SOUND DISPERSION IN A RELAXING POLYMER FLUID WITH BUBBLES

Z. P. Shul'man and S. P. Levitskii UDC 536.24:532.135

Results are presented of an investigation of the singularities of sound wave propagation in a polymer solution with bubbles.

Two-phase systems of a polymer fluid (solution or melt) and gas bubbles are formed during production and polymer reworking processes because of chemical gas liberation, low wettability of the solid boundaries of the carrying medium, etc. The stability of such systems is determined by the high Newtonian viscosity of the liquid phase which hinders spontaneous gravitational phase separation [1]. An important condition in the progress of many processes is the preliminary evacuation of free gas, with which the problem of bubble diagnostics is related directly. One method of determining the gas content in a fluid is acoustic [1, 2], which raises the interest in studying weak pressure perturbation propagation in polymer media with bubbles. The purpose of this paper is to derive a dispersion equation for a relaxing polymer fluid with gas inclusions that takes account of all fundamental dissipative effects accompanying bubble fluctuation in the wave.

Sound propagation in a medium with bubbles will be considered as a process of multiple scattering of the fundamental signal [3]. In a first step we construct the wave field scattered by a single bubble. It is convenient to execute the solution in dimensional variables by selecting the quantities ρ_{20}, p_{20}, T_0 and the equilibrium radius of one of the bubbles R_0 (we consider the mixture polydisperse) as the characteristic parameters. We write the linearized equations of motion and continuity in the form

$$\frac{\partial \mathbf{v}_2}{\partial \tau} = -\nabla p_2 + \nabla \cdot \pi, \quad \frac{\partial \rho_2}{\partial \tau} = -\nabla \cdot \mathbf{v}_2. \tag{1}$$

For a monochromatic incident wave, the perturbations of the hydrodynamic quantities in (1) can be represented in the form

$$\{\mathbf{v}_2,\ p_2,\ \rho_2,\ \pi\} = \{\mathbf{v}_2^*,\ p_2^*,\ \rho_2^*,\ \pi^*\} \exp i\omega\tau, \tag{2}$$

where the asterisk denotes complex amplitudes of the perturbations. The tensor π and the scalar p_2 are defined according to the general hereditary models for a polymer medium relaxing under shear and volume deformations based on the Boltzmann–Volterra principle [4]:

$$\pi_{ij} = 2\int\limits_{-\infty}^{\tau} G_1(\tau-\tau')\, s_{ij} d\tau' + 2\eta_s^* s_{ij},\ p_2 = \eta_v^* \frac{\partial \rho_2}{\partial \tau} - \int\limits_{-\infty}^{\tau} G_2(\tau-\tau')\, e_{kk} d\tau',$$

$$G_\alpha(\tau) = G_{\alpha 0} + \int\limits_0^\infty F_\alpha(\lambda)\, e^{-\tau/\lambda}\, d\lambda,\ G_{10} = 0,\ G_{20} = K^* = K_a p_{20}^{-1}. \tag{3}$$

Sound wave propagation in a fluid is later considered an adiabatic process while the deviations from adiabaticity, as a consequence of interphasal heat transfer, are taken into account only in direct proximity to the phase interfaces, which is perfectly justifiable in the case of gas bubbles [5].

Let us introduce the velocity potential ψ_0 in the incident wave by setting $\mathbf{v}_2 = \nabla\psi_0$. Then we obtain the Helmholtz equation for ψ_0 from (1)-(3):

$$\nabla^2\psi_0 + (\omega^2/G)\,\psi_0 = 0, \tag{4}$$

$$G = \frac{4}{3}(G_1^* + i\omega\eta_s^*) + G_2^* + i\omega\eta_v^*,\ G_\alpha^* = G_\alpha' + iG_\alpha''.$$

A. V. Lykov Institute of Heat and Mass Transfer, Academy of Sciences of the Belorussian SSR. Lenin Komsomol Voronezh State University. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 48, No. 1, pp. 63-69, January, 1985. Original article submitted October 21, 1983.

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Let ψ_{k0} denote the fluid velocity potential at the point $\mathbf{r}_k$ of the fundamental wave field. Let a single bubble with equilibrium radius R_{k0} be located at this point of the space. We take the long-wave approximation ($l_2 \gg R_{k0}$) usual in acoustics problems of two-phase media, and we express the scattered wave field ψ_{ks} in terms of the potential ψ_{k0}. We introduce a spherical coordinate system connected with the center of the bubble. Then taking the symmetry of the fluid flow around a radially fluctuating bubble into account, we obtain the solution corresponding to a divergent wave for ψ_{ks} from (4):

$$r\psi_{ks} = A_k \exp[i(\omega\tau - mr)], \quad m = \omega G^{-1/2}, \tag{5}$$

where A_k is an arbitrary constant. The amplitudes of the fluid velocity perturbation v^*_{2s} and the pressure p^*_{2s} of the scattered wave are determined in the form

$$v^*_{2s} = -r^{-2}A_k(1 + imr)\exp(-imr), \tag{6}$$

$$p^*_{2s} = -i\omega r^{-1}\left[1 - \frac{4}{3}G^{-1}(G^*_1 + i\omega\eta^*_s)\right]A_k\exp(-imr).$$

We find the constant A_k from the condition for junction of the solutions of the external and internal problems for the fluctuating bubble. We use the homobaricity condition ($l_1 \gg R_{k0}$), which is valid in a broad frequency range [6], in formulating the gas dynamics equations for the gas in the bubble. Then the linearized system of dimensionless equations of the internal problem is written in the form

$$\frac{\partial\rho_1}{\partial\tau} + r^{-2}\frac{\partial}{\partial r}(r^2 v_1) = 0, \quad p_1 = p^*_{10}(\rho_1 + \theta), \tag{7}$$

$$\frac{\partial\theta}{\partial\tau} = (\gamma - 1)(\gamma p^*_{10})^{-1}\frac{\partial p_1}{\partial\tau} + \frac{1}{\mathrm{Pe}}\nabla^2\theta,$$

$$\frac{1}{p^*_{20}}\frac{\partial p_1}{\partial\tau} = 3\gamma\left(\frac{1}{\mathrm{Pe}}\left(\frac{\partial\theta}{\partial r}\right)_{r=R^*_k} - \frac{dR^*_k}{d\tau}\right).$$

We seek the bubble radius in the form of the real part of the expression

$$R^*_k = s_k(1 + \delta_k\exp(i\omega\tau)), \quad s_k = R_{k0}/R_0,$$

and the small perturbations of the gasdynamic parameters analogously to (2)

$$\{\rho_1, p_1, v_1, \theta\} = \{\rho^*_1, p^*_1, v^*_1, \theta^*\}\exp(i\omega\tau). \tag{8}$$

We denote the pressure perturbation in an incident wave on the bubble surface by $p^*_{k0}\exp(i\omega\tau)$ and we formulate the dynamic, kinematic, and thermal boundary conditions

$$p_2 - \pi_{rr} = p_1 + 2\sigma^* s_k^{-1}\delta_k\exp(i\omega\tau), \tag{9}$$

$$\sigma^* = \sigma(p_{20}R_0)^{-1}, \quad p_2 = (p^*_{k0} + p^*_{2s})\exp(i\omega\tau),$$

$$v_1 = v_2 = i\omega s_k\delta_k\exp(i\omega\tau), \quad \theta = 0, \quad r = s_k; \quad \theta < \infty, \quad r = 0.$$

Taking account of (6), (8), and (9), the solution of system (7) yields

$$A_k = -i\omega s_k^3\delta_k, \quad \delta_k = \bar{\delta}_k p^*_{k0}, \quad \bar{\delta}_k = D_k/\Delta_k, \quad D_k = \beta^2 - 3\alpha_k s_k^{-1}(1 - \gamma), \tag{10}$$

$$\Delta_k = D_k(2\sigma^* + \omega^2 s_k^2(1 + ims_k)^{-1} - 4(G^*_1 + i\omega\eta^*_s)) - 3p^*_{10}\gamma\beta^2,$$

$$\beta^2 = i\omega\mathrm{Pe}, \quad \alpha_k = \beta\coth\beta s_k - s_k^{-1}.$$

We express the pressure perturbation p^*_{k0} in the incident wave at the point $\mathbf{r} = \mathbf{r}_k$ and the scattered wave potential ψ_{ks} in terms of the value ψ_{k0} of the fundamental wave potential at this point by using (1)-(3). We obtain

$$p^*_{k0} = B\psi_{k0}\exp(-i\omega\tau), \quad B = i\omega G^{-1}\left[\frac{4}{3}(G^*_1 + i\omega\eta^*_s) - G\right],$$

$$r\psi_{ks} = \Phi_k\psi_{k0}\exp(-imr_k), \quad \Phi_k = -i\omega s_k^3\bar{\delta}_k B, \quad r_k = |\mathbf{r} - \mathbf{r}_k|. \tag{11}$$

We consider sound wave interaction with the collective of bubbles. In the polymer volume V under consideration let there be N bubbles with the equilibrium radii R_{k0} ($k = 1, 2, \ldots, N$). Following [7], we introduce the probability distribution function for the bubble configuration

$$P = \left(\frac{1}{N}\right)^N q(\mathbf{r}_1, s_1)\, q(\mathbf{r}_2, s_2) \ldots q(\mathbf{r}_N, s_N), \tag{12}$$

where $q(\mathbf{r}, s)ds$ is the mean value of the number of bubbles per unit volume with center at the point $\mathbf{r}$, whose radii are in the band $(s, s + ds)$. Then the configuration mean of the arbitrary quantity $f = f(\mathbf{r}_j, s_j)$ that depends on the bubble location in the system can be determined from the formula

$$\langle f \rangle = \int_{V^*} \ldots \int\int_{V^*} \int \ldots \int\int f(\mathbf{r}_j, s_j)\, P(\mathbf{r}_j, s_j)\, ds_1 ds_2 \ldots$$
$$\ldots ds_N d\mathbf{r}_1 d\mathbf{r}_2 \ldots d\mathbf{r}_N, \quad V^* = V/R_0^3. \tag{13}$$

Let $\psi(\mathbf{r})$ be the potential of the resultant field due to the superposition of the fundamental and scattered fields in the medium with bubbles, taking multiple scattering into account. The factor $\exp(i\omega\tau)$ will henceforth be omitted, for brevity. Then, by representing the resultant field in the neighborhood of each bubble as the superposition of an incident wave and a wave scattered by a bubble, we arrive at the equations

$$\psi_k = \psi(\mathbf{r}_k) - \psi_{ks} = \psi(\mathbf{r}_k) - r_k^{-1}\Phi_k\psi_k \exp(-imr_k), \tag{14}$$

$$\psi(\mathbf{r}) = \psi_0(\mathbf{r}) + \sum_{k=1}^{N} \psi_{ks} = \psi_0(\mathbf{r}) + \sum_{k=1}^{N} \Phi_k\psi_k r_k^{-1} \exp(-imr_k), \tag{15}$$

where ψ_k is the potential of the wave acting on the k-th bubble. On the other hand, the potential ψ_k can be represented in the form

$$\psi_k = \psi_0(\mathbf{r}_k) + \sum_{\substack{n=1\\ n \neq k}}^{N} \Phi_n\psi_n r_{nk}^{-1} \exp(-imr_{nk}), \quad r_{nk} = |\mathbf{r}_n - \mathbf{r}_k|. \tag{16}$$

System (14)-(16) determines the desired quantities as characteristics of the self-consistent field. Evaluating the configuration mean (13) from both sides of (14) with (4), (15), and (16) taken into account, we obtain an equation for the potential of the mean sound wave field in a polymer fluid with bubbles

$$\nabla^2 \langle \psi(\mathbf{r}) \rangle + n^2 \langle \psi(\mathbf{r}) \rangle = 0, \tag{17}$$

$$n^2 = \omega^2 G^{-1} - 4\pi\omega^2 \left(1 - \frac{4}{3} G^{-1}(G_1^* + i\omega\eta_s^*)\right) \int s_k^3 n(\mathbf{r}, s_k)\, \bar{\delta}_k ds_k. \tag{18}$$

Equation (18) solves the problem formulated. The specific nature of the wave dispersion and absorption can be analyzed if the bubble distribution law is known in space and by size. This equation takes account of the influence of all the fundamental factors on the sound wave propagation process: the rheological features of the polymer fluid, the nonequilibrium interphasal heat transfer, the losses in sound radiation by the bubbles, the semidispersity of the mixture. For a monodisperse mixture of homogeneously distributed inclusions $s_k \equiv 1$; here [8]

$$\int s_k^3 q(s_k)\bar{\delta}_k ds_k = (4\pi)^{-1} 3\alpha_0(1 - \alpha_0)\,\bar{\delta}, \quad \bar{\delta}_k \equiv \bar{\delta}\ (k = 1, 2, \ldots, N).$$

Then the dispersion equation (18) takes the form

$$\frac{n^2}{\omega^2} = \frac{1}{c_0^2}\left[1 - 3\alpha_0(1 - \alpha_0)\left(G - \frac{4}{3}(G_1^* + i\omega\eta_s^*)\right)\bar{\delta}\right]. \tag{19}$$

If components characterizing the rheological and thermal dissipation are neglected in (19), we find by setting $G_1^* = 0$, $G_2^* = K^*$, $\eta_S^* = \eta_V^* = 0$ and $|\beta| \gg 1$ or $|\beta| \ll 1$

$$\frac{1}{c^2} = \frac{1}{c_0^2} + \frac{3\alpha_0(1 - \alpha_0)}{\omega_0^2 - \omega^2 + i\omega^3/c_0}, \quad c_0^2 = K^*, \quad \omega_0^2 = 3p_{10}^*\gamma - 2\sigma^*, \tag{20}$$

which agrees with the classical dispersion equation of the theory of multiple scattering for a fluid with gas bubbles [8] with just acoustic losses taken into account during fluctuation of the inclusions.

As follows from (19), the "frozen" speed of sound in the system is in agreement with the speed of acoustic wave propagation in a pure relaxing fluid. The magnitude of the equilibrium sound velocity is independent of the rheological parameters of the medium since $\{G_1^*, \omega\eta_S^*, \omega\eta_V^*\} \to 0$ and $G_2^* \to K^*$ as $\omega \to 0$.

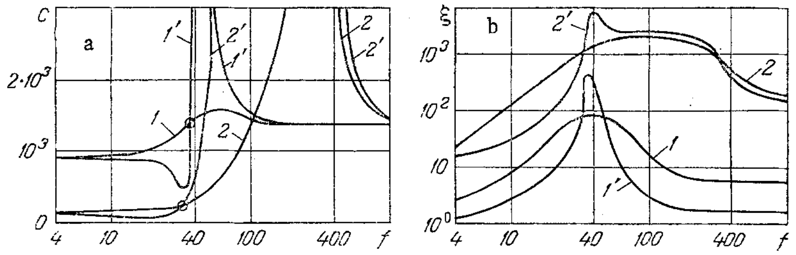

Fig. 1. Sound dispersion and absorption in a polymer solution with air bubbles ($f = (\omega/t_0)/(2\pi\cdot10^3)$, $\xi = -\mathrm{Im}\{n\}/R_0$, $C = (\omega/\mathrm{Re}\{n\})(p_{20}/\rho_{20})^{1/2}$. $R_0 = 10^{-4}$ m, $\nu = 10^3$, $K_a = 1.61\cdot10^9$ Pa, $\rho_{20} = 850$ kg/m³, $\sigma = 0.022$ N/m, $\lambda_1' = 10^{-2}$ sec): 1, 1') $\alpha_0 = 10^{-4}$; 2, 2') $\alpha_0 = 10^{-2}$. C, m/sec; f, kHz.

For numerical computations on an electronic computer, the dimensional frequencies f, the sound speed C and the wave damping factor ξ were determined from (19). The results of the computations are presented in the figure, *a* and b, for a polymer solution with air bubbles at $T_0 = 20°C$ and $p_{20} = 10^5$ Pa. The relaxation spectrum was taken in the form

$$F_1 = (\eta_p^* - \eta_s^*)\left(\sum_{k=1}^{\nu}\lambda_k\right)^{-1}\sum_{k=1}^{\nu}\delta(\lambda - \lambda_k),\ \lambda_k = \lambda_1/k^2.$$

The maximal relaxation time in the spectrum λ_1 and the quantity ν were estimated by the Kargin—Slonimskii—Rauth model. The values taken correspond in order of magnitude, for instance, to a 2.5% solution of polystyrene with molecular weight $2\cdot10^6$ in toluene [9]. Computations showed that taking account of the bulk viscoelasticity of the solution in the presence of bubbles does not affect the values of C and ξ. Curves 1, 2 in the figure correspond to a Newtonian fluid with viscosity of the solution ($\eta_p = \eta_s = 0.5$ Pa·sec); 1', 2' to the polymer solution ($\eta_p = 0.5$ Pa·sec, $\eta_s = 0.5\cdot10^{-3}$ Pa·sec).

As follows from the graphs, taking account of the viscoelastic properties of the liquid phase in the example considered changes the nature of sound dispersion qualitatively in a medium with gas bubbles. A computation by a Newtonian rheological model of a fluid yields growth in the sound velocity with frequency on the low-frequency branch of the dispersion curve, while the maximal velocity of wave propagation only exceeds the "frozen" sound velocity insignificantly and can be in agreement with it in the case of high fluid viscosity and sufficiently small size and concentration of the inclusions (curve 1 in Fig. 1a). Such a kind of dependence C = C(f) for curve 1 is due to the large dissipation in the system because of the high Newtonian viscosity of the medium resulting in this case in not only the disappearance of the "window of opacity" on the dispersion curve which is characteristic for a system without dissipation [10], but also in the fact that the least velocity of sound propagation in a bubble mixture agrees with the equilibrium value $C_e = \lim_{f\to0} C$. However, the influence of the relaxation properties of the liquid phase results in a change in the sign of the dispersion in the left branch of the dispersion curve, whereupon the minimal velocity of sound wave propagation turns out to be less than C_e. The sound propagation velocity in the high-frequency range simultaneously grows significantly; here the location of the dispersion zone on the frequency axis is conserved.

As computations show, growth in the amplitude of the gas bubble fluctuations in a wave that is due to viscoelasticity effects results in a significant increase in the sound absorption in a fluid with bubbles near the resonance frequencies (Fig. 1b). Far from the resonance frequency, the sound damping in a relaxing medium with bubbles can be substantially lower than in an analogous viscous fluid. As the concentration of inclusions, as well as the bubble size, increase, the role of the rheological factors weakens and the differences between the results of computations using the relaxation and Newtonian models of the liquid phase level off. Let us mention that the change in sign of sound dispersion in the preresonance frequency domain when taking account of the viscoelastic properties of the liquid phase for the curves in Fig. 1a results in the existence of a frequency at which the velocity of wave propagation in the relaxing and analogous viscous fluid with gas bubbles agrees. The corresponding point is marked with a circle on the dispersion curves.

The results obtained are explained physically by a change in the contribution of the rheological dissipative mechanism to the total dissipation during sound propagation in a rheologically complex relaxing medium with bubbles as compared with an analogous viscous fluid. Since the magnitude of the dissipative losses in a polymer medium turns out to be substantially less during bubble fluctuations than in an analogous viscous fluid [11], the sound dispersion and absorption do not grow in the resonance zone.

NOTATION

Dimensional quantities: k, c_p, heat-conduction coefficient and specific heat of the gas at constant pressure; T_0, equilibrium temperature; p_{i0}, ρ_{i0}, equilibrium pressure and density; T, gas temperature; η_s, η_v, shear and bulk viscosities of the solvent; K_a, adiabatic volume modulus; σ, surface tension coefficient; λ_1', maximal relaxation time; $t_0 = R_0(\rho_{20}/p_{20})^{1/2}$, $\eta_0 = R_0(\rho_{20}p_{20})^{1/2}$, characteristic time and viscosity coefficient; η_p, Newtonian viscosity of the solution; l_1, l_2, sound wavelength in the gas and fluid, respectively. Dimensionless quantities: $Pe = R_0(p_{20}/\rho_{20})a^{-1}$, Peclet number; $a = k/(\rho_{10}c_p)$, thermal diffusivity factor; γ, gas adiabatic index; v_i, p_i, ρ_i, π, velocity, pressure, density, and stress tensor deviator perturbations in the wave; τ, time; $\lambda = \lambda'/t_0$, relaxation time; ω, wave frequency; e_{ij}, s_{ij}, strain rate tensor and its deviator, respectively; $F_\alpha(\lambda)$, G_α', G_α'' ($\alpha = 1, 2$), relaxation time distribution function and the components of the complex dynamical elastic moduli; $\theta = T/T_0 - 1$, $p_{10}^* = p_{10}/p_{20}$, temperature perturbation and equilibrium pressure in the gas; $R_k^* = R_k/R_0$, running bubble radius; r, radius of the spherical coordinate system; n, $c = \omega/n$, complex wave number and speed of sound in a fluid with bubbles; c_0, speed of sound in a pure fluid; η_s^*, η_v^*, $\eta_p^* = \{\eta_s, \eta_v, \eta_p\}/\eta_0$, dimensionless viscosity coefficients; α_0, volume bubble concentration; $\delta(\lambda - \lambda_k)$, delta function. Subscripts: $i = 1$, gas phase; $i = 2$, fluid.

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