International Journal of Multiphase Flow 85 (2016) 177–185

Contents lists available at ScienceDirect

International Journal of Multiphase Flow

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

Experimental investigation of wall film renewal in liquid–liquid slug flow

Linda Arsenjuk[a,*], Florian Kaske[a], Joachim Franzke[b], David W. Agar[a]

[a] Department of Biochemical and Chemical Engineering, Laboratory of Chemical Reaction Engineering, Technical University Dortmund, Germany
[b] ISAS – Leibniz Institute for Analytical Sciences, Dortmund, Germany

ARTICLE INFO

Article history:
Received 8 October 2015
Accepted 6 June 2016
Available online 21 June 2016

Keywords:
Capillary micro-reactor
Interfacial area
Liquid–liquid slug flow
Mass transfer
Wall film

ABSTRACT

Liquid–liquid slug flow offers the unique characteristics of high heat and mass transfer combined with a narrow residence time distribution in continuous flow and has thus attracted considerable attention in the field of microfluidics. To exploit its advantages in the successful design and operation of micro-reactors, a precise understanding of the mass transfer processes is essential. In the present work, the role of the thin continuous liquid film formed on the capillary wall in mass transfer is investigated. Fluorescence microscopy is used to determine the exchange between wall film and continuous phase segments to determine if the film is continuously renewed and can therefore be considered to contribute interfacial area available for mass transfer. The distinct wetting properties of different capillary materials are utilized in the experimental set-up to achieve a reproducible and non-invasive release of tracer. The degree of wall film mass transfer as a function of velocity, interfacial area and wall-film thickness is established.

1. Introduction

High rates of heat and mass transfer, resulting from large surface to volume ratios, combined with low hold-ups and fast responses have made micro-reactors a promising tool for process intensification (Roberge et al., 2009, Wiles and Watts, 2011, Yao et al., 2015). In two phase flow in capillaries the dominance of surface forces over gravitational forces leads to the prevalence of the so-called segmented, slug, plug or Taylor flow pattern, characterized by alternating segments of either gas–liquid or liquid–liquid immiscible fluids (Burns and Ramshaw, 2001), with lengths larger than the capillary diameter, as schematically depicted in Fig. 1.

This flow pattern has many advantages over single phase laminar flow. The well-defined and repetitive flow pattern creates a large and reproducible interfacial area for interphase mass transfer (Ufer et al., 2011). This is enhanced still further by the recirculation patterns arising within the liquids segments, the so called Taylor vortices, which are induced by shear with the capillary wall (Abiev, 2013, Kashid et al., 2005, Kreutzer et al., 2005, Taylor, 1961). In addition, the confinement in discrete fluid segments greatly reduces axial dispersion (Muradoglu et al., 2007).

Depending, inter alia, on the contact angles of the fluids with the capillary material, one forms a continuous phase while the other one is dispersed within it, adopting a characteristic bullet shape. This leads to the formation of a continuous film sheath at the capillary wall, enclosing the dispersed slugs (Bretherton, 1961).

1.1. Wall film in gas–liquid slug flow

The existence of a continuous wall film in gas–liquid slug flow was experimentally observed long before the concept micro-reactors emerged. In 1935 Fairbrother and Stubbs (1935) found that in a bubble flowmeter, the bubble and liquid exhibited different average velocities, owing to a thin liquid film at the capillary wall. These investigations were extended to more viscous liquids by Taylor (1961), who observed an increase in film thickness h_{wf} with rising velocity and viscosity. In the same year Bretherton (1961) first modeled this phenomenon and ascertained that the thickness of the liquid film depends on the relationship between surface tension and viscous forces, which can be described by the Capillary number Ca. He proposed the following relationship, which is valid at very low Capillary numbers ($Ca < 5 \cdot 10^{-3}$).

$$\frac{h_{wf}}{R} = 0.643\left(3 \cdot \frac{\eta \cdot V}{\sigma}\right)^{\frac{2}{3}} = 0.643(3 \cdot Ca)^{\frac{2}{3}} \tag{1.1}$$

Since then, this relationship has been refined and its validity extended by numerous experimental and numerical investigations, due to renewed interest in this topic resulting from the growing application of biphasic flow microfluidics. Irandoust and Andersson (1989) measured film thickness by photometric detection in vertical gas-liquid flow and described it empirically as a function

* Corresponding author.
E-mail address: linda.arsenjuk@bci.tu-dortmund.de (L. Arsenjuk).

http://dx.doi.org/10.1016/j.ijmultiphaseflow.2016.06.009
0301-9322/

Symbols

A	lateral surface area bulk region, [mm²]
c	fluorescein concentration, [mmo·L^{-1}]
c_{max}	maximal concentration, [mmol·L^{-1}]
C*	normalized concentration (c/c_{max}), [-]
D_{ax}	dispersion coefficient, [m² s^{-1}]
δ_{ex}	flux between wall film and bulk region, [mm s^{-1}]
h_{wf}	wall film thickness, [µm]
I	fluorescence Intensity, [-]
L_R	length reactor, [m]
P	polynomial factor, [-]
r	radial coordinate, [mm]
$\hat{r}$	normalized radial coordinate, [-]
R	capillary radius, [mm]
u	average flow velocity, [mm·s^{-1}]
V	volume, [µl]
$\dot{V}_{ax}$	axial volume flow in film region, [mm² s^{-1}]
$\Delta\dot{V}_S$	overall segment volume exchanged per second, [% s^{-1}]
ρ	density, [kg·m^{-3}]
η	viscosity, [mPa·s]
σ	interfacial tension, [mN·m^{-2}]

Subscripts

aq	aqueous phase
b	continuous bulk region
c	relating to concentration
flour	aqueous fluorescein phase
n	segment number
org	organic phase
r	relating to capillary radius
s	segment
wf	wall film region

Dimensionless numbers

Ca	Capillary number, $\frac{\eta \cdot u}{\sigma}$
Re	Reynolds number, $\frac{\rho \cdot u \cdot 2R}{\eta}$
VDN	Vessel dispersion number, $\frac{D_{ax}}{u \cdot L_R}$
We	Weber number, $\frac{\rho \cdot u^2 \cdot 2R}{\sigma}$

of the capillary number. Aussillous and Quéré (2000) used Taylor's data supported by their own experiments to develop an empirical correlation for film thickness, known as Taylor's law.

$$\frac{h_{wf}}{R} = \frac{1.34 \cdot Ca^{2/3}}{1 + 1.34 \cdot 2.5 \cdot Ca^{2/3}} \tag{1.2}$$

$$Ca^{*} \sim \left(\frac{\eta^2}{\rho \cdot \sigma \cdot R}\right)^{3/4} \tag{1.3}$$

They further found that this correlation only holds for low Capillary numbers and that above a threshold Capillary number of Ca* one enters a visco-inertial regime, in which inertial effects exert a considerable influence on film thickness. Klaseboer et al. (2014) refined and extended Bretherton's model analytically and developed a correlation quite similar to that of Aussillous and Quéré (2000) valid for Ca < 2. Han and Shikazono (2009) determined film thickness in gas–liquid slug flow with a laser focus displacement measurement combined with refractive index matching and an analytical correction for refraction errors between capillary material and the working liquids. Through diligent observations of the film thickness for a wide range of Reynolds numbers, they proposed a correlation for liquid film thickness dependent on *Re*, We (Weber Number) and Ca, which covers the visco-inertial regime as well. Howard and Walsh (2013) established the validity of this correlation up to Capillary numbers of 1.9 by microscopic image analysis and showed that, due to the high viscosity ratio in gas–liquid flow, the velocity within the liquid film is negligible and that it can therefore be considered as stationary.

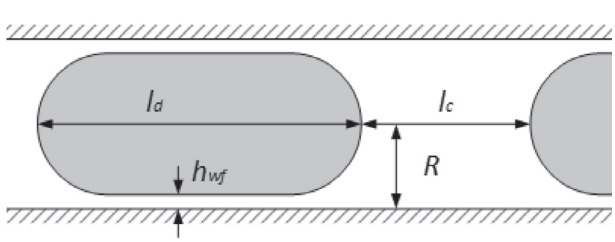

Fig. 1. Schematic of segmented two phase flow in capillaries, the so-called slug, plug or Taylor flow.

1.1.1. Axial dispersion in gas–liquid slug flow

The existence of a wall film and difference in fluid velocities encountered in segmented flow consequently leads to a backflow of the continuous fluid and thus axial dispersion. Its extent in segmented gas–liquid flow was already investigated decades ago in order to characterize the lag phase in continuous flow analysis, where gas has been deliberately introduced to reduce axial dispersion of the liquid phase being analyzed. Thiers et al. (1971) developed an analytical equation for the concentration development of a tracer introduced in a single continuous segment spreading along the subsequent slugs using a simplified model with the assumption of perfect mixing within the segments. Pedersen and Horvath (1981) also used a lumped parameter model to simulate axial dispersion in gas–liquid flow, but included the effect of radial mass transfer by separating the flow into two perfectly mixed zones of a film and a bulk region with an adjustable parameter controlling the mass transfer between them. He found that slower radial mass transfer in the slugs leads to greater axial dispersion along the segments. Thulasidas et al. (1997) modeled the extent of axial dispersion in gas–liquid slug flow on the basis of flow profiles, assuming diffusive mass transfer from the wall film to the outer streamline of the Taylor vortices and validated their model by experiments employing conductimetric measurements. The results showed a strong contribution of convective effects on axial dispersion, which increased with rising velocity. Salman et al. (2004) numerically investigated axial backmixing through the liquid film in gas–liquid flow at Ca < 10^{-3}. By employing a simplified model assuming total exchange of wall film with the two perfectly mixed subsequent slugs, vessel dispersion numbers (VDN) of a magnitude of 10^{-5} were found, which tended to rise with increasing capillary numbers due to a thickening of the wall film. Abiev (2013) used a similar modeling approach and introduced an additional model for mass transfer in gas–liquid slug flow based on a three-layered flow structure, dividing the slugs into a film region, a bulk region and an intermediate layer connecting the two. Muradoglu et al. (2007) studied axial dispersion in gas–liquid flow more precisely using CFD simulations with a finite-volume/front-tracking method and using Lagrangian tracer particles to visualize and quantify mass transfer. By investigation of the effect of Péclet number *Pe, Ca* and slug length on axial dispersion, three regimes were identified, where mass transfer was convection controlled for $Pe > 10^3$, diffusion controlled for $Pe < 10^2$ and in a transitional regime for $10^2 < Pe < 10^3$.

1.2. Wall film in liquid–liquid slug flow

Although the existence and decisive influence of the wall film in gas–liquid slug flow has long been known and characterized