19th Australasian Fluid Mechanics Conference
Melbourne, Australia
8-11 December 2014

Single Bubble Sonoluminescence and Bubble Surface Stability in Surfactant Solutions

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Abstract

The radial dynamics of a single sonoluminescing bubble has been investigated in surfactant solutions. Experimental results show that an increase in the surfactant concentration leads to a decline in the oscillation amplitude and hence light emission intensity. Numerical simulations support this result, showing that under the driving pressures required to achieve single bubble sonoluminescence (SBSL), the surface properties, namely the surface elasticity and dilatational viscosity, contribute to the damping of the radial amplitude in the bubble oscillation. In most cases this stabilises the bubble surface, but leads to a decreased light intensity due to smaller oscillation amplitude. The application of a stronger driving pressure in an attempt to produce equivalent light emission to a surfactant-free bubble, leads to a decrease in the surface stability, making it practically very difficult for a bubble to achieve high SBSL intensities in concentrated surfactant solutions. Although the bubble oscillates at a smaller amplitude, the instability mechanism for a surfactant-coated bubble at higher ambient radii and surfactant concentrations, is more likely to be of the Rayleigh-Taylor type than that of a clean bubble at the same given acoustic parameters. This can lead to bubble disintegration before correcting mechanisms can bring the bubble back into the stable SL regime.

Introduction

A single bubble levitated in a standing wave can, under specific conditions, experience nonlinear pulsations that result in light emission. This phenomenon is called single bubble sonoluminescence (SBSL) [6]. A bubble smaller than the resonance size injected into a standing wave field, will be drawn towards the pressure antinode due to the *Primary Bjerknes* force [3, 7]. The light intensity emitted by this pulsating bubble, is dependent on various factors that include the amount and type of dissolved gases in the liquid [20], the frequency of the applied ultrasound [2], the applied sound pressure amplitude, hydrostatic pressure and addition of particular solutes [1, 14, 16, 19].

Surface active solutes, i.e., surfactants, have been shown experimentally to influence the behaviour of SBSL. Ashokkumar et al. [1] showed that micromolar concentrations of non-volatile surfactants such as sodium dodecyl sulphate (SDS), dodecyl trimethyl ammonium chloride (DTAC) and decyl ammonium propane sulfonate (DAPS) did not significantly affect the dynamics or SL of a single bubble. Numerical simulations performed by Yasui [18] explained that the effect of the surfactant was to inhibit the condensation of water vapour at the bubble wall during bubble collapse which lowers the achievable temperature inside the bubble.

The shape stability of a bubble is another important consideration. Instabilities arise from perturbations of the surface during oscillation that disrupt the spherical shape of the bubble such that the curvature of the liquid becomes non-uniform and form a local surface tension pressure associated with each point of the surface [8]. Under stable conditions, these perturbations are dampened and the bubble returns to its equilibrium condition (spherical). However, sometimes, dramatic overshoot can occur which can propagate over a large number of cycles (parametric instability), leading to experimentally observed phenomena such as shape mode oscillations [15]. In some cases, dramatic oscillations occur at the point of a strong bubble collapse and persist only for a single cycle (Rayleigh-Taylor instability) which may cause a bubble to move chaotically (dancing motion) and to pinch-off daughter bubbles [5] or to disintegrate completely, as the bubble usually does not have enough time to correct the strong perturbation to its surface. In surfactant rich environments, the bubble surface will have viscoelastic properties that may dampen or enhance the shape stability.

The effect of higher concentrations of surfactant on SBSL, has recently been studied by Leong et al. [10] experimentally and theoretically. This conference presentation will report on the findings of this work.

Materials and Methods

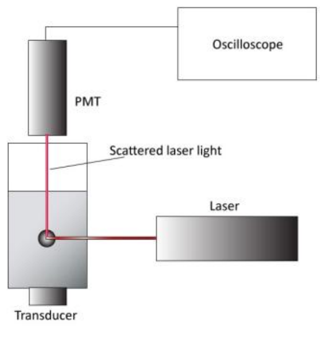

Figure 1. Set-up for measuring the SBSL intensity and radial dynamics from Leong et al. [10] Copyright (2014) by the American Physical Society.

Further details of the experimental setup and methods can be found in Leong et al. [10] The surfactants used in the SBSL experiments were of the purest grades available: Sodium dodecyl sulphate (SDS) (VWR international, purity>99%) and dodecyl trimethyl ammonium chloride (DTAC) (TCI Japan, purity>99%). Sodium chloride (NaCl) was supplied by Merck Germany (purity>99.5%).

The same apparatus set-up as detailed for single-bubble rectified diffusion experiments by Leong et al. [9] was used with minor adjustments in these experiments (figure 1).

A vacuum pump was used to partially degas the solution.

To determine the bubble's radial dynamics, light emitted from a low power laser diode (633 nm) was directed at the bubble and its scattered intensity was measured using a photomultiplier tube (PMT) (Hamamatsu E849-35 amplified by Canberra H.V. Supply Model 3002). The same PMT was also used to measure the sonoluminescence intensity. The PMT signal was relayed to an oscilloscope (LeCroy WaveSurfer 452) and an average over 50 sweeps was taken.

A driving pressure between 1.1 to 1.3 bar and frequency of between 22.23 and 22.31 kHz were used.

The maximum bubble radii were determined using images processed in ImageJ. The minimum bubble radii could not be determined from the images taken. Instead, an approximate R_{max}/R_{min} ratio was determined from the data obtained by the oscilloscope for the reflected laser light, which was plotted in Matlab for further analysis. The estimated R_{min} in this case, was in the order of 5 µm radius.

Equations

The equation of motion used to calculate the radius of a bubble in an acoustic field is a modified Keller Equation adapted from Yasui [17]:

$$\left(1-\frac{\dot{R}}{c_\infty}+\frac{\dot{m}}{c_\infty\rho_L}\right)R\ddot{R}+\frac{3}{2}\dot{R}^2\left(1-\frac{\dot{R}}{3c_\infty}+\frac{2\dot{m}}{3c_\infty\rho_L}\right) = \frac{1}{\rho_L}\left(1+\frac{\dot{R}}{c_\infty}\right)\left[p_B-p_s\left(t+\frac{R}{c_\infty}\right)-p_\infty\right] + \frac{\ddot{m}R}{\rho_L}\left(1-\frac{\dot{R}}{c_\infty}+\frac{\dot{m}}{c_\infty\rho_L}\right) + \frac{\dot{m}}{\rho_L}\left(\dot{R}+\frac{\dot{m}}{2\rho_L}+\frac{\dot{m}\dot{R}}{2c_\infty\rho_L}\right)+\frac{R}{\rho_L}\frac{dp_B}{dt} \quad (1)$$

Here time derivatives are denoted by a dot with R the radius, $\dot{m}$ the net rate of evaporation (or condensation) of water vapour in the bubble, c_∞ the speed of sound in the bulk (1483 m/s), ρ_L the bulk liquid density (1000 kg/m^3), p_s the acoustic field defined as $P_a cos(\omega t)$ where P_a is the acoustic driving pressure and ω is the angular frequency, and p_∞ the static pressure.

The liquid pressure on the external surface of the bubble is $p_B(t)$ and is related to $p(t)$ by [12]:

$$p_B(t) = p(t) - \frac{2\sigma(R_o)}{R} - \frac{4\mu}{R}\left(\dot{R}-\frac{\dot{m}}{\rho_L}\right) - \dot{m}^2\left(\frac{1}{\rho_L}-\frac{1}{\rho_G}\right) - 4\chi\left(\frac{1}{R_0}-\frac{1}{R}\right) - \frac{4\kappa_s}{R^2}\left(\dot{R}-\frac{\dot{m}}{\rho_L}\right) \quad (2)$$

where, χ is the surface elasticity, κ_s is the surface dilatational viscosity and μ is the bulk liquid viscosity.

The instabilities caused by small distortions of the spherical interface are modelled by equation (3) [13]:

$$R_D = R(t) + a_n(t)Y_n(\theta,\phi) \quad (3)$$

where R_D is the bubble radius distorted by bubble oscillations, $R(t)$ is the instantaneous bubble radius governed by the radial time behavior of the oscillating bubble (i.e. the solution to equation (1)), Y_n is a spherical harmonic of degree n with $n \geq 0$ and a_n is the radial distortion amplitude for mode n.

The aim is to determine $a_n(t)$, the radial distortion amplitude, which can be used to determine the stability of a bubble. In response to an initial radial perturbation, a stable bubble will have $a_n(t)$ always smaller than $R(t)$ and converging to a finite value, whereas an unstable bubble will have $a_n(t)$ diverge to a value larger than $R(t)$.

The value of $a_n(t)$ is determined by solving the second order ordinary differential equation (ODE),

$$\ddot{a}_n + B_n(t)\dot{a}_n - A_n(t)a_n = 0 \quad (4)$$

for $n \geq 0$.

Loughran et al. [11] presents equations for $A_n(t)$ and $B_n(t)$ as follows:

$$A_n = (n-1)\frac{\ddot{R}}{R} - \frac{(n-1)(n+1)(n+2)\sigma(R_o)}{\rho_L R^3} - \left[\frac{2\mu\dot{R}}{\rho_L R^3}\right]\left[(n-1)(n+2) + 2n(n+2)(n-1)\frac{\delta}{R}\right] - \frac{2n(n+2)\dot{R}\mu^s}{(n+1)R^4\rho_L}\left(n(n+1)+(n-1)\right) - \frac{2\kappa_s\dot{R}}{(n+1)\rho_L R^4}(n^2(n+2)(n+1)+7n^3 + 9n^2-n-4) - \chi\left(\frac{1}{R^3\rho}(n+2)(n^2+4n+2) + \frac{2(R-R_o)}{(n+1)\rho_L R^4}(n^2(n+2)(n+1)+7n^3 + 9n^2-n-4)\right) - G^s\left(\frac{n(n+2)}{(n+1)R^3\rho_L}(n(n+1)-2) + \frac{2n(n+2)(R-R_o)}{(n+1)R^4\rho_L}\left(n(n+1) + (n-1)\right)\right) \quad (5)$$

$$B_n = \frac{3\dot{R}}{R} + \frac{2\mu}{\rho_L R^2}\left[(n+2)(2n+1) - 2n(n+2)^2\frac{\delta}{R}\right] + \frac{n(n+2)\mu^s}{(n+1)R^3\rho_L}(n(n+1)-2) + \frac{\kappa_s}{(n+1)R^3\rho_L}(n+2)(n+1)(n^2+4n + 2) \quad (6)$$

Here ρ_L is the density of the liquid and δ is the diffusive boundary layer thickness around the bubble approximated by

$$\delta = \min\left(\sqrt{\frac{\eta}{\rho_L\omega}}, \frac{R}{2n}\right) \quad (7)$$

These expressions include the effect upon the bubble stability of the surface elasticity (χ), the surface dilatational viscosity (κ_s) as well as the equilibrium surface tension $\sigma(R_0)$ and the bulk liquid viscosity (μ). Loughran et al. [11] indicate that these terms are appropriate for describing simple outward expansion of the shell but do not describe the shell bending and flexing. They include the surface shear viscosity (μ^s) and the surface shear modulus (G_s) to account for this bending and flexing.

The mode n = 2 is considered since it is the least stable mode of oscillation, and a_2 is solved using Euler's Equation. Stability thresholds were determined by trial and error calculation with a minimum of 5 oscillation periods used (in some cases >30 oscillation periods were required) to judge the convergence/divergence of the plotted radial distortion amplitude.

Results and discussion

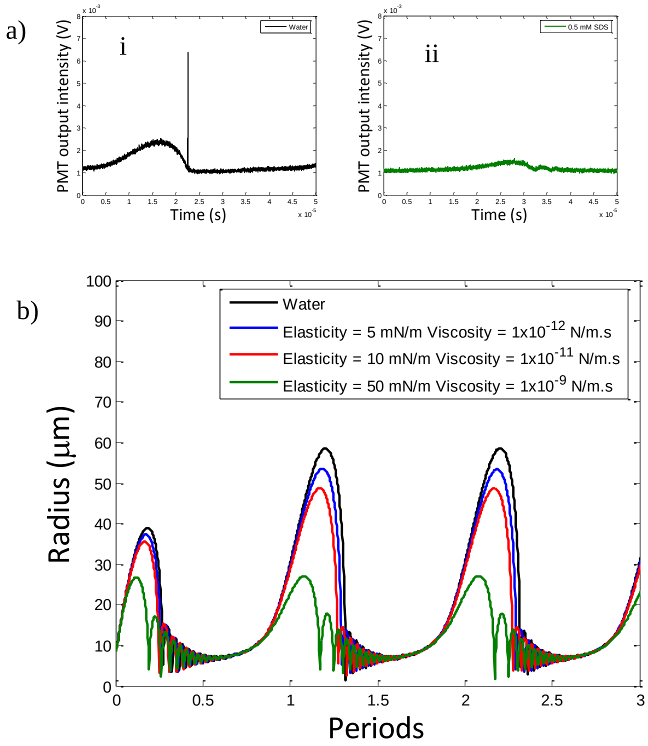

Figure 2. a) Bubble radius as a function of time during SBSL for a bubble in i) water ii) 0.5 mM SDS. b) Decrease in oscillation amplitude with increasing surface elasticity and viscosity determined from numerical calculations. Driving frequency of 20.6 kHz, Pa = 1.3 bar and R_0 = 10 μm are used. Equilibrium surface tension of 73 mN/m used for water and a constant value of 68 mN/m for the surfactant-coated bubble (estimated concentration range from ~ 0.01 mM to 0.5 mM SDS) Figures taken from Leong et al. [10] Copyright (2014) by the American Physical Society.

Figure 2 (a) shows typical PMT outputs for the radial oscillation amplitude of a bubble in water and 0.5 mM SDS solution. The sharp peak associated at the point of collapse is the SBSL emission. Note that with the addition of surfactant, the bubble oscillation amplitude is reduced. The consequence is a 'quenching' of the light emission with higher surfactant concentration, since lower maximum radial amplitude (R_{max}) is achieved. The likely reason for this lower amplitude in the presence of surfactant is the contribution of the bubble surface elasticity and surface dilatational viscosity, which restricts the oscillation amplitude of the bubble as shown in the calculated bubble radius during bubble oscillation (figure 2 (b)).

In general, the damping of radial oscillations due to an enhanced surface elasticity and/or viscosity will lead to a stable bubble surface. The surface stability threshold for a bubble in pure water and one in a surfactant solution (~ 4.0 mM SDS) is shown in figure 3. As can be seen, the presence of surfactant results in a 'more-stable' bubble surface evidenced by an increased stability threshold.

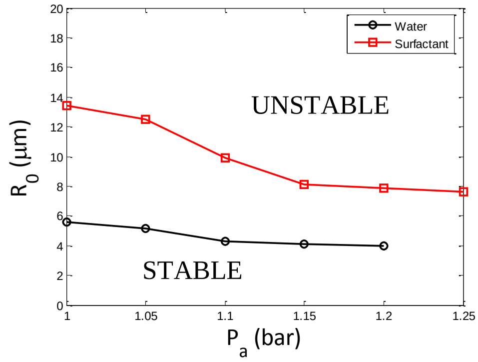

Figure 3. Stability thresholds of the n=2 mode for a surfactant-coated bubble with surface elasticity = 100 mN/m and surface dilatational viscosity of 1 x 10^{-10} N/m.s compared with pure water. A surface tension of 50.0 mN/m (~ 4.0 mM SDS) is used for the surfactant-coated bubble and 73.0 mN/m is used for the clean bubble. In the case of the surfactant-coated bubble, G_s and μ_s are estimated to be 25% of the surface elasticity and viscosity respectively. An acoustic driving frequency of 20.6 kHz is used. Figure taken from Leong et al. [14] Copyright (2014) by the American Physical Society.

Despite a more stable bubble surface however, the ability for a surfactant-coated bubble to achieve SBSL is reduced. This is demonstrated in the numerical calculations presented in figure 4.

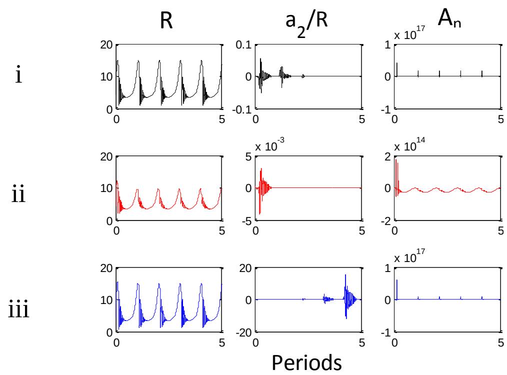

Figure 4. Radius, radial distortion amplitude and A_n as a function of oscillation period of a bubble in i) water driven at 1.1 bar and surfactant solution driven at a pressure of ii) 1.1 bar and iii) 1.208 bar. For the surfactant solution, the equilibrium surface tension = 68 mN/m, surface elasticity = 10 mN/m and surface dilatational viscosity = 1x10^{-11} N/m.s (~ 0.05 mM SDS). An equilibrium surface tension of 73 mN/m is used for water. For all cases, the driving frequency is 20.6 kHz and an ambient radius of 4.25 μm is used. Figure taken from Leong et al. [10] Copyright (2014) by the American Physical Society.

Here, an equivalent driving pressure of 1.1 bar is used for a bubble in water and a surfactant solution ~0.05 mM SDS (surface tension 68 mN/m, surface elasticity 10 mN/m, surface dilatational viscosity 1x10^{-11} N/m.s). Note that these surface properties are lower in magnitude to those used in the calculations for figure 3, i.e. lower surfactant concentration, so the bubble has a lower stability threshold. The radial oscillation of the bubble in the surfactant solution achieves a lower R_{max} compared with the clean bubble. The ratio of the radial distortion amplitude of the bubble to the bubble radius in both water and surfactant are less than unity in both cases, indicating a stable bubble surface.

An increase of the driving pressure to 1.208 bar allows the surfactant-coated bubble to experience a similar oscillation amplitude to the surfactant-free bubble. However, when this occurs, the ratio of the radial distortion amplitude to the bubble radius increases above unity, indicating bubble instability.

The implication is that forcing the surfactant-coated bubble to expand in amplitude by increasing pressure (to increase the SBSL intensity), will cause the bubble to become unstable. This has been observed experimentally by strong dancing motion during this present study.

It is clear that the higher driving pressure, required to achieve an equivalent radial amplitude to that observed in water, causes the surfactant-coated bubble to become shape-unstable. However, in this case the instability is parametric (i.e. several periods of oscillation occur before the radial distortion amplitude exceeds unity), and so there is likely to be sufficient time (~5-10 cycles) for corrective mechanisms to bring the bubble into a regime where it can emit SBSL. One of the possible mechanisms for this correction is the re-adjustment of the ambient radius by enhanced diffusion during the non-spherical period of the bubble oscillation [4]. This demonstrates that SBSL emission is possible in the presence of less concentrated surfactant concentrations, as shown by workers in earlier experimental studies [1]. At higher surfactant concentrations (results not shown), the instability mechanism is more likely to be of a Rayleigh-Taylor type, which can cause the bubble to enter into a chaotic state or to disintegrate completely.

Conclusions

The radial oscillation of a sonoluminescing single bubble decreases in the presence of a surfactant. This decrease in oscillation also results in a decrease in emitted light intensity. The viscoelastic properties of the surfactant layer contribute to these effects. A numerical study of the behaviour of the bubble surface stability including the effect of surface viscoelasticity shows that in the 20 kHz frequency region, the presence of the surfactant reduces the oscillation amplitude and is the dominating effect that leads to a more stable bubble. However, this in turn reduces the SBSL intensity, and makes it practically more difficult to attain strong SBSL emission in concentrated surfactant solutions.

It should be noted that our analysis assumes that neither shell buckling nor rupture occurs with such soluble surfactants. Further work is required to confirm whether this indeed is the case.

Acknowledgments

The authors would like to acknowledge the Postgraduate Overseas Research Experience Scheme awarded by the University of Melbourne, the Research Travel Conference Assistance Scheme awarded by the School of Engineering (The University of Melbourne), and the Travel Scholarship award by the Particulate Fluids Processing Centre (The University of Melbourne), which provided the funds that enabled this work to be performed. The National Institute of Advanced Industrial Science and Technology Nagoya is also acknowledged for hosting the work.

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