Effect of Power and Frequency on Bubble-Size Distributions in Acoustic Cavitation

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Acoustic bubble-size distributions have been determined using a pulsed ultrasound method at different ultrasound powers and frequencies. It was observed that the mean bubble size increased with increasing acoustic power and decreased with increasing ultrasound frequency. It was also recognized that the mean size of bubbles emitting sonoluminescence was greater than those producing sonochemiluminescence indicating that the two processes take place in different populations of cavitation bubbles in the system.

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Introduction.—Acoustic cavitation is the phenomenon observed when ultrasound of sufficient intensity is transmitted through a liquid causing micron-sized gas bubbles to oscillate, grow, and violently implode giving rise to extreme, but localized, conditions within the collapsed cavities (extremely high temperatures and pressures) [1,2]. Such conditions are primarily responsible for the chemical effects associated with acoustic cavitation in liquids [3,4].

Under certain conditions bubble collapse can also result in light emission, sonoluminescence (SL), originating from the core of the bubble during the final stages of collapse. The sonochemically active region and time period, however, are considerably broader [5]. Therefore, SL studies yield information that is limited both spatially and temporally with respect to sonochemistry. A more indicative method of quantifying the sonochemical activity of cavitation bubbles is by measurement of the sonochemiluminescence (SCL) from luminol solution [6,7]. Recent studies indicate that SL and sonochemistry originate from different regions of a sonochemical reactor [8] and in certain cases from bubbles of different sizes [9].

An important parameter of a multibubble cavitation field is the size distribution of bubbles that can undergo inertial collapse to produce SL and/or sonochemistry. There have been several recent studies [10–17] investigating the cavitation bubble size and size distributions quantitatively using different techniques, with the majority of work conducted at 20 kHz. However, to date, there has not been a comprehensive study examining the effect of ultrasound frequency on the cavitation bubble-size distribution. To address this, the effect of ultrasonic frequency and also the effect of the applied acoustic power on the bubble-size distribution in water have been examined. In addition, this work provides a comparison between the size distributions of SL emitting and sonochemistry producing cavitation bubbles.

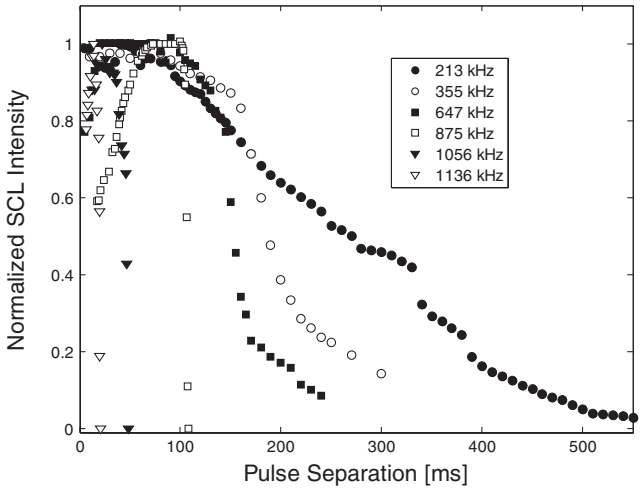
Experimental details.—Luminol (Sigma-Aldrich) solutions were prepared with distilled water and sodium hydroxide (pH $\approx$ 11). Distilled water was used for SL measurements. The sample volume was 200 ml and fresh solutions, saturated with air, were used for each experiment.

Two different ultrasound arrangements were used in this study. Both employed a Hameg function generator (model HM8131-2) triggered by an external pulse generator (Datapulse 100A). In the first arrangement, a T&C Power Conversion, Inc. amplifier was used and connected to an Allied Signal transducer. Two transducers were used: one with resonances at 213 and 647 kHz and the other with resonances at 355 and 1056 kHz. In the second arrangement, a Meinhardt amplifier and transducer (575, 856, or 1156 kHz) were used. Light emission was measured using a Hamamatsu photomultiplier tube and LeCroy oscilloscope (WaveSurfer 452).

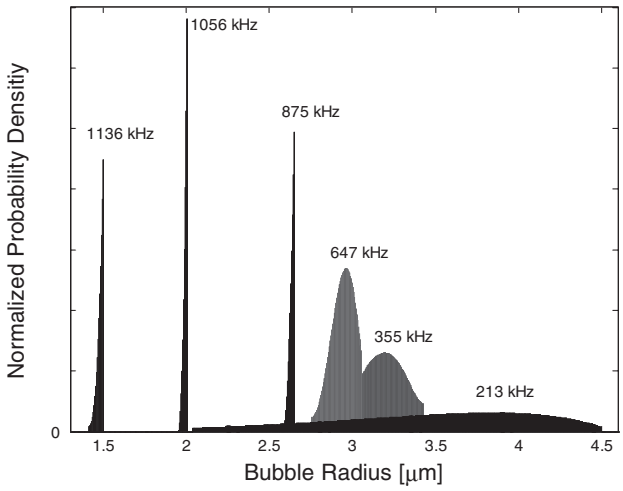
A 4 ms ultrasound pulse width was chosen based on the observation that at short pulse separations this width resulted in the greatest SL-SCL signal. Experiments were performed by measuring the steady-state SL-SCL intensities at different pulse separation times. SL was measured from distilled water (no luminol present). SCL was measured from an alkaline luminol solution. The intensity of luminol SCL emission was several orders of magnitude greater than SL emission, so any SL contribution to the SCL signal could be considered insignificant.

The acoustic power delivered was determined calorimetrically. For the study of the effect of frequency and for the comparison of SCL and SL data, the acoustic power was set slightly above the threshold for detectable emission (in the range of 1.5–3.0 W). The technique used to determine the bubble-size distribution is based on the dissolution of cavitation bubbles in a pulsed sound field and has been described elsewhere [13].

Results and discussion.—In order to determine the size distribution of the cavitation bubbles at different frequencies, the SCL intensity was measured as a function of pulse separation for a constant pulse width of 4 ms and acoustic power of about 1.5 $\pm$ 0.4 W for various ultrasonic frequencies. The observed results are shown in Fig. 1(a). The intensities reported have been normalized with respect to the maximum intensity obtained at each respective frequency. The absolute intensities at 355 kHz were much greater than at 1136 kHz, for example, but it is the relative change in intensity with pulse separation that is of interest in this experiment.



(a)



(b)

FIG. 1. (a) Normalized sonochemiluminescence intensity from luminol as a function of pulse separation (with a constant pulse time of 4 ms) and (b) the bubble-size distributions for 213, 355, 647, 875, 1056, and 1136 kHz. In (b) the data for 875, 1056, and 1136 kHz have been scaled down by a factor of 4. The acoustic power of all frequencies in (a) and (b) is 1.5 ± 0.4 W. Values for 1136 and 1056 kHz had a high accuracy with a standard deviation of less than 5%, whereas the size determined at lower frequency (213 and 355 kHz) had a standard deviation of about 25%.

First, it can be noted that the SCL intensity increases with an initial increase in the pulse separation times. Similar behavior was observed for SL and SCL at all frequencies. This was found to occur with or without preconditioning the solution by sonicating prior to pulse sonication (1 min high-power continuous mode sonication followed by 4 min silent) and for different methods of measurement (using fresh solutions for each measurement or reusing the same solution). We also note that this behavior was not observed by Lee *et al.* [13]. This might be due to the different experimental conditions (different transducer, cell geometry, volume, power, etc.) used in the former study. We speculate that the intensity behavior we observed might be related to partial degassing under very short pulse separation conditions resulting in a lower number of active bubbles. With an initial increase in the pulse off time, the extent of degassing may decrease. Tuziuti *et al.* [18] have shown that a decrease in degassing increases the acoustic amplitude. This would eventually increase the number of active bubbles leading to an increase in the cavitation activity. However, at longer pulse off times, the dissolution of the bubbles would dominate leading to a decrease in the SL-SCL intensity. In Fig. 1(a), it can be clearly observed that at each frequency, the SCL intensity decays as a function of the pulse separation. This occurs over a relatively long time period for the lower frequencies and over a relatively shorter period for the higher frequencies.

Equation 1 in [13] was used to convert the pulse separation times into bubble radii. The fit of the experimental radii data was then used to develop a histogram of the relative bubble population [19]. The resulting bubble-size distributions calculated are shown in Fig. 1(b). It should be noted that the radii calculated from our experimental data refer to the initial radii of the bubbles (that can be grown to a maximum size by the acoustic field) prior to expansion and then bubble collapse.

Mean bubble radii and the full width at half height (FWHH) determined in this study are presented in Table I in conjunction with the theoretical bubble radii as calculated using the Minnaert equation [20] and those from Yasui's investigation [21], along with other relevant experimentally determined values. One of the key observations that can be made is of the relatively smaller cavitation bubble sizes compared with the theoretically predicted values (linear resonance size). Yasui's numerical calculations of equilibrium bubble radii at three frequencies (20 kHz, 140 kHz, and 1 MHz) yield values that are more consistent with the experimental data [21].

Considering the data shown in Fig. 1(b), two features are apparent: first, as the ultrasound frequency increases it is quite clear that the mean bubble size (mean of the distribution) becomes smaller, and the distribution itself becomes narrower with increasing frequency [22].

In addition to the active bubble size decreasing with increasing acoustic frequency, Yasui [21] has also predicted that the size range of SL producing bubbles should be narrow, which is precisely what we observe in the present investigation for SCL producing cavitation bubbles.

The effect of acoustic power on the size of sonochemically active bubbles is shown in Fig. 2 for sonication at 1056 kHz. It is clear that the mean size increases as a function of acoustic power up to a limiting value of about 4.5 μm. This experiment has been conducted at other frequencies, but reproducibility was found to be greatest at 1056 kHz. Nevertheless, the general trend of increasing

TABLE I. Theoretical and experimentally determined cavitation bubble radii at different acoustic frequencies.

Frequency (kHz)	Linear resonance radius (μm)[a]	Theoretical radius (μm)[b]	Mean measured bubble radius and FWHH (μm)[c]	Previous experimental values (μm)
20	150	0.1–100	···	3–25[d]; 2–5[e]
140	21	0.1–10	···	···
213	14	···	3.9 (1.6)	···
355	8.5	···	3.2 (0.6)	···
515	5.8	···	···	2.8–3.7[f]
647	4.6	···	2.9 (0.2)	···
875	3.4	···	2.7 (0.02)	···
1000	3.0	0.1–3	-	···
1056	2.8	···	2.0 (0.04)	···
1100	2.7	···	···	0.9, 1.4[g]

[a]Calculated using Minnaert's equation: $R_{\mathrm{res}} \approx 3/f$.
[b]Reference [21].
[c]Experimental results of this study.
[d]Reference [11].
[e]Reference [15].
[f]Reference [13].
[g]Reference [12].

size with power is also observed at the other frequencies. The observed trend with acoustic power is consistent with previous studies of single-bubble systems [23,24]. Multibubble studies suggest, however, that due to stronger nonlinear pulsations at higher acoustic power, the radius should decrease with an increase in acoustic pressure [9]. This, however, may not come into effect over the relatively low power range of this study. Experimentally, Lee [25] observed no change in radius with a 50% change in acoustic power.

Figure 3(a) shows the SL and SCL intensities as a function of pulse separation at 575 kHz (acoustic power 2.5 ± 0.5 W). It can be seen that the SL signal decays at longer pulse separations compared with the SCL signal and also that the rate of decay is more rapid. These data were used to calculate the bubble-size distributions shown in Fig. 3(b). It is clear that the SL bubbles and SCL bubbles belong to distinctly separate populations, with the SL bubbles being considerably larger in size and confined within a narrower range. It should be noted that these experimental conditions were chosen to accentuate this difference between the two populations. Under different frequency and power conditions this difference was found to be less pronounced and at higher frequencies (i.e., 1056 kHz) not noticeable at all. Such narrowness in the size of SL bubbles was observed at all frequencies (data not shown), in contrast to a relatively broad range of SL bubbles (2.8–3.7 μm) that has been observed previously [13] at 515 kHz. Theoretical work predicts a much broader size range for SL bubbles than we observe at the frequencies used in this study [9,21], and also that the width of the size range increases with increasing acoustic power. It should be noted that the acoustic power used by Lee *et al.* [13] was an order of magnitude greater than that of this study. In our system, it is the SCL bubbles which exhibit a broad size distribution, at 575 kHz, as is evident in Fig. 3(b) and also at 213, 355, and 647 kHz [Fig. 1(b)].

The temperature requirements for sonochemical reactions and SL are quite different. Sonochemistry (bond dissociation) requires only modest bubble temperatures whereas SL (ionization) will only occur under much hotter core conditions. Sonochemistry will therefore occur well below the threshold temperature for SL emission, which can be attained by smaller cavitation bubbles or bubbles undergoing pronounced asymmetric collapse. It may be extrapolated from this study that the relatively larger SL

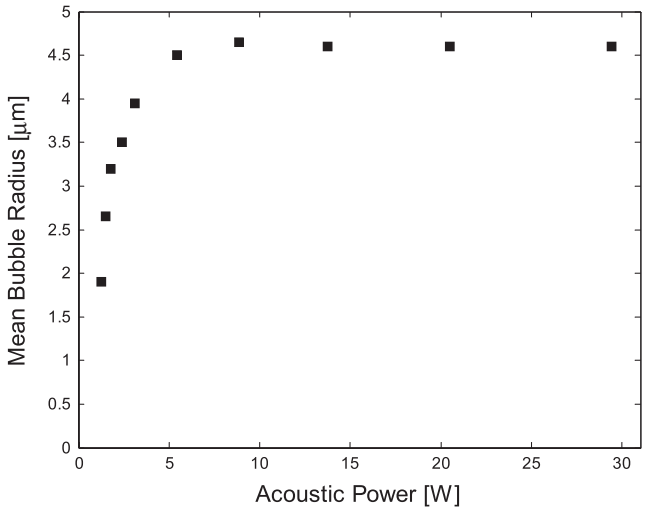


FIG. 2. Bubble radii (mean of the size distribution) under 1056 kHz sonication as a function of acoustic power.

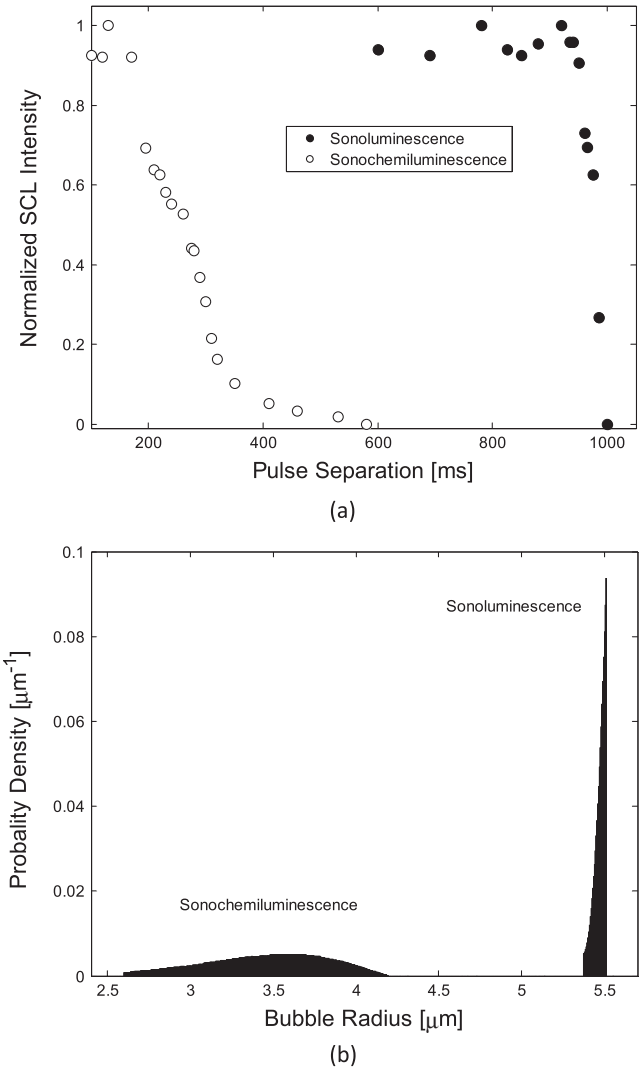


FIG. 3. (a) Normalized sonochemiluminescence intensity from luminol and sonoluminescence intensity as a function of pulse separation (with a constant pulse time of 4 ms) at 575 kHz and (b) the bubble-size distributions determined from the sonochemiluminescence and sonoluminescence data. The acoustic power in (a) and (b) was 2.8 ± 0.5 W.

emitting bubbles are attaining a higher temperature compared with the SCL emitting bubbles.

Conclusions.—Bubble-size distributions of SCL emitting bubbles have been determined using a pulsed ultrasound technique as a function of both acoustic frequency and power. It was found that the bubble size increased with increasing power and decreased with increasing frequency. In addition, a comparison was made between the size distributions of SL and SCL bubbles. The results of this investigation show that two distinct classes of bubbles exist: the larger one producing SL, the smaller one producing sonochemistry.

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