Investigation of surfactant effect on the bubble shape and mass transfer in a milli-channel using high-resolution microfocus X-ray imaging

Mohammadreza Haghnegahdar[a,*], Stephan Boden[a], Uwe Hampel[a,b]

[a] *Institute of Fluid Dynamics, Helmholtz-Zentrum Dresden - Rossendorf, Bautzner Landstr. 400, 01328 Dresden, Germany*
[b] *AREVA Endowed Chair of Imaging Techniques in Energy and Process Engineering, Technische Universität Dresden, 01062 Dresden, Germany*

ABSTRACT

In this paper we present an experimental study on the influence of surface active agents (surfactants) on Taylor bubble flow in a vertical millimeter-size channel. Moreover we give a short review on the subject and previous investigations. We investigated the shape and dissolution rate of individual elongated carbon dioxide Taylor bubbles, which were hydraulically fixed in a downward flow of water. Bubble shape and dissolution rate was determined from microfocus X-ray radiographs. From the shrinking rate we calculated the liquid side mass transfer coefficient.

The results show that the presence of surfactants causes a change of the bubble shape and leads to a slight increase of the liquid film thickness around the bubble and as a result the elongation of contaminated bubbles. In addition, the comparison of clean and contaminated bubbles indicate that presence of surfactant has a more significant impact on the dissolution rate of small bubbles. Furthermore, applying different concentrations of surfactant reveals that in our case, where surface coverage ratio of surfactant on the bubbles is high, increase of contamination does not have a noticeable influence on the mass transfer coefficient of bubbles.

© 2016 Elsevier Ltd. All rights reserved.

1. Introduction

Monolith froth reactors and millimeter-sized reactors have gained a great research interest from the industry and academia because of their advantages such as large interfacial area, high mass transfer rates, low pressure drop, and ease of scale-up over the conventional reactor technology (Donaldson et al., 2011). To improve the efficiency of this kind of reactors, it is required to understand the relationship between the mass transfer rate and other important parameters. For gas-liquid two-phase systems, one of the main factors, which is known to have a significant influence both on the hydrodynamics and mass transfer rate of phases, is presence of surface active agents (surfactants). Surfactants are adsorbed at gas–liquid interfaces and decrease the surface tension. The presence of surfactants in multiphase systems, either in the form of unavoidable impurities or as additives, has a great effect on the shape and the dynamics of the interfaces (Bothe and Prüss, 2010). Great attention has been paid to investigate the ef-

fect of surfactants on small and/or spherical bubbles in infinite liquid both theoretically and experimentally, while contamination of large/elongated bubbles in small channels are subject of only few studies. In the following, the most relevant studies on the effect of surfactants on the gas-liquid systems are shortly reviewed.

1.1. Theoretical investigations

Regarding theoretical studies, Weber (1975) investigated the effect of surfactant on the mass transfer of spherical-cap bubbles at Reynolds number ($Re = \rho D U_b/\mu$) higher than 100, where U_b is bubble rise velocity, D the channel diameter, ρ the liquid density and μ the liquid dynamic viscosity. It was supposed that the surfactant forms a stagnant film on the spherical surface near the rim of the bubble and its effect was attributed to a balance between surface forces and shear forces. Comparison of their results with experimental data showed agreement within 25%. Stokes flow due to the motion of a liquid drop or bubble in a contaminated immiscible fluid was analyzed by Sadhal and Johnson (1983). An exact solution was found for the resulting problem for an arbitrary cap angle and an expression for the drag force in terms of viscosities and the cap angle was for the first time developed. The dissolution of spherical bubbles at low Reynolds numbers was studied by

* Corresponding author. Fax: +49 351 260 1 2772, Phone: +49 351 260 3773.
E-mail addresses: m.haghnegahdar@hzdr.de (M. Haghnegahdar), s.boden@hzdr.de (S. Boden), u.hampel@hzdr.de (U. Hampel).

Notation

A	bubble surface area based on the bubble equivalent diameter
C^*	concentration of gas at interface
C	concentration of gas at the liquid bulk
Ca	Capillary number $(=\mu U_b/\sigma)$
C_L	water concentration
C_s	concentration of surfactant at the liquid bulk
d	bubble diameter
d_{eq}	sphere-volume equivalent bubble diameter
D	channel hydraulic diameter
D_c	gas molecular diffusion coefficient
d_{max}	bubble diameter at its maximum cross section in the channel
dev_{max}	the maximum deviation in absolute distance between the interface sampling points extracted from the x-ray images and their nearest points on the fitted polynomial curve
E	radiographic extinction image
Eo	Eötvös number $(=\Delta\rho g D^2/\sigma)$
g	acceleration due to gravity
h	distance from the liquid surface
H	Henry's constant
I	X-ray intensity
k_L	liquid side mass transfer coefficient
$k_L a$	liquid-phase volumetric mass transfer coefficient
k_s	calibration function
k_v	calibration function
La	Laplace number $(=C_s\beta/\alpha)$
L_b	bubble length
n	total moles of gas inside the bubble
P	pressure inside of the bubble
P_{atm}	atmospheric pressure
Pe	Peclet number $(=DU_b/D_c)$
r	radial direction, bubble radius
R	universal gas constant
Re	Reynolds number $(=\rho DU_b/\mu)$
S_e	surface coverage ratio
S_b	bubble's interfacial area
Sh	Sherwood number $(=Dk_L/D_c)$
t	time
T	bubble temperature
U_b	bubble terminal velocity
V_b	bubble volume
y	mole fraction of CO_2 inside of gas phase
z	axial direction
α	desorption rate constant
β	adsorption rate constant
ρ	liquid density
μ	liquid dynamic viscosity
μd	radiographic attenuation
σ	surface tension of liquid

Dani (2006) using Direct Numerical Simulation (DNS) for fully contaminated, partially contaminated and clean bubbles by applying the conventional stagnant cap model. The comparison of results with classical relations showed the good scaling of Sherwood number $(Sh = Dk_L/D_c)$ with $Pe^{1/3}$ and $Re^{1/2}$ respectively for solid sphere and clean bubble in creeping flow, where Pe $(=DU_b/D_c)$ is Peclet number, k_L the liquid side mass transfer coefficient and D_c the gas molecular diffusion coefficient.

In a comprehensive study, Muradoglu and Tryggvason (2008) developed a finite-difference/front-tracking method to simulate the interfacial flows with soluble surfactants. In their method both the interface and bulk surfactant concentration evolution equations were solved and coupled with the incompressible flow equations. The simulation was done to predict the influence of surfactant on the hydrodynamics of buoyant viscous bubbles in a straight channel. It was found that the contaminated bubble behaves like a solid sphere in the limit of very low Reynolds number $Re < 1$ and the results were found to be in a good agreement with the experimental correlations collected by Clift et al. (1978). Tasoglu et al. (2008) used the same approach to study the unsteady motion and deformation of a bubble rising in an otherwise quiescent liquid. They showed that the surfactant generally decreases the terminal velocity of the bubble but this reduction is most noticeable in the nearly spherical regime in which the bubble behaves like a solid sphere and its terminal velocity reaches that of a solid sphere.

Cuenot et al. (1997) considered the similar problem and confirmed the validity of the well-known stagnant-cap model for describing the flow around a bubble contaminated by surfactants. Their results indicated a considerable rise of the drag which in several cases reaches the value corresponding to a rigid sphere. Hayashi and Tomiyama (2012) investigated the effect of surfactant on the terminal velocity of Taylor bubbles in vertical pipes. They applied an interface tracking method and simulated for various Eötvös numbers $(Eo = \Delta\rho g D^2/\sigma)$, different surfactant concentration and two different surfactants, where g is acceleration by gravity, σ surface tension and $\Delta\rho$ the density difference between two phases. They showed that the terminal velocity of bubbles increased because of reduction of surface tension near the bubble nose and since the bubbles at high Eötvös numbers are independent of surface tension, the presence of surfactant does not affect the terminal velocities of high Eötvös number bubbles.

Ghadiali and Gaver (2003) utilized a numerical model of semi-infinite air bubbles in a capillary to analyze the continual interfacial expansion dynamics that occur during the opening of collapsed pulmonary airways. They showed that the surfactant properties can strongly affect the interfacial pressure drop through modification of the surface tension and the creation of Marangoni effect. In addition, they showed that, depending upon the range of parameters, either film thickening or film thinning responses are possible.

In case of liquid film thickness around the bubble, Ginley and Radke (1989) presented a regular perturbation expansion in large adsorption rates within the low capillary number, singular perturbation hydrodynamic theory of Bretherton. They considered surface transport of the surfactants and neglected all concentration gradients in the bulk phase and showed that by addition of soluble surfactant to the liquid phase, the surface concentration increases in the thin film region and leads to a decrease in the film thickness compared to the surfactant-free case. In contrary, in a more comprehensive analysis and for a semi-infinite bubble in a capillary, Ratulowski and Chang (1990) carried out an asymptotic analysis for various convective, diffusive and kinetic timescales and showed that, if transport in the film is mass-transfer limited, the film thickness increases by a maximum factor of $4^{2/3}$ over Bretherton's mobile result at low bubble speeds. Stebe and Barthès-Biesel (1995) considered the same problem for a viscous surfactant solution at high concentration. They considered the case where the surfactant flux is adsorption-desorption controlled and the equations of momentum and mass transfer are coupled to leading order. The results showed that interfaces with surface viscosities require larger pressure drops to aspirate the flow and leave thicker wetting layers along the capillary walls. For finite length bubbles in capillaries, Park (1992) studied the influence of soluble surfactant on the steady motion of inviscid bubbles. Both front and rear ends of the bubble were examined in the limit of small capillary number and it was shown that due to the accumulation of the surfactant at the rear end of the bubble, the film thickening effect of the surfactant