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Initial growth of sonochemically active and sonoluminescence bubbles at various frequencies

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ABSTRACT

The initial growth of acoustic cavitation activity is important in some applications such as therapeutic and diagnostic medicine. The initial growth of cavitation activity has been investigated using sonoluminescence and sonochemical activity (sonochemiluminescence) at 358 kHz, 647 kHz and 1062 kHz and at 5 W, 15 W and 30 W applied power levels. The growth of sonochemically active bubble population is found to be much faster than that of sonoluminescence bubble population at 358 kHz and 647 kHz whereas almost similar growth rate is observed at 1062 kHz for both bubble populations. This suggests that the cavitation bubble resonance size ranges of sonoluminescence and sonochemically active bubbles are different at 358 kHz and 647 kHz, whereas they have similar size range at 1062 kHz. At 358 kHz and 647 kHz, relatively smaller bubbles become chemically active. Possible reasons for such observations have been discussed. The data presented and discussed in this study may be useful in controlling the growth of cavitation bubble population in addition to enhancing the knowledge base in cavitation science.

1. Introduction

Acoustic cavitation has been increasingly used in several applications [1–3]. Syntheses of nanomaterials [4,5], biomaterials [6,7], sonoprocessing [8,9] and therapeutic medicine [10] are applications some that could be highlighted. While the efficiency of these processes depends upon the overall cavitation activity, the initial growth of cavitation activity is crucial in some applications. For example, pulsed acoustic field is used in diagnostic and therapeutic medicine [11–14] to control the growth and intensity of cavitation activity. The development of active cavitation bubble population at the initial stages of sonication is important in such applications. For example, in imaging applications, minimisation of cavitation activity is necessary to reduce any health risks associated with acoustic cavitation. It is known that cavitation process generates intense chemical and physical effects [15]. It has been shown in previous studies that an induction time is necessary to initiate cavitation and grow the active bubble population to a steady-state in an acoustic field [16–18].

Often, sonoluminescence (SL) or acoustic emission is used to characterise and determine cavitation intensity/activity [19,20]. It has been also shown that sonochemical activity could occur in the absence of sonoluminescence [21]. In addition, the detection of acoustic emission from cavitation bubbles needs special transducers. The homolysis of water molecules within collapsing bubbles due to high temperature conditions leads to the generation of highly reactive redox radicals. It has been suggested that a relatively lower temperature within the cavitation bubbles could lead to sonochemical activity whereas significantly higher temperatures are needed for SL to occur. Sonochemical activity (quantified in terms of sonochemiluminescence, SCL) could occur at much lower acoustic power levels compared to that required for SL activity [21]. It could be seen in Fig. 1 that a very small region of the cavitation bubbles sonoluminesce whereas significantly larger proportion of the cavitation bubbles are sonochemically active under the same experimental conditions. The SL and sonochemical activity are correlated to the size of cavitation bubbles. Sonochemically active bubbles are shown to be relatively smaller compared to SL bubbles [22].

Henglein and coworkers and our group have reported on the initial growth of SL and SCL [16–18]. However, a direct comparison between the initial growth of SL and SCL bubble populations has never been reported. As evidenced from Fig. 1, SCL activity provides relatively more accurate information on cavitation activity. The main aim of the current work is to expand the knowledge base on acoustic cavitation by studying the initial growth of SL and SCL

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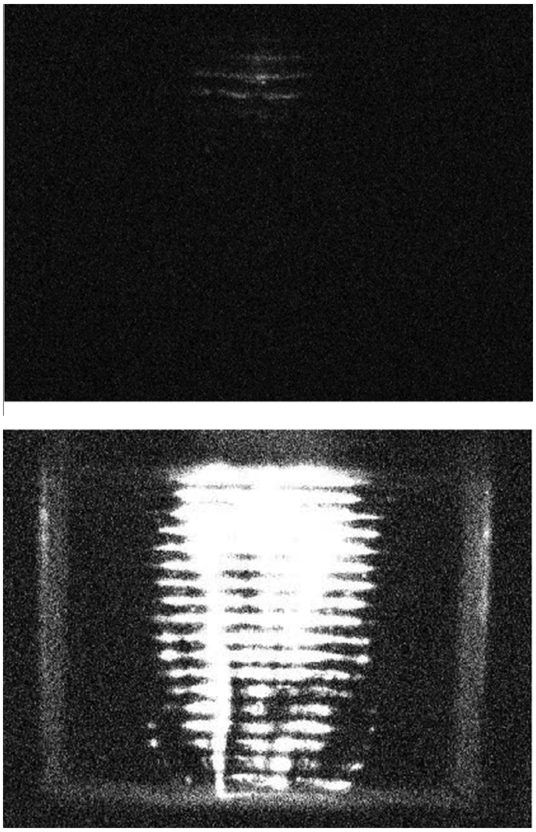

Fig. 1. Spatial distribution of sonoluminescence and sonochemically active cavitation bubbles at 170 kHz at 8 W applied power (Top: SL; Bottom: SCL) [21].

bubble population, which may have relevance to various applications including diagnostic and therapeutic medicine.

2. Experimental details

Analytical grade sodium hydroxide and luminol were from Sigma–Aldrich. High-purity Milli-Q filtered water was obtained from a Millipore system (18.2 MΩ cm at 25 °C). An ELAC (Germany) ultrasonic generator/transducer system was used in all the experiments. A cylindrical Pyrex reaction cell filled with 200 mL solution was mounted over a plate transducer. The reaction cell together with an end-on photomultiplier tube were kept in a light-shielded housing. The generator was modified in-house to allow for pulsed acoustic energy delivery where both pulse on and off time could be varied. For this work, on and off times of the acoustic pulse "train" were fixed at 4 ms and 12 ms, respectively – these conditions were chosen since similar conditions were used in previous studies [17,22,23]. Previous experiments [18] have shown that the transmission of electrical pulse into acoustic pulse is instantaneous and hence the observed initial growth pattern is not related to the Q-factor of the transducer. A variation in pulse on or off time would affect the number of pulses required to reach a steady state population reported in this study – however, the overall trend in terms of frequency and power effects may not be affected. For this reason, further experiments varying the pulse on–off times were not performed. Three different ultrasound frequencies, viz., 358 kHz, 647 kHz, 1062 kHz at 5 W, 15 W and 30 W applied power were used in this experiment. 1 mM aqueous NaOH solution containing 0.1 mM luminol was used to measure SCL intensity. High-purity Milli-Q filtered water was used to measure SL intensity. Air-saturated water/aqueous solutions were used in all experiments. Individual growth measurements took less than a few seconds and hence degassing did not occur. In other words, the gas concentration was not affected by the measurement procedure. In addition, the solutions were pre-conditioned by sonicating for 2 min followed by 5 min resting time to maintain the gas concentration at the same level independent of initial gas concentration. Such procedures were known to give reproducible results based on our previous work [18,22]. The SL and SCL intensities were recorded using a photomultiplier tube (PMT) in a dark enclosure. PMT used had a response in the wavelength range 300 nm–600 nm. We used cells that were made of Pyrex. Hence, PMT signals correspond mostly to visible region. However, it should be noted that we are looking at relative signals. The PMT was positioned at the same place collecting emission from the entire liquid. The PMT signal was transmitted to an oscilloscope for display and the data were stored on a computer. All SCL and SL experiments were repeated at least 3 times for reproducibility check.

3. Results

Fig. 2 shows the initial growth of SL and SCL growth observed at 647 kHz at various power levels.

There are several interesting observations that could be made:

- As in previous studies [16–18], it takes several acoustic pulses before steady-state populations are reached for both SL and SCL. The total SL/SCL intensity is a function of photons emitted by each bubble and the total number of bubbles. Hence, the total intensity observed could be correlated to the "SL/SCL bubble population".
- The relative number of acoustic pulses required to reach a steady-state SL intensity is higher than that required to reach steady-state SCL intensity at all power levels and the difference in the number of pulses required becomes less obvious at higher power levels. This is clearly demonstrated in Figs. 2 and 3.
- With increasing acoustic power level, the number of pulses required to reach the steady state decrease with an increase in acoustic power for both SL and SCL as shown in Fig. 3. It should be noted that there is no difference in the number of pulses required between 15 W and 30 W for SCL.
- With increase in acoustic power, the steady state intensity increases for both SL and SCL. The steady state SCL intensity is about 30–50 times greater than that of SL for the power levels used (Table 1).
- The rate of growth of SL bubble population is low compared to that of SCL bubble population. For better clarity, ratio between intensity of 10th pulse to steady-state intensity could be used. At 5 W, this ratio for SL is about 16 whereas SCL ratio is about 2. In other words, at 5 W, about 50% SCL population (to that of steady state) is reached in 10 pulses whereas only about 6% SL population is reached for the same number of pulses (also see Table 1 for the ratios observed at other power levels and other frequencies).

It should be noted that there seems to be some fluctuation when a steady-state is reached, in particular at low power levels (Fig. 2). SL and SCL bubbles are located in different regions of a reactor as shown in Fig. 1. In a given system, both standing and travelling waves exist. The strength of travelling waves could be stronger closer to the transducer. Since most SL bubbles are away from the radiating source, they may not be influenced by the travelling waves to the same extent to SCL bubbles. This could be the reason for the observed fluctuations in steady-state intensity for SCL bubbles.