Absorption and dispersion of ultrasound and hypersound in glycerine

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The velocity and absorption of ultra- and hypersound in glycerine are measured by a pulse technique at frequencies between 3 and 2000 MHz and at temperatures between −70 and +180°C. A dispersion of the sound velocity of the order of 40–60% and an absorption maximum of sound variation of the temperature are observed experimentally. The phenomenon cannot be described by the relaxation theory with a single relaxation time but can be satisfactorily described by the nonlocal theory of propagation of sound in viscous media.

Recent acoustic investigations have demonstrated that the absorption and velocity of ultrasound and hypersound in low-viscosity liquids can be described satisfactorily by the relaxation theory of L. M. Mandel'shtam and M. A. Leontovich, using one or several relaxation times. The situation is quite different in the case of viscous and highly viscous liquids (viscosities ranging from a few poises to the glassy state).

Experimental investigations of the absorption and velocity of ultrasound and hypersound in viscous and highly viscous liquids are still few in number. They have been carried out mainly in the frequency range 2—54 MHz but in a wide range of viscosities.[1-10] In some cases,[2-4] these investigations have been accompanied by a study of the propagation of longitudinal and transverse sound in viscous liquids and this has made it possible to distinguish the bulk and shear viscosities and to establish that considerable changes in the velocity and absorption occur in practically the same range of frequencies. Measurements of the temperature dependences of the absorption of sound have revealed the presence of a maximum which decreases and shifts to higher temperatures when the frequency is raised. Similar behavior has also been reported for highly viscous liquids at higher frequencies.[11,12]

Fabelinskiĭ et al.[5-7] observed a dispersion of the velocity of hypersound of the order of 40—70% in glycerin, triacetin, and 1,2-propylene glycol. The experimental studies[5,6] demonstrated that the relaxation theory, which describes so well the propagation of sound in liquids characterized by a low shear viscosity and a high bulk viscosity, could not provide even a qualitative description of highly viscous liquids. The main relationships were established and analytic expressions were obtained that would have to be predicted by a future theory describing the propagation of sound in viscous liquids. It was also pointed out in[5] that a description of the velocity and absorption of sound in viscous media based on multiple-valued functions with a continuous set of relaxation times would be physically meaningless.

The relationships obtained by Krivokhizha and Fabelinskiĭ[5] were used by Isakovich and Chaban[13] to develop a nonlocal theory of the propagation of sound in a viscous medium on the assumption that a viscous liquid is microinhomogeneous medium.

The experimental relationships[5] and theoretical formulas[13] describe satisfactorily all the experimental results obtained so far. Our task was to extend the range of the experimental investigations by measuring the velocity and absorption of sound in glycerin in a wide range of frequencies and in a wide range of viscosities (this has not yet been done) and then to compare the results with theoretical calculations.

We carried out acoustic measurements in glycerin in the frequency range 3—2000 MHz at temperatures from −70 to +180° C. The absorption and velocity of ultrasound were measured by a pulse method to within 5—10% at low frequencies and temperatures and to within 0.1—3% at high frequencies and temperatures. At low temperatures and microwave frequencies, when the absorption of sound in glycerin was strong and a standing wave could not be established, we used the method of coherent shift of a delayed radio-frequency signal and an acoustic signal.[14]

In the temperature range 1—180°C the viscosity was measured with an Ostwald capillary viscometer; in the range from 0 to −30°C the viscosity was determined with a Hoeppler viscometer and the density with a pycnometer. Between −30 and −70°C the density was found by extrapolation of the experimental straight lines and the viscosity was calculated from the formula

$$\lg \eta = -A + B/(T - T_\infty). \qquad (1)$$

The results of measurements of the shear viscosity and of the density are collected in Table I.

The temperature was maintained to within 0.1—0.2 deg C with the aid of a U-10 ultrathermostat or with an N180 ultracryostat.

At high and microwave frequencies small displacements were measured with an optical system in which the interference of light was employed.[14] A helium-neon laser (λ = 6328 Å) was used as the monochromatic light source.

Measurements of the absorption of ultrasound and hypersound in glycerin showed that, in the investigated range of frequencies and temperatures, the absorption coefficient α deviated from the quadratic dependence. When the temperature was lowered the relaxation frequency shifted toward lower frequencies (Fig. 1). The temperature dependence of the ratio α/f^2, obtained at various frequencies f, had a definite maximum whose amplitude decreased with increasing frequency and shifted toward higher temperatures (Fig. 2).

TABLE I. Temperature dependences of the density and viscosity of glycerin

t, °C	η, P	ρ, g/cm³	t, °C	η, P	ρ, g/cm³	t, °C	η, P	ρ, g/cm³
180	—	1.140	100	—	1.196	20	5.7	1.252
170	—	1.146	90	0.50	1.203	10	15.9	1.259
160	0.1	1.153	80	—	1.210	0	90	1.267
150	—	1.160	70	0.71	1.217	−10	505	1.272
140	—	1.167	60	0.82	1.224	−20	1300	1.280
130	0.24	1.170	50	0.96	1.231	−30	10 000	1.288
120	—	1.182	40	1.18	1.241			
110	0.36	1.189	30	1.57	1.246			

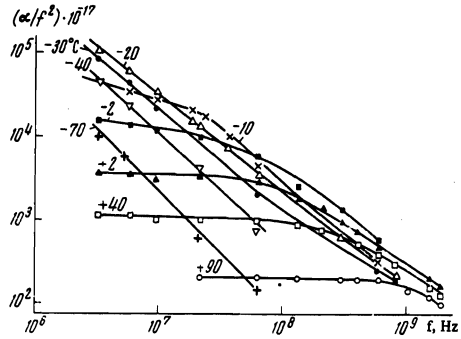

FIG. 1. Frequency dependences of the ratio α/f^2 for glycerin at various temperatures which are given alongside the curves and expressed in °C.

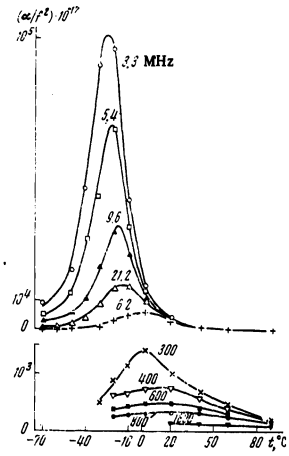

FIG. 2

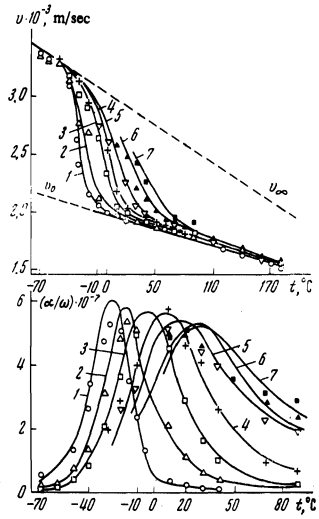

FIG. 3

FIG. 2. Temperature dependences of α/f^2 for glycerin obtained at different frequencies given alongside the curves in MHz.

FIG. 3. Temperature dependences of the velocity of sound v and of the ratio α/ω for glycerin obtained at various frequencies f(MHz): 1) 3.3; 2) 21.2; 3) 62; 4) 200; 5) 600; 6) 1000; 7) 1800. The continuous lines represent the results of calculations based on the Isakovich-Chaban theoretical formulas. [13]

Extrapolation of the velocity of sound v_0 to the range of temperatures where the rapid change of the velocity became a linear function $v_\infty(t)$ (Fig. 3) gave the dispersion of the velocity of sound. The dispersion $d = (v_\infty - v_0)/v_0$ was of the order of 40–60% when regarded as a function of temperature. It was greater at low temperatures than at high temperatures. Our results were in satisfactory agreement with the experimental data obtained for the absorption and velocity of sound in glycerin obtained by the Rayleigh spectroscopy method.[15,16]

We compared our experimental results with the calculation formulas deduced by Isakovich and Chaban.[13] According to the Isakovich-Chaban theory, the absorption coefficient, velocity of sound, and delay time τ are given by the formulas

$$\frac{\alpha}{\omega}=\frac{1}{v_\infty}\frac{1}{\sqrt{2}}\left\{\left[\left(1-\frac{v_\infty^2-v_0^2}{v_0^2}\operatorname{Im}F\right)^2+\left(\frac{v_\infty^2-v_0^2}{v_0^2}\operatorname{Re}F\right)^2\right]^{1/2}-\left(1-\frac{v_\infty^2-v_0^2}{v_0^2}\operatorname{Im}F\right)\right\}^{1/2}, \tag{2}$$

$$\frac{1}{v}=\frac{1}{v_\infty}\frac{1}{\sqrt{2}}\left\{\left[\left(1-\frac{v_\infty^2-v_0^2}{v_0^2}\operatorname{Im}F\right)^2+\left(\frac{v_\infty^2-v_0^2}{v_0^2}\operatorname{Re}F\right)^2\right]^{1/2}+\left(1-\frac{v_\infty^2-v_0^2}{v_0^2}\operatorname{Im}F\right)\right\}^{1/2}, \tag{3}$$

$$\tau=\frac{5}{3}\frac{v_\infty^2}{\rho v_0^2}\frac{\eta+{}^3/_4\eta'}{v_\infty^2-v_0^2}. \tag{4}$$

The values of the limiting velocities v_∞ and v_0, found by linear extrapolation of the experimental results, were substituted into Eqs. (2)–(4) and the values of α/ω, v, and τ were calculated. The continuous lines in Fig. 3 represent the results of such calculations and the points are the experimental values. It is clear from the figure that over a wide range of frequencies the agreement between the experiment and theory is good. At low frequencies the calculated frequency dependence is in satisfactory agreement with the experimental results but at high frequencies a discrepancy of the order of 15–25% is observed. Obviously, the shear modulus of glycerin is high and it is possible that allowance for this modulus may improve the agreement between the calculated and experimental values of the velocity of sound and the absorption coefficient.

It follows from the results reported here that calculations based even on the simplified formulas of the Isakovich-Chaban nonlocal theory, which uses just one parameter τ that has a definite physical meaning for individual viscous liquids, are in qualitative agreement with the experimental results obtained in a wide range of frequencies and temperatures.

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