Investigation of the nonlinear propagation of ultrasound through a bubbly medium including multiple scattering and bubble-bubble interaction: Theory and experiment

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Abstract—Understanding of the propagation of ultrasound through a bubbly medium is a challenging task because of the nonlinear dynamics of the bubbles and their effect on the attenuation and sound speed of the medium. The majority of the studies on this subject apply linear models, which will generate inaccurate results, especially at higher-pressure excitations. These studies have also ignored the effect of bubble-bubble interaction and nonlinear multiple scattering.

In this work, we have numerically simulated the attenuation and sound speed of a bubbly medium by solving our recently developed nonlinear model. An efficient method to investigate the nonlinear bubble-bubble interaction and multiple scattering is developed, and this phenomenon is included the numerical investigations through considering a cluster of 130 randomly distributed interacting bubbles with sizes derived from experimental measurements. Broadband experimental attenuation measurements of monodisperse lipid-coated microbubble solutions were performed with peak acoustic pressures ranging within 10-100kPa. The bubble solutions had mean diameters of 4-6 micron and peak concentrations of 1000 to 15000 bubbles/ml.

At lower concentrations (with minimal bubble-bubble interactions), predictions of the model (attenuation and sound speed vs frequency) in the absence of interaction are in good agreement with experimental measurements. At higher concentrations, secondary peaks in the attenuation and sound speed diagrams as a function of frequency appear. Through considering the bubble-bubble interactions, the numerical results can predict the quantitative and qualitative changes in the attenuation and frequency as well as the generation of secondary peaks.

I. Introduction

Acoustically excited bubbles exhibit nonlinear and chaotic dynamics [1-5]. The investigation of the nonlinear dynamics of the bubbles has been the focus of numerous studies [1-5]. In spite of the complex and nonlinear dynamics, bubbles are one of the important parts of several applications and disciplines ranging from underwater acoustics [6-7] and sonochemistry [8] to diagnostic and therapeutic applications of ultrasound [9].

Knowledge on the behavior of the bubbles is key in controlling and optimizing the phenomena and applications associated with bubble dynamics. However, the nonlinear dynamics of the bubbles is complex and is not fully understood. The presence of the stabilizing shell (e.g. albumin, polymer or lipid), adds to the complexity of the bubble behavior by introducing the nonlinear shell behavior (e.g. buckling and rupture [10]).

Additionally, the presence of the bubbles in an ultrasound field changes the attenuation and sound speed of the medium. The changes in the attenuation and sound speed are nonlinear and they depend on the nonlinear oscillations of the bubbles. Several models have been developed to understand the effect of bubble oscillations on the attenuation and sound speed; however, the majority of the studies employ linear models (e.g. 11) which are only valid for small amplitude bubble oscillations. Application of linear approximations can lead to inaccurate predictions; this is because most of the applications employ higher acoustic pressures which results in nonlinear large amplitude bubble oscillations. To address this problem, various models have been developed recently [6-8,12]. Although the models in [6,8] employ the nonlinear oscillations of the bubbles by incorporating the nonlinear scattering cross section of the bubble; however, they still employ the linear approximations for the other damping factors (e.g. thermal, viscous, etc.). Additionally, they neglect the influence of the changes of the sound speed. The model in [8] employs the nonlinear changes of the damping factors; however, it still employ the linear approximations for the changes of the sound speed.

In addition, the existence approaches, neglect the effect of the bubble-bubble interaction. Bubble-bubble interaction and multiple scattering are nonlinear and significantly influence the oscillations of the bubbles [13-15]. Effect of the bubble-bubble interaction becomes more significant for higher concentrations (due to smaller bubble-bubble distances). To accurately predict the attenuation and sound speed of the bubbly medium, the large amplitude nonlinear oscillations of the bubbles as well as the interaction between the bubbles should be included in the model.

Recently we have developed a model that incorporated the nonlinear large oscillations of the bubbles [16]. The predictions of the model have been validated with the predictions of the existing models [16] as well as with experiments [17]. Additionally, we have recently developed an efficient approach to simulate the dynamics of large clusters of interacting bubbles [14, 15].

In this work, we will investigate the changes of the attenuation and sound speed through solving the nonlinear model [16] and incorporating the bubble-bubble interactions. Broadband experimental attenuation measurements of monodisperse lipid-coated microbubble (MB) solutions will be performed with peak acoustic pressures ranging within 10-100kPa for various concentrations of MBs.

II. METHODS

A. Numerical methods

The Marmattont model [10] was modified by adding the interaction term and then used to numerically investigate the oscillations of the lipid coated interacting MBs:

$$R_i\ddot{R}_i + \frac{3}{2}\dot{R}_i^{\,2} = A_i - \sum_{\substack{j=1 \\ j\neq i}}^{N} \frac{R_j}{d_{ij}}\left(R_j\ddot{R}_j + 2\dot{R}_j^2\right) \qquad i = 1,2,\dots,N \tag{1}$$

Where A_i is given by

$$A_i = \frac{1}{\rho}\left(\left[P_0 + \frac{2\sigma(R_{i0})}{R_{i0}}\right]\left(\frac{R_{i0}}{R_i}\right)^{3\gamma}\left(1 - \frac{3\gamma}{c}\dot{R}_i\right) - \frac{2\sigma(R_i)}{R_i} - \frac{4\mu\dot{R}_i}{R_i} - \frac{4\kappa_s\dot{R}_i}{R_i^2} - P_0 - P_a\sin\omega t\right) \tag{2}$$

Here R_0 is the initial radius, ρ_L is the density of the liquid, P_0 is the equilibrium pressure inside the bubble, Γ is the polytropic exponent, μ_L is the viscosity of the surrounding liquid, μ_L is the liquid viscosity, κ_s is the surface dilatational viscosity of the shell and d_{ij} is the distance between MB (i) and (j). The initial surface tension is given by equation 3:

$$\sigma(R) = \begin{cases} 0, & \text{if } R \leqslant R_{\text{buckling}} \approx R_0 \\ \chi\left(R^2/R^2_{\text{buckling}} - 1\right), & \text{if } R_{\text{buckling}} \leqslant R \leqslant R_{\text{breakup}}. \\ \sigma_{\text{water}} = 0.072\ \text{N m}^{-1}, & \text{if } R > R_{\text{rupture}} \end{cases} \tag{3}$$

where χ is the shell elasticity and:

$$R_b = \frac{R_0}{\sqrt{1 + \frac{\sigma(R_0)}{\chi}}}$$

$$R_r = R_b\sqrt{1 + \frac{\sigma_{break-up}}{\chi}}$$

The dynamics of the interacting MBs, were numerically simulated for the sizes derived from the experimental measurements and pressures of (1kPa-100 kPa) within a frequency range of 500 kHz to 3 MHz. The attenuation and sound speed was then, calculated through solving equation equations 4 and 5 [16]:

$$< \Re(k^2) >= \frac{-\omega^2}{{C_l}^2} - \frac{\rho_l}{T * 0.5 * |P|^2}\int_0^T \Re(P)\frac{\partial^2\beta}{\partial t^2}dt \tag{4}$$

$$< \Im(k^2) >= -\frac{\rho_l}{T * 0.5 * |P|^2}\int_0^T \Im(P)\frac{\partial^2\beta}{\partial t^2}dt \tag{5}$$

The integration was performed and summed over the ranges of the sizes derived from the monodisperse populations in the experiments.

B. Experiments

The monodisperse lipid coated MBs were produced using the method described in [18-20]. Broadband experimental attenuation measurements (with 2.25 MHz center frequency) of the monodisperse lipid-coated microbubble solutions were performed with peak acoustic pressures ranging within 10-100kPa using the method described in [18-20]. The bubble solutions in our experiments had mean diameters of 4-6 micron and peak concentrations of 1000 to 15000 bubbles/ml. Figure 1, shows a sample of the produced lipid coated MBs. This sample was used for the experimental measurements in this paper. Desired concentrations were achieved by injecting 5 µl (sample 1) and 45 µl (sample 2) of this sample in a holder of 2.5*4.5*4.5 cm.

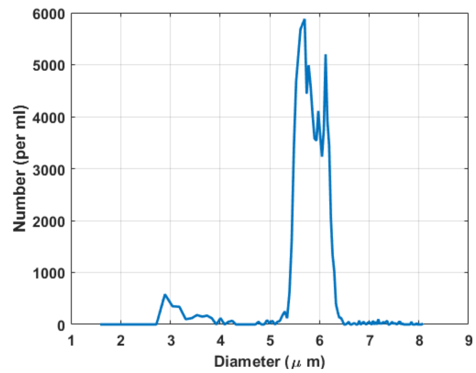

Figure 1: The size distribution of the lipid coated MBs in our experiments.

The numerical simulations were performed by solving equation 1, for 10 formations of 130 randomly distribuetd MBs in a cube of different lenghts (the lebght of the cubes were chosen to match the concenterations in the experiments) at each freqeuncy and pressure. The interaction between the MBs were simulated using the method described in [14-15] Then, equations 4 and 5 were solved and integrated over the considered MB sizes to calucate the attenuation and sound speed.

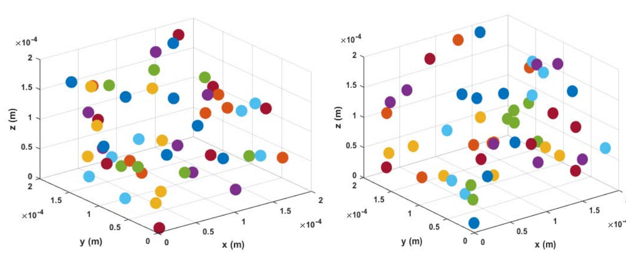

Figure 2: Two different random formations derived from Fig. 1. For simplicty we have shown only 50 MBs in each case.

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III. RESULTS

Figures 3a-b show the measured attenuation of the solutions of sample 1 and sample 2 at acoustic pressures of 12.5, 25, 50 and 100 kPa.

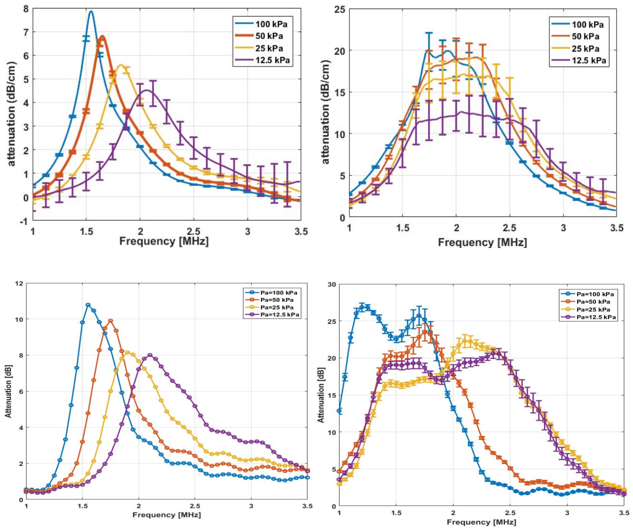

Figure 3: Attenuation of the bubbly medium at various pressures. a) Experimentally measured attenuation of sample 1, b) Experimentally measured attenuation of sample 2, c) Numerically calculated attenuation of sample 1 and d) Numerically calculated attenuation of sample 2.

For the lower concentration (Fig. 3a, sample 1), there is a clear attenuation peak at each pressure. As the pressure increases the frequency of the maximum peak (resonance frequency) decreases (~ 2.15 MHz to ~1.55 MHz) and the magnitude of the peak increases (4.6 dB/cm to ~ 8 dB/cm). For the higher concentration (Fig 3b , sample 2), the interaction between MBs changes the dynamics of the cloud and the attenuation peaks broadens such that there is no identifiable attenuation peak at the investigated pressures. At the higher pressure (100 kPa), secondary peaks are generated in the attenuation diagrams vs frequency. Compared to Fig. 3a, the magnitude of the attenuation increases nonlinearly with concentration increase.

Results of the numerical simulations are in good agreement with the experimental measurements (Fig 3c-d). Fig 3c shows the results of simulations for the concentration of sample 1. The results of the simulations are very identical to the case where the interaction between the MBs is neglected (not shown here). Fig. 3d shows the simulated attenuation for the sample 2. By considering the MB-MB interaction and the concentration of sample 2, the model predicts the qualitative changes of the attenuation such as the broadening of the attenuation peak and the generation of secondary peaks at higher pressures (100 kPa). These changes were not observed when the MB-MB interaction was not incorporated in the model.

IV. DISCUSSION

The presence of the MBs in a medium changes the attenuation and sound speed of the medium. The changes in the attenuation and sound speed are nonlinear and depend on frequency and pressure of the ultrasonic pulse, nonlinear oscillations of the MBs and the interaction between the MBs.
In this work, we have investigated the effect of pressure and frequency and MB interactions both theoretically and experimentally. We have shown that, at lower concentrations (with minimal bubble-bubble interactions), predictions of the nonlinear model (attenuation and sound speed vs frequency) in the absence of interaction are in good agreement with experimental measurements. At each investigated pressure (10 kPa-100 kPa); the bubbly medium has one narrow peak, which indicates the resonance frequency. At higher concentrations, the attenuation peak broadens such that there is no distinguishable peak at pressures below 50 kPa. At 100 kPa, secondary peaks in the attenuation and sound speed diagrams as a function of frequency appear. Through considering the bubble-bubble interactions, the numerical results can predict the quantitative and qualitative changes in the attenuation and frequency as well as the generation of the secondary peaks.

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