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ACOUSTIC SPECTRA IN A DISPERSE SYSTEM OF AEROSIL AND POLYPROPYLENEGLYCOL (PPG-425)

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We have studied the absorption and speed of sound in disperse systems based on aerosil and polypropyleneglycol (PPG-425) within the range of concentrations from 1 to 10 vol.% and of frequencies from 5 to 1250 MHz. It is shown that the aerosil particles exist in the form of clusters; their dimensions have been determined.

Investigation of the acoustic properties (the absorption coefficient α and the speed of sound C) of disperse systems is of interest for establishing a correlation between the thermodynamic and kinematic parameters and the structure of such systems. Measurements of the coefficient of absorption and speed of sound in disperse systems make it possible to obtain information about the mechanisms of relaxation processes and clustering of particles and about the interactions between the particles and the dispersion medium [1–5].

In [1–3] it is shown that the propagation of acoustic waves in disperse systems is accompanied by greater absorption than such propagation in the dispersion medium. A theoretical analysis of the excess absorption was carried out in [2–4] within the framework of the following model: the particles are spherical, the concentration of the disperse medium is insignificant, and the absorption of sound in the disperse medium is independent of the frequency. The absorption of acoustic waves in disperse systems as a function of the concentration was investigated experimentally in a number of works [3–5], and the excess absorption $\alpha_{exc} = \alpha - \alpha_0$ was determined (α and α_0 are the coefficients of absorption of sound in the disperse system and in the dispersion medium, respectively). From an analysis of the excess absorption, the contributions of various mechanisms responsible for the losses were determined, and the effect exerted by the dimensions of the disperse-phase particles on the magnitude of the absorption was established. However, as a rule, these works were conducted with coarsely dispersed systems in a narrow range of frequencies and temperatures.

In the present work we give results of measurements of the absorption of sound in a disperse system based on polypropyleneglycol and methylaerosil (A-300). The methylaerosil consists of particles of radius $R \simeq 5 \cdot 10^{-9}$ m. The polypropyleneglycol (PPG) is chromatographycally pure and has a molecular mass $M = 425$. The acoustic properties of PPG-425 were studied in [6, 7].

The absorption of sound was measured within the range of frequencies from 3 to 1200 MHz by the pulse method with an error of 3 to 5%; the speed of sound was measured at a frequency of 5 MHz with an error of 0.1%. The density (ρ) was determined by the pycnometric method with an error of 0.05%. The coefficient of shear viscosity (η_s) was measured by a capillary viscosimeter with an error of 1–2%. We measured the values of c, α, ρ, and η_s along the equilibrium curve within the temperature range 283–343 K. The objects of investigation were prepared using a UZDN-1 ultrasonic disperser directly before performing the experiments. This made it possible to obtain disperse systems with reproducible parameters. The measurements were carried out in the range of solid-phase concentrations of 0.5–10 vol.%.

The experimentally determined dependences of absorption α/f^2 on the volumetric concentration φ are linear only in the region of small values of φ ($\varphi \simeq 3$ vol.%). In the region of $\varphi > 3$ vol.% these functions are nonlinear.

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TABLE 1. Dependence of the Properties of a Disperse System of PPG-425 in Aerosil (A-300) on the Temperature and Concentration

T, K	ρ, kg/m^3	$\eta_s \times 10^3$, Pa·sec	C, m/sec	A_1	A_2	B	$\alpha_{cl} f^{-2}$	f_1	f_2	b_1	b_2	$\bar{R}_1 \cdot 10^9$, m
				10^{15}, m^{-1}·sec^2				MHz		10^3		
$\varphi = 1\%$												
283	1023.3	204.3	1425	–	–	–	1784	–	–	–	–	–
293	1019.5	107.6	1398	854	900	360	1016	20.0	280	7.6	111	–
303	1008.2	57.3	1371	668	610	220	583	24.5	390	7.1	105	–
313	1000.7	33.1	1344	500	350	179	267	30.0	670	6.4	100	800
333	985.51	13.8	1290	180	210	120	171.5	50.0	1196	3.7	92.3	–
$\varphi = 5\%$												
283	1056.6	362	1428	–	–	–	3094	–	–	–	–	–
293	1043.4	216	1402	1350	1600	340	1994	18.6	260	11.2	186	2750
303	1036.2	100	1376	990	750	240	974	22.2	480	9.6	158	1300
313	1029.0	57	1350	720	480	185	592	27.0	660	8.3	136	700
343	1007.6	20.0	1272	320	240	110	253	56.0	1240	7.2	120	300
$\varphi = 10\%$												
293	1072.2	320	1408	–	–	–	2812	–	–	–	–	
303	1064.6	176	1383	–	–	–	1644	–	–	–	–	
313	1057.0	124	1358	1800	840	400	1232	5.1	270	4.0	98	
323	1049.4	88	1333	840	560	380	931	7.7	380	2.7	90	
333	1041.6	58	1308	420	400	350	654	10.5	480	1..8	80	

In the disperse systems investigated the absorption of sound depends on the frequency, i.e., acoustic relaxation is observed. The separation of the regions of dispersion (the determination of their number and parameters) was carried out on the basis of a nonlinear version of the least-squares method using statistical criteria of conformity of experimental and predicted values.

The method of processing is basically as follows. Suppose there are several values ($i > 3$) of the absorption quantities ($y_i = \alpha_i / f_i^2$) for the corresponding frequencies (f_i). Assuming that the dependence on the frequency has the form

$$y(f_i) = \frac{\alpha}{f^2} = \sum_{j=1}^{n} \frac{A_i}{1 + (f_i/f_j)^2} + B,$$

we determined the coefficients A_j, f_j, and B of this approximating function for the optimum correspondence with experimental values, i.e., with points having the coordinates f_i and y_i.

The acoustic spectra of the investigated disperse systems, just as of PPG-425, consist of two simple regions of dispersion. The dependence of α/f^2 on the frequency is described by an equation [6, 7] that takes into account two relaxation times:

$$\alpha f^{-2} = \frac{2\pi^2 C}{C_0^2} \left(\frac{b_1 \tau_1}{1 + (\omega\tau_1)^2} + \frac{b_2 \tau_2}{1 + (\omega\tau_2)^2} \right) + B.$$

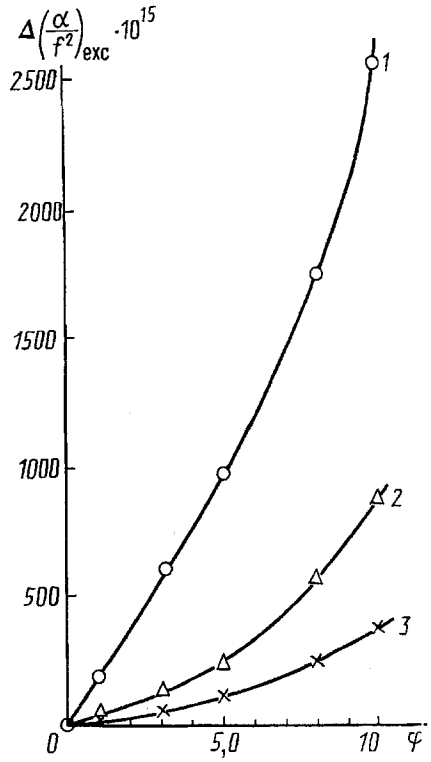

Fig. 1. Excess absorption as function of the concentration in a disperse system of aerosil and PPG-425 at $T = 293$ and the frequencies 5.0 MHz (1), 15.0 (2), and 90 (3). $\Delta(\alpha/f^2)_{exc} \cdot 10^{15}$, $m^{-1} \cdot sec^2$.

Here C is the speed of sound at the frequency $\omega = 2\pi f$; b_i and τ_i are the relaxation forces and times of the i-th region of dispersion ($i = 1, 2$); B is the high-frequency limit ($\omega\tau_2 >> 1$) of the quantities; C_0 is the speed of sound at frequencies $\omega\tau_1 << 1$.

The values of the quantities that characterize the simple regions of dispersion of the quantities αf^{-2} are listed in Table 1. Figure 1 presents the excess absorption ($\Delta(\alpha/f_2)$) as a function of the concentration in comparison to absorption in PPG-425 for waves of different frequencies. In the region of frequencies above 400 MHz the excess absorption for concentrations $\varphi \leq 5\%$ is equal zero to within the experimental error. From this it follows that with an increase in the concentration of the aerosil in the system investigated the excess absorption increases. This allows us to assume that the increase in absorption is attributable to clustering of the particles. As a result, the magnitude of absorption and the acoustic spectrum of the system investigated are determined by the parameters of a cluster.

In [1–5] it is shown that the excess absorption of sound in disperse systems is basically determined by the friction of clusters against the fluid. The functional relationship between the absorption and the dimensions of the clusters has the form [4, 5]

$$\frac{\alpha}{f^2} = A\frac{3.14}{Cf}\left(\frac{R}{R_1}\right)^{0.67}\left(\frac{2.2}{\rho_0}-1\right)^2 \frac{Y(1+\sqrt{Y})}{(1+\sqrt{Y})^2 + Y\left[1+\frac{0.98}{\rho_0}\left(\frac{R}{R_1}\right)^{0.67}\sqrt{Y}\right]}.$$

Here A is a factor that takes into account the dimensionality of the quantities in the equation; $y = \pi f \rho_0 R^2/\eta_0$; ρ_0 is the density of the system; R is the radius of the particles; R_1 is the radius of a cluster; C is the speed of sound at the frequency f; η_0 is the shear viscosity coefficient of the dispersion medium.

The mean size of a cluster R_1 is calculated using a computer. The results of a calculation are given in Table 1. With account for the assumptions underlying the computational model, the values of the mean radius of the clusters found by means of the acoustic spectroscopy method are in good agreement with those obtained by an optical method. It can be seen from Table 1 that the size of a cluster depends on the temperature.

It follows from experimental data that in a disperse system of aerosil and PPG-425 the amplitude of the second region of acoustic dispersion $A_2 + B$ exceeds the absorption $\alpha_{cl} f^{-2}$ attributable to the coefficient of shear viscosity. The value of the quantities $\alpha_{cl} f^{-2}$ is calculated from the equation

$$\frac{\alpha_{cl}}{f^2} = 26.3 \frac{\eta_s}{\rho C_0^3}.$$

The inequality $B < \alpha_{cl}/f^2$ indicates that relaxation of the volumetric viscosity (η_v) is accompanied by relaxation of the shear viscosity. Since in this case the dependence of the quantity α/f^2 (the second region of relaxation) on the frequency is described by an equation with one time of relaxation; the times of relaxation of the volumetric and shear viscosities are indistinguishable within the limits of experimental errors.

The frequency dependence of the absorption of sound for the second region of relaxation coincides with the analogous dependence for PPG-425. Consequently, the second region of relaxation in the objects investigated, just as in PPG-425, is caused by processes of breakdown and formation of intermolecular bonds.

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