Experimental study of liquid-liquid plug flow in a T-shaped microchannel

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Abstract. Plug flow regime in immiscible liquid-liquid flow in a T-shaped microchannel is experimentally studied in the present work. For three different immiscible liquids plug length is measured using high-speed visualization of the flow. Micro-PIV technique is applied for instantaneous velocity field measurements in plugs. Velocity circulation in plugs is calculated for different bulk flow rates.

1. Introduction

Nowadays immiscible liquid-liquid flows in microchannels take place in a number of biological, chemical, medical and industrial processes [1,2]. Therefore the hydrodynamics investigation of immiscible liquid-liquid flows on a microscale is very important. A plug flow pattern is most suitable for conducting chemical reactions efficiently. In order to design and optimize liquid-liquid flow in microdevices the conditions required for plug flow formation, plug length, velocity and circulation inside plugs for liquids with different properties are needed to know.

Jovanovich et al. [3] measured a plug length depending on a plug velocity and a flow rate of dispersed and continuous phases for two different sets of immiscible liquids and two different channel diameters. The relationship between plug length and flow rate ratio was proposed by Garstecki et al. [4]. The corresponding model is valid for so called squeezing mode of plug formation when interfacial forces dominate shear stresses, i.e. Capillary number Ca << 1. This model was expanded by Xu et al. [5] who identified three different mechanisms of a plug formation process. There are the squeezing mode where a plug length is described by the correlation of Garstecki et al., the dripping mode where shear stresses dominate and a plug or droplet length is proportional to inversed Capillary number of a continuous phase, and the transient mode for which plug length dependence from flow rate ratio and Ca was also proposed in [5].

The circulation inside plugs is another important parameter required to determine a mixing efficiency. Kashid et al. [6] calculated circulation time for two models of plug flow: with film between a plug and a channel wall and without it. Velocity fields were obtained by means of CFD and validated by micro-PIV results in a narrow range of velocities. Dore et al. [7] investigated circulation inside water plugs in an ionic liquid for microchannels with circular cross section by micro-PIV technique. Three distinct circulation modes were found depending on plug velocity. Minimal
circulation time and maximal velocity inside plugs were measured for different Ca and ionic liquid volume fractions.

In the present paper the peculiarities of the plug flow in a T-shaped microchannel were studied for different liquid-liquid sets. In order to access circulation inside plugs micro-PIV technique is used for velocity measurements for one of liquid-liquid sets.

2. Experimental setup
In the present work three different liquid-liquid sets were used for plug flow visualization in a T-shaped microchannel, i.e. kerosene - water, paraffin oil – water and paraffin oil – castor oil. The liquid properties measured are represented in Table 1. The T-shaped microchannel used is made of SU-8. The inlet channels have 200x200 um cross-section, outlet channel has 400x200 um cross-section. The flow was organized such that two opposing streams of liquids used were flowing horizontally in outlet channel. The length of inlet and outlet channels is 11.5 and 22.5 mm respectively which is sufficient to get steady plug flow. The photo of experimental setup for flow visualization and micro-PIV measurements inside plugs is presented in figure 1. There are pco.1200 hs high speed camera and inverted Zeiss Axio Observer.Z1 microscope with 5x magnification lens used for flow visualization. Frame rate varied from 5 to 1000 Hz. Halogen lamp is used for flow illumination. Flow is organized by KDS Gemini 88 double syringe pump. Flow visualization is done in two different channel areas: T-zone and at the end of the channel, which is 56 channel hydraulic diameters far from T-zone.

| Physical properties | Kerosene | Water | Paraffin oil | Castor oil |
|---------------------|----------|-------|--------------|------------|
| Density, kg/m³      | 745      | 997   | 845          | 935        |
| Viscosity, mPa•s    | 0.820    | 0.894 | 110          | 650        |
| Interfacial tension, mN/m | 45     | 48    | 14           |

Table 1. Physical properties of liquids

For kerosene – water flow velocity field measurements are done inside water plugs by means of micro-PIV technique using POLIS PIV system. Water phase was seeded by 3 um fluorescent polystyrene particles. Flow was illuminated by double pulsed Nd:YAG laser. Particle images were captured using double exposure 4 Mpix CCD camera. Instantaneous velocity fields were calculated using iterative cross-correlation algorithm. The final spatial resolution of the measurements was 18 um for every velocity vector.

Figure 1. The photo of experimental setup for flow visualization and micro-PIV measurements.
3. Results

3.1. Plug length measurements

ImageJ software was used for plug length measurements. Uncertainty of measurements was about 5 pixels, which corresponds to 0.01 mm. The averaging over a number of plugs was done for every value of volumetric flow rate. In figure 2 a plot of the dimensionless plug length versus flow rate ratio for all three sets of liquids is shown. To change the flow rate ratio the flow rate of a dispersed phase was changed while the flow rate of a continuous phase was constant. The standard deviation of the measured plug length is presented by error bars in figure 2. One could see that plug length increases with the flow rate ratio increase but dependence for castor oil – paraffin oil set differs from one for kerosene – water and paraffin oil – water sets. Moreover, the results for castor oil – paraffin oil do match well with the relation given in Garstecki et al. [4] while for two other sets of liquids there is no good agreement, in spite the fact that the squeezing mode of plug formation takes place in all cases. This difference between results and the prediction probably caused by adhesion forces which affect the plug formation process [8].

![Figure 2](image)

**Figure 2.** Non-dimensional plug length versus the ratio of rate of the flow for different liquid-liquid sets. Solid line is a plug length according to the scaling law by Garstecki et al. [4].

3.2. Velocity fields and circulation inside plugs

Velocity field measurements in water plugs are done in the area of plug formation and at the end of the channel. Figure 3 illustrates instantaneous velocity fields in central plane of the channel in different phases of water plug formation. One can see that after contact line formation between the plug and the channel wall velocity inside the plug increases significantly although the plug is still connected with the inlet channel by the dispersed fluid neck. This phenomenon could explain higher values of the plug length for kerosene – water and paraffin oil – water liquid sets.
In figure 4 velocity fields inside water plug at the end of the channel are presented. The maximum velocity is at the center of the plug and it decreases towards channel walls. In figure 4 c the difference between instantaneous velocity field and plug velocity is shown. One can see circulation inside plug with different sign left and right from plug axis.

Based on measured velocity fields velocity circulation inside plugs was calculated for different flow rates. Velocity circulation was calculated using Green’s theorem by integrating vorticity over the surface bounded by closed contour shown in figure 4 c. In all cases the boundaries of integrating area were chosen nearby channel walls and channel center. Calculated circulation was normalized by integrating surface area. This allowed avoiding dependence of circulation on the length of integration surface. Circulation was calculated for two different contours shown in figure 4 c (black and red contours). The difference in velocity circulations for two contours did not exceed 10%.

Normalized velocity circulation values in different flow rates are presented in figure 5. When plug
velocity increase circulation growths too. It is obvious that circulation inside plugs defines intensity of convective mass transfer. In case of chemical reactions circulation is responsible for convective transfer of reagents. Thus circulation increase or, as follows from obtained result, plug velocity increase leads to decrease of reaction time. Nevertheless one should notice that reaction rate by microreactor unit length remains constant.

4. Conclusion
The plug flow in a T-shaped microchannel was studied in details. The experiments were carried out for three sets of immiscible liquids with different physical properties: kerosene – water, paraffin oil – water and castor oil – paraffin oil. The plug length was measured for all liquid-liquid sets. The plug length was compared with plug length scaling low proposed by Garstecki et al. [4]. It was found that for castor oil – paraffin oil set where adhesion forces for dispersed phase are negligible and plugs of paraffin oil do not wet channel walls the measured plug length matches well with the scaling law. But for kerosene – water and paraffin oil – water liquid sets there is mismatch between the measured plug length and the prediction given by scaling law. The discrepancy of the measured plug length with the model can be explained by existing of the contact line and an adhesion force which should be taken into account.

The plug formation process was studied by means of micro-PIV. The instantaneous velocity fields inside plugs were obtained for characteristic moments of plug formation in the T-junction. It is shown that after contact line formation between the plug and the channel wall velocity inside the plug increases significantly although the plug is still connected with the inlet channel by the dispersed fluid neck. This phenomenon could explain bigger values of the plug length for kerosene – water and paraffin oil – water liquid sets. The circulation inside the plugs was calculated from the velocity fields for different flow rates and shown to be linear with the plug velocity.

Acknowledgements
This work performed in the Kutateladze Institute of Thermophysics SB RAS by a grant from the Russian Science Foundation (project No 16-19-10519).

References
[1] Benavides J, Aguilar O, Lapizco-Encinas B H and Rito-Palomares M 2008 Extraction and Purification of Bioproducts and Nanoparticles using Aqueous Two-Phase Systems Strategies Chem. Eng. Technol. 31 pp 838–45
[2] Tran T M, Lan F, Thompson C S and Abate A R 2013 From tubes to drops: droplet-based microfluidics for ultrahigh-throughput biology J. Phys. D. Appl. Phys. 46 114004
[3] Jovanovic J, Zhou W, Rebrov E V, Nijhuis T A, Hessel V and Schouten J C 2011 Liquid-liquid slug flow: Hydrodynamics and pressure drop Chem. Eng. Sci. 66 pp 42–54
[4] Garstecki P, Fuerstman M J, Stone A and Whitesides G M 2006 Formation of droplets and bubbles in a microfluidic T-junction — scaling and mechanism of break-up 2006 Lab Chip 6 pp 437–46
[5] Xu J H, Li S W, Tan J and Luo G S 2008 Correlations of droplet formation in T-junction microfluidic devices: from squeezing to dripping Microfluid. Nanofluid. 5 pp 711–17
[6] Kashid M, Renken A and Kiwi-Minsker L 2011 Influence of flow regime on mass transfer in different types of microchannels Ind. Eng. Chem. Res. 50 pp 6906–14
[7] Dore V, Tsaoulidis D and Angeli P 2012 Mixing patterns in water plugs during water/ionic liquid segmented flow in microchannels Chem. Eng. Sci. 80 pp 334-41
[8] Yagodnitsyna A A, Kovalev A V and Bilsky A V 2016 Flow patterns of immiscible liquid-liquid flow in a rectangular microchannel with T-junction Chem. Eng. J. 303 pp 547-54