Ultrasonics Sonochemistry 38 (2017) 168–173

Contents lists available at ScienceDirect

Ultrasonics Sonochemistry

journal homepage: www.elsevier.com/locate/ultson

A derivation of the stable cavitation threshold accounting for bubble-bubble interactions

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ARTICLE INFO

Article history:
Received 6 January 2017
Received in revised form 16 February 2017
Accepted 7 March 2017
Available online 9 March 2017

Keywords:
Stable cavitation
Bubble dynamics
Subharmonic emission
Bubble clouds

ABSTRACT

The subharmonic emission of sound coming from the nonlinear response of a bubble population is the most used indicator for stable cavitation. When driven at twice their resonance frequency, bubbles can exhibit subharmonic spherical oscillations if the acoustic pressure amplitude exceeds a threshold value. Although various theoretical derivations exist for the subharmonic emission by free or coated bubbles, they all rest on the single bubble model. In this paper, we propose an analytical expression of the subharmonic threshold for interacting bubbles in a homogeneous, monodisperse cloud. This theory predicts a shift of the subharmonic resonance frequency and a decrease of the corresponding pressure threshold due to the interactions. For a given sonication frequency, these results show that an optimal value of the interaction strength (i.e. the number density of bubbles) can be found for which the subharmonic threshold is minimum, which is consistent with recently published experiments conducted on ultrasound contrast agents.

1. Introduction

Cavitation bubbles, whether generated by ship propellers, ultrasound or used in sonochemistry and material surface cleaning, disintegrate at bubble collapse because a shape instability develops [1]. However, under certain conditions, disintegration need not occur, and one can achieve controlled and stable cavitation with weakly or strongly oscillating bubbles, with applications in diverse medical and engineering fields. In medical and therapeutic applications, bubbles can act as vectors for directed drug delivery and gene transfection within living cells [2]. Even if bubble collapses were thought to be the key element of cell permeabilization, recent works demonstrate the possibility of transfecting cells by gentle oscillating bubbles, possibly resulting in lower tissue damage, with numerous applications such as the permeabilization of central nervous system capillaries [3,4], or sonothrombolysis enhancement [5]. For engineering applications such as material cleaning, soft or gentle bubble dynamics recently appear as an additional mechanism of surface cleaning, without involving well-known erosive bubble collapse [6,7]. Amongst the potential underlying mechanisms of cleaning, the possibility of steadily fatiguing the surface contamination by the oscillatory component of the bubble-induced liquid flow has been demonstrated, requiring control of the fast boundary flows induced by bubbles oscillation near surfaces [8].

A commonly used indicator for the stable cavitation regime is the subharmonic component emitted by the bubble population [9,10]. Indeed the subharmonic component is the first nonlinear frequency component that arises uniquely from the bubble oscillations in opposite to harmonics possibly induced by nonlinear propagation. The diverse mechanisms underlying subharmonic generation have been theoretically and experimentally investigated, and can be classified as: (i) the large bubble theory for which the bubble population contains bubbles around twice the resonant diameter [11]; (ii) for larger amplitudes, volumetric oscillations of near-resonant sizes bubbles can bifurcate with period-doubling oscillations as bubble behavior becomes more chaotic [1]. (iii) the occurence of shape modes through parametric instability could also provide subharmonic component but with weak radiative efficiency [12] and (iv) insights of subharmonic generation through the periodic collapse of an acoustically-driven bubble cloud have been proposed [13]. Amongst these mechanisms, driving bubbles at twice their resonance frequency if the one requiring both moderate acoustic amplitudes and avoidance of bubble collapses. In this case subharmonic component arises above a pressure threshold whose theoretical predictions for subharmonic existence have been extensively studied [14–17] for free or coated bubbles. Particularly, this subharmonic threshold possesses a minimum value at twice the resonance frequency for free bubbles, and when

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http://dx.doi.org/10.1016/j.ultsonch.2017.03.010
1350-4177/

using coated bubbles a significant decrease and low-frequency shifting of this threshold are achieved depending on the microbubble shell properties [15]. However, it is worth noting that all these works considered only subharmonic emission from single bubble, or the collective result of simultaneous but independent contributions of a bubble distribution [16].

When it comes to considering bubble cloud dynamics, even linear oscillation of bubble clouds significantly differs from the physics of single bubbles, as internal spatial resonant modes occurs [18]. Particularly the first (spatial) cloud mode involves in-phase oscillations of the bubbles within the cloud, and thus bubble cloud dynamic is no more adequately described by the summed independent response of individual bubbles as significant bubble interactions occur. Therefore by assuming the bubbles to oscillate in phase and remaining spherical, several authors proposed a modification of the well-known Rayleigh-Plesset equation accounting for the bubble–bubble interaction [19–21]. Further works then extended this result to the case of N interacting bubbles to investigate bubble clusters dynamics [22–24]. Most of these works were devoted to the understanding of transient/collapsing activities of bubble clouds for sonochemistry applications, and almost none focused on stable cavitation activity of bubbles clouds. It can however be expected that for low bubble concentration, the generated subharmonic signal comes from the summed contribution of each individual bubble [16], while at higher concentration strong bubbles interactions would affect overall dynamics and therefore the stable cavitation emission, possibly explaining recent experimental observation of an optimal microbubble concentration for the promotion of stable cavitation dose [25]. Thus, to get first insight in the stable cavitation activity of a dense bubble cloud, as encountered in therapeutical or sonochemical applications, we propose in this study an extension of the derivation of the subharmonic threshold to the ideal case of a homogeneous and monodisperse bubble cloud.

2. Theory

2.1. Amplitude of the subharmonic response

We consider a homogeneous, monodisperse cloud of N bubbles in a liquid of density ρ and viscosity μ, where all the bubbles pulsate in phase, so that the dynamics of each bubble can be described by the modified Rayleigh-Plesset equation in its simplest form [22]:

$$\rho\left(R\ddot{R}+\frac{3}{2}\dot{R}^2\right)=p_0\left(\frac{R_0}{R}\right)^{3\gamma}-p_\infty-p_a(t)-\frac{2\sigma}{R}-4\mu\frac{\dot{R}}{R}-\rho S\left(R^2\ddot{R}+2R\dot{R}^2\right), \quad (1)$$

where R is the time-varying radius of one bubble around its static radius R_0, the dot denotes time derivative, p_∞ is the static pressure in the liquid, $p_0=p_\infty+2\sigma/R_0$ is the static pressure inside the bubble, p_a is the acoustic pressure of amplitude P_a and angular frequency $\Omega, p_a(t)=P_a\cos(\Omega t), \sigma$ is the surface tension, γ is the gas polytropic index, and S is the coupling strength between the bubble and all the surrounding ones, defined as function of the distances d_i between bubbles:

$$S=\sum_{i=1}^{N-1}\frac{1}{d_i}. \quad (2)$$

Unlike the theoretical works mentioned above, we are not interested in the violent dynamics in strong acoustic fields but in the weakly nonlinear oscillations of bubbles, thus the effect of liquid compressibility is discarded in Eq. (1). Also note that the hypothesis of in-phase bubble oscillations rests on the liquid incompressibility assumption. Including the effect of liquid compressibility in a more realistic description of the bubble cloud dynamics would also imply to account for the finite value of the speed of sound, and time delay effects should be introduced in Eq. (1) as the radiated acoustic wave takes time to travel along the distance d_i [26,27].

In the weakly nonlinear oscillations framework, approximate solutions of Eq. (1) can be found using an asymptotic approach [14]. We briefly recall the principle of such perturbation method, resulting in the analytical expression of the subharmonic oscillations. First, it is convenient to introduce new spatial and temporal coordinates:

$$R=R_0(1+x), \quad (3)$$

$$\tau=\sqrt{\frac{p_0}{\rho}}\frac{t}{R_0}, \quad (4)$$

and to transform Eq. (1) using a power series expansion in x by assuming small radial displacements around the static radius R_0 ($x \ll 1$). The proper derivation of the threshold of subharmonic oscillations requires to retain terms up to the cubic order in the power series expansion. Neglecting terms of fourth order or higher, this leads to the following equation for x:

$$(1-s)^{-1}\ddot{x}+\omega_0^2x=\zeta\cos(\omega\tau)+\left[\alpha_{s1}x^2-\frac{3}{2}\upsilon_1\dot{x}^2-e_1x\zeta\cos(\omega\tau)-2b\dot{x}\right]+\left[-\alpha_{s2}x^3+\frac{3}{2}\upsilon_2\dot{x}^2x+e_2x^2\zeta\cos(\omega\tau)+4b(1+s/2)\dot{x}x\right], \quad (5)$$

where the different parameters are given in Appendix A. In this set of parameters, ω and ω_0 are the (dimensionless) driving frequency and bubble eigen frequency, respectively, ζ is the effective driving pressure, b is the viscous damping constant, and $s=SR_0/(1+SR_0)$ is the coupling parameter which captures the effect of bubble-bubble interactions. When the latter is set to zero, one recovers the single bubble case [14].

The form of Eq. (5) shows the shift of the linear resonance frequency $\omega=\omega_0\sqrt{1-s}$ due to bubble interactions, in agreement with Ref. [28]. Note that because s has values ranging from 0 ($S=0$, no interactions) to 1 ($S\to\infty$), the linear resonance frequency of the bubble cloud is necessarily lower than the single bubble resonance frequency. Following the Bogolyubov-Krylov perturbation method, an approximate solution of Eq. (5) to the first order is obtained in the first subharmonic region ($\omega\simeq 2\omega_0\sqrt{1-s}$):

$$x(\tau)=a\cos(\omega\tau/2+\phi)+\zeta\Lambda\cos(\omega\tau)+a^2(q_1+q_2\cos(\omega\tau+2\phi))+\zeta^2(q_3+q_4\cos 2\omega\tau)+q_5\zeta a\cos(3\omega\tau/2+\phi)+q_6b\zeta\sin\omega\tau, \quad (6)$$

where a and ϕ denote the amplitude and phase of the subharmonic component, and the coefficients Λ and q_i's are given in Appendix A. A set of differential equations for the unknowns a and ϕ is obtained by cancelling the secular terms up the cubic order in Eq. (5). In the steady-state regime, three solutions exist for the amplitude a (together with the corresponding solutions for ϕ), either $a(\omega)=0$, or

$$a(\omega)=\left(\frac{\omega_0^2-\omega^2/4(1-s)+h_1\zeta^2\pm\Delta}{h_0}\right)^{\frac{1}{2}}, \quad (7)$$

with