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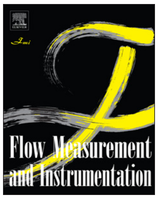

Measurement of Taylor bubble shape in square channel by microfocus X-ray computed tomography for investigation of mass transfer

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ABSTRACT

This paper addresses the measurement of the dynamic evolution of Taylor bubble volume and interfacial area during mass transfer. Carbon dioxide (CO_2) Taylor bubbles were placed in countercurrent water flow in a square channel. Microfocus X-ray radioscopy enabled the measurement of volumetric dissolution rates. The measurements were calibrated by microfocus X-ray computed tomography scans of non-dissolving air Taylor bubbles. The reconstruction algorithm was adapted to correct the slight position shift of the Taylor bubble during the tomographic scan. The obtained three-dimensional representation of the Taylor bubble's shape enabled the measurement of Taylor bubble's true volume and interfacial area, which were correlated to the two-dimensional radioscopic measurements. The volumetric dissolution rates were measured in a square milli-channel with hydraulic diameter of $d_h = 6$ mm. Furthermore, the thin film region of the constricted Taylor bubble's surface near the planar channel walls was extracted from the data and the ratio to the total Taylor bubble surface was computed.

1. Introduction

Mass transfer is of importance in many applications in chemical engineering. During mass transfer molecules of particular species are transported due to differences in species concentration. The mass flux along the interfaces between a disperse gas phase and the surrounding liquid phase is of particular interest. One selected example is Taylor bubble flow [1] in small channels, which is the desired operation state in microreactor applications [2]. The key advantages of Taylor bubble flow are good mixing of species within the bubble, large interfacial area per unit volume and the thin liquid film between bubble and wall, reduced axial dispersion of liquid, good mixing and recirculation in the liquid slug and wall-normal convective transport in laminar flow. These features make Taylor bubble flow an ideal flow regime to improve mass transfer performance between the gaseous and liquid species [3,4].

So far several experimental and theoretical studies are known in which the effect of various parameters on mass transfer between Taylor bubbles and liquid slugs were investigated (e.g. most recently by [5,6]). The most common measuring technique was photographic observation of the change of the bubble's volume [7]. Photographic measurement of bubble shape, however, requires careful refractive index matching to correct for image distortions due to optical refraction at the glass and liquid interfaces, it does not reveal the three-dimensional shape of the bubble and it is not applicable for opaque materials like ceramic monolithic structures. These drawbacks may be overcome by the application of microfocus X-ray radioscopy and tomography.

Microfocus X-ray visualization techniques rely on the utilization of X-ray sources with a micrometer sized focal spot [8]. An object placed in the divergent X-ray beam originating from the source is projected onto a detector plane with a spatial uncertainty in the object space of a few micrometers corresponding to the size of the X-ray source. High magnification ratios may be employed to acquire the X-ray image with readily available two-dimensional flat panel X-ray image detectors with rather coarse pixel spacing of a few hundred micrometers edge length. As hard X-rays penetrate materials on straight paths, no refractive index matching is necessary, and even the internal structure of opaque objects is revealed. Although the measured attenuation images correlate to the length of the X-ray paths in the material, radioscopy is still a two-dimensional (2D) visualization technique. However, the acquisition of a set of projection images around the object enables the high-resolution three-dimensional (3D) reconstruction of the internal object shape by the application of computational

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tomographic reconstruction algorithms [9]. Laboratory microfocus X-ray sources provide only low X-ray photon flux, therefore the projection image acquisition is a lengthy task and thus only images averaged over time can be measured. Only high flux synchrotron radiation sources may be applied to decrease acquisition time to capture images of instantaneous gas bubble shape even in the case of fast motion [10].

The microfocus X-ray visualization technique was used for investigation of the mass transfer from Taylor bubbles in circular milli-channels previously [5]. In the current paper we present a method to measure the volumetric dissolution rate of a dissolving Taylor bubble in countercurrent liquid flow in milli-channels with square cross-section by means of high-resolution X-ray radioscopy, whereby the method was calibrated by microfocus X-ray computed tomography measurements. To investigate the mass transfer between a Taylor bubble and the liquid flowing around the bubble the shape of a dissolving CO_2 Taylor bubble was radioscopically monitored. The acquired X-ray images of the bubble were analyzed with respect to the integral X-ray attenuation of the gas bubble. These dynamic integral X-ray attenuation measurements were quantitatively correlated to true volume and surface area of the Taylor bubble through a set of tomographically measured 3D shapes of non-dissolving air Taylor bubbles. The results might be used to gain further knowledge on the mass transfer characteristics of Taylor bubble flow and to validate corresponding CFD models [11].

2. Materials and methods

2.1. Experimental setup

The experimental setup to study the behavior of dissolving Taylor bubbles in vertical channels by microfocus X-ray radioscopy and tomography consisted of a microfocus X-ray source operated at 135 kV tube voltage and 5 W tube power and a two-dimensional flat panel X-ray image detector with a pixel size of $200 \times 200\ \mu m^2$. A milli-channel mounted at the hollow shaft of a rotary table to enable channel rotation between $\pm 180°$ was placed near the X-ray tube as shown in Fig. 1. Liquid flow from an elevated reservoir through the channel was driven by gravity only to eliminate pump induced flow fluctuations. A Taylor bubble was generated by injection of some finite amount of gas into the liquid through a metallic needle mounted at a T-junction at the bottom of the channel. The amount of gas injected into the liquid was controlled by a remotely operated fast acting solenoid valve as described elsewhere [10]. By precisely adjusting the liquid flow-rate with a remotely controlled motorized needle valve the position of the buoyant Taylor bubble was kept constant in countercurrent flow conditions. A borosilicate glass channel was used with square cross section and with a hydraulic inner diameter of $d_h = 6$ mm and a length of 300 mm. The liquid was deionized water and the gas was either carbon dioxide (CO_2) or pressurized air.

2.2. Radioscopic monitoring

The shrinking Taylor bubble was radioscopically monitored at exposure times of 100 ms and frame repetition rates of 5 Hz. From the acquired radioscopic data, attenuation images A were computed. Each pixels' value in these images was computed as the negative logarithm of the ratio of measured intensity I_{ij} with the channel in the beam and the reference intensity $I_{0,ij}$ without any object in the X-ray beam,

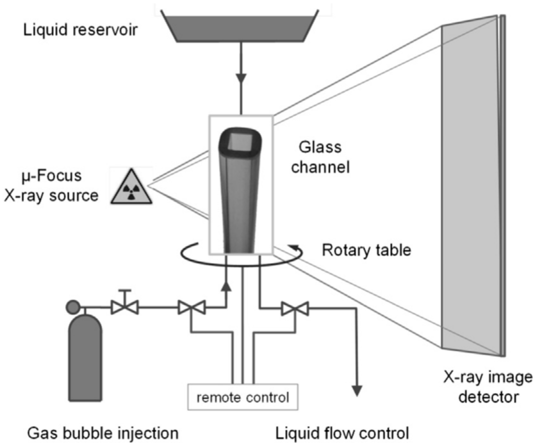

Fig. 1. Schematic drawing of the experimental setup.

$$A_{ij} = -\log\left(\frac{I_{ij}}{I_{0,ij}}\right) \approx \int_0^{\mathbf{x}_{ij}} \mu(s)ds. \tag{1}$$

The attenuation A_{ij} only approximatively is equal to the line integral over the attenuation coefficient distribution $\mu(s)$ along each individual X-ray ray path between source and detector pixel ij at position $\mathbf{x}_{ij}$ as given by Beer–Lamberts law due to beam hardening and scattering inherent in cone-beam microfocus X-ray measurement setups. However, the inverse of Eq. (1) is the basis of all X-ray tomographic reconstruction algorithms to recover a given attenuation coefficient distribution. Moreover, the inherent non-linear imaging properties are only minor in the setup used here, and with $A_{ref,ij}$ being an attenuation image showing the liquid filled channel only without any bubble in view, attenuation images $A_{b,ij}$ showing the Taylor bubble only were computed by subtraction (Fig. 2),

$$A_{b,ij} = A_{ij} - A_{ref,ij}. \tag{2}$$

2.3. Measurement of bubble volume and interfacial area from 2D radioscopic images

The bubble's volume V_b and interfacial area S_b can be estimated from a solid body of rotation through the virtual revolution of an extracted contour profile of the Taylor bubble out of a 2D radioscopic projection assuming rotational symmetry of the bubble. In the case of a circular channel, measured V_b and S_b represent the true bubble volume and interfacial area, if rotational symmetry of the Taylor bubbles can be ensured. However, it turned out that the absolute fixation of the vertical position of a dissolving Taylor bubble by remote controlling of the countercurrent liquid flow was difficult. The resulting bubble motion resulted in noticeable motion blurring in the radioscopic projections at the front and rear tip of the bubble and thus invalidated extracted 2D contour profiles. But, the integral attenuation $\sum_{ij} A_{b,ij}$ appeared as independent on bubble motion, thus instead the linear correlations

$$V_{b,2D} = k_V \sum_{ij} A_{b,ij} \tag{3}$$

$$S_{b,2D} = k_S \sum_{ij} A_{b,ij} \tag{4}$$

were utilized with k_V and k_S being constant factors. These factors were evaluated from radioscopic projections of non-dissolving air Taylor bubbles with no apparent bubble motion, U_B=0, only, and utilizing $V_{b,2D} = V_b$ and $S_{b,2D} = S_b$ as described in detail by Haghnegadhar et al. [5].