Chemical Engineering Journal 328 (2017) 717–736

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Experimental and numerical hydrodynamic studies of ionic liquid-aqueous plug flow in small channels

Qi Li, Panagiota Angeli *

Department of Chemical Engineering, University College London, Torrington Place, London WC1E 7JE, UK

HIGHLIGHTS

- Detailed plug flow characteristics in liquid-liquid microchannel flows.
- Velocity profiles and circulation patterns in plugs were obtained with bright field PIV.
- CFD simulations predicted well experimental plug lengths.
- Variation of pressure along the microchannel during plug flow.

ARTICLE INFO

Article history:
Received 31 March 2017
Received in revised form 1 July 2017
Accepted 6 July 2017
Available online 8 July 2017

Keywords:
Liquid-liquid
Plug flow
PIV, CFD
Microchannels

ABSTRACT

The hydrodynamic characteristics of liquid-liquid plug flow were studied in microchannels with 0.2 and 0.5 mm ID both experimentally and numerically. For the experiments high speed imaging and bright field micro-Particle Image Velocimetry were used, while the numerical simulations were based on the volume-of-fluid (VOF) method. The two immiscible liquids were a 1 M HNO_3 aqueous solution which formed the dispersed plugs and a mixture of 0.2 M n-octyl(phenyl)-N,N-diisobutylcarbamoylmethyphosphine oxide (CMPO) and 1.2 M Tributylphosphate (TBP) in the ionic liquid 1-butyl-3-methylimidazolium bis[(trifluoromethyl)sulfonyl]amide ([C_4min][NTf_2]). The thickness of the film surrounding the plugs, and the plug velocity and length were measured and compared against literature correlations. For the cases studied (0.0224 < Ca < 0.299) it was observed that the liquid film was largely affected by the changes in the shape of the front cap of the plug. The plug length was affected by both the Capillary number and the ratio of the aqueous to ionic liquid phase flow rates while the plug volume depended on the channel diameter and the mixture velocity. The numerical simulations showed that, in agreement with the measurements, a parabolic velocity profile develops in the middle of the plugs while the circulation patterns in the plug are affected by the channel size. The pressure profile along the channel with a series of plugs and slugs was predicted numerically while the pressure drop agreed well with a correlation which included the dimensionless slug length and the ratio Ca/Re.

1. Introduction

Technical and environmental reasons are driving the development of small scale and miniaturised equipment within the frame of process intensification. The small scales reduce the molecular diffusion distances and increase the importance of surface/interfacial forces which bring new research challenges in the study of the hydrodynamic features and transport phenomena, particularly for multiphase systems [1–3]. Taylor (plug/segmented) flow in gas-liquid and liquid-liquid systems, where a continuous liquid phase separates elongated bubbles or drops (plugs) is a preferred pattern because the plug sizes are regular and can be controlled via the choice of inlet geometry. In addition, recirculation patterns establish in the liquid phases while the thin films that separate the bubbles/plugs from the channel wall enhance interphase and bubble (plug)-wall mass transfer [4–7]. Liquid-liquid microfluidic systems have found applications in solvent extraction [8–10], dispersion/emulsion formation [11], chemical synthesis/catalysis and biomedical analysis.

For the design of microfluidic devices operating in liquid-liquid plug flow, knowledge of hydrodynamic parameters such as film thickness, plug length/velocity and pressure drop is particularly important [12]. The significance of the film, which is formed due to viscous effects between the liquid plug and the wall is that the whole enclosed plug surface can participate in mass transfer.

* Corresponding author.
E-mail address: p.angeli@ucl.ac.uk (P. Angeli).

http://dx.doi.org/10.1016/j.cej.2017.07.037
1385-8947/

Nomenclature

Ca	Capillary number, $Ca=\frac{\mu_{IL}U_p}{\sigma}$, dimensionless
f	friction factor, dimensionless
ID	channel internal diameter, mm
L	length, m
L^*	dimensionless length, dimensionless
n	Refractive index, dimensionless
Q_{aq}	volumetric flow rate of aqueous phase, $ml \cdot h^{-1}$
Q_{IL}	volumetric flow rate of ionic liquid phase, $ml \cdot h^{-1}$
r	radius distance, m
r^0	location of recirculation centre, m
R	channel internal radius, mm
Re	Reynolds number, $Re = \frac{\rho U_{mix} ID}{\mu}$, dimensionless
R_p	plug width, m
U_f	film velocity, $m \cdot s^{-1}$
U_{mix}	mixture velocity, $m \cdot s^{-1}$
U_p	plug velocity, $m \cdot s^{-1}$
$U_{x,p}$	horizontal velocity profile in aqueous plug, $m \cdot s^{-1}$
$U_{x,f}$	horizontal velocity profile in continuous film, $m \cdot s^{-1}$
W	channel width, m
We	Weber number, $We = \frac{\rho U_{mix}^2 ID}{\sigma}$, dimensionless
ΔP	Pressure drop, Pa

Greek Symbol

α	fitting parameter, dimensionless
β	fitting parameter, dimensionless
σ	interfacial tension, $N \cdot m^{-1}$
δ	film thickness, m
ε	dispersed phase fraction, dimensionless
λ	viscosity ratio, dimensionless
μ	dynamic viscosity, cp
ρ	density, $kg \cdot m^{-3}$
φ	droplet effect parameter, $\varphi = \frac{\Delta P_p D}{\mu U_{mix}}$, dimensionless

Subscript

aq	aqueous phase
c	continuous phase
d	dispersed phase
IL	ionic liquid phase
max	maximum value
mix	mixture phase
p	plug
s	slug

The film thickness depends on a number of parameters, including interfacial tension, the viscosity ratio between the two phases, and the phase flowrates, and can vary along the length of the plug [13–16]. The length of the dispersed plugs and continuous phase slugs is also very important for heat and mass transfer processes and depends on many parameters such as fluid properties and superficial velocities, represented by dimensionless groups such as Re and Ca [6,17]. It has been shown, however, that the inlet configuration has a significant effect on the plug or slug size and attempts have been made to predict it by studying the plug formation mechanisms. In the squeezing regime (10^{-4}<Ca < 0.002, L_p/W > 2.5), when the shear forces dominate over the interfacial ones, the plug break-up process is mainly controlled by the pressure drop across the plug, and the plug size is determined solely by the volumetric flow rate ratio of the two immiscible fluids [18]. In the dripping regime (0.01 < Ca < 0.3, L_p/W < 1), the formation process is dominated by both the shear and the interfacial forces, and the size depends on the Capillary number only [19,20]. In the transition regime (0.002 < Ca < 0.01, 1 < L_p/W < 2.5), the plug size depends on all these forces [21].

Apart from the geometric characteristics of the plug, pressure drop in plug flows is important for the design of the pumps and for the choice of the flowrates while it can even affect the operation of the separation units at the channel outlet. In multiphase flows, the presence of the interfaces contributes an additional Laplace pressure term to the frictional component. The total pressure drop can then be calculated as a sum of the pressure drop due to the interfaces and of the frictional pressure drop of the various parts of the plug flow, i.e. film, slug and plug [12,22,23]. In these models it is assumed that the film is uniform along the plug. In simplified approaches, the overall pressure drop can also be calculated by adding in the continuous single phase pressure drop a term which accounts for the presence of the second phase [24–26].

The hydrodynamics of liquid-liquid plug flow in microchannels have been studied numerically with Computational Fluid Dynamics (CFD) simulations by a number of investigators. Different aspects of plug flow were considered, such as plug formation at the inlet [27,28], film thickness [12], velocity profiles and circulation inside the plugs [29], and pressure drop [23,30]. These studies have used either a fixed reference frame when plug formation is considered at the inlet or a moving one and periodic conditions when an isolated plug is investigated [31]. Kurup and Basu [32] compared simulated and experimental velocity profiles obtained with micro-Particle Image Velocimetry (μPIV) and found that the circulation patterns largely depend on the Capillary number. The simulated recirculation times, defined as the average time to displace material from one to the other end of the plug/slug, were found to be affected by a number of factors such as flow velocity, plug length, channel size, and viscosity ratio between the two phases [7,33,34].

To improve the understanding of liquid-liquid microchannel plug flows and to validate the numerical predictions detailed velocity measurements are needed. Non-intrusive optical techniques, such as micro-Particle Image Velocimetry, can extract multipoint information in velocity fields with high accuracy and spatio-temporal resolution. Few studies are available with μPIV in liquid-liquid microchannel flows which explore the plug formation process [35] and the local mixing inside the plugs or slugs [36,37]. In common μPIV approaches, laser light is used to volume illuminate the microchannels and the plane of measurement is defined by the focus of the camera; for temporally resolved measurements high speed lasers or a combination of continuous lasers with high speed cameras are required. A simpler approach, which does not require the use of lasers, is bright field (micro) PIV, where white light is used to illuminate the microchannel and the shadows of the tracing particles are captured by camera. For temporally resolved measurements high speed cameras can be used.

The improved heat and mass transfer rates in microscale units increase efficiencies and can reduce the overall solvent volumes required in two-phase processes such as liquid-liquid extractions. This enables novel and sometimes expensive solvents, such as ionic liquids, to be used economically. We have shown in previous work that the use of ionic liquids in small channel extraction units can enhance significantly mass transfer [10,38]. In particular, mass transfer coefficients as high as 0.05 s^{-1} were found during the extraction of Eu(III) from nitric acid solutions into an ionic liquid mixture (CMPO/TBP/[C_4min][NTf_2]) in microchannels operating in plug flow. In the current paper, the detailed hydrodynamics of the plug flow regime for the same liquid-liquid system are studied systematically using bright field μPIV. The velocity fields and