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# Influences of pressure amplitudes and frequencies of dual-frequency acoustic excitation on the mass transfer across interfaces of gas bubbles<sup>\*</sup>



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#### A R T I C L E I N F O

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#### ABSTRACT

In the present paper, the influence of pressure amplitudes and frequencies of dual-frequency acoustic excitation on the mass transfer across interfaces of gas bubbles are numerically investigated. The size of bubble growth region is proposed as a criterion for the optimization of the dual-frequency approach. Our simulation reveals that a. there exists a particular point at which the threshold of mass transfer is independent of the power allocation between two sonic waves; b. for the promotion of the effects of mass transfer, more energies should be allocated to the low-frequency sonic wave rather than the high-frequency sonic wave; and c. the benefits of dualfrequency approaches decrease with the increase of the frequency ratio of the two sonic waves.

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#### 1. Introduction

Acoustic cavitation can significantly promote and enhance the mechanical, chemical and biological processes in liquids [1–5]. For example, acoustic cavitation plays an important role during non-invasive therapeutic processes using high intensity focused ultrasonic beams by enhancing and monitoring the treatment [5]. Hence, the bubble dynamics and the cavitation effects under acoustic excitation are of great interests to the researchers from many disciplines (e.g., acoustical, biomedical and chemical engineering). To further promote the cavitation effects, a system with multiple (dual or triple) frequencies is being intensively studied [6-25]. Compared with cavitation under singlefrequency acoustic excitation, multiple-frequency system has many advantages, such as facilitation of the nucleation of bubble embryos, enhancement of the bubble growth rate and generation of more violent forces during bubble collapse. Therefore, the multi-frequency system has been employed to increase the intensity of sonoluminescence [6–11], to enhance the efficiency of sonochemical reactors [12–21], to improve the accuracy of ultrasound imaging [22,23] and to advance the techniques of the tissue ablation [24] and the tumor therapy [15].

Compared with the single-frequency acoustic excitation, more parameters (e.g., amplitudes and frequencies of the added acoustic waves, phase differences between component acoustic waves, power allocation between component sonic waves) are involved in the multiplefrequency excitation, leading to a complicated phenomenon. As a large set of parameters is involved, it is essential to optimize these parameters to achieve the best performance of the cavitation. Various criteria have been proposed for this optimization process, e.g., the maximum bubble

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radius during bubble oscillations [10], the minimum bubble radius during bubble collapse [6], the bubble collapse time [19,21] and the maximum velocity of the bubble wall [11]. Unfortunately, those criteria are only applicable to very violent bubble oscillations induced by acoustic excitation with considerably large amplitudes (termed as "transient cavitation" by Neppiras [26]). Currently, there is no criterion available in the literature for the more controllable and stable bubbles when bubbles are excited by acoustic waves with small or medium amplitudes (termed as "stable cavitation" by Neppiras [26]). During this process, mass transfer through rectified diffusion across bubble interfaces plays a key role on the bubble behavior [1,26–29].

In the present study, influences of two paramount parameters (i.e., the pressure amplitudes and the frequencies of external acoustic excitation) of the dual-frequency approach on the mass transfer across bubble-liquid interfaces are studied numerically. The size of the region in which bubbles grow through the effects of mass transfer during stable cavitation is proposed as a criterion for the optimization of the multiple-frequency system.

#### 2. Theory and equations

In this section, the equations employed in the present paper for the simulations of bubble dynamics under dual-frequency acoustic excitation are given. Here, dynamics of spherical gas bubbles in Newtonian, compressible and viscous liquids are considered. The equation of bubble motion with liquid compressibility is [30],

$$\begin{pmatrix} 1 - \frac{\dot{R}}{c_l} \end{pmatrix} R\ddot{R} + \frac{3}{2} \left( 1 - \frac{\dot{R}}{3c_l} \right) \dot{R}^2 = \left( 1 + \frac{\dot{R}}{c_l} \right) \frac{p_{ext}(R, t) - p_s(t)}{\rho_l} + \frac{R}{\rho_l c_l} \frac{d[p_{ext}(R, t) - p_s(t)]}{dt},$$
(1)

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where

$$p_{ext}(R,t) = P_{in} - \frac{2\sigma}{R} - \frac{4\mu_l}{R}\dot{R}, \qquad (2$$

$$P_{in} = \left(P_0 + \frac{2\sigma}{R_0}\right) (R_0/R)^{3\kappa},\tag{3}$$

$$P_{s}(t) = P_{0} + P_{A1}\cos(\omega_{1}t) + P_{A2}\cos(\omega_{2}t).$$
(4)

Here, *R* is the instantaneous bubble radius; the overdot denotes the time derivatives;  $c_l$  is the speed of sound in the liquid;  $\rho_l$  is the density of the liquid; *t* is the time;  $\sigma$  is the surface tension coefficient;  $\mu_l$  is the viscosity of the liquid;  $P_0$  is the ambient pressure; and  $P_{A1}$  and  $P_{A2}$  are the amplitudes of external sound field with angular frequencies  $\omega_1$  and  $\omega_2$  respectively (assuming that  $\omega_1 < \omega_2$  for convenience).

The bubble growth rate can be given as [27],

$$\frac{dR_0}{dt} = \frac{DR_g T_* C_0}{R_0 P_0} \left[ \langle R/R_0 \rangle + R_0 \left( \frac{\left\langle (R/R_0)^4 \right\rangle}{\pi t D} \right)^{1/2} \right] \\
\times \left( 1 + \frac{4\sigma}{3P_0 R_0} \right)^{-1} \left( \frac{C_i}{C_0} - \frac{\left\langle (R/R_0)^4 (P_{in}/P_0) \right\rangle}{\left\langle (R/R_0)^4 \right\rangle} \right).$$
(5)

Here,  $R_g$  is the universal gas constant;  $T_*$  is the ambient temperature in the liquid;  $C_0$  is the saturation concentration of the gas in the liquid;  $C_i$ is the concentration of the gas in the liquid at infinity; D is the diffusion constant; and  $\langle \rangle$  denotes time-averaged terms, which can be determined based on the solution of the equations of bubble motion [i.e., Eqs. (1)–(4)]. Up to the second order of ( $P_{A1}/P_0$ ), the three timeaveraged terms can be described as [25],

$$\langle R/R_0 \rangle = 1 + B_2 (P_{A1}/P_0)^2,$$
 (6)

$$\langle (R/R_0)^4 \rangle = 1 + \left[ 4B_2 + 3 \left( A_{11}^2 + A_{12}^2 \right) \right] (P_{A1}/P_0)^2,$$
 (7)

$$\left\langle \left(R/R_{0}\right)^{4}\left(P_{in}/P_{0}\right)\right\rangle = \left(1 + \frac{2\sigma}{P_{0}R_{0}}\right) \left\langle \left(R/R_{0}\right)^{4-3\kappa}\right\rangle = \left(1 + \frac{2\sigma}{P_{0}R_{0}}\right) \left\{1 + \left[\left(4 - 3\kappa\right)B_{2} + \frac{(4 - 3\kappa)(3 - 3\kappa)}{4}\left(A_{11}^{2} + A_{12}^{2}\right)\right]\left(P_{A_{1}}/P_{0}\right)^{2}\right\},$$

$$(8)$$

where

$$A_{11} = -\frac{P_0}{M\rho_l R_0^2} \left[ \frac{1 + (\omega_1 R_0 / c_l)^2}{(\omega_0^2 - \omega_1^2)^2 + 4\beta_{tot}^2 \omega_1^2} \right]^{1/2},\tag{9}$$

$$A_{12} = -\frac{P_0 P_{A2}}{M \rho_l R_0^2 P_{A1}} \left[ \frac{1 + (\omega_2 R_0 / c_l)^2}{(\omega_0^2 - \omega_2^2)^2 + 4\beta_{tot}^2 \omega_2^2} \right]^{1/2},$$
(10)

$$B_{2} = -\frac{1}{4\omega_{0}^{2}M} \left( A_{11}^{2}\omega_{1}^{2} + A_{12}^{2}\omega_{2}^{2} \right) + \frac{1}{4} \frac{A_{11}^{2} + A_{12}^{2}}{4\rho_{l}R_{0}^{2}\omega_{0}^{2}M} \left[ 3\kappa(3\kappa+1) \left( P_{0} + \frac{2\sigma}{R_{0}} \right) - \frac{4\sigma}{R_{0}} \right],$$
(11)

with

$$\begin{split} \omega_{0}^{2} &= \frac{1}{M\rho_{l}R_{0}^{2}} \bigg[ 3\kappa \bigg( P_{0} + \frac{2\sigma}{R_{0}} \bigg) - \frac{2\sigma}{R_{0}} \bigg], \\ \beta_{tot} &= \frac{2\mu_{l}}{M\rho_{l}R_{0}^{2}} + \frac{R_{0}}{2c_{l}}\omega_{0}^{2}, \\ M &= 1 + \frac{4\mu_{l}}{\rho_{l}R_{0}^{2}} \frac{R_{0}}{c_{l}}. \end{split}$$

Hence, the bubble growth or dissolution rate can be obtained by integration of Eq. (5). According to Eq. (5), there exists a threshold of the pressure amplitude, the value of which corresponds to the balance of the amount of gas diffused into and out of the bubbles [i.e.,  $dR_0/dt = 0$  in Eq. (5)].

#### 3. Results and discussions

In this section, the influences of two paramount parameters (amplitudes and frequencies respectively) of the dual-frequency acoustic excitation on the mass transfer across bubble–liquid interfaces are numerically studied with demonstrating examples. For simplicity, air bubbles in water are considered in the following sections. The constants employed in the simulations are:  $P_0 = 1.013 \times 10^5$  Pa;  $\rho_l = 998.20$  kg/m<sup>3</sup>;  $c_l = 1486$  m/s;  $\mu_l = 0.001$  Pa · s;  $\sigma = 72.8$  dyn/m;  $D = 2.4 \times 10^{-9}$  m<sup>2</sup>/s;  $R_g = 8.314$  J/mol/K;  $T_{\infty} = 293.15$  K;  $\kappa = 1.33$ ;  $\omega_1 = 0.001$  Pa · s;  $\sigma = 72.8$  dyn/m;  $\sigma = 1.013 \times 10^{-9}$  m<sup>2</sup>/s;  $R_g = 8.314$  J/mol/K;  $T_{\infty} = 293.15$  K;  $\kappa = 1.33$ ;  $\omega_1 = 0.001$  Pa · s;  $\sigma = 72.8$  dyn/m;  $\sigma = 0.001$  Pa · s;  $\sigma = 72.8$  dyn/m;  $\sigma = 0.013 \times 10^{-9}$  m<sup>2</sup>/s;  $R_g = 8.314$  J/mol/K;  $T_{\infty} = 293.15$  K;  $\kappa = 1.33$ ;  $\omega_1 = 0.001$  Pa · s;  $\sigma = 0.00$ 



**Fig. 1.** The predicted threshold of the total acoustic pressure amplitude of rectified mass diffusion under single-frequency [marked as " $\omega_1$ " (dotted line) and " $\omega_2$ " (dashed line)] and dual-frequency [marked as " $\omega_1 + \omega_2$ " (solid line), N = 2] acoustic excitation.  $\omega_1 = 5 \times 10^5 s^{-1}$ .  $\omega_2 = 3\omega_1 = 1.5 \times 10^6 s^{-1}$ .  $R_{r1}$  and  $R_{r2}$  are the resonance bubble radii of gas bubbles under acoustic excitation with frequencies  $\omega_1$  and  $\omega_2$  respectively.  $R_s$  and  $R_E$  are the start and the end bubble radii of bubble growth region respectively. For other marks, readers are referred to the text in Section 3.

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