Bubble cluster dynamics in an acoustic field

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SUMMARY
Mathematical model describing the dynamics of a bubble cluster composed of gas bubbles of various radii oscillating in an unbounded, slightly compressible viscous liquid under the action of an external acoustic field is presented. The proposed model is used for an analytical study of small (linear) bubble oscillations in monodisperse and polydisperse clusters, for a numerical investigation of large (nonlinear) bubble oscillations, and for a diffusion stability analysis of gas bubbles in the cluster. The following phenomena have been revealed:

1. synchronization of the collapse phases of bubbles with different radii;
2. collapse intensification for bubbles of one size in the presence of bubbles of other size;
3. bubbles in a monodisperse cluster are more diffusively stable than a single bubble in an infinite liquid;
4. polydisperse (two-fraction) cluster tends to become a one-fraction cluster due to the diffusion.

INTRODUCTION
Cavitation is the formation of bubble clusters or bubble clouds in a liquid subject to tension. Direct experiments for studying the dynamics of bubbles in clusters or clouds represent a significant technical complexity. Therefore mathematical modeling of the dynamics of bubbles in the cluster is of great importance. One can distinguish two complementary approaches to study bubble clouds:

1. analysis of dynamics of the bubble cluster in which a mixture of liquid and gas bubbles is considered as a continuum;
2. analysis of motion of single bubbles in the cluster.

In the first approach the cluster is considered as bubbly liquid. One of the first who considered the dynamics of bubble clusters but was van Wijngaarden [1] who found that the collapse of a large number of bubbles close to a smooth wall leads to increase in pressure near the wall as the result of bubble interactions. After that the bubble cloud dynamics was studied theoretically by many authors (see for example, [2-6]).
The main assumptions are following:
(1) the size of the cluster $R$ is small in comparison with the wavelength of the acoustic field in bubbly liquid $\lambda$;
(2) the scattering cross sections of neighboring bubbles don’t overlap [7] ($\sigma^{1/2} << l$, where $\sigma$ is the value of the bubble scattering cross section; $l$ is the average distance between the bubbles);
(3) the gas pressure inside the bubbles is spatially uniform for each bubble;
(4) bubbles are spherically–symmetric;
(5) there is no liquid evaporation/condensation phase transition in the cluster, the uniform gas pressure inside the bubbles obey the adiabatic law and mass transfer is absent.

Note that under assumptions (1) and (2), the liquid pressure inside the cluster $p_c$ can be considered as uniform and time-dependent, i.e. $p_c = p_c(t)$.

**MATHEMATICAL MODEL OF BUBBLE CLUSTER**

When a polydisperse cluster contains bubbles of $n$ different sizes, the dispersed phase can be divided into $n$ fractions, each characterized by its initial bubble radius ($a_{ik}$ for $k$-th fraction). The radial motions of a conventional cluster boundary and bubble radii in general case obey the following set of equations ([8], [9]):

\[
\frac{a_k}{2} \ddot{a}_k + \frac{3}{2} \dot{a}_k^2 = \frac{p_{ak} - p_a}{\rho_i}, \quad k = \overline{1,n}, \tag{1}
\]

\[
\frac{RR}{2} + \frac{3}{2} R^2 = \frac{p_a - p_t}{\rho_i} + \frac{R}{\rho_i C_i} \frac{d}{dt}[p_c - p_l], \tag{2}
\]

\[
\sum_{k=1}^{n} N_k a_k^2 = R^2 \dot{R}, \tag{3}
\]

\[
p_{ak} = \left( p_0 + \frac{2\sigma}{a_{ok}} \right) a_k^{-3/2} - \frac{4\mu \dot{a}_k}{a_k} - \frac{2\sigma}{a_k}, \quad k = \overline{1,n}, \tag{4}
\]

\[
p_l = p_0 - \Delta \rho \sin(\omega t). \tag{5}
\]

Here $p_{ak}$ is the pressure of gas at bubble wall of $k$-th fraction; $p_l$ is the driving incident pressure; $p_t$ is the liquid density; $C_i$ is the sound speed in the liquid; $p_0$ is the static ambient pressure; $\Delta \rho$ is the pressure amplitude; $\omega$ is the driving frequency; $\sigma$ is the surface tension; $\mu$ is the liquid viscosity; $N_k$ is the number of bubbles in the cluster of $k$-th fraction and $t$ is the time.

The ranges of parameters of applicability of this model have been determined. On Figure 2 two different initial bubble sizes $a_0 = 5\ mkm$ and $a_0 = 25\ mkm$ are presented. Areas of mathematical model validity are shaded by parallel lines. The area shaded by parallel vertical lines on Figure 2(b) is an area where a cluster does not exist due to geometrical constraints. Number I denotes the level line $R_0 = \rho$. (see assumption (1)) and number II shows level curves $\sigma^{1/2} = l$ (see assumption (2)). It is seen that increase of the initial bubble radius from $a_0 = 5\ mkm$ to $a_0 = 25\ mkm$ leads to an increase and shift to the areas, where the condition $\sigma^{1/2} << l$ is not satisfied.

**RESULTS**

The analytical study of small bubble oscillations based on the bubble cluster model proposed in this paper showed that the natural frequency of bubble oscillations in a monodisperse cluster is expressed by formula

\[
\omega_n^2 = \frac{3p_0 + 2\sigma(3\gamma - 1)}{\rho_0 a_{0}^2(1 + N_{a_{0}}/R_0)} < \omega_M^2
\]

This is lower than the natural frequency of a single bubble (Minnaert frequency) with the same initial radius.

**Figure 2:** Areas of validity of mathematical model shaded by parallel lines for: (a) $a_0 = 5\ mkm$ and (b) $a_0 = 25\ mkm$. The area shaded by parallel vertical lines indicates the area were no cluster exist with these parameters. Level curves: I – $R_0 = \lambda$; II – $\sigma^{1/2} = l$.

**Figure 3:** Amplitude-frequency characteristics for monodisperse (top left) clusters with $N = 5000$ each and two-fraction (bottom left) clusters with $N_1 = N_2 = 5000$, and typical bubbles oscillations in two-fraction cluster in the vicinity of the main resonance (top right) and the secondary resonance (bottom right). Vertical dotted lines indicate values of Minnaert frequencies $\omega_M^{(3)}$ and $\omega_M^{(10)}$ for a single bubble of initial radius of 5 mkm and 10 mkm respectively. Solid lines indicate curves for bubble of 5 mkm initial radius; dashed lines indicate curves for bubble of 10 mkm initial radius.

The expression for bubble amplitude as function of frequency where the acoustic field is taken into account has
been obtained. Figure 3(left) shows the amplitude-frequency functions for monodisperse cluster (top graph) and two-fraction cluster (bottom graph). Physical parameters correspond to the parameters of air and water: $\rho_l = 10^3 \text{kg/m}^3$, $C_l = 1500 \text{m/s}$, $p_0 = 10^5 \text{Pa}$, $\sigma = 0.073 \text{N/m}$, $\mu = 10^{-3} \text{Pa's}$, $\gamma = 1.4$. Initial cluster radius is $R_0 = 10^{-3} \text{m}$.

One can see that for two-fraction cluster besides the main resonance, which takes place at low frequency, there is a secondary resonance at higher frequency. Figure 3(right) demonstrates the nature of nonlinear oscillations of bubbles in cluster in an acoustic field of $\Delta P = 5 \times 10^5 \text{Pa}$ in the vicinity of these two resonances. It is seen that in the vicinity of the main resonance (at $\omega = 2.9 \times 10^5 \text{s}^{-1}$) bubbles in both fractions oscillate in phase. In the vicinity of the second resonance (at $\omega = 4 \times 10^6 \text{s}^{-1}$) they oscillate out of phase, while the period of expansion of bubbles of the first fraction coincides with the period of contraction of bubbles of the second one.

The equations (1–5) have been solved numerically using the Runge-Kutta method based on Dorman-Prince formulas with automatic time-step control [10] for the analysis of nonlinear oscillations of bubbles in the cluster.

The phenomenon of phase synchronization of bubbles collapse in polydisperse cluster is shown in Figure 4. The Figure 4(a) shows that in two separate monodisperse clusters with different initial bubble radii ($a_{01} = 5 \text{ mkm}$ and $a_{02} = 10 \text{ mkm}$) the bubbles reach their minimum sizes at different times. But if these bubbles are placed in one cluster making two-fraction cluster, they reach their minimum sizes almost at the same time. Moreover, it is evident that the amplitude of a bubble expansion in the first fraction became lower, while the amplitude of a bubble in the second fraction increased. For clarity there is a graph in Figure 4(b) illustrating the phase difference of the collapse of bubbles of various initial radii calculated by the formulae

$$\Delta \phi = \phi_{1}^{(\text{col})} - \phi_{2}^{(\text{col})}, \quad \phi_{k}^{(\text{col})} = a t_{k}^{(\text{col})}, \quad k = 1, 2,$$

Here $t_{k}^{(\text{col})}$ is the time when the bubble of initial radius of $a_{0k}$ reaches its minimal value. The difference between the collapse phases for two monodisperse clusters is shown by dots (left scale), and for two-fraction cluster is shown by triangles (right scale).

Figure 5 shows the phenomenon of an intensification of the bubbles collapse, which is the result of energy exchange between the fractions. Namely, adding a certain number of bubbles with specific radius in the cluster containing bubbles with another radius can lead to a deeper collapse of the latter ones. This result is consistent with experiments [11], where it was found that in a cluster with two bubble sizes, small bubbles collapse deeper in the presence of large bubbles.

![Figure 5: Maximum temperature in the bubble of initial radius of $a_{01} = 5 \text{ mkm}$ vs. total number of bubbles in cluster at $\Delta P = 5 \times 10^5 \text{Pa}$.
For the case $n = 2$: $N_1$ is fixed, $N_2$ is variable, $a_{02} = 10 \text{ mkm}$.

Let us assume now that the mass transfer due to rectified diffusion is taken into account (assumption (5) is not valid).

Diffusion between the bubble and the liquid is calculated using diffusion averaging approximation developed in [12]. The approximation is carried out for large Peclet numbers ($Pe = a_0^3 \alpha / D \gg 1$, where $D$ is the diffusion coefficient of gas in liquid) by transforming to the normalized Lagrangian coordinate $\zeta = (r^3 - a^3(t))/[3a_0^3]$, where $r$ is the distance to the centre of the bubble, and by averaging over time.

Then the rate of gas mass transfer through the bubble interface averaged over the period of bubble oscillation is represented as follows:

$$\frac{d \bar{m}}{dt} = \frac{\bar{c}_e - \langle \bar{c} \rangle}{T_{rd}}, \quad T_{rd} = \frac{\int_0^T d \eta}{\int_0^T [3\eta + \bar{a}(\eta)]}, \quad (6)$$

where

$$\bar{c} = \frac{c(a(t), t)}{c_0}, \quad \bar{c}_e = \frac{c_{e_0}}{c_0}, \quad \bar{m} = \frac{m}{m_0}, \quad \bar{a} = \frac{a}{a_0}.$$
Here \( \tau = tD/a_0^2 \) is the slow diffusion time scale; \( T_{\text{nd}} \) is the dimensionless characteristic time of the growth rate of bubble mass due to diffusion; \( \bar{\gamma} \) is the period of external acoustic field;
\( c(a(t),t) = \frac{2\gamma}{a_0} \frac{a(t)}{a_0} \),

\[ \langle \bar{c} \rangle_t = \frac{1}{T} \int_0^T a^4(t)c(t)dt. \tag{7} \]

Gas concentration dissolved in the liquid at the bubble interface was calculated by the formula

\[ c(a(t),t) = H \left( p_0 + \frac{2\gamma}{a_0} \frac{a(t)}{a_0} \right)^{-3/2}, \tag{8} \]

where \( H = c_0 / p_0 \) is the Henry constant.

For the numerical calculations of mass transfer through the surface of the bubble in the cluster the approximation formulas (6), (7) are used, where gas concentration near the bubble wall is found from the boundary condition (8) and the radius of the bubble \( a(t) \) is found from the solution of the system of equations (1-5) for \( n = 1 \).

Normalized maximum bubble radius \( a_{\text{max}}/a_0 \) and curves of the average gas concentration near the bubble wall \( \langle \bar{c} \rangle_t \), for different amplitudes of the acoustic field \( \Delta P \) are shown in a Figure 6. Figure 6(b) allows to compare results for a bubble in a monodisperse cluster with similar results for a single bubble (see Figure 6(a)) calculated in [13]. Maximum responses for a bubble in a monodisperse cluster (Figure 6(top)) for the amplitudes of pressure \( \Delta P = 1.1 \times 10^5 \) Pa occur for about the same size of initial radius as for a single bubble. However the magnitude of the response is much smaller. For example, at \( \Delta P = 1.5 \times 10^3 \) Pa resonant radius is about \( a_0 \approx 1.5 \text{ mkm} \) both for a single bubble and for a bubble in a cluster, but the value of the response in the former case is 6.5 times larger.

Curves of the averaged gas concentration \( \langle \bar{c} \rangle_t \), near the bubble interface are shown in a Figure 6(bottom). These curves are calculated by formula (7) for a single bubble (see Figure 6(a)) and for a bubble in the cluster (see Figure 6(b)) depending on the initial bubble radius for different amplitudes of the pressure. In this range of initial radius values the character of curves for the averaged concentration of a bubble in the cluster (see Figure 6(b)) is the same as in the case of a single bubble. Namely, the concentration decreases monotonically while initial radius increases to certain threshold value of the driving pressure amplitude \( \Delta P \).

**Figure 6**: Normalized maximum radius (top) and the average concentration of gas near the bubble wall (bottom), depending on the initial radius for different pressure amplitudes:

(a) single bubble;

(b) bubble in a monodisperse cluster.

Solid curves: I – \( \Delta P = 1.1 \times 10^5 \) Pa; II – \( \Delta P = 1.2 \times 10^5 \) Pa; III – \( \Delta P = 1.3 \times 10^5 \) Pa; IV – \( \Delta P = 1.4 \times 10^5 \) Pa; V – \( \Delta P = 1.5 \times 10^5 \) Pa. The horizontal dotted lines represent different levels of initial concentration of gas in a liquid, open circle denotes an unstable equilibrium radius, filled circle denotes a stable equilibrium radius.

The averaged rate of the gas mass in a bubble depends only on the difference between gas concentration in a liquid \( \bar{c}_0 \) and averaged concentration near the bubble interface \( \langle \bar{c} \rangle_t \) (see formula (6)). There is a range of values \( \bar{c}_0 \) for given pressure amplitudes, when there is one equilibrium point \( \langle \bar{c} \rangle_t = \bar{c}_0 \), which is unstable (the open circle at intersection of the curve at \( \Delta P = 1.2 \times 10^5 \) Pa and the upper dash line). This equilibrium point corresponds to the initial radius value \( a_{\text{un}}^{(1)} \). For \( a_0 < a_{\text{un}}^{(1)} \) bubbles dissolve and for \( a_0 > a_{\text{un}}^{(1)} \) bubbles grow up until they are destroyed by the shape instability. Once the certain threshold value of \( \Delta P \) is reached the concentration curve \( \langle \bar{c} \rangle_t(a_0) \) is getting non-monotous as a result of non-monotonous bubble response shown at the top of Figure 6. With such a non-monotous dependence \( \langle \bar{c} \rangle_t \) vs. \( a_0 \) there is a range of values \( \bar{c}_0 \) for which there are two equilibrium points \( \langle \bar{c} \rangle_t = \bar{c}_0 \). Unstable point (the open circle at intersection of the curve at \( \Delta P = 1.5 \times 10^5 \) Pa and the under dash line) corresponds to radius \( a_0 = a_{\text{st}}^{(2)} \) and stable point (the filled circle) corresponds to radius \( a_0 = a_{\text{st}}^{(1)} \). For \( a_0 < a_{\text{st}}^{(1)} \) bubbles dissolve until they disappear, for \( a_{\text{st}}^{(2)} < a_0 < a_{\text{st}}^{(1)} \) bubbles grow up until they reach equilibrium radius \( a_{\text{st}}^{(1)} \) and, finally, for \( a_0 > a_{\text{st}}^{(2)} \) bubbles dissolve until they reach \( a_{\text{st}}^{(2)} \). However, the equilibrium in bubble cluster is possible at larger (few hundred
times larger) initial concentrations of dissolved gas than in the case of a single bubble. It means that purified liquid separated well from the gas is required to obtain a diffusively stable single bubble, whereas in not purified liquid the appearance of diffusively stable bubble cluster is most likely to occur.

The bubble maximum response and the averaged gas concentration at interface for a bubble initial radius values up to \( (a_0 = 1\pm 15 \text{ mkm}) \) for amplitude of the external pressure \( \Delta P = 1.5 \times 10^5 \text{ Pa} \) are presented in Figure 7. While initial radius increases the curve of bubble maximum response becomes non-monotonic (Figure 7(top)) and new equilibrium points appear on the curve of averaged gas concentration at interface (Figure 7(bottom)). With further increase in the radius the period doubling bifurcation is observed. The shape of the curve of gas averaged concentration near the bubble interface also changes (Figure 7(bottom)). With period doubling of bubble oscillation the averaged over period of acoustic driving concentration curve is also bifurcates. So averaging over the period of the bubble oscillation (dashed curve) must be performed rather than over the period of the acoustic field. One can see that at low concentrations \( \bar{c}_a \) there are no equilibrium points at all. With increase of \( \bar{c}_a \) there are two equilibrium points (stable and unstable). If \( \bar{c}_a \) increases further then four points of equilibrium appear (two stable and two unstable). This case is shown in Figure 7: filled circles correspond to stable points, open ones correspond to unstable points. Maximum radii corresponding to them are shown by vertical dotted lines. For given values of initial concentration one has the following. For \( a_0 < a^{(1)}_m \) bubbles dissolve until they completely disappear; for \( a^{(1)}_m < a_0 < a^{(2)}_m \) and \( a^{(1)}_m < a_0 < a^{(2)}_m \) bubbles grow or dissolve, respectively, until they reach an equilibrium size \( a^{(1)}_m \); for \( a^{(2)}_m < a_0 < a^{(2)}_m \) and \( a_0 > a^{(2)}_m \) bubbles grow or dissolve, respectively, until they reach an equilibrium size \( a^{(2)}_m \).

In case of two-fraction cluster there is an interaction between bubbles of different radii through pressure \( p_c \) inside the cluster. Consequently, the averaged gas concentration near the bubble interface for each fraction will depend not only on the radius of the bubble in this fraction, but also on the radius of the bubble in the other fraction. Thus, the averaged gas concentrations near the bubble walls can be represented as a function of two radii:

\[
\langle \bar{c}_a (a_{01}, a_{02}) \rangle_c, \langle \bar{c}_a (a_{01}, a_{02}) \rangle_c.
\]

For two-fraction cluster at a fixed value of \( \bar{c}_a \) the equilibrium curves \( \langle \bar{c}_a \rangle = \bar{c}_a \) will be obtained instead of the equilibrium points. Figure 8 shows these equilibrium curves for \( \bar{c}_a = 0.12, \Delta P = 1.5 \times 10^5 \text{ Pa} \). The amounts of bubbles in the fractions are \( N_1 = 3000 \) and \( N_2 = 7000 \). The arrows show the direction of initial radius alteration (growth or dissolution of bubble): thick arrows are for the first fraction, thin arrows are for the second fraction. One can see that the equilibrium curves of different fractions cross when \( a_{01} = a_{02} \) (the case of monodisperse cluster). Thus there are three different scenarios for a given initial gas concentration in the liquid:

(a) the bubbles in both fractions will dissolve (this is implemented in a very small region, not visible in the figure, where the initial bubble radii in both fractions less than 1 mkm);

(b) the bubbles of one fraction will dissolve or grow until they disappear, while the bubbles of another fraction converge to its equilibrium radius;

(c) the bubbles of both fractions will tend to the same radius.

\[\begin{align*}
\text{Figure 7: Normalized maximum radius (top) and averaged gas concentration near the bubble wall (bottom) vs. initial radius for pressure amplitude } \Delta P = 1.5 \times 10^5 \text{ Pa.}
\end{align*}\]

\[\begin{align*}
\text{Figure 8: The equilibrium curves for bubbles in two-fraction cluster for } \bar{c}_a = 0.12.
\end{align*}\]

CONCLUSION

In this paper a mathematical model of bubble cluster is presented that allows studying:

- overall dynamics of the cluster as single mechanical object driven by external acoustic field;
- dynamics of individual bubbles inside the cluster;
- bubbles of different radii in the cluster and their interaction;
- gas diffusion between bubbles and liquid in the cluster.

The ranges of parameters of applicability of this model have been determined. The analytical study of small bubble oscillations based on the bubble cluster model proposed in this paper showed that the natural frequency of bubble oscillations
in a monodisperse cluster is lower than the natural frequency of a single bubble (Minnaert frequency) with the same initial radius. It was also found that in a two-fraction cluster the secondary resonance in addition to the main resonance is present. In the range of the main resonance bubbles of different fractions oscillate in phase whereas in the range of the secondary resonance they oscillate out of phase.

The numerical analysis of nonlinear bubble oscillations in a polydisperse cluster revealed synchronization of the collapse phases of bubbles with different radii and collapse intensification for bubbles of one size in the presence of bubbles of other size.

The study of gas diffusion between bubble and liquid has shown that the diffusion stability of bubbles in a monodisperse cluster differs from the one for a single bubble in an infinite liquid. Namely an equilibrium radius in a cluster may be observed at higher values of dissolved gas concentration than for a single bubble. It means that a bubble in a monodisperse cluster is more diffusively stable than a single bubble. The numerical study of the diffusion in two-fraction cluster shows that the cluster has a tendency to become monodisperse or to disappear under the action of the acoustic field.

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