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# Dynamic behaviour of a two-microbubble system under ultrasonic wave excitation 

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#### Abstract

Acoustic bubbles have wide and important applications in ultrasonic cleaning, sonochemistry and medical ultrasonics. A two-microbubble system (TMS) under ultrasonic wave excitation is explored in the present study, by using the boundary element method (BEM) based on the potential flow theory. A parametric study of the behaviour of a TMS has been carried out in terms of the amplitude and direction of ultrasound as well as the sizes and separation distance of the two bubbles. Three regimes of the dynamic behaviour of the TMS have been identified in terms of the pressure amplitude of the ultrasonic wave. When subject to a strong wave with the pressure amplitude of 1 atm or larger, the two microbubbles become non-spherical during the first cycle of oscillation, with two counter liquid jets formed. When subject to a weak wave with the pressure amplitude of less than 0.5 atm ,


[^0]two microbubbles may be attracted, repelled, or translate along the wave direction with periodic stable separation distance, depending on their size ratio. However, for the TMS under moderate waves, bubbles undergo both non-spherical oscillation and translation as well as liquid jet rebounding.

Keywords:
microbubble dynamics, ultrasonic wave, boundary element method

## 1. Introduction

Cavitation bubbles are associated with the erosion and noise of hydraulic machines, propellers and turbines [1, 2]. Acoustic cavitation bubbles are applied in cavitation cleaning [3, 4], extracorporeal shock wave lithotripsy (ESWL) [5], drug delivery and sonoporation [6].

A two-microbubble system (TMS) is a basic unit of bubble clusters. Theoretical studies were carried out for two spherical bubbles subject to acoustic waves. Crum derived the formula for calculating the 'secondary Bjerknes force' for the interaction of two oscillating spherical bubbles subject to an acoustic wave [7]. This model was later developed by considering the viscous and thermal effects $[8,9]$, bubbles at different sizes [10] and under dualfrequency ultrasound [11], as well as in viscoelastic fluid [12]. Microstreaming generated by two acoustically induced spherical bubbles was studied by Doinikov and Bouakaz [13]. The interaction of two approximate spherical encapsulated bubbles was studied by using perturbation theory based on the potential flow theory by Liu and Wang [14].

The interaction of non-spherical transient bubbles was modelled by numerical methods. The domain methods coupled with the interface capturing
techniques present good results for computing the collapse, coalescence, and splitting of three-dimensional (3-D) bubbles $[15,16,17]$. The interaction of two bubbles was modelled by Han et al. by using the boundary element method (BEM) [18, 19]. Multiple bubbles dynamics was simulated by using the BEM coupled with fast multiple expansions [20, 21, 22]. Multibubble surface cavitation can be achieved by using a strong negative impulsive liquid pressure wave to expand the nuclei in the microcavities. Interaction of two or more pulsating microbubbles formed in this way was investigated both experimentally and numerically by Bremond et al. [23, 24]. Jet formation during the evolution of two microbubbles near a solid wall has been observed [23].

In this paper we consider the interaction of two initially spherical microbubbles subject to ultrasound with different pressure amplitude, which may become non-spherical due to close interactions and/or strong ultrasonic wave (Wang and Manmi [25, 26]; Ochiai and Ishimoto [4]). The dynamics of a single non-spherical bubble subject to ultrasound was simulated by using the BEM (Klaseboer et al. [27]; Calvisi et al. [28, 29]; Wang and Blake [30, 31]; Wang and Manmi [25, 26]). The physical and numerical models are introduced in Section 2. In Section 3, the numerical model is firstly validated against an axisymmetric model, and a parametric study of the behaviour of a TMS is then carried out in terms of the amplitude and direction of ultrasound, the sizes, and separation distance of the two bubbles. Finally, concluding remarks are given in Section 4.

## 2. Boundary element method

Consider a two-microbubble system (TMS) subject to an ultrasonic wave, as illustrated in Fig. 1. The two bubbles are labeled as bubble 1 and bubble 2 with the initial radii $R_{01}$ and $R_{02}$ respectively. The initial separation distance between their centres $C_{1}$ and $C_{2}$ is denoted as $D$. A Cartesian coordinate system is set with its origin $O$ at the midpoint of the initial centreline $C_{1} C_{2}$ and the $x$-axis is in the direction of wave propagation. The centreline $C_{1} C_{2}$ lies in the $O x z$ plane, with an angle $\beta$ with the $x$-axis.
(Figure 1)
The far-field pressure $p_{\infty}$ is given by

$$
\begin{equation*}
p_{\infty}(x, t)=p_{0}+p_{\mathrm{a}} \sin (k x-2 \pi f t) \tag{1}
\end{equation*}
$$

where $p_{0}$ is the hydrostatic pressure, $p_{\mathrm{a}}$ the pressure amplitude of the wave, $k$ the wave number, and $f$ the wave frequency.

We assume that the flow is incompressible and potential, the velocity potential $\varphi$ thus satisfies Laplace's equation

$$
\begin{equation*}
\nabla^{2} \varphi=0 \tag{2}
\end{equation*}
$$

Assuming that each bubble undergoes adiabatic process during the motion, the internal gas pressure $p_{\mathrm{b} i}$ of bubble $i$ is given by

$$
\begin{equation*}
p_{\mathrm{b} i}=p_{\mathrm{v}}+p_{\mathrm{g} i 0}\left(\frac{V_{0 i}}{V_{i}}\right)^{\alpha} \tag{3}
\end{equation*}
$$

where $p_{\mathrm{v}}$ is the saturated vapor pressure, $p_{\mathrm{g} i 0}$ the initial partial pressure of a non-condensible gas in bubble $i, V_{0 i}$ and $V_{i}$ are its initial and transient volumes, respectively, $\alpha$ is the polytropic index of the bubble gas, which is set as 1.4 in this work.

The liquid pressure $p$ on bubble surface $S_{\mathrm{b} i}$ satisfies the Young-Laplace equation

$$
\begin{equation*}
p=p_{\mathrm{b} i}-2 \sigma \kappa \quad \text { on } S_{\mathrm{b} i}, \tag{4}
\end{equation*}
$$

where $\sigma$ is surface tension and $\kappa$ the local curvature of the bubble surface.
The Bernoulli equation for incompressible potential flows reads

$$
\begin{equation*}
\rho \frac{\partial \varphi}{\partial t}+\frac{\rho}{2}|\nabla \varphi|^{2}+p-p_{\infty}=0 \tag{5}
\end{equation*}
$$

where $\rho$ is the liquid density. Substituting eqs. (1), (3) and (4) into (5) yields the dynamic boundary condition on bubble surfaces

$$
\begin{equation*}
\rho \frac{\partial \varphi}{\partial t}+\frac{\rho}{2}|\nabla \varphi|^{2}+p_{\mathrm{v}}+p_{\mathrm{g} i 0}\left(\frac{V_{0 i}}{V_{i}}\right)^{\alpha}-2 \sigma \kappa-p_{0}-p_{\mathrm{a}} \sin (k x-2 \pi f t)=0 . \tag{6}
\end{equation*}
$$

Using matched asymptotic expansions, Wang and Blake [30, 31] showed that the flow far away from the bubble can be described by the linear wave equation to second order in terms of the Mach number while the flow near the bubble can be described by Laplace's equation to second order. This weakly compressible theory shows that the incident wave effects can be approximated to second order as the last term of the boundary condition (6).

To perform non-dimensionalization, the length scale is chosen as the initial radius $R_{01}$ of bubble 1 , the pressure scale $\Delta p=p_{0}-p_{\mathrm{v}}$, and the reference velocity $U=\sqrt{\Delta p / \rho}$. The corresponding reference time is $T=R_{01} / U$. The dimensionless parameters and variables are labeled by the superscript ${ }^{*}$, , as follows

$$
\begin{gather*}
D^{*}=\frac{D}{R_{01}}, t^{*}=\frac{t}{T}, f^{*}=f T,  \tag{7a}\\
\varphi^{*}=\frac{\varphi}{U R_{01}}, \sigma^{*}=\frac{\sigma}{R_{01} \Delta p}, k^{*}=k R_{01} . \tag{7b}
\end{gather*}
$$

The dimensionless dynamic boundary condition (6) becomes

$$
\begin{equation*}
\frac{\mathrm{d} \varphi^{*}}{\mathrm{~d} t^{*}}=1-p_{\mathrm{g} i 0}^{*}\left(\frac{V_{0 i}^{*}}{V_{i}^{*}}\right)^{\alpha}+\frac{1}{2}\left|\nabla \varphi^{*}\right|^{2}+2 \sigma^{*} \kappa^{*}+p_{\mathrm{a}}^{*} \sin \left(k^{*} x^{*}-2 \pi f^{*} t^{*}\right) \tag{8}
\end{equation*}
$$

Note that the partial derivative in eq. (6) is converted to the material derivative using the relation $\mathrm{d} \varphi^{*} / \mathrm{d} t^{*} \equiv \partial \varphi^{*} / \partial t^{*}+\boldsymbol{v}^{*} \cdot \nabla \varphi^{*}$, where $\boldsymbol{v}^{*}$ is the velocity. The kinematic boundary condition is essential for tracking the position $\boldsymbol{r}^{*}$ of each particle on bubble surfaces:

$$
\begin{equation*}
\frac{\mathrm{d} \boldsymbol{r}^{*}}{\mathrm{~d} t^{*}}=\boldsymbol{v}^{*} \tag{9}
\end{equation*}
$$

The velocity potential $\varphi^{*}$ satisfies the following boundary integral equation (BIE)

$$
\begin{equation*}
c\left(\boldsymbol{r}^{*}\right) \varphi^{*}\left(\boldsymbol{r}^{*}\right)=\iint_{S}\left[\frac{\partial \varphi^{*}\left(\boldsymbol{q}^{*}\right)}{\partial n} G\left(\boldsymbol{r}^{*}, \boldsymbol{q}^{*}\right)-\varphi^{*}\left(\boldsymbol{q}^{*}\right) \frac{\partial G\left(\boldsymbol{r}^{*}, \boldsymbol{q}^{*}\right)}{\partial n}\right] \mathrm{d} S \tag{10}
\end{equation*}
$$

where $S$ denotes all surfaces of bubbles; $c\left(\boldsymbol{r}^{*}\right)$ is solid angle, $\boldsymbol{r}^{*}$ and $\boldsymbol{q}^{*}$ are the field and source points, respectively and $\boldsymbol{n}$ is the unit outward normal vector of bubble surfaces. The Green function is $G\left(\boldsymbol{r}^{*}, \boldsymbol{q}^{*}\right)=\left|\boldsymbol{q}^{*}-\boldsymbol{r}^{*}\right|^{-1}$.

The surfaces of the initially spherical bubbles are meshed into triangles based on regular icosahedrons [32]. The bubbles are at rest at the beginning of simulation. Hence, both the velocity potential $\varphi^{*}$ and the velocity $\boldsymbol{v}^{*}$ of each node on bubble surfaces equal zero at $t^{*}=0$. The normal velocity $\partial \varphi^{*} / \partial n$ of each node is calculated using eq. (10) in every time step. The tangential velocity is calculated using the interpolation of the bubble surface and the velocity potential on the surface [33].
$\varphi^{*}$ and $\boldsymbol{r}^{*}$ are then updated by integrating the dynamic and kinematic boundary conditions (8) and (9), respectively. An explicit fourth-order RungeKutta (RK4) scheme is used in the numerical integration. A weighted least-
squares smoother [19] is used every 30 time steps to keep the bubble surfaces smooth.

A time step $\Delta t^{*}=\min \left(\Delta t_{\boldsymbol{v}}^{*}, \Delta t_{\varphi}^{*}\right)$ is used, with

$$
\begin{equation*}
\Delta t_{\boldsymbol{v}}^{*}=\frac{\min \left(l_{\text {edge }}^{*}\right)}{\max \left(\left|\boldsymbol{v}^{*}\right|\right)}, \quad \Delta t_{\varphi}^{*}=\frac{\varphi_{\mathrm{cr}}^{*}}{\max \left(\mathrm{~d} \varphi^{*} / \mathrm{d} t^{*}\right)^{\prime}}, \tag{11}
\end{equation*}
$$

where $l_{\text {edge }}^{*}$ denotes the length of bubble element edge, $\varphi_{\mathrm{cr}}^{*}$ is a constant used to restrict the increment of the velocity potential, and the maximum and minimum in (11) are taken for all nodes on bubble surfaces. In this paper, we set $\varphi_{\mathrm{cr}}^{*}=0.01$, unless specified otherwise.

The field pressure $p_{\mathrm{f}}^{*}$ is given by the Bernoulli equation

$$
\begin{equation*}
p_{\mathrm{f}}^{*}=1+p_{\mathrm{v}}^{*}+p_{\mathrm{a}}^{*} \sin \left(k^{*} x^{*}-2 \pi f^{*} t^{*}\right)-\frac{\partial \varphi^{*}}{\partial t^{*}}-\frac{1}{2}\left|\nabla \varphi^{*}\right|^{2} . \tag{12}
\end{equation*}
$$

It is a well-known challenge to calculate the pressure field associated with bubble dynamics. Calculating the term $\partial \varphi^{*} / \partial t^{*}$ in (12) using a finite difference method often results in unacceptable errors, due to very small time steps usually used for simulating a violent collapsing bubble. We calculate $\partial \varphi^{*} / \partial t^{*}$ using the BEM model since $\partial \varphi^{*} / \partial t^{*}$ satisfies Laplace's equation, too, following Tanizawa [34] and Wu [35]. We also calculate $\nabla \varphi^{*}$ using the BEM since $\partial \varphi^{*} / \partial x^{*}, \partial \varphi^{*} / \partial y^{*}$ and $\partial \varphi^{*} / \partial z^{*}$ satisfy Laplace's equation as well.

## 3. Numerical Analysis

In the following calculations, the surface tension is taken as $\sigma=0.073 \mathrm{Nm}^{-1}$, the liquid density $\rho=998 \mathrm{~kg} \mathrm{~m}^{-3}$, the hydrostatic pressure $p_{0}=101.3 \mathrm{kPa}$ and the saturated vapor pressure $p_{\mathrm{v}}=2339 \mathrm{~Pa}$.

### 3.1. Comparison with axisymmetric BEM

The case is for a single bubble with an initial radius $R_{0}=4.5 \mu \mathrm{~m}$, subject to a travelling ultrasound with a pressure amplitude $p_{\mathrm{a}}^{*}=1.6$ and a frequency $f^{*}=0.136$ (in dimensional, 0.3 MHz ). The polytropic index of the bubble gas is $\alpha=1.667$. Fig. 2 compares the bubble shapes calculated from the present 3-D model and the axisymmetric model [28]. Agreement between the two models improves as the mesh density increases.
(Figure 2)

### 3.2. General features

Bubbles may become non-spherical when subject to acoustic waves. We use the sphericity $\psi$ to describe the deviation extent of a bubble from a spherical shape [36]:

$$
\begin{equation*}
\psi=\frac{A_{\mathrm{b}}}{\pi^{1 / 3}\left(6 V_{\mathrm{b}}\right)^{2 / 3}}, \tag{13}
\end{equation*}
$$

where $A_{\mathrm{b}}$ and $V_{\mathrm{b}}$ denote the surface area and the volume of a bubble, respectively. Notice $\psi=1$ for a sphere and $\psi>1$ for all the non-spherical geometries.

Fig. 3 shows the overall dynamic behaviour of the TMS under ultrasonic wave forcing in terms of the maximum sphericity of bubble 1 and the number of cycles that the TMS has oscillated (the average volume oscillation cycles of the two bubbles) before the termination of simulation. The simulation is either terminated at $t^{*}=30$ or at the moment when any liquid jet impacts on the opposite side of the bubble surface. It can be seen from Fig. 3 that the bubble dynamic behaviour can be divided into three categories: high pressure amplitude ( $p_{\mathrm{a}}^{*}>1.0$ ), moderate pressure amplitude $\left(0.5 \leqslant p_{\mathrm{a}}^{*} \leqslant 1.0\right)$
and low pressure amplitude $\left(p_{\mathrm{a}}^{*}<0.5\right)$. When under strong acoustic wave ( $p_{\mathrm{a}}^{*}>1.0$ ), the bubbles become significantly non-spherical associated with a larger maximum sphericity $\psi_{\max }$ and the liquid jets form in the first cycle of oscillation and impact on the opposite bubble surface. These inertial collapse behaviour is associated with applications in surface cleaning [37] and lithotripsy [38]. For a weak acoustic wave ( $p_{\mathrm{a}}^{*}<0.5$ ), the bubbles stay nearly spherical for a relative long time and oscillate for many cycles without jet formation. The maximum sphericity $\psi_{\text {max }}$ stays at the value 1 while the oscillation cycles exceed 6 as can be seen in the figure. The number of bubble oscillation cycles is 9 for $p_{a}^{*}=0.1-0.3$ up to dimensionless time $t^{*}=30$ (equivalent roughly to 4.53 acoustic cycles), as the dimensionless natural frequency of the bubble, being 0.32 , is about two times of the driving wave frequency 0.151 . On this occasion, their translational motion and oscillation are worth studying. This kind of stable repeated oscillation of bubbles are associated with sonography [38] and sonochemistry [39]. The cases shown in Fig. 3 are for the dimensionless initial spacing $D^{*}=6$ between two bubbles. Our calculations for $D^{*} \in[5,20]$ with remaining parameters unchanged have shown that the TMS presents similar behaviour shown in Fig. 3 for both strong and weak wave amplitude regions.
(Figure 3)
(Figure 4)
As a TMS is subject to moderate acoustic wave forcing, the behaviour of a TMS in both strong and weak wave regions may be observed. Fig. 4 shows a case for $p_{\mathrm{a}}^{*}=0.8$. The acoustic wave induces a liquid jet with bubble 1 at the end of the first cycle (Fig. 4(b)). However, the wave is not strong
enough to cause a jet penetration through the opposite side of the bubble, like in Fig. 2 for a strong wave. Instead, the bubble start rebounding during jet development and the jet disappears before it reaches maximum volume (Fig. 4(c)). The liquid jet re-appears at the end of bubble re-collapse (Fig. $4(\mathrm{~d})$ ). Likewise, this time the jet is still not fast enough to penetrate the bubble; it bounces back again when the bubble starts to expand in the third cycle. When the bubble 1 reaches its maximum volume in this cycle (Fig. $4(\mathrm{e})$ ), two bubbles are close to each other and are about to coalesce. In this process, bubble 2 behaves similarly to bubble 1 with a slight delay. In this category, bubbles undergo both non-spherical oscillation and translation.

### 3.3. Subject to strong ultrasound

(Figure 5)
(Figure 6)

## (Figure 7)

(Figure 8)
Consider two bubbles having initial radii $5 \mu \mathrm{~m}$ and the initial spacing $D^{*}=5$ subject to an acoustic wave with $p_{\mathrm{a}}^{*}=1.2$ and $f^{*}=0.151$ (in dimensional, 0.3 MHz ). Single acoustic bubble dynamics in this parameter family was widely studied [28, 40, 25, 26]. The interaction of two microbubbles near the solid wall induced by a stronger negative pressure impulse was investigated by an axisymmetric BEM model [24]. In the present case, bubble 1 reaches its maximum volume at $t^{*}=3.43$ (see Fig. 5(b)), and both bubbles keep spherical shapes until this moment. The bubbles become non-spherical at the end of collapse due to the acoustic wave as well as the interaction between bubbles. Dominated by the secondary Bjerknes force, the two bubbles
are attracted to each other with the formation of two counter jets. While the two microbubbles investigated by Bremond et al. [23] are symmetrical to each other, the two microbubbles investigated here clearly undergo different dynamical processes. Bubble 1, located in front to the traveling wave, presented much larger volume oscillation, loss of sphericity, and jet formation than bubble 2 .

Fig. 6 shows the time histories of the two jet velocities. The jets of bubble 1 and bubble 2 reach their maximum velocities at $t^{*}=4.84$ and $t^{*}=4.90$ respectively, during the early stage of development of the two jets, as shown in Figs. 5(c) and 5(d), respectively. After that, the jet velocities firstly drop slightly and then do not vary significantly before jet impact.

Fig. 7 shows the convergence tests for the case in Fig. 6, for the jet velocity histories in the late stage of the collapse phase of bubble 1 in terms of mesh sizes and time steps. It can be seen that a smaller mesh size and/or a shorter time step lead to the liquid jet forming slightly later with a higher speed. The results converge approximately for 980 elements and $\varphi_{\mathrm{cr}}^{*}=0.01$. These two values for the mesh size and time step are used in the following cases to ensure the accuracy and efficiency.

Fig. 8 shows the pressure field around bubble 1 at the $O x z$-plane during jet development under strong wave forcing. At this time, each bubble is more affected by the other bubble than by the ultrasonic wave, due to the strong interaction between the bubbles. A high pressure zone is formed at the base of the jet and its amplitude increases with time as the jet develops, whilst the pressure on the other side of the bubble is relatively smaller. The pressure difference across the bubble drives the motion of the jet. During the
formation of the jet, the bubble surface on the other side of the jet does not deviate significantly from the spherical form.

Fig. 9 displays the time histories of the volumes of two bubbles. One can see from Figs. 8(d) and 9 that bubble 1 reaches its minimum volume before the jet penetrates through the other side. We believe that this 'early maximum jet velocity' and 'early bubble rebound' is due to the 'not-so-strong' wave pressure amplitude $p_{\mathrm{a}}^{*}$. According to our calculations, the maximum jet velocity occurs at the early stage of jet formation, for the wave pressure amplitude $p_{\mathrm{a}}^{*} \in[1.0,1.5]$, and the initial spacing $D^{*} \in[5,20]$. The jet velocity will keep increasing until jet penetration for $p_{\mathrm{a}}^{*}>1.6$.
(Figure 9)
Note the jet of bubble 2 forms later than that of bubble 1 with a higher maximum velocity (Fig. 6), and has a larger maximum volume and a longer period (Fig. 9). Fig. 9 also displays the time history of the pressure at the initial centre of the TMS, whose amplitude is one order larger than that of the wave pressure.

In the above cases, the two bubbles are placed along the wave direction, i.e. $\beta=0$. Fig. 10 shows the maximum jet velocities and the corresponding time when maximum jet velocities occur in function of $\beta$. As $\beta$ increases from 0 to $\pi / 2$, the difference of the maximum dimensionless jet velocities between bubble 1 and 2 decreases from 4 (in dimensional value, $40 \mathrm{~m} \mathrm{~s}^{-1}$ ) to 0.
(Figure 10)

### 3.4. Subject to weak ultrasound

When subject to weak ultrasound, i.e. $p_{\mathrm{a}}^{*}<0.5$, a TMS undergoes steady oscillation for many cycles. We terminate the simulation at $t^{*}=30$ here, which is sufficient for studying the dynamics of the TMS.

### 3.4.1. Equal sized TMS

The case considered is for $p_{\mathrm{a}}^{*}=0.3$ and $\beta=0$, with the remaining parameters being the same as in Fig. 4. Both bubbles are nearly spherical during the whole simulation process, as their sphericity $\psi$ is less than 1.001 . As shown in Fig. 11, each violent oscillation cycle is followed by a mild one and the bubbles oscillate approximately twice per acoustic period. The natural frequency $f_{\mathrm{n}}^{*}$ of a bubble is given by Wang and Blake [30]

$$
\begin{equation*}
f_{\mathrm{n}}^{*}=\frac{1}{2 \pi R_{0}^{*}} \sqrt{3 \alpha}, \tag{14}
\end{equation*}
$$

where $R_{0}^{*}$ is the equilibrium radius of a bubble. The natural frequency of the bubbles considered being 0.32 is about two times of the driving wave frequency 0.151.
(Figure 11)
The time history of the centroid displacement $C_{x}^{*}$ of the two bubbles is also shown in the figure. The magnitude of $C_{x}^{*}$ denotes the displacement of a bubble from its initial position and the sign depicts the translational direction. The bubbles migrate towards the initial centre of configuration with their speeds increasing with time. The secondary Bjerknes force between the two bubbles is predominant in this case.

As $\beta \neq 0$, the bubbles translate along both the $x$ - and $z$-axes. Fig. 12
displays the time histories of the average displacement of the bubble centroids

$$
\begin{equation*}
C_{\mathrm{as}}^{*}=\frac{\int_{0}^{t_{1}} C_{s}^{*} \mathrm{~d} t}{t_{1}} \tag{15}
\end{equation*}
$$

where the subscript ' $s$ ' denotes ' $x$ ' or ' $z$ ' and $t_{1}$ is the total time used in simulation. The two bubbles translate in opposite directions with approximately the same speed. The bubble translation in this case is again dominated by the secondary Bjerknes force.
(Figure 12)

### 3.4.2. Unequal sized TMS

In this section, translation of two bubbles with unequal initial sizes is investigated. The initial radius of bubble 1 is $5 \mu \mathrm{~m}$ while that of bubble 2 varies with a larger size. The initial spacing is set as $D^{*}=20$, to examine a relative larger range of $R_{02}^{*}$. The remaining parameters are the same as in Fig. 11.
(Figure 13)
Fig. 13 presents the results of the case $R_{02} / R_{01}=2$. Since the larger bubble dominates the behaviour of the TMS, only the volume history of bubble 2 is depicted here. The natural frequency $f_{\mathrm{n}}^{*}$ of bubble 2 calculated using (14) being 0.163 is close to the wave frequency 0.151 . Bubble 2 undergos resonance with its oscillation amplitude increasing monotonically. Fig. 13 also displays the time history of the displacement of the bubble centroids. Both bubbles migrate along the wave direction at similar amplitude, with much smaller relative motion. The separation distance between the bubbles is periodic stable, which is consistent with the conclusion of Barbat et al.
[8]. In this case, the primary Bjerknes force [41] due to the acoustic wave is predominant.
(Figure 14)
Fig. 14 shows the time history of the displacement of the bubble centroids for $R_{02}^{*}=8$. The initial volume ratio of two bubbles here reaches 512 . In this case, the dimensionless time scale only covers one cycle of oscillation of bubble 2. During the time period, the center of bubble 2 does not migrate significantly while bubble 1 is pushed away by bubble 2 and undergoes oscillating translation following the oscillating bubble 2 .
(Figure 15)
Fig. 15 shows the time histories of the bubble centroids for bubble 1 and bubble 2 for 4 different size ratios $R_{02}^{*}=1,2,4,8$. The two bubbles are attracted to each other for $R_{02}^{*}=1$, translate along the wave direction for $R_{02}^{*}=2$, and the small bubble leaves away from the large one which does not translate significantly for $R_{02}^{*}=4,8$. These characteristics were also predicted by the linear analysis of two spherical bubbles subject to an acoustic wave $[7,8]$.

## 4. Summary and conclusions

The dynamics of a two-microbubble system (TMS) subject to an ultrasonic wave is investigated by using the boundary element method (BEM) based on the potential flow theory. The numerical model is verified by comparing with an axisymmetric BEM as well as convergence test. The pressure distribution in the flow field is calculated by using the Bernoulli equation, where the partial derivative of the velocity potential in time is calculated
using the BEM to avoid numerical instabilities.
Numerical analysis is performed for a two bubble system for bubbles with equilibrium radii about $5 \mu \mathrm{~m}$ and initial spacing from 25 to $100 \mu \mathrm{~m}$ subject to the wave with a frequency of $f=0.3 \mathrm{MHz}$ and pressure amplitude from $0.5-2 \mathrm{~atm}$. Three regimes of the dynamic behaviour of the TMS have been identified in terms of the pressure amplitude of ultrasound as following.
a) When the wave pressure amplitude $p_{\mathrm{a}} \gtrsim 1 \mathrm{~atm}$, both bubbles become non-spherical during first few cycles of oscillation, forming two counter jets, generated by high pressure zones behind the jets. The bubble hit by the wave front earlier has a shorter oscillation cycle and a relative lower speed liquid jet. The field pressure can be one order of magnitude larger than the wave pressure amplitude $p_{\mathrm{a}}$. The bubble behaviour is dominant by the secondary Bjerknes between the two bubbles.
b) When the wave pressure amplitude $p_{\mathrm{a}} \lesssim 0.5 \mathrm{~atm}$, bubble translation is the main characteristic behaviour. Two equal sized bubbles are attracted to each other due to the secondary Bjerknes force. For two unequal sized bubbles, the small bubble leaves away from the large one and the large one does not translate significantly. When the resonance happens, the primary Bjerknes force may lead to the obvious translation of a TMS along the wave direction. However, the separation distance between the bubbles is periodic stable.
c) However, for a TMS under moderate wave amplitude, bubbles undergo both non-spherical oscillation and translation.

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## Figure Captions

Figure 1 Sketch of a TMS subject to a travelling ultrasonic wave and the coordinate system.

Figure 2 Comparison of bubble shapes at the end of the collapse phase ( $t^{*}=5.637$ ) with an axisymmetric BEM (dashed line, Calvisi et al. [28]). (a) 720 elements, (b) 980 elements, and (c) 1280 elements. $p_{\mathrm{a}}^{*}=1.6, R_{0}=4.5 \mu \mathrm{~m}$, $f^{*}=0.136$ (in dimensional, 0.3 MHz ), $\alpha=1.667$ and $\rho=998 \mathrm{~kg} \mathrm{~m}^{-3}$.

Figure 3 Oscillation cycles of a TMS and the maximum sphericity $\psi_{\text {max }}$ of bubble 1 at different ultrasonic wave pressure amplitude $p_{\mathrm{a}}^{*} . \beta=0, R_{01}^{*}=$ $R_{02}^{*}=1($ in dimensional, $5 \mu \mathrm{~m}), f^{*}=0.151(0.3 \mathrm{MHz})$ and $D^{*}=6$.

Figure 4 TMS under moderate wave forcing when bubble 1 reaches its first, second and third maximum volumes (frames (a), (c) and (e)) and its first and second minimum volumes (frames (b) and (d)). $p_{\mathrm{a}}^{*}=0.8, \beta=0$, $R_{01}^{*}=R_{02}^{*}=1($ in dimensional, $5 \mu \mathrm{~m}), f^{*}=0.151(0.3 \mathrm{MHz})$ and $D^{*}=6$.

Figure 5 Bubble shapes of a TMS under strong wave forcing. $p_{\mathrm{a}}^{*}=1.2$, $\beta=0, R_{01}^{*}=R_{02}^{*}=1$ (in dimensional, $\left.5 \mu \mathrm{~m}\right), f^{*}=0.151(0.3 \mathrm{MHz})$ and $D^{*}=5$.

Figure 6 Jet velocities of a TMS in the compression phase as functions of time for the case in Fig. 5.

Figure 7 Jet velocity histories during the late stage of the collapse phase of bubble 1 for the case in Fig. 6 with different mesh sizes and time steps.

Figure 8 Pressure field $p_{\mathrm{f}}^{*}$ near bubble 1 as the bubble jet develops for the case in Fig. 5.

Figure 9 Time histories of the bubble volumes and pressure at the initial centre $O$ of the configuration for the case in Fig. 5.

Figure 10 The maximum jet velocity $v_{\mathrm{jet}}^{*}$ of each bubble and their corresponding occurrence time as functions of $\beta$. The remaining parameters are the same as in Fig. 5.

Figure 11 Time histories of the geometric centres of the bubbles in the $x$ direction and the volume of bubble 1 for $p_{\mathrm{a}}^{*}=0.3, \beta=0, R_{01}^{*}=R_{02}^{*}=1$, $f^{*}=0.151$ and $D^{*}=6$.

Figure 12 Time histories of the displacement of the bubble controids $C_{\mathrm{a} x}^{*}$ and $C_{\mathrm{a} z}^{*}$ in terms of $\beta$. The remaining parameters are the same as in Fig. 11.

Figure 13 Time histoires of the centroids $C_{x}^{*}$ of the two bubbles and the volume $V^{*}$ of bubble 2 for $R_{02}^{*}=2$ and $D^{*}=20$. The remaining parameters are the same in Fig. 11.

Figure 14 Time histories of the centroids $C_{x}^{*}$ of the two bubbles and the volume $V^{*}$ of bubble 2 for $R_{02}^{*}=8$ and $D^{*}=20$. The remaining parameters are the same in Fig. 11.

Figure 15 Time histories of the centroids $C_{x}^{*}$ for (a) bubble 1 and (b) bubble 2 for $R_{02}^{*}=1,2,4,8$. The remaining parameters are the same as in Fig. 14.

bubble 1

Figure 1: Sketch of a TMS subject to a travelling ultrasonic wave and the coordinate system.


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(a) $t^{*}=3.04$

(b) $t^{*}=4.49$

(d) $t^{*}=7.01$

(e) $t^{*}=9.75$

Figure 4: TMS under moderate wave forcing when bubble 1 reaches its first, second and third maximum volumes (frames (a), (c) and (e)) and its first and second minimum volumes (frames (b) and (d)). $p_{\mathrm{a}}^{*}=0.8, \beta=0, R_{01}^{*}=R_{02}^{*}=1$ (in dimensional, $5 \mu \mathrm{~m}$ ), $f^{*}=0.151(0.3 \mathrm{MHz})$ and $D^{*}=6$.

(a) $t^{*}=0$

(b) $t^{*}=3.43$

(c) $t^{*}=4.84$

(d) $t_{2}^{*}=4.90$

Figure 5: Bubble shapes of a TMS under strong wave forcing. $p_{\mathrm{a}}^{*}=1.2, \beta=0, R_{01}^{*}=$ $R_{02}^{*}=1($ in dimensional, $5 \mu \mathrm{~m}), f^{*}=0.151(0.3 \mathrm{MHz})$ and $D^{*}=5$.


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