Improvement of acoustic theory of ultrasonic waves in dilute bubbly liquids

Keita Ando, Tim Colonius, and Christopher E. Brennen
Division of Engineering and Applied Science, California Institute of Technology, Pasadena, California 91125
kando@caltech.edu, colonius@caltech.edu, brennen@caltech.edu

Abstract: The theory of the acoustics of dilute bubbly liquids is reviewed, and the dispersion relation is modified by including the effect of liquid compressibility on the natural frequency of the bubbles. The modified theory is shown to more accurately predict the trend in measured attenuation of ultrasonic waves. The model limitations associated with such high-frequency waves are discussed.

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1. Introduction

The acoustics of bubbly liquids have been extensively studied for many years. The traditional theory for a dilute bubbly mixture assumes that mutual interactions among the bubbles are negligible. The bubble/bubble interactions can never be ignored at resonance even in the dilute limit,[1,2] and the theory is known to overestimate attenuation under the resonant condition. The theory is also known to be inaccurate in estimating the attenuation of ultrasonic waves. So far, to the authors' knowledge, the cause of the discrepancy under the ultrasonic condition has not been revealed.

Herein, we briefly review the theory and modify the dispersion relation by including the effect of liquid compressibility on the natural frequency of the bubbles and validate this modified theory by comparing to experimental data. Finally, we discuss the model limitations associated with ultrasonic waves.

2. Review of the theory

In the classic papers of Foldy[3] and Carstensen and Foldy,[4] wave propagation in a bubbly mixture was treated as a problem of multiple scattering by randomly distributed isotropic scatterers representing the spherical bubbles, and the dispersion relation for the mixture was derived. An alternative approach is to treat the mixture as a single phase (continuum) medium. van Wijngaarden[5] defined volume-averaged quantities in order to remove the local fluctuations due to scattering and derived the averaged equations based on heuristic, physical reasoning. By linearizing van Wijngaarden's equations, Commander and Prosperetti[2] derived the dispersion relation

$$\frac{1}{c_m^2} = \frac{1}{c_l^2} + 4\pi n \int_0^\infty \frac{R_0 f(R_0) dR_0}{\omega_N^2 - \omega^2 + i2\delta\omega}, \tag{1}$$

where c_m is the complex sonic speed in the mixture, c_l is the sonic speed in the liquid alone, n is the total bubble number density, δ is the bubble-dynamic damping constant, ω is the temporal angular frequency ($\omega = 2\pi f$), ω_N is the natural frequency of the bubbles, R_0 is the equilibrium bubble radius, and $f(R_0)$ is the density function for the size distribution of the equilibrium bubble radius, which satisfies $\int_0^\infty f(R_0) dR_0 = 1$. In the derivation of the dispersion relation (1), the void fraction,

$$\alpha = \frac{4\pi}{3} n \int_0^\infty R_0^3 f(R_0) dR_0, \tag{2}$$

is assumed to be small ($\alpha \ll 1$) and relative motion between the phases is ignored. The relative motion has been shown to have minimal impact on the acoustic problem.[6]

To complete the dispersion relation (1), the bubble-dynamic constant and the natural frequency need to be specified. Commander and Prosperetti[2] used the following expressions for δ and ω_N,

$$\delta = \frac{2\mu_l}{\rho_l R_0^2} + \frac{p_{g0}}{2\rho_l \omega R_0^2} \Im\{\Upsilon\} + \frac{\omega^2 R_0}{2c_l}, \tag{3}$$

$$\omega_N^2 = \frac{p_{g0}}{\rho_l R_0^2} \left(\Re\{\Upsilon\} - \frac{2S}{p_{g0} R_0} \right). \tag{4}$$

Here, μ_l is the liquid viscosity, S is the surface tension, and p_{g0} is the internal (gas) bubble pressure (vapor pressure is typically negligible) given by $p_{g0}=p_{l0}+(2S/R_0)$, where p_{l0} is the ambient pressure. The quantity Υ is a function of the Peclet number $\mathrm{Pe}=\omega R_0^2/\alpha_{\mathrm{th}}$, where α_{th} is the thermal diffusivity of the gas,

$$\Upsilon = \frac{3\gamma}{1 - i3(\gamma-1)\mathrm{Pe}^{-1}(\sqrt{i\mathrm{Pe}} \coth \sqrt{i\mathrm{Pe}} - 1)}, \tag{5}$$

where γ is the ratio of specific heats. The effective polytropic index for thermal behavior of the gas is then given by $\kappa=\Re\{\Upsilon\}/3$. Since $\kappa \to 1$ as $\omega \to 0$ or $\mathrm{Pe} \to 0$, the isothermal natural frequency (defined below) is obtained in the quasistatic limit and is generally very close to the resonant frequency.

$$\omega_N^2 \big|_{\kappa=1} = \frac{p_{g0}}{\rho_l R_0^2} \left(3 - \frac{2S}{p_{g0} R_0} \right). \tag{6}$$

It should be noted that the effect of liquid compressibility is ignored in Eq. (4) and is negligible unless the frequency is extremely high compared to the resonant frequency.[7]

We define the phase speed V and attenuation A (in decibels per unit length) as

$$V = \left[\Re\left\{ \frac{1}{c_m} \right\} \right]^{-1}, \tag{7}$$

$$A = -20(\log_{10} e)\omega \Im\left\{ \frac{1}{c_m} \right\}. \tag{8}$$

The estimated phase velocity (7) is known to yield quantitative agreement with experimental data in a wide frequency range below and above the resonance.[2,8] However, the estimated attenuation (8) under resonant and ultrasonic conditions appears to deviate from the experimental values.

Before concluding this review, we examine the model limitations. In order that the mixture be considered homogeneous and the wave structure be well resolved, we need to choose a physically appropriate averaging volume, $\Delta\mathcal{V}$, and presuppose the scale separation[9]

$$l = n^{-1/3} \ll \Delta\mathcal{V}^{1/3} \ll L, \tag{9}$$

where l is the mean bubble spacing and L is the wavelength in the mixture. Note that $R_0 \ll l$ in the dilute limit (i.e., $\alpha \to 0$). Since the wavelength of ultrasonic waves may be comparable to or shorter than the mean bubble spacing, the continuum model may be invalid. In addition, neglect of the acoustic contribution to the bubble natural frequency (4) may also give rise to a discrepancy in theory and experiment between the attenuation of high-frequency waves. In Sec. 3, we discuss the effect of liquid compressibility on the attenuation of ultrasonic waves.

3. Modification to the theory

3.1 Linearized dynamics of the spherical bubbles

To obtain the formulas for the bubble-dynamic damping constant and the bubble natural frequency, we need to linearize the spherical bubble dynamics. It follows from Prosperetti[7] and Prosperetti *et al.*[10] that the linearized dynamics are described by

$$\ddot{x} + 2\delta\dot{x} + \omega_N^2 x = -\epsilon \frac{p_{l0}}{\rho_l R_0^2} e^{i\omega t}, \tag{10}$$

where ϵ is the infinitesimal amplitude of sinusoidally oscillating (farfield) liquid pressure ($|\epsilon| \ll 1$) and x is the corresponding perturbation in the bubble radius ($|x| \ll 1$):

$$p_l = p_{l0}(1 + \epsilon e^{i\omega t}), \tag{11}$$

$$R = R_0(1 + x). \tag{12}$$

Here, the damping constant and the natural frequency are

$$\delta = \frac{2\mu_l}{\rho_l R_0^2} + \frac{p_{g0}}{2\rho_l \omega R_0^2}\Im\{\Upsilon\} + \frac{\dfrac{\omega^2 R_0}{2c_l}}{1 + \left(\dfrac{\omega R_0}{c_l}\right)^2}, \tag{13}$$

$$\omega_N^2 = \frac{p_{g0}}{\rho_l R_0^2}\left(\Re\{\Upsilon\} - \frac{2S}{p_{g0} R_0}\right) + \frac{\left(\dfrac{\omega R_0}{c_l}\right)^2}{1 + \left(\dfrac{\omega R_0}{c_l}\right)^2}\omega^2, \tag{14}$$

where the last terms on the right-hand sides of the above equations represent the contributions associated with liquid compressibility. To quantify the impact of liquid compressibility on the ultrasonic waves, we compute the dispersion relation (1) based on Eqs. (13) and (14) instead of Eqs. (3) and (4) and validate the modification by comparing to experimental data below.

For future use, we develop the asymptotic limits of Eqs. (13) and (14). In the quasi-static limit (i.e., $\omega \to 0$), it follows from Prosperetti[7,11] that

$$\delta \approx \frac{2\mu_l}{\rho_l R_0^2} + \frac{\gamma - 1}{10\gamma}\frac{p_{g0}}{\rho_l \alpha_{\mathrm{th}}}, \tag{15}$$

$$\omega_N \approx \omega_N|_{\kappa=1}, \tag{16}$$

where $\omega_N|_{\kappa=1}$ is the isothermal natural frequency (6) so that liquid compressibility is unimportant. On the other hand, in the limit of $\omega \gg \omega_N|_{\kappa=1}$, it is readily shown that

$$\delta \approx \frac{c_l}{2R_0}, \tag{17}$$

$$\omega_N \approx \omega. \tag{18}$$

In this limit, the damping due to liquid compressibility dominates over the viscous and thermal contributions and the natural frequency is independent of the bubble size.

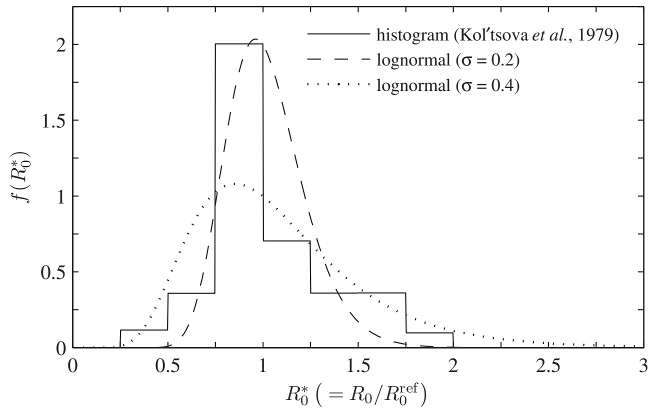

Fig. 1. Normalized histogram of the bubble size distribution of Kol'tsova *et al.* (Ref. 12) and lognormal distributions with a standard deviation σ. The probable size, R_0^{ref}, is set to be 20 μm. The measured values are based on a hydrogen/water mixture of α=0.03% at 15 °C and 1 atm.

3.2 Validation of the modification

We compare the dispersion relation (1) to the experiment of Kol'tsova *et al.*[12] who measured the attenuation in a high-frequency range up to 30 MHz. In those experiments, hydrogen bubbles were produced using electrolysis and had a size distribution with a mean radius of 15–20 μm. The histogram of the size distribution for α=0.03% (probable size, R_0^{ref}=20 μm) is plotted in Fig. 1. We assume that the size distribution for different values of α is similar to that in Fig. 1. The actual distribution may be smooth, as shown in Fig. 1, and we model it using a lognormal density function with standard deviation σ,

$$f(R_0^*) = \frac{1}{\sqrt{2\pi}\sigma R_0^*} \exp\left(-\frac{\ln^2 R_0^*}{2\sigma^2}\right), \tag{19}$$

where $R_0^* = R_0/R_0^{\mathrm{ref}}$.

Using the size distributions in Fig. 1, the phase velocity (7) and attenuation (8) are computed using Eqs. (3) and (4) or Eqs. (13) and (14) and are plotted in Fig. 2. The attenuation of Kol'tsova *et al.*[12] is also plotted (α=0.004%, $\omega_N|_{\kappa=1}/2\pi$=0.142 MHz for R_0^{ref}). It follows

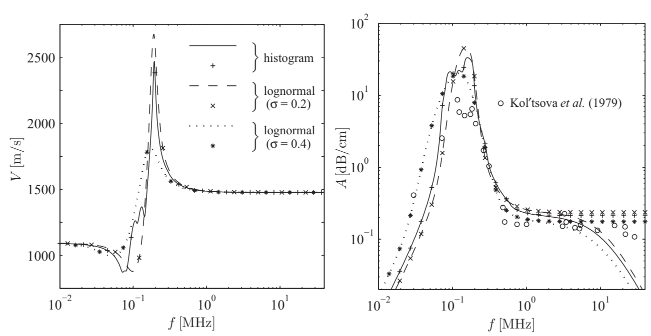

Fig. 2. Phase velocity (left) and attenuation (right) for a hydrogen/water mixture of α=0.004% and R_0^{ref}=20 μm at 15 °C and 1 atm. The lines and symbols (plus, cross, and asterisk) are computed using the dispersion relation (1) with Eqs. (3) and (4) and with Eqs. (13) and (14), respectively. The symbols (circle) denote the experimental data of Kol'tsova *et al.* (Ref. 12).

from the phase velocity plot that the present modifications to δ and ω_N have negligible impact on V. It is also found that the size distribution tends to smooth the transition in V at the resonant frequency.

However, the present modification does lead to a striking change in the attenuation for $\omega \gg \omega_N|_{\kappa=1}$. The dispersion relation (1) with the present modification predicts attenuations at high frequencies that agree with the experimental measurements. That is, liquid compressibility has major impact on the attenuation of the ultrasonic waves. As a result of Eqs. (17) and (18), the phase velocity and the attenuation for $\omega \gg \omega_N|_{\kappa=1}$ asymptote to the constant values,

$$V \approx c_l, \tag{20}$$

$$A \approx 20(\log_{10} e)\frac{3\alpha}{2R_0^{\mathrm{ref}}}C_1, \tag{21}$$

where the constant C_1 is

$$C_1 = \frac{\int_0^\infty R_0^{*2} f(R_0^*)dR_0^*}{\int_0^\infty R_0^{*3} f(R_0^*)dR_0^*}. \tag{22}$$

For the lognormal $f(R_0^*)$, $C_1=\exp(-2.5\sigma^2)$ so that the attenuation decreases as σ increases. It should be pointed out that Kol'tsova *et al.*[12] presented the different data sets of the attenuation (with different void fractions) which remains almost constant under the ultrasonic condition. It is therefore concluded that the modified theory is superior to the previous theory when it comes to predicting this trend.

Furthermore, we notice that the size distribution increases the attenuation below the resonant frequency. From Eqs. (15) and (16), the asymptotic values at low frequency become

$$V \approx \frac{c_l}{\sqrt{1 + \frac{\alpha \rho_l c_l^2}{p_{l0}}}}, \tag{23}$$

$$A \approx 20(\log_{10} e)\frac{\alpha \rho_l^2 V \delta \omega^2}{3p_{l0}^2}C_2, \tag{24}$$

where the constant C_2 is

$$C_2 = \frac{\int_0^\infty R_0^{*5} f(R_0^*)dR_0^*}{\int_0^\infty R_0^{*3} f(R_0^*)dR_0^*}. \tag{25}$$

Here, we have neglected the viscous contribution in Eq. (15) since the thermal damping generally dominates over the viscous damping. In addition, it is assumed that the surface tension is negligible in Eq. (16). For the lognormal $f(R_0^*)$, $C_2=\exp(8\sigma^2)$ so that the attenuation increases as σ increases. To interpret this tendency, consider linear bubble oscillations under a sinusoidal forcing ($p_l - p_{l0} \propto \sin(\omega t)$) of the farfield liquid pressure. The corresponding perturbation in the bubble radius oscillates with the forcing frequency ω and with a phase shift ϕ such that

$$\cos\phi = \frac{\omega_N^2(R_0) - \omega^2}{\sqrt{(\omega_N^2(R_0) - \omega^2)^2 + 4\delta^2(R_0)\omega^2}}. \tag{26}$$

Therefore, phase cancellations due to the different phases among the different-sized bubbles occur in the low-frequency regime since $\omega_N \approx \omega_N|_{\kappa=1} \neq \omega$. The phase cancellations can be regarded as apparent damping of the wave propagation in the polydisperse mixture, and the damping mechanism becomes more effective as the bubble size distribution broadens.[13,14] As a result, the size distribution increases the attenuation, as seen in Fig. 2. However, in the ultrasonic limit, all the different-sized bubbles oscillate with the same phase due to the fact that $\omega_N \approx \omega$ (regardless of the bubble sizes); hence, in this case, the phase cancellations do not occur.

Finally, we check the model limitation (9). The mean bubble spacing in Fig. 2 is

$$l = \sqrt[3]{\frac{4\pi\int_0^\infty R_0^{*3} f(R_0^*) dR_0^*}{3\alpha}} R_0^{\mathrm{ref}} \approx 1100\ \mu\mathrm{m}, \tag{27}$$

where the histogram in Fig. 1 is used for $f(R_0^*)$. At $f=10$ MHz, the wavelength is approximated by

$$L \approx \frac{c_l}{f} \approx 150\ \mu\mathrm{m}, \tag{28}$$

which is larger than the mean bubble radius but shorter than the mean bubble spacing. Hence, the continuum assumption is invalidated, while bubble/bubble interactions may be ignored. However, despite this violation, as seen in Fig. 2, the continuum theory accurately predicts the trend in the attenuation around $f=10$ MHz. This implies that the validity of the dispersion relation (1) may extend beyond the limitation (9).

4. Conclusion

A modification to the traditional dispersion relation of linear waves in dilute bubbly liquids is made to take into account the effect of liquid compressibility (which is very important far above the resonant frequency) on linearized dynamics of spherical bubbles. The modified dispersion relation is found to accurately predict the trend in measured attenuation of ultrasonic waves. The agreement between the modified theory and experiment implies that the validity of the dispersion relation (1) may extend beyond the continuum model limitation (9).

Acknowledgments

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