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Mass transfer measurement in a square milli-channel and comparison with results from a circular channel

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ABSTRACT

In this work, we report on an experimental investigation of mass transfer from stagnant Taylor bubbles in a small square channel via measurement of the dissolution rate of an individual elongated bubble of carbon dioxide into water. As a measurement technique we used high resolution X-ray radiography and tomography. The changes in the size of the bubble at constant pressure obtained from the high-resolution X-ray images were used to calculate the liquid side mass transfer coefficient. The bubbles were continuously monitored by hydrodynamic fixation of the bubble in a down flow of the liquid. The results are compared with the available recently published data for circular channels.

The results show that the bubble dissolution curves in square channels are relatively even while the dissolution curves for bubbles in circular channels show some noticeable change in the slope. Furthermore, it is shown that the calculated liquid side mass transfer coefficient based on the measured data show good agreement with the data predicted by the penetration theory when the contact time between two phases is defined as the ratio of bubble length to the relative velocity. In addition, the comparison of the results with the data for circular channels showed that despite the fact that the rise velocity of bubbles in square channel is about three times faster than in circular channel, the liquid side mass transfer coefficients are approximately the same.

1. Introduction

In many industrial processes involving disperse gas and liquid, the mass transfer rate between the contacting phases is an essential parameter for the efficient design and control of the processes. Mass transfer between gas and liquid phases depends upon various parameters such as bubble size, fluid properties, relative velocity between the two phases, degree of liquid contamination, bubble shape, presence of a channel wall and many more.

For milli- and micro-reactors the bubble shape and relative velocity between the two phases are mainly governed by the cross-sectional shape of the channel. For channels with circular cross section (pipes) great attention has been paid in the last decades and many studies on hydrodynamics and mass transfer are to be found in the literature. However, other channel cross sections than circular ones were a subject of only a few studies [1].

Regarding the hydrodynamic investigations, Kolb and Cerro [2] presented experimental data for the flow of an open ended air bubble in a square capillary. They looked at the fluid–fluid interface in both the axial and radial directions for a wide range of capillary numbers (Ca) and showed that the transition from a non-axisymmetric to axisymmetric bubble occurs at $Ca \sim 0.1$. Thulasidas et al. [3] performed extensive experimental investigations in circular and square capillaries to measure bubble size and shape, bubble velocity, and volume fraction of gas inside capillaries for a large range of capillary numbers and also developed a mass balance model to compute the flow parameters with an iteration scheme. In another work [4] they used high-speed video imaging and particle image velocimetry (PIV) to characterize liquid flow patterns and velocity distributions inside liquid slugs. They observed that depending on the capillary number of the flow, counter rotating vortices or a complete bypass flow inside the liquid slug exist. Fries et al. [5] used Laser Induced Fluorescence (LIF) and confocal Laser Scanning Microscopy (LSM) to characterize gas–liquid phase distribution in rectangular microchannels. They did their experiments for low Ca in the range $2 \times 10^{-4} \leqslant Ca \leqslant 1 \times 10^{-2}$. They showed that the film thickness in the corners slightly decreases with Ca and that for low Ca values the film thickness at the wall is nearly constant. Taha and Cui [1] used the volume of fluid (VOF) method to study the hydrodynamics of slug flow inside square and circular capillaries containing

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Notation

A	bubble surface (interfacial) area
A_L	available cross sectional area for the liquid between the bubble and the channel wall
C^*	concentration of gas at interface
C	concentration of gas at the liquid bulk
Ca	capillary number ($\frac{\mu u_b}{\sigma}$)
C_L	water concentration
d_{eq}	sphere-volume equivalent bubble diameter
D	channel internal diameter
D_c	gas molecular diffusion coefficient
E	radiographic extinction image
E_g	radiographic extinction image of a bubble in the liquid-filled tube
E_{ref}	radiographic extinction image of liquid-filled tube
g	acceleration due to gravity
h	distance from the liquid surface
H	Henry's constant
I	X-ray intensity
$I(L/D)$	integral function
k_L	liquid side mass transfer coefficient
k_La	liquid-phase volumetric mass transfer coefficient
k_s	calibration function
k_v	calibration function
L_b	bubble length
M	magnification ratio
n	total moles of gas inside the bubble
P	pressure inside of the bubble
P_{atm}	atmospheric pressure
Q_L	liquid flow rate
R	universal gas constant
$s_{\sum E_b}$	standard deviation of the integral extinction signal
S_b	bubble surface (interfacial) area
SO	object distance
SD	detector distance
t	time
t_c	contact time between gas and liquid
T	bubble temperature
u_b	bubble terminal velocity
u_r	relative velocity between the bubble and liquid in the channel (Q_L/A_L)
V_0	bubble initial volume
V_b	bubble volume
y	mole fraction of CO_2 inside of gas phase
ρ	liquid density
μ	liquid dynamic viscosity
μd	radiographic attenuation
σ	surface tension of liquid

Newtonian liquids by numerical computations. Their computed values of the velocity field and bubble diameter were in good agreement with published experimental data. Recently, Kuzmin et al. [6] reported on a simulation study of three-dimensional channels with square cross sections in the range $0.05 \leqslant Ca \leqslant 6.0$. By resolving the liquid film thickness as twice the interface thickness, they showed that the predicted axial and the diagonal bubble radius are consistent with those reported by Hazel and Heil [7]. They also investigated the existence of a vortex in front of the bubble and the transition from the non-axisymmetric to symmetric case, and showed that the lattice Boltzmann binary liquid model are capable to be used for simulation of gas bubbles in microchannels.

Regarding mass transfer studies, the dissolution of oxygen (O_2) into water in 1, 2 and 3 mm square and circular capillaries was investigated by Vandu et al. [8]. They used oxygen absorption dynamics to measure the volumetric mass transfer coefficient of O_2 as a function of channel diameter and showed that the experimental k_La values are in good agreement with the model developed by van Baten and Krishna [9] for circular capillaries when the dominant mass transfer contribution is assumed to be from the film surrounding the bubble. Dietrich et al. [10] focused on the characterization of gas–liquid mass transfer in a straight millimetric square channel. They developed a new colorimetric technique using an oxygen sensitive dye. They studied various hydrodynamic conditions, compared the equivalent oxygen concentration fields and calculated from them the mass transfer coefficients. Their results were satisfactorily comparable with the measurements using oxygen microsensors. Recently Yue et al. [11] measured flow and mass transfer properties of air–water Taylor flow in square microchannels. Their data showed a large deviation from the available correlations in the literature, which mainly were developed for millimeter-sized channels. They attributed this discrepancy to short liquid slugs produced in their microchannel and rather poor mixing between the liquid film and the liquid slug, which was not in accordance with assumptions associated with the available mass transfer correlations. As a result they applied modifications to the available correlations to be applicable for microchannels with reasonable predicting accuracy.

Concerning the role and importance of square channels in various existing and potential industrial applications such as microelectromechanical systems and monolith froth reactors, there exist still some gaps particularly in related aspects of transport phenomena in these channels and further experimental work is needed to provide detailed heat and mass transfer data for model validation.

In the work reported in this paper, the dissolution rate of a single CO_2 Taylor bubble into the water inside of a square millimeter size channel was investigated using a microfocus X-ray radiography and tomography technique and the results are compared with the circular channel data which was published previously [12]. The X-ray method was chosen since it is not dependent on refractive index. Therefore it is most accurate in comparison with other conventional optical methods. Furthermore, this technique allows tomography for square channels, while full 3D shape determination by optical techniques is difficult in square channels.

2. Materials and methods

2.1. Experimental

2.1.1. Setup

The apparatus and procedure used in this study have been described in detail elsewhere [12]. Thus only a brief description will be given here. The experimental setup is schematically shown in Fig. 1. A glass channel with square cross section (300 mm length, 6 mm hydraulic diameter, made of borosilicate glass) is placed between a microfocus X-ray source and a two-dimensional flat panel X-ray image detector. The capillary is mounted at the hollow shaft of a rotary table which enables rotation between ±180°. Degassed–deionized water with electrical conductivity <1 μS/cm, surface tension 72.7×10^{-3} N/m and pH 6.1 was fed into an open-air overhead reservoir. The reservoir was covered by a plastic head to prevent intrusion of dust particles; however air could dissolve in the water during the experiments. Counter-current liquid flow through the capillary originating from the upper reservoir to the lower reservoir enables the fixation of the bubble at a given