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Instability of a charged bubble with a gas soluble in the liquid

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The problem of stability of a charged gas bubble containing gas soluble in liquid is considered. The stability condition of the system „gas bubble—liquid"is obtained. The linear analysis is carried out, on the basis of which the influence of the effects of radial inertia, viscosity of liquid and the process of diffusion on the development of the instability process is studied.

Keywords: Instability, charged bubble, increment, linear analysis, carbon dioxide.

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Introduction

The study of bubble stability in a liquid is of interest not only from a theoretical, but also from a practical point of view, since it is associated with many technological applications, such as, for example, flotation, cavitation, heat exchange during boiling, underwater acoustics, bubbling, etc. [1].

The stability of bubbles with different gas phase compositions is studied in Ref. [2–5].

It was shown in Ref. [2] that bubbles of a liquid-soluble gas, which make small radially symmetric vibrations in an acoustic field, are unstable in amplitude. An expression is obtained for the increment characterizing the rate of instability development.

The diffusion stability of gas bubbles in a one- and two-fraction cluster when exposed to an acoustic field is studied in Ref. [3]. Ranges of values of initial gas concentrations in a liquid are numerically obtained in this study for a single-fraction cluster, at which the bubble tends to one of two equilibrium states due to diffusion processes occurring between it and the surrounding liquid. It is found that the two-fraction cluster tends to become single-fraction.

The stability of an superheated liquid containing insoluble gas nuclei is studied in Ref. [4]. Critical conditions for the mass of gas nuclei, their radii, and volume concentrations in the case of a stable state of the liquid—vapor-gas nuclei system have been determined. The theory of spontaneous solutions is constructed, describing the release of an superheated bubble vapor-gas-liquid system from an unstable state. The dynamics of the transition of an superheated liquid to a stable state has been studied based on such solutions.

Boiling of an superheated liquid containing a steam bubble (or a system of steam bubbles) is studied in Ref. [5]. It was found in this study that the state of a mixture of liquid and bubbles is unstable due to the action of capillary forces. Linear and nonlinear solutions are constructed that describe the system's exit from an unstable state, as well as the unlimited growth of a single bubble and the transition to a stable vapor-liquid state in the presence of volume-distributed bubbles in the initial state.

Gas bubbles play a negative role in the dielectric strength of insulating liquids. Depending on their shape and localized electric field, bubbles can significantly reduce the dielectric strength of the insulating liquid.

We would like to note the papers that study the stability of charged bubbles, [6–10].

A dispersion equation is derived in Ref. [6] for capillary movements in a viscous liquid surrounding a spherical bubble carrying a surface charge that can lead to instability of the interface. There are critical conditions for such instability. The problem is solved using the scalarization method in a spherical coordinate system.

The equilibrium states of a charged spherical bubble in a dielectric liquid are studied for stability with respect to virtual centrally symmetric changes in its volume based on the analysis of a nonlinear equation describing radial oscillations of such a bubble in the vicinity of singular points [7]. It is shown that of the two possible equilibrium states of the bubble, only one is stable. The boundaries of the ranges of values of physical parameters separating stable and unstable states are found. It turned out that the presence of an electric charge on the bubble leads to an expansion of the ranges of values of physical parameters in which there are equilibrium states of the bubble.

It is shown in Ref. [8] that experimentally detected nanoscale bubbles in an aqueous medium arise spontaneously due to the minimization of Gibbs energy, taking into account the electrostatic component, of a gas-liquid dispersed system. The increased gas pressure inside the nanobubble gradually equalizes (according to Henry's law) with the atmospheric pressure of air dissolved in water. The radius of the bubble decreases to some extent, and the bubble becomes stable.

The stability mechanism of charged bulk nanobubbles is described in Ref [9] based on theoretical analysis. The strong attraction of negative charges to the surface of the nanobubble leads to an accumulation of charge, as a result of which the energy of the electric field creates a local minimum for the free energy required for bubble formation, which leads to thermodynamic metastability of charged nanobubbles. The excess surface charges mechanically create a size-dependent force that balances the Laplace pressure and acts as a restoring force when the nanobubble thermodynamically deviates from its equilibrium state. Using this negative feedback mechanism, the stability of a nanobubble as a function of surface charge and gas supersaturation is discussed. The theoretical prediction was compared with experimental observations and a good agreement was found.

Theoretical concepts of the existence of stable gas bubbles in pure water and aqueous solutions of electrolytes in equilibrium with the external gas environment are developed in Ref. [10]. A theoretical model of ion adsorption on the water surface is proposed, and a quantitative description of the resulting double electric layer is given on its basis. These results also made it possible to conduct a thermodynamic description of the Babston structure in the „water–external gas medium" system. It is shown that the appearance of such a structure at certain values of temperature and concentration of dissolved impurity ions is a phase transition of the first kind. In this problem, the unique role of helium as an external gas medium has been established: in this case, the Babston structure does not occur at any initial ion concentrations, and the solubility of helium itself increases with increasing temperature. The mechanism of formation of experimentally observed Babston clusters is considered.

We would like to note the monographic paper [11]. It states that nanobubbles filled with air or various pure gases persist in water for several weeks and months. Nanoemulsions consisting of oil droplets in water are also surprisingly resistant to coagulation and can last up to several weeks, even if they are not coated with surfactants. A reverse system consisting of nanodrops of water in oil is also available for study and application. Nanoscale voids are formed when modeling water under strong tension and are stable throughout the entire simulation time. The stability of these nanoobjects is ultimately determined by the structure of their surfaces at the molecular level. However, thermodynamic theory can also provide some insight into this. Therefore, we consider spherical gas nanobubbles, immiscible liquid nanodrops, and nanocavities formed in water at negative pressure at the same level and conduct a unified thermodynamic analysis of these systems. In all cases, the mechanical equilibrium (local maximum or minimum of free energy) is expressed by the Laplace equation, and the thermodynamic stability (local minimum of free energy) follows from the dependence of surface tension on the radius. All of them would be unstable if their surface tension were constant. Data from the literature allow constructing numerical examples for cavities and gas nanobubbles. Spectroscopic data are provided to confirm that the structure of water at the interface of gas nanobubbles and water droplets in oil differs from their counterparts on a flat surface. It was believed that the observed durability of nanobubbles, in particular, violates the fundamental principles of diffusion and solubility. A close look at Laplace's equation and its derivation shows why this widely held view is incorrect.

Capillary vibrations and the stability of a charged bubble in a viscous incompressible dielectric liquid with respect to infinitesimal distortions of volume and shape are studied in Ref. [12]. It defines the regions of physical parameters at which the instability of centrally symmetric radial and axisymmetric surface movements of the bubble is observed. Analytical asymptotic expressions for attenuation decrements of axisymmetric capillary oscillations of a bubble in low and high viscosity approximations are obtained.

This paper considers the instability of a single charged bubble in carbon dioxide-saturated water when the bubble is in dynamic and thermal equilibrium in its initial state. The influence of radial inertia, fluid viscosity, and diffusion on bubble instability is analyzed.

1. Problem statement and basic equations

Let there be a gas bubble with a radius a_0 in a liquid at a temperature T_0 and a pressure p_0, on the surface of which a charge Q_0 is evenly distributed. We will assume that the liquid is a dielectric with a permeability ε. We will also assume that the „gas bubble–liquid" system is in dynamic and thermal equilibrium. We can write the following relation based on this assumption:

$$p_{g0} + p_{el0} = p_0 + \frac{2\sigma}{a_0}, \tag{1}$$

where p_{g0}, p_{el0} is the initial partial vapor pressure in the bubble and the pressure force of the electric field, σ is the coefficient of surface tension of the liquid.

The gas inside the bubbles will be considered soluble. The gas pressure will be considered homogeneous and obeying the Clapeyron–Mendeleev equation:

$$p_{g0} = \rho_g^0 R_g T_0,$$

where ρ_g^0 is the initial gas density, R_g is the reduced gas constant.

The lower indices (l) and (g) denote the parameters of the liquid and gas in the following description of the problem; the additional lower index corresponds to the initial state of equilibrium; the index (el) refers to the parameters indicating the effect of the electric field of charge on the surface of the bubble.

Let us consider the radially symmetric motion of the system in the vicinity of the equilibrium state defined by equation (1).

Let us study the dynamics of the „exit“ of an electrically charged gas bubble from equilibrium by presenting the basic equations describing the radial motions of a bubble in an incompressible liquid ($\rho_l^0 = \text{const}$ — liquid density).

The equation of pulsation motion of a bubble — the Rayleigh–Lamb equation has the form [13]:

$$\rho_l^0 \left(a\ddot{a} + \frac{3}{2}\dot{a}^2 + \frac{4\nu_l^{(\mu)}\dot{a}}{a} \right) = p_g + p_{el} - p_l - \frac{2\sigma}{a}. \quad (2)$$

Considering the hypothesis of homobaricity, it is possible to obtain an equation for the change in gas pressure in the following form [14]:

$$\frac{dp_g}{dt} = -3\frac{p_g}{a}\frac{da}{dt} + 3\frac{p_g}{a}\frac{\rho_l^0}{\rho_g^0} D_l \left(\frac{\partial g}{\partial r} \right)_a. \quad (3)$$

Parameters $\nu_l^{(\mu)}$, D_l and g — kinematic viscosity of the liquid, diffusion coefficient and mass concentration of gas in the liquid.

The lower index a in the equation (3) for the gas concentration gradient corresponds to the bubble boundary. The second term in (3) is responsible for the intensity of gas dissolution, which is limited by the process of gas diffusion from the liquid into the bubble near the interfacial surface.

Let us write down the diffusion equation in a liquid to determine the intensity of mass transfer [13]:

$$\frac{\partial g}{\partial t} + w_l \frac{\partial g}{\partial r} = D_l \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial g}{\partial r} \right), \quad (4)$$

where $w_l = \frac{a^2}{r^2}\dot{a}$ is the radial velocity of the liquid.

Boundary conditions for the diffusion equation (4) in a liquid have the form

$$g = g_a \text{ if } r = a \text{ and } g = g_0 \text{ if } r = \infty. \quad (5)$$

Here g_a is the concentration of gas on the surface of the bubble, which is related to the gas pressure according to Henry's law:

$$g_a = G p_g. \quad (6)$$

The initial pressure of the electric field forces of the bubble is taken in the following form [7]:

$$p_{el0} = \frac{kQ_0^2}{8\pi\varepsilon a_0^4}, \quad (7)$$

where $k = \frac{1}{4\pi\varepsilon_0}$ is the proportionality coefficient, $\varepsilon_0 = 8.85 \cdot 10^{-12}$ F/m is the electrical constant.

We assume that the law of conservation of charge on the bubble surface holds:

$$Q = Q_0. \quad (8)$$

Based on this assumption, we obtain the relationship between the current pressure and the initial pressure:

$$p_{el} = p_{el0}\left(\frac{a_0}{a}\right)^4. \quad (9)$$

2. Linear analysis

Let the pressure in the liquid p_l be constant and equal to the initial value p_0 ($p_l = p_0$). Let's consider small deviations of the bubble radius from the initial value, which will entail a change in the remaining parameters. Expressions for the radius, gas pressure, pressure of the electric field force of the charge on the surface of the bubble, density and concentration of the gas are represented as:

$$a = a_0 + a', \qquad p_g = p_{g0} + p_g', \qquad p_{el} = p_{el0} + p_{el}',$$

$$\rho_g = \rho_{g0} + \rho_g', \qquad g = g_0 + g',$$

where the parameters with strokes are small deviations of the parameters from the equilibrium state, which are values of the first order of smallness [15].

Linearizing the system of equations (2)–(9) near the equilibrium state (neglecting the values of the second order of smallness, for example, the product of parameters with strokes), we obtain

$$\rho_l^0 \left(a_0 \frac{\partial^2 a}{\partial t^2} + 4\frac{\nu_l^{(\mu)}}{a_0}\frac{\partial a}{\partial t} \right) = p_g + p_{el} + \frac{2\sigma}{a_0^2}a,$$

$$\frac{\partial a}{\partial t} = w, \quad (10)$$

$$\frac{\partial p_g}{\partial t} = -3\frac{p_{g0}}{a_0}\frac{\partial a}{\partial t} + 3\frac{p_{g0}}{a_0}\frac{\rho_l^0}{\rho_{g0}} D_l \left(\frac{\partial g}{\partial r} \right)_{a_0}, \quad (11)$$

$$\frac{\partial g}{\partial t} = D_l \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial g}{\partial r} \right)_{a_0}, \qquad (a_0 < r < \infty), \quad (12)$$

$$g = g_a \text{ if } r = a_0 \text{ and } g = 0 \text{ if } r = \infty, \quad (13)$$

$$g_a = G p_g, \quad (14)$$

$$p_{el} = -4p_{el0}\frac{a}{a_0}. \quad (15)$$

In the resulting equations and further, the stroke sign indicating parameter perturbations is omitted.

We will look for the solution of system (10)–(15) in the form

$$a = A_a e^{\lambda t}, \qquad p_g = A_p e^{\lambda t}, \qquad g = A_g(r) e^{\lambda t}. \quad (16)$$

It should be noted that with this type of solution, the inverse λ indicates the time it takes for the amplitude of the disturbances to increase by e times ($\tau = 1/\lambda$).

From the diffusion equation (12) based on (13), (14) for the amplitude of the gas concentration around the bubble, we have

$$A_g(r) = A_p G \frac{a_0}{r} \exp\left(Y\left(1 - \frac{r}{a_0}\right)\right),$$

$$Y = a_0 k = \sqrt{a_0^2 \lambda / D_l}. \tag{17}$$

From equation (15), the pressure amplitude of the electric field forces has the form

$$A_{el} = -4 \frac{p_{el0}}{a_0} A_a. \tag{18}$$

Using this solution based on equations (10), (11) from the condition for the existence of a nontrivial solution of the form (16), we obtain the equation for determining λ:

$$\psi(\lambda) = \rho_l^0 \lambda^2 a_0^2 + 4\rho_l^0 \lambda \nu_l^{(\mu)} + \\ + \frac{3p_{g0}Y^2}{Y^2 + 3Os(1+Y)} + 4p_{el0} - \frac{2\sigma}{a_0} = 0, \tag{19}$$

where Os is the Ostwald number, indicating the value of the volume of gas that can dissolve in a unit volume of liquid [16].

Equation (19) has a positive root λ if the condition is satisfied

$$p_{el0} < \frac{\sigma}{2a_0}, \tag{20}$$

therefore, the bubble is unstable when this condition is met.

If the pressure of the electric field forces of the charge does not satisfy the condition (20), then the bubble state is stable. In this case, the value of the root, called the increment, determines the rate of instability development at the initial linear stage. The existence of a positive root means that expressions of the form (16) represent spontaneous solutions [17] for which the initial state of equilibrium is reached at $t \to -\infty$.

If there is no charge on the bubble ($Q_0 = 0,\ p_{el0} = 0$) from the condition (1) it is possible to find the value of the bubble radius at given liquid pressure values p_0:

$$a_0^{(M)} = \frac{2\sigma}{p_{g0} - p_0}. \tag{21}$$

It follows from the condition of mechanical equilibrium (1) that if there is a charge on the bubble surface ($Q_0 > 0,\ p_{el0} > 0$), the radius a_0 is always less than the value determined by the expression (21) ($a_0 < a_0^{(M)}$). Let us determine the lower limit value of the bubble radius in the equilibrium state in the case when the bubble is unstable, i.e. the condition (20) is fulfilled. Obviously, for this radius, in addition to (1), the condition must be fulfilled

$$p_{el0} = \frac{\sigma}{2a_0}. \tag{22}$$

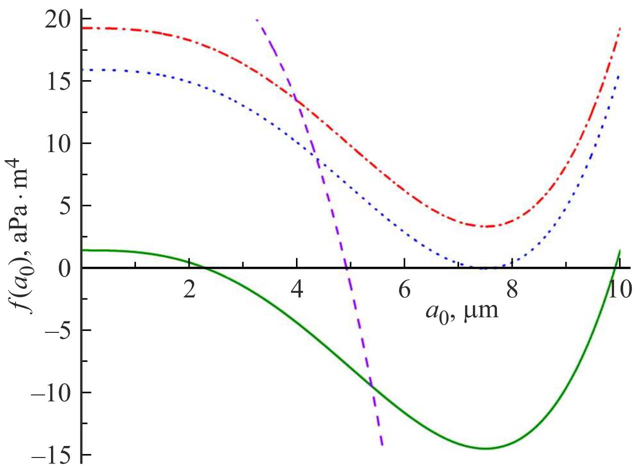

Figure 1. Dependence of the function $f(a_0)$ on the initial radius a_0, for different values of bubble charges: 1st case — dashed line, 2nd case — dotted, 3rd case — dotted, 4th case — solid.

Excluding from (1), (22) p_{el0}, we find the expression for the minimum radius $a_0^{(m)}$ in the form

$$a_0^{(m)} = \frac{3}{4} a_0^{(M)} = \frac{3\sigma}{2(p_{g0} - p_0)}. \tag{23}$$

It is possible to obtain an expression for the critical charge of a gas bubble using expressions (7), (23)

$$Q_{cr} = \sqrt{\frac{27\pi\varepsilon\sigma^4}{2k(p_{g0} - p_0)^3}}. \tag{24}$$

Substituting the expression (7) in the condition of mechanical equilibrium (1), we obtain the equation for determining the equilibrium radius a_0 for a given charge value p_{el0} of a gas bubble

$$f(a_0) = (p_{g0} - p_0)a_0^4 - 2\sigma a_0^3 + \frac{kQ_0^2}{8\pi\varepsilon} = 0. \tag{25}$$

Let's analyze the expression (25). The following cases are possible, which are shown in Fig. 1:

1) for $p_{g0} < p_0$, the equation has one positive root for any value Q_0;

2) for the case $p_{g0} > p_0$ and for $Q_0 > Q_{\text{cr}}$, there are no valid roots;

3) If $p_{g0} > p_0$ and for $Q_0 = Q_{\text{cr}}$, the equation has one real root;

4) and finally, if $p_{g0} > p_0$ for $Q_0 < Q_{\text{cr}}$, the equation has two real roots.

In addition, for $0 < Q_0 < Q_{\text{cr}}$, the equation has two positive roots $a_0^{(m)} < a_{01} < a_0^{(M)}$ and $0 < a_{02} < a_0^{(m)}$, and for a larger radius a_{01} the „bubble-liquid system" is unstable for the case of positive roots of the equation (19), and it is stable for a smaller radius a_{02} for the case of complex conjugate roots of equation (19).

We obtain the value $Q_{\text{cr}} \approx 2\,\text{pC}$ from formula (24) for the critical charge at parameters $\varepsilon = 87.9$,

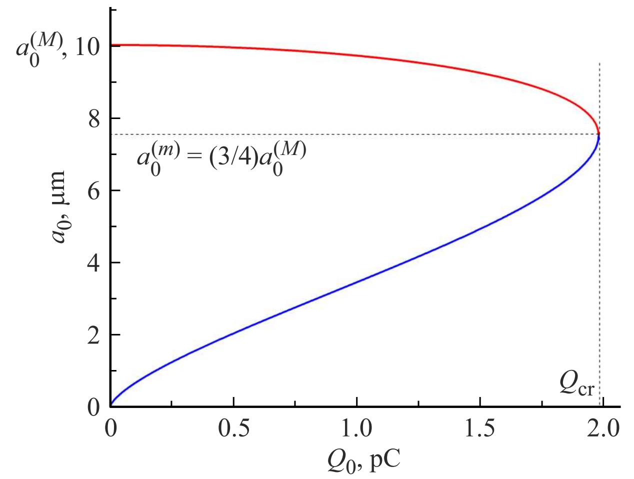

Figure 2. Dependence of the equilibrium radius on the charge of the bubble at $p_0 = 0.1$ MPa, $T_0 = 273$ K, $Os = 1.7$.

$\sigma = 75.64 \cdot 10^{-3}$ Pa · m, $a_0 = 10^{-5}$ m. In the case when the charge of the bubble is zero ($Q_0 = 0$), the maximum value of the equilibrium radius is $a_0^{(M)} = 10^{-5}$ m. All necessary values of the thermophysical parameters are taken from Ref. [18].

For a given charge value Q_0 , based on equation (25), it is possible to determine the values of the radii that correspond to the unstable and stable equilibrium state of the gas bubble. Fig. 2 shows the dependence of the equilibrium radii of a gas bubble on its charge in water saturated with carbon dioxide. The upper branch of the curve of the radius dependence on the charge corresponds to the actual values of the roots of equation (19); they also correspond to the unstable equilibrium state of the bubble. The lower branch of the curve corresponds to the complex conjugate roots of equation (19) corresponding to stable bubble states. These roots correspond to the decaying natural oscillations. The upper and lower branches are separated by a horizontal line $a_0 = a_0^{(m)}$.

Fig. 3 shows the dependence of the increment (of the positive root of equation (19)), corresponding to the upper branch of the dependence shown in Figure 2, on the charge of the gas bubble. In the left-hand side of equation (19), the first, second, and third terms take into account the influence of radial inertia, fluid viscosity, and gas diffusion on the development of instability at the linear stage, when the bubble radius is equal to the equilibrium value.

In the case when the development of instability is limited by radial inertia, i.e. in equation (24) the second and third terms related to viscosity and diffusion are omitted, for the increment value we have

$$\lambda^{(R)} = \sqrt{\frac{2\sigma/a_0 - 4p_{el0}}{\rho_l^0 a_0^2}}. \qquad (26)$$

Assuming that the increment value is determined by the viscosity of the liquid, we obtain

$$\lambda^{(\mu)} = \frac{\sigma/a_0 - 2p_{el0}}{2\rho_l^0 \nu_l^{(\mu)}}. \qquad (27)$$

We also give a formula for the increment, when the diffusion process is the determining factor in the development of instability

$$\lambda_D = \frac{D_l}{a_0^2}\left(\frac{A}{2} + \sqrt{\frac{A^2}{4} + A}\right)^2,$$

$$\left(A = 3Os\frac{\Sigma}{1-\Sigma},\ \Sigma = \frac{2\sigma/a_0 - 4p_{elo}}{3p_{g0}}\right). \qquad (28)$$

Dotted, dashed, and dotted lines in Fig. 3 are obtained by formulas (26), (27), and (28), respectively.

An analysis of Fig. 3 shows that the increment values determined by radial inertia, viscosity, and diffusion are independent of the charge value Q_0 up to Q_{cr}. At $Q_0 = Q_{cr}$, the increment value is $\lambda \to 0$ regardless of the factor affecting the bubble instability, hence $\tau \to \infty$. This means that it takes an infinite amount of time for a bubble with a charge Q_{cr} on its surface to grow. Under such conditions, the charged bubble is close to a stable state. The increment is most important for the case when the instability of the bubble is limited by viscosity (dashed line, Fig. 3). It should be noted that the increment determined by diffusion (dotted line) coincides with the general solution of equation(19) (solid line), i.e., the diffusion effect plays a major role in the development of instability for any values of the charge on the bubble up to Q_{cr}.

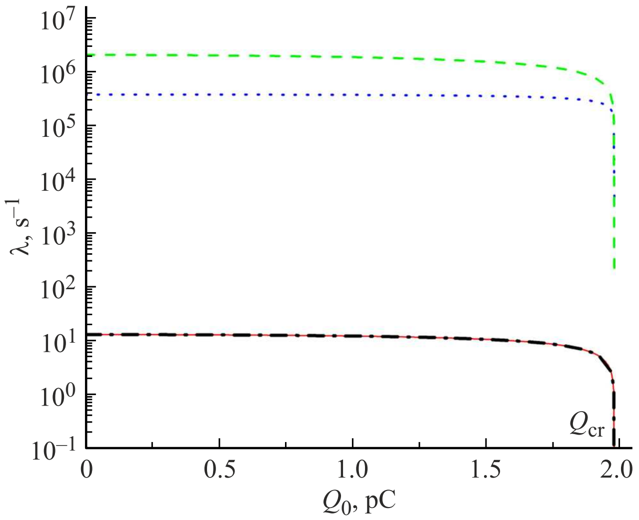

Figure 3. Dependence of increments of unstable equilibrium radii on the charge of the bubble at $\varepsilon = 87.9$, $\sigma = 75.64 \cdot 10^{-3}$ Pa·m, $a_0 = 10^{-5}$ m, $\rho = 1000$ m/s, $\nu = 1.787 \cdot 10^{-6}$ m^2/s, $D = 3.53 \cdot 10^{(-9)}$ m^2/s, the remaining parameters are the same as in Fig. 2: solid line — general solution of equation (19), dotted, dashed and dotted lines — solutions of equation (19) (dotted — taking into account radial inertia, dashed — viscosity, dashed — diffusion).

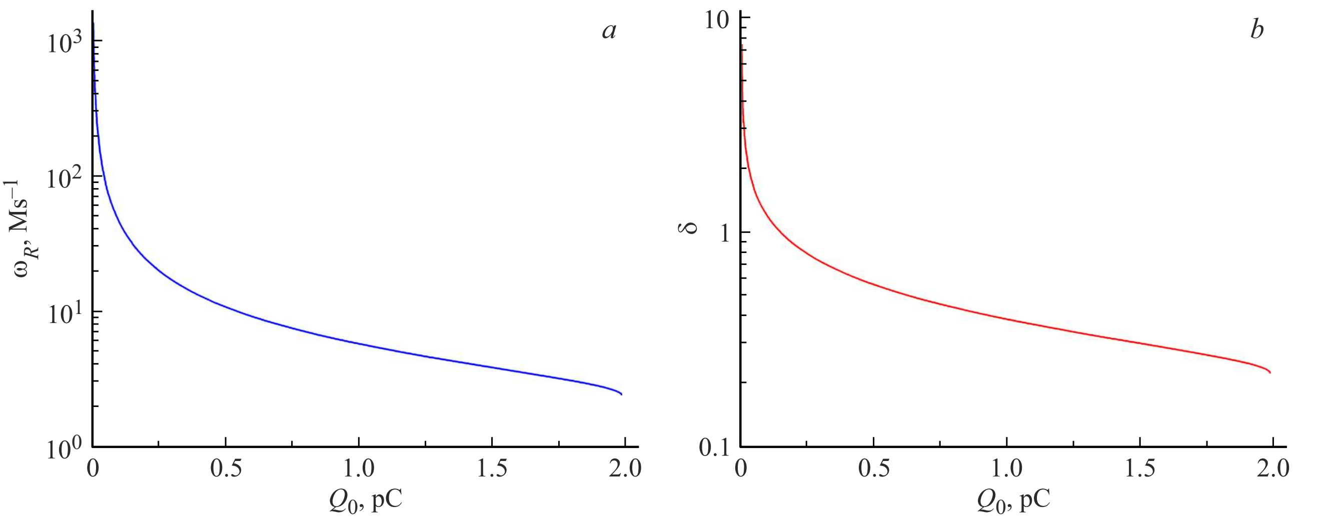

Figure 4. Dependences of the natural oscillation frequency (a) and the attenuation decrement (b) for stable radii on the charge on the bubbles. The calculation parameters are the same as in Fig. 3.

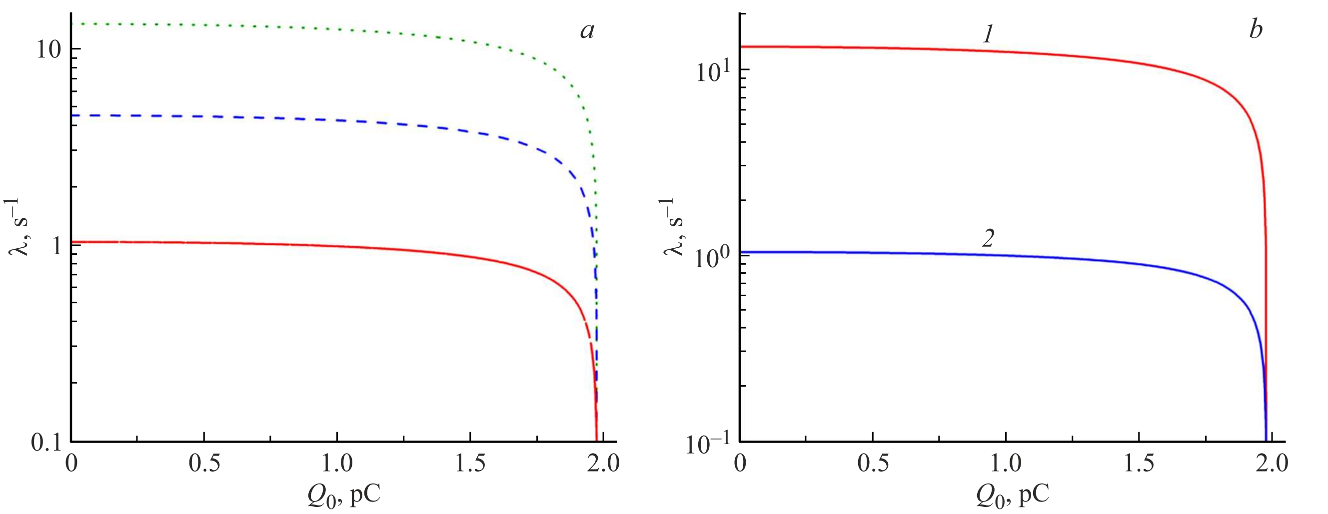

Figure 5. a — dependence of increments of unstable radii on the charge of the bubble. Gas — carbon dioxide, liquid — water at $p_0 = 0.1$ MPa for various values of Ostwald number and temperatures: solid line — $Os = 0.51$, $T_0 = 323$ K; dashed — $Os = 1$, $T_0 = 288$ K; point —$Os = 1.7$, $T_0 = 273$ K. b — dependence of increments of unstable radii on charge a bubble for carbon dioxide in water at $T_0 = 273$ K, $Os = 1.7$. Lines *1* and *2* correspond to the static liquid pressures $p_0 = 0.1$ and 1 MPa.

Fig.4 shows the dependences of the natural oscillation frequency $\omega_R = \mathrm{Im}(\lambda)$ and the logarithmic decay decrement $\delta = -2\pi\mathrm{Re}(\lambda)/\mathrm{Im}\lambda$ on the charge of the gas bubble. It follows from Fig. 4 that as the charge on the bubble surface increases, the natural oscillation frequency and attenuation decrement decrease.

Fig. 5, a shows the dependences of the increment that determines the rate of development of the output of a charged carbon dioxide bubble on the charge Q_0. Dotted, dotted, and solid lines correspond to the solution of equation (19). It can be seen that the higher the temperature of the liquid, the more stable the bubble is. It also follows from the graph that as the Ostwald number increases, the value of the critical charge increases. Fig. 5, b shows the dependences of the increment of unstable radii on the charge Q_0 at various static pressures. The lines *1* and *2* are obtained according to the solution of equation (10). The line *2* is located below the line *1*, this means that for the temperature $T_0 = 273$ K, the higher the static pressure of the liquid, the more stable the bubble is.

Conclusion

The paper shows that in water saturated with carbon dioxide, a single charged bubble can be in dynamic and thermal equilibrium if the charge of the bubble is less than a critical value.

Linear analysis of the instability of a single bubble in carbon dioxide-saturated water has shown that the instability is mainly limited by the effect of gas diffusion into the bubble.

When the bubble charge is less than the critical value, there are two values of the equilibrium radius. Moreover, an unstable state corresponds to a higher value of the radius, and a stable state corresponds to a lower value.

Conflict of interest

The authors declare that they have no conflict of interest.

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