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## Gas hold-up and liquid film thickness in Taylor flow in rectangular microchannels

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### Abstract

The gas hold-up in nitrogen/water Taylor flows in a glass microchannel of rectangular cross-section  $(100 \,\mu\text{m} \times 50 \,\mu\text{m})$  was shown to follow the Armand correlation. The validity of the Armand correlation implies that the liquid film thickness is not a function of the bubble velocity, which was varied between 0.24 and 7.12 m/s. Images of the Taylor flow were captured at a rate of 10,000 frames per second and were used to obtain the bubble and liquid slug lengths, the bubble velocity, and the number of bubbles formed per unit of time. A mass balance-based model was developed for Taylor flow with negligible liquid film velocities. The model describes the gas hold-up as a function of the liquid film thickness, the bubble and liquid slug lengths, the liquid superficial velocity, and the bubble formation frequency. © 2007 Elsevier B.V. All rights reserved.

Keywords: Taylor flow; Gas hold-up; Microchannel; Liquid film

### 1. Introduction

Taylor flow is the main flow regime of interest for performing gas/liquid/solid reactions in small channels (diameter <1 mm). It consists of sequences of a gas bubble and a liquid slug. The length of the gas bubbles is larger than the channel diameter and a thin liquid film separates the gas bubbles from the channel walls. The liquid film ensures a short diffusion path length for the gas phase diffusing through the film to the channel wall, where the catalyst is often located. The liquid in the slugs forms circulation cells when the capillary number ( $Ca = \mu u_b / \sigma$ ) is smaller than 0.5 [1,2]. The circulation patterns within the liquid slugs improve radial mass transfer in the liquid as compared to laminar flow [3]. The thin liquid film and the liquid circulation cells make Taylor flow a suitable flow regime for three-phase reactions where mass transfer to the wall is of influence on the reaction rate.

The thickness of the liquid film and the liquid velocity therein are key parameters, not only for mass transfer, but also for describing the hydrodynamics of Taylor flow. The gas hold-up is an important parameter in reactor design since it determines the mean residence times of the phases in the reactor and is related to the thickness of the liquid film. Due to the presence of the liq-

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uid film, the gas bubbles move through a smaller cross-sectional area than the combined gas and liquid flows. Continuity then requires that the velocity of the gas bubbles is larger than the total superficial velocity in the channel. Because of this, the gas hold-up differs from the flow quality, which is defined as the volumetric fraction of gas in the feed stream. The relation between film thickness and gas hold-up also depends on the flow rate of the liquid in the film.

Bretherton [4] showed that the film thickness is a function of the capillary number for capillaries with a circular crosssection. Kolb and Cerro [2] expanded on this work by analyzing Taylor flow in tubes of square cross-section, also showing the film thickness to be a function of capillary number. However, these observations are only valid when inertia does not play a significant role. The conditions in small reactor channels operated in Taylor flow are often such that inertia has to be accounted for when estimating the film thickness. When taking inertia into account, it is reported that the film thickness is a function of both the capillary and Reynolds ( $Re = \rho u_b W_b^2 / \mu$ ) numbers and is therefore dependent on the bubble velocity [5–7]. Aussillous and Quere [5] found that inertial effects give rise to a thicker liquid film than predicted by Bretherton's theory and provide a qualitative explanation for this effect. They also stated that the thickening effect is superimposed by a geometric effect which makes the film thickness converge to a finite fraction of the tube radius. However, they provide no quantitative analysis for predicting this limit in the film thickness.

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Nomenciature	
Α	area of the channel cross-section $(m^2)$
$A_{\rm b}$	area of the bubble cross-section $(m^2)$
Bo	Bond number
Ca	capillary number
$F_{\rm b}$	frequency of bubbles (1/s)
g	gravitational constant $(m/s^2)$
$\tilde{L}_{\rm b}$	length of a bubble (m)
L <sub>nose</sub>	length of the nose of a gas bubble (m)
$L_{\rm s}$	length of a liquid slug (m)
$L_{\text{tail}}$	length of the tail of a gas bubble (m)
Re	Reynolds number
ub	velocity of a bubble (m/s)
$U_{g}$	superficial gas velocity (m/s)
$U_1$	superficial liquid velocity (m/s)
$V_{\rm b}$	volume of a gas bubble (m <sup>3</sup> )
$V_{\mathrm{f}}$	volume of the liquid film in a unit cell (m <sup>3</sup> )
$V_{\rm s}$	volume of a liquid slug (m <sup>3</sup> )
$V_{\rm uc}$	volume of a unit cell (m <sup>3</sup> )
$W_{\rm b}$	width of the gas bubble (m)
We	Weber number
Greek	symbols
δ	correction of slug length for the liquid in the slug
	surrounding the nose and tail of bubble (m)
εσ	gas hold-up
$\mu^{\rm s}$	viscosity of the liquid (Pas)

density of the liquid  $(kg/m^3)$ ρ

surface tension (N/m) σ

Experimentally determining the liquid film thickness or gas hold-up from images of the flows is difficult, especially for channels with a rectangular cross-section and the relatively large bubble velocities used in this work. The cross-sectional bubble shape is not axisymmetrical and cannot be obtained directly from images of the flow. Therefore, in this work, a mass balance-based model for Taylor flow is developed. It describes the gas hold-up as a function of bubble and liquid slug lengths, the number of bubbles formed per unit of time, the liquid superficial velocity, and the cross-sectional area of the bubbles relative to the channel cross-section. This model is applied to experimental data obtained by imaging techniques from which the dimensionless cross-sectional bubble area is determined. This allows for calculation of the gas hold-up, which is then shown to be a function of the flow quality according to Armand's experimentally obtained correlation [8]. The model presented in this work is similar, but not identical, to that of Thulasidas et al. [9]. The differences with the Thulasidas model will be addressed explicitly in the next section on Taylor flow model assumptions.

#### 1.1. Taylor flow model assumptions

Before the Taylor flow model is described in detail, the main assumptions and their motivation are discussed.

- (1) At any specific location in the channel, there is no variation in gas bubble and liquid slug sizes.
- (2) There is a uniform, continuous liquid film surrounding the gas bubbles as well as the liquid circulation cells that form the liquid slugs.
- (3) There is no flow in the liquid film.

Ad 1: For any single Taylor flow considered in this work, there is no variation in the amount of gas per bubble and the amount of liquid per slug. However, due to the pressure drop over the channel and the compressibility of the gas phase, the volume of a gas bubble varies with the location in the channel. The pressure dependence of the solubility of the gas phase in the liquid can also cause a small bubble volume change along the length of the channel, but this is not accounted for in this work. Thus, when considering a single location in the channel, all bubbles passing that location have the same volume for a given set of gas and liquid flow rates. Since there is no convective flow in or out of a liquid slug and the liquid phase is assumed to be incompressible all liquid slugs have the same volume for a given set of gas and liquid flow rates.

Ad 2: Up to certain values for the capillary number Ca, liquid circulation cells form between the gas bubbles. These liquid circulation cells are separated from the wall by a thin liquid film. Thus, the liquid film is not only present around the gas bubbles, but continues into the liquid slugs forming a uniform, continuous liquid film throughout the length of the channel. For capillaries with a square cross-section there is both theoretical and experimental evidence [1,2] that these circulation patterns exist for Ca < 0.5. For the experiments in channels with a rectangular cross-section described in this work, the capillary numbers are Ca < 0.1, and smaller than this threshold value. Even though the channels have a rectangular cross-section, it can be assumed that bypass flow does not occur and there is a continuous and uniform liquid film along the length of the channel.

Ad 3: The boundary conditions for the liquid flow in the film are: no shear stresses at the gas-liquid interface and no slip at the channel wall [2,9]. Shear between the gas bubble and the liquid film is therefore not a driving force for any flow in the film. It is also reported [2,6] that there is no pressure gradient in the liquid film in the uniform bubble region, eliminating another potential source for liquid flow in the film. For vertically oriented systems with respect to the gravity vector, gravity can cause flow in the film region, especially for channels with a rectangular crosssection. However, in this work horizontally oriented channels are used and the Bond number  $(Bo = \rho g W_{\rm b}^2 / \sigma)$  is in the order of  $10^{-3}$ , so the effect of gravity is not significant. It is therefore assumed that there is no liquid flow in the film surrounding the gas bubbles. This is also assumed in the work of Thulasidas et al. [9] in the absence of gravity as a driving force.

Shear stress between the liquid in the slug and that in the film can induce flow in the film surrounding the liquid slug. Provided that the liquid slugs are longer than 1.5 times the channel diameter, this will result in fully developed laminar flow of the liquid at some point between two bubbles [1]. In the model developed by Thulasidas et al. [9] it is therefore assumed that there is a fully developed laminar flow in the liquid between two gas bubbles.

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