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VIBRATIONS OF A LIQUID MICROSPHERICAL SHELL FILLED WITH GAS

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The characteristic vibrations of a liquid spherical microshell are investigated with account for the acoustic vibrations of the gas filling the interior of the shell. It is shown that the presence of the gas influences strongly the vibrational spectrum of the shell.

In recent years, considerable interest has been shown in the study of the thermohydrodynamics of the formation and propagation of spherical hollow shells (microcapsules) having submillimeter sizes [1–4]. This is explained by the fact that interesting effects arise in the process of formation and propagation of such microcapsules in different media and in external fields and by the possible fields of their application. We should note some important cases where microcapsules are used: glass, polymer, and metal microsphere-capsules as targets for controlled thermonuclear fusion [5, 6]; special composite materials with preassigned unique properties [3]; microcapsules for pharmacological preparations [4]; systems of microencapsulation of liquid fuels, in particular, hydrogen [7].

At the present time, the most efficient method of production of hollow submillimeter microcapsules is the method of forced capillary breakdown of hollow jets [1], although there are also other nontrivial methods of production of microspheres with submillimeter sizes [8]. Among the main properties of the produced spherical liquid shell (that can subsequently be transformed a solid shell with a filler) are high degrees of sphericity and uniformity of the shell thickness. At the same time, in the process of formation of liquid shells from hollow jets, liquid microspheres experience vibrations that can have a fairly large amplitude. In consequence of this, in the case of hardening or oxidation (where the microsphere is found in an oxidizing atmosphere and is capable of being oxidized) microspherical shells of irregular shape are formed. It is generally believed that to obtain high-quality microspheres, it is necessary to produce them in weightlessness or in reduced gravity. Much research is devoted to these problems [9–11]. Nevertheless, the authors of the above works did not take into account the possible influence of the gas that is found inside a microspherical shell. Even though a number of technologies make it possible to obtain hollow microspheres inside which there is no gas, for example, [3, 4], this, nonetheless, is quite unnecessary, and it is simpler to produce microspheres filled with gas. It is shown in the present work that vibrations of spherical shells filled with gas are attended by a number of important effects that can be used directly in technologies of microencapsulation.

As has been indicated above, at present, capillary breakdown of hollow jets seems the most promising method for production of liquid hollow microspheres filled with gas. Traditionally, in such systems, the liquid arrives through a special-purpose cylindrical coaxial spinneret, forming a coaxial jet that contains a gas under pressure (the gas arrives through a special hole in the central part of the spinneret). Thus, a coaxial jet containing a gas is formed. The problems of capillary instability of such coaxial jets (or of compound jets, as they are sometimes called) and the dynamics of hollow droplets have been the object of a fairly large number of works (see, for example, [1, 3, 8–13]). At the same time, the influence of the compressible gas inside the

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produced hollow drops on their parameters, in particular, on the acoustic properties of such liquid microspheres, has not been systematically investigated so far. It is shown below that such liquid microspheres posses unique acoustic properties.

The technologies of microencapsulation considered make it possible to produce hollow microspheres with characteristic dimensions of less than 2 mm. Therefore, in the capillary breakdown of this a jet the surface-tension forces on both the interior and exterior surfaces of the jet are of great importance.

When droplets containing a cavity filled with gas break away, at the site of breakage of the neck a local hydrodynamic shock arises. This action initiates vibrations of the formed microsphere. In the presence of the compressible gas inside the microcapsule and outside it, the spectrum of these vibrations differs from the spectrum of capillary vibrations of a continuous liquid droplet [14] due to the presence of the compressible medium inside it.

The present work seeks to determine the spectrum of vibrations of a liquid microcapsule containing a cavity filled with gas.

Formulation of the Problem. We will assume that the vibrations of the interior and exterior surfaces are related and have a small amplitude such that a linear approximation can be used to determine the frequency of vibrations of the system. Assuming that the liquid is ideal, we restrict our consideration to possible potential motions in it.

Introducing the velocity potentials in the liquid and gas phases (Φ_{liq}, Φ_1, Φ_2) and assuming that the liquid is incompressible, we write the system of equations for the velocity potentials in each phase in a spherical coordinate system

$$\frac{\partial^2 \Phi_i}{\partial t} = \frac{1}{c^2}\Delta\Phi_i\,, \quad i = 1, 2\,, \tag{1}$$

$$\Delta\Phi_{\text{liq}} = 0\,, \tag{2}$$

where

$$\Delta = \frac{1}{r^2}\frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r} + \frac{1}{r^2 \sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) + \frac{1}{r^2\sin^2\theta}\frac{\partial^2}{\partial\varphi^2}$$

is the Laplacian operator.

We must set the conjugation conditions for the free surfaces. On the surface separating the interior cavity filled with gas (I) from the liquid shell (II) ($r = R_1$) we have:

a) the kinematic condition

$$\frac{\partial\xi_1}{\partial t} = \frac{\partial\Phi_1}{\partial r}\,, \tag{3}$$

where $\xi_1 = \xi_1(t, \theta, \varphi)$ is the surface deformation; and the running value of the outside radius is determined as $R_1(t, \theta, \varphi) = R_1 + \xi_1(t, \theta, \varphi)$; $\Phi_1 = \Phi_1(t, R_1, \theta, \varphi)$;

b) the dynamic condition

$$\rho_1\frac{\partial\Phi_1}{\partial t} - \frac{\sigma}{R_1^2}\left\{2\xi_1 + \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\sin\theta\frac{\partial\xi_1}{\partial\theta} + \frac{1}{\sin^2\theta}\frac{\partial^2\xi_1}{\partial\varphi^2}\right\} = \rho_{\text{liq}}\frac{\partial\Phi_{\text{liq}}}{\partial t}\,; \tag{4}$$

c) the condition of continuity of the normal component of the velocity

$$\frac{\partial \Phi_1}{\partial r} = \frac{\partial \Phi_{\text{liq}}}{\partial r} . \tag{5}$$

On the surface separating the liquid shell (II) from the outer space filled with gas (III) ($r = R_2$) we have:

a) the kinematic condition

$$\frac{\partial \xi_2}{\partial t} = \frac{\partial \Phi_{\text{liq}}}{\partial r} , \tag{6}$$

where $\xi_2 = \xi_2(t, \theta, \varphi)$, and the running value of the outside radius is determined as $R_2(t, \theta, \varphi) = R_2 + \xi_2(t, \theta, \varphi)$, $\Phi_{\text{liq}} = \Phi_{\text{liq}}(t, R_2, \theta, \varphi)$;

b) the dynamic condition

$$\rho_{\text{liq}} \frac{\partial \Phi_{\text{liq}}}{\partial t} - \frac{\sigma}{R_2^2} \left\{ 2\xi_2 + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \frac{\partial \xi_2}{\partial \theta} + \frac{1}{\sin^2 \theta} \frac{\partial^2 \xi_2}{\partial \varphi^2} \right\} = \rho_2 \frac{\partial \Phi_2}{\partial t} ,$$

$$\Phi_2 = \Phi_2 \, (t, R_2, \theta, \varphi) \, ; \tag{7}$$

c) the condition of continuity of the normal component of the velocity

$$\frac{\partial \Phi_{\text{liq}}}{\partial r} = \frac{\partial \Phi_2}{\partial r} . \tag{8}$$

It is evident that in the internal gas region (I), for $r \to 0$, the condition of boundedness of the solution in the form $\partial \Phi_1 / \partial r = 0$ should be fulfilled, and in the external gas region (III), for $r \to \infty$, we will assume that the vibrations of the sphere are responsible only for diverging spherical waves.

Solution of the Problem. It is known [15] that Eq. (1) has bounded solutions in the form of standing waves

$$\Phi_1 \, (t, r, \theta, \varphi) = a \exp \, (- i\omega t) \sqrt{\frac{2\pi k}{r}} \, J_{l+1/2} \, (kr) \, P_l^m \, (\cos \theta) \exp \, (- im\varphi) , \tag{9}$$

where $k = \omega / c$, $J_{l+1/2}$ is a Bessel function of the first kind of order $l + 1/2$ and P_l^m is a Legendre polynomial.

In region III, the diverging waves correspond to the following solution of Eq. (1):

$$\Phi_2 \, (t, r, \theta, \varphi) = p \exp \, (- i\omega t) \sqrt{\frac{\pi k}{2r}} \, H_{l+1/2}^{(1)} \, (kr) \, P_l^m \, (\cos \theta) \exp \, (- im\varphi) , \tag{10}$$

where p is the wave amplitude and $H_{l+1/2}^{(1)}$ is a Hankel function of the first kind of order $l + 1/2$.

Equation (2) has the solution

$$\Phi_{\text{liq}} \, (t, r, \theta, \varphi) = \exp \, (- i\omega t) \, (qr^l + dr^{-l-1}) \, P_l^m \, (\cos \theta) \exp \, (- im\varphi) , \tag{11}$$

where q and d are constants.

We assume that the deformations of the two surfaces can be represented in the form

$$\xi_1 \, (t, \theta, \varphi) = b \exp \, (- i\omega t) \, P_l^m \, (\cos \theta) \exp \, (- im\varphi) , \tag{12}$$

$$\xi_2 (t, \theta, \varphi) = g \exp(-i\omega t) P_l^m (\cos\theta) \exp(-im\varphi) , \tag{13}$$

where b and g are the amplitudes of the surface deformation.

Substituting the expressions for the sought solution (9)–(13) into the boundary conditions (3)–(8), we obtain a system of equations for the amplitudes a, b, q, d, p, and g. The condition of its solvability is the equality to zero of the determinant of the matrix of coefficients, which represents a dispersion relation of the form

$$A_l (R_1, R_2, k)\, \omega^4 + \frac{\sigma}{\rho_{\text{liq}}} (1 - l)(1 + l)\, B_l (R_1, R_2, k)\, \omega^2 +$$

$$+ C (R_1, R_2) \frac{\sigma^2}{\rho_{\text{liq}}} l (1 + l)(1 - l)^2 (2 + l)^2 = 0 , \tag{14}$$

where $k = \omega/c$. The coefficients in formula (14) can be represented as follows:

$$A_l (R_1, R_2, k) = \frac{R_2^{2l+1}}{R_1^{2l+1}} \left[1 + \frac{\rho_1 \zeta_l (kR_1)(l + 1)}{\rho_{\text{liq}}} \right] \left[1 - \frac{\rho_2 l \mu_l (kR_2)}{\rho_{\text{liq}}} \right] -$$

$$- \left[1 - \frac{\rho_1 l \zeta_l (kR_1)}{\rho_{\text{liq}}} \right] \left[1 + \frac{\rho_2 \mu_l (kR_2)(l + 1)}{\rho_{\text{liq}}} \right],$$

$$B_l (R_1, R_2, k) = \frac{R_2^{2l+1}}{R_1^{2l+1}} \left\{ (l + 1) \frac{\rho_{\text{liq}} - \rho_2 l \mu_l (kR_2)}{\rho_{\text{liq}} R_1^3} + l \frac{\rho_{\text{liq}} + \rho_1 \zeta_l (kR_1)(l + 1)}{\rho_{\text{liq}} R_2^3} \right\} +$$

$$+ l \frac{\rho_{\text{liq}} + \rho_2 \mu_l (kR_2)(l + 1)}{\rho_{\text{liq}} R_1^3} + (l + 1) \frac{\rho_{\text{liq}} - \rho_1 l \zeta_l (kR_1)}{\rho_{\text{liq}} R_2^3} ,$$

$$C (R_1, R_2) = \frac{1}{R_2^3 R_1^3} \left(\frac{R_2^{2l+1}}{R_1^{2l+1}} - 1 \right),$$

here

$$\zeta_l (kR_1) = \frac{j_l (kR_1)}{l j_l (kR_1) - kR_1 j_{l+1} (kR_1)} ,$$

where $j_l(kR_1)$ are spherical Bessel functions [16] and

$$\mu_l (kR_2) = \frac{H^{(1)}_{l+1/2} (kR_2)}{l H^{(1)}_{l+1/2} (kR_2) - kR_2 H^{(1)}_{l+3/2} (kR_2)} .$$

Analysis of the Results Obtained. Relation (14) represents an equation for the angular frequency of vibrations ω, the solution of which gives an idea of the vibrational spectrum of a sphere filled with gas. Analysis of the dispersion relation shows that when there is no gas inside the sphere ($\rho_1 = 0$), the zero har-

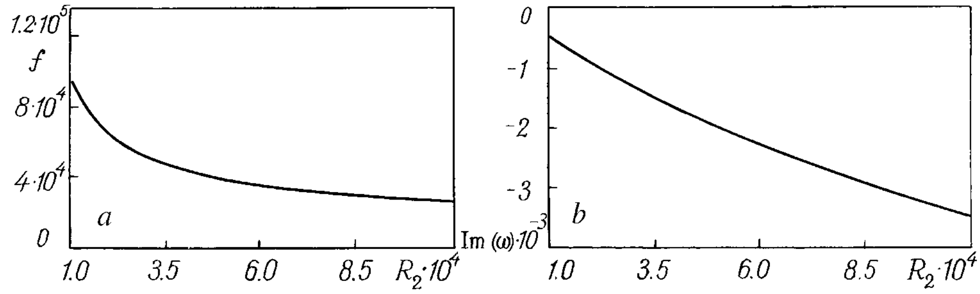

Fig. 1. Dependence of the frequency (a) and the decrement of damping (b) of vibrations of a shell on the outside radius for δ = 10 μm and l = 0. f, Hz; Im (ω), $\sec^{-1}$; R_2, m.

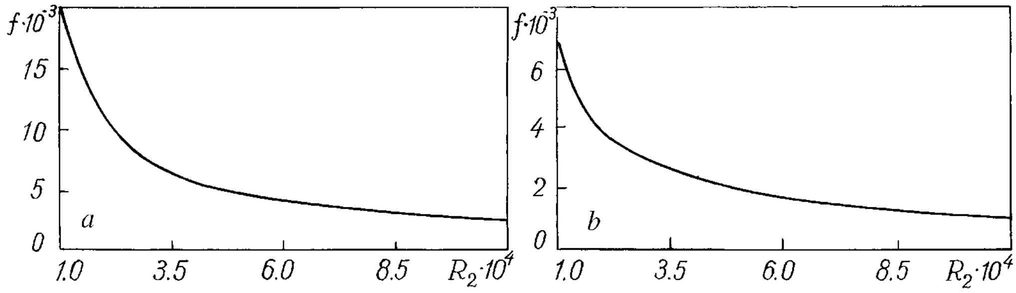

Fig. 2. Dependence of the frequency of vibrations of a shell of δ = 10 μm (a) and δ = 100 μm (b) on its radius for l = 2.

monic (l = 0) is unstable, i.e., a motion of the liquid that leads to the collapse of the hollow spherical cavity arises.

As an example, Fig. 1 shows results of calculating the spherical deformations of the zero harmonic (l = 0) for water shells 10 μm thick in the presence of air at atmospheric pressure.

The calculation results show that as the dimensions of the sphere increase, the frequency of its vibrations decreases and the rate of their damping increases, which is evidenced by a decrease in the imaginary part of the solution of the dispersion relation. An increase in the thickness of the shell causes a slight decrease in the frequency of the vibrations and a marked decrease in the rate of their damping. For example, for shells with an inside radius of 200 μm and a thickness δ = 100 μm the vibration frequency is $f \approx 22$ kHz and the time of vibration damping is $\tau \approx 0.16$ sec.

If after breakdown of the jet this shell moves with a velocity of ≈ 3 m/sec, it continues to vibrate and emit acoustic waves along a flight length equal to ~0.5 m. If a monodisperse regime of breakaway of droplet-sis selected, the system of shells that move successively one after another can serve as an audio-signal generator with a strictly determined frequency.

Let us compare damping of vibrations due to emission of acoustic waves into the surrounding space with viscous vibration damping. The decrement for the latter can be determined from the relation [15]

$$\gamma = \frac{\frac{\eta}{2}\int\limits_V \left(\frac{\partial v_i}{\partial x_k} + \frac{\partial v_k}{\partial x_i}\right)^2 dV}{\rho \int\limits_V v^2 \, dV}. \tag{15}$$

Substituting the solution of the dispersion relation into (15) and using (11), we obtain an estimate for the decrement of vibrations. The calculations show that the decrement of viscous vibrations for the harmonic with l = 0 is equal to zero, because tangential stresses are absent in the case of radial vibrations.

Figure 2 shows results of calculating the solution of the dispersion relation for the second harmonic. The calculation results show that for the second harmonic the vibration damping is very weak (Im (ω) ≈ -9 at $\delta = 10$ μm and Im (ω) ≈ -0.18 at $\delta = 100$ μm). Estimation of the decrement of viscous damping of vibrations (15) shows that for the second harmonic it is larger than the decrease in the vibration amplitude due to emission of sound. The decrement of vibration damping decreases with increase in the radius of the shell and lies within the range 2–25 sec^{-1}. Moreover, with increase in the thickness of the shell, the frequency of its vibrations decreases fairly rapidly to a value corresponding to the frequency of vibrations of a bubble in a large volume of liquid. With increase in the radius of the sphere and a constant thickness of the shell the vibration frequency also decreases.

The investigations performed in the present work show that the presence of a gas inside microspheres formed in forced capillary breakdown of hollow jets allows them to generate weakly damped acoustic waves (within time intervals of interest from the of technology viewpoint). On the one hand, this makes it possible to design an audio-signal generator with a definite frequency, and on the other, it suggests the possibility of the existence of collective effects of interaction of liquid microspheres with gas through acoustic waves. Specific effects of generation of sound were observed earlier in analysis of capillary breakdown of hollow jets [4, 12, 14]; however, they escaped the attention of the researchers. It follows from the results of the present investigation that there is a need to investigate the acoustic effects that arise in the production and propagation of spherical liquid shells formed by forced capillary breakdown of hollow jets.

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NOTATION

Φ_1, Φ_2, and Φ_{liq}, velocity potentials in the internal and external gas regions (I, III) and in the liquid, respectively; R_1 and R_2, inside and outside radii of the hollow liquid shell, respectively; ρ_{liq}, ρ_1, and ρ_2, densities of the liquid and of the gas inside the shell and outside it, respectively; σ, coefficient of surface tension; ξ_1 and ξ_2, surface deformations of the external and internal boundaries of the liquid shell; ω, angular frequency of vibrations; k, wave number; c, velocity of sound; l, number of the harmonic; a, wave amplitude; δ, thickness of the shell; f, vibration frequency; τ, time of vibration damping; r, radial coordinate; θ and φ, angles in the spherical coordinate system; γ, decrement of the viscous damping of vibrations; η, coefficient of dynamic viscosity; v_i, v_k, and v, i-th and k-th components of the velocity and velocity modulus, respectively; V, volume over which the integration is performed. Subscripts: liq, liquid; 1, gas inside the liquid shell; 2, gas outside the shell.

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