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In situ X-ray tomography of aqueous foams: Analysis of columnar foam generation

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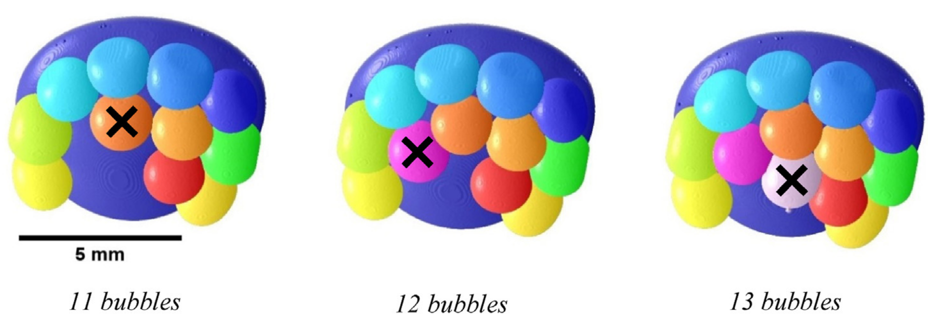
HIGHLIGHTS

- Bubble rearrangements and displacements within layers, ejections out of a layer as well as bubble chain and cluster rotation at the meniscus took place.
- Fcc or hcp ordering of bubbles at the outer layer.
- A volume increase of the bubbles in time, possibly due to a temperature increase caused by X-ray irradiation.
- A decreased of sphericity of bubble layers with time with increasing bubble number, i.e. with increasing foam height and liquid drainage.

GRAPHICAL ABSTRACT

3D rendering of a sequence of tomographies of an aqueous foam made by adding one bubble every minute to a liquid/gas interface (big blue surface). By analysing the images one can follow the build-up of the foam structure in detail. Crosses mark the bubbles that have arrived last in each image.



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ABSTRACT

The accumulation of bubbles rising in a liquid and forming a foam is a complex process which is difficult to observe and describe in three dimensions due to the poor visibility of bubbles in the depth of a foam when using optical microscopes. We applied in situ X-ray tomography in phase-contrast mode to visualise how bubbles of almost equal size accumulate and form a foam confined within a vertical tube in an aqueous liquid. By capturing a full 3D image of the foam after the arrival of each bubble and tracking each bubble individually, collective movements of bubbles, displacements and changes of positions could be described unambiguously.

1. Introduction

To study bubble arrangements in foams in three dimensions, to measure the local liquid fraction within, order parameter of foams or other properties, X-ray tomography is a suitable and accepted method [1–3]. Recent developments at synchrotron X-ray tomography beamlines have led to such high tomography acquisition rates that liquid foams can now be studied in situ and dynamic processes such as bubble movements, coarsening or drainage observed in the entire foam volume with high time resolution. [4–11] Such time-resolved tomography is also called '4D tomography'.

The arrangement of monodisperse aqueous bubbles produced by gas injection into a liquid is of special interest [12–16]. Opti-

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cal observation allows only for the study of the structure of the first three flat surface layers in crystalised monodisperse foams [13,17], but can hardly reproduce the inner structure of a bulk foam as X-ray tomography permits [18]. Light scattering experiments probe a deeper volume but only average information about bubble sizes in the bulk is provided and other limitations apply [3,19–21]. Simulations based on packings of hard spheres have been used to understand the structure of monodisperse bubbles in cylindrical columns [22–25], and recently also combined with experiments [26]. But such simulations are restricted to ratios of the cylinder diameter D to the bubble diameter d of $D/d<2.9$ [22] and recently until $D/d=4$ [25] as computation time increases exponentially with increasing ratio. Tomographic images of monodisperse aqueous foams constrained in columns were recorded and helped to illustrate arrangements of real bubble in such systems [27]. Such experiments open access to larger D/d ratios and provide the positions of all bubbles inside a specimen. The structures found differ from ideal structures due to several reasons: i) a slight deviation from monodispersity of bubble sizes, ii) the deformation of the bubbles compared to hard spheres, iii) an alignment of the bubbles along a curved instead of a flat surface due to the meniscus shape of the liquid/gas interface, and iiii) defects and rearrangements created by the interaction between resting and injected bubbles, i.e. by the impact of incoming bubbles accelerated by buoyancy forces [28].

In the work presented here, the generation of a foam column was studied by observing the attachment of individual bubbles to a liquid gas interface in situ using X-ray tomography. Such in situ 4D tomography with moderate time resolution has been used to research dynamic effects such as coarsening [3] or crystallisation [29] and was used in the present work, with increased time resolution, to study the build-up of a foam constrained in a cylindrical tube.

2. Experimental

2.1. In situ generation of individual bubbles

Bubbles were produced by injecting air through a stainless steel cannula of 200 mm inner diameter into a vertically arranged thin-walled tube of diameter $D\approx 8.5$ mm filled with an aqueous solution (Fig. 1). The tube was made of polyimide that transmits X-rays very well. To obtain stable foam we used an aqueous solution containing 5 vol.% of the commercial detergent *Fairy Liquid* and to reduce the coarsening rate we enriched the blowing air with perflourohexane vapor that is insoluble in the liquid [30]. Individual bubbles of a controlled size were generated by defining pressure pulses using a very fast magnetic valve (type MHE2-MS1H-3/2G-M7-K from Festo, Germany) for the opening and closing of the gas supply during a period of time as short as 1.7 ms. The valve was controlled by self-developed hardware, which allows for adjusting the opening time and the interval between the pulses independently and also to generate single pulses. Adjustment of the opening time on the one hand and the gas pressure in the line with a needle valve on the other allowed us to control the gas volume of each pulse. Single bubbles of a similar pre-set diameter d ranging from 1.5 mm to 5 mm could be produced. The pre-set gas volume $V\approx 3$ mm^3 used in the experiment corresponded to a spherical bubble diameter of $d\approx 1.7$ mm, i.e. to a nominal cylinder-to-bubble diameter ratio $\lambda = D/d\approx 5$, going beyond the simulation limits [25]. For our experiment, we chose an interval between gas valve activation of one minute, i.e. produced one individual bubble every minute. This time interval allowed us to record a complete tomography in 30 s, inject the next bubble and let the foam column relax for a few seconds, during which the gas-liquid interface that had been slightly pushed up during gas injection by the liquid in the line returned to its original position, before recording the next tomography. In total 48 bubbles were produced in the tube, after which the experiment was terminated.

2.2. In situ 4D tomography

To allow for 4D tomography, the whole gas injection setup including the cylindrical tube containing the aqueous solution, a washing bottle containing perflourohexane, the injector cannula, the magnet valve and a source of pressurised air (a small electrical air pump) was fixed on a rotation stage and rotated continuously (Fig. 1). Several electrical sliding contacts allowed us to connect the devices on the rotating table to the external hardware without limiting the maximal amount of continuous rotations.

The experiment was carried out at the imaging beam line ID 19 of the European Synchrotron Radiation Facility (ESRF), Grenoble, France. The beam line provides different polychromatic configurations with high photon flux density in the energy range of 18 – 250 keV. The radiation transmitted through the foam was transformed to visible light by a 100-μm thick LuAG:Ce scintillator, after which the image was guided to the camera sensor using a mirror and lenses. A pco.dimax high-speed camera from PCO AG, Kelheim, Germany, with 2016 × 2016 pixels together with an optical magnification of 2 allowed us to obtain a field of view of (10.2 × 10.2) mm^2 with each pixel representing an area of 5.06 μm edge length in the sample. Although faster image acquisition rates than the one chosen would have been feasible, we selected a polychromatic beam with narrow bandwidth of around 2% (undulator U17-6C, gap 29.5 mm) with an energy of 19 keV and a limited flux to prevent radiation damages of the foam sample. In particular, we recorded a tomography in 30 s (300 projections/180°) with 35 ms exposure time per projection (Fig. 2). A drift space between sample and detector allowed for the application of propagation-based phase contrast in order to visualise the weakly attenuating cell walls.

4D tomography does not only visualise the foam structure and its evolution from one bubble injection to the other, but also allows us to quantify bubble positions, bubble volumes, local or integral liquid fractions, foam inner surfaces, number of neighbours, order parameters, etc. [18]. As in our experiment bubble sizes were in the mm range, they could be deformed and then deviated from the ideal spherical shape depending on their position and the number of their neighbours. Therefore, for each bubble we defined its bubble diameter as the equivalent diameter of a sphere of the same volume as the bubble extracted from the tomographic data set.

As shown in Fig. 3, individual bubbles could be separated and labelled with different numbers or colours. As the gas volume of each bubble could be measured very precisely, individual bubble tracking due to the slightly different bubble volumes was possible, e.g. allowing us to monitor whether a single bubble was displaced or exchanged its position with another bubble or whether a whole cluster of bubbles was rotated under the influence of the incoming bubble.

3. Results and discussion

A total number of 48 bubbles were injected and corresponding tomographies acquired after each injection. Bubble rearrangements of different magnitude, bubble displacements within a bubble layer as well as rotations of bubble chains and clusters during the filling of the cylinder were observed. Furthermore, the bubble sphericity and diameter evolution of the bubbles was studied.

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