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Untersuchung der Einfluss von Tensiden (Oberflächenaktive Stoffe) auf die Dynamik der akustischen Kavitationsblasen in Wandnähe mithilfe der Hochgeschwindigkeitsaufnahmen

Effect of surfactant concentration on the dynamics of oscillation of acoustic cavitation bubbles near a rigid surface using high speed photography

H. A. Vaidya[1, 3], Ö. Ertunç[1, 2, 3], T. Lichtenegger[1], J. Hachmann[1], A. Delgado[1, 3]
[1]Institute of Fluid Mechanics, University of Erlangen-Nuremberg, D-91058 Erlangen, Germany
[2]Department of Mechanical Engineering, Ozyegin University, 34662 Istanbul, Turkey
[3]Erlangen Graduate School in Advanced Optical Technologies (SAOT), D-91054 Erlangen, Germany

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Summary

In the present work, high-speed video imaging has been used to investigate the effect of surfactant concentration on the oscillation characteristics of the bubbles generated near a rigid surface due to acoustic cavitation. A gaseous bubble excited using ultrasonic waves oscillates at the exciting frequency and can undergo different types of oscillations, viz. stable or transient. In stable cavitation the oscillations last for multiple cycles before the bubble collapses. The bubbles undergoing transient cavitation, on the other hand, expand within couple of cycles and explode violently. The presence of surface active solutes in the medium has a profound effect on the stability and growth of the bubbles. The resistance to mass transfer provided by a layer of surfactant around the bubble is found to be one cause of enhanced rectification. During bubble expansion the surfactant molecules are less densely packed and as a result mass transfer is high. On the other hand, during the contraction, the mass transfer is reduced due to the dense packing of surfactant molecules. As a result the rectified diffusion is enhanced by the presence of a surfactant. Similarly their presence hinders bubble coalescence. The dynamics of the bubbles near a rigid surface shall be investigated using a high-speed camera with a frame rate of one million frames per second. The surfactant used is SDS (Sodium dodecyl sulfate) at different concentrations and the medium is water.

Introduction

Acoustic cavitation is a physical phenomenon which involves the formation of cavities in a liquid when a sound wave imposes a time-varying sinusoidal pressure on the steady ambient pressure. A liquid contains pre-existing nucleation sites where gas or vapor may be trapped. As a result of reduced pressure during the negative part of the cycle, these gas pockets grow and subsequently detach to form so called “cavitation” bubbles. Under the influence of an external sound field, these bubbles can grow either into “stable” bubbles which oscillate non-linearly around an equilibrium radius for many cycles or “transient” bubbles which expand to many times their original size within one pressure cycle and collapse violently. This peculiar nature of cavitation bubbles gives rise to interesting phenomena such as sonochemistry (Suslick 1990, Suslick et al. 1990), acoustic microstreaming (Elder 1959, Davidson and Riley

1971, Collis et al. 2010, Doinikov and Bouakaz 2010 Manasseh et al. 2010), molecular degradation, erosion (Naudé and Ellis 1961, Benjamin and Ellis 1966, Lindau and Lauterborn 2003; Tervo et al. 2004), etc.

Surface active materials are present in solutions in many ultrasonic applications. Surfactants are surface active agents which reduce the surface tension. Their presence in a liquid is expected to affect the interaction between individual bubbles (Lee et al. 2005). However this represents a very complex problem governed by extremely large number of parameters, which makes their modeling cumbersome (Segebarth et al. 2002) . As a result these parameters need to be determined experimentally and controlled accordingly. The knowledge of the effect of these parameters on the ultrasound field would help in optimizing the processes using ultrasound, such as ultrasonic cleaning (Apfel 1997, Krefting et al. 2004), medical ultrasound (Barnett et al. 1997), sonochemistry (Suslick 1990, Suslick et al. 1990), etc.

Most of the work reported in the literature focuses on the study of cavitation noise and its relation to the chemical yields, sonoluminescence intensities, etc. The acoustic cavitation field can be characterized using Sonoluminescence (Tronson et al., 2002, Lee et al., 2005) and Passive cavitation detection (PCD) (Atchley et al. 1988, Holland and Apfel 1990). Sonoluminescence (Suslick et al. 1990, Löfstedt et al. 1993, Barber et al. 1997) is the emission of light resulting due to bubble collapse, whereas PCD is the detection of the acoustic signals emitted due to bubble oscillations. However, to the best of our knowledge, high speed visualizations of oscillating bubbles in surfactant solutions has not been reported yet in literature. This would lead to quantitative information about the number of bubbles and bubble size distribution. Apart from that the oscillation characteristics can also be determined by this method.

In this study, sodium dodecyl sulfate (SDS), an anionic surfactant is used in varying concentrations. It is a commonly used surfactant and its surface properties have been thoroughly characterized. The effect of SDS concentration on the acoustic cavitation activity has been investigated by Tronson et al. 2002 and Ashokkumar et al. 1997. They have shown that addition of small concentration of SDS leads to increased SL activity. Segebarth et al., 2002, on the other hand have made use of the acoustic noise to find the effect of SDS concentration on the sonochemical yield of Peroxide. They correlated the width of the second harmonics of the acoustic signal with SDS concentration. Their experiments concluded that for SDS concentration up to 1mM increases the width of the second harmonic as well as the background noise around it decreases. However above 1 mM the second harmonic becomes large again. This trend continues up to 10 mM, after which no more changes occur. Thus it is clear that the presence of SDS in the solution does have an effect on the cavitation activity. There are many hypotheses which explain these effects, however they need to be quantified. Segebarth et al. 2002 have shown a clear-cut correlation between an acoustic parameter and measurements of sonochemistry and sonoluminescence. Similarly, Lee et al. 2005 too have shown a strong correlation between SDS concentration and SL activity and inertial cavitation. From these studies, they have come up with some possible explanations for the effect of surfactant concentration on cavitation activity.

One of the characteristics of any surfactant is its ability to adsorb at the air/water interface of the bubbles and decrease surface tension. Critical micelle concentration (cmc) is an indicator for the surface activity of any surfactant. For SDS, the equilibrium cmc is approximately 8 mM (Tajima et al. 1970). According to Segebarth et al. 2002 there is no correlation between surface tension and sonochemistry data.

Another hypothesis by Lee et al. 2005 is that the adsorption of surface active solutes on the bubble interface may inhibit coalescence. In case of charged surfactants like SDS, electrostatic repulsion between the charged head groups of surfactants also hinder bubble coalescence. It is this repulsion which results in an increase in the number of bubbles but a reduction in their average size. It is very likely that this may lead to a narrow size distribution. Bubble coalescence (Neppiras 1980, Agrawal 2013) and rectified diffusion (Hsieh and Plesset 1961, Strasberg 1961, Eller 1969, Crum 1980, 1984) are the two ways by which a bubble grows in size in an acoustic field. Thus, if the bubble coalescence is hindered by SDS, then the bubbles have to grow by rectified diffusion. This leads to a slower growth rate, which influences the bubble population and size distribution. Ashokkumar and Grieser 2007 have shown that this results in smaller size distribution in SDS solution. Since bubble coalescence is hindered due to the presence of SDS, the collapse of cavitation bubbles occurs at a relatively smaller size range. Crum (Crum 1980) has measured the growth rate of bubbles by rectified diffusion in a 22.2 kHz sound field as a function of surface tension of the liquid. He concluded that the addition of surfactant leads to substantial increase in the rate of growth of bubble due to rectified diffusion. In an attempt to come up with a suitable explanation to these phenomena, he suggested that the surface active monolayer may be responsible for the large growth rates observed in rectified diffusion. During expansion of the bubble, the surfactant molecules are sparsely distributed on the bubble surface, which allows more penetration of molecules, thus leading to increase in mass transfer. On the contrary, during contraction, the surfactant molecules cover the bubble surface densely, which creates a resistance to mass transfer. As a result the resultant mass flux from the liquid to the bubble is higher than in the absence of surface active layer. This explanation is in agreement with the theory of rectified diffusion.

Similarly, Lee et al. 2005 have observed that in case of multibubble cavitation, the cavitation intensity observed in the presence of surfactant is substantially higher than pure water. They propose that the electrostatic repulsion between bubbles causes them to separate and thus the bubble cluster becomes more open. This leads to increased penetration of ultrasound in the cluster.

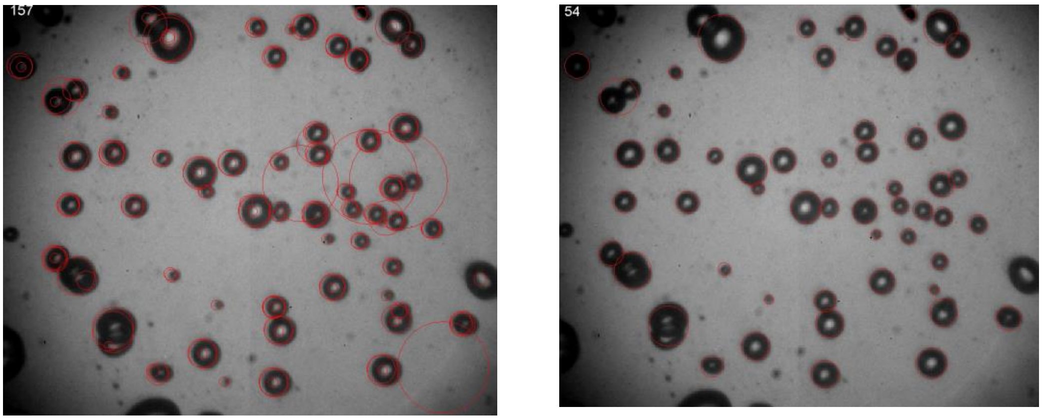
The studies available in the literature correlate the effect of the presence of surfactants to the acoustic activity by means of indirect methods mentioned above, like SL intensity, acoustic noise, sonochemical yield, etc. Although it yields quantitative data about the cavitation activity in general, not much information about the bubble size and size distribution can be obtained. Moreover, being an extremely complex process governed by many parameters, it cannot be easily modeled. In order to address this problem, the present study is concerned with the high speed visualization of cavitation bubbles in the presence of surfactant in water. The current work is a part of an ongoing project to study the dynamics of acoustic cavitation bubbles near a rigid surface. This has already been explained elsewhere (Vaidya et al. 2013). The visualizations of the cavitation bubble cluster created near a rigid surface have been carried out at one million frames per second.

Experimental Setup

The visualization experiments are carried out using a high speed IS-CCD camera (Shimadzu Corporation, HPV-2) capable of up to one million frames per second with a resolution of 312 X 260. This high speed implies that the time for which the shutter is open can be as short as 1µs. A long distance microscope (W.D = 23.5 mm) consisting of infinity corrected objective (Mitutoyo Plan Apo 20X), infinity corrected zoom lens (Navitar, Inc., Ultrazoom 6.5X) and F-Mount adapter tube (2X, Navitar, Inc.) is used with the camera. Such short time scales demand extremely powerful light source for reasonable illumination. A super pressure short arc mercury lamp (200 W) light source fitted with a 320 nm – 700 nm filter is used for illumination. This light source is coupled to a liquid light guide and a collimating lens. A cylindrical piezoelectric Langevin-type transducer dia = 25.4 mm and height = 26 mm) with a resonance frequency of 75 kHz has been used in the current work. The detailed experimental setup has been explained elsewhere (Vaidya et al. 2013). SDS special purity grade (98%) was obtained from Sigma Aldrich. Ultrapure water (Biochrom AG, Germany) at 293 K was used for the experiments.

Image processing

The images obtained from the high speed camera need to be evaluated for bubble size distribution and number of bubbles. Similarly the oscillation characteristics of the cluster need to be determined. This requires intensive image processing, which is made cumbersome by the large amount of data produced. Numerous problems are encountered while evaluating the images. One main problem is the uneven background illumination. This arises due to the presence of large number of bubbles in the cluster. The long distance microscope used here has a depth of focus 3.5 µm, which implies that there are bubbles on either side of the focal plane. These bubbles scatter the light falling on the focus plane which makes the illumination highly inhomogeneous. Some regions of the image are dark, whereas the others are over illuminated. This hindrance is overcome by carrying out the intensity thresholding of the image, where the histogram of the image is equalized. After leveling the illumination, the next task is to identify the bubbles which are in the focal plane. This is done by using the Hough transform (Duda and Hart 1972; Illingworth and Kittler 1988), which is a method of detecting complex pattern of points in an image data (Hough 1962). This is achieved by determining specific values of parameters which characterize these patterns. Spatially extended patterns are transformed so that they produce spatially compact features in a space of possible parameter values. The HT converts a difficult global detection problem in image space into a more easily solved local peak detection problem in a parameter space (Illingworth and Kittler 1988). In the present study the image processing has been carried out using Matlab® and the Hough transform code written by Peng 2005 has been used for bubble detection to detect circles on grey scale images. This algorithm works with a gradient based algorithm for edge detection and is capable of handling bubbles with distorted circular shapes. However, an inherent disadvantage of this method is that it detects many false bubbles, as shown in Figure 1a. In order to overcome this problem, a systematic sorting algorithm is implemented. The algorithm looks for the center detected by the Hough transform and measures the intensity within the circle. Simultaneously the intensity is measured in a circle with the same center but thrice the radius. If the difference in intensities of the two circles is less than the threshold value, then the circle is selected, otherwise it is dropped, as shown in Figure 1b.



(a) Before intensity thresholding

(b) After intensity thresholding

Fig. 1: Bubble detection using Hough transform and intensity thresholding

Data evaluation and correlation

The acoustic measurements in the set-up have been carried out using a pre-calibrated TC 4038 broad band miniature probe hydrophone (Teledyne-Reson A/S, Denmark) coupled to a VP1000 pre-amplifier (Teledyne-Reson A/S, Denmark). The data from the hydrophone was recorded using an oscilloscope SDS-200A (softDSP Co., Ltd.) at a real-time sampling rate of 50 MS/sec. Thus the the Nyquist sampling rate criterion for 75 kHz is met easily and the pressure data is recorded every 20 ns. The active element measures 4 mm in diameter, which is much smaller than the wavelength of sound to be used (20 mm).
Each frame obtained by image processing as described above, possesses information about the location and size of the cavitation bubbles. The next step is to correlate the image data with the pressure data in order to examine the oscillation characteristics of the bubble cluster. Due to the fact that camera and hydrophone are triggered simultaneously, each image corresponds to a specific phase of the sine wave. In order to correlate the acquired images and the pressure information obtained from the hydrophone, it is necessary to trigger them at the same time. The entire process is executed as illustrated in Figure 2.

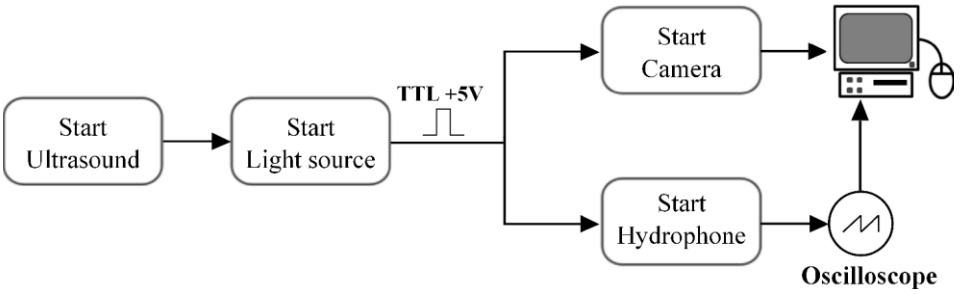
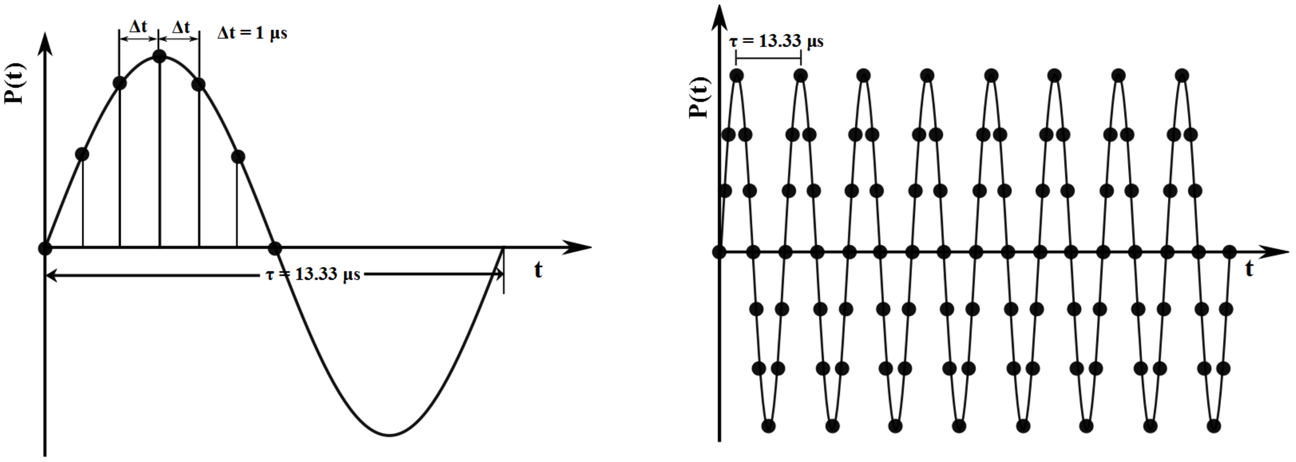


Fig. 2: Process execution

As mentioned earlier, the hydrophone records pressure data every 20 ns, whereas an image is recorded every 1 µs. Thus every 50th pressure data corresponds to a single image. After triggering, the camera records 100 images in one sequence, which corresponds to 100 µs. At a frequency of 75 kHz, the incoming ultrasound wave has a period (τ) of 13.33 µs. Thus in one sequence approximately eight ultrasound cycles are captured. This has been demonstrated in Figure 3a. Similarly, Figure 3b shows one period of the incident pressure wave. The solid circles indicate the instants at which the images are captured. The corresponding pressure data is obtained from the hydrophone measurements. Thus each image has a pressure amplitude and phase value associated with it. This proves to be vital in determining the oscillation characteristics of the acoustic cavitation bubbles, since there is perfect synchronization between the pressure or phase data and images.



(a) Time instants of the incoming ultrasound wave (f_0 = 75 kHz) where the images are captured, as shown by the solid circles. The time between two successive dots is 1 μs

(b) One acoustic pressure cycle and the corresponding instants where the images are captured. The period is 13.33 μsand the time between two recordings is 1 μs.

Fig. 3: Correlation of the high-speed recordings with the corresponding pressure data.

Results and discussions

For each visualization experiment 96 images were obtained and the time between two successive images is 1 μs. Using Hough transform, for each image Hough transform was used and the position of center of each bubble and the radius of the bubble were calculated. By means of an optical scale, the correlation between pixel data and actual data was calculated and it turned out that 45 pixels on the image corresponded to 100 μm. The two important parameters for the evaluation of individual images are the bubble volume and number of bubbles. For each image the volume of all the bubbles are summed up. As mentioned earlier each image can be correlated with a particular plase. The total bubble volume and number of bubbles for a very narrow phase interval of three degrees is evaluated. This means that among all the video sequences, the volume of all the bubbles in images which fall in a certain phase interval is summed up. The total number of bubbles are calculated in a similar fashion.

Figure 3 is the variation of total bubble volume as a function of SDS concentration. It can be clearly seen that in the absence of SDS, period doubling(A. Eller and Flynn 1969, Esche 1952, Lauterborn and Cramer 1981, Neppiras 1980, J. T. Tervo) takes place. The curve in red has twice the period of the incident sound wave. In the presence of SDS, as mentioned above, the bubble coalescence is hindered due to the electrostatic repulsion. As a result the bubbles are unable to coalesce and thus cannot reach the resonance size (W Lauterborn 1969, Lauterborn and Kurz 2010; Minnaert 1933) in this manner. Another possibility is that the bubbles grow by rectified diffusion, which in turn is slower. However in the absence of SDS the bubbles can freely coalesce and reach the resonance size and even beyond. Period doubling suggests that bubbles with size twice the resonance size are present in the system. However the presence of SDS in the solution prevents the formation of bubbles twice the resonant size. Moreover, for all the three SDS concentrations, a similar trend is observed. Another important observation, which can be made from Figure 3 is that the total bubble volume in the absence of SDS is much higher than when SDS is present in the system. The bubble volume is dominated by the larger bubbles, which are generated greater in number in the absence of SDS. The presence of SDS prevents the bubble coalescence and hence instead of large bubbles, many bubbles with smaller size are formed. These, although plenty in number, do not have much influence on the total volume. Thus from Figure 3 is in agreement with the observations of (Ashokkumar and Grieser 2007; Lee et al. 2005; Segebarth et al.

2002) that the presence of SDS leads to large number of small bubbles. This is well reflected in the bubble volume data shown in Figure 3

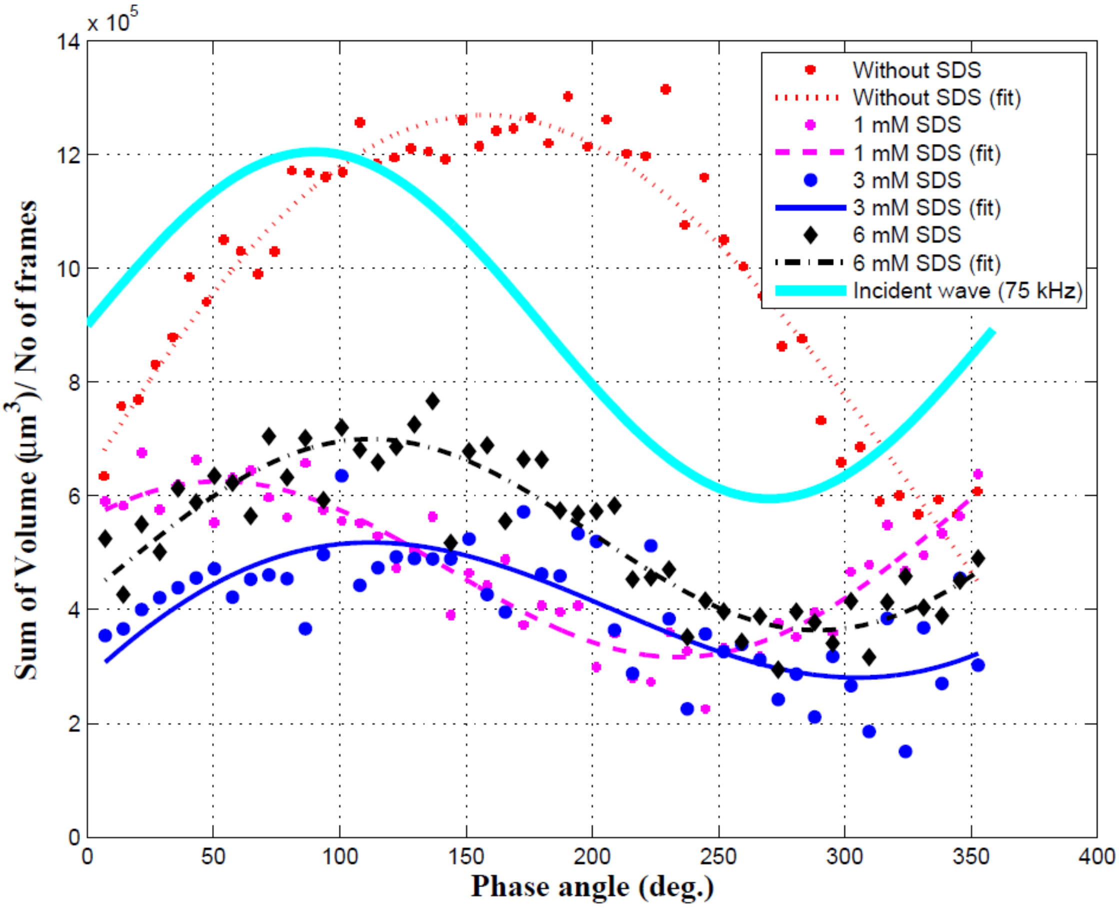


Fig. 4: Variation of the total volume with the phase angle

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