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Short communication

Enhancement and quenching of high-intensity focused ultrasound cavitation activity via short frequency sweep gaps

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ABSTRACT

This letter reports on the use of frequency sweeps to probe acoustic cavitation activity generated by high-intensity focused ultrasound (HIFU). Unprecedented enhancement and quenching of HIFU cavitation activity were observed when short frequency sweep gaps were applied in negative and positive directions, respectively. It was revealed that irrespective of the frequency gap, it is the direction and frequency sweep rate that govern the cavitation activity. These effects are related to the response of bubbles generated by the starting frequency to the direction of the frequency sweep, and the influence of the sweep rate on growth and coalescence of bubbles, which in turn affects the active bubble population. These findings are relevant for the use of HIFU in chemical and therapeutic applications, where greater control of cavitation bubble population is critical.

The high temperatures and pressures generated within cavitation bubbles gave birth to the phenomenon of sonochemistry, a unique class of high-energy chemistry [1,2]. Unlike the traditional high-energy chemistry such as radiation and laser chemistry, sonochemistry is capable of stimulating chemical and biochemical processes by generating high-energy conditions localised on a microscopic scale [3,4]. The most desired effect of ultrasound is to increase efficiency by enhancing cavitation activity for sonochemical reactions (e.g., degradation of organic pollutants) and sonoprocesses (e.g., emulsification and extraction) without increasing power input [4,5]. One method of concentrating energy is to use a high-intensity focused ultrasound (HIFU) which allows acoustic energy to be focused, and even without cavitation the absorption of the acoustic energy generates tremendous heating at the focal point. This has allowed HIFU to find success in medical and therapeutic applications such as non-invasive device for localised necrosis of diseased tissue [6], hemostasis [7], drug delivery [8,9] and imaging [8].

Although cavitation can be both beneficial and detrimental [10], it is generally undesired in some medical applications. For this, HIFU research is mainly focused on restricting or suppressing cavitation at the focal region by methods of dual frequency [10–12] and over-pressurisation [13,14] to alter cavitation thresholds; and wave form manipulation such as pseudorandom signals [15] and frequency sweeps [16,17] to disrupt the standing wave patterns. However, Bailey et al. [10] hypothesised that the attenuation in cavitation activity by linear sweeps (from 250 kHz to 290 kHz) is caused by bubbles not being able to oscillate in resonance during the sweep. Others [18–20] have probed the resonance response of bubbles by monitoring the radial oscillation of shelled microbubbles that has been subjected to either a pulse of “upward” or “downward” frequency sweep for a given frequency range. The focus of these studies is to increase signal to noise ratio to improve contrast agent imaging and the results indicated that “downward” frequency sweep is more efficient when the transmitted frequency is chosen above the resonance frequency of the interrogated microbubbles. In these studiesa much lower acoustic power, below the threshold of acoustic cavitation, was used.

In this study, the acoustic power is operated above the cavitation threshold where cavitation bubbles, unlike the shelled microbubbles, undergo complex bubble dynamics involving bubble growth, dissolution, coalescence, and inertial collapse to emit sonoluminescence and chemical reactive species. A similar “upward” and “downward” frequency sweep is employed to probe the cavitation bubble dynamics, except in this work the same starting frequency is used to generate the initial bubble population. To distinguish this difference, we have denoted the different sweep

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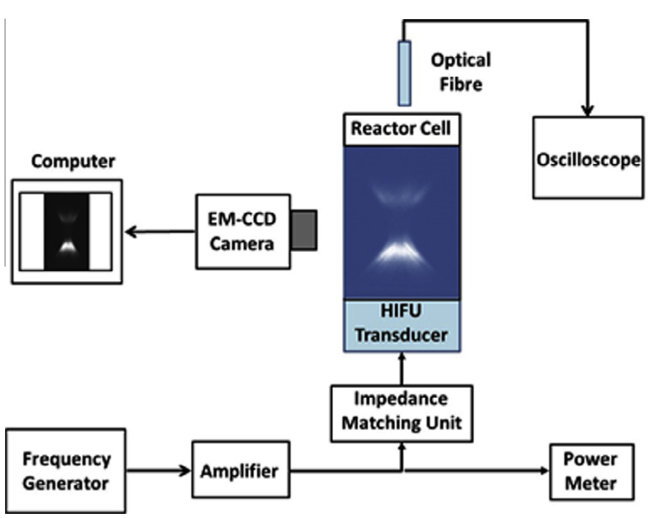
direction as "negative" and "positive" frequency sweep and varied the sweep rate. This letter will demonstrate the unprecedented enhancement and quenching of the inertial cavitation activity, as well as the sonochemical yield, by altering the direction of the sweep. The results provide an alternative means to enhance sonochemical activity without compromising input power for the development of energy efficient sonochemical reactors [21] and other industrial applications such as ultrasonic processing of dairy systems [22], wastewater treatment [23] and surface treatments [24].

Scheme 1 depicts the experimental setup where a 4 MHz HIFU (IMASONIC), with 40 mm focal length, is positioned at the bottom of a cylindrical reactor. The excitation signal of the HIFU transducer is generated by a Function Waveform Generator (33500B, Agilent Technology) and amplified by an Amplifier (150A100B 10–100 MHz, Amplifier Research). The forward and reflected powers delivered to the HIFU were measured by a wattmeter (NRVD Rhode & Schwartz). A cooled EM-CCD camera (iXon 885, Andor Technology) is positioned perpendicular to the transducer to capture the spatial distribution of the sonochemiluminescence (SCL) and the total integrated intensity of the images for a given exposure time was then evaluated. The total SCL intensity was also measured using an optical fibre linked to a photomultiplier tube (PMT) (H10721-210 Hamamatsu), and the signals were recorded by an oscilloscope (Lecroy WaveSurfer 44Xs, 400 MHz, 2.5 GS/s) and post-treated with Matlab®. Complementary measurements were performed with a cavitometer ICA 3 MHz from the University of Minsk.

Conventional frequency sweeps are performed in the positive direction with large frequency ranges varying from 20% to 20,000% of the starting frequency [10,25]. This renders the system inefficient considering the performance of transducers is usually limited by a small bandwidth. In this study, the wave form of the frequency sweeps conducted is depicted Fig. 1. The starting frequency was fixed at 3.6 MHz and the stop frequency was varied in both positive and negative direction, with a frequency sweep range between 0% and 5% of the starting frequency. These frequency ranges fall within the bandwidth of the transducer, which is between 3.4 and 3.8 MHz as shown in Fig. 2. 500 mL of solution was used for all experiments and presonicated for 2 min at 3.6 MHz to ensure all the initial solution condition is the same, i.e., to ensure there is a consistent number of initial population of stable cavitation nuclei. After the presonication, ultrasound was stopped and the frequency sweep was conducted from a rest period and therefore the first point of cavitation generation will be the



Scheme 1. Schematic diagram of the experimental setup.

first f_{start}. To verify that the results are not influenced by any variations in temperature or in dissolved gas concentration, SCL was checked for a continuous sonication at a single frequency of 3.6 MHz before and after the frequency sweep. A continuous sonication was applied and unless stated otherwise, the sweep time and power absorbed by the insonated solution (measured by calorimetry) was fixed at 1 ms and 17 W, respectively. The acoustic pressure was measured using a hydrophone and confirms that, if the focal location moves during the sweep mode, the pressure level remains at the same range of magnitude. Cavitation activity was quantified by sonochemiluminescence (SCL) intensity, generated using Luminol solution (1 mM of 3-aminophthalhydrazide and 0.1 M NaOH), and potassium iodide (KI) dosimetry, carried out by sonicating 0.1 M of KI. The concentration of I_3^- formed was determined by measuring the adsorbance of I_3^- at 355 nm (ε = 26,303 $dm^3\,mol^{-1}\,cm^{-1}$) every 5 min for 30 min.

Fig. 3 shows the enhancements in both the SCL intensity and I_3^- formation by a factor of two when a short negative frequency sweep range is performed. Further increase in the frequency sweep range longer than 0.1 MHz decreases the sonochemical yield even though the frequency range is still within the bandwidth of the transducer which yields 93–100% of the maximum SCL as shown in Fig. 2. Alternatively, if the frequency sweep is performed in the positive direction, the sonochemical activity is quenched, as reported by Bailey et al. [10]. Sonoluminescence intensity as a function of the sweep frequency range was also measured and exhibited similar relative changes as a function of the frequency sweep (data not shown). One may notice an important difference between SCL and I_3^- measurements at frequency sweep around −0.15 and −0.18, which may be attributed to experimental reasons such as difference in test duration: the luminol emission is from the fluorescence decay and the intensity collected is the integration of the spatial distribution of SCL captured in 1 s exposure whereas I_3^- formation is evaluated over a period of sonication (30 min). Nevertheless, the global curve pattern is beyond question.

It is hypothesised that the observed effect is related to the variations in the resonance bubble size, which decreases with increasing frequency [26] and the time required to grow the bubbles to the resonance size [27]. It is well known that bubbles should reach a resonance size range [28] prior to growing to a maximum size followed by collapse [29]. During the positive frequency sweep, bubbles generated by the starting frequency are too large (larger than the resonance size range) to act as bubble nuclei for the subsequent sweep frequencies, thus resulting in the observed quenching of the SCL activity. Conversely, when the sweep is performed in the negative direction, bubbles generated by the starting frequency can act as cavitation nuclei. The size of nuclei generated at the start frequency is large enough to be grown to the resonance size within a short frequency range (0.1 MHz). However, as the frequency range increases, the rate of change of frequency within 1 ms is too fast for the bubble nuclei to grow to the resonance size and the SCL activity attenuates. Although it is assumed that the switch from f_{stop} to $f_{start,}$ is instantaneous, it is possible that bubble fragmentation can occur at the f_{stop} to produce daughter bubbles which can further act as cavitation nuclei for the subsequent f_{start}.

To confirm the effect of the frequency sweep rate, the sweep time was increased for two extreme frequency sweep ranges in which attenuation in the SCL was observed in Fig. 3. The SCL intensities are then plotted in terms of absolute frequency sweep rate (Fig. 4). It can be seen in Fig. 4 that there exists two correlations between the cavitation activity and the sweep rate, distinguished by the direction of the sweep, and independent from the frequency range. For all positive frequency sweeps, the SCL decreases with increasing sweep rate. On the other hand, when a negative frequency sweep is applied there exits an optimum frequency