# Behaviour of a bubble cluster in an ultrasound field

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#### SUMMARY

Ultrasound medical applications, such as an ultrasound imaging with micro-bubble contrast agents, and high intensity focused ultrasound (HIFU) therapy have attracted much attention in recent years. These applications have a close relation to the motion of micro-bubbles, so that it is essential to understand their dynamics. The bubble motion is influenced by the internal phenomena and its interaction with the surrounding medium, such as the thermal diffusion, the mist formation, the mass diffusion, the heat and mass transfer through the bubble wall. It is important that the oscillation of micro bubbles generates a strong acoustic pressure for these medical applications. The strong pressure also appears in the case of a bubble cluster, like a cloud cavitation. As these dynamics of bubbles are strongly influenced by the thermal phenomena inside them, it is necessary to construct the model taking these phenomena into account to analyse the behaviour of the bubble cluster precisely. Thus, the following effects are considered: the evaporation and condensation of the liquid at the bubble wall, the heat transfer through the bubble wall, and the compressibility of the liquid. Then the spherical bubble cluster is numerically simulated. When the frequency of the ultrasound is sufficiently high, the bubble cluster hardly oscillates. On the contrary, when the frequency of the ultrasound is at the resonance of the bubble cluster, the pressure wave generates the shock wave and it focuses to the cluster centre. As a result, the pressure inside the bubble at the cluster centre becomes much higher than that of a single bubble. Though this extreme high pressure causes the severe cavitation erosion, it is thought that this high energy concentration has the potential to be utilized for medical applications. Copyright  $\odot$  2004 John Wiley & Sons, Ltd.

KEY WORDS: micro bubble; bubble cluster; ultrasound; non-linear acoustics; medical application

# 1. INTRODUCTION

Ultrasound is widely applied in the clinical field today, such as ultrasonography, extracorporeal shock wave lithotripsy (ESWL), high intensity focused ultrasound (HIFU), sonodynamic therapy. Some of these have close relation to the dynamic behaviour of micro bubbles. In an ultrasound imaging, micro bubbles are used as contrast agents. In ESWL, the focusing of the

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shock wave causes the cavitation and the impact pressure with bubble collapse damages not only the stone but also the surrounding body tissues. It is required to understand precisely the amplitude and the power spectrum of acoustic emission from micro bubbles to visualize the tissues and organs clearly and to prevent the tissue damages accompanied with the lithotomy.

Many researchers have investigated the motion of a single bubble in the infinite liquid and there are also many studies where additional effects, such as the compressibility of the liquid, deformation of the bubble from the spherical shape near a solid wall and thermal phenomena are considered. It is well understood that the thermal phenomena inside the bubble significantly influence the cavitation process. Fujikawa and Akamatsu [1] discussed the effects of the non-equilibrium phase change at the bubble wall on the bubble motion. Nigumatulin *et al.* [2] and Kamath and Prosperetti [3] analysed the bubble motion numerically taking into accounts thermal diffusion and so on. Matsumoto and Takemura [4] simulated the collapsing bubble motion numerically by using the full equations for the mass, momentum and energy in the gas and liquid phases. Takemura and Matsumoto [5] calculated the bubble growth with the surrounding pressure reduction by taking into account the internal phenomena such as the thermal and mass diffusion and mist formation due to the homogeneous condensation.

Not only a single bubble but also a bubble cloud has been investigated previously. Mørch [6] has shown that the collapse of the cavitating bubble cloud forms an inward propagating shock wave and its focusing causes severe cavitation damage. Omta [7] studied the oscillation of the bubble cloud and described the relationship between the oscillation and the acoustic emission. Chahine and Duraiswami [8] investigated the dynamics of the multi-bubble cloud consisting of several bubbles and compared with analytical results obtained by asymptotic expansions. Reisman *et al.* [9] investigated the dynamics and acoustics of the cloud cavitation. They suggested that the formation and focusing of bubbly shock waves, which are formed during the collapse of a cloud, are responsible for the severe noise and damage potential associated with this form of cavitating flows. Wang [10] studied the dynamics of a spherical cloud of bubbles with the nuclei size distribution and showed that the strength of the shock and the relaxation behind the shock front are suppressed due to the effects of multiple bubble sizes. The behaviour of cavitation cloud has been investigated in connection with the severe cavitation damage [11, 12] using the set of governing equations for the spherical bubble cloud, where the internal phenomena of each bubble and the compressibility of liquid are taken into account. Inwardly propagating shock wave, which associates a precursor, is formed during the collapse of the bubble cloud and focused in the cloud centre.

In this paper, the single bubble dynamics and the bubble cluster dynamics are discussed. The simulation of the single bubble motion is extended to the pressure wave phenomena coupled with the averaged equation for a bubbly liquid, where thermal phenomena inside the bubble and the liquid compressibility are taken into account. Using this set of equations, the dynamics of bubble cluster is investigated. The numerical results reveal that the pressure waves emitted from bubbles in the cluster centre are amplified when the surrounding pressure oscillates in the resonant frequency of the cluster. The maximum pressure emitted from the bubble at the cluster centre becomes much higher than that of a single bubble and a high frequency pressure oscillation is observed in the centre region. However, the maximum pressure of cluster is much lower than that of a single bubble when the applied ultrasound frequency is at the natural frequency of a single bubble.

#### 2. A MICRO BUBBLE OSCILLATION IN AN ULTRASOUND FIELD

#### *2.1. Non-linear oscillation of a micro bubble*

Non-linear behaviour of a micro bubble in an ultrasound field is investigated. The ultrasound frequency is 1 MHz and the amplitude is 100 kPa. The initial bubble radius,  $R_{b0}$  is 3  $\mu$ m, which is a typical bubble radius of the contrast agent for the medical ultrasound imaging. The initial ambient pressure is 100 kPa, and the initial temperature is 293 K.

The bubble oscillation in an ultrasound field is simulated by using the numerical model  $[12]$ , where the following phenomena are taken into account: (1) effect of the compressibility of the surrounding bubbly liquid and the phase change at the bubble wall for the bubble motion. (2) change of the pressure and temperature inside the bubble due to the internal phenomena, such as non-equilibrium heat and mass transfer by evaporation and condensation on the bubble wall, diffusion between vapor and non-condensable gas and mist formation by homogeneous nucleation, its growth, evaporation and deposition onto the bubble wall.

The following assumptions are employed to formulate the above model: (1) The bubble maintains spherical shape. (2) The pressure and temperature inside the bubble are uniform except the thin boundary layer near the bubble wall, which is thin compared with the bubble radius. (3) Vapour and non-condensable gas obey the perfect gas law. (4) The temperature of the bubble wall is equal to that of the liquid. (5) Non-condensable gas obeys the Henry's law at the bubble wall. (6) Coalescence and fragmentation of the mist and slippage between the mist and the surrounding gas are ignored.

The simulated results are shown in Figure 1. The abscissa is time and the ordinates are the normalized radius, ambient pressure and acoustic emission from the bubble in Figure 1(a). The abscissa is the frequency and the ordinate is the amplitude in Figure 1(b). In the FFT (fast Fourier transform) analysis, the Hamming window function is employed. The acoustic pressure emitted from a bubble is estimated from the bubble oscillation taking into account the liquid compressibility [1] at the bubble surface.

Due to the non-linearity of the bubble motion, the time history of the bubble radius has a cusp shape at the minimum point. In other words, the bubble oscillates repeatedly with the cycle consisted of slow expansion, rapid shrinkage and rebound. In this case, the bubble oscillates resonantly with the ultrasound field. The phase of the bubble motion is  $\pi/2$  backward



Figure 1. Time history of the bubble oscillation: (a) time history of the ambient pressure, bubble radius and acoustic pressure emitted from the bubble; and (b) FFT analysis of the acoustic pressure emitted from the bubble.

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of the surrounding pressure. The acoustic pressure from a micro bubble shows a high pressure when a bubble shrinks and rebounds.

The waveform has an impulsive shape and it contains the higher harmonic components [13]. The FFT analysis shows the second harmonics has similar amplitude to the fundamental frequency of 1 MHz. Utilizing of this second harmonics mode excited by the volumetric oscillation of a micro bubble is now widely applied to the ultrasound contrast imaging in a medical field.

# *2.2. Acoustic turbulence from a micro bubble*

The bubble motion in an ultrasound field shows the chaotic behaviour due to the non-linearity of a bubble motion when the sound amplitude is increased. This phenomenon is called Acoustic Turbulence [14]. The bifurcation and chaotic behaviours of the bubble oscillation have been calculated assuming the interior gas obeys a polytropic relationship which neglects thermal dissipation [15].

To investigate such behaviour in the case of micro bubbles, the bubble motion is simulated when the ambient ultrasound pressure increases linearly from 0 to 1 MPa in 5000 cycles. The initial bubble radius is  $2 \mu m$ , and the ultrasound frequency is 1.34 MHz, which is the 80% of the natural frequency of  $2 \mu m$  micro bubble. Other conditions are the same with the former analysis.

Figure 2 shows the time history of the power spectrum of the acoustic emission from a micro bubble. The abscissa indicates the ambient ultrasound pressure corresponding to time and the ordinate indicates the normalized frequency. When the ultrasound irradiation starts, the bubble begins its oscillation in the fundamental mode. As the increase of the ultrasound amplitude, the emitted acoustic pressure increases and it gradually involves non-linearity with the higher



Figure 2. Power spectrum of the acoustic pressure from the bubble.

harmonics modes. The bifurcation occurs and the  $1/2$  harmonic modes begin to appear in each higher harmonics modes, when the ambient pressure reaches up to 200kPa. Further bifurcation appears and the  $1/4$  harmonics modes emerge at  $400$  kPa. After these processes, the micro bubble oscillation becomes chaotic. When the ambient ultrasound pressure reaches around 650 kPa the windows are formed for a short time, and then the bubble motion returns to the chaotic behaviour. In this case, the sub harmonics modes are hardly observed at the relatively low amplitude. The sub harmonics modes are not generated by the ultrasound propagation in the body tissues. In the ultrasound imaging, it is possible to intensify the noise signal ratio of the image by extracting the sub harmonics modes. However, it is required to apply relatively higher amplitude of the ultrasound than the case for the higher harmonics modes.

#### 3. A BUBBLE CLUSTER IN AN ULTRASOUND FIELD

In considering medical applications of the ultrasound, an acoustic cavitation induced by the focusing ultrasound should be discussed. It is said that the cavitation makes serious traumas to the human body tissues [16] and its occurrence disturbs the ultrasound propagation by scattering and absorbing the ultrasound energy. To the contrary, the cavitation can be used as an energy transducer if it is controlled well. To utilize their energy efficiently, it is needed to understand their behaviour under the ultrasound field.

In this section, the volumetric change of a micro bubble cluster is simulated numerically. The acoustic emission from the bubble cluster behaves much more intricately than that from a single bubble.

# *3.1. Bubble cluster model*

Figure 3 shows the bubble cluster model [12]. The simulation domain of the spherical bubble cluster is divided into three regions: (1) the outside of the bubble cluster, which is the liquid single phase region, (2) the inside of the bubble cluster, which is the bubbly liquid, and



Figure 3. Concept of the bubble cluster model.

(3) the inside of each bubble. Three sets of governing equations consisting of the equation for the motion of the bubble cluster interface, the equations for the bubbly liquid and the equations for bubble motion are formulated. To calculate the bubble cluster motion, Keller equation [17] is applied. To estimate the pressure at the bubble cluster, the pressure wave phenomena are solved using the set of equations formulated under the following assumptions: (1) Bubble cluster maintains a spherical shape. (2) Bubbly liquid inside the cluster is treated as a continuum fluid, whose mass and momentum are assumed to be equal to those of the liquid phase, because the mass in the unit volume of the gas phase is much smaller than that of the liquid phase. (3) Bubbles move with the surrounding liquid. Bubbles are small enough to ignore the slippage between the bubble and the liquid. (4) Coalescence and fragmentation of bubbles in the cluster are ignored. (5) Viscosity of the bubbly mixture is ignored in the bubble cluster because it has little influence on the wave phenomena. (6) The mass and momentum of the gas phase are ignored to estimate those of the bubbly liquid, because those of the gas phase are much smaller than those of the liquid phase due to the low void fraction. (7) A bubble is assumed to be located at the centre of the cluster to avoid the singularity at the centre of the cluster. Each bubble motion is calculated using the numerical model, which is almost the same as that in the former section. In this section, it is assumed that mass of non-condensable gas inside each bubble is constant and gases inside each bubble obey the van der Waals gas law.

The pressure wave propagates inward and outward inside the bubble cluster. Kameda et al. clarified that the internal thermal phenomena have much influence on the shock wave propagation [18] and in certain cases the compressibility of the liquid cannot be ignored depending on the condition of the bubble motion [19]. It is said that very high pressure of  $O(10^8)$ – $O(10^9)$  Pa emerges in the cloud cavitation when it collapses violently. In such a case the compressibility of the liquid must be taken into account. Therefore, to analyse the collapsing phenomena of the bubble cluster precisely, the internal thermal phenomena of each bubble and the liquid compressibility must be taken into account. The state of liquid is estimated by the Tait equation. Considering such phenomenon, Shimada *et al.* numerically simulated bubble cluster. They concluded that when the bubble cluster collapses, very high pressure emitted from each of the bubbles near the centre of bubble cluster and the frequency of the pressure wave is very high [12].

#### *3.2. Frequency response of the bubble cluster*

The acoustic cavitation induced by an ultrasound is considered to be much dependent on the ultrasound frequency. In a medical applications, the typical frequency of the ultrasound is around 0.5–5 MHz. When the ultrasound frequency is 4 MHz, wavelength is about 0:4 mm in water or a human body. The focal region is considered to be around 2–4 times of the wavelength. In this simulation we assume that the region of the acoustic cavitation is 0.75 mm in radius, and the radius of each bubble that consist the bubble cluster is  $1\mu m$ , whose resonance frequency is about 4 MHz. The simulation conditions are shown in Table I.

Figure 4 shows the frequency response of the bubble cluster from 1 kHz to 10 MHz. Figure  $4(a)$  shows the radius of the bubble cluster, and Figure  $4(b)$  shows the maximum pressure inside the bubble at the centre of the bubble cluster. Resonance frequency response of the bubble cluster for this case is about 110 kHz, which is less than the natural frequency of the cluster, which is 163 kHz. The variation of the bubble cluster radius is very small in

Initial cluster radius, $R_{c0}$	$0.75$ mm
Initial bubble radius, $R_{b0}$	$1 \mu m$
Initial void fraction, $\alpha_0$	$0.1\%$
Ambient pressure, $p_{\infty}$	101.3 kPa
Temperature, $T_0$	293 K
Amplitude of the ambient pressure, $\Delta p$	10, 50, 100 kPa
Variation range of the frequency	$1$ kHz $-10$ MHz
Natural frequency of the bubble cluster	$163 \text{ kHz}$

Table I. Calculation conditions.



Figure 4. Response curves of the bubble cluster: (a) radius of the bubble cluster; and (b) internal pressure of the center bubble.

comparison with a single bubble motion. This is because the changes of the cluster radius are mainly caused by the volumetric changes of the bubbles in the cluster and the initial void fraction is so small. To the contrary, the pressure inside the bubble at the cluster centre reaches around 500 MPa in the case where the surrounding acoustic frequency is the resonance one and the amplitude is 100 kPa. It is far surpassing the case of a single bubble. The pressure wave propagates toward the centre of the bubble cluster. Finally the pressure wave focuses at the centre and the bubbles near the centre collapse violently. One of the authors discusses in detail on the process of the shock wave propagation in the bubble cluster when the surrounding pressure increases stepwisely [20].

Figure 4 also shows that the frequency response of the bubble cluster has much dependence on the amplitude of the surrounding ultrasound pressure. When it is 10 kPa in the amplitude, the maximum pressure inside the bubble at the centre of bubble cluster is very small. In the case of 50kPa, although the maximum pressure reaches up to 2MPa, the range of the resonance frequency where the pressure exceeds a few atmospheric pressures is very narrow, which is less than 100 kHz. However, at the case of 100 kPa, the range of the resonance frequency becomes broad. The frequency range where the maximum pressure exceeds 10 MPa becomes from 50 to 300 kHz. This means that it is able to control the collapsing phenomenon of cavitation bubbles in the ultrasound field. The size of the bubble cluster and those of bubbles in it are determined by the frequency of exciting ultrasound. If the frequency is selected carefully, it is technically possible to concentrate the high energy in the spatial restricted area in the water or the human body.



Figure 5. Time history of the bubble cluster oscillation (frequency: 110 kHz): (a) ambient pressure cluster radius and acoustic pressure; (b) FFT analysis of ambient pressure and acoustic emission from the cluster; and (c) ambient pressure radius and internal pressure of the centre bubble.

#### *3.3. Non-linear oscillation of the bubble cluster*

The bubble cluster which is analysed in the last section has the resonance frequency of  $110 \text{ kHz}$ . The single bubble whose size is  $1 \mu m$  in radius has the natural frequency of  $4 \text{ MHz}$ . The bubble cluster and the bubbles in it have very different features in their oscillations. Figures 5 and 6 show the oscillation of the bubble cluster when the ultrasound is applied on the cluster. In each figure, (a) shows the time history of the ambient pressure, cluster radius and emitted acoustic pressure, (b) shows the FFT analysis of the ambient pressure and the acoustic emission from the cluster, and (c) shows the time history of the ambient pressure, the radius and the internal pressure of the bubble at the centre of the cluster. The calculation conditions are the same as those in Table I. The surrounding ultrasound amplitude is 100 kPa.



Figure 6. Time history of the bubble cluster oscillation (frequency 4 mHz): (a) ambient pressure, cluster radius and acoustic pressure; (b) FFT analysis of ambient pressure and acoustic emission from the cluster; and (c) ambient pressure radius and internal pressure of the centre bubble.

The acoustic pressure from the bubble cluster is estimated from the motion of the cluster surface [1].

At the resonance frequency of the cluster, which is 110 kHz, the centre bubble collapses violently, the bubble radius decreases with extremely high speed and the pressure inside the centre bubble exceeds 500 MPa. The acoustic emission is far beyond the amplitude of the ambient pressure. Higher harmonics are also emitted from the high speed rebound process.

The bubble cluster hardly oscillates under the higher frequency of 4 MHz. The phase of the acoustic emission and the ambient pressure are completely inverted. The bubbles in the cluster also hardly oscillate. The FFT analysis reveals that the acoustic emission from the bubble cluster includes almost only the fundamental component, which is 4 MHz.

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# 4. CONCLUDING REMARKS

Micro bubbles are utilized in the medical applications, for example, ultrasound imaging with contrast agents. A single bubble motion in an ultrasound field is simulated taking the internal thermal phenomena into account. It is revealed that the bubble motion is influenced by the internal phenomena, such as the thermal diffusion, mist formation, mass diffusion, heat and mass transfer through the bubble wall. The acoustic pressure emitted from a single micro bubble is analysed by FFT. The higher harmonics of the ultrasound field are observed in the emitted sound from the micro bubbles, when the natural frequency of the bubble is almost the same of the ultrasound field. When the amplitude of the ultrasound field is increased, the sub-harmonics sound emitted from the bubble are observed and the acoustic turbulence appears.

A set of governing equations for the motion of a spherical bubble cluster is formulated taking into account the internal phenomena of each bubble and the compressibility of the liquid. Pressure wave phenomena in a bubble cluster are simulated numerically in the ultrasound field. An inwardly propagating shock wave is formed during the collapse of the bubble cluster and focused in the cluster centre. This creates the violent bubble collapse, which is more than several hundreds times larger than that of a single bubble collapse. These numerical results reveal that the pressure waves emitted from bubbles in the cluster centre area are amplified when the surrounding pressure oscillates in the resonance frequency of the cluster. The maximum pressure emitted from the bubble at the cluster centre becomes much higher than that of a single bubble and a high frequency pressure oscillation is observed in the centre region. However, the maximum pressure of the cluster is much lower than that of a single bubble when the applied ultrasound frequency is at the natural frequency of a single bubble. The response curve of the emitted pressure from the bubble in the centre of the cluster is broadened as a function of the ultrasound amplitude.

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