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EQUATIONS OF DYNAMICS OF A MIXTURE COMPOSED OF A GAS AND HOLLOW SELECTIVE-PERMEABLE MICROSPHERES

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Based on the laws of conservation of mass, momentum, and energy, equations of dynamics of multiphase systems, which are gas mixtures with hollow microspheres with selectively permeable shells, are obtained under the assumption of quasi-steadiness of the process of filling the microspheres by the gas. Acoustic characteristics of the system composed of a uniform gas and hollow permeable microspheres are studied using a simplified (*one-velocity and one-temperature*) *model. The frequency dependences of velocity and damping coefficient of sound are determined with regard for gas density* (*pressure*) *relaxation inside the microspheres.*

Introduction. Flows of two-phase mixtures of a gas and solid particles are frequently encountered in nature and engineering, and this is reflected in numerous publications (for example, see the review in [1]). These systems also include gas mixtures with disperse hollow selective-permeable (with a shell of a membrane material [2, 3]) microspheres [4, 5]. The suspended particles, which are hollow spheres 10–1000 μm in diameter with a shell thickness of 0.5–10 μm, are made of various glasses, corundum, plastics, organic substances, and other materials [4, 5]. Glass or ceramic hollow microparticles may be formed as industrial wastes of burning of some coal brands [6, 7]. Microspheres (which are also called microballoons or microcapsules) are used as targets in experiments on laser nuclear fusion [8], fillers in obtaining light and high-strength composite materials [4], and microvessels for storage of a hydrogen fuel and its injection into combustion chambers of engines [9]. Microspheres are used in medicine for highly efficient transportation of medications to certain types of tissues [5] and in applied acoustics as an effective method of noise reduction [10] (by microspheres with a perforated shell). A method was proposed to separate gas mixtures by means of microspheres with selectively permeable (membrane) shells, which are transported in the form of a suspension in the separated mixture along the pipeline [11, 12].

It should be noted that the problems of mathematical simulation of such complicated media have not been adequately considered in the literature. To understand the processes in the mixture and perform calculations, it is necessary to formulate a mathematical model that takes into account gas-dynamic and kinetic phenomena. Equations of dynamics of the above mixtures are derived in the present work on the basis of laws of conservation of mass, momentum, and energy of individual components. In addition, acoustic properties of this type of mixtures were calculated using a simplified model that assumes the temperature and velocity equilibrium of disperse and carrier phases.

The equations of dynamics of the mixtures were derived using the model of interacting and interpenetrating continua in which a multiphase medium is considered as a combination of several effective continua that occupy the same volume and are characterized by volume-averaged parameters. Interaction of continua is manifested in the processes of mass, momentum, and energy exchange, which may be qualitatively characterized by considering the interaction of an individual solid particle with an ambient gas medium. Assumptions typical of most models of this type are accepted [1]: 1) the size of solid particles is much greater than the mean free path of molecules, which allows one to use the equations of mechanics of continuous media in considering the processes at the microsphere surface; 2) the size of solid particles is much smaller than the distances at which the macroscopic parameters of

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the phases and the mixture change significantly, which allows one to describe the mixture by averaged parameters; 3) the particles do not contribute to the pressure of the medium; 4) if the carrier medium is a mixture of gases, the components of this mixture move with an identical velocity; 5) viscosity and thermal conductivity are ignored, though they are assumed to determine the interaction of the carrier continuum with particles suspended in it; 6) the carrier phase is an ideal gas.

In addition, the following assumptions caused by the presence of cavities inside the particles and permeability of their shells are made: 1) the parameters of the gas inside the microspheres are always uniform (ideal mixing); 2) the gas temperature inside the microspheres is equal to the temperature of microsphere shells; 3) the temperature and velocity of the gas contacting the external surface of the microspheres coincide with the corresponding parameters of the microspheres; 4) the gas flow in terms of the microsphere shells is quasi-stationary, which allows one to express the flux of molecules through the difference in the current values of pressure on both sides of the shell (membrane), thickness, and permeability of the shell material [9]; 5) all microspheres are identical, as well as the processes inside them and near them; 6) the microspheres have an absolutely rigid shell and unchanged volume.

Mass Exchange of a Suspension of Hollow Selective-Permeable Microspheres with a Carrier Gas Mixture. To obtain the equations of dynamics of the mixtures, it is necessary to use the expression for the intensity of mass exchange between the microsphere and the carrier medium due to penetration of gas molecules through the membrane shell. The quasi-stationary approximation is used for these purposes. Then, the gas diffusion in the microsphere shell is described by the equation

$$\frac{1}{z^2}\frac{d}{dz}\Big(z^2 D_i\,\frac{d\eta_i}{dz}\Big) = 0, \tag{1}$$

where η_i is the number density of molecules of the ith gas inside the shell, D_i is the coefficient of diffusion of gas molecules in the shell material, and z is the radial coordinate counted from the center of the microsphere. In an immediate vicinity of the external boundary of the microsphere, the number density of molecules of the ith gas in the shell material is $B_i p_i^{\rm ext}$, where B_i is the temperature-dependent coefficient of solubility of the ith gas in the microsphere material [7] and $p_i^{\rm ext}$ is the partial pressure of the ith gas outside the microsphere. According to the same model, in an immediate vicinity of the internal boundary of the microsphere shell, we have $\eta_i = B_i p_i^{\rm int}$, where $p_i^{\rm int}$ is the partial pressure of the ith gas inside the microsphere (hereinafter, the superscripts "int" and "ext" refer to the gas parameters inside the microspheres and in the carrier phase). The boundary conditions for Eq. (1) may be represented as

$$\eta_i(R_+) = B_i p_i^{\rm ext}, \qquad \eta_i(R_-) = B_i p_i^{\rm int}, \tag{2}$$

where R_+ and R_- are the radii of the external and internal surfaces of the microsphere, respectively.

The solution of Eq. (1) with the boundary conditions (2) has the following form:

$$\eta_i(z) = \frac{C_1}{z} + C_2, \qquad C_1 = \frac{R_+R_-}{R_+ - R_-}\,B_i(p_i^{\rm int} - p_i^{\rm ext}), \qquad C_2 = \frac{R_+p_i^{\rm ext} - R_-p_i^{\rm int}}{R_+ - R_-}.$$

The flux of molecules of the ith gas (if the positive direction of the z axis is that from the center of the microsphere to its surface) is determined by the expression

$$j_i = -D_i\,\frac{d\eta_i}{dz} = \frac{C_1D_i}{z^2} = \frac{B_iD_i}{z^2}\,\frac{R_+R_-}{R_+ - R_-}\,(p_i^{\rm int} - p_i^{\rm ext}) = \frac{q_i}{z^2}\,\frac{R_+R_-}{R_+ - R_-}\,(p_i^{\rm int} - p_i^{\rm ext}),$$

where $q_i = B_iD_i$ is the permeability [7] of the shell material [q_i is in molecules $\cdot$ m/(m$^2\cdot$ sec $\cdot$ Pa)].

The rate J_i of variation of the number of molecules of the ith gas inside the microsphere equals the product of the specific flux j_i taken with the opposite sign and the total area of some spherical surface of radius z ($R_- \leqslant z \leqslant R_+$):

$$J_i = 4\pi z^2(-j_i) = \frac{4\pi R_+R_-}{R_+ - R_-}\,q_i(p_i^{\rm ext} - p_i^{\rm int}) = \frac{q_iS_{\rm eff}}{\delta}\,(p_i^{\rm ext} - p_i^{\rm int}).$$

Here $S_{\rm eff} = 4\pi R_+R_-$ is the effective area of the shell surface [4] and δ is its thickness. The mass-growth rate $æ_i$ of the ith gas in the microsphere cavity is

$$æ_i = \mu_iJ_i = (q_iS_{\rm eff}\mu_i/\delta)(p_i^{\rm ext} - p_i^{\rm int}), \tag{3}$$

where μ_i is the mass of the gas molecule.

The rate of variation of the mass of the ith gas inside the microspheres contained in a unit volume of the mixture is

$$K_i = n_s æ_i = (n_s S_{\rm eff} T_s Q_i R_u/\delta)(\rho_{0i}^{\rm ext} - \rho_{0i}^{\rm int}), \tag{4}$$

where $R_u = kN_{\rm A}$ is the universal gas constant, k is the Boltzmann constant, ρ is the mass density, the subscript 0 refers to true values of quantities and its absence indicates the reduced (i.e., effective, averaged over a small microvolume) parameters, $N_{\rm A}$ is the Avogadro number, and $Q_i = q_i/N_{\rm A}$ is the permeability of the microsphere material for the ith gas, which has the dimension of kmole $\cdot$ m/(m$^2\cdot$ sec $\cdot$ Pa).

In deriving (4), we took into account the equation of state of the ideal gas $p = nkT$, where n is the number density of molecules. Substituting the relations $\rho_i^{\rm ext} = (1-m)\rho_{0i}^{\rm ext}$ and $\rho_i^{\rm int} = (1-m)\beta^3\rho_{0i}^{\rm int}$ ($\beta = R_-/R_+$) into Eq. (4) and taking into account that the number density of microspheres is $n_s = 3m/(4\pi R_+^3)$ (m is the volume fraction of microspheres in the mixture), we finally obtain

$$K_i = \frac{3Q_iR_uT_s}{\beta^2R_+^2(1-\beta)}\Big(\frac{m\beta^3}{1-m}\,\rho_i^{\rm ext} - \rho_i^{\rm int}\Big).$$

Equations of Dynamics of a Suspension Composed of a Gas Mixture and Hollow Selective-Permeable Microspheres. Based on the relations of mass, momentum, and energy balance for a chosen portion of individual components [1, 13] and Eqs. (3), (4) for the quasi-stationary rate of filling the microsphere cavities by gases penetrating through the membrane shell, we obtained the following differential equations in partial derivatives:

$$\frac{\partial m}{\partial t} + \nabla(m\boldsymbol{U}_s) = 0, \qquad \frac{\partial \rho_i^{\rm int}}{\partial t} + \nabla(\rho_i^{\rm int}\boldsymbol{U}_s) = K_i, \qquad \frac{\partial \rho_i^{\rm ext}}{\partial t} + \nabla(\rho_i^{\rm ext}\boldsymbol{U}^{\rm ext}) = -K_i,$$

$$\frac{\partial \rho_s^+\boldsymbol{U}_s}{\partial t} + \nabla(\rho_s^+\boldsymbol{U}_s\cdot\boldsymbol{U}_s) = n_s\boldsymbol{f} - m\nabla p_0^{\rm ext} + \sum_{i=1}^N K_i\boldsymbol{U}_s,$$

$$\frac{\partial \rho^{\rm ext}\boldsymbol{U}^{\rm ext}}{\partial t} + \nabla(\rho^{\rm ext}\boldsymbol{U}^{\rm ext}\cdot\boldsymbol{U}^{\rm ext}) = -n_s\boldsymbol{f} - (1-m)\nabla p_0^{\rm ext} - \sum_{i=1}^N K_i\boldsymbol{U}_s,$$

$$\frac{\partial}{\partial t}\Big[\rho_s^+\Big(e_s^+ + \frac{\boldsymbol{U}_s^2}{2}\Big)\Big] + \nabla\Big[\rho_s^+\boldsymbol{U}_s\Big(e_s^+ + \frac{\boldsymbol{U}_s^2}{2}\Big)\Big] = -m\boldsymbol{U}_s\nabla p_0^{\rm ext} + n_sq + n_s\boldsymbol{f}\boldsymbol{U}_s + \sum_{i=1}^N K_i\Big(e_i(T_s) + \frac{\boldsymbol{U}_s^2}{2}\Big),$$

$$\frac{\partial}{\partial t}\Big[\rho^{\rm ext}\Big(e^{\rm ext} + \frac{(\boldsymbol{U}^{\rm ext})^2}{2}\Big)\Big] + \nabla\Big[\rho^{\rm ext}\boldsymbol{U}^{\rm ext}\Big(e^{\rm ext} + \frac{(\boldsymbol{U}^{\rm ext})^2}{2}\Big)\Big]$$

$$= -\nabla(p_0^{\rm ext}\boldsymbol{U}^{\rm ext}) + m\boldsymbol{U}_s\nabla p_0^{\rm ext} - n_sq - n_s\boldsymbol{f}\boldsymbol{U}_s - \sum_{i=1}^N K_i\Big(e_i(T_s) + \frac{\boldsymbol{U}_s^2}{2}\Big),$$

$$p_0^{\rm ext} = \sum_{i=1}^N p_i^{\rm ext}, \qquad p_i^{\rm ext} = \rho_{0i}^{\rm ext}R_iT^{\rm ext} = \frac{\rho_i^{\rm ext}R_iT^{\rm ext}}{1-m} \quad (i = 1, 2, \ldots, N).$$

Here $\boldsymbol{U}$ is the velocity, T is the temperature, q is the heat flux to the external surface of an individual microsphere, $\boldsymbol{f}$ is the drag force acting on the microsphere from the side of the carrier gas medium, e is the internal energy of a unit mass of the gas, R_i is the gas constant of the ith gas, and N is the number of components of the gas mixture; the superscript plus indicates quantities corresponding to composite particles, i.e., the microsphere and the gas contained inside it.

Relaxation of the Gas Density Inside Microspheres in a Suspension Composed of a Uniform Gas and Hollow Permeable Microspheres. The simplest case of motion of systems of the type considered is a temperature and velocity equilibrium flow of a suspension of hollow gas-permeable microspheres in a uniform gas. As an example, we solved the problem of dispersion and absorption coefficient of acoustic perturbations propagating in a quiescent uniform mixture of hollow permeable glass microspheres and helium. In solving this problem, it is convenient to consider this mixture as homogeneous [14] with a given mass fraction of the solid phase. Along with the parameters conventional for this model (pressure, density, temperature, and velocity), there is an additional parameter: (true) gas density inside the microspheres. Based on the reasoning used in studying homogeneous two-phase systems with continuous solid particles [14], the following equation of state can be obtained for the system considered:

$$p = \frac{\rho R T}{1-\varphi_s\rho/\bar{\rho}_s}\Big(1 - \frac{\varphi_s\beta^3}{1-\varphi_s}\,\frac{\rho_0^{\mathrm{int}}}{\bar{\rho}_s}\Big).$$

Here p is the pressure in the mixture (outside the microspheres), $\bar{\rho}_s$ is the volume-averaged density of the microsphere shell, φ_s is the mass fraction of the solid phase, ρ_0^{int} is the (true) gas density inside the microsphere, and $R = (1-\varphi_s)R_0$ is the effective gas constant of the mixture (R_0 is the gas constant of the uniform gas). In this model, the equations of continuity, momentum, and energy have the same form as those for uniform gas flows, and the dynamics of the mass of the gas located in microsphere cavities is described by the relaxation equation

$$\frac{\partial\rho_0^{\mathrm{int}}}{\partial t} + \boldsymbol{U}\nabla\rho_0^{\mathrm{int}} = -\frac{(1-\varphi_s)\rho - [1-\varphi_s(1-\beta^3)\rho/\bar{\rho}_s]\rho_0^{\mathrm{int}}}{\tau},$$

where the effective time τ of gas-density relaxation inside the microspheres is

$$\tau = (1-\varphi_s\rho/\bar{\rho}_s)\tau_0 = (1-m)\tau_0 \qquad [\tau_0(T) = (1-\beta)\beta^2R_+^2/(3R_uQT)].$$

Acoustic Properties of a Suspension Composed of a Uniform Gas and Hollow Permeable Microspheres. Writing down the equations of continuity, momentum, energy, and density of the gas mass inside the microspheres for uniform flows in a dimensionless form, linearizing them, and considering infinitely weak sinusoidal perturbations of an arbitrary parameter y of the type $y = y_0\{1+\delta y\exp[i(sx-\omega t)]\}$ ($\omega = \Omega R_+/c_0$ is the dimensionless circular frequency of oscillations, Ω is the dimensional circular frequency, c_0 is the velocity of sound in a uniform gas, s is the dimensionless wavenumber, and $i = \sqrt{-1}$), we obtain the following dispersion relation for acoustic waves:

$$\Big[\frac{A}{b}\Big(1+\frac{R}{c_v}\Big) - i\omega\Big(\frac{1}{b}+\frac{R_0}{c_v}\Big)\Big]s^2 = \frac{\omega^2}{a^2}\Big(-i\omega + \frac{(1-\varphi_s)A}{b}\Big).$$

Here $A = 3MQ\sqrt{R_0T_0}/[(1-\beta)\beta^2R_+]$, M is the mass of one kilomole of the gas, $c_v = (1-\varphi_s)c_{v0} + \varphi_sc_s$ (c_v, c_{v0}, and c_s are the specific heat capacities at constant volume of the mixture, uniform gas, and microsphere material, respectively), $a = 1-\varphi_s[1-(1-\beta^3)r]$ and $b = 1-\varphi_s(1+\beta^3r)$, where $r = (\rho_0^{\mathrm{int}}/\bar{\rho}_s)_{\mathrm{eq}}$ is the ratio of the gas density inside (or outside) the microsphere to the volume-averaged density of the solid substance under equilibrium conditions of the undisturbed medium. The dispersion relation yields the following expressions for the frequency dependences of the dimensionless velocity of sound $\bar{c}$ and the coefficient γ of its damping at a distance equal to the wavelength:

$$\bar{c} = a/(\sqrt{\text{\ae}}\,\Phi\cos\alpha), \qquad \gamma = 2\pi\tan\alpha.$$

Here the velocity of sound is normalized to the velocity of sound in the uniform gas and $\text{\ae}$ is the ratio of specific heat capacities at constant pressure and volume for the uniform gas,

$$\Phi = \frac{\{[((1-\varphi_s)A^2/b^2)(1+R/c_v) + (1/b+R_0/c_v)\omega^2]^2 + (\omega A\beta^3\varphi_sr/(ba^2))^2\}^{1/4}}{[(A/b)^2(1+R/c_v)^2 + (1/b+R_0/c_v)^2\omega^2]^{1/2}},$$

$$\alpha = 0.5\arctan\frac{\omega A\beta^3\varphi_sr/b^2}{(1-\varphi_s)(A/b)^2(1+R/c_v) + (1/b+R_0/c_v)\omega^2}.$$

Calculation Results. The calculations were performed for hollow microspheres with glass porous shells with a permeability coefficient $Q = 3.08\cdot10^{-16}$ kmole$\cdot$m/(m$^2\cdot$sec$\cdot$Pa). The undisturbed conditions correspond to the values $T_0 = 300$ K and $p_0 = 10^5$ Pa. The values $c_s = 750$ J/(kg$\cdot$K) and $\rho_s = 2500$ kg/m^3 were borrowed from [15].

Figures 1 and 2 show the frequency dependences of the damping coefficient at a distance equal to the wavelength and the relative velocities of sound for $\beta = 0.98$ and a volume fraction of the solid phase in the mixture $m = 0.1$ (which corresponds to a mass fraction $\varphi_s = 0.989$). Curves 1, 2 and 3 refer to $R_+ = 2\cdot10^{-5}$, $3\cdot10^{-5}$, and $4\cdot10^{-5}$ m, respectively. The form of these curves is typical of media with relaxation processes of various nature (vibrational and rotational relaxation [16] or temperature and velocity relaxation of particles suspended in the gas [17, 18]). A typical feature is a clear maximum of γ at $\Omega_{\max} = 1/\tau$ (see Fig. 1) and the transition from the equilibrium to the frozen value of the velocity of sound with increasing frequency near the threshold mentioned

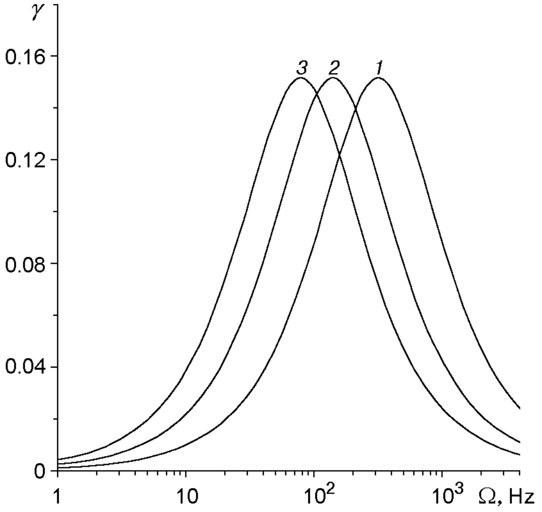

Fig. 1

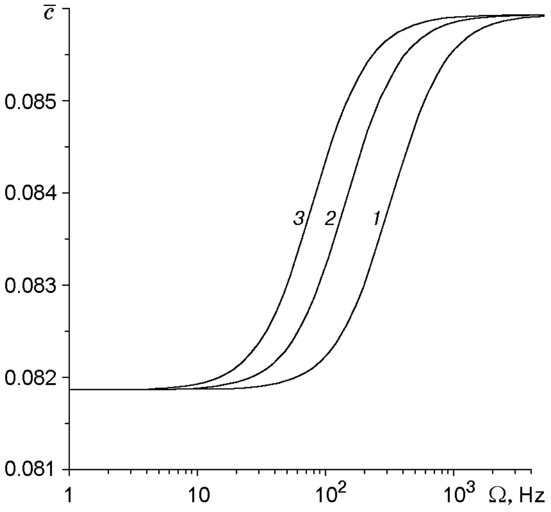

Fig. 2

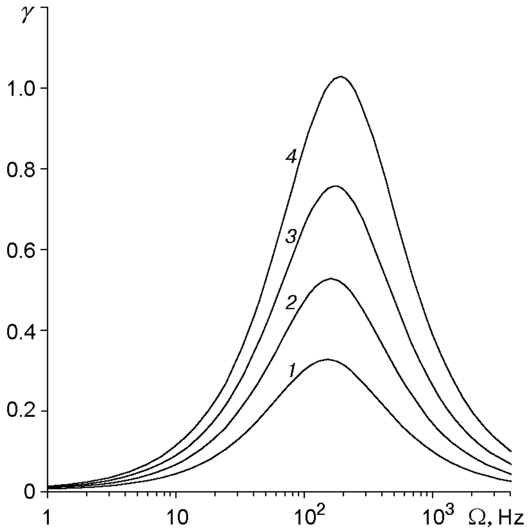

Fig. 3

(see Fig. 2). With increasing size of the microspheres, the time of internal-pressure relaxation increases, and the maximum value of the damping coefficient is shifted to lower frequencies (correspondingly, the transition from the equilibrium to the frozen velocity of sound occurs at a lower frequency). With increasing volume fraction of the microspheres, a significant decrease in the velocity of sound is observed both in the equilibrium (low-frequency) and frozen (high-frequency) ranges, as compared to its value in pure helium. It should be noted that the maximum values of the damping coefficient remain unchanged; they are only shifted along the frequency axis with changing radius of the microspheres.

The asymptotic values of the equilibrium $\bar c_{\rm eq}$ and frozen $\bar c_{\rm f}$ relative velocity of sound correspond to the low-frequency and high-frequency limits in the expression for $\bar c(\Omega)$:

$$\bar c_{\rm eq} = \sqrt{\frac{a^2}{(1-\varphi_s)æ}\Big(1+\frac{R}{c_v}\Big)}, \qquad \bar c_{\rm f} = \sqrt{\frac{a^2}{æ}\Big(\frac{1}{b}+\frac{R_0}{c_v}\Big)}.$$

The damping coefficient increases with increasing mass fraction of the microspheres (Fig. 3). Curve 1–4 in Fig. 3 refer to a volume fractions of the microspheres $m = 0.2$, 0.3, 0.4, and 0.5, respectively. The microsphere radius is constant and equal to $3 \cdot 10^{-5}$ m, and $\beta = 0.98$. For small volume fractions ($m \leqslant 0.1$), an increase in m leads to a proportional increase in the maximum value of γ, whereas the value of $\Omega_{\max} \approx 1/\tau_0$ remains almost constant.

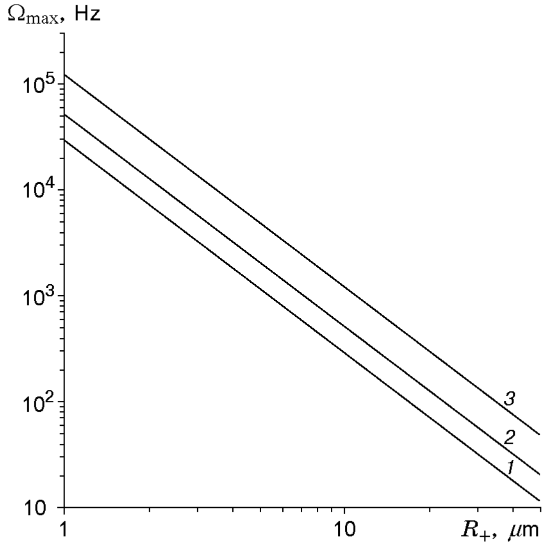

Fig. 4

A further increase in the volume fraction of the microspheres ($m > 0.1$) is also accompanied by an increase in the maximum values of the damping coefficient (Fig. 3), but the value of $\Omega_{\max}$ is shifted toward higher frequencies. The reason is the influence of the volume fraction of particles on the relaxation time τ. The volume fraction has a much weaker effect on the behavior of the corresponding curves of dispersion of the velocity of sound, and they are not given here.

For various values of β, Fig. 4 shows the circular frequency $\Omega_{\max}$ versus R_+ (calculated for small concentrations of microspheres by the formula $\Omega_{\max} = 1/\tau_0$), for which the maximum value of the damping coefficient is reached at a distance equal to the wavelength. Curves 1–3 refer to $\beta = 0.90$, 0.95, and 0.98, respectively. It is seen that one can change the frequency of the most effective absorption of sound within wide limits (by four orders of magnitude) by changing the parameter β. These variations may also be performed by changing the microsphere size or the permeability of the shell material, which is reached by modification of the porous structure of the material or by using another material [2, 3].

The characteristic times of microsphere temperature and velocity relaxation in helium and the time of gas mixing inside the microspheres due to diffusion were estimated using the formulas derived in [1]. The greatest of these times, which is the time of velocity relaxation for particle sizes (less than 50 μm) and value of β (greater than 0.9) studied in the present work, is always approximately an order of magnitude smaller than the characteristic time of equalization of gas densities inside and outside the microspheres, which allows one to use the assumption on the temperature and velocity equilibrium of particles at frequencies of acoustic oscillations lower, equal, or slightly higher than $\Omega_{\max}$.

Thus, the calculations show that the damping coefficient and the velocity of sound in one-temperature, one-velocity mixtures composed of a gas and hollow permeable microspheres have the same frequency dependences as most media with relaxation phenomena. In our case, the relaxation process is the equalization of gas densities (pressures) inside and outside the microspheres due to penetration of molecules through membrane shells of the microspheres. The relaxation time of this process may be controlled by changing the microsphere size, the ratio of the internal to the external radius, or the permeability of the shell. This allow one to vary the frequency of the most effective absorption of low-frequency acoustic oscillations ($\Omega < 1000$ Hz) within a wide range.

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