Sonoluminescence, sonochemistry (H$_2$O$_2$ yield) and bubble dynamics: Frequency and power effects

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Abstract

In the present work, comprehensive experimental and numerical investigations on the effects of ultrasound frequency and acoustic power on sonoluminescence (SL) and H$_2$O$_2$ yields have been carried out. The multibubble SL and H$_2$O$_2$ yields have been examined for four frequencies (213, 355, 647 and 1056 kHz) and over a wide range of acoustic powers. The observed experimental results have been discussed with respect to single bubble dynamics and the number of active cavitation bubbles.

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Keywords: Sonoluminescence; Multibubbles; Bubble dynamics; Acoustic cavitation

1. Introduction

The formation, growth and the violent collapse of bubbles, under the influence of ultrasound, is known as acoustic cavitation [1]. Extreme temperature and pressure conditions are associated with acoustic cavitation and thus it has shown applicability in almost every field of science and engineering, namely, medicine, chemistry, biotechnology, etc. Cavitation bubbles, due to the very high temperatures generated at the final stages of bubble collapse, emit light which is known as sonoluminescence (SL). Multibubble sonoluminescence (MBSL) was discovered in 1933 by Marinesco and Trillat [2]. The SL had caused fogging of a photographic plate immersed in a liquid irradiated by ultrasonic waves. Frenzel and Schultes [3], in 1934, documented MBSL as the light emission from cavitating liquids. Single bubble sonoluminescence (SBSL) was first reported by Gaitan and Crum [4]. In their experimental investigation, a standing wave was generated in a liquid medium and a single bubble was trapped at the pressure antinode because of the radiation force known as Bjerknes force [5] and the mass transport of gas across the liquid/bubble interface.

Theoretical and experimental investigations of both SBSL and MBSL are related to sonochemistry, as both SL and sonochemical reactions originate from the high temperature conditions inside the collapsing bubbles. Recently, a substantial amount of work has been carried out on acoustic cavitation. Some of these studies include the investigation of the mechanism of SL [6–8], the effects of different experimental conditions on SL [9–18], and instrumentation and modeling aspects of cavitation [19,20]. A thorough understanding of the effect of different operating parameters on cavitation and/or SL is very important in order to enhance sonochemical reaction efficiencies and to develop large scale sonochemical reactors.

There are still many processes involved in the production of SL that are unresolved. Nevertheless, it has been acknowledged that SL probably involves a combination of blackbody radiation [21], bremsstrahlung [22] and excited state emission [23]. On the basis of numerical simulations, Yasui [24] has pointed out that the origin of SL emission depends on the acoustic frequency. He has
suggested that the light emission at 1 MHz is mainly plasma emission, whereas light emission at 20 kHz is due to both plasma emission and chemiluminescence. Many previous investigations have reported on the effect of variations in different experimental parameters on SL intensity and sonochemical efficiency [12–16,24–28].

Ashokkumar and Grieser [29] have discussed in detail the phenomenon of SL quenching by solutes in a liquid. They have proposed that the SL quenching by solutes is due to a reduction of the core temperature within a bubble as a consequence of the evaporation of the solutes from the bubble–liquid interface, followed by the endothermic reactions within a bubble during bubble collapse.

Beckett and Hua [30] have performed an experimental investigation of ultrasound frequency and its role in controlling sonochemical yield and MBSL intensity. They found 358 kHz to be an optimum frequency for observing a maximum SL intensity and the highest chemical reaction rates. They concluded that nonlinear bubble implosions play a major role at lower frequencies, whereas higher flux rates (mass transport) of solutes and radicals influence chemical reactivity at higher frequencies.

The main aim of the present work was to examine MBSL and sonochemistry under different ultrasound frequencies at varying acoustic power levels. A number of previous studies [30–33] have attempted to explore the effect of ultrasound frequency on cavitation events. However, the combined effect of frequency and acoustic power has not been previously investigated. Also, most of the previous studies in the field of sonochemistry and/or SL, either theoretical or experimental, have been carried out over a limited range of operating parameters. In the present work, a comprehensive assessment of the frequency and acoustic power effects on SL and sonochemical activity under the same experimental conditions has been carried out. The experimentally obtained results have been explained on the basis of numerical calculations based on single bubble dynamics and the number of active cavitation bubbles in the system.

### 2. Experimental details

#### 2.1. Sonoluminescence (SL) measurement

Purified water (100 or 200 mL) was used as the cavitating medium to investigate the effect of varying frequency and acoustic power on the SL intensity. The ultrasonic waves of 213, 355, 647 and 1056 kHz were generated by an ELAC RF-generator and delivered through plate-type allied signal transducers (irradiating surface area of 24 cm²) that were operated at the above-mentioned frequencies. A Pyrex reaction cell was mounted over the transducer. The SL intensity was measured with the use of a Hamamatsu end-on photomultiplier tube (PMT), which captured the SL signal and transferred it to an oscilloscope, where the intensity, in terms of mV, was read off the display. In order to avoid the initial and variable effect of dissolved air in water, the solution was exposed to 30 s of operating frequency ultrasound irradiation (pre-treatment). The solution was allowed to equilibrate for 2 min prior to each measurement. This procedure ensured reproducibility and consistency in the sonication experiments. For each reading, the displayed signal was averaged over 256 pulses and measured once a steady-state level had been reached, typically in less than 30 s. For each measurement, to maintain accuracy, five different readings were noted and an average of the readings was taken as the final reading. The temperature of the sonication medium was maintained at approximately 25 °C (±2 °C) using external cooling. The acoustic power delivered to the cavitating medium was estimated calorimetrically.

#### 2.2. Hydrogen peroxide yield measurement

Purified water (100 mL) was irradiated for 10 min at the same operating conditions as that used for the measurement of SL. After each irradiation, the yield of H₂O₂ was measured using an UV–Vis absorption spectrophotometer (Shimadzu), using standard methodology [34]. The iodide

### Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>c</td>
<td>velocity of sound in the medium (m/s)</td>
</tr>
<tr>
<td>f</td>
<td>frequency of first ultrasound source (kHz)</td>
</tr>
<tr>
<td>I</td>
<td>intensity of irradiation (W/m²)</td>
</tr>
<tr>
<td>P₀</td>
<td>ambient pressure (N/m²)</td>
</tr>
<tr>
<td>P₁</td>
<td>initial pressure inside the bubble (N/m²)</td>
</tr>
<tr>
<td>Pₚₘᵦₓ</td>
<td>maximum radius of the bubble (m)</td>
</tr>
<tr>
<td>Rₘᵦₓ</td>
<td>initial radius of the bubble (m)</td>
</tr>
<tr>
<td>Rₘᵦₓ</td>
<td>minimum radius of the bubble (m)</td>
</tr>
<tr>
<td>P₀</td>
<td>ambient pressure (N/m²)</td>
</tr>
<tr>
<td>P₁</td>
<td>resultant time varying pressure field due to two waves (N/m²)</td>
</tr>
<tr>
<td>Pᵦ</td>
<td>vapour pressure (N/m²)</td>
</tr>
<tr>
<td>P₟</td>
<td>driving pressure amplitude of ultrasound (N/m²)</td>
</tr>
<tr>
<td>P∞</td>
<td>pressure in the surrounding liquid (N/m²)</td>
</tr>
<tr>
<td>R</td>
<td>radius of cavity/bubble (m)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Greek letters</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>γ</td>
<td>specific heat ratio (Cₚ/Cᵥ)</td>
</tr>
<tr>
<td>ρ</td>
<td>density of the liquid medium (Kg/m³)</td>
</tr>
<tr>
<td>µ</td>
<td>viscosity of liquid (Ns/m²)</td>
</tr>
<tr>
<td>σ</td>
<td>surface tension of liquid (N/m)</td>
</tr>
</tbody>
</table>
Reagent was prepared by mixing equal volumes (1 mL) of solution A (0.4 M KI, 0.05 M NaOH, 0.00016 M (NH₄)₆Mo₇O₂₄•4H₂O) and solution B (0.1 M KHC₈H₄O₄). One milliliter of the sonicated sample was added in the iodide reagent, and after mixing, absorbance of I₂ was measured spectrophotometrically using e = 26,400 M⁻¹ cm⁻¹ at 353 nm.

3. Simulation methodology

When ultrasound passes through a liquid medium, the time-dependent variation in the pressure field can be described by the following equation [5]:

\[ P_t = P_0 - P_a \sin(2\pi ft), \]

where \( P_0 \) is the ambient pressure, \( P_a \) is the pressure amplitude of the ultrasound wave and \( f \) is the frequency of ultrasound. The pressure amplitude, \( P_a \), can be estimated as follows [1]:

\[ P_a = (2I/\rho c)^{1/2}, \]

where \( I \) is the intensity of ultrasound (W/m²), \( \rho \) is the density of the liquid medium (kg/m³, water in the present work) and \( c \) is the speed of sound in the medium (m/s).

The bubble dynamics in the acoustic field is described by the well-known Rayleigh–Plesset equation as

\[ R \frac{d^2R}{dt^2} + \frac{3}{2} \left( \frac{dR}{dt} \right)^2 = \frac{1}{\rho} \left[ P_i - P_\infty - \frac{2\sigma}{R} - \frac{4\mu}{R} \left( \frac{dR}{dt} \right) \right], \]

where \( R \) is the radius (m) of cavitation bubble at any time, \( \mu \) is the viscosity of the liquid medium (Ns/m²), \( \sigma \) is the surface tension (N/m), \( P_i \) is the pressure inside the bubble (N/m²) and \( P_\infty \) is the pressure in the liquid far from the bubble (N/m²).

The effects of mass and heat transfer on the general trends of bubble dynamics are not known to be significant [5,35,36]. These effects have not been considered in the present study and it should be recognized that the inclusion of these effects will generate maximum bubble temperatures \( T_{\text{max}} \) that are significantly different to those calculated in the present study. However, the qualitative trends of \( T_{\text{max}} \) with variation in the operating parameters will not be affected (this is expanded upon in Section 5).

The simulations that were performed were terminated when the calculated speed of the bubble wall reached the speed of sound in water (1485 m/s). However, for many simulations this condition was never reached and thus simulations were terminated after 60,000 iterations. The termination of the numerical simulations after 60,000 iterations was chosen arbitrarily. Sixty thousand iterations were carried out in order to check whether bubble wall speed reaches \( \approx 1485 \text{ m/s over several acoustic cycles.} \)

The time varying fluctuating pressure field, \( P_t \), as obtained from Eq. (1) was substituted for \( P_\infty \) in Eq. (3). Eq. (3) was solved using the Runge–Kutta fourth-order method with the following initial conditions:

At \( t = 0 \), \( \frac{dR}{dt} = 0 \).

The maximum bubble temperature [5] was estimated with the following equation:

\[ T_{\text{max}} = T_0 \left( \frac{R_{\text{max}}}{R_{\text{min}}} \right)^{3(\gamma-1)}, \]

where \( T_0 \) is the temperature of the cavitating medium (298 K in the present study), \( \gamma \) is the specific heat ratio, \( R_{\text{max}} \) is the maximum radius of the bubble prior to the start of collapse and \( R_{\text{min}} \) is the minimum radius of the bubble at the collapse.

The following assumptions were adopted in the numerical simulations study:

1. A single bubble in isolation and of spherical shape during its entire lifetime.
2. An initial radii of the bubble = 4.5 or 5 \( \mu \text{m} \) (for 213 kHz), 4.5 or 5 \( \mu \text{m} \) (for 355 kHz), 2.5 or 3 \( \mu \text{m} \) (for 647 kHz) and 1.5 or 2 \( \mu \text{m} \) (for 1056 kHz) [37–39].
3. A spatially uniform pressure and temperature within the bubble.
4. Gravity and other body forces were neglected.
5. The liquid is incompressible.

4. Results

The overall cavitation activity can be correlated to the total MBSL intensity or the total amount of primary radicals produced during sonication. The total MBSL intensity can be monitored by the procedure discussed in the experimental section. However, due to their high reactivity, the total number of primary radicals produced by the acoustic bubbles cannot be directly measured. It is generally accepted that the yield of H₂O₂ can be considered as an indicator of the sonochemical activity. This is a valid practice, in particular for comparative studies. The reaction between primary radicals results in the formation of H₂O₂ as described elsewhere [1]. Thus, both the MBSL intensity and H₂O₂ yields have been considered in this study for the comparison of the cavitation activity under different experimental conditions.

The MBSL intensities observed from sonicated water (continuous mode) at different acoustic powers and frequencies are shown in Fig. 1. Whereas the results shown are for a 200 mL solution volume, similar trends were observed (results not shown) for a 100 mL solution volume. Although the intensities were found to be affected by the solution volume chosen, the trends observed (relative changes) in terms of frequency and power effects, were similar for both solution volumes.

The results observed in Fig. 1 have been summarized below:
The acoustic power threshold for SL to occur at 213, 355, 647 and 1056 kHz is about 2.5, 3.5, 5 and 7 W, respectively.

The SL intensity increases with an increase in the acoustic power for all ultrasound frequencies.

For a given acoustic power above the SL threshold, the SL intensity decreases with an increase in the frequency in the order 213 > 355 > 647 > 1056 kHz.

The acoustic power absorbed by the cavitating medium was measured calorimetrically, however, we are conscious of the limitations of this parameter. Nevertheless, we also note that the trends are considered on a relative basis and therefore should not be affected by the inherent weakness in the measured calorimetric values, nor any loss of acoustic energy by other mechanical effects.

The variation in the experimentally obtained H₂O₂ yields with acoustic power, for different ultrasound frequencies, is shown in Fig. 2. The salient features are:

- For approximately the same acoustic power level, there is not a significant difference in the H₂O₂ yields at 213, 355 and 647 kHz. The yield is the lowest at 1056 kHz.

The variation in the numerically calculated $T_{\text{max}}$ as a function of acoustic frequency and power. The points to note are:

- $T_{\text{max}}$ increases with an increase in acoustic power at all frequencies.
- For a fixed acoustic power level, $T_{\text{max}}$ decreases with an increase in ultrasound frequency. However, there is not a significant difference in $T_{\text{max}}$ between 213 and 355 kHz. $T_{\text{max}}$ is significantly less for 647 and 1056 kHz compared with the lower frequencies.
- For 1056 kHz, $T_{\text{max}}$ only shows a significant increase beyond about 12 W.

5. Discussion

5.1. Effect of acoustic power

It can be observed from Figs. 1 and 2 that both the MBSL intensity and H₂O₂ yield increase with an increase in the acoustic power. This trend is in line with previous reports on the effect of acoustic power on the sonochemistry reaction efficiency. For example, Sehgal and Wang [40] have reported an increase in the decomposition of thymine (in terms of % change in the thymine concentration) in aqueous solutions over the intensity range of 0.5–3 W/cm² at 990 kHz. Gutierrez and Henglein [41] have observed an increase in the rate of oxidation of iodide with an increase in the acoustic power levels in the range 0.3–2 W/cm² at 1 MHz. They reported that 2 W/cm² was an optimum power intensity, at which a maximum rate for the sonochemical oxidation of iodide was observed.

The possible reasons for the general trend observed in our study (Figs. 1 and 2) and that reported in the literature.
[40,41] on the acoustic power effect are addressed in the following discussion. For a given ultrasound frequency, an increase in the acoustic power can be expected to increase the number of active cavitation bubbles and also the size ($R_{\text{max}}$) of the individual bubbles. In single bubble systems, it has been experimentally [9] shown that an increase in the acoustic intensity leads to an increase in the size of the bubble. This increase in $R_{\text{max}}$ can be expected to result in an increase in the maximum collapse temperature.

When higher acoustic powers are applied, the bubbles are exposed to greater negative pressures during the rarefaction cycle of ultrasound wave and also greater positive pressures in the subsequent compression cycle. This results in higher $R_{\text{max}}$ and lower $R_{\text{min}}$ values. A cavitation bubble possesses its maximum potential energy at its maximum size, $R_{\text{max}}$. This potential energy, during bubble collapse, is partly converted into chemical reactions (i.e., formation of radicals and ions) and partly into heat, light and sound emission. The higher the $R_{\text{max}}$, the higher will be the potential energy available and thus would produce a higher $T_{\text{max}}$ and a greater sonochemical yield. It can be seen from Table 1 that as the power is increased, the $R_{\text{max}}/R_0$ and $R_{\text{max}}/R_{\text{min}}$ are increased leading to a corresponding increase in $T_{\text{max}}$ and the SL intensity.

Fig. 3 is the graphical presentation of the numerically obtained $T_{\text{max}}$ at different operating power. It can be noted that as the power is increased, $T_{\text{max}}$ is also increased significantly for all frequencies used in this study. Table 1 gives the numerically and experimentally obtained results for 213 kHz irradiation at different acoustic powers.

Another important observation that can be made from Figs. 1–3 is that the relative increase in SL and $T_{\text{max}}$ with acoustic power is more or the less same, whereas the H$_2$O$_2$ yield does not show the same relative increase. For example, in case of 213 kHz irradiation, when the power is increased from 4.2 to 9.3 W (i.e., 2.2 times), the SL is increased from 15 to 184 mV (i.e., 12 times) and $T_{\text{max}}$ from 1000 to 10,000 K (i.e., 10 times), whereas the H$_2$O$_2$ yield is increased from 6.7 to 18 mM (i.e., 2.5 times). It should also be noted at this stage that the SL intensity is mainly a consequence of $T_{\text{max}}$, whereas the H$_2$O$_2$ yield (i.e., sonochemistry) is a consequence of the average temperature of the bubbles during collapse as well as associated mass transfer effects.

The number of active cavitation bubbles may increase with an increase in power. Whereas the results shown in Figs. 1 and 2 have been interpreted on the basis of $T_{\text{max}}$ of a single bubble, an increase in the number of bubbles may also be responsible for the observed trends. However, it is difficult to estimate the number of active bubbles at different power levels using the available experimental techniques. Also, the values obtained for $T_{\text{max}}$ are overestimated, as has been already been noted, because mass and heat transfer effects have not been considered. Further details concerning these issues will be discussed in the following section.

5.2. Effect of frequency

In order to understand the results presented in Figs. 1 and 2 in terms of a frequency effect, it is necessary to consider the factors that influence both sonochemistry and MBSL. As noted previously, sonochemical reactions depend upon the average bubble temperature and the number of active bubbles, whereas the MBSL intensity depends on the $T_{\text{max}}$ of the collapsing bubbles and the number of active bubbles. The active bubbles are those bubbles that collapse violently and are capable of producing sonochemistry and SL. It should be noted that there will be some bubbles that produce sonochemistry but not emit SL, as the temperature threshold for SL is much higher than the threshold for radical formation. $T_{\text{max}}$, which is attained at the end of bubble collapse, determines the intensity of SL emitted from individual bubbles. $T_{\text{max}}$ depends on the severity of the collapse, which in turn depends on a number of factors, such as the symmetry of the collapse, maximum size of the bubble prior to collapse ($R_{\text{max}}$), the minimum size of the bubble at the collapse ($R_{\text{min}}$), and the amount and the type, of the gaseous material within the bubble.

The population, i.e., number and size, of cavitation bubbles also depends on the frequency of irradiation and the acoustic power. It is acknowledged that higher irradiation frequencies produce a greater number of cavitation bubbles but comparatively higher powers are required to produce active bubbles. This is due to the fact that bubbles grow to a lesser extent and undergo lower compression at comparatively higher irradiation frequencies.

It can be observed from Fig. 1 that the SL intensities, over the whole acoustic power range, are decreased significantly as the irradiation frequency is increased (213 > 355 > 647 > 1056 kHz). This may be as a consequence of a number of factors, such as a decrease in $T_{\text{max}}$, and an increase in amount of gaseous material within the bubbles and a decrease in the number of active bubbles. How these factors vary as a function of frequency is a complex issue. However, possible reasons can be suggested in order to interpret the observed results.

<table>
<thead>
<tr>
<th>Power (W)</th>
<th>$P_0$ (atm)</th>
<th>SL (mV)</th>
<th>Experimental H$_2$O$_2$ ($\mu$M)</th>
<th>$R_{\text{max}}/R_0$</th>
<th>$R_{\text{max}}/R_{\text{min}}$</th>
<th>$T_{\text{max}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.5</td>
<td>1.0</td>
<td>124</td>
<td>11</td>
<td>1.6</td>
<td>3.3</td>
<td>1260</td>
</tr>
<tr>
<td>9.3</td>
<td>1.1</td>
<td>184</td>
<td>18</td>
<td>2.6</td>
<td>19</td>
<td>10,200</td>
</tr>
<tr>
<td>15</td>
<td>1.4</td>
<td>170</td>
<td>26</td>
<td>2.8</td>
<td>29</td>
<td>16,800</td>
</tr>
<tr>
<td>20</td>
<td>1.6</td>
<td>286</td>
<td>28.5</td>
<td>2.7</td>
<td>28.5</td>
<td>16,700</td>
</tr>
</tbody>
</table>

Table 1

SL, bubble dynamics parameters, $T_{\text{max}}$ and H$_2$O$_2$ yields at 213 kHz irradiation; $R_0 = 4.5$ $\mu$m
First, let us consider an ideal case of a single bubble at these frequencies. The numerically obtained results at 9.3 W for different frequencies are shown in Table 2. The typical radius–time profiles are presented in Figs. 4 and 5 for different irradiation frequencies at a fixed acoustic power. It can be seen from Table 2 that as the frequency is increased, the \( \frac{R_{\text{max}}}{R_{\text{min}}} \) ratio is decreased. The \( \frac{R_{\text{max}}}{R_{\text{min}}} \) is indicative of change in the volume of the bubble upon collapse. Thus, \( \frac{R_{\text{max}}}{R_{\text{min}}} \) is the compression ratio of the bubble, which represents the severity of the collapse. At a higher frequency, bubbles have comparatively less time to grow (during the rarefaction cycle of the ultrasound) and to collapse (during the compression cycle of the ultrasound). Thus, relatively higher acoustic power is needed at a higher frequency in order to show the same effect as at lower frequency from acoustic cavitation. If the applied acoustic power is decreased, then bubbles will experience a lower negative pressure during the rarefaction cycle as well as a lower positive pressure during the compression cycle. This results in a lower \( \frac{R_{\text{max}}}{R_{\text{min}}} \), which in turn results in a lower \( T_{\text{max}} \).

Thus, just by considering single bubble dynamics, the observed trend in the SL intensity can be qualitatively explained. However, a quantitative comparison between the \( T_{\text{max}} \) and the SL intensity as a function of frequency cannot be established simply by considering numerical calculations based on single bubble dynamics. For example, when the frequency is increased from 213 to 1056 kHz at 9.3 W, \( T_{\text{max}} \) and the SL intensity are decreased by 85% and 98%, respectively. When the frequency is increased from 213 to 355 kHz, \( T_{\text{max}} \) is decreased by 3–5%, whereas the SL intensity is decreased by 86%.

At this stage, it must be noted that the experimentally obtained MBSL is also controlled by the number of active cavitation bubbles. It has been predicted [30] that the number of active cavitation bubbles increases with an increase in the ultrasound frequency, which indicates that cavitation activity and hence the MBSL intensity can be expected to increase with an increase in frequency. However, the observed MBSL results show the opposite trend. What this indicates is that of the two factors, the \( T_{\text{max}} \) and the number of active bubbles, \( T_{\text{max}} \) must be the dominant factor that governs the overall MBSL intensity trend.

The additional factors that should be considered in the discussion are the heat and mass transfer processes at the bubble/liquid interface as well as within bubbles. These parameters were not taken into account when the \( T_{\text{max}} \) values were calculated (Table 2, Fig. 3). It can be expected that the heat and mass transfer processes would play a significant role in controlling the \( T_{\text{max}} \) as well as the SL intensity. As the frequency increases, the extent of stable cavitation is known to increase [30]. At higher frequencies, the bubbles undergo hundreds perhaps thousands of oscillations before their complete collapse. During the growth of the bubble, the amount of water vapor increases due

![Fig. 4. Radius-time profile for a single bubble exposed to 213 and 1056 kHz ultrasound at 9.3 W \([R_0 = 4.5 \mu m (213 kHz), 1.5 \mu m (1056 kHz)]\).](image)

![Fig. 5. Radius-time profile for a single bubble exposed to 355 and 647 kHz ultrasound at 9.3 W \([R_0 = 4 \mu m (355 kHz), 2.5 \mu m (647 kHz)]\).](image)

<table>
<thead>
<tr>
<th>Frequency (kHz)</th>
<th>SL (mV)</th>
<th>Experimental ( \text{H}_2\text{O}_2 ) (( \mu \text{M} ))</th>
<th>Assumed ( R_0 ) for simulation (( \mu \text{m} )) ( a )</th>
<th>( \frac{R_{\text{max}}}{R_0} )</th>
<th>( \frac{R_{\text{max}}}{R_{\text{min}}} )</th>
<th>( T_{\text{max}} ) (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>213</td>
<td>184</td>
<td>18</td>
<td>4.5, 5</td>
<td>2.6, 2.7</td>
<td>19, 18.5</td>
<td>10,300, 9900</td>
</tr>
<tr>
<td>355</td>
<td>25</td>
<td>20</td>
<td>4, 4.5</td>
<td>2.6, 2.5</td>
<td>18, 16.7</td>
<td>9700, 8700</td>
</tr>
<tr>
<td>647</td>
<td>23</td>
<td>17</td>
<td>2.5, 3</td>
<td>2.2, 1.8</td>
<td>11, 6</td>
<td>5400, 2600</td>
</tr>
<tr>
<td>1056</td>
<td>3</td>
<td>4</td>
<td>1.5, 2</td>
<td>1.3, 1.6</td>
<td>2, 3.7</td>
<td>700, 1400</td>
</tr>
</tbody>
</table>

\( a \) Previous experimental investigations [37–39] indicate that the initial size of bubbles in cavitating medium is not a single value whereas it has a range. Thus, the numerical simulations have been carried out at two different values of \( R_0 \).
to the evaporation at the bubble–liquid interface primarily caused by a decrease in the pressure inside the bubble. During the bubble collapse, however, the amount of water vapor decreases due to condensation at the interface because of an increase in the pressure inside the bubble. At the final stage of compression, the speed of the bubble collapse becomes so high that the condensation becomes a non-equilibrium process and water vapor does not get time to escape [42]. Thus, the evaporation and the condensation at the bubble wall and the amount of vapor trapped, at the end of the collapse, may play a major role in determining the amount of \( \text{H}_2\text{O}_2 \) yield. When endothermic chemical reactions are considered, a lower temperatures are realized compared to that obtained by a simple adiabatic compression of the bubble [35,43,44]. As suggested by Ashokkumar and Grieser [29], endothermic reactions consume heat energy, which results in a lower SL intensity.

From Eq. (4), for higher \( R_{\text{max}} \), \( T_{\text{max}} \) would be higher. However, a higher \( R_{\text{max}} \) is also accompanied by a larger amount of vapor being trapped during the collapse phase of a bubble and hence can lower \( T_{\text{max}} \). Lohse and co-workers [43], based on a comprehensive numerical model, reported that at low frequencies (<16 kHz) the latter mechanism dominates. Thus, for frequencies >16 kHz, \( T_{\text{max}} \) can be considered to be directly proportional to \( R_{\text{max}} \).

From the above discussion, it is clear that the numerically obtained \( T_{\text{max}} \) values are overestimated. However, the obtained trends of \( T_{\text{max}} \), with operating power and frequency, should not change significantly with inclusion of mass and heat transfer effects. Thus, the numerical results of the present study need to be considered on a qualitative basis rather than on a quantitative basis.

It is observed in Table 2 and Fig. 2 that the experimental yield of \( \text{H}_2\text{O}_2 \) is more or less the same for 213, 355, and 647 kHz irradiations, whereas it is considerably less for 1056 kHz irradiation. This trend is different to that observed (within experimental errors) for the frequencies 213, 355 and 647 kHz.

The decrease expected based on both the time availability for evaporation and a lower \( T_{\text{max}} \) must have been compensated by an increase in the number of active bubbles (i.e., effective cavitation events per unit time). In addition, whereas the \( T_{\text{max}} \) decreases with an increase in the frequency, the average temperature of the bubble may not vary significantly between these frequencies.

Yasui et al. [45], in their numerical work, have obtained an optimum temperature (about 5500 K) for the sonochemical production of oxidants from an air bubble. They have reported an increase in the amount of oxidants up to 5500 K and a decrease beyond this temperature. This is because at higher temperatures oxidants can be consumed by oxidizing nitrogen. Based on this study the temperature within the bubble may also play a role for the observed trends of the \( \text{H}_2\text{O}_2 \) yield in the case of the 213, 355 and 647 kHz ultrasonic irradiations.

6. Summary and conclusions

The effects of ultrasound frequency and acoustic power on MBLS intensity and \( \text{H}_2\text{O}_2 \) yield have been experimentally investigated. The observed experimental results have been discussed on the basis of single bubble dynamics, that are obtained from numerical simulations, and the number of active cavitation bubbles. Whereas the effect of acoustic power on SL and sonochemical reactions could be accounted for by considering the above mentioned parameters, the effect of frequency could only be speculated upon due to the complexities involved. The dependence of \( \text{H}_2\text{O}_2 \) yield on frequency is governed by the average bubble temperature, heat and mass transfer effects and the number of cavitation bubbles.

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References
