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Liquid–liquid segmented flows in polycarbonate microchannels with cross-sectional expansions ☆

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ABSTRACT

Liquid–liquid segmented flows in microchannels fabricated on polycarbonate test chips were investigated experimentally. Three different cross-sectional expansion ratios of 16, 4, and 2 from the injection to the test channels were used to observe the effect of different cross-sectional areas of the injection channel on the segmented flow regimes in the test channel. Flow regime maps and the transitions between flow regimes were determined for the different expansion ratios. The effect of the cross-sectional expansion ratio was determined by mutual comparison of these results. The measured length of the dispersed fluid segments was found to scale inversely with the carrier fluid volumetric flow ratio (β_C). A simplified geometric model for the volume of the dispersed and the carrier fluid plugs provided good estimates of the carrier plug length, which scaled inversely with $(1-\beta_C)$. Velocity and pressure drop measurements of the liquid–liquid segmented flows were carried out for all flow regimes, and the associated trends were correlated with changes in the flow topology. Most previous homogeneous and separated flow models based on the viscous frictional pressure drop underestimated the measured pressure drops. Including the contribution of the capillary pressure drop in addition to the frictional pressure drop improved the agreement with the measured data.

1. Introduction

Liquid–liquid segmented flows in microchannels have attracted considerable attention from the viewpoint of handling fluids in biochemical analytical devices because they potentially offer numerous advantages over single-phase flows. In biochemical reactions, heat and mass transfer are improved with increasing surface to volume ratios in miniaturized fluidic systems (Burns and Ramshaw, 2001). However, the reaction efficiency is limited by the slow diffusive mixing due to the laminar, low Reynolds number (Re) flow in microchannels. The parabolic velocity profile of a single-phase flow driven by pressure gradients under no-slip conditions (i.e., Poiseuille flow) causes the broad axial dispersion of target molecules along the channel. The molecules at different radial locations of the flow experience different residence times during the reaction process. Dispersed molecules close to the channel walls also tend to nonspecifically adsorb on the wall surface. The adsorption of target molecules on the walls reduces the reaction efficiency because of the associated loss of working molecules (Prakash et al., 2008). This could reduce the accuracy when identifying rare targets such as mutations. However, segmenting the single-phase flow by using another immiscible fluid induces spontaneous encapsulation of the fluid carrying the reactant molecules. The resulting recirculation in the encapsulated fluid plugs enhances mixing and prevents the molecules of interest from dispersing along the channels. In a gas–liquid two-phase flow, target molecules are distributed within the liquid plug and mixing is intensified by the recirculation. However, the dispersion and adsorption of molecules across neighboring liquid plugs, although less than in a single-phase flow, still occur through liquid films and corner flows between the gas bubble and channel walls (Muradoglu et al., 2007). For a liquid–liquid segmented flow, dispersed fluids encapsulated in spherical or elongated droplets serve as independent biochemical reactors owing to the fact that the target molecules are confined in the droplets (Günther et al., 2004; Song et al., 2003). The interaction of shear and surface tension forces at the interface between the dispersed and carrier fluids induces recirculation inside both the dispersed and the carrier fluid plugs. This, in turn, intensifies mixing (Song et al., 2003), which is normally reduced by the laminar characteristics of microchannel

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flows. If neighboring dispersed fluid droplets do not coalesce, the contents of the encapsulated dispersed fluids are preserved without cross-contamination. This is one of the reasons why a liquid–liquid segmented flow is a potential candidate for use in high throughput screening. Another important characteristic of such flows is the presence of a continuous thin film of the carrier fluid separating the dispersed fluid droplets from the channel wall, which prevents target molecules from being adsorbed on the channel wall. The adsorbed molecules are the principal sources of possible cross-contamination between successive dispersed plugs containing different reagent mixtures.

Owing to these advantages, liquid–liquid segmented flows have been employed in many practical applications including continuous segmented flow polymerase chain reaction (PCR) (Beer et al., 2008; Curcio and Roeraade, 2003; Mohr et al., 2007), enzyme encapsulation in lipid vesicles using microfluidic jetting (Jeanne et al., 2008), protein crystallization (Li et al., 2006), and fabrication of magnetic hydrogel micro-particles (Hwang et al., 2008). The manipulation of dispersed droplets and plugs (Adamson et al., 2006; Song et al., 2003; Yamada et al., 2008) has been studied in order to replace conventional micro-titer plates with a series of continuous separated droplets in microchannels for high throughput screening in drug discovery (Chen and Ismagilov, 2006). Each encapsulated droplet serves as an independent micro-reactor with increased mass and heat transfer and maintains its distinct contents without cross-contamination between neighboring droplets.

In order to design microfluidic bioanalytical systems using liquid–liquid segmented flows in an effective and predictable manner, it is necessary to understand the fundamental behavior of these microscale segmented flows. Thus, an experimental investigation of liquid–liquid segmented flows in polymer microfluidic channels was carried out. Using different cross-sectional expansion ratios from the injection part of the system to the test channel enabled an examination of the influence of this geometrical transition on the evolution of the segmented flow in the test channel. The study includes identification of segmented flow regimes and their maps, measurement of the length of the dispersed and carrier fluid segments, development of predictive geometrical models for these quantities, and measurement of the dispersed fluid segment velocities. Segmented flow pressure drop in microfluidic systems is higher compared to that in single-phase flow. Therefore it is necessary to be able to predict it with satisfactory accuracy. For this reason, the present study has also focused on the comparison of measured pressure drop with prediction of classical models reported in the literature.

2. Experimental

2.1. Test chip configurations

Fig. 1 shows a schematic of the base configuration of the microchannels fabricated on polycarbonate chips with three different expansion ratios (ER = 16, 4, and 2) of the cross-sectional area from the injection channel to the test channel. These microfluidic chips are referred to as ER16, ER4, and ER2, respectively, throughout this paper. The nominal dimensions of the injection and test channels are shown in Table 1. Carrier and dispersed fluids were introduced into each inlet and met at a cross-junction channel. Carrier fluid from the sides periodically pinched the elongated fluid thread from the central branch to form the dispersed phase. Shear and surface tension forces at the interface between the dispersed and carrier fluids generated a continuous string of mono-dispersed droplets into the central branch after the cross-junction (Thorsen et al., 2001; Utada et al., 2005). This segmented flow entered the test channel, which was deeper (ER16) and wider (ER16, ER4 and ER2) in a serpentine configuration along the test channel. In the ER4 chip, two 50 μm square channels were used to connect pressure transducers to pressure taps, which enabled measurement of the pressure drop across the test section. These were located at either end of the serpentine test channel (see Fig. 1). The serpentine configuration was chosen due to its compact footprint and because it allowed simultaneous observation of flow over almost the full test section channel length. The total channel length between the two pressure taps was ~120 mm. The U- and 90°-bends in the path of the test channel had a nominal centreline radius of 105 μm (±6%) for all chips. The cross-junction of the injection section and expansion area from the injection to the test channels had rounded corners as shown in Fig. 1a. This was a result of the 100 μm cutting tool radius (r) used in micromachining of the mold insert which produced the chips.

2.2. Microfabrication

All polycarbonate chips were replicated by hot embossing with micro-milled brass mold inserts. This thermoforming of polymer microfluidic chips enabled rapid production of multiple copies with stable production quality. A mold insert with the fluidic features was micro-machined on a 4.75″ diameter brass substrate (Alloy 353, McMaster-Carr, Atlanta, GA) using a KERN MMP 2522 (KERN Micro- und Feinwerktechnik GmbH, Eschenlohe, Germany) micro-milling machine. The milling bit radii (PMT, Janesville, WI) ranged from 25 μm to 100 μm. The brass mold insert was used to thermoform multiple chips in polycarbonate sheets (GE LEXAN® 9034, 2.36 mm thick, Modern Plastics, Bridgeport, CT) using a hot embossing machine (HEX02, JENOPTIK Mikrotechnik, Jena, Germany). After post-processing, including cutting and drilling holes at the inlet and outlet, chips were cleaned using the following sequence: (1) soaking in a weak (1%) mild detergent solution (5 min); (2) ultrasonic agitation (2 min) to remove the organic lubricant used during demolding; and (3) rinsed in deionized water (>120 min). After machining and cleaning, the chips were dried at 95 °C (20 min residence) in a temperature-controlled oven (HAFO 1602, VWR, West Chester, PA) and thermally bonded to 250 μm thick PC cover slips (CT301325, Goodfellow, Oakdale, PA) through a controlled process. A bonding jig designed to apply controlled, uniformly distributed clamping pressure was used. Thermal bonding was carried out at 160 °C (20 min residence) with two cooling steps, to 95 °C over 30 min and then to ambient over 150 min. This thermal bonding process induced changes in channel geometries from the nominal values because the temperature of the bonding process is set slightly above the glass transition temperature (Tg) of polycarbonate under the applied clamping pressure. The resulting channel width and depth after the thermal bonding were measured using a microscope equipped with a digital height gage (ME-50 IA DigiMicro, Nikon Instruments Inc. Melville, NY) and a digital readout (Quadra-Chek 2000, Metronics Inc., Bedford, NH). The measured channel dimensions are listed in Table 1 along with the design dimensions.

All microchips were made from the same PC stock and batch and processed identically using the same parameters. The unavoidable use of a detergent during the cleaning of the microchannel could introduce a surfactant contamination of the microchannel walls so, prior to running experiments, the channels were soaked and flushed for at least 1–2 days using deionised water after the thermal bonding was completed. Capillary tubes with a 175 μm inner diameter (1577 PEEK™ tube, Upchurch Scientific Inc., Oak Harbor, WA) were connected to the drilled holes at the inlet and outlet and sealed with epoxy (5minute® Epoxy, Devcon, Danvers, MA). The measured root mean square (RMS) surface roughness of the fabricated channel walls using an optical profiler (Wyko NT 3300, Veeco instruments Inc., Woodbury, NY) ranged from ~200 to