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Energy analysis during acoustic bubble oscillations: Relationship between bubble energy and sonochemical parameters

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ABSTRACT

In this work, energy analysis of an oscillating isolated spherical bubble in water irradiated by an ultrasonic wave has been theoretically studied for various conditions of acoustic amplitude, ultrasound frequency, static pressure and liquid temperature in order to explain the effects of these key parameters on both sonochemistry and sonoluminescence. The Keller–Miksis equation for the temporal variation of the bubble radius in compressible and viscous medium has been employed as a dynamics model. The numerical calculations showed that the rate of energy accumulation, dE/dt, increased linearly with increasing acoustic amplitude in the range of 1.5–3.0 atm and decreased sharply with increasing frequency in the range 200–1000 kHz. There exists an optimal static pressure at which the power w is highest. This optimum shifts toward a higher value as the acoustic amplitude increases. The energy of the bubble slightly increases with the increase in liquid temperature from 10 to 60 °C. The results of this study should be a helpful means to explain a variety of experimental observations conducted in the field of sonochemistry and sonoluminescence concerning the effects of operational parameters.

1. Introduction

A direct consequence of ultrasound transmission through a liquid is the growth of adventitious bubble nuclei, which may collapse violently, generating localized hot-spots of extreme pressure and temperature (~1000 atm and ~5000 K respectively [1,2]). This phenomenon, termed acoustic cavitation, is responsible for the vast majority of applications involving ultrasound (e.g. materials synthesis, environmental remediation, food technology, etc.) and also for the emission of broad wavelength light: sonoluminescence [3,4].

Cavitation systems (multibubble) are extremely complex in nature with numerous interdependent parameters that influence the bubble dynamics and thus the overall efficiency (chemical and/or physical) of the system. From an engineering point of view, cavitation systems would be ideally studied with a single cavitation bubble with known size pulsating in known acoustic pressure field. Such studies of single bubble cavitation provide the means to understand the dependence of operational parameters (such as ultrasonic frequency, acoustic amplitude, static pressure, liquid temperature,. . .) on the bubble dynamics and thus, to explain the efficiency of sonochemical processes utilizing cavitation phenomena. A lot of research studies in the literature have addressed the matter with this approach. Gogate et al. [5,6] have correlated the iodine liberation rate to the extremes of temperature and pressure obtained at the transient collapse using a bubble dynamics model. Sivasankar et al. [7] also took a similar approach and explained some trend in sonochemistry of KI oxidation. Prasad Naidu et al. [8] explain the trend in the rate of iodine liberation with the sonication of aqueous KI solution of various concentrations and different gas atmospheres using the Rayleigh–Plesset equation for the radial motion with Flynn's criterion [9] that suggests the adiabatic collapse phase of the cavity on the basis of the assumption of partial pressure of gas in the bubble equals the vapor pressure of the cavitating media at the operating parameters. The used form of the Rayleigh–Plesset equation in Prasad Naidu's study does not take into account the compressibility of the liquid medium, and hence, Prasad Naidu et al. terminated their simulations at the instance when the bubble wall velocity reaches the velocity of sound in water. Gogate and Pandit [10] added the liquid compressibility to Prasad Naidu's assumptions and developed an empirical correlation that predicts the collapse pressure as function of intensity, ultrasound frequency and initial nuclei size. Kanthale et al. [11] used simulation results provided by a single bubble dynamics model to discuss their results on the effect of ultrasound frequency and acoustic power on both sonochemistry (H_2O_2 yield) and sonoluminescence.

However, most reported studies (including those cited above) correlated their experimental results to the maximum temperature or pressure reached in the bubble at the end of the collapse. In this

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study, we related the efficiency of both sonochemical reactions and sonoluminescence to the energy of the cavity which includes simultaneously the temperature and pressure inside a bubble, the collapse duration and other dynamic factors. The energy of the collapsing bubble has been theoretically estimated for various operating conditions of ultrasonic frequency, acoustic amplitude, static pressure and liquid temperature.

2. Model

Before describing the model, we would like to mention that the phenomenon of bubble collapse and subsequent fragmentation depends on many factors such as the surface instability, local flow conditions and the bubble population in the vicinity of the bubble [7]. We assume that in sonochemistry applications, the cavitation is transient and the bubble breaks apart (fragments) at the first collapse after an initial expansion. Therefore, we are only interested in the first expansion and subsequent violent collapse.

The bubble dynamics model used in the present study and the assumptions therein have been fully described in our previous works [12,13]. The following is a brief description of the model. A gas and vapor filled spherical bubble isolated in water oscillates under the action of a sinusoidal sound wave. The radial dynamics of the bubble is described by the Keller–Miksis equation that includes first order terms in the Mach number $M = \dot{R}/c$ [14–16]:

$$\left(1-\frac{\dot{R}}{c}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3c}\right)\dot{R}^2=\frac{1}{\rho_L}\left(1+\frac{\dot{R}}{c}+\frac{R}{c}\frac{d}{dt}\right)\left[p-p_\infty-\frac{2\sigma}{R}-4\mu\frac{\dot{R}}{R}+P_A\sin(2\pi ft)\right] \tag{1}$$

in this equation dots denote time derivatives (d/dt), R is the radius of the bubble, c is speed of sound in the liquid, ρ_L is density of the liquid, σ is surface tension, μ is liquid viscosity, p is pressure inside the bubble, p_∞ is ambient static pressure, P_A is acoustic amplitude and f is the sound frequency. The acoustic amplitude P_A is correlated with the acoustic intensity I_a, or power per unit area, as $P_A = (2I_a\rho_L c)^{1/2}$ [3,10].

The expansion of the bubble is assumed as isothermal and its total compression is considered as adiabatic [17–19]. We also assume that the vapor pressure in the bubble remains constant during the bubble expansion phase and there is no gas diffusion during expansion and no mass and heat transfer of any kind during collapse.

On the basis of the above assumptions, the pressure and temperature inside the bubble at any instant during adiabatic phase can be calculated from the bubble size, using the adiabatic law:

$$p=\left[P_v+P_{g0}\left(\frac{R_0}{R_{\max}}\right)^3\right]\left(\frac{R_{\max}}{R}\right)^{3\gamma} \tag{2}$$

$$T=T_\infty\left(\frac{R_{\max}}{R}\right)^{3(\gamma-1)} \tag{3}$$

where P_v is the saturated vapor pressure, $P_{g0} = p_\infty + (2\sigma/R_0) - P_v$ is the gas pressure in the bubble at its ambient state ($R = R_0$), R_0 is the ambient bubble radius, T_∞ is the bulk liquid temperature, $R_{\max}$ is the maximum radius of the bubble and γ is the ratio of specific heats capacities (c_p/c_v) of the vapor/gas mixture. The maximum internal temperature ($T_{\max}$) and pressure ($p_{\max}$) reached in the bubble at the end of the bubble collapse are approximated by:

$$T_{\max}=T_\infty\left(\frac{R_{\max}}{R_{\min}}\right)^{3(\gamma-1)} \tag{4}$$

$$p_{\max}=\left[P_v+P_{g0}\left(\frac{R_0}{R_{\max}}\right)^3\right]\left(\frac{R_{\max}}{R_{\min}}\right)^{3\gamma} \tag{5}$$

where $R_{\min}$ is the minimum radius of the bubble at the collapse.

The Keller–Miksis equation (Eq. (1)) that describes the dynamic of the cavitation bubble is a second-order nonlinear differential equation. This equation has been numerically integrated using the fourth-order Runge-Kutta method. All the physical properties (saturated vapor pressure, density, surface tension, viscosity and sound velocity) in the above equations are calculated for water as function of liquid temperature T_∞ and static pressure p_∞. The equations for the physical properties have been described in our previous work [13].

In the present study, energy analysis is carried out on the basis of internal energy (ΔE). The variation in the internal energy of the bubble ΔE during its lifetime is given as the sum of internal energies of the two parts, isothermal (noted ΔE_{iso}) and adiabatic (noted ΔE_{ad}), and as $\Delta E_{\text{iso}} = 0$, the internal energy of the bubble resumed to (ΔE_{ad}), which is given as:

$$\Delta E=\Delta E_{\text{ad}}=\Delta W+\Delta Q \tag{6}$$

where ΔQ is the heat exchanged with the liquid during adiabatic phase ($\Delta Q = 0$) and ΔW is the work done to the system (the bubble). ΔW is given for an instant t of the adiabatic phase by:

$$\Delta W=\frac{p(t)V(t)-p_{\min}V_{\max}}{\gamma-1} \tag{7}$$

here $p(t)$ and $V(t)$ are the internal pressure and temperature at any instant t during adiabatic phase and $p_{\min}$ and $V_{\max}$ are, respectively, the maximum volume of the bubble and the minimum internal pressure achieved in the bubble, (both $p_{\min}$ and $V_{\max}$ are corresponding to $R_{\max}$).

The time scale in which the energy is accumulated in the cavity is of primary importance in order to compare the efficiency of the cavities oscillating with different operating parameters. So, we describe the power w as ΔE divided by the duration of bubble collapse τ_c:

$$w=\frac{\Delta E}{\tau_c} \tag{8}$$

3. Results and discussion

It is well established that the ambient bubble radius R_0 for an active bubble depends on experimentally controllable parameters, particularly on the ultrasonic frequency and acoustic amplitude [20–23]. The active bubbles are those collapsing violently and which are capable of producing sonochemistry and sonoluminescence. Recently, Brotchie et al. [23] demonstrated that the ambient radius for an active bubble increased with increasing acoustic amplitude and decreased with increasing ultrasound frequency. In our previous work [12], we have studied in detail the influence of ultrasound frequency in the range of 200–1000 kHz and acoustic amplitude (up to 3 atm) on the ambient radius R_0 for an active bubble in sonochemical reaction using the same dynamics model adopted in this investigation. The obtained theoretical results agree well with the experimental results. Thus, in the present study, the ambient bubble radius R_0 for the numerical simulations of the bubble dynamics were selected as function of frequency and acoustic amplitude according to our previous study. The selected values are presented in Table 1.

3.1. Bubble dynamics and energy analysis

In Fig. 1a and b, the calculated results during the collapse of the bubble are shown as function of time for an ultrasonic frequency of 300 kHz and acoustic amplitude of 2.5 atm. In Fig. 1a, the reduced bubble radius and internal energy ΔE are shown. During the

Table 1
Selected values of ambient bubble radius (R_0) as function of ultrasound frequency and acoustic amplitude [12].

Frequency	200 kHz	300 kHz	500 kHz	1000 kHz
Amplitude (P_A) (atm)	R_0 (μm)			
1.5	5.0	3.5	2.7	1.85
2.0	6.6	4.25	2.5	1.55
2.5	8.0	5.25	3.0	1.4
3.0	9.5	6.0	3.5	1.5

collapse phase, the bubble compresses (collapses) very violently up to 7% of its maximum radius ($\tau_c \approx 0.82$ μs). However, the internal energy of the bubble starts with a slow increase during the first stage and then sharply increases at the end of the collapse (~3.02 μs). The same behaviors have been observed for the internal temperature and pressure variations during the implosion phase as shown in Fig. 1b. At the end of the bubble collapse, the temperature and pressure inside a bubble increase drastically up to 4800 K and 1500 atm respectively. Thus, as ΔE is related to the conditions of implosion (Eq. (7)), the reason for the rise of internal energy during adiabatic collapse is the increase in temperature and pressure inside a bubble during its collapse.

3.2. Effect of acoustic amplitude

In Fig. 2a, the effect of acoustic amplitude on the power w is shown for an ultrasonic frequency of 300 kHz. As can be seen, the power w increases linearly with increasing acoustic amplitude from 1.5 to 3 atm.

While the present model focuses on single bubble, experimental results reported in the literature were obtained from a bulk solution (multibubble system). Therefore, an exact comparison between this model and experimental results is impossible. This is in fact due to the complexity of the multibubble system which includes also the bubble–bubble interactions phenomenon (coalescence, clustering, etc.) that is not considered in the present model. However, although all the considerations, various studies reported in the literature clearly support the observed trends of the variation of the power w with acoustic amplitude. Koda et al. [24] analyzed the variation in the sonochemical efficiency in various laboratory scale sonochemical reactors of operating power in the range of 35–220 W with irradiation frequency in the range of 25–1200 kHz. The obtained results indicate that sonochemical efficiency increases linearly with increasing ultrasonic power. At 300 kHz, the sonochemical oxidation of bisphenol A was found to be higher at higher acoustic power [25]. Other similar results have been also reported by other researchers [26–28]. The sonoluminescence intensity was also found to be increased with acoustic power [11,29].

When higher acoustic amplitudes are applied, the bubble is 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_{max} and lower R_{min}

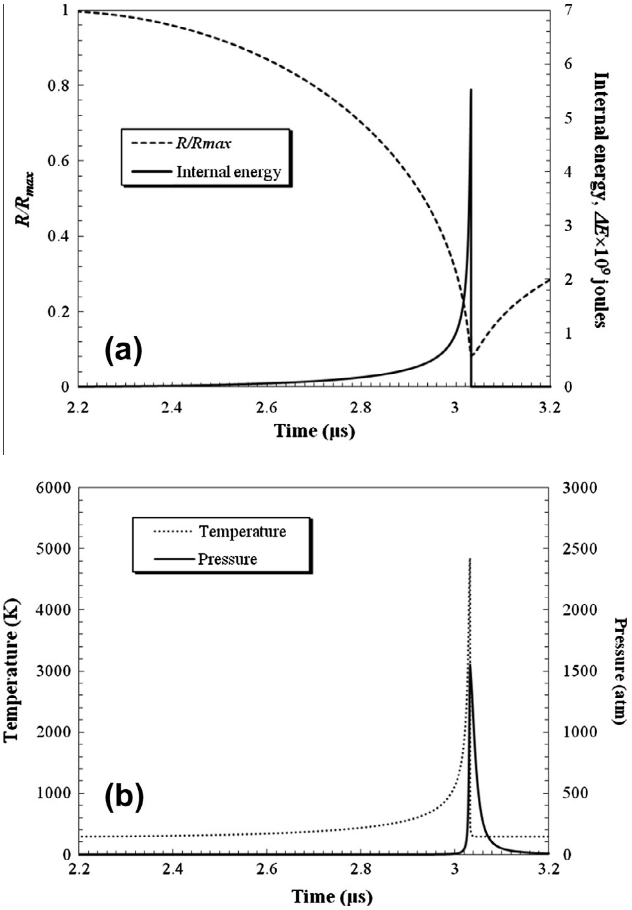

Fig. 1. Reduced bubble radius (R/R_{max}) and internal energy of the bubble (a) and temperature and pressure inside a bubble (b) as function of time during the collapse (conditions: ambient bubble radius: 5.25 μm; frequency: 300 kHz; acoustic amplitude: 2.5 atm; liquid temperature: 20 °C; static pressure: 1 atm).

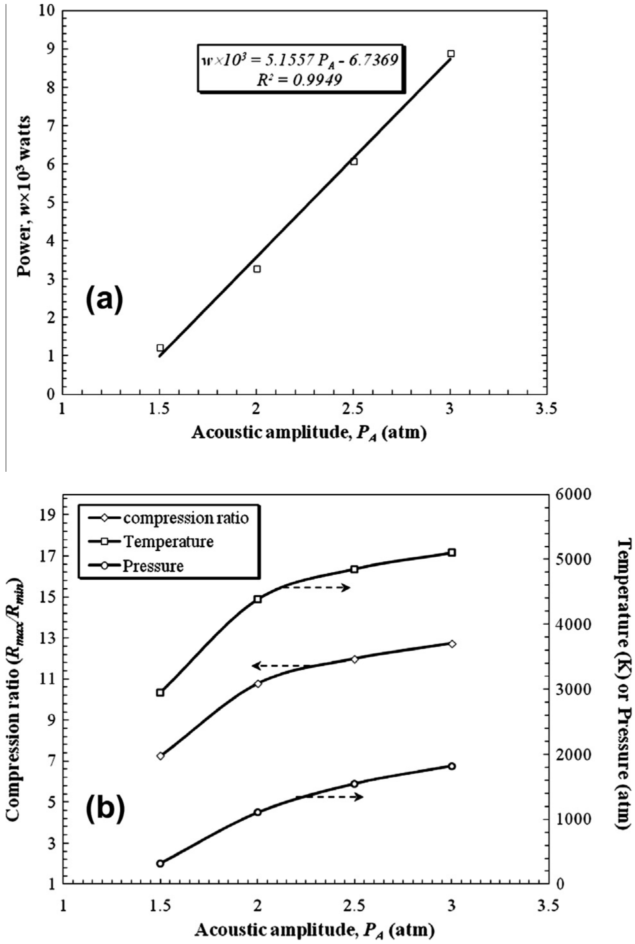

Fig. 2. The calculated power w for one collapse (a) and the compression ratio (R_{max}/R_{min}) of the bubble and the collapse temperature and pressure (b) as function of acoustic amplitude (conditions: ambient bubble radius: Table 1; frequency: 300 kHz; liquid temperature: 20 °C; static pressure: 1 atm).

values (higher compression ratios $R_{\max}/R_{\min}$). The higher the $R_{\max}/R_{\min}$, the higher will be the $T_{\max}$ and $p_{\max}$ (Eqs. (4) and (5)) and thus would produce a higher energy (ΔE) in the bubble at the collapse. It can be seen from Fig. 2b that as the acoustic amplitude increases, the $R_{\max}/R_{\min}$ increases leading to a corresponding increase in $T_{\max}$ and $p_{\max}$. But increasing acoustic amplitude leads also to an increase in the collapse duration τ_c. However, the order of ΔE increase with acoustic amplitude is greater than that of τ_c which results in higher power w at higher acoustic amplitudes as shown in Fig. 2a. We should noted that the increase in τ_c may also attributed to the increase in ambient radius R_0 of the bubble with acoustic amplitude but, however, the results have shown that the major part of τ_c increase is due to the acoustic amplitude itself. So as a logical conclusion, the cavities are more effective at higher acoustic amplitudes. In incompressible medium, Vichare et al. [17] concluded that the cavities are less effective at higher acoustic amplitudes.

3.3. Effect of ultrasound frequency

In order to seek a correlation between the power w and the driving ultrasound frequency, numerical simulations have been performed for various frequencies (200, 300, 500 and 1000 kHz) at different acoustic amplitudes (1.5, 2, 2.5 and 3.0 atm). The liquid temperature was 20 °C and the static pressure was 1 atm. The obtained results are presented in Fig. 3a. The vertical axis in this figure is in logarithmic scale. As can be seen, the power w is higher at lower ultrasound frequency for all the employed acoustic amplitudes. This result may be as a consequence of a number of factors, such as a decrease in $T_{\max}$ and $p_{\max}$, an increase in amount of water vapor trapped at the collapse and a decrease of the lifetime and the collapse duration τ_c. How ultrasound frequency affects these factors is a complex issue. However, a possible explanation can be made basing on simulation results. Because a smaller frequency gives the bubble more time to expand, it leads to a larger expansion ratio $R_{\max}/R_0$ and so higher compression ratio $R_{\max}/R_{\min}$ as can be seen in Fig. 3b. As a result, the collapse will be stronger and generates higher $T_{\max}$ and $p_{\max}$. On the other hand, the expansion to a larger expansion radius is accompanied by a larger amount of vapor being trapped during collapse Fig. 3b), which decreases γ of the mixture and thus decreases $T_{\max}$ and $p_{\max}$. For high frequencies (above 200 kHz) the first mechanism dominates, and the net effect of decreasing frequency is an increase in $T_{\max}$ and $p_{\max}$ (Fig. 3b). The decrease of $T_{\max}$ and $p_{\max}$ and the increase of γ with increasing frequency would thus result in lower internal energy ΔE at higher ultrasound frequency (Eq. (7)). In addition, at higher frequency, the bubble has not enough time to store energy because the collapse duration τ_c is short at higher frequency. However, the decrease of ΔE with ultrasonic frequency is greater than that of τ_c, which results in lower power w at higher ultrasonic frequency as indicated Fig. 3a. Thus, it can be concluded, on the basis of single bubble dynamics, that cavities are less effective at higher operating frequency.

Pétrier and co-workers examined the sonochemical degradation of phenol [30], bisphenol A [25] and chlorophenols [31] at various frequencies in the range of 200–800 kHz. They found that the efficiency of sonochemical degradation decreases as the frequency of ultrasound increases. Kanthale et al. [11] found that both sonochemistry (H_2O_2 yield) and sonoluminescence intensity decreases with increasing frequency in the range 213–1056 kHz. The same trend of decreasing sonoluminescence intensity with increasing ultrasonic frequency in the range of 358–1071 kHz was observed by Beckett and Hua [32]. Thus, there are several reports in the literature that support the observed variation of the power w for single bubble with ultrasound frequency.

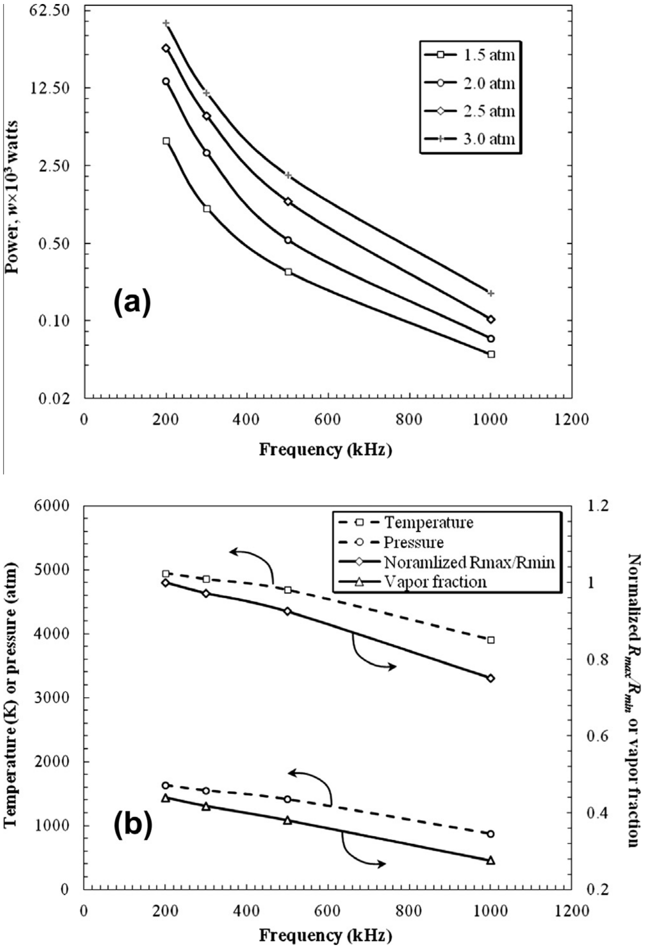

Fig. 3. The calculated power w for one collapse (a) and the temperature and pressure, vapor fraction and normalized $R_{\max}/R_{\min}$ (b) as function of ultrasound frequency for various acoustic amplitudes [2.5 atm for (b)]. Vertical axis in (a) is in logarithmic scale (conditions: ambient bubble radius: Table 1; frequency: 200–1000 kHz; liquid temperature: 20 °C; static pressure: 1 atm).

3.4. Effect of static pressure

The effect of static pressure p_∞ in the range of 0.3–3.0 atm on the power w has been studied for an ultrasonic frequency of 300 kHz and for various acoustic amplitudes (1.5–3.0 atm). The obtained results are shown in Fig. 4. As can be seen an optimum of static pressure is observed. This optimal value shifts toward higher value of static pressures as acoustic amplitude increases. At the acoustic amplitude of 3 atm, the optimal static pressure is 1.5 atm. This optimal value of the static pressure decreases to 1, 0.75 and 0.5 atm as the acoustic amplitude decreases respectively to 2.5, 2.0 and 1.5 atm. A similar behavior has been observed in our previous work for the production of the oxidants [13].

The obtained maxima of the power w is certainly a consequence of competing factors, such as amount of gas content within the bubble, the expansion and compression ratios and the collapse duration τ_c which are strongly affected by the variation in static pressure. However, how these factors produce maximum energy in the bubble is difficult to explain. Although this, there exist in the literature a certain number of studies that support the existence of a maximum static pressure in both sonochemistry and sonoluminescence. Henglein and Gutierrez [33] found at 1 MHz that as the static pressure increases, the efficiency of KI oxidation increases to a maximum and then decreases with continuous increase in static pressure. The maximum shifts toward higher values

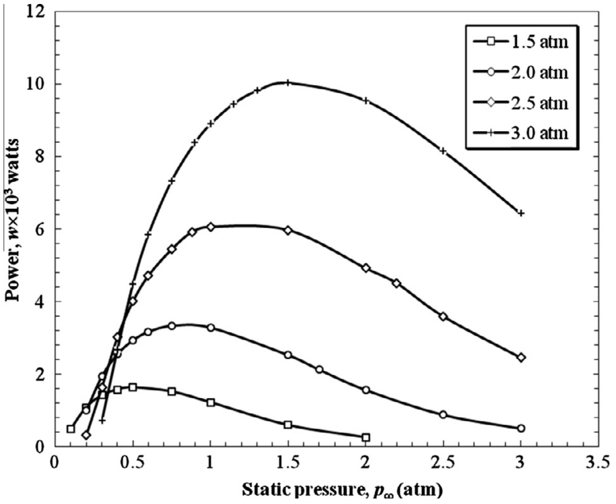

Fig. 4. The calculated power *w* for one collapse as function of static pressure for various acoustic amplitudes (conditions: ambient bubble radius: 3.5 μm for 1.5 atm, 4.25 μm for 2 atm, 5.25 μm for 2.5 atm and 6 μm for 3 atm; frequency: 300 kHz; liquid temperature: 20 °C).

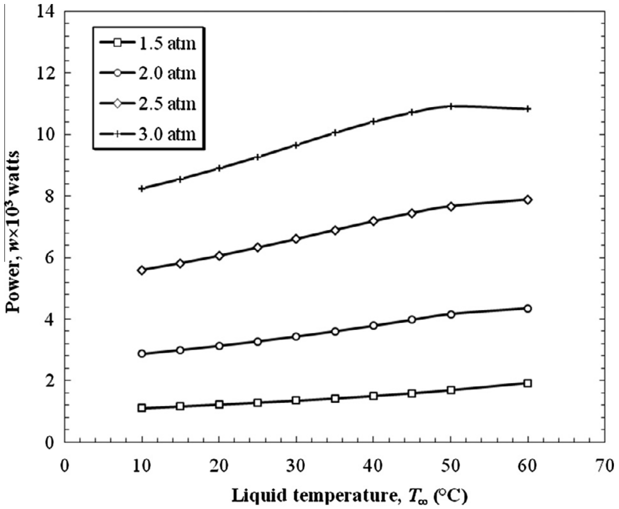

Fig. 5. The calculated power *w* for one collapse as function of liquid temperature for various acoustic amplitudes (conditions: ambient bubble radius: 3.5 μm for 1.5 atm, 4.25 μm for 2 atm, 5.25 μm for 2.5 atm and 6 μm for 3 atm; frequency: 300 kHz; liquid temperature: 20 °C).

of static pressure as the acoustic amplitude increases. Near atmospheric pressure, the disintegration of starch paste by ultrasound occurs at a maximum rate [34]. The amount of hydrogen peroxide, nitrate and nitrite produced by ultrasound irradiation of water is zero at air pressures less than 100 mm H_2O, above which it increases to maximum at 1520 mm H_2O, finally decreasing to zero at 4180 mm H_2O [34]. Similar results were reported by measurement of sonoluminescence intensity, but the maximum intensity of luminescence shifts toward higher static pressures when the ultrasonic power was increased [34], which is the same trend observed in Fig 4. Chendke [35] found at 20 kHz that sonoluminescence intensity of nitrogen saturated water increases with static pressure, reaching a maximum and subsequently decreases with a further increase in the static pressure.

We note at the end of this section that the variation of static pressure, p_∞, over the liquid is known to affect strongly the inertial cavitation threshold and the strength of inertial cavitation events [36,37]. The inertial cavitation threshold increases linearly with increasing static pressure at 25.5 kHz [36]. But does the static pressure affect the equilibrium size, R_0, of the bubble? Experimental studies in this subject are very limited and mostly carried out at low frequencies and at only some points of static pressures. Dan et al. [38] measured R_0 of the bubble in water/glycerine mixture for an acoustic amplitude $P_A = 1.29$ atm and a frequency of around 17.5 kHz. They found that decreasing the ambient static pressure p_∞ over the liquid from 1 atm to 0.9 atm increases R_0 from 7.3 to 9.0 μm, which is in obvious contradiction with the results of Kondic et al. [39] that predict a decrease of R_0 when p_∞ decreased. Thus, the change of R_0 with the static pressure p_∞ is not well established. To address this subject, we are preparing a detailed theoretical analysis of the $R_0 - p_\infty$ dependence for various ultrasound frequencies, including those <200 kHz. The preliminary results performed at 1 MHz showed that as the static pressure increases from 0.5 to 1 atm, the range of R_0 decreases and shifts toward higher values of R_0, but the optimal radius, which is the typical radius used in the simulations, was not affected by the increase of static pressure in this range of p_∞ (e.g. at $p_\infty = 0.5$ atm, the range of R_0 is ~0.3–3.2 μm and is ~0.5–3.2 when $p_\infty = 1$ atm; whereas the optimal radius remained always at 1.4 μm). The same trend is observed when p_∞ increases above 1 atm, only a slight increase of the optimal radius is observed. Thus, if the ambient radius R_0, which is the optimum radius, increases with increasing p_∞ above 1 atm, this results in lower corresponding T_{max} and P_{max} and thus to more lower energy. So, the slight increase in R_0 with p_∞ above 1 atm may change the absolute value of the bubble energy but definitively will not change the trend observed in Fig. 4.

3.5. Effect of liquid temperature

Fig. 5 shows the variation of the power *w* as function of liquid temperature (10–60 °C) for various acoustic amplitudes (1.5–3 atm) when the ultrasound frequency is 300 kHz. As can be seen from this figure, the power *w* is not strongly affected by the elevation of liquid temperature from 10 to 60 °C for all acoustic amplitudes.

Effectively, the ambient liquid temperature does not appear to be a critical factor in many sonochemical effects. For example, the ultrasonic disintegration of starch paste [34] and the sonochemical degradation of CCl_4 [40] are hardly affected by temperature over the range 20–60 °C. Similarly, the liberation of iodine from KI-CCl_4 aqueous solution shows only a slight decrease with temperature from 2 to 60 °C [41]. However, the elevation of liquid temperature appears to reduce the sonoluminescence intensity [35]. One of the possible explanations given on the basis of single bubble system [11] is that sonochemical reactions depend upon the average collapse temperature, whereas sonoluminescence intensity depends only on the T_{max} of the collapsing bubble, which decreases as the liquid temperature increases [13].

4. Conclusion

A model for the bubble dynamics in acoustic field have been used to estimate the energy of the bubble at various conditions of ultrasound frequency, acoustic amplitude, static pressure and liquid temperature in order to explain some observations in the field of sonochemistry and sonoluminescence. The energy of the bubble was found to be increased with increasing acoustic amplitude and decreased with increasing ultrasound frequency. There exists an optimal static pressure which shifts toward a higher value as the acoustic amplitude increases. The energy of the bubble slightly increased with the increase in liquid temperature in the range 10–60 °C. The results of this theoretical study, although are obtained from a single bubble system, are able to explain the qualitative trend observed in some experimental studies in the field of sonochemistry and sonoluminescence phenomena.

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