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A MECHANISM OF FORMATION OF GAS-LIQUID SYSTEMS

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A mathematical model is constructed for the process of formation of a disperse liquid-gas bubble system as a result of evolution of dissolved gas due to changes in the external pressure. The model is implemented numerically for chlorine and air and is used to explain the difference in the rates of bubble growth at small and large excess pressures.

At present the sphere of both research and application of nonlinear wave processes in gas-liquid media is expanding rapidly owing to their highly nonlinear properties. Nonlinear and especially resonance hydroacoustic effects are most pronounced in monodisperse mixtures. However, creation of these mixtures with preset concentrations and sizes of bubbles is a complicated engineering problem. In the present work formation of a monodisperse mixture during evolution of a dissolved gas from a supersaturated solution produced by an instantaneous drop in the external pressure is considered. The goal of the study is determination of parameters of the mixture formed.

It is assumed that at the initial moment the continuous medium is a solution of gas in water saturated at an increased pressure. After the pressure drops to the normal value, a supersaturated solution is formed, from which the excess of the dissolved gas evolves relatively slowly on nuclei. It is assumed that the growth rate of bubbles is limited by diffusion of the supersaturated solution through depleted liquid layers adjacent to the bubble. Since the temperature of the gas entering the bubble is equal to that of the liquid, the process is considered in the isothermal approximation. Because the rate of bubble growth is low compared to the velocity of sound in the liquid, hydrodynamic effects are neglected.

At the initial moment the liquid is a solution of gas saturated at the pressure p_1. After the pressure drops to the normal value p_0, a supersaturated solution is formed with an excess concentration equal to

$$c_0 = K_s (p_1 - p_0) ,$$

where K_s is the solubility of this gas. Then, nuclei of future bubbles appear on microinclusions. Since the pressure of the gas in a bubble of radius R is

$$p = p_0 + \frac{2\sigma}{R} , \tag{1}$$

where σ is the coefficient of surface tension, the initial radius of a bubble is

$$R_{\text{in}} = \frac{2\sigma}{p_1 - p_0} . \tag{2}$$

Within the isothermal approximation the gas density ρ_g is

$$\rho_g = \frac{\rho_{0g}}{p_0} p , \tag{3}$$

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where ρ_{0g} is the gas density at normal pressure. The mass of the gas in a bubble is

$$m = \frac{4}{3}\pi R^3 \rho_g .$$

Differentiation of the above expression with respect to the time t gives

$$\frac{dm}{dt} = 4\pi R^3 \rho_g R + \frac{4}{3}\pi R^3 \frac{d\rho_g}{dp} p . \tag{4}$$

Substitution of expressions (1) and (3) into (4) gives

$$\frac{dm}{dt} = 4\pi \rho_{0g} R \left(R + \frac{4\sigma}{3p_0} \right) R . \tag{5}$$

On the other hand, according to Nernst's law [1], the mass flow of gas through the bubble surface is caused by diffusion and is equal to

$$\frac{dm}{dt} = 4\pi R^2 D \frac{\partial c}{\partial r}\Big|_R \rho_{0g} , \tag{6}$$

where D is the diffusion coefficient of the gas in the liquid; $\partial c/\partial r|_R$ is the concentration gradient on the bubble surface.

Describing the diffusion process in Euler coordinates, it is necessary to take into consideration that expansion of a bubble results in a divergent flow of the liquid. Since the growth rate of the bubbles is low, the kinematics of the liquid can be considered within the incompressibility model. In this case the velocity field in the liquid is

$$V = \frac{R^2 R}{r^2} . \tag{7}$$

In the general case, the diffusion equation in a medium with the velocity field $\mathbf{V}(r;\ t)$ has the form [2]

$$\frac{\partial c}{\partial t} = D\,\Delta c - \mathbf{V}\ \mathrm{grad}\ c - c\ \mathrm{div}\ \mathbf{V} . \tag{8}$$

Since $\mathrm{div}\ \mathbf{V} = 0$ for an incompressible liquid, Eq. (3) can be written for the case of spherical symmetry [3] and with account for (7) as

$$\frac{\partial c}{\partial t} = D \left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r} \right) - \frac{R^2 R}{r^2}\frac{\partial c}{\partial r} . \tag{9}$$

The initial and boundary conditions for Eq. (9) are evident:

$$c(r;\ 0) = c_0 ; \quad c(R;\ t) = 0 ; \quad \lim_{r \to \infty} c(r;\ t) = c_0 . \tag{10}$$

From relations (5) and (6) the equation of the growth of a bubble can be obtained:

$$\dot{R} = \frac{DR}{R + \frac{4\sigma}{3p_0}} \frac{\partial c}{\partial r}\Big|_R . \tag{11}$$

Thus, Eq. (9) with conditions (10) and Eq. (11) with initial condition (2) form a closed mathematical model of the growth of a gas bubble in a supersaturated solution. Numerical implementation of this model was carried out for

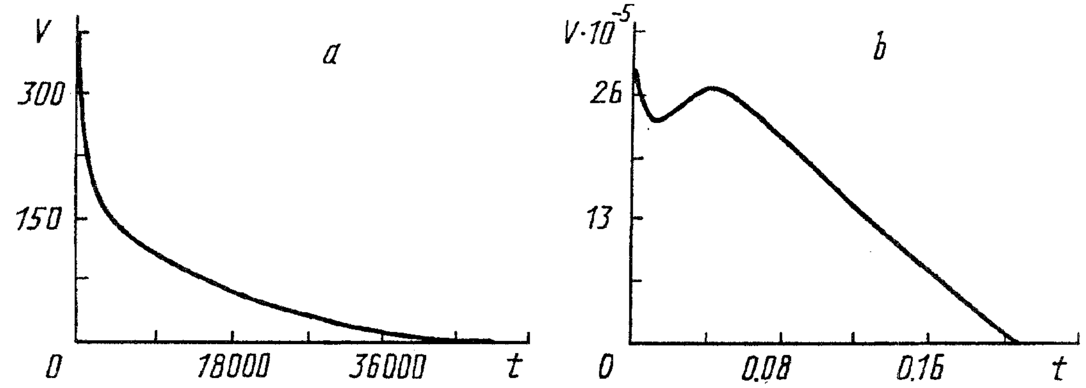

Fig. 1. Plots of the rate of chlorine bubble growth in a supersaturated solution at different excess pressures versus time: a) $\Delta p = 0.1 \cdot 10^5$; b) $50 \cdot 10^5$ Pa. V, μm/sec; t, μsec.

TABLE 1. Quantities Characterizing the Process of Chlorine Bubble Growth in a Supersaturated Solution

$\Delta p \cdot 10^{-5}$, Pa	R_{in}, μm	R_{f}, μm	V_{in}, μm/sec	$\tilde{t}$, μsec	ε	N, cm^{-3}	l, μm
0.1	14.7	16.5	360	45000	0.19	10^7	47
1	1.5	2.9	23000	440	0.7	$7 \cdot 10^9$	5
10	0.15	0.7	$5 \cdot 10^5$	9	0.96	$7 \cdot 10^{11}$	1.1
0.3350	0.03	0.2	$3 \cdot 10^6$	0.2	0.99	$3 \cdot 10^{13}$	0.33

chlorine and air using an explicit three-point scheme. Results of the calculation are presented in Tables 1 and 2. Here $\Delta p = p_1 - p_0$ is the initial excess pressure, at which the gas was dissolved in water; R_{in} is the initial radius of a bubble defined by Eq. (2), R_{f} is its final radius; $\tilde{t}$is the time of the process; ε is the concentration of the gas occurring in the liquid in the form of bubbles. In this case, because the bubbles are small and so is their rate of ascent to the surface, at the initial moment the decrease in the concentration of gas inclusions due to their emergence on the surface can be neglected, and then ε is defined by the relation

$$\varepsilon = c\,(p_{\text{in}}) - c\,(p_{\text{f}}),$$

where p_{in} and p_{f} are the initial and final pressures of water with dissolved gas in the general case.

Table 1 contains calculation results for chlorine, the solubility of which in water is very high (up to 2.3 volumes of chlorine can be dissolved in a unit volume of water).

Fist of all, it should be noted that the growth rates are always small. Even for an excess pressure of 5 MPa, the rate is only 3 m/sec at the initial moment. Thus, our assumption is correct and use of the model of an incompressible liquid is quite justified.

From the results obtained it is possible to assess the validity of neglecting the hydrodynamic effects. Describing the relation between the rate of bubble expansion and the hydrodynamic pressure on its surface by Rayleigh's equation

$$R\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{p_{\text{d}}}{\rho_{\text{liq}}}$$

and considering that $|R\ddot{R}| \ll \dot{R}^2$ and $R < 0$, we can write that $p_{\text{d}} \sim 3/2\rho_{\text{lig}}V^2$. In the most "unfavorable" case $\Delta p = 5$ MPa, $V = 3$ m/sec, the hydrodynamic pressure on a bubble is as low as 0.014 MPa.

It is of interest to plot the rate of bubble growth versus the time with different excess pressures (Fig. 1). Figure 1 shows that the change in the rates of bubble growth at small and large excess pressures is qualitatively

TABLE 2. Quantities Characterizing the Process of Air Bubble Growth in a Supersaturated Solution

$\Delta p \cdot 10^{-5}$, Pa	R_{in}, μm	R_{f}, μm	V_{in}, μm/sec	$\tilde{t}$, μm	ε	N, cm^{-3}	l, μm
0.1	14.6	14.65	3	450	0.002	$15 \cdot 10^4$	190
1	1.46	1.47	190	20	0.02	$15 \cdot 10^8$	9
10	0.14	0.16	4100	9	0.17	10^{13}	0.47
100	0.014	0.02	47000	0.1	0.67	$2 \cdot 10^{16}$	0.04

different. In the former case the rate decreases monotonically, and in the latter case the growth rate increases in one section. This difference can be explained as follows. One can see from Table 1 that at a small excess pressure, on the one hand, the initial bubble size is large and, on the other, the excess concentration of dissolved gas is relatively small. Therefore, from the physical standpoint it should be expected that the final bubble size differs but slightly from the initial size. Indeed, the growth rate of the bubbles decreases gradually due to deceleration of diffusion through the gas-depleted liquid layers.

On the other hand, at a high excess pressure the initial bubble radius is very small and the concentration of gas is high. The final radius will differ substantially from the initial one. Initially, in the manner described above, a bubble grows due to diffusion of gas from adjacent layers and the growth rate decreases. Then, it is likely that these layers are compressed by the rather rapidly growing bubble and become thinner, the concentration of gas in them increases, the rate of diffusion rises, and, consequently, the growth rate of the bubble increases. Finally, the size of the bubble becomes so large that the relative decrease in the thickness of the adjacent layers becomes small and the decrease in the rate of diffusion due to a decrease in the concentration of the gas in the water surrounding the bubble becomes dominant again. It should be noted that as the pressure increases further, this so-called "effect of compression of the layers" is increasingly important, increasing the growth rate almost to the initial value at a certain moment.

It is of interest to consider calculated data on the number of bubbles possible in a unit volume of the mixture, which is sufficiently large at both low and high pressures. As is known, the appearance of a bubble with a sufficiently large initial radius characteristic of small excess pressures requires the presence of a center in the liquid, a solid particle, as a rule. However, ordinary water does not contain a sufficient number of relatively large particles, and therefore in practice at a small excess pressure the gas evolution is not complete. The number of bubbles formed is much smaller than the number possible and the liquid remains a supersaturated solution of gas even after formation of bubbles.

Therefore, it is natural that stirring of the liquid or introduction of additional centers of gas formation (powder, dust) will result again in intense evolution of gas. This effect is well known for water saturated with CO_2, whose solubility, like that of chlorine, is very high. In this case large bubbles are formed only on relatively large suspended particles and they can entrain them when rising to the surface. This is the well known phenomenon of flotation.

Next, as is seen from Table 1, at a low excess pressure the final size of a bubble determined by diffusion differs very slightly from the initial size, i.e., bubble growth is quite insignificant. Precisely such is the case when the number of gas-formation centers in the liquid is sufficient, for example, in flotation. However, if the water is relatively pure and, as was mentioned above, the number of bubbles is much smaller than the number possible, further growth of the bubbles can be effected by the buoyancy process rather than by diffusion.

At a high excess pressure, for example, at $50 \cdot 10^5$ Pa, the number of bubbles per cm^3 is very high and equal to $3 \cdot 10^{13}$ (Table 1). However, in this case the initial radius of a bubble is as small as 0.03 μm, and therefore evolution of gas can take place not only on a solid particle as the center but also on a bacterium and even on a fluctuation of the liquid density. Consequently, in practice, better conditions for complete evolution of excess dissolved gas are created at a high pressure than at a low pressure. Moreover, the bubble is so small that its growth during ascent can be neglected and the final size of the bubble determined by diffusion is assumed to be the same as that given in Table 1.

It should be noted that at pressures $\Delta p \geq 10^5$ Pa the inequality $l < 2R_{cr}$ means that in this case the mixture is foam.

In Table 2 results are given for dissolved air. Just as in the case of chlorine, here at a small pressure the initial radius of the bubble is fairly large and growth caused by diffusion is insignificant. However, the maximum number of bubbles is as small as $15 \cdot 10^4$. This number of centers of gas formation can be present in relatively pure water, and therefore in this case virtually complete evolution of the dissolved gas can be affected by diffusion, and subsequent ascent does not affect substantially bubble growth. Thus, unlike the data for chlorine, the data of Table 2 are probably in better agreement with the actual parameters of the mixture at both high and low pressures.

The present results can be useful in creating gas-liquid mixtures with characteristics required for technological purposes.

NOTATION

c, gas concentration in the liquid; K_s, gas solubility in the liquid; l, average distance between the centers of bubbles; m, mass of gas in a bubble; N, number of bubbles per cm^3 of mixture; p, gas pressure in a bubble; p_d, hydrodynamic pressure on the bubble surface; p_1, p_0, initial increased and normal external pressures, respectively; $\Delta p = p_1 - p_0$, initial excess pressure; R, radius of a gas bubble; r, instantaneous radius; T, temperature; t, time; $\tilde{t}$, total time of the process; $V = \dot{R}$, growth rate of a bubble in the liquid; Δ, Laplacian; ε, concentration of gas contained in the liquid in the form of bubbles; ρ_g, gas density in a bubble; ρ_{liq}, liquid density. Subscripts "1" and "0" refer to values based on the pressures p_1 and p_0, respectively. Subscripts "in" and "f" refer to initial and final states of the bubble growth process.

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