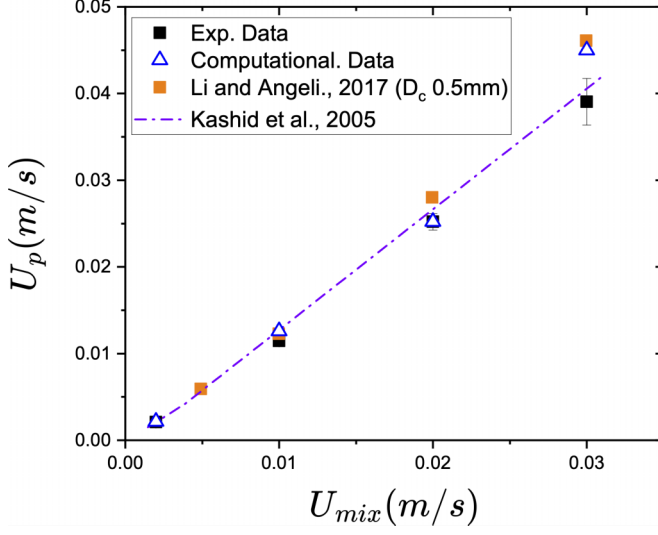


FIG. 4. Plug velocity, U_p , as a function of the mixture velocity U_{mix} from our μ PIV experiments and numerical simulations; also shown are the experimental results of Li and Angeli [10] and the experiment-based correlation of Kashid *et al.* [33].

in the present work. Balestra *et al.* [15] proposed the following relationship, which captures the dependence of δ/R on Ca_p and λ :

$$\frac{\delta}{R} = \frac{\bar{P}(\lambda)(3Ca_p)^{2/3}}{1 + \bar{P}(\lambda)\bar{Q}(\lambda)(3Ca_p)^{2/3}}, \quad (5)$$

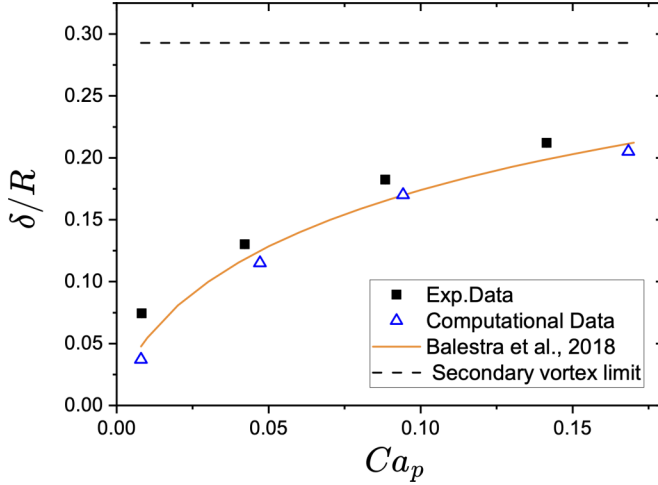


FIG. 5. Effect of the capillary number Ca_p on the film thickness δ normalized by the channel radius R : a comparison is shown of the results generated from μ PIV data, numerical simulations, and predictions obtained using Eq. (5) [15]. Also shown is the threshold δ/R beyond which no secondary vortices form obtained from the work of Balestra *et al.* [15].

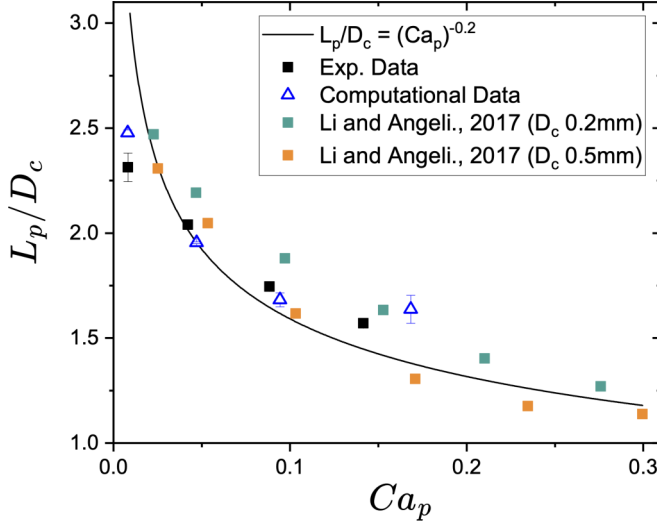


FIG. 6. Effect of the capillary number Ca_p on the plug length L_p normalized by the channel diameter D_c obtained from our μ PIV data and numerical simulations. Also shown are the experimental results of Li and Angeli [10] and our proposed correlation of $L_p/D_c = Ca_p^{-0.2}$.

where the functions $\bar{P}(\lambda)$ and $\bar{Q}(\lambda)$ are given by

$$\bar{P}(\lambda) = 0.322[1 + 2^{2/3} + (2^{2/3} - 1) \tanh[1.28 \log \lambda + c_2]], \quad (6)$$

$$\bar{Q}(\lambda) = \frac{a_3 \lambda^3 + a_2 \lambda^2 + a_1 \lambda + a_0}{\lambda^3 + b_2 \lambda^2 + b_1 \lambda + b_0}, \quad (7)$$

wherein $c_2 = -2.36$ in the axisymmetric case, and $a_0 = 2.21$, $a_1 = 111.25$, $a_2 = 33.84$, and $a_3 = 1.37$ and $b_0 = 0.89$, $b_1 = 44.86$, and $b_2 = 54.5$ are constants. For our viscosity ratio $\lambda = 0.117$ and $10^{-3} < Ca_p < 1$, $\bar{P}(\lambda) = 0.64$ and $\bar{Q}(\lambda) = 2.28$. Figure 5 demonstrates the agreement between the predictions obtained from Eq. (5) and the experimental data, with a 5% discrepancy. In addition, our numerical results are a nearly exact match with these predictions. The close agreement between the numerical results and those of Balestra *et al.* [15] provides validation that the hydrodynamics of the plug have reached a steady state and the plug has developed fully after pinching off near the channel inlet. Equation (5) highlights the importance of including the dependence of δ/R not only on Ca_p but also on λ in the case of two-phase liquid-liquid microchannel flows.

Figure 5 shows that the dimensionless film thickness tends toward a plateau at high capillary numbers. It is important to note that consideration here is only given to the uniform film thickness region; however, as shown by Li and Angeli [10] and Chinaud *et al.* [32], the film thickness exhibits nonuniformity at $Ca_p > 0.1$. In reality, the overall film thickness is greater than that in the uniform region and increases near the plug caps. With increasing Ca_p , the region of nonuniformity increases, and the predictions made using Eq. (5) deteriorate.

Balestra *et al.* [15] also predicted the existence of a critical film thickness, beyond which the secondary vortices are lost for low λ as shown in Fig. 5. With increasing film thickness, the influence of wall shear and viscous drag decreases, diminishing the extent of the secondary vortices. For the current system with $\lambda = 0.117$, the maximum dimensionless film thickness is $\delta/R \approx 0.2$, which is well below the critical thickness of $\delta/R \approx 0.29$ (see Fig. 5). Thus, there should be no loss of secondary circulation in the capillary number range studied here.

We have also examined the dependence of the plug length L_p on Ca_p . In Fig. 6, we show the variation of the normalized L_p/D_c with Ca_p obtained experimentally (error 0.9–5.6%) and

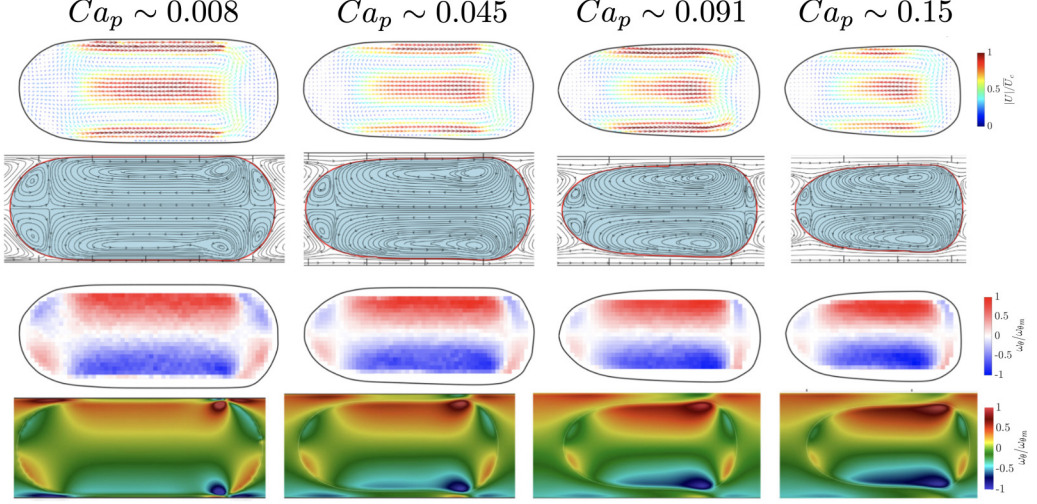


FIG. 7. Velocity vector fields and θ component of the vorticity ω_θ obtained from our μ PIV measurements, shown in the top and third rows, respectively, for $Ca_p \approx 0.008, 0.045, 0.091, 0.15$. Also shown in the second and bottom rows are the corresponding streamlines and vorticity fields obtained from our numerical simulations, respectively. For every plug and associated Ca_p , the velocity and ω_θ have been normalized by the maximum velocity at the plug center ($\bar{U}_c$) and by the maximum vorticity (ω_{θ_m}) of the vortex pairs within the plug, respectively.

numerically (error 0.2–4.1%). It is seen that there is excellent agreement between our experimental and numerical predictions with a deviation that ranges from 4–7%, which decreases with increasing Ca_p . We also see good agreement between our results and those of Li and Angeli [10]. Taken together, these results demonstrate that the normalized plug length decreases with Ca_p . Li and Angeli [10] proposed a fit of the form $L_p/D_c \sim Ca_p^{-0.273}$ for their data, which they collected in 0.2 and 0.5 mm channels and in a capillary number range of $0.025 < Ca_p < 0.3$. Given the goodness of fit (see Fig. 6) and the agreement with our data, we propose $L_p/D_c = Ca_p^{-0.2}$ as a valid correlation for the range $0 < Ca < 0.3$. Notably, the increase in the viscosity ratio between our experiments for which $\lambda = 0.117$ and those of Li and Angeli [10], with $\lambda = 34.12$, has a negligible effect on L_p/D_c . This is in contrast to the case of the film thickness as highlighted above. This observation provides justification for disregarding the dependence of L_p/D_c on λ in our proposed correlation, where the length of the plug depends only on Ca_p . Next, we turn our attention to the structure of the flow field inside the plugs.

C. Velocity and vorticity fields

The velocity fields inside the plugs were measured for Ca_p in the range 0.008–0.15 (which corresponds to a range of mixture velocities of 0.002–0.03 m/s). These fields capture the evolution of the primary and secondary vortices characterized by their geometry, velocity, vorticity, and circulation time.

1. Velocity and vorticity profiles

Figure 7 highlights the influence of varying Ca_p on the velocity field obtained from our μ PIV measurements (top row), the streamlines from our simulations (second row), and the vorticity field from our experimental (third row) and numerical predictions (fourth row). The results are shown in the (x, r) plane and in a frame of reference moving with the plugs. The experimental velocity

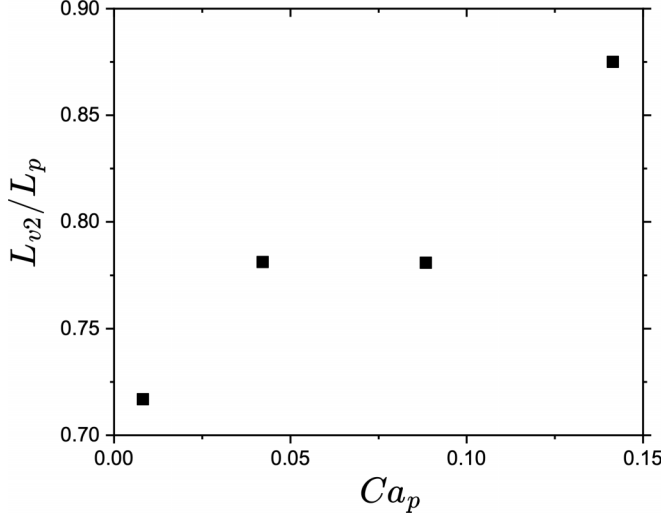


FIG. 8. Effect of the capillary number Ca_p on L_{v2} , the length of the primary vortex pair V_2 (as defined in Fig. 3), normalized by the plug length L_p , obtained from our μ PIV measurements.

profiles were scaled on the maximum velocity at the center of the plug. The vorticity is given by

$$\omega_\theta = \frac{1}{2} \left(\frac{\partial u_r}{\partial x} - \frac{\partial u_x}{\partial r} \right), \quad (8)$$

and normalized using the maximum vorticity of the three vortex pairs within the plug given by $\omega_\theta/\omega_{\theta m}$. It is seen clearly from the streamline plots in Fig. 7 that there are three counter-rotating pairs of vortices in the plug, which are particularly pronounced for low Ca_p : a primary vortex pair (labeled V_2 in Fig. 3) that occupies a large proportion of the plug, flanked by two smaller vortex pairs at the leading and trailing edges of the plug (labeled V_1 and V_3 in Fig. 3, respectively).

Inspection of the velocity field shows that the maximum velocity is seen at the plug center, and for $Ca_p \sim 0.008$ this high-velocity region occupies most of the plug length. With increasing Ca_p , this maximum shifts toward the rear of the plug as shown in Fig. 7 in agreement with the work of Hodges *et al.* [22]. Furthermore, as Ca_p increases, the plug becomes more bullet-shaped and the thickness of the film surrounding it becomes increasingly nonuniform with a local film thickness minimum at the rear of the plug. This, in turn, results in uneven shear stresses along the plug interface, leading to a perceived displacement of the central region of high velocity toward the plug rear altering the velocity distribution in the plug. For $Ca_p \sim 0.008$, the secondary vortices are a near mirror image of each other. As Ca_p increases, the vortices V_1 maintain their shape whereas the vortices V_3 flatten into a pancakelike geometry with their height exceeding their width. Both sets of secondary vortices diminish in size and all but vanish for $Ca_p \sim 0.15$.

2. Vortex dimensions

In Fig. 8, we plot the dependence on Ca_p of the axial length of the primary vortex pair V_2 , L_{v2} , normalized by the plug length L_p . It is seen that L_{v2}/L_p exhibits an overall increasing trend with Ca_p . This increase in the relative proportion occupied by the primary vortex pair comes at the expense of the smaller, leading, and trailing vortices, which retreat toward the front and rear of the plug, respectively. Additional evaluation of the dependence on Ca_p of the axial displacement of the vertex extremities, X_1 and X_2 of V_2 (defined in Fig. 3), from the plug front, F, is shown in Fig. 9. As can be seen, the rear vertex X_2 retreats toward the rear of the plug at a greater rate than the front vertex X_1 moves toward the front of the plug, where X_2 is near that of the plug rear at $Ca_p \sim 0.14$.

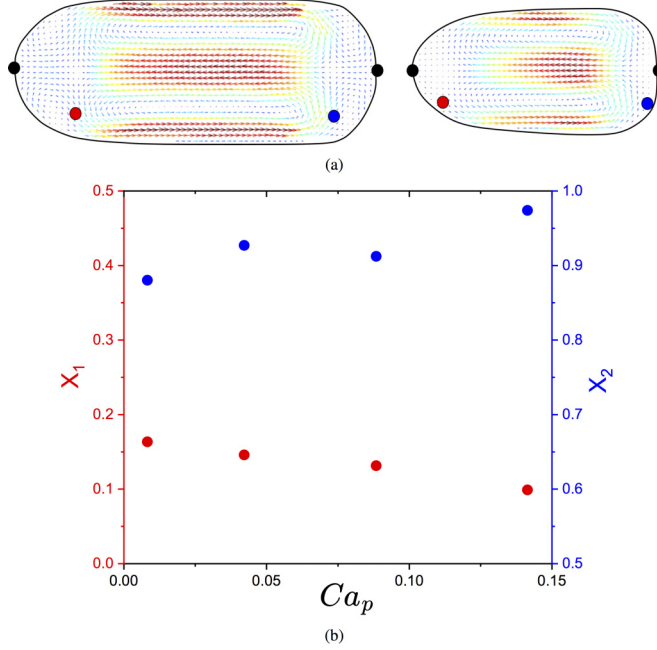


FIG. 9. (a) Illustration of the axial locations of the front and rear vertices (X_1 and X_2 , as defined in Fig. 3), of the vortex pair V_2 for $Ca_p = 0.008$ (left) and $Ca_p = 0.014$ (right) from our μ PIV measurements. (b) Variation in the vertex point location, X_1 and X_2 from the plug front with capillary number, Ca_p . $X=0$ is defined as the plug front, F, and $X=1$ is defined as the plug rear, R, as represented in Fig. 3.

(which coincides with the flattening of the rear vortex pair into a pancakelike shape at $Ca_p \sim 0.15$, as discussed above and shown in Fig. 7). This observation highlights the fact that the evolution of vortex pair V_2 has greater influence on vortex pair V_3 than it does on V_1 .

From Fig. 9, it is possible to extrapolate that the front and rear vortices will vanish, that is, $X_1 \rightarrow 0$ and $X_2 \rightarrow 1$ (taking $X=0$ as point F in Fig. 3) and the vertices of vortex pair V_2 will reach the interface, for $Ca_p \sim 0.35$ and $Ca_p \sim 0.19$, respectively. Using Eq. (5), we can estimate that this corresponds to $\delta/R = 0.32$ and $\delta/R = 0.26$, respectively. The loss of the rear vortex pair before that of the front one is in agreement with the findings of Afkhami *et al.* [23] and Balestra *et al.* [15]. The film thicknesses where the secondary vortices vanish agree reasonably well with the critical film thickness for the loss of secondary circulations given by Balestra *et al.* [15] for $\delta/R = 0.29$.

We now examine the influence of Ca_p on the locations of the interface stagnation points associated with the secondary vortex pairs using the results of our numerical simulations. For $Ca_p \sim 0.008$, the trailing vortex pair stagnation points, $(X, R)_r$, are located closer to the plug rear than those of the leading vortex pairs, $(X, R)_f$, are to the plug front, as shown in Fig. 10. With increasing Ca_p , $(X, R)_f$ and $(X, R)_r$ shift toward the front and rear of the plug, located at points F and R in Fig. 3, respectively, in line with the findings of Fig. 9.

We have also measured the influence of Ca_p on the dimensionless radial extent R_0/R (see its definition in Fig. 3) of the primary vortex V_2 . In Fig. 11, it is seen that R_0/R exhibits a weak dependence on Ca_p and remains approximately constant around $R_0/R \sim 0.45$. This is slightly below the predictions by Thulasidas *et al.* [21] of $R_0/R \sim 0.65$ based on the following relation Thulasidas *et al.* [21]:

$$R_0 = \frac{R}{\sqrt{2}} \sqrt{2 - \frac{U_p}{U_{\text{mix}}}}, \quad (9)$$

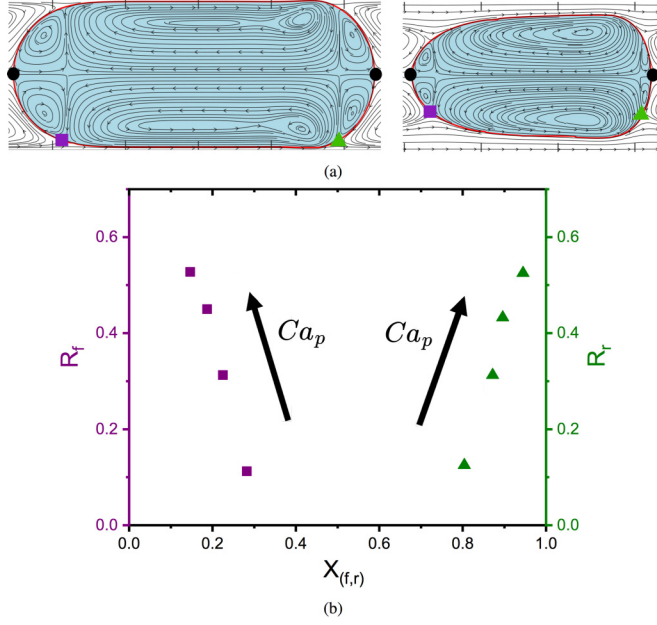


FIG. 10. (a) Illustration of the locations of the interface stagnation points $(X, R)_f$ and $(X, R)_r$, purple squares and green triangles, of the secondary vortex pairs V_1 and V_3 (as defined in Fig. 3) for $Ca_p = 0.008$ (left) and $Ca_p = 0.016$ (right) obtained from our numerical simulations; also shown by the black circles are the axial locations of the front and rear leading edges of the plug at the plug axial center. (b) Variation in the stagnation point location from the axial center plug front ($X=0$), F, with capillary number Ca_p . X is normalized by the plug length, L_p , and R is normalized by the channel radius, R . $Ca_p = 0.008, 0.047, 0.094, 0.168$.

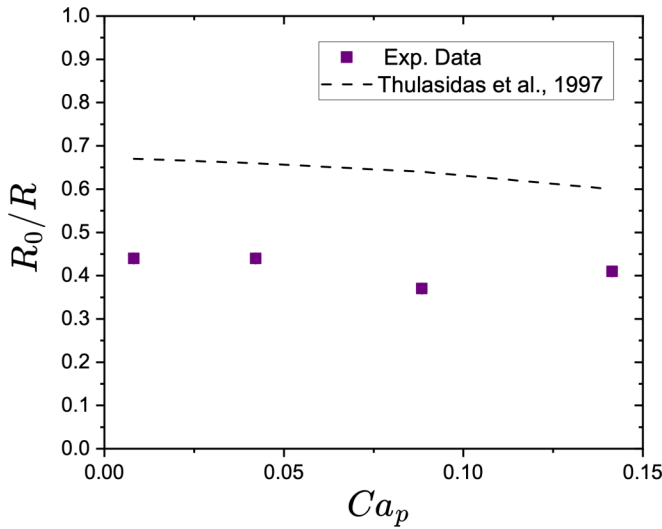


FIG. 11. Effect of the capillary number Ca_p on the radial location R_0 normalized by the channel radius R of the stagnation point of the primary central vortex V_2 (as defined in Fig. 3) from our μ PIV measurements. Also shown are the predictions obtained from Eq. (9) [21].

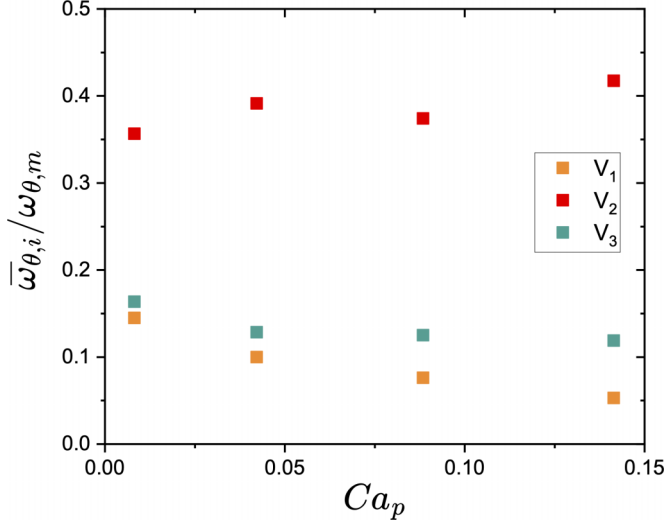


FIG. 12. Effect of the capillary number Ca_p on the normalized vorticity $\bar{\omega}_{\theta,i}/\omega_{\theta,m}$ of each vortex from our μ PIV experiments.

adapted to our case by replacing the bubble speed with the plug speed U_p and, similarly, the superficial velocity with U_{mix} , which is independent of the capillary number. The shift in R_0 toward the plug center in comparison to Thulasidas *et al.* [21] could be attributed to the change in fluids from gas-liquid to liquid-liquid and channel geometry from square to cylindrical. The weak dependence on Ca_p also implies a similar dependence on the film thickness, in agreement with previous findings [21,22,24]. Thus, the apparent plug squeezing and increased non-uniformity in film thickness has no significant effect on the radial geometry of the circulation.

3. Vortex intensity

In Fig. 12, we show the dependence of the normalized vorticity, obtained experimentally, of the three vortices discussed in the previous sections on the capillary number Ca_p . It can be seen that the vorticity associated with the primary vortex pair V_2 is significantly larger than that of the secondary vortex pairs, V_1 and V_3 . Whereas the vorticity of V_3 plateaus with increasing Ca_p following a decrease at low Ca_p values, that of V_2 and V_1 increases and decreases with Ca_p , respectively. These trends can be explained by the change in the shape of the plug as Ca_p increases, which is accompanied by a thickening (thinning) of the film near the front (rear) of the plug with a concomitant decrease (increase) of the shear in these regions. This, in turn, leads to a decrease (plateau) in the vorticity of the front (rear) vortices.

It is possible to extrapolate the curve associated with the normalized vorticity of the front secondary vortex V_1 to obtain an estimate of Ca_p for which this vortex vanishes; this gives a value of $Ca_p = 0.24$, which is lower than the $Ca_p = 0.35$ value previously estimated based on the vertex retreat of the primary vortex V_2 . The corresponding estimate for the normalized film thickness obtained from Eq. (5) is $\delta/R = 0.24$, which is in good agreement with the prediction of Balestra *et al.* [15] for the loss of the secondary vortices. Despite the decrease in its size with Ca_p , the rear vortex is predicted to be maintained at higher Ca_p because the locally thin film at the rear of the plug increases the shear in the region.

Based on the reduction in their size, it was shown in Fig. 9 that both secondary vortices were predicted to vanish with increasing Ca_p , the rear vortex pair being lost before the front one. In contrast, based on vorticity, it is predicted that with increasing Ca_p the front secondary vortex pair, V_1 , will be lost and the trailing pair, V_3 , will remain. This is in contrast to the geometric evaluations

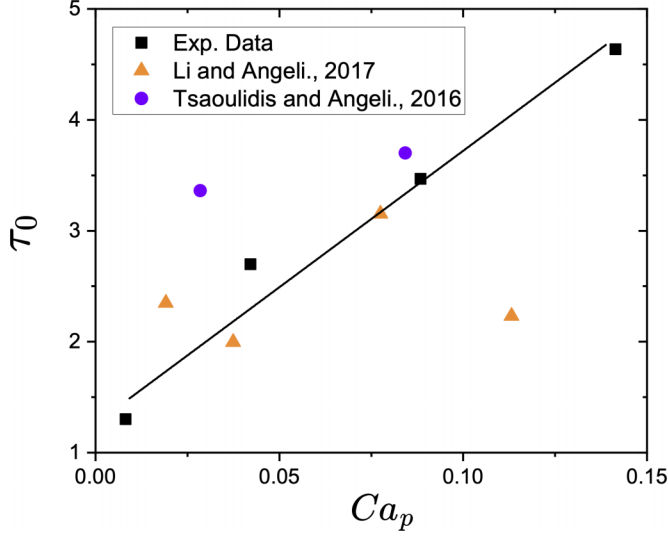


FIG. 13. Effect of the capillary number Ca_p on the normalized circulation time τ_0 of the primary central vortex V_2 obtained from our μ PIV experiments. Also included are the experimental results of Li and Angeli [10] and Tsaoulidis and Angeli [24]. A line of best fit of $\tau_0 = 1.36 + 23.79Ca_p$ is also shown.

and the investigations of Balestra *et al.* [15] and Afkhami *et al.* [23] who observed the opposite trend. Given that the plug is surrounded by a film of nonuniform thickness, there is a difference in shear between the leading and trailing regions of the plug, resulting in decreased (increased) vorticity at the plug front (rear). This is in line with the findings of Afkhami *et al.* [23] who highlighted the increase in wall shear at the plug rear but did not quantify its effect on the vorticity. The nonuniformity of the film thickness has a greater influence on vorticity in contrast to the geometry of the vortices resulting in a difference in which vortex is lost first. In addition, differences in the critical capillary number at which secondary vortices are lost may also be attributed to the differences in the domain of investigation between Balestra *et al.* [15], Afkhami *et al.* [23], and the current study.

4. Primary vortex circulation time

The dimensionless circulation time in the primary vortex V_2 , τ_0 , defined as the average time to displace material from one end of the plug to the other normalized by the plug travel time L_p/U_p , is calculated from the experimental results using the following relation:

$$\tau_0 = \frac{L_p R_0}{\int_0^{R_0} v(x, r) dr} \left(\frac{U_p}{L_p} \right). \quad (10)$$

Note that this circulation time is determined in the (x, r) plane [25]. In Fig. 13, we plot the variation of τ_0 with Ca_p . In this plot, we compare our experimental results with those of Tsaoulidis and Angeli [24] and Li and Angeli [10], demonstrating broad agreement in terms of the overall trend despite the fact that τ_0 in the previous works is associated with aqueous plugs moving in an IL continuous phase with $\lambda = 34.12$. From our experimental results, it is possible to show that τ_0 exhibits a linear dependence on Ca_p over the capillary number range studied. As Ca_p increases, the circulation time increases in line with previous findings that an increase in capillary number leads to a decrease in circulation velocities [15,22]. Additional investigations of the circulation time were performed analytically by Kececi *et al.* [34] and Abadie *et al.* [35] exhibiting an average discrepancy of 10% with our results. This discrepancy can be attributed to the fact that the works of Kececi *et al.* [34]

and Abadie *et al.* [35] were concerned with gas-liquid systems compared to the liquid-liquid system studied here.

IV. CONCLUSIONS

This paper draws on experimental and computational investigations of liquid-liquid plug flow in a microchannel. High-speed imaging and μ PIV experiments were used in conjunction with numerical simulations to quantify the film thickness, plug length, and velocity and vorticity fields. The experimental data were also used to validate the numerical predictions.

Experimental and numerical results for film thickness and plug length aligned increasingly with Ca_p to within 3% and 4%, respectively. Three counter-rotating vortex pairs were observed in a frame of reference moving with the steady translation speed of a fully developed plug. These pairs comprise a primary central vortex pair and two secondary pairs located near the front and rear of the plug. The size of the secondary vortices diminished with increasing capillary number, Ca_p . Furthermore, estimates of the critical capillary number and of the film thickness were obtained in which the secondary vortices would vanish. It was possible to estimate the loss of the vortices based on the reduction of both their size and vorticity. The reduction in vortex size showed that the rear vortex pair, V_3 , is lost before the front pair, V_1 , in agreement with Balestra *et al.* [15] and Afkhami *et al.* [23], at $Ca_p = 0.17$ computationally and $Ca_p = 0.19$ experimentally. The $Ca_p \sim 0.18$ (corresponding to the arithmetic mean of these capillary numbers) where the rear vortex pair is lost may explain the plateau in the mass transfer coefficient observed by Pheasey and Angeli [18] at $Ca_p \sim 0.17$. The front vortex pair was lost at $Ca_p = 0.3$ computationally and $Ca_p = 0.35$ experimentally. In contrast, based on vorticity, the rear vortex pair was maintained while the front one was lost at $Ca_p = 0.24$.

By analyzing the vorticity, we further quantify secondary vortices above that of Balestra *et al.* [15], Afkhami *et al.* [23] to highlight additional complexities to their diminishment. These results may explain the discrepancies seen in previous studies on the conditions and order of the loss of secondary vortices. However, in general, the size and vorticity of secondary vortices diminished with increased Ca_p , leading to a decrease in mass transfer rates, as observed by Van Stee *et al.* [16], Vereycken *et al.* [17], and Pheasey and Angeli [18]. Analysis of velocity and vorticity fields using a combination of μ PIV measurements and accurate numerical simulations provides valuable insight into the vortex evolution in plug flow with significant effects on mass transfer applications in small channels.

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The authors report no conflicts of interest.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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