Experimental study of ionic liquid-water flow in T-shaped microchannels with different aspect ratios

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Abstract. Flow regimes of immiscible ionic liquid - water flow in T-shaped microchannels with 160 um hydraulic diameter and 1:2 and 1:4 aspect ratios are experimentally studied in the present work. Plug length and velocity were measured using high-speed visualization of the flow. Flow pattern maps were drawn for two channels. Parallel flow was shown to prevail for 1:4 aspect ratio channel in comparison to 1:2.

1. Introduction

In the recent years, one of the most promising immiscible liquid-liquid systems for industry applications is ionic liquid – water. Ionic liquids are salts with relatively low melting point (usually < 100°C), and many of them are liquids at room temperature. Unique properties of ionic liquids like resistance to high temperatures and radiation, low evaporation rate and vapor pressure make them very attractive for using in power plants and nuclear waste recycling [1-3]. The main limitation for ionic liquids is a high cost, thus development of efficient low-loss systems for handling liquid flow is essential. For this purpose promising microfluidic technology can be adopted. Microfluidic systems can strongly enhance heat and mass transfer because of high surface to volume ratio. In addition, using of microchannels instead of conventional devices reduces consumption of expensive chemicals or fluids [4,5].

However, there is a lack of fundamental knowledge about liquid – liquid flows on microscale and particularly about ionic liquid – water flows. To properly design and optimize microfluidic devices more data on hydrodynamics and fluid flows in microchannels is needed. Nowadays only a few research articles on immiscible liquid flows with ionic liquid as one of the phases are available in the literature.

Novak et al. performed a study of extraction reaction for ionic liquid based two-phase system in the case of parallel flow pattern when liquids occupy equal volumes [6]. A model for mass transfer in this particular case was proposed and compared with experimental results. Experimental research of flow structure inside water plugs during ionic liquid – water flow was done by Dore et al. [7]. Presence of two recirculation areas in plugs was shown and circulation time was calculated from velocity fields and plotted as function of plug velocity and length. Flow pattern maps for microchannels made of three different materials were obtained in [8]. Flow pattern and pressure drop were strongly depend on the liquid which first entered a channel. Influence of channel diameter and flow velocity on extraction...
process in plug flow regime was reported by Li et al. [9]. Extraction efficiency tends to decrease with increasing of flow velocity or channel diameter. As one can see from existing studies, hydrodynamic properties play important role in chemical reaction efficiency. But there is still not enough data about influence of channel surface wettability, fluids viscosities, channel geometry etc. on flow pattern map and properties of particular flow patterns such as plug flow.

In this work ionic liquid – water flow in T-shaped rectangular microchannels with different aspect ratios and equal hydraulic diameters was studied by means of high speed visualization. In order to determine influence of aspect ratio on liquid – liquid flow properties flow pattern maps for different channels were plotted and plug length and velocities were measured.

2. Experimental setup

In the present work 1-Butyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide ionic liquid and distilled water were used for flow regimes visualization in T-shaped microchannels. Liquid properties are noted in Table 1. Ionic liquid was held in contact with water long enough to rich a saturation limit before using. Corresponding properties in Table 1 are taken from work [8] for water saturated ionic liquid. The T-shaped microchannels used are made of SU-8. Two channels with different aspect ratios were used in the experiments. A, b and c dimensions are shown in figure 1 (right) were equal to 120 um, 240 um and 120 um for the first channel and 100 um, 400 um and 200 um for the second one, which corresponds to 1:2 and 1:4 aspect ratios, respectively. The length of inlet and outlet channels is 11.5 and 22.5 mm, respectively which is sufficient to get steady flow. The photo of experimental setup for flow visualization is presented in figure 1. For flow visualization pco.1200 hs high speed camera and inverted Zeiss Axio Observer.Z1 microscope with 5x magnification lens were used. Frame rate varied from 1 to 1000 Hz. Halogen lamp was used for flow illumination. Liquids were injected in the channel by KDS Gemini 88 double syringe pump. Flow visualization was 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.

Table 1. Physical properties of liquids

| Physical properties | Ionic liquid (water saturated) | Water |
|---------------------|-------------------------------|-------|
| Density, kg/m³      | 1420                          | 997   |
| Viscosity, mPa·s    | 41                            | 0.894 |
| Interfacial tension, mN/m | 12.3                      |       |

Figure 1. The photo (left) and the scheme (right) of experimental setup for flow visualization.
3. Results and discussion

3.1. Flow patterns

Typical flow patterns for two types of microchannels with 120x240 um and 100x400 um outlets are presented in figure 2. The water was a dispersed phase in this ionic liquid -water system inside SU8 channel because of wettability properties. It was found that flow patterns are independent from the phase which penetrates the channel first. Four basic flow patterns viz. plug, droplet, slug and parallel were observed for both microchannel sizes. In addition to those flow regimes there were throat-annular and rivulet flows for 120x240 um channel.

A plug flow corresponds to low superficial velocities, $U_{il} = 0.3 \text{ mm/s}$ and $U_{w} = 0.15 \text{ mm/s}$ for the case depicted in figure 2 a. Interfacial tension and adhesion forces dominate in this type of flow and tend water to form plugs with contact line on the channel wall. With increase of superficial velocities contact line is breaking and plugs are flowing surrounded by continuous phase film as presented in figure 2 b for $U_{il} = 5.8 \text{ mm/s}$, $U_{w} = 5.8 \text{ mm/s}$. The transition from plugs with contact line to non-contact line plugs is defined by dynamic contact angle dependence on contact line velocity. Usually only one type of plugs is observed in experiments [10], thus two different types for one set of liquids in this work indicate strong contact angle dependence from contact line velocity. Further increasing in superficial velocity of ionic liquid leads to droplet flow shown in figure 2 c for $U_{il} = 14.5 \text{ mm/s}$, $U_{w} = 2.9 \text{ mm/s}$. In this type of flow the water forms almost spherical drops that smaller than channel diameter. The mechanism of droplet formation is known in literature as the “dripping” mode in contrast to the “squeezing” mode inherent for the plug flow [11]. The slug flow was observed for high water to ionic liquid superficial velocities ratio, for instance $U_{il} = 0.06 \text{ mm/s}$ and $U_{w} = 115.7 \text{ mm/s}$ in figure 2 d. The parallel flow occurred when inertia force was strong enough to prevent plug formation. Typical pictures of this flow are represented in figure 2 e for $U_{il} = 14.5 \text{ mm/s}$ and $U_{w} = 290 \text{ mm/s}$.

Throat-annular and rivulet flows were found only for 120x240 um channel. Throat-annular pattern also known as liquid ring was mentioned before in gas - liquid [12] and ionic liquid – water [8] flows and looks like core-annular flow with waves propagating over the interface. This flow pattern was realized in very narrow area of phase superficial velocities values, in figure 2 f $U_{il} = 14.5 \text{ mm/s}$ and $U_{w} = 28.9 \text{ mm/s}$. In the rivulet flow a thin thread of ionic liquid flows upon one of channel walls. Rivulet flow is shown in figure 2 g for $U_{il} = 1.16 \text{ mm/s}$ and $U_{w} = 580 \text{ mm/s}$.

Figure 2. Typical flow patterns in 120x240 um (left column) and 100x400 um (right column) microchannels: a) plug flow, b) plug flow (no contact line), c) droplet, d) slug, e) parallel, f) throat-annular, g) rivulet.
3.2. Flow pattern maps
Based on visualization results flow pattern maps for both microchannel sizes were constructed. The dimensionless parameter proposed in [13] was used to plot flow pattern maps as diagrams presented in figure 3. It should be noted that almost every experimental point in the map for 120x240 μm microchannel has corresponding point obtained for same superficial velocity in the map for 100x400 μm microchannel. Consequently, dimensionless parameters such as Capillary number, Reynolds number and Weber number (We) written in terms of superficial velocity and hydraulic diameter have the same values in these couples of points. This fact allows us to specify aspect ratio influence on flow pattern maps keeping other parameters constant.

Physical significance of the using parameter Weber number multiplied by Ohnesorge number (Oh) is a ratio of forces that contribute to interface formation – capillary forces – to inertia forces and viscous dissipation acting in opposite direction. According to this definition and experimental results from [13] flow pattern maps are divided into several regions where different forces are prevail. There is a good match between experimental results and proposed regions borders. The plug flow points correspond to low WeOh values for both phases. Plug flow without contact line and droplet flow points hit the region with low WeOh of water and high WeOh of ionic liquid. There is parallel flow in the central region and the region of high values. Slug and rivulet points are fall in regions with high WeOh of water and low or medium WeOh values of ionic liquid respectively.

As one can note the main difference between flow pattern maps is that parallel flow in 100x400 μm channel (figure 3 b) takes more area than in 120x240 μm channel (figure 3 a). It spreads into plug and droplet regions in comparison to figure 3 a, and completely replaces rivulet flow. Probably, decrease in aspect ratio from 1:2 to 1:4 can enhance the role of viscous dissipation and make parallel flow more preferential. Therefore, we can conclude that channel with smaller aspect ratio is more appropriate for parallel flow applications such as various continuous chemical reactions.

![Figure 3](image)

3.3. Plug flow properties
For plug length and velocity measurements special software developed in the Kutateladze Institute of Thermophysics was used. Uncertainty of plug length measurements was about 2 pixels, which
corresponds to 4 um. Plug velocity was calculated using plug leading and trailing edge positions for different time steps. The averaging over a number of plugs was done for every value of superficial velocity. In figure 4 (left) a plot of plug velocity versus bulk velocity is shown for both microchannels. Standard deviation of plug velocity is shown by error bars with 3 times higher magnitude in order to bring more clarity. It was found that for both channels equation (1) fits the experimental plug velocity data well with coefficient of determination R-squared = 0.992.

\[ u_{plug} = 2.34 U_{bulk}^{1.087} \]  

(1)

The power fitting function corresponds to the dependence of dynamic contact angle on contact line velocity for ionic liquid-water system. Analysis of plug length for different flow rates showed that plug length increases with the flow rate ratio increase and decreases with bulk flow increase. Therefore in figure 4 (right) non-dimensional plug length divided by channel hydraulic diameter is presented as a function of the ratio of bulk velocity to flow rate ratio for both channels. Similar trend can be easily seen for both channels with different coefficients in fitting functions.

Figure 4. Left: plug velocity versus bulk velocity for 120x240 um and 100x400 um microchannels. Right: plug length as a function of the ratio of bulk velocity to flow rate ratio.

4. Conclusion

Experimental study of ionic liquid–water flow in T-shaped microchannels with different aspect ratios was done. The following flow regimes were obtained during flow visualization: plug, slug, parallel, throat-annular, rivulet and droplet flow. Two types of plugs, i.e. plugs with contact line and without one were observed which point out to the strong dependence of dynamic contact angle on contact line velocity. Flow maps were plotted for both microchannels in We*Oh numbers. It was shown that parallel flow in 100x400 um replaces rivulet and part of plug and droplet flows in contrast to 120x240 um channel. This fact is probably explained by aspect ratio difference and should be studied in detail more precisely. Plug length and velocity measurements were performed for a wide range of superficial velocities. For both microchannels plug length can be described as a function of bulk velocity very well and does not depend on the aspect ratio of the channels. Plug length can be fitted by power functions of bulk velocity and flow rate ratio with different coefficients for both channels.

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