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DIFFUSION STABILITY OF BUBBLES IN A CLUSTER

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The diffusion stability of gas bubbles in one-fraction and two-fraction clusters subjected to an acoustic field is studied. For a one-fraction cluster, numerical values were obtained for the initial gas concentrations in the liquid at which the bubble tends to one of two equilibrium states because of diffusion processes between the bubble and the ambient liquid. It is found that a two-fraction cluster tends to become a one-fraction cluster.

Key words: *diffusion, bubble cluster, stability, equilibrium radius.*

Introduction. Acoustic cavitation experiments have shown that in the presence of an acoustic field, small gas bubbles grow with time. This phenomenon is explained as follows [1–4]. In the absence of an acoustic field, the bubble pressure is higher than the pressure of the ambient liquid; as a result, the gas diffuses from the bubble into the liquid and the bubble dissolves slowly. Thus, in the state of rest, the bubble is diffusively unstable. When an acoustic field is switched on and reaches a certain threshold amplitude, the bubble begins to grow. During oscillations of the bubble, its expansion leads to gas diffusion from the liquid into the bubble and bubble compression leads to gas diffusion from the bubble into the liquid. During expansion, the amount of the gas passing from the liquid into the bubble is larger than the amount of the gas leaving the bubble during compression since the surface area of the bubble increases during expansion. Therefore, for the total period of oscillation, diffusion should result in the growth of the bubble. This process was called directional diffusion. The mathematical theories describing directional diffusion for the case of small amplitudes of bubble oscillation are considered in [1, 2, 4]. It has been established that the threshold amplitude of the acoustic field at which the bubble begins to grow depends on the gas concentration, the bubble radius, and the frequency of the acoustic field. Calculations of the threshold value for various values of these parameters were performed in [2–4].

After the discovery of single-bubble sonoluminescence in 1991 [5, 6], it has been found that in a strong acoustic field, a single bubble could oscillate without changing size for several days. Because this phenomenon cannot be explained by the theory set forth above, further investigation of the problem in question is required. There is no analytical solution of the complete system of equations of the diffusion problem; therefore, we will solve this problem numerically [7]. However, the change in the bubble mass with time is very small and comparable to the calculation error; therefore, it is impossible to uniquely determine whether the bubble grows or the result is affected by the calculation error and the bubble size does not change over the oscillation period. In addition, numerical solution of the complete system of equations requires considerable computation time. In this connection, an adiabatic approximation of the diffusion problem was proposed [8, 9], which allows large-amplitude oscillations to be calculated for large Peclet numbers. Hilgenfeldt et al. [7] compared the solution for the adiabatic approximation and the solution for the complete model and obtained good agreement. In addition, the phase diagram in the space of concentration and pressure parameters was calculated, which can be used to determine which of the three cases occurs: diffusively stable sonoluminescence, diffusively unstable sonoluminescence, or the absence of sonoluminescence. Akhatov et al. [10] developed a theory for stable oscillations of bubbles subjected to a strong acoustic field. This theory takes into account the effect of surface tension and directional diffusion on the dynamics

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of small bubbles and explains the existence of small stably oscillating single bubbles observed in sonoluminescence experiments.

Previous papers [1–10] dealt with the diffusion process only for single gas bubbles but diffusion in bubbles located in large aggregates (bubble clouds or bubble clusters) is also an important problem. This problem is related, for example, to the study of the processes involved in multibubble sonoluminescence. The existing mathematical models describing the bubble dynamics in bubble clusters (ignoring diffusion) can be divided into two main groups [11]. In the first group [12–14], a mixture of a liquid and gas bubbles is treated as a continuum, the cluster dynamics is studied using averaged equations of the bubbly liquid model, and the dynamics of single bubbles is investigated using linearized Rayleigh–Plesset equations. The second group of studies deal primarily with the dynamics of a single bubble in a cluster and the interaction between bubbles [15, 16] without considering the dynamics of the cluster as whole. Thus, this approach is confined to a small number of bubbles arranged in special configurations. In a previous study [11], we proposed a mathematical model for a bubble cluster which allows one to study not only the dynamics of the cluster as a whole but also the dynamics of single bubbles in the cluster, taking into account the presence of bubbles of various radii in the cluster and their interaction.

The present work is a continuation of [11]. The mathematical model proposed previously is used to study the diffusive stability of gas bubbles in bubble clusters subjected to an acoustic field. The diffusion processes between the bubble and liquid are examined using an approximation of the diffusion problem [8], and the variation of the bubble radius using the bubble cluster model [11].

1. Formulation of the Problem. We consider a set of gas bubbles of various radii which perform oscillations in a finite volume of an unbounded, slightly compressible viscous liquid under the action of an acoustic field. A spherical region filled with bubbles — a bubble cluster — is distinguished in the liquid. In this case, the cluster can be treated as a large drop which contains a liquid and a set of microbubbles. Under the assumption that the cluster size is smaller than the length of the acoustic wave, the pressure in the cluster is considered uniform [17]. It is also assumed that the bubbles perform spherically symmetric radial motion, the gas pressure in the bubble varies adiabatically, and there are no heat and mass transfer processes in the system of the gas bubbles and liquid.

The bubbles in the cluster are divided into a finite number of fractions, each of which is characterized by the size of the initial bubble radius. Then, the equations describing the oscillations of the cluster and bubble oscillations in the cluster are written as follows [11]:

$$a_i\ddot{a}_i + 3\dot{a}_i^2/2 = (p_{ai} - p_c)/\rho_l, \qquad i = \overline{1,n}; \tag{1}$$

$$R\ddot{R} + 3\dot{R}^2/2 = (p_c - p_I)/\rho_l + R(\dot{p}_c - \dot{p}_I)/(\rho_l C_l); \tag{2}$$

$$\sum_{i=1}^{n} N_i a_i^2 \dot{a}_i = R^2\dot{R}; \tag{3}$$

$$p_{ai} = \Big(p_0 + \frac{2\sigma}{a_{0i}}\Big)\Big(\frac{a_i}{a_{0i}}\Big)^{-3\gamma} - \frac{4\mu\dot{a}_i}{a_i} - \frac{2\sigma}{a_i}, \qquad i = \overline{1,n}; \tag{4}$$

$$p_I = p_0 - \Delta P \sin(\omega t). \tag{5}$$

Here $a_i = a_i(t)$ is the bubble radius in the ith fraction, p_{ai} is the gas pressure at the wall of a bubble of the ith fraction, $p_c = p_c(t)$ is the liquid pressure in the cluster, ρ_l is the density of the liquid, n is the number of fractions, $R = R(t)$ is the cluster radius, p_I is the external pressure, C_l is the sound velocity in the liquid, N_i is the number of bubbles in the ith fraction, p_0 is the initial pressure in the liquid, σ is the surface-tension coefficient, a_{0i} is the initial bubble radius in the ith fraction, γ is the adiabatic exponent, μ is the dynamic viscosity of the liquid, ΔP is the external-pressure amplitude, ω is the angular frequency, and t is time; the point above a variable denotes the time derivative. For $n = 1$, system (1)–(5) corresponds to the case of a monodisperse (or one-fraction) cluster, and the subscript $i = 1$ is omitted.

We consider the diffusion problem for a bubble in a monodisperse cluster. In the present paper, the diffusion processes between the bubble and the liquid are studied using an approximation of the diffusion problem [8]. Performing approximation for large Peclet numbers $\mathrm{Pe} = a_0^2\omega/D \gg 1$ (D is the diffusion coefficient for the gas in the liquid) by transforming to the normalized Lagrangian coordinate $\eta = (r^3 - a^3(t))/(3a_0^3)$ (r is the distance to the center of the bubble) and averaging over time, we can represent the rate of gas mass transfer through the bubble surface averaged over the period of bubble oscillation in the form

$$\frac{d\bar{m}}{d\tau} = \frac{\bar{c}_\infty - \langle \bar{c} \rangle_\tau}{T_{rd}}, \qquad T_{rd} = \int\limits_0^\infty \Big(\frac{1}{T} \int\limits_0^T [3\eta + \bar{a}^3(t)]^{4/3}\, dt \Big)^{-1} d\eta,$$
$$\bar{c} = \frac{c(a(t), t)}{c_0}, \qquad \bar{c}_\infty = \frac{c_\infty}{c_0}, \qquad \bar{m} = \frac{m}{m_0}, \qquad \bar{a} = \frac{a}{a_0}. \tag{6}$$

Here $\tau = tD/a_0^2$ is the slow diffusion time scale, T_{rd} is the dimensionless characteristic time of the rate of diffusion growth of the bubble mass, $a = a(t)$ is the bubble radius, T is the oscillation period of the acoustic field, $c(a(t), t)$ is the mass concentration of the gas dissolved in the liquid at the bubble wall, c_0 is the gas-saturation concentration in the liquid at a pressure p_0, c_∞ is the initial uniform gas concentration in the liquid, and m_0 is the mass of the gas dissolved in the liquid that occupies the volume of the unperturbed bubble. The average gas concentration at the bubble wall for the oscillation period of the acoustic field is given by the expression

$$\langle \bar{c} \rangle_\tau = \int\limits_0^T a^4(t) \bar{c}\, dt \Big/ \int\limits_0^T a^4(t)\, dt \tag{7}$$

and the gas concentration on the bubble boundary is calculated by the formula

$$c(a(t), t) = H(p_0 + 2\sigma/a_0)(a(t)/a_0)^{-3\gamma}, \tag{8}$$

where $H = c_0/p_0$ is the Henry constant.

2. Numerical Calculations for a Monodisperse Cluster. Numerical calculations of the mass transfer through the bubble surface in the cluster are made using the approximation formulas (6) and (7), in which the gas concentration at the bubble wall is found from boundary conditions (8) and the bubble radius $a(t)$ from system (1)–(5) for $n = 1$. In the calculations, we used the values of the physical parameters typical of water and air: $\rho_l = 10^3$ kg/m^3, $C_l = 1500$ m/sec, $p_0 = 10^5$ Pa, $\sigma = 0.073$ N/m, $\mu = 10^3$ Pa$\cdot$ sec, and $\gamma = 1.4$; the initial cluster radius was $R_0 = 10^{-3}$ m, and the number of bubbles in the cluster was $N = 10^4$.

Figures 1 and 2 shows curves of the normalized maximum bubble radius $a_{\max}/a_0$ and the average gas concentration at the bubble wall $\langle \bar{c} \rangle_\tau$ versus initial radius a_0 for various amplitudes of the acoustic field ΔP. Figure 1 compares the results for a bubble in a monodisperse cluster with similar results for a single bubble given in [10]. In Fig. 1a, it is evident that for pressure amplitudes $\Delta P = 1.1 \cdot 10^5$–$1.5 \cdot 10^5$ Pa, the maximum responses for the bubble in a monodisperse cluster occur for the same size of the initial radius as for a single bubble but the magnitude of the response is much smaller. For example, at $\Delta P = 1.5 \cdot 10^5$ Pa, the resonant radius $a_0 \approx 1.5$ μm for both the single bubble and the bubble in the cluster, but the value of the response in the former case is 6.5 times larger. Figure 2a shows the normalized maximum radius for a bubble in a stronger acoustic field ($\Delta P = 10^5$–$4 \cdot 10^5$ Pa). It is evident that as the pressure amplitude increases, the maximum response grows.

Figure 1b shows that in the given range of the initial radius, the behavior of the curves of the average concentration for a single bubble is similar to that for the case of the bubble in the cluster: as initial radius increases, the concentration decreases monotonically to a certain threshold value of the external pressure amplitude ΔP. From relation (6) it follows that the average rate of variation in the mass of the bubble gas depends only on the difference between the gas concentration in the liquid $\bar{c}_\infty$ and the average concentration at the bubble wall $\langle \bar{c} \rangle_\tau$. Then, for these pressure amplitudes there is a range of values $\bar{c}_\infty$ with one equilibrium point $\langle \bar{c} \rangle_\tau = \bar{c}_\infty$, which is unstable (point I in Fig. 1b) and corresponds to the initial radius $a_{\text{un}}^{(1)}$. For $a_0 < a_{\text{un}}^{(1)}$, the bubbles dissolve, and for $a_0 > a_{\text{un}}^{(1)}$, they grow until they collapse because of surface instability. Once the threshold amplitude of the external pressure ΔP is reached, the average gas concentration curve becomes nonmonotonic, and in the range of small values of the initial bubble radii there is a global minimum, which corresponds to the global maximum in Fig. 1a. For this nonmonotonic dependence $\langle \bar{c} \rangle_\tau(a_0)$, there is a range of values of $\bar{c}_\infty$ which contains two equilibrium points $\langle \bar{c} \rangle_\tau = \bar{c}_\infty$. The unstable point II in Fig. 1b corresponds to the value of the radius $a_0 = a_{\text{un}}^{(2)}$, and the stable point III to the value of the radius $a_0 = a_{\text{st}}^{(2)}$. For $a_0 < a_{\text{un}}^{(2)}$, the bubbles dissolve until complete disappearance, and for $a_{\text{un}}^{(2)} < a_0 < a_{\text{st}}^{(2)}$, the bubbles grow until they reach the size of the radius $a_{\text{st}}^{(2)}$, which corresponds to the equilibrium state. For $a_0 > a_{\text{st}}^{(2)}$,

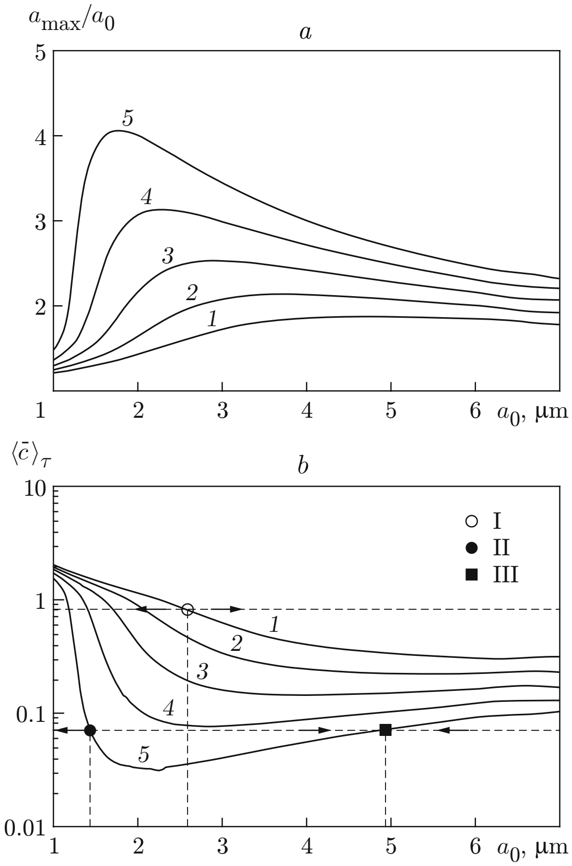

Fig. 1

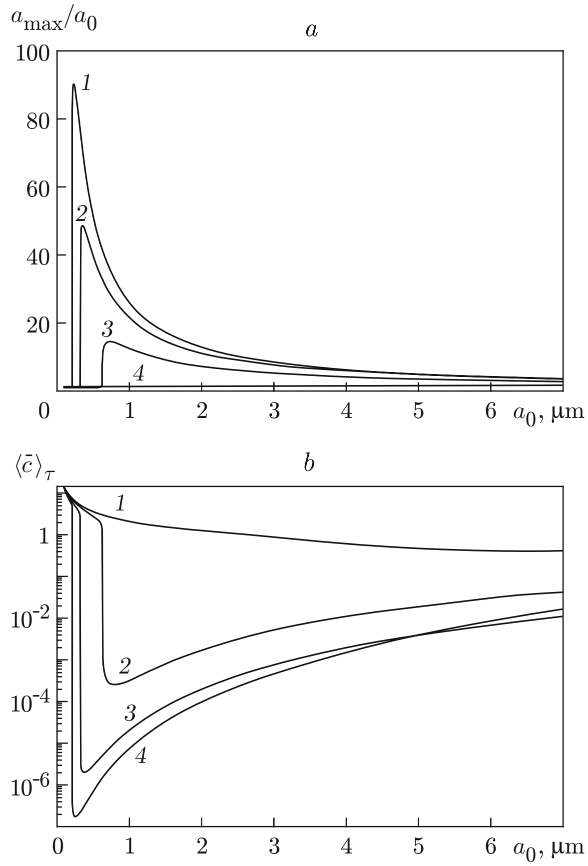

Fig. 2

Fig. 1. Normalized maximum radius (a) and average gas concentration at the bubble wall (b) in a monodisperse cluster versus initial radius for pressure amplitudes $\Delta P = 1.1 \cdot 10^5$ (1), $1.2 \cdot 10^5$ (2), $1.3 \cdot 10^5$ (3), $1.4 \cdot 10^5$ (4), and $1.5 \cdot 10^5$ Pa (5); the horizontal lines are the gas concentration levels $\bar{c}_\infty$ in the liquid; point I refers to the initial unstable equilibrium radius $a_{\text{un}}^{(1)}$, point II to the unstable equilibrium radius $a_{\text{un}}^{(2)}$, and point III to the stable equilibrium radius $a_{\text{st}}^{(2)}$; the arrows show the direction of change of the initial bubble radius.

Fig. 2. Normalized maximum radius (a) and average gas concentration at the bubble wall (b) in a monodisperse cluster versus initial radius for pressure amplitudes: $\Delta P = 10^5$ (1), $2 \cdot 10^5$ (2), $3 \cdot 10^5$ (3), and $4 \cdot 10^5$ Pa (4).

the bubbles dissolve until they reach the size of the radius $a_{\text{st}}^{(2)}$. In the bubble cluster, however, equilibrium becomes possible at higher (a few hundred times) initial gas concentrations than in the case of a single bubble. Consequently, a purified liquid separated well from the gas is required to obtain a diffusively stable single bubble, whereas in an unpurified liquid, a bubble cluster, also diffusively stable, is most likely to occur.

Figure 2b shows the average gas concentration at the bubble wall at high pressures. It is evident that as the oscillation amplitude of the acoustic field increases, the range of concentrations $\bar{c}_\infty$ in which the diffusion stability is possible is extended. In addition, as the concentration decreases, the distance between the stable and unstable points of equilibrium decreases considerably.

Figure 3 shows curves of the equilibrium radius of a single bubble and a bubble in a cluster (stable equilibrium point) versus pressure amplitude for various initial gas concentrations in the liquid. For the single bubble, this dependence is monotonic; in addition, for pressure amplitudes $\Delta P > 3 \cdot 10^5$ Pa, the bubble radius in the equilibrium state becomes very large, and, because of surface instability, the bubble can collapse without reaching the equilibrium state. It should be noted that this dependence is observed only for small values of $\bar{c}_\infty$. Therefore, the single bubble is

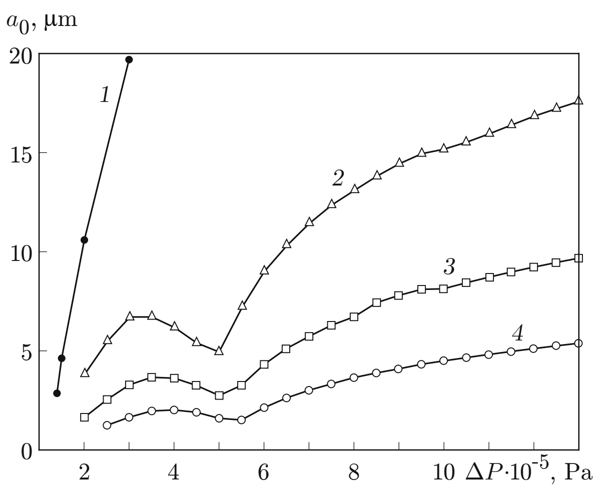

Fig. 3

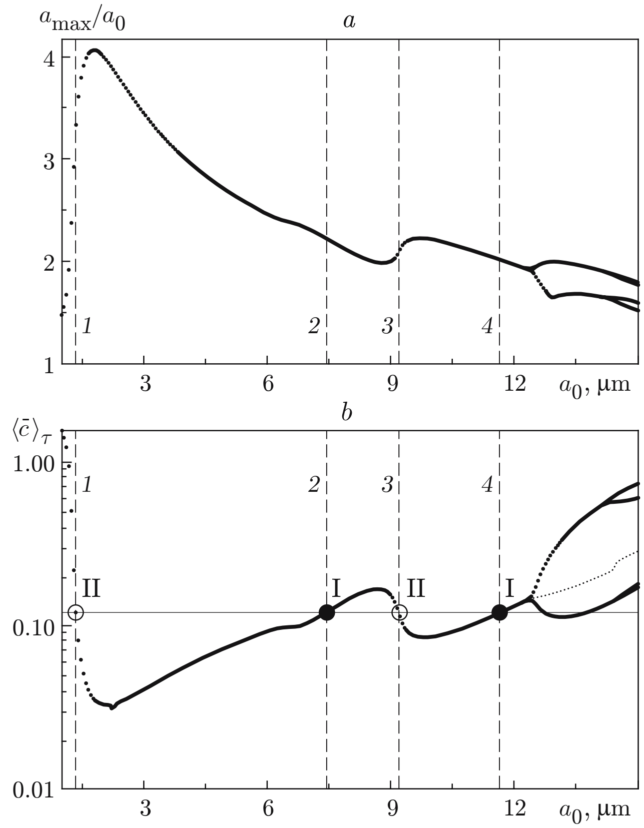

Fig. 4

Fig. 3. Equilibrium bubble radius versus pressure amplitude for various initial gas concentrations in the liquid: curve 1 refers to the single bubble ($\bar{c}_\infty = 10^{-4}$) and curves 2–4 refer to the bubble in the cluster [$\bar{c}_\infty = 10^{-2}$ (2), 10^{-3} (3), and 10^{-4} (4)].

Fig. 4. Normalized maximum radius (a) and average gas concentration at the bubble wall (b) in a monodisperse cluster versus initial radius for $\Delta P = 1.5 \cdot 10^5$ Pa: solid curves refer to averaging over the period of the acoustic field; dashed curves refer to averaging over the bubble oscillation period; dashed curves refer to the maximum radii $a_{\rm un}^{(3)}$ (1), $a_{\rm st}^{(3)}$ (2), $a_{\rm un}^{(4)}$ (3), and $a_{\rm st}^{(4)}$ (4); points I refer to stable equilibrium points and points II refer to the unstable equilibrium points.

in principle diffusively unstable. In the cluster, a nonmonotonic dependence of the bubble radius in the equilibrium state on the pressure amplitude is observed. Furthermore, even at large oscillation amplitudes of the external field, the bubble radius in the equilibrium state is not large enough for the bubble to collapse because of surface instability. Thus, even in a strong acoustic field, the bubble in the cluster is diffusively stable.

Figure 4 shows the calculation results for the parameter $a_0 = 1$–15 μm and the oscillation amplitudes of the external pressure $\Delta P = 1.5 \cdot 10^5$ Pa. It is evident that as the initial radius increases, the curve of the maximum response of the bubbles becomes nonmonotonic (Fig. 4a) and new extreme points appear. With further increase in the radius, doubling of the period is observed (bifurcation). The behavior of the average gas concentration curves at the bubble wall also varies (Fig. 4b). If the bubble oscillation period is doubled, the concentration curve is also bifurcated; therefore, the averaging is performed over the oscillation period of the bubble rather than over the oscillation period of the acoustic field. Consequently, the concentration curve calculated for the bubble oscillation period is between the concentration curves calculated for the oscillation period of the acoustic field. In addition, Fig. 4b shows the range of values of $\bar{c}_\infty$ in which equilibrium points (two stable and two unstable) exist. The dashed curves show the maximum radii corresponding to this points. For $a_0 < a_{\rm un}^{(3)}$, the bubbles dissolve until complete disappearance; for $a_{\rm un}^{(3)} < a_0 < a_{\rm st}^{(3)}$ and $a_{\rm st}^{(3)} < a_0 < a_{\rm un}^{(4)}$, the bubbles grow or dissolve, respectively, until they reach

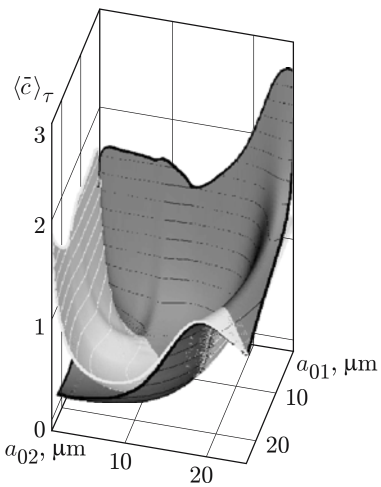

Fig. 5

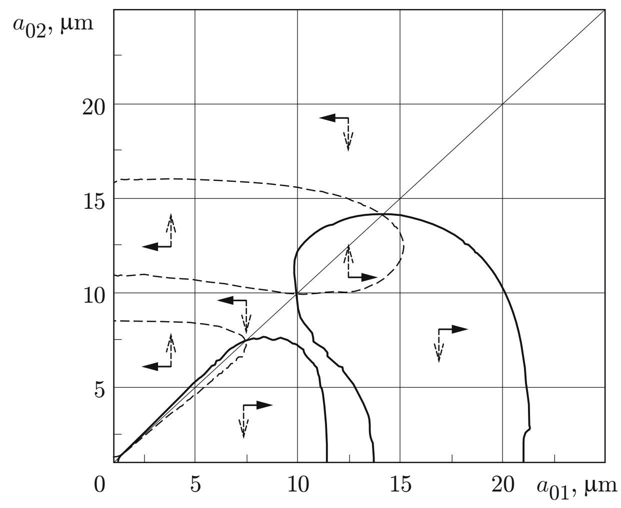

Fig. 6

Fig. 5. Average gas concentration at the bubble wall in a two-fraction cluster versus initial radii for $\Delta P = 1.5 \cdot 10^5$ Pa: the dark regions are the first fraction, the light regions are the second fraction.

Fig. 6. Equilibrium curves for bubbles in a two-fraction cluster ($\bar{c}_\infty = 0.12$): solid curves refer to the first fraction, and dashed curves to the second fraction; solid arrows shows the direction of change of the initial bubble radius for the first fraction, and the dashed arrows the same for the second fraction.

the equilibrium size $a_{\rm st}^{(3)}$; for $a_{\rm un}^{(4)} < a_0 < a_{\rm st}^{(4)}$ and $a_0 > a_{\rm st}^{(4)}$, the bubbles grow or dissolve, respectively, until they reach the equilibrium size $a_{\rm st}^{(4)}$.

3. Numerical Calculations for a Two-Fraction Cluster. Setting $n = 2$ in system (1)–(5), we consider a two-fraction cluster. In each of the fractions, the bubble concentration forms a pressure p_c in the cluster. At the same time, the pressure in the cluster influences the nature of oscillation of the bubbles. Thus, the given model assumes interaction between bubbles of various radii in the cluster; therefore, for each fraction, the average gas concentration at the bubble wall depends not only on the bubble radius in this fraction but also on the bubble radius of the other fraction.

In Fig. 5, the average gas concentrations for each fraction is shown as a function of the initial radii a_{01} and a_{02} for an external-pressure amplitude $\Delta P = 1.5 \cdot 10^5$ Pa. The number of bubbles in the first and second fractions was set equal to $N_1 = 3000$ and $N_2 = 7000$, respectively. For a two-fraction cluster with a fixed value $\bar{c}_\infty$, instead of the equilibrium points we obtain equilibrium curves $\langle\bar{c}\rangle_\tau = \bar{c}_\infty$, which are shown in Fig. 6. It is evident that the equilibrium curves for the different fractions are intersected only for $a_{01} = a_{02}$ (the case of a monodisperse cluster). Thus, for the given initial gas concentration in the liquid, three scenarios are possible: 1) in both fractions, the bubbles dissolve (this occurs in a very small region for $a_0 < 1$ μm in both fractions); 2) bubbles of one fraction dissolve or grow, and bubbles of the other fraction tend to the size of the bubble radius in the equilibrium case; 3) the bubbles of both fractions tend to the same radius. This implies that with time the cluster becomes monodisperse or vanishes. This result agrees with the result of Kedrinskii [18], who found transition of a polydisperse bubble structure to a monodisperse structure in strong ultrasonic fields using a two-phase model of a cavitating liquid.

Conclusions. It was established that a bubble in a monodisperse cluster is more diffusively stable than a single bubble. In an unpurified liquid containing gas, a single bubble continuously grows (until collapse due to surface instability), whereas the bubble in a cluster tends to a definite radius corresponding to its equilibrium state. In addition, in a certain range of initial gas concentrations in the liquid, for a bubble in a cluster, unlike for a single bubble, there are two stable radii corresponding to the equilibrium state. The initial gas concentrations in the liquid are found at which a two-fraction cluster becomes monodisperse with time.

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