International Journal of Multiphase Flow 54 (2013) 1–10

Contents lists available at SciVerse ScienceDirect

International Journal of Multiphase Flow

journal homepage: www.elsevier.com/locate/ijmulflow

Flow patterns and pressure drop of ionic liquid–water two-phase flows in microchannels

Dimitrios Tsaoulidis [a], Valentina Dore [a], Panagiota Angeli [a,*], Natalia V. Plechkova [b], Kenneth R. Seddon [b]

[a] Department of Chemical Engineering, University College London, Torrington Place, London WC1E 7JE, UK
[b] QUILL Research Centre, School of Chemistry and Chemical Engineering, The Queen's University of Belfast, Belfast BT9 5AG, UK

ARTICLE INFO

Article history:
Received 18 March 2012
Received in revised form 7 December 2012
Accepted 11 February 2013
Available online 19 February 2013

Keywords:
Micro-fluidics
Liquid–liquid
Pressure drop
Flow patterns
Ionic liquids

ABSTRACT

The two-phase flow of a hydrophobic ionic liquid and water was studied in capillaries made of three different materials (two types of Teflon, FEP and Tefzel, and glass) with sizes between 200 μm and 270 μm. The ionic liquid was 1-butyl-3-methylimidazolium bis{(trifluoromethyl)sulfonyl}amide, with density and viscosity of 1420 kg m^{-3} and 0.041 kg m^{-1} s^{-1}, respectively. Flow patterns and pressure drop were measured for two inlet configurations (T- and Y-junction), for total flow rates of 0.065–214.9 cm^3 h^{-1} and ionic liquid volume fractions from 0.05 to 0.8. The continuous phase in the glass capillary depended on the fluid that initially filled the channel. When water was introduced first, it became the continuous phase with the ionic liquid forming plugs or a mixture of plugs and drops within it. In the Teflon microchannels, the order that fluids were introduced did not affect the results and the ionic liquid was always the continuous phase. The main patterns observed were annular, plug, and drop flow. Pressure drop in the Teflon microchannels at a constant ionic liquid flow rate, was found to increase as the ionic liquid volume fraction decreased, and was always higher than the single phase ionic liquid value at the same flow rate as in the two-phase mixture. However, in the glass microchannel during plug flow with water as the continuous phase, pressure drop for a constant ionic liquid flow rate was always lower than the single phase ionic liquid value. A modified plug flow pressure drop model using a correlation for film thickness derived for the current fluids pair showed very good agreement with the experimental data.

1. Introduction

Operations in microchannels have emerged as an important area of research and have found numerous applications in (bio)chemical analysis and synthesis, intensified reactors, micropower generation, fuel cells and thermal management systems (Angeli and Gavriilidis, 2008). Many of these systems involve two phases, gas–liquid or liquid–liquid. Understanding of the flow characteristics and flow patterns, pressure drop and mass/heat transfer is essential for the design and the precise control of multiphase micro-devices. Although there are many studies concerning gas–liquid flows, only limited ones have reported on the flow behaviour of two immiscible liquids in small channels.

In liquid–liquid systems, depending on the fluid properties and the channel material, either phase can wet the channel wall, and for phases with similar wettabilities both phases can intermittently adhere to the wall, rendering ordered, stable and well-defined patterns more difficult to form than in gas–liquid flows (Wegmann and von Rohr, 2006). Controlling the hydrodynamics could decrease pressure drop, improve mass transfer, and facilitate product separation from the reaction mixture (Dessimoz et al., 2008). Two-phase liquid flows in large channels are mainly dominated by inertia forces and have been investigated using experimentation together with numerical and theoretical modelling (Angeli and Hewitt, 2000; Brauner and Maron, 1992). In the case of two-phase flow in microchannels, the interfacial tension and viscous forces are significant because of the small characteristic distances and the low Re numbers (Re < 2000), while gravity and inertia effects become negligible (Kreutzer et al., 2005; Foroughi and Kawaji, 2011).

Different patterns can be obtained in microchannels depending not only on operational conditions, such as flow rates, phase ratio and properties of the fluids (Lin and Tavlarides, 2009), but also on the geometry of the mixing zone and the channel, and the channel wall roughness and wettability (Jovanovic et al., 2011). A highly viscous oil–water system has been investigated by Salim et al. (2008) in microchannels made of quartz and glass and different flow configurations were observed depending on the fluid that was first injected into the test channel. The main flow patterns which have been observed are plug (or segmented), drop, annular and parallel flow. The formation of plug and parallel flow is controlled by the competition between viscous and surface tension forces. Plug flow in particular has been studied by many investigators (Kashid and

* Corresponding author. Tel.: +44 (0)20 7679 3832; fax: +44 (0)20 7383 2348.
E-mail address: p.angeli@ucl.ac.uk (P. Angeli).

0301-9322/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijmultiphaseflow.2013.02.002

Agar, 2007; Garstecki et al., 2006; Dessimoz et al., 2008), utilising both Y- and T-junctions as mixing zones. Moreover, Kashid and Agar (2007) observed that by having fluid mixing zones with different channel diameters, significant changes on the plug size and thus interfacial area were obtained. They found that capillary microreactors provided very large specific interfacial areas in comparison to other contactors, which enhanced mass transfer rates (for water–iodine–kerosene system, $k_L\alpha = 1311–9815 \times 10^{-4}\ s^{-1}$, where $k_L\alpha$ is the volumetric mass transfer coefficient). Kashid et al. (2005) reported that the intensity of the internal circulations in slugs, during plug flow, and therefore the overall mass transfer rate, depended on slug geometry. Plug flow has been used to enhance mass and heat transfer in a few reactions, such as nitration (Dummann et al., 2003). Annular and parallel flows were observed when the inertial forces dominated over the interfacial forces at We > 1 (see Table 1 for a definition of dimensionless numbers) which, however, were easily destabilised by changing flow rates and volumetric flow ratios (Zhao et al., 2006; Dessimoz et al., 2008). A general criterion has been suggested by Kashid and Kiwi-Minsker (2011) for an a priori flow pattern identification based on the parameter $Re_D d_{ch}/\varepsilon_D$, where Re_D is the Reynolds number of the dispersed phase, d_{ch} is the internal diameter of the microchannel, and ε_D is the volumetric flow fraction of the dispersed phase. According to the authors, flow patterns were classified into three regions based on the dominant forces. Segmented (plug) flow would occur in the surface tension dominated region for $Re_D d_{ch}/\varepsilon_D < 0.1$ m; annular flow would occur in the inertia dominated region for $Re_D d_{ch}/\varepsilon_D > 0.35$ m; transitional patterns (between plug and annular flow) would appear in the transition region for $0.1\ m < Re_D d_{ch}/\varepsilon_D < 0.35\ m$. The proposed criteria were applied to different microchannel geometries i.e. rectangular, trapezoidal, and concentric, and were found to be independent of contacting geometry and cross sectional geometry of the microchannels.

Knowledge of pressure drop during two-phase flow in microchannels is also essential for the design of energy efficient systems. There are, however, relatively few studies available on pressure drop in liquid–liquid microchannel flows (e.g. Kashid et al., 2007; Chakrabati et al., 2005; Jovanovic et al., 2011) compared to those available for gas–liquid flows (e.g. Kreutzer et al., 2005; Chen et al., 2002; Triplett et al., 1999; Kawahara et al., 2002). The models which have been developed for the plug liquid–liquid flow pattern in a microchannel are based on two basic contributions, i.e. the hydrodynamic pressure drop of the two individual phases and the pressure drop due to capillary phenomena. Plug flow is usually modelled as a series of unit cells, composed of a dispersed and a continuous phase (Kashid and Agar, 2007; Jovanovic et al., 2011).

One of the common applications involving the flow of two immiscible liquids is extraction, which is conventionally carried out using organic solvents. Recently, ionic liquids (ILs) have been suggested as alternatives to organic solvents because of their negligible volatility and flammability at common industrial conditions (Freemantle, 2010; Plechkova and Seddon, 2008), which reduce solvent loss and make them inherently safe and environmentally friendly. Ionic liquids are salts with low melting points (below 100 °C) composed exclusively of ions (Freemantle, 2010). Their properties can be tuned by the choice of the anion and/or the cation, allowing them to be optimised for a particular application (Seddon et al., 2000; Stark and Seddon, 2007). The hydrophobicity of the ionic liquids depends both on the alkyl chain length of the associated cation, and on the nature of the anion. Imidazolium ions, especially 1-alkyl-3-methylimidazolium ($[C_n mim]^+$), are often used as the cation. The bis{(trifluoromethyl)sulfonyl}amide anion, $[N(SO_2CF_3)_2]^-$ (also known as bistriflamide, and abbreviated to $[NTf_2]^-$), has become a popular anion choice for synthesising hydrophobic ionic liquids, that are chemically and thermally robust (Bônhote et al., 1996).

Although ionic liquids can find a wide variety of industrial applications (chemical industry, pharmaceuticals, nuclear reprocessing, etc.), there is a perception that the industrial use of ionic liquids is limited by their high costs bringing both real and psychological economic barriers to their wide use (Birdwell et al., 2006; Deetlefs and Seddon, 2006). One approach to circumvent these barriers is by operating within microchannels which require small solvent hold-up. The reduction in solvent volume is compensated by the high efficiencies achieved, because of the thin fluidic films formed in the confined spaces of the small channels, which can significantly reduce mass transfer resistances. Reactions involving ionic liquids have already been tested in microchannels, and yields much higher than in intensely mixed batch processes were found (Pohar et al., 2009). The flow patterns and associated pressure drop of two-phase flows involving ionic liquids are expected to be different to those of common organic solvents, because of their generally high viscosities and their higher densities compared to water.

The present work aims to investigate the flow patterns and the corresponding pressure drops during the flow of an ionic liquid, 1-butyl-3-methylimidazolium bistriflamide ($[C_4mim][NTf_2]$) and de-ionised water in microchannels made from materials that have different wetting characteristics. Moreover, a comparison with existing pressure drop models during plug flow was attempted and a modified model was suggested that agreed well with the experimental results. This is the first time that such a study has been presented.

Table 1
Dimensionless numbers for the characterisation of the two-phase flow.

Dimensionless numbers	Definition
Reynolds number	$\mathrm{Re}_i = \frac{\rho_i U_i \mathrm{ID}}{\mu_i} = \frac{\text{inertia forces}}{\text{viscous forces}}$
Capillary number	$\mathrm{Ca}_i = \frac{\mu_i U_i}{\gamma} = \frac{\text{viscous forces}}{\text{interfacial forces}}$
Weber number	$\mathrm{We}_i = \frac{\rho_i U_i^2 \mathrm{ID}}{\gamma} = \frac{\text{inertia forces}}{\text{interfacial forces}}$
Bond number	$\mathrm{Bo} = \frac{\mathrm{ID}^2 \Delta\rho g}{\gamma} = \frac{\text{buoyancy forces}}{\text{interfacial forces}}$

2. Experimental set-up and procedure

2.1. Experimental set-up

A schematic of the experimental set-up used for the two phase ionic liquid–water flow in microchannels is depicted in Fig. 1. It comprises of three main sections: the fluid delivery section in the mixing zone, the flow visualisation section, and the pressure drop measurement section. Two syringe-pumps (Aladdin-1000, WPI) fed the two liquids to the mixing zone. The pumps were calibrated and the maximum uncertainty of the flowrates was ±2%. Two inlet configurations (Y- and T-junction) were used for mixing the fluids, both made of PTFE with all the branches having the same ID (0.5 mm). In the T-junction, the two fluids entered the mixing zone perpendicularly with the water injected along the test channel axis. The angles of the inlets of the Y-junction were 120°. The test channels used in this work were made of two types of Teflon, PFA and Tefzel, with internal diameter (ID) of 220 μm and 270 μm respectively, and of borosilicate glass with an internal diameter of 200 μm. The length (L) of all test channels was 100 mm. The internal diameter of the microchannels was measured using a microscope.

The flow visualisation section comprised a high-speed camera (Phantom Miro 4) connected to a computer for data storage and a light source. Images were acquired at a distance 80 mm downstream the inlet. For the pressure drop measurements, a