Experimental study on the effects of phase change during a bubble collapse

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Abstract

A planar shock front produced in a shock tube provides an instantaneous pressure rise within a water-like gelatin mixture that contains bubbles filled with two different gases. Air represents a gas that reacts non-condensable under the experimental conditions, while refrigerant vapor R1233zd-E is a gas with a vapor pressure slightly above atmospheric pressure under the experimental conditions. We compare both types of bubbles during an aspherical collapse and highlight similarities and differences in order to analyze the effect of the gas content on collapse dynamics. The experimental set-up is presented and preliminary results are discussed. An image sequence of a recorded video and normalized radius data show the dynamic response of both investigated bubble types positioned next to each other close to a solid wall. A difference in the rebound behavior is indicated with the R1233zd-E bubble expanding more omnidirectional and to a lower relative radius.

Keywords: shock-induced bubble collapse; aspherical collapse dynamics; cavitation

Introduction

Collapsing bubbles or bubble clusters may be used in medical applications for targeted drug or gene delivery [1], for microbubble-cell interaction [2], to non-invasively dissolve blood clots [3] or to treat urinary stones in extracorporeal shock wave lithotripsy [4]. But first, the microbubbles need to be transported unharmed and in a controlled way to the desired locations in the human body and so a long residence time of the microbubbles in blood must be assured. To achieve that, bubbles are usually coated and filled with insoluble gases as for example perfluorocarbon gases like perfluorobutane or perfluoropentane. Interestingly, the vapor pressure of these gases is near atmospheric pressure [1] and hence, acoustic pressure pulses in medical treatments that are used to collapse bubbles, can also trigger condensation. It is thus possible that both effects interact and influence the bubble collapse. Consequently, the present experimental study aims at investigating this effect in the case of aspherical collapse dynamics by comparing bubbles filled with air, as a gas that reacts non-condensable under the experimental conditions, and R1233zd-E refrigerant vapor, a gas with a vapor pressure slightly above atmospheric pressure at the experimental conditions. R1233zd-E is a new generation of working fluid that is non-toxic, non- or mildly-flammable and has similar thermodynamic properties as the more common refrigerant R245fa [5]. To achieve good comparability, the collapse is initiated simultaneously for both bubbles under identical and homogeneous experimental conditions.

A wide number of high-quality experimental investigations with respect to the collapse behavior of bubbles is present in the literature, but mostly focuses on cavities produced through optical breakdown, e.g. [6, 7], or electric discharge, e.g. [8-10], where the effect of different gases cannot be investigated. Experiments investigating preexisting gas bubbles are difficult because the bubbles preferably need to be of spherical shape and must be positioned and kept at a point that can easily be observed during the collapse. Different ways to manage the problem were applied and some are mentioned here exemplarily. Tomita and Shima [9] examine bubble-shock wave interaction using a spark discharge to create a shock wave that impacts on an air bubble attached to a wall. The bubbles collapse aspherically and show the formation of a liquid jet that leaves an imprint on an impact specimen. Other groups investigate bubbles below a gelatin layer impacted by an underwater shock wave generated via a micro explosive pellet [11] or optical breakdown [12]. Rising air bubbles interacting with a high-pressure shock wave are also studied [13, 14]. Ohl and lkink [15] analyze jetting and jet breakup of micro bubbles in water that are subject to a shock wave by an extracorporeal lithotripter, while Kodama and Takayama [16] focus on tissue-damage mechanisms during extracorporeal shock wave lithotripsy. Philipp, et al. [17] also use a lithotripter to collapse air bubbles attached to a plastic foil. Gases other than air are comparatively scarcely used, but Tomita et al. [18] mention the collapse process of a hydrogen bubble generated at the top of a needle through electrolysis.

This introduction shows that the aspherical bubble collapse is usually analyzed either for cavities or air bubbles. Other gases are rarely used and their effect on the collapse is not adequately analyzed. Nevertheless, this aspect should not

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be discarded completely as it has been shown, for example in the context of sonoluminescence, that oscillating bubbles with different gas contents show a different behavior during the collapse [19].

With this ongoing study, we will contribute further experiments to the community in order to identify the influence of different gases and the effect of phase change on bubble collapse behavior. The chosen experimental set-up offers two particular advantages: First, it allows the simultaneous investigation of diverse bubbles containing different gases and hence ensures a high comparability. Second, the homogeneous and well-defined initial conditions allow for comparison to successive numerical simulations.

Experimental equipment and procedure

A schematic diagram of the experimental set-up is shown in fig. 1. The shock tube, with an overall length of 22.5 m and an inner diameter of 290 mm, consists of three parts: the driver, the driven and the test section. A diaphragm separates the high-pressure driver section from the driven section at atmospheric pressure. After the diaphragm breaks, a shock wave forms and propagates towards the test section, where experiments are conducted by using the instantaneous pressure increase caused by the incident shock wave and subsequent shock reflection.



Figure 1: Experimental arrangement of the shock tube for bubble dynamics experiments.

The concept of analyzing bubble dynamics via shock tube experiments has been proofed by other groups before. Beylich and Gülhan [20], for example, investigate transient wave phenomena for glycerin-gas mixtures or shock wavebubble interaction for single bubbles, bubble chains and bubble clouds. Even the different effect of He, N₂ and SF₆ on shock wave propagation is considered, but the influence on the collapse is not stressed. In another study, the free oscillation of bubbles after a shock tube generated pressure jump is analyzed with a needle hydrophone by monitoring acoustic waves radiated by the bubble [21]. Fujikawa and Akamatsu [22] use the expansion wave generated in a shock tube to expand gas nuclei that collapse under a following compression wave.

In the present set-up, the shock tube-generated shock wave arrives in a test section filled with gelatin. Figure 2 shows shock propagation and reflection exemplarily for a repeatedly investigated operating point (labeled as standard test case). The incident shock wave in the air arrives at the gelatin interface where it reflects almost ideally due to the high difference in acoustic impendence. However, the pressure at the interface must be equal and hence a compression wave forms and propagates into the gelatin. At the solid end of the test section, the compression wave is reflected, which further increases the pressure. Starting from that point in time, the ambient conditions around the bubbles remain constant at almost zero velocity and with a well-defined high pressure field until the compression wave reflects at the air-gelatin interface and returns as an expansion wave to the bubble position. A test time Δt_{test} of around 0.5 ms results for 400 mm gelatin and a speed of sound in gelatin similar to water. According to estimates with the Rayleigh-Plesset equation for 10 bar surrounding pressure, gas bubbles at atmospheric pressure of up to 5 mm collapse, rebound and collapse a second time within that time span, which confirms the validity of the concept for bubble dynamic experiments.

GelriteTM Gellan Gum for microbiological applications is used in combination with magnesium sulfate as soluble salt to form a clear, agar-like gel. The GelriteTM is dissolved in distilled water and the mixture is heated until the boiling point is reached and the magnesium sulfate is added. Subsequently, the liquid gelatin is decanted, cools down slowly and solidifies. Gelatin has already been used for bubble dynamics investigation, namely for the two-dimensional gel

technique [23-25], but is now applied in a three-dimensional set-up. Bubbles are produced by carefully inserting the needle of a syringe into the partly solidified gelatin and releasing a defined amount of air or R1233zd-E.



Figure 2: Sketch of the test section with a wave diagram; Pressure values and test time are given for a standard test case with 8.5 bar pressure in the driver section and atmospheric pressure in the driven section of the shock tube.

For visualization, we apply a Z-type schlieren system with a 150 W constant Xenon light source to create either schlieren images by cutting light at the second focal point with a knife edge or 'focused shadowgraph' images without the knife edge [26]. Using the knife edge is useful for shock visualization, while the direct shadowgraph provides more light and thus allows a higher frame rate or more optical zoom. The optical system projects the visualized section along the line-of-sight on a camera focal plane and thus gives a two-dimensional representation of the three-dimensional bubble. The entire collapse procedure is recorded with a Shimadzu HyperVision HPV-X ultra-high-speed camera that produces videos consisting of 128 consecutive frames with a resolution of 400x250 pixels at up to 5 million frames per second. In addition, PCB Piezotronics ICP® fast-response pressure sensors are connected to a LTT transient recorder and record the pressure during the collapse at up to 16 locations at a frequency of 4 million samples/s per channel.

A new test section was planned and is built, featuring a design that relies on experience gathered during preliminary experiments. Due to delays in deliveries and the manufacturing process, the section will be finished within January 2018 and then will allow to conduct experiments according to the simplified sketch shown in fig. 2. Despite the delay of the new test section, preliminary experiments have been conducted using acrylic boxes that are filled with gelatin and placed inside an already existing test section. This workaround did not allow for valid pressure measurement and undesired effects like deformation of the acrylic box and additional wave motion within the surrounding gas of the box occurred. The following preliminary results should thus be handled with care, but nevertheless show that the basic features of the concept work well. New results without the mentioned uncertainties are presented at the conference.

Preliminary results

Using the mentioned workaround, experiments with air bubbles have been conducted on a regular basis and capture well-known effects like the development of a liquid jet that penetrates the bubble. An image sequence of a single air bubble collapsing next to a solid wall is presented in fig. 3. The top row shows the aspherical collapse of the bubble towards the solid wall on the right. In the fourth image, a flattening of the bubble on the far side of the wall is already

visible and a distinct liquid jet develops in the following images. The bottom row shows the rebound of the bubble including delicate flow features like Rayleigh-Taylor instabilities at the bubble surface causing bubble fission. Planar shock waves in the surrounding gas of the acrylic box are observed in frame 6-8, but since the new test chamber will exclude these additional waves no further attention is payed to this effect here.



Figure 3: Image sequence of an aspherically collapsing air bubble close to a solid boundary (interframe time 30 µs).

As a next step, experiments for bubbles with different gas content were conducted and an image sequence of the recorded video of an exemplary experiment is presented in fig. 4. It shows a R1233zd-E bubble in the upper part and an air bubble in the lower part of each frame. The bubbles are positioned in the gelatin next to a solid wall in such a way that both bubbles can be observed simultaneously. The refrigerant vapor bubble is smaller and closer to the wall than the air bubble so consequently, a shorter collapse time results. However, the different bubble sizes and positions result in a similar stand-off parameter $\gamma = s/R_0$, with *s* as the distance between the solid boundary and the bubble center and R_0 as the bubble radius at collapse initiation. Thus, a similar collapse pattern is expected.



Figure 4: Image sequence of a collapsing R1233zd-E gas bubble and an air bubble close to a solid boundary (interframe time 42µs).

Comparable to the experiment with the single bubble, both bubbles collapse aspherically because of the asymmetric pressure field caused by the solid wall. The top row shows that the primary collapse occurs in a similar fashion for both bubbles, but following images indicate that there are some conspicuous differences during the rebound. The bubble with refrigerant gas creates a splash and rebounds almost uniformly in all directions while the air bubble shows a rebound clearly directed towards the solid wall. In addition, the air bubble produces a defined, mushroom-like shape after the second collapse, as can be seen in the last images, while the R1233zd-E bubble's shape remains rather arbitrary and omnidirectional.

An interesting aspect apart from the qualitative analysis is the quantitative value of the radius change. A time dependent equivalent radius R(t) (determined by counting dark pixels and assuming spherical symmetry) for both bubbles of fig. 4 is extracted from the video and shown in fig. 5. The radius is normalized by the equivalent radius R_0 at collapse initiation so that the figure allows to directly compare the evolution of two bubbles with different gas content under equal experimental conditions. The R1233zd-E bubble collapses to a slightly smaller minimum normalized radius and the subsequent rebound is not as strong as for the air bubble. While the first difference is rather small and can solely depend on inaccuracies in the radius estimation, the second observation confirms the impression of the qualitative analysis that differences in the rebound behavior exist. The air bubble rebounds to a larger normalized

radius than the vapor bubble and keeps a high amplitude for the oscillation during the completely observed time span. In contrast, the oscillation of the bubble filled with R1233zd-E seems to be dampened. A likely explanation is that the bubble undergoes partial condensation. Furthermore, a difference in oscillation time is observed that is caused by the different initial radii.



Figure 5: Temporal development of the equivalent radii of the R1233zd-E and air bubble for the experiment shown in fig. 4. Radii are normalized with the respective equivalent radii at collapse initiation.

As a final remark, the uncertainties of the preliminary workaround should be mentioned once more. The acrylic box filled with gelatin is placed in an existing non-ideal test section. The incident shock wave propagates much slower in the surrounding gas and interacts with the geometry of the box and the test section. This causes additional wave motion (visibly in fig. 4, frame 4, 7-10, 12) and a deformation of the acrylic box and both effects affect the bubble collapse. However, both bubbles undergo almost equal ambient conditions during the collapse, because the wave motion is much faster than the low-Mach number liquid flow.

Conclusion and outlook

First results prove that the concept of using a shock tube to create an instant pressure increase within gelatin is suitable to investigate bubble dynamics. Especially advantageous is the possibility that bubbles of arbitrary gas content at variable positions can be exposed to equal ambient conditions and analyzed simultaneously. For the shown experiment with an air bubble and a R1233zd-E gas bubble, a different behavior, notably concerning the bubble rebound, is indicated. A likely explanation is that partial condensation occurs whereas a complete condensation during the investigated collapse time is not observed.

To exclude the mentioned experimental uncertainties of the current workaround, further experiments will be conducted with a new test section throughout 2018 and presented at the conference. New experiments will feature a constantly high ambient pressure during the collapse and the collapse induced wave motion will be monitored by high frequency pressure measurements at distinct positions at the boundaries and near the bubble. To ensure a better comparability, these experiments will feature an equal bubble size and equal stand-off distance, allowing a more detailed analysis of the influencing parameters with a focus on the gas content. Furthermore, the non-Newtonian behavior of the gelatin and the influence on the bubble collapse will be analyzed via a separate test series with different pressures in the gelatin. The simple geometry and the well-defined initial and boundary conditions of the new configuration along with temporally highly resolved pressure distributions and schlieren pictures will provide extensive reference data to the community of bubble dynamics research. This renders a straightforward reproduction via numerical simulations possible and results can serve as numerical test cases for validation of computational models and methods.

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