

One of the main factors affecting the dynamics of homogeneous solution type pulse reactors is the formation of gas bubbles on the fission-fragment tracks [1, 2]. The behavior of the reactor depends very considerably on the size (10^{-5} cm) and growth rate of these bubbles [2], and it is, accordingly, a very important matter to study these properties. One convenient means of doing this lies in the acoustic method. The behavior of gas bubbles in the field of a sound wave has been studied in a large number of papers and reviews [3, 4]. In this paper we shall see the approximation of a sound wave of small amplitude to consider the dissipation of sound-wave energy in a gas bubble, at the same time allowing for inertia, surface tension, viscosity, heat transfer, and the diffusion of gas through the surface of the bubble.

As in the majority of the papers already indicated, we shall, in general, consider sound waves in which the wavelength is much greater than the dimensions of the bubble $\lambda \gg R$ (i.e., over a distance equal to the dimensions of the bubble, the density and pressure may be regarded as constant); we shall assume that the changes in gas pressure inside the bubble follow the changes in wall pressure almost immediately. This corresponds to the case in which the velocity of the bubble boundary is much smaller than the velocity of sound in the gas in question.

It was indicated earlier [5] that, in order to describe the concentration of the dissolved gas at the moving boundary of a spherical gas bubble on the approximation of a thin diffusion boundary layer ($l \ll R$), the following expression might be used:

$$c(t) - c_0 = \eta(t) - \left(\frac{D}{\pi}\right)^{1/2} \int_0^t \frac{R^2(x) \left(\frac{\partial c}{\partial r}\right) R(x)}{\left\{\int_x^t R^4(y) dy\right\}^{1/2}} dx, \quad (1)$$

this having been obtained in [6] for the temperature at a moving spherical boundary. Here c and c_0 are the concentrations of the gas dissolved in the liquid close to the side of the bubble and a long way from the latter, respectively, D is the diffusion coefficient of the gas in the liquid, R is the radius of the bubble, r is the radial coordinate in a spherical coordinate system, the center of the system coinciding with the center of the bubble, t is the time, and η represents the sources of dissolved gas in unit volume of the liquid.

The approximate thickness of the boundary diffusion layer over which the fall in the concentration of the dissolved gas takes place is determined by the relation $l \sim (Dt)^{1/2}$; this gives the condition for the frequencies of the sound waves falling on the bubble at which Eq. (1) is valid:

$$\omega \gg D/R^2. \quad (2)$$

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Regarding the gas in the bubble as ideal we may write its pressure

$$p_2 = F \rho_2 T_2, \quad (3)$$

where F is the universal gas constant referred to 1 g-mole of the particular, T_2 is the gas temperature in the bubble and ρ_2 is the gas density.

At the bubble boundary the concentration gradient of the gas dissolved in the liquid may be written [5]

$$\left(\frac{\partial c}{\partial r}\right)_R = \frac{1}{3D} R \dot{\rho}_2 + \frac{\rho_2}{D} \dot{R}. \quad (4)$$

The equation of motion for the spherical boundary of the bubble in the liquid takes the form [7, 8]

$$p_2 = p_1 + \frac{2\alpha}{R} + \frac{3}{2} \rho_1 \dot{R}^2 + \rho_1 R \ddot{R} + 4\rho_1 \nu \frac{\dot{R}}{R}. \quad (5)$$

Here p_1 is the pressure in the liquid at the boundary of the bubble; α is the surface tension; ν is the kinematic viscosity of the liquid and ρ_1 is its density.

The pressure in the liquid is determined by the sum of the equilibrium pressure p_{01} , the pressure of the monochromatic traveling sound wave $\Delta p_{01} \exp \{i(kr - \omega t)\}$, and the pressure of the scattered spherical wave having its center in the bubble $(\Delta p_s/r) \exp \{i(kr - \omega t)\}$ [9, 10]; the pressure at the bubble boundary may be written

$$p_1 = p_{01} + \Delta p_1 e^{-i\omega t},$$

where Δp_1 is determined by means of an approximation analogous to that used in [9]:

$$\Delta p_1 = \Delta p_{01} + \Delta p_s / R_0 + ik \Delta p_s.$$

Here R_0 is the equilibrium radius of the gas bubble.

Let us consider a bubble for which the radius averaged over the period of the sound wave remains constant under steady oscillation conditions. Physically, this corresponds to the case in which the dissolved gas concentrations (averaged over the period) at the actual surface of the bubble is equal to the concentration a long way from the latter.

When the bubble oscillates steadily in the field of the sound wave, the time dependence of its radius, pressure, and density, and the temperature of the surrounding liquid may be expressed in the form

$$f(t) = f_0 + \Delta f e^{-i\omega t}, \quad (6)$$

where $\Delta f \ll f$, while for the temperature we have $T_{01} = T_{02} = T_0$ (T_0 is the equilibrium temperature).

Using (3)-(5) with due allowance for (6) and neglecting quantities of higher orders of smallness than ΔR , Δp , ΔT , we obtain

$$\left(\frac{\partial c}{\partial r}\right)_R = -\frac{R_0}{3DFT_0} \left\{ p_{01} + \frac{2\alpha}{R_0} \right\} \dot{T}_2 + \frac{1}{DFT_0} \left\{ p_{01} + \frac{4\alpha}{3R_0} \right\} \dot{R} + \frac{R_0}{3DFT_0} \left\{ p_1 + \rho_{01} R_0 \ddot{R} + 4\rho_{01} \nu \frac{\dot{R}}{R_0} \right\}. \quad (7)$$

Taking account of (6) and confining attention to terms of the first order of smallness, let us express (5) in the form

$$p_2(t) = p_{01} + \Delta p_1 e^{-i\omega t} + \frac{2\alpha}{R_0} \left(1 - \frac{\Delta R}{R_0} e^{-i\omega t} \right) - \omega^2 \rho_{01} R_0 \Delta R e^{-i\omega t} - i4\omega \nu \rho_{01} \frac{\Delta \dot{R}}{R_0} e^{-i\omega t}. \quad (8)$$

For a stationary bubble, according to (5) $p_{01} + 2\alpha/R_0 = p_{02}$. Substituting the latter in (8) and using Henry's law for the solubility of the gas $c = Bp_2$ (B is a proportionality factor), we find

$$c(t) - c_0 = B \left\{ \Delta p_1 - \frac{2\alpha}{R_0^2} \Delta R - \omega^2 \rho_{01} R_0 \Delta R - i4\omega \nu \rho_{01} \frac{\Delta R}{R_0} \right\} e^{-i\omega t}. \quad (9)$$

Here $c_0 = Bp_{02}$.

The change in the density of the liquid under the influence of the sound wave is expressed in terms of the change in pressure [11] $\Delta \rho_1 = \Delta p / u_1^2$, where u_1 is the velocity of sound in the liquid. The amount of gas dissolved in 1 g of liquid a long way from the bubble c_∞ , remains constant in the field of the sound wave. The amount of gas dissolved in unit volume of liquid is $c_0 = c_\infty \rho_{01}$ where $c_0 + \Delta c(r) \exp(-i\omega t)$ is the concentration of dissolved gas in the equilibrium state; $\Delta c(r) = (c_\infty / u_1^2) \{ \Delta p_{01} + (\Delta p_s / r) \exp(ikr) \}$. The change in the concentration of dissolved gas under the influence of the sound wave may be formally expressed as the action due to the sources

$$\eta = \Delta c(r) \exp(-i\omega t). \quad (10)$$

Since the condition $l \ll R$ is satisfied by the thickness of the diffusion layer adjacent to the bubble over which the concentration of the dissolved gas falls, we may put

$$\Delta c = \frac{c_\infty}{u_1^2} \left(\Delta p_{01} + \frac{\Delta p_s}{R_0} + ik \Delta p_s \right) = c_\infty \Delta p_1 / u_1^2 \quad (11)$$

within this layer (without allowing for any change in concentration due to the diffusion of the gas).

Substituting (7), (10), and (11) into (1) with due allowance for (6) and confining attention to terms of the first order of smallness, for the difference $c(t) - c_0$ we obtain an equation in terms of the Fresnel integrals $S[(\omega t)^{1/2}]$, $C[(\omega t)^{1/2}]$, which, subject to the condition $(\omega t)^{1/2} \gg 1$, i.e., steady oscillation conditions, tends to a limit of 1/2. Using the expression so obtained for the difference $c(t) - c_0$ together with Eq. (9) we obtain

$$\begin{aligned} B \Delta p_1 - \frac{2\alpha B}{R_0^2} \Delta R - \omega^2 B \rho_{01} R_0 \Delta R - i4\omega B \nu \rho_{01} \frac{\Delta R}{R_0} = \\ = \Delta c + i \frac{(2\omega)^{1/2}}{6D^{1/2} F T_0} \left\{ -\frac{R_0}{T_0} \left[p_{01} + \frac{2\alpha}{R_0} \right] \Delta T_2 + R_0 \Delta p_1 - \right. \\ \left. - \omega^2 \rho_{01} R_0^2 \Delta R + 3p_{01} \Delta R + 4 \frac{\alpha}{R_0} \Delta R - i4\omega \rho_{01} \nu \Delta R \right\} (1 + i). \end{aligned} \quad (12)$$

If we use E to denote the internal energy of the gas in the bubble, for the amount of heat liberated in a bubble of volume V_2 we obtain [12]

$$\frac{dQ}{dt} = \rho_2 V_2 D_{V_2} \frac{dT_2}{dt} + T_2 \left(\frac{\partial p_2}{\partial T_2} \right)_V \frac{\partial V_2}{\partial t},$$

where C_{V_2} is the specific heat of the gas at constant volume. The amount of heat passing into the liquid through the surface of the bubble in unit time is

$$\frac{dQ}{dt} = 4\pi \lambda_1 R^2 \left(\frac{\partial T}{\partial r} \right)_R,$$

where λ_1 is the thermal conductivity of the liquid. Equating the two latter relationships with due allowance for the equations $V_2 = (4\pi/3)R^3$; $\dot{V}_2 = 4\pi R^2 \dot{R}$ and confining attention to terms

of the first order of smallness, we obtain

$$\left(\frac{\partial T}{\partial r}\right)_R = \frac{1}{3\lambda_1} \left\{ \rho_{02} C_{V2} \dot{T}_2 + 3T_0 \left(\frac{\partial p_2}{\partial T_2}\right)_V \dot{R} \right\}.$$

As in [13] we shall consider that the temperature at the bubble boundary equals the temperature inside the bubble T_2 . The temperature of the moving spherical bubble boundary is described by Eq. (1) [6, 13] on the assumption of a thin thermal boundary layer ($l \ll R$ thick), except that instead of the concentrations $c(t)$, c_0 and the concentration gradient $(\partial c/\partial r)$ we now have to put the temperature T_2 , T_0 , and the gradient $(\partial T/\partial r)_R$, respectively, while the diffusion coefficient D must be replaced by the thermal diffusivity χ_1 . We may derive a frequency limitation analogous to (2) for which the foregoing considerations are valid

$$\omega \gg \chi_1/R_0^2.$$

The change taking place in the temperature of the liquid under the influence of the sound wave, like the concentration of the dissolved gas, may be expressed as the action due to the heat sources, i.e., $\eta(t) = \Delta T_1 \exp(-i\omega t)$. In this case the temperature a long way from the bubble may be regarded as constant and equal to the equilibrium temperature T_0 . Since the condition $\lambda \gg R$ is satisfied by the sound-wave length over the thermal boundary layer of thickness $l \ll R$ in which the temperature drop takes place $\eta(t)$ may be regarded as a uniformly distributed quantity, depending solely on the time. Proceeding as in the determination of $c(t) - c_0$, we find

$$\Delta T_2 - \Delta T_1 = -\frac{(2\omega\chi_1)^{1/2}}{6\lambda_1} \left\{ \rho_{02} C_{V2} R_0 \Delta T_2 + 3T_0 \left(\frac{\partial p_2}{\partial T_2}\right)_V \Delta R \right\} (1-i),$$

for $(\omega l)^{1/2} \gg 1$, whence

$$\Delta T_2 = (\Gamma_1 + i\Gamma_2) \Delta T_1 - (\Lambda_1 - i\Lambda_2) \Delta R, \quad (13)$$

where

$$\begin{aligned} \Gamma_1 &= \frac{1}{8} \{ 36\lambda_1^2 + 6(2\omega\chi_1)^{1/2} \lambda_1 \rho_{02} C_{V2} R_0 \}; \\ \Gamma_2 &= \frac{6}{8} (2\omega\chi_1)^{1/2} \lambda_1 \rho_{02} C_{V2} R_0; \\ \Lambda_1 &= \frac{1}{8} \left\{ 18(2\omega\chi_1)^{1/2} \lambda_1 T_0 \left(\frac{\partial p_2}{\partial T_2}\right)_V + 12\omega\chi_1 \rho_{02} C_{V2} T_0 \left(\frac{\partial p_2}{\partial T_2}\right)_V R_0 \right\}; \\ \Lambda_2 &= \frac{18}{8} (2\omega\chi_1)^{1/2} \lambda_1 T_0 \left(\frac{\partial p_2}{\partial T_2}\right)_V; \\ \delta &= 36\lambda_1^2 + 12(2\omega\chi_1)^{1/2} \lambda_1 \rho_{02} C_{V2} R_0 + 4\omega\chi_1 \rho_{02}^2 C_{V2}^2 R_0^2. \end{aligned}$$

The quantity ΔT_1 may be expressed in terms of Δp_1 [11]: $\Delta T_1 = (T_0/C_{p1})(\partial v_1/\partial T_1)_p \Delta p$, where C_{p1} is the specific heat of the liquid at constant pressure, and v_1 is the specific volume of the liquid.

Substituting (13) into (12) with due allowance for (11), we obtain

$$\Delta R = (G - iH) \Delta p_1, \quad (14)$$

Here

$$\begin{aligned} G &= \frac{ag + bd}{g^2 + d^2}; \quad H = \frac{bg - ad}{g^2 + d^2}; \\ a &= 6D^{1/2} F T_0 R_0^2 \left(B - \frac{c_\infty}{a_1^2} \right) + (2\omega)^{1/2} \left\{ R_0^3 - \frac{R_0^2}{C_{p1}} \left(\frac{\partial v_1}{\partial T}\right)_p (\Gamma_1 + \Gamma_2) [p_{01} R_0 + 2\alpha] \right\}; \end{aligned}$$

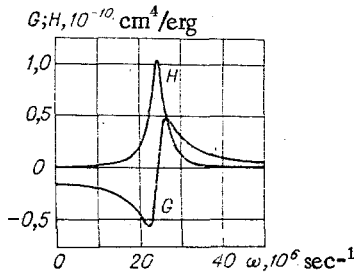


Fig. 1

Calculations show that the amplitude of the vibrations of the bubble undergoes resonance at a certain frequency. The greater the radius of the bubble, the narrower is the frequency range of the resonance.

By way of example, Fig. 1 shows the frequency dependence of G and H in the resonance region for a gas bubble (molecular hydrogen) with a radius of $R = 10^{-4}$ cm appearing in water at $p_{01} = 1$ atm and $T_0 = 293^\circ\text{K}$.

No explicit expression can be obtained for the resonance frequency ω_0 in view of the rather complicated relationships existing for G, H, Γ_1 , Γ_2 , Λ_1 , Λ_2 and δ in terms of the frequency ω of the external sound field and the radius of the bubble. However, the difference between the results obtained on the basis of such calculations and the well-known classical expression [9, 10]

$$\omega_0 = \frac{c_2}{R_0} \sqrt{\frac{3\rho_2}{\rho_1}}$$

is reasonably small and only becomes appreciable in the range $R \sim 10^{-5}$ cm. For $R = 10^{-3}$ cm the discrepancy is 8%, for $R = 10^{-4}$ cm, 13%, and for $R = 10^{-5}$ cm, 60%

In view of the foregoing restrictions regarding the wavelength and the thickness of the thermal and diffusion layers, the results obtained for the vibration amplitude of the bubble apply simply to cases in which the frequencies satisfy the inequalities $\omega \ll 2\pi u_1/R_0$; $\omega \gg D/R_0^2$; $\omega \gg \chi_1/R_0^2$. Since we always have $\chi_1 \gg D$, the strict condition for the frequency at which the equations in question are valid may be written

$$\chi_1/R_0^2 \ll \omega \ll 2\pi u_1/R_0. \quad (15)$$

For example, in the case of a hydrogen bubble with a radius of $R = 10^{-5}$ cm, we must satisfy $10^7 \ll \omega \ll 10^{11}$, while for $R = 10^{-3}$ cm $10^3 \ll \omega \ll 10^9$. However, calculations show that, in all practical cases, in the region of the lower limit of (15) the Γ_1 in (13) is close to unity, while the remaining terms are negligible. Thus even in the region of the lower boundary $\Delta T_2 \simeq \Delta T_1$, which corresponds to the isothermal approximation. Since the heat capacity of unit volume of liquid is much greater than that of unit volume of gas, while the temperature fluctuations in the bubble under the influence of a sound wave of small amplitude are negligible ($\Delta T_2 \ll T_0$), the equation $T_2 = T_1$ means (physically) that in the region of fairly low frequencies almost all the heat liberated in the bubble during its compression is absorbed in the boundary layer. This result may be predicted on the basis of the actual form of the expression for Γ_1 , Γ_2 , and Λ_1 , Λ_2 , since as the frequency diminishes Γ_1 tends to unity and Γ_2 to zero; the terms containing Λ_1 and Λ_2 , both diminish.

Thus in solving Eq. (12) in the frequency range between $\omega \sim \chi_1/R_0^2$ and $\omega \sim D/R_0^2$ we may use the approximation $\Delta T_2 = \Delta T_1$, and since this approximation is obtained automatically from Eq. (15) all the results will be valid for the frequencies

$$D/R_0^2 \ll \omega \ll 2\pi u_1/R_0$$

(for $R = 10^{-5}$ cm and $R = 10^{-3}$ cm we have $10^5 \ll \omega \ll 10^{11}$ and $10 \ll \omega \ll 10^9$, respectively).

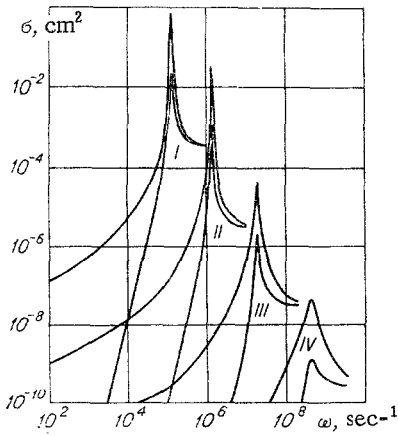


Fig. 2

The radial velocity component should remain continuous at $r=R$ [9, 10], from which [using (6) and (14) for $R(t)$ and expanding $\exp(ikr)$ in series in accordance with the condition $\lambda \gg R$] we obtain

$$\left(-\frac{A_s}{R_0^2} + ik\frac{A_s}{R_0}\right)(1 + ikR_0) = -i\omega(G - iH)\Delta p_1$$

to an accuracy of terms of the second order of smallness.

Defining Δp_1 in terms of $\Psi = \Psi_0 + \Psi_s$ and substituting this into the preceding relationship, we finally obtain

$$A_s = \omega^2 \rho_{01} R_0^2 \frac{G - iH}{-q + ih} A_0, \quad (16)$$

where

$$q = 1 + \omega^2 \rho_{01} R_0 G + \omega^2 k \rho_{01} R_0^2 H + k^2 R_0^2;$$

$$h = \omega^2 \rho_{01} R_0 H - \omega^2 k \rho_{01} R_0^2 G.$$

The flow of sound energy is reduced after striking the bubble, on account of both scattering and absorption. The total attenuation cross section may be determined in the following way [9]:

$$\sigma_t = E/I_0, \quad (17)$$

where I_0 is the intensity of the sound in the wave falling on the bubble, E is the average energy absorbed in the bubble per unit time, equal to the work done per unit time by the sound wave striking the bubble. This includes the energy both dissipated and absorbed in the bubble. The average work executed on the bubble in unit time when its volume alters is

$$E = -\frac{1}{2} \operatorname{Re} \left\{ \Delta p_{01}(t) \overline{\dot{V}_2} \right\}.$$

Defining $\Delta p_{01}(t) = -\rho_{01}(d\Psi_0/dt)$ and expressing \dot{V}_2 in terms of (14) we obtain

$$E = 2\pi R_0^3 \omega^3 \rho_{01}^2 |A_0|^2 \left\{ H + \omega^2 \rho_{01} R_0 \frac{h(G^2 - H^2) - 2qGH + kR_0q(G^2 - H^2) + 2kR_0hGH}{q^2 + h^2} \right\}.$$

Remembering that the velocity of the liquid in the incident wave is equal to $W_0 = \nabla\Psi_0$, we have

$$I_0 = \frac{1}{2} \rho_{01} u_1 |W_0|^2 = \frac{1}{2u_1} \rho_{01} \omega^2 |A_0|^2.$$

It should be noted that the terms allowing for gas diffusion in the equations for α , d , and g make an insignificant contribution at high frequencies, while the role of the terms allowing for inertial forces and viscosity increases with rising frequency. In the same way, a phase shift develops in the temperature fluctuations of the bubble, which also follows immediately from (13).

The temperature distribution inside a pulsating bubble was considered in [14] (the temperature on the surface of the bubble was assumed constant), and here also a phase shift was observed between the oscillations of the bubble boundary and the oscillations of the thermal flux through this boundary.

It is convenient to define the plane monochromatic traveling wave in terms of the potential $\Psi_0 = A_0 \exp\{i(kr - \omega t)\}$, and the scattered spherical wave correspondingly [10, 11] in terms of $\Psi_s = (A_s/r) \exp\{i(kr - \omega t)\}$. The excess pressure created by the sound wave in the liquid and the velocity of the liquid in the sound wave are described by the expressions $\Delta p(t) = -\rho_1(\partial\Psi/\partial t)$; $W = \nabla\Psi$.

Substituting the last two equations in (17) we find

$$\sigma_t = 4\pi R_0^2 \omega \rho_{01} u_1 \left\{ H + \omega^2 \rho_{01} R_0 \frac{(h + kR_0 q)(G^2 - H^2) - 2(q - kR_0 h)GH}{q^2 + h^2} \right\}.$$

The scattering cross section $\sigma_s = 4\pi \left| \frac{A_s}{A_0} \right|^2$ [9]. On using (16) we thus obtain

$$\sigma_s = 4\pi \omega^4 \rho_{01}^2 R_0^4 \frac{G^2 + H^2}{q^2 + h^2}.$$

The sound absorption cross section σ_α is determined by the difference $\sigma_t - \sigma_s$.

Using the foregoing equations, we calculated the attenuation and scattering cross sections for gas bubbles of different radii in water at $p_{01} = 1$ atm and $T_0 = 293^\circ\text{K}$. As the gas component we took molecular hydrogen. The results are presented in Fig. 2. The pairs of curves I-IV correspond in sequential order to bubble radii of $10^{-2} - 10^{-5}$ cm. The upper curve in each pair reproduces the total attenuation cross section, and the lower curve represents the scattering component. We see from Fig. 2 that the cross sections have a sharp peak. The width of the peak in the cross sections, and also the width of the resonance in the vibrations of the bubble, increase as the bubble becomes smaller.

The existence of peaks in the scattering and absorption cross sections offers extensive possibilities for the experimental analysis of the dynamics of gas bubbles, and, in particular, the mechanism underlying the boiling of homogeneous water pulse reactors, since it enables bubbles of a specific diameter to be separated out by reference to the sound-absorption maximum, so that their behavior may be studied as a function of time.

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