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Mixing and recirculation characteristics of gas–liquid Taylor flow in microreactors

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ABSTRACT

The effects of operating parameters (capillary and Reynolds numbers) and microchannel aspect ratio ($\alpha = w/h =$ [1; 2.5; 4]) on the recirculation characteristics of the liquid slug in gas–liquid Taylor flow in microchannels have been investigated using 3-dimensional VOF simulations. The results show a decrease in the recirculation volume in the slug and an increase in recirculation time with increasing capillary number, which is in good agreement with previous results obtained in circular and square geometries (Thulasidas et al., 1997). In addition, increasing the aspect ratio of the channel leads to a slight decrease in recirculating volumes but also a significant increase in recirculation times.

Keywords: Gas–liquid Taylor flow; Microchannel; Microreactor; Mixing; Recirculation; CFD; VOF

1. Introduction

Over the last decade, micro reaction technology has become of much interest to both academics and the process industries for the intensification of chemical processes. Taylor or slug flow is a commonly encountered flow regime for gas–liquid microchannel flows and has the advantage of providing high interfacial area and good liquid mixing in the liquid slug, thereby enhancing transport processes. These features of microreactors are particularly interesting for fast and highly exothermic gas–liquid reactions – amongst other applications – and allow an increase in reaction performance whilst working under safe operating conditions (Hessel et al., 2005; Kashid and Kiwi-Minsker, 2009). A number of studies have focused on the understanding of hydrodynamics as well as heat and mass transfer enhancement in these flows (see reviews Kreutzer et al., 2005; Shao et al., 2009; Gupta et al., 2010) but often independently. Indeed the transport efficiency appears to be closely related to the recirculation in the liquid phase, which depends on the operating conditions, fluid properties and reactor geometry.

A major feature of gas–liquid Taylor flow is the recirculation flow pattern generated in the liquid slug in the moving frame of reference as shown in Fig. 1 and detailed by Taylor (1961). The recirculating flow pattern is characterized by the position of the center of the circulation loop [x_0, y_0], and the position of the streamline separating the recirculation zone and the liquid film at the channel wall [x_1, y_1]. As the bubble velocity increases, both the loop center and the outer streamline of the recirculation zone move towards the center of the channel (Thulasidas et al., 1997). This leads to a reduction of the volume of the recirculating zone and an increase in the volume of liquid in the film region until complete bypass flow occurs at $U_B \geq U_{max}$. Studies on the recirculating flow in circular and square capillaries have been conducted previously (Taylor, 1961; Thulasidas et al., 1997), however little information on the characteristics of the recirculation zone in rectangular channels is available. Recently, it has been shown theoretically (Kececi et al., 2009) that the cross-sectional area occupied by the recirculation zone in the liquid slug is generally smaller for rectangular channels than for circular channels due to the increased film thickness in the channel corners.

The recirculation time in the liquid slug in Taylor flow is defined as the time required for an element of fluid to complete one revolution in the recirculating slug. This characteristic time is particularly relevant for transport processes

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Fig. 1 – Streamline pattern in the frame of reference moving with the bubble (Ca = 0.06 and Re = 16.8). The flow is directed from the left to the right. The positive part of the recirculating pattern of volume V_0 is delimited by the coordinates [x_0, y_0] in the channel width and depth; the negative part of the recirculating zone of volume $V_1 = V_{rc} - V_0$ is located between [x_0, y_0] and [x_1, y_1]; and the film region of volume $V_2 = V_{slug} - V_{rc}$ is located between [x_1, y_1] and the walls.

occurring in the system, such as mass transfer between the bubble and slug or wall and slug, and heat transfer with the channel wall. The rate of flow recirculation can be calculated via the surface integration of the relative velocity profile across the microchannel cross-section, similarly to what is done in conventional stirred tanks to calculate circulation induced by the mechanical impeller Jaworski et al. (1996). The recirculation flow rate through the microchannel can be divided into three parts (illustrated in Fig. 1):

- a positive flow rate, Q_0, in the main flow direction at central core of the microchannel occupying a volume V_0 with a cross-sectional area A_0,
- a negative flow rate, Q_1, with a volume V_1 and a cross-sectional area A_1 that corresponds to the recirculating liquid in the slug,
- a negative flow rate, Q_2 of volume V_2 in area A_2, that is close to the channel wall and contributes to axial mixing between slugs instead of radial mixing within the slug.

The recirculation time is then defined as $t_{rc} = V_{rc}/Q_{rc}$, where the recirculating volume V_{rc} corresponds to the volume of liquid within the limit of the separating streamline and Q_{rc} is the recirculation flow rate equal to Q_0 (and $|Q_1|$). t_{rc} can be made non dimensional by dividing it by the time taken for the bubble to travel a distance equal to the slug length: $\tau_{rc} = t_{rc}/(L_S/U_B)$.

In our previous work we have explored the effects of fluid properties, operating conditions and microchannel geometry on the size of Taylor bubbles (Abadie et al., 2012) and the flow patterns in the liquid slug using micro Particle Image Velocimetry ($\mu - PIV$) (Zaloha et al., 2012). The objective of this work is to explore the effects of operating parameters (capillary, Ca, and Reynolds numbers, Re) and microchannel aspect ratio ($\alpha = w/h = [1; 2.5; 4]$) on the mixing and recirculation characteristics of the liquid slug in gas–liquid Taylor flow in microchannels. To do this, 3-dimensional VOF simulations of gas–liquid Taylor flow in microchannels have been performed. Using an approach that is analogous to the determination of circulation rate in stirred tanks, the recirculation rate in the liquid slug, as well as the size of the recirculating zone have been evaluated from the 3-dimensional numerical data. An attempt has been made to relate these characteristics of the recirculating liquid slug to the enhanced transport phenomena observed in Taylor flow in microreactors. Finally, recommendations on the design and operation of microreactors employing Taylor flow are given.

2. Methodology

2.1. *Theoretical developed velocity profile in rectangular capillaries*

In the limit of infinite slug length, the analytical solution of the velocity profile in a cross-section of a rectangular capillary can be derived. The corresponding velocity profile in a rectangular capillary with a cross-section $2w \times 2h$ is given by Eqs. (1) and (2). This theoretical velocity profile has been used to evaluate the effects of aspect ratio and dimensionless bubble velocity on characteristic parameters of recirculation motion in gas–liquid Taylor flow: recirculating volumes and recirculation times. Under the assumption that the slug is long when compared with development lengths of velocity profile at the rear and the nose caps of the bubbles, the recirculation volume can be approximated by the cross-sectional area of the recirculating zone multiplied by the slug length: $V_{0,1} \sim A_{0,1} \times L_S$.

$$u(x, y) = -\frac{16c_1w^2}{\pi^3}\sum_{n=1,3,\ldots}^{\infty}\frac{-1^{(n-1)/2}}{n^3}\left[1-\frac{\cosh(n\pi y/2w)}{\cosh(n\pi h/2w)}\right]\cos\left(\frac{n\pi x}{2w}\right) \quad (1)$$

$$U_{TP} = -\frac{c_1w^2}{3}\left[1-\frac{192}{\pi^5}\frac{w}{h}\sum_{n=1,3,\ldots}^{\infty}\frac{1}{n^5}\tanh\left(\frac{n\pi h}{2w}\right)\right] \quad (2)$$

Numerical integration of the profiles (1) and (2) for aspect ratios $\alpha = [1 ; 2.5 ; 4]$ have been performed on fine uniform grids of 400 × 400, 1000 × 400 and 1600 × 400, respectively, to calculate the area A_0 where the velocity in the moving coordinate system is positive. Following this, an iterative integration of A_1 (cross-section of the negative part of the recirculating liquid) has been performed until the flow rates in the positive and negative parts of the recirculating zone are balanced. Integration of these profiles allows the prediction of recirculating volumes and recirculation times as a function of the aspect ratio and the dimensionless velocity $W = (U_B - U_{TP})/U_B$ assuming long liquid slugs.

2.2. *Numerical simulations*

The numerical code used for this study is the *JADIM* code, which has been developed to simulate dispersed two-phase flows and used to simulate various multiphase flows systems (Bonometti and Magnaudet, 2007; Dupont and Legendre, 2010; Sarrazin et al., 2006a,b; Abadie et al., 2012). The interface capturing technique implemented in this code is the Volume of Fluid method (VOF), which consists of a Eulerian description of each phase on a fixed grid. Under the assumptions that (i) the fluids are Newtonian and incompressible, (ii) there is no mass