



Flow hydrodynamics of immiscible liquids with low viscosity ratio in a rectangular microchannel with T-junction

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HIGHLIGHTS

- Hydrodynamics of liquid-liquid flow with low viscosity ratio have been studied experimentally.
- Scaling laws on plug velocity and length have been developed.
- Velocity profiles and circulation patterns in plugs have been obtained with PTV.
- New hydrodynamic features of the plug flow have been found.

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ABSTRACT

We present an experimental study of the liquid-liquid system with extremely low viscosity ratio (10^{-3}) in a T-shaped microchannel with $120 \times 120 \mu\text{m}$ inlets and a $240 \times 120 \mu\text{m}$ outlet channel. Six different flow patterns have been observed: plug, droplet, slug, throat-annular and parallel flow. A specific plug flow pattern was found where micron-sized droplets or even a jet breaks off from the rear meniscus of a plug. In addition to typical Taylor-shaped plugs the dumbbell-like plugs were observed. Flow visualization data were summarized in the flow pattern maps. Plug length and velocity were measured based on flow visualization results. It was found that the plug velocity can be fitted by power function rather than the linear function, which disagrees with experimental data at a low plug velocity. Front and tail plug surface curvature was scaled using dimensionless parameter, the flow rate ratio multiplied by Capillary number based on bulk velocity ($Q_d/Q_c \cdot Ca_{\text{bulk}}$). Instantaneous velocity vector fields inside water plugs were measured by means of PTV technique. Different flow structures were found and discussed. As a result, it is proposed to use the values of $Q_d/Q_c \cdot Ca_{\text{bulk}}$ for distinguishing different plug shapes and circulation patterns inside the plugs.

1. Introduction

Two-phase flows in microchannels, particularly liquid-liquid flows, have been studied extensively during last decades. The interest of the scientific community is caused by numerous applications and advantages of performing technological procedures in microchannels rather than in conventional macroscale devices. Microchannels are distinguished by extremely large surface to volume ratio that leads to intensification of heat and mass transfer. Flows of immiscible liquids in microchannels are proved to enhance different kinds of processes such as emulsion production [1], chemical reactions [2–4] and biological analysis [5]. In addition, microchannels are suitable for operations with tiny fluid volumes, thus decreasing a cost of tests with expensive reagents and improving safety of exothermic or hazardous chemical

reactions. Mass transfer coefficients are shown to be significantly higher for microchannels compared to conventional batch systems [6]. Moreover, Kashid et al. [7] in their work reported that numbering-up microchannels allows achieving the throughput of conventional reactors. In medicine and bio-applications, droplet microfluidics is considered to be a very promising technique that allows to analyze proteins [8], cells [9] and other biological objects. An increase of throughput by several orders of magnitude, compared to standard techniques, can be achieved in such devices [10].

Depending on two-phase system parameters, i.e. channel geometry, phase superficial velocities, phase viscosities, etc., different flow patterns may be obtained. Often it is vital to operate within one specified flow pattern or even under certain conditions, such as droplet size and velocity. For example, a droplet flow with narrow distribution of

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Nomenclature

Roman symbols

| | |
|--|---|
| d_h | hydraulic diameter, m |
| u | superficial velocity, m s^{-1} |
| v | flow velocity in plug, |
| U_{plug} | plug velocity, m s^{-1} |
| Q | flow rate, $\text{m}^3 \text{s}^{-1}$ |
| l | plug length, m |
| w | channel width, m |
| R | curvature radius, m |
| U_{bulk} | bulk velocity, m s^{-1} |
| $Ca = \mu u \sigma_i^{-1}$ | Capillary number |
| $Ca_{bulk} = \mu_c U_{bulk} \sigma_i^{-1}$ | Capillary number based on bulk velocity |
| $Re = \rho u d_h \mu^{-1}$ | Reynolds number |

$$Oh = \mu (\rho \sigma_i d_h)^{-1/2} \quad \text{Ohnesorge number}$$

$$We = u^2 d_h \rho \sigma_i^{-1} \quad \text{Weber number}$$

Greek symbols

| | |
|-------------------------|--|
| θ | contact angle |
| μ | dynamic viscosity, Pa s |
| ρ | density, kg m^{-3} |
| σ_s | surface tension, N m^{-1} |
| σ_i | interfacial tension, N m^{-1} |
| $\lambda = \mu_d/\mu_c$ | viscosity ratio |

Subscripts

| | |
|-----|------------------|
| c | continuous phase |
| d | dispersed phase |

droplet sizes may be used in microparticle production [11]. Chemical reactions can be performed either in the parallel flow [12,13] or in the plug one [14]. The plug flow was shown to provide the controlled residence time and to speed up the mass transfer due to internal circulation inside the plugs [6]. A variety of channel geometries, working fluids and, consequently, their properties along with a strong influence of adhesion and interfacial tension makes two-phase microfluidic flows difficult to predict. Therefore, many studies on microfluidics aim at characterizing the influence of system properties on the fluid flow in microchannels. There are two primary directions in this research field. The first one deals with the description of two-phase flow patterns, and the second one refers to the plug or Taylor flow regime properties.

Liquid-liquid flows in microchannels were studied in the context of flow patterns, for instance, in [15–18]. Typical flow patterns for liquid-liquid flows in microchannels are parallel, dispersed, annular, plug and slug regimes [15]. Flow regimes of liquid-liquid and gas-liquid flows demonstrate similarity to some extent. However, low viscosity and density ratio in the case of gas-liquid flows is responsible for some specific flow patterns such as churn flow, which was observed in gas-liquid cases [19]. In addition, the increased influence of a channel material and wettability properties was shown in the case of liquid-liquid flows. Salim et al. [20] studied a flow of water and mineral oil in microchannels made of glass and quartz, and reported a flow pattern dependence on the liquid, which entered the channel first. The same effect was observed by Tsaoulidis et al. [17] in the case of ionic liquid – water flows: both liquids can be either continuous or dispersed phase depending on which one was injected first.

To characterize two-phase flows, flow pattern maps based on superficial velocities or flow rates of phases are usually applied. There were a few attempts to generalize the flow pattern map for an arbitrary system, using experimental data and dimensional analysis. Waelchli and van Rohr developed the flow pattern map for gas-liquid flows based on dimensionless parameter $Re^{0.2} We^{0.4}$, which can be used as universal [21]. However, discrepancies were noted when compared with liquid-liquid flows. Kashid et al. studied liquid-liquid flows in channels with different inlet geometries and proposed the generalized map based on continuous phase Laplace number and dispersed phase dimensionless parameter $Re_d * d_h/\epsilon_d$, where d_h is the hydraulic diameter, Re_d and ϵ_d are the dispersed phase Reynolds number and the volume fraction, respectively [16]. This approach to the flow pattern description based on the dispersed phase inertia has given satisfactory results for some systems, even for gas liquid flows. Yagodnitsyna et al. conducted experiments with different sets of liquids and obtained dimensionless complex $We*Oh$ as the most appropriate generalization parameter for liquid-liquid flows, because it takes into account viscosity ratio of liquids [18].

The Taylor or plug flow was thoroughly studied in literature for

both liquid-liquid and gas-liquid flows. The most interesting parameters for applications are the plug length and velocity as well as the flow structure inside the plugs. Further, we dwell on the flow structure inside the plugs in the liquid-liquid case, which is more relevant to the current study. Studies of plug length, velocity and corresponding models and correlations may be found in the articles [22–25].

It was shown by Tice et al., that mixing in plugs can be strongly enhanced by the internal circulation, but the effect of fluid properties on a flow structure and circulation patterns should be studied in more details [26]. Kashid et al. numerically investigated a flow structure inside the plugs with verification by micro-PIV measurements. The flow structure with two counter rotating vortices was observed in their work [27]. To estimate the mixing efficiency, they utilized the non-dimensional circulation time, introduced earlier by Thulasidas et al. for liquid segments in gas-liquid flows [28]. The non-dimensional circulation time was shown to have a significant dependence on the presence of continuous phase film over the channel walls. The later measurements of the circulation time in [29] and [30] have shown contradictory results: the increase of circulation time with plug velocity in the first case, and the absence of clear dependence in the second one. However, it may be explained by some differences in the flow structure, because Dore et al. observed additional vortices at the front cap of a plug. Kinoshita et al. revealed a three-dimensional structure inside plugs using PIV technique along with confocal microscope [31]. Although the same flow structure with two counter rotating vortices prevailed, the authors pointed out at three-dimensional nature of the flow inside the plugs. Likewise, the three-dimensional flow structure and small additional vortices in the plug front cap were identified by Sarrazin et al., who numerically modeled the plug flow and verified it based on PIV data [32].

The viscosity ratio of liquids was proved to be a very important parameter not only for the two-phase flow pattern transitions, but also for the flow structure inside plugs. Hodges et al. predicted a possible appearance of additional vortices for viscosity ratio $\lambda < 1$ in the simplified model of a drop moving through a capillary [33]. Additionally, Lac et al. discovered a strong plug shape dependence on λ [34]. Jakiela et al. experimentally observed different scaling laws of plug velocity with capillary number (Ca) for different λ [35], and a sharp transition from the flow structure with two counter rotating vortices to the structure with additional vortices in the front and the rear plug caps that depends on λ as well [36]. Changes and transitions in a flow topology inside plugs depending on λ were also observed numerically and experimentally by other researchers [37].

There are not many works concerning this topic even though the influence of viscosity ratio on two-phase microflows is very strong. Most of existing studies on liquid-liquid flows deal with viscosity ratio in the range $0.01 < \lambda < 100$, and an important case, when

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