Chemical Engineering Science 174 (2017) 56–66

Contents lists available at ScienceDirect

Chemical Engineering Science

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

Liquid-liquid flow patterns and slug hydrodynamics in square microchannels of cross-shaped junctions

Zan Wu, Zhen Cao, Bengt Sundén *

Department of Energy Sciences, Lund University, Box 118, Lund SE-22100, Sweden

HIGHLIGHTS

- Flow regimes were mapped for square microchannels of cross-shaped junctions.
- Smaller channel size expands the tubing regime and narrows the dripping regime.
- Effects of physical properties on flow regime transitions were revealed.
- Slug length can be scaled with the Capillary number of the continuous phase.
- Slug velocity linearly depends on the average flow velocity of the mixture.

ARTICLE INFO

Article history:
Received 7 May 2017
Received in revised form 15 July 2017
Accepted 30 August 2017
Available online 1 September 2017

Keywords:
Microreactor
Liquid-liquid flow
Flow pattern map
Slug hydrodynamics
Dripping
Cross-shaped junction

ABSTRACT

Flow patterns for water-butanol, water-toluene, water-hexane, water-oil and water/glycerol (weight ratio 60:40) mixture-oil two-phase flows were visualized in the cross-shaped junctions of three square glass microchannels with hydraulic diameters of 200 μm, 400 μm and 600 μm. The aqueous phase is the continuous phase contacting the channel walls while the organic phase is the dispersed phase in the experiments. Three main flow pattern groups were observed, including the tubing/threading regime group, the dripping regime and the jetting regime. The flow regimes were mapped based on the Capillary number of the continuous phase and the Weber number of the dispersed phase. The flow rate ratio and the Capillary number of the dispersed phase were also employed to present flow patterns. The effects of hydraulic diameter of the square microchannels, flow rates, and physical properties, e.g., the interfacial tension and the viscosities of the aqueous and organic phases on flow pattern transitions were clarified. Besides, in the dripping regime, the dimensionless slug length can be scaled as a function of the Capillary number of the continuous phase for cross-shaped junctions. The slug velocity is linearly dependent on the average flow velocity in the dripping regime.

1. Introduction

The European Roadmap of Process Intensification (European Commission, 2014) identifies miniaturization as a promising direction for process intensification in terms of resource consumption, yield, selectivity and economic output. The principle behind intensification of thermal and chemical processes is the enhancement of transport processes by minimizing transport distances and residence time to reduce transport resistances, which leads to large increases in related transport capacities. Microreactors have been the subject of interest due to the high surface area to volume ratios leading to intensified heat and mass transfer rates (Kashid et al., 2011; Sattari-Najafabadi et al., 2017). Compared to conventional chemical reactors, the desired drop/slug size distribution in microreactors can be controlled precisely to tailor the transfer rates and output.

Liquid-liquid systems are of importance in applications like extraction, polymerization, nitration and pharmaceutical chemistry. There are relatively few studies focusing on liquid-liquid flow patterns and relevant hydrodynamics in the literature (e.g., Zhao et al., 2006; Kashid and Agar, 2007; Ghaini et al., 2011; Jovanović et al., 2011; Dore et al., 2012; Tsaoulidis et al., 2013; Wehking et al., 2014; Biswas et al., 2015; Chinaud et al., 2015; Plouffe et al., 2016; Tsaoulidis and Angeli, 2016). Generally, dispersed and continuous liquid phases flow into microfluidic devices from separate microchannels. The channels usually meet at a junction, which depends on the specific microfluidic device geometry, and the shape of the junction helps define the local flow fields that deform the two-fluid interface (Nunes et al., 2013). Four of the

---

* Corresponding author.
E-mail address: bengt.sunden@energy.lth.se (B. Sundén).

http://dx.doi.org/10.1016/j.ces.2017.08.032

Nomenclature

Ca	Capillary number
D	depth of the microchannel, μm
d_h	hydraulic diameter of the microchannel, μm
j	superficial velocity, m/s
L	slug length, length of the main channel, mm
Q	volumetric flow rate, ml/h
q	flow rate ratio of the organic phase to the continuous phase
t	elapsed time, s
u	velocity, m/s
w	width of the microchannel, μm
We	Weber number

Greek symbols

γ	interfacial tension, N/m
μ	dynamic viscosity, Pa s
ρ	density, kg/m^3

Subscript

ave	average
c	continuous phase, aqueous phase
d	dispersed phase, organic phase

most common microfluidic devices for droplet/slug production are the coaxial, flow-focusing, T-junction and Y-junction designs (Zhao and Middelberg, 2011). The cross-shaped junction, which consists of four microchannels with the same geometry and size that intersect at right angles, is a typical hydrodynamic flow-focusing geometry. When two immiscible phases meet at the cross-shaped junction, different flow patterns might appear depending on the junction and microchannel geometries, flow rates of the two phases and properties of the two phases. For liquid-liquid two-phase flow in microchannels, flow pattern transitions are mainly controlled by the relative magnitudes of the interfacial tension, the viscous shear force and the liquid inertia, while these forces depend on the channel geometry, flow rates and physical properties of the dispersed and continuous phases. The wetting characteristics of the liquids with respect to the microchannel walls in some way determine the dispersed phase and the continuous phase and the corresponding flow structures (Foroughi and Kawaji, 2011; Kawahara et al., 2002). Various flow patterns in microchannels of different inlet junctions have been observed and mapped in the literature by employing superficial velocities of the dispersed phase and the continuous phase or using dimensionless numbers such as Capillary numbers, e.g., Kashid and Agar (2007), Fu et al. (2012), Yagodnitsyna et al. (2016). For example, threading, jetting, dripping, tubing and viscous displacement have been observed for microfluidic cross-shaped junctions by Cubaud and Mason (2008). Flow pattern maps using dimensionless numbers are preferred to those employing dimensional parameters as the latter ones have limited generality.

Slug flow is a favorable flow regime for various heat and mass transfer processes owning to its high degree of control over slug size distribution and high interfacial surface-to-volume ratio. In slug flow, the dispersed phase does not wet the wall and is surrounded by a thin film of the continuous phase. The length of the slug is larger than the diameter, width or depth of the microchannel. Slug hydrodynamics such as slug length and slug velocity control the heat and mass transfer rates. Different scaling relations have been proposed to predict the slug length. Garstecki et al. (2006) and Bai et al. (2016) stated that the length of slugs produced in a T junction linearly depends on the flow rate ratio of the dispersed phase to the continuous phase in the squeezing regime. Liu and Zhang (2011) performed a three-dimensional lattice Boltzmann simulation for slug formation in microfluidic cross-shaped junctions and found that the slug length depends on both the flow rate ratio and the Capillary number of the continuous phase.

During the scale-up using a numbering-up approach, rectangular and square microchannels are preferable compared to circular microchannels in terms of easier integration of the former with a less volume. Besides, glass microchannels might be preferred to plastic microchannels because they wet most aqueous solutions. In general, flow hydrodynamics are pre-requisites to better characterization of heat and mass transport at microscale as flow hydrodynamics and relevant flow patterns are closely coupled with heat and mass transport processes. Thus, the present work aims to experimentally study the liquid-liquid two-phase dynamics and flow patterns at/near the cross-shaped junctions of square glass microchannels with hydraulic diameters of 200 μm, 400 μm and 600 μm by employing five different liquid-liquid systems.

2. Experiments

The test rig is shown in Fig. 1a. It mainly consists of two high-precision syringe pumps (New Era, NE-4000) equipped with 20 mL syringes for flow delivery, microchannels, and a stereo microscope (Motic, SMZ-171) with a camera (Olympus OM-D E-M1) for flow visualization. The reservoir is connected to the atmosphere. Three square microchannel sets (microchips), manufactured by Little Things Factory GmbH, were used as horizontal test sections. They were fabricated in borosilicate glass and sealed using a thermal bonding technique that resulted in excellent chemical resistance. The cross sectional dimensions of the three microchannels were 200 μm × 200 μm (MC-200), 400 μm × 400 μm (MC-400) and 600 μm × 600 μm (MC-600). The accuracy of the width and depth of the three microchannels is within ± 10 μm. The length of the tested microchannels is 105 mm. Fig. 1b presents the geometrical details of the microchannels.

Five immiscible liquid-liquid systems were tested in order to understand the effect of physical properties on flow patterns at the inlet junction and slug hydrodynamics in the main channel. The organic phases include 1-butanol (Acros Organics, ≥99.5%), toluene (Acros Organics, ≥99.99%), *n*-hexane (Acros Organics, ≥99.36%) and oil (Mogul Trafo CZ-A Paramo). The aqueous phases include de-ionized water and a mixture of 60.0 wt% of de-ionized water and 40.0 wt% of highly purified glycerol (MP Biomedicals, LLC). Although 1-butanol is soluble in water, it is assumed that the soluble nature of water-butanol flow may not affect the two-phase flow patterns at/near the inlet junctions due to a short contact time. During the tests, special care was taken to make sure that the aqueous phase is the continuous phase contacting the microchannel walls in the test section while the organic phase is the dispersed phase. Minute amount of phenol red was added to the aqueous phase to get clear images. The flow dynamics and patterns in and near the cross-shaped junctions were visualized for different liquid-liquid systems and flow rates. In the dripping flow pattern, the slug length and slug velocity were extracted and averaged from five snapshots and videos captured at a position 40 mm away from the junction for each operating condition. The