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CALCULATION OF THE SOUND VELOCITY AND THE ABSORPTION FACTOR IN LIQUIDS WITH QUASI-SPHERICAL MOLECULES

A short review of theoretical and experimental works dealing with the research of acoustic parameters of some classical liquids and their solutions is made. On the basis of the analytical expressions obtained in the framework of molecular-kinetic theory for the sound velocity $c(\nu)$ *and the absorption factor* $\alpha(\nu)$*, as well as the choice of a model for the potential of intermolecular interaction* $\Phi(|\mathbf{r}|)$ *and the radial distribution function* $g(|\mathbf{r}|)$*, the frequency dispersion of acoustic parameters for liquid* N_2*,* O_2*, and* CH_4 *was numerically calculated in wide intervals of density and temperature.*

Keywords: sound velocity, sound absorption coefficient, translational and structural relaxations, friction coefficient, density, temperature, frequency.

The propagation velocity and the absorption coefficient of sound waves in a liquid are those parameters, which directly depend on the liquid structure. The mentioned quantities are studied as functions of the temperature, pressure, density, frequency, and so on. In many cases, the absorption and the velocity of sound comprise an important tool to study rapid molecular processes. The frequency dispersion of the propagation velocity and the absorption coefficient of sound waves in liquids can be expressed in terms of dynamic elastic moduli and transport coefficients. On the basis of relevant data on the dispersion, it is possible to obtain a valuable information concerning the molecular structure of a liquid and the interparticle interaction in it, to study the kinetics of irreversible processes running in liquids under an external action, and to reveal specific features in the character of relaxation processes in them.

In classical work [1], the Stokes–Kirchhoff, Mandelshtam–Leontovich, and Kneser theories, and some general relaxation theories were reviewed, as well as results of experimental measurements of the coefficient of ultrasonic wave absorption in liquids, which is related to the structure and the transport coefficients in liquids. It was noticed that the frequency dependence of the absorption coefficient (and, on the same footing, the sound velocity) can actually turn out much more complicated, because a variety of relaxation processes with different relaxation times can run in liquids.

In works [2, 3], the sound velocity in liquids was studied in the framework of molecular theory. The corresponding problem was formulated for the model of one-dimensional liquid, which allowed the sound velocity to be accurately expressed in terms of the molecular parameters of a liquid. In their further researches, the cited authors generalized the problem to a three-dimensional model of liquid represented as a system of hard noninteracting spheres,

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which can be characterized by an arbitrary density value.

The detailed reviews of researches aimed at studying the liquid structure, the nature of internal relaxation processes, the transfer phenomena, the acoustic properties of liquids, and the corresponding results obtained in the framework of the general theory of relaxation processes by applying temporal correlation functions, projection operators, kinetic equations, nonequilibrium static operators, and, mainly, the molecular acoustics method were presented in works [4–6]. The results of measurements of the ultrasound velocity and the absorption coefficient in some viscous liquids and their dependences on the pressure (at 1–1000 atm) and temperature (at 8–50°C) were discussed in work [7], where the dependences of the relative variation of relaxation times were plotted. It was pointed out that a growth of the relaxation time under the pressure action may probably by typical of only the structural relaxation.

In works [8, 9], the sound velocity in liquid carbon dioxide was measured in the temperature interval from the ternary point to 293 K along the liquid phase saturation curve. Numerical results which were obtained for the adiabatic compressibility β_S, the specific heat capacities C_P and C_V, the isobaric coefficient of thermal expansion α_P, and the temperature coefficient of pressure γ_V in the indicated temperature interval are in a satisfactory agreement with the literature data. On the basis of sound wave absorption measurements, the frequency dispersion of the bulk viscosity coefficients of liquid N_2, CO, CH_4, and CD_4 in the temperature interval from 77 to 300 K were determined. A relation between the rotational relaxation times was established, and the corresponding kinetic transverse cross-section was found.

Using the reverberation technique [10], the absorption coefficient of ultrasonic waves in cottonseed and linseed oils was measured in vessels 0.5 l in volume in the frequency interval from 50 kHz to 4 MHz. The relaxation of a bulk viscosity was found in both oils. The quantity α/ν^2 was demonstrated to become constant at frequencies below 50 kHz.

The main results of researches concerning the temperature dependences of the propagation velocity and the absorption coefficient for longitudinal and transverse hypersounds in liquids obtained by analyzing the spectra of molecular light scattering were reported in work [11]. Using the shift and the width of Mandelstam–Brillouin components, the velocity and absorption coefficient of hypersound 10^9–10^{10} Hz in frequency were measured in glycerol, benzophenone, salol, and β,β'-dichlorodiethyl ether in a wide interval of temperatures. The results were compared with the data of numerical calculations according to the formula derived for the sound velocity and the absorption coefficient in the framework of a nonlocal diffusion theory, and a satisfactory agreement was obtained. The character of transverse hypersound propagation was studied by analyzing the spectra with a fine structure of the Rayleigh line wing obtained in salol and benzophenone in the geometry, when the exciting light was polarized in the scattering plane, and the scattered light normally to it. The obtained results and their comparison with experimental data show that the molecular light scattering is a convenient method to study the propagation of longitudinal and transverse high-frequency sounds in liquids.

The sound absorption coefficient, the bulk viscosity, and the relaxation times in liquid nitrogen (N_2) and oxygen (O_2) were studied in work [12]. The sound absorption in liquid N_2 and O_2 was measured at frequencies of 5–145 MHz along the saturation curve in a wide interval of temperatures, and the values of bulk viscosity were calculated. The vibrational relaxation does not affect experimental results in this case, and the presence of rotational degrees of freedom leads to a certain acceleration of the relaxation over the momenta (the translational one). On the basis of the theory of effective collisions, it was shown that the "forgetting" time of a coordinate at low temperatures substantially exceeds the period between "ordinary" collisions.

In work [13], the absorption coefficient α and the sound velocity C were measured in liquid methane (CH_4) and tetrafluoromethane (CF_4) along the saturation curve at frequencies from 5 to 126 MHz and temperatures from 90 to 210 K. In CF_4, the dispersion of the quantity α/ν^2 was observed in the whole examined temperature and frequency regions, and the sound velocity revealed a weak dispersion at low temperatures. In CH_4, the dispersion of α/ν^2 was observed only at $T > 140$ K. In the whole temperature region, the absorption considerably exceeded dissipative losses associated with external degrees of freedom of the thermal motion. The analysis of the results obtained brings about a conclusion that the phenomena observed in both substances are related to the excitation of intramolecular vibrational degrees of freedom. The obtained characteristics of the vibrational relaxation testify to a different kinetics of this process in each of the examined liquids, which is associated with the different characters of a rotational motion of particles.

The researches of the acoustic properties of classical liquids and their solutions with regard for the contributions of various internal relaxation processes were successfully carried out in recent decades as well. The frequency dispersion and the temperature dependence of ultra- and hypersound velocities in aqueous solutions of electrolytes and mixed solutions were studied in works [14–16]. In those works, along with the temperature dependence, the negative dispersion of the sound velocity in a narrow interval of hypersound frequencies was revealed. The calculated deviations of the sound velocity in a mixed solution from the corresponding value in pure water are in good agreement with the corresponding experimental data.

In work [17], the propagation of acoustic waves in chemically nonequilibrium media was considered. It was shown that, in this case, the inversion of the frequency dispersion of the velocity and the sound absorption, as well as the second viscosity, is possible.

In works [18–21], the analytical expressions for the velocity and the absorption coefficient of sound waves in classical liquids with regard for the contributions of internal relaxation processes were derived in the framework of molecular-kinetic theory. For a certain choice of the potential energy of interaction and the radial distribution function, numerical calculations of the dependences of those acoustic parameters on the density, temperature, concentration, and frequency were carried out. The obtained results are in a satisfactory agreement at the quantitative level with experimental data.

This work is aimed at calculating the velocity c and the absorption factor α/ν^2 for liquids N_2, O_2, CH_4 of sound in a wide interval of density ρ, temperature T, and frequency ν, as well as at comparing the obtained results with experimental data. Here, for the potential of intermolecular interaction $\Phi\left(|\mathbf{r}|\right))$ and the radial distribution function $g\left(|\mathbf{r}|\right)$, we use the most optimum initial model considered in work [22].

In work [21], the following analytical expressions were derived for the velocity $C(\omega)$ and the absorption coefficient $\alpha(\omega)$ of sound waves:

$$\begin{aligned} C\left(\omega\right) &= C_0\left\{1+\frac{1}{2\rho C_0^2}\left[\frac{4}{3}\mu\left(\omega\right)+\mathrm{K}_r\left(\omega\right)\right]\right\},\\ \alpha\left(\omega\right) &= \frac{\omega^2 C}{2\rho C_0^4}\left[\frac{4}{3}\eta_S\left(\omega\right)+\eta_V\left(\omega\right)\right], \end{aligned} \tag{1}$$

where $c_0=\sqrt{K_S/\rho}$ is the adiabatic sound velocity, ρ the density of a liquid, $\gamma = C_P/C_V$, $K_r(\omega) = K(\omega) - K_S$, and

$$K_S = \rho\left(\frac{\partial P}{\partial \rho}\right)_T + \frac{T_0}{\rho C_V}\left(\frac{\partial P}{\partial T}\right)_\rho^2$$

is the adiabatic bulk elastic modulus. The expressions obtained describe the dispersions of the sound velocity and the absorption coefficient in liquids in wide intervals of thermodynamic state parameters and frequencies, and make allowance for the translational and structural relaxations.

For numerical calculations, let us apply the following analytical expressions derived for the dynamic coefficients of bulk, $\eta_V(\omega)$, and shear, $\eta_S(\omega)$, viscosities in a liquid and for the dynamic elastic moduli of a liquid $\mu(\omega)$ and $K(\omega)$ [21]:

$$\begin{aligned} K(\omega) = K_s + \frac{2\pi n^2\sigma^3\omega}{3}\int\limits_0^\infty dr\ r^3\ \frac{\partial\Phi(r)}{\partial r}\times\\ \times\int\limits_{-\infty}^{\infty} G_2(r,\ r_1,\omega)\varphi_0(r_1)d\mathbf{r}_1, \end{aligned} \tag{2}$$

$$\begin{aligned} \mu\left(\omega\right) = \frac{nkT(\omega\tau)^2}{1+(\omega\tau)^2} + \frac{2\pi n^2\sigma^3\omega}{15}\int\limits_0^\infty dr\, r^3\ \frac{\partial\Phi(r)}{\partial r}\times\\ \times\int\limits_{-\infty}^{\infty} G_2(r,\ r_1,\omega)\frac{\partial g(r_1)}{\partial r_1}\ r_1 d\mathbf{r}_1, \end{aligned} \tag{3}$$

$$\begin{aligned} \eta_V(\omega) = \frac{2\pi\, n^2\, \sigma^3}{3}\int\limits_0^\infty dr\, r^3\ \frac{d\Phi(|\mathbf{r}|)}{dr}\times\\ \times\int\limits_{-\infty}^{+\infty} G_1(r,r_1,\omega)\ \varphi_0(|\mathbf{r}_1|)\, d\mathbf{r}_1 \end{aligned} \tag{4}$$

$$\begin{aligned} \eta_S(\omega) = \frac{n\,k\,T\,\tau}{1+(\omega\tau)^2} + \frac{2\pi\, n^2\, \sigma^3}{15}\int\limits_0^\infty dr\, r^3\ \frac{d\Phi(|\mathbf{r}|)}{dr}\times\\ \times\int\limits_{-\infty}^{+\infty} G_1(r,r_1,\omega)\ \frac{\partial g(|\mathbf{r}_1|)}{\partial r_1}\ r_1\, d\mathbf{r}_1, \end{aligned} \tag{5}$$

where

$$\varphi_0(|\mathbf{r}_1|) = \frac{r_1}{3}\frac{\partial g(r_1)}{\partial r_1} - \left[n\left(\frac{\partial g(r_1)}{\partial n}\right)_T + \gamma T\left(\frac{\partial g(r_1)}{\partial T}\right)_n\right],$$

$$\gamma = \frac{1}{nc_V}\left(\frac{\partial P}{\partial T}\right)_\rho,$$

$$G_{1,2}(r, r_1, \omega) = \pm\frac{\tau_0(2\omega\tau_0)^{-1/2}}{4\pi\, r\, r_1}\times$$

$$\times\left[e^{-\varphi_1}\left(\sin\varphi_1 \mp \cos\varphi_1\right) - e^{-\varphi_2}\left(\sin\varphi_2 \mp \cos\varphi_2\right)\right],$$

$$\varphi_1 = \varphi_1(r, r_1, \omega) = \left(\frac{\omega\tau_0}{2}\right)^{-1/2}(r - r_1);$$

$$\varphi_2 = \varphi_2(r, r_1, \omega) = \left(\frac{\omega\tau_0}{2}\right)^{1/2}(r + r_1),$$

$\tau = m/(2\beta)$ and $\tau_0 = \beta\sigma^2/(2kT)$. In the formulas above, m, σ, and $n = \frac{N}{V}$ are the mass, diameter, and concentration, respectively, of particles in a liquid; $\mathbf{r}_{12} = \mathbf{q}_2 - \mathbf{q}_1$ and $r = \mathbf{r}_{12}/\sigma$ are the relative and reduced distances between them; β is the friction coefficient; k the Boltzmann constant, T the absolute temperature, and $\omega = 2\pi\nu$ is the cyclic frequency of the process. It should be noted that expressions (2) and (3) for the dynamic elastic moduli and expressions (4) and (5) for the viscosity coefficients were obtained for liquids with spherical molecules. The parameter of eccentricity for liquids with quasi-spherical molecules is known to be small [23]. However, the referral of those liquids to Lennard-Jones ones can be made only in a certain approximation. Hence, let us use expression (1) and, taking Eq. (2)–(5) into account, numerically calculate $c(\nu)$ and $\alpha(\nu)$ for N_2, O_2, and CH_4 in wide intervals of density, temperature, and frequency.

For this purpose, according to expressions (1)–(5), we need to know the molecular parameters of a liquid such as the mass m and the diameter σ of a particle, the potential well depth ε, which is known from the literature [24–28], and the relaxation times τ and τ_0, which can be determined through the coefficient of friction in a liquid β (the latter cannot be determined in the framework of the considered theory). The right-hand sides of kinetic equations, which are responsible for the irreversibility of those equations in time (in our case, this is the Fokker–Planck collision operator), describe the dissipation processes in liquids and contain the friction coefficient β. In work [29], by determining the autocorrelation function of momenta and finding the average force acting on the probe particle moving at a constant velocity, the following expression for β was obtained:

$$\beta^2 = (4\pi/3)\rho\sigma\int\limits_0^\infty \nabla^2\Phi\left(|\mathbf{r}|\right)\, g\left(|\mathbf{r}|\right) r^2 dr, \tag{6}$$

where ρ is the density of a liquid, and $\nabla^2 = \frac{1}{r^2}\times \times\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right)$ the radial part of the Laplace operator. Hence, the study of the origin and the determination of the dependences of the coefficients β, $c(\nu)$, and $\alpha(\nu)$ on the state parameters ρ, T, and P requires a choice of the potential $\Phi\left(|\mathbf{r}|\right)$ and the radial distribution function $g\left(|\mathbf{r}|\right)$. Let us select the most optimal initial model considered in work [22]:

$$\Phi\left(|\mathbf{r}|\right) = \begin{cases}\infty & \text{if } r < 1,\\ 4\varepsilon\left(r^{-12} - r^{-6}\right) & \text{if } r \geqslant 1,\end{cases} \tag{7}$$

$$g\left(|\mathbf{r}|\right) = y\left(\overset{*}{\rho}\right)\, \exp\left(-\Phi\left(|\mathbf{r}|\right)/kT\right), \tag{8}$$

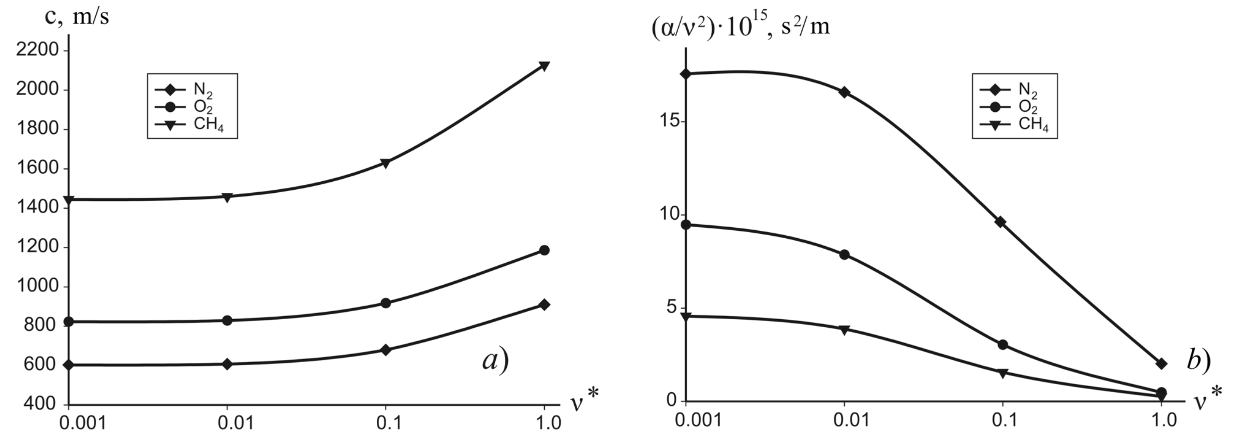
where $y(\overset{*}{\rho}) = (2-\overset{*}{\rho})/[2(1-\overset{*}{\rho})^3]$ is the Carnahan–Starling function, $\rho^* = (\pi/6)\, N_0\sigma^3\rho/M$ is the reduced density, N_0 the Avogadro constant, M the molar mass, and ρ the density of a liquid.

Expressions (7) and (8) make it possible to perform a theoretical research and to execute numerical calculations of the friction coefficient β, the relaxation times defined as $\tau = m/(2\beta)$ and $\tau_0 = \beta\sigma^2/(2kT)$, the velocity $c(\nu)$, and the absorption coefficient $\alpha(\nu)$ of sound waves in liquids in a wide interval of the frequency $\nu = \omega/2\pi$ as functions of the thermodynamic state parameters (ρ, T). The results of numerical calculations of the frequency dependences of the velocity $c(\nu)$ and the absorption coefficient $\alpha(\nu)$ of sound waves on the state parameters are given in Tables 1 and 2, respectively.

Hence, the theoretical calculations of the sound velocity $c(\nu)$ and the sound absorption coefficient α/ν^2 in liquid N_2, O_2, and CH_4 give the numerical values that are in a satisfactory agreement with experimental data. An insignificant difference between them may probably follow from the neglecting of contributions inserted by orientational angles to the interparticle interaction potential. As one can see from Tables 1 and 2, as well as from Figure, the region of frequency dispersion of the quantities $c(\nu)$ and α/ν^2 for sound in the liquids with quasi-spherical molecules (N_2, O_2, and CH_4) is rather wide (about 10^4 Hz). It

Table 1. **Frequency dependence of the sound velocity $c(\nu)$ in liquid nitrogen, oxygen, and methane ($\nu = 10^6 \div 10^{12}$ Hz)**

T, K	ρ, kg/m^3 [12]	$c(\nu)$, m/s							
		c, m/s [12]	$\nu^* = 10^{-6}$	$\nu^* = 10^{-5}$	$\nu^* = 10^{-4}$	$\nu^* = 10^{-3}$	$\nu^* = 10^{-2}$	$\nu^* = 10^{-1}$	$\nu^* = 1$
					nitrogen				
80	794	831	831.00	831.00	831.02	831.25	838.37	937.06	1231.06
90	744	717	717.00	717.00	717.02	717.23	722.49	806.93	1068.87
100	688	603	603.00	603.00	603.02	603.25	607.63	679.88	909.60
110	623	475	475.00	475.00	475.03	475.32	479.57	543.38	744.31
115	581	405	405.00	405.00	405.04	405.38	409.83	470.13	653.29
120	527	318	318.00	318.01	318.05	318.49	323.64	383.63	549.68
					oxygen				
80	1190	986	986.00	986.00	986.02	986.41	997.42	1114.01	1440.60
90	1142	905	905.00	905.00	905.02	905.26	913.01	1013.95	1310.45
100	1090	823	823.00	823.00	823.02	823.22	829.06	917.29	1185.98
110	1035	732	732.00	732.00	732.02	732.22	737.04	815.60	1061.14
120	974	643	643.00	643.00	643.02	643.25	647.50	717.05	937.69
130	903	542	542.00	542.00	542.03	542.30	546.39	608.73	805.27
140	813	422	422.00	422.00	422.04	422.39	426.83	484.73	657.16
146	741	338	338.00	338.01	338.05	338.49	343.61	401.44	557.42
T, K	ρ, kg/m^3 [13]	c, m/s [13]	$\nu^* = 10^{-6}$	$\nu^* = 10^{-5}$	$\nu^* = 10^{-4}$	$\nu^* = 10^{-3}$	$\nu^* = 10^{-2}$	$\nu^* = 10^{-1}$	$\nu^* = 1$
					methane				
100	440	1444	1444.00	1444.00	1444.03	1444.54	1459.62	1632.55	2127.73
120	413	1245	1245.00	1245.00	1245.02	1245.36	1254.93	1398.56	1835.34
140	380	1040	1040.00	1040.00	1040.03	1040.38	1047.56	1166.35	1541.54
150	361	926	926.00	926.00	926.04	926.42	933.14	1041.88	1387.60
160	339	797	797.00	797.00	797.05	797.49	804.16	904.85	1221.85
170	312	646	646.00	646.01	646.06	646.62	653.82	749.82	1039.00
180	277	483	483.00	483.01	483.08	483.84	492.54	588.98	844.61



Dependences of the sound velocity (*a*) and sound absorption coefficient (*b*) on the reduced frequency for liquid N_2, O_2, and CH_4 at $T = 100$ K

Table 2. **Frequency dependence of the sound absorption coefficient α^2/ν in liquid nitrogen, oxygen, and methane ($\nu = 10^6 \div 10^{12}$ Hz)**

T, K	ρ, kg/m^3 [12]	α/ν^2, 10^{-15} s^2/m							
		[12]	$\nu^* = 10^{-6}$	$\nu^* = 10^{-5}$	$\nu^* = 10^{-4}$	$\nu^* = 10^{-3}$	$\nu^* = 10^{-2}$	$\nu^* = 10^{-1}$	$\nu^* = 1$
nitrogen									
80	794	10.3	12.022	12.005	11.949	11.757	10.548	4,697	0.865
90	744	13.6	13.881	13.872	13.843	13.725	12.736	6.448	1.297
100	688	19.9	17.741	17.731	17.697	17.565	16.585	9.496	2.017
110	623	34.2	27.562	27.543	27.482	27.268	26.043	16.692	3.623
115	581	52.6	37.838	37.812	37.731	37.450	36.008	24.411	5.268
120	527	84.0	64.864	64.826	64.705	64.296	62.344	44.650	9.373
oxygen									
80	1190	7.5	9.64	9.62	9.56	9.36	8.27	3.55	0.63
90	1142	7.8	10.00	9.99	9.97	9.86	9.02	4.29	0.83
100	1090	8.7	10.12	10.07	9.93	9.48	7.87	3.04	0.48
110	1035	11.7	11.51	11.50	11.48	11.38	10.61	5.56	1.14
120	974	15.9	13.88	13.87	13.84	13.72	12.93	7.44	1.59
130	903	23.9	18.78	18.76	18.72	18.54	17.62	11.07	2.40
140	813	44.2	31.30	31.27	31.19	30.92	29.67	20.30	4.36
146	741	89.6	51.47	51.44	51.32	50.94	49.27	35.38	7.36
T, K	ρ, kg/m^3 [13]	α/ν^2, 10^{-15} s^2/m							
		[13]	$\nu^* = 10^{-6}$	$\nu^* = 10^{-5}$	$\nu^* = 10^{-4}$	$\nu^* = 10^{-3}$	$\nu^* = 10^{-2}$	$\nu^* = 10^{-1}$	$\nu^* = 1$
methane									
100	440	3.38	4.82	4.80	4.74	4.57	3.88	1.56	0.26
120	413	3.80	5.00	5.00	4.98	4.93	4.50	2.12	0.41
140	380	5.76	6.18	6.18	6.16	6.12	5.74	3.16	0.66
150	361	7.30	7.48	7.47	7.45	7.39	6.99	4.13	0.89
160	339	10.70	9.91	9.90	9.88	9.79	9.31	5.90	1.28
170	312	18.40	15.39	15.37	15.33	15.20	14.57	9.88	2.13
180	277	41.00	29.70	29.68	29.62	29.40	28.47	20.54	4.25

is a result of the power law of damping of relaxing fluxes, which in turn corresponds to the diffusion mechanism. In other words, it is a contribution of the structural relaxation, when, according to the exponential law of damping, on the basis of the general relaxation theory [4, 5], the region of dispersion of the acoustic parameter is narrow (about 10^2 Hz).

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РОЗРАХУНОК ШВИДКОСТІ І КОЕФІЦІЄНТА ПОГЛИНАННЯ ЗВУКУ В РІДИНАХ З КВАЗІСФЕРИЧНИХ МОЛЕКУЛ

Резюме

Наведено короткий огляд теоретичних та експериментальних робіт з дослідження акустичних параметрів класичних рідин і їх розчинів. На основі аналітичних виразів швидкості і коефіцієнта поглинання звуку, отриманих згідно з молекулярно-кінетичною теорією і вибором моделі для потенціалу міжмолекулярної взаємодії і радіальної функції розподілу, проведений чисельний розрахунок частотної дисперсії акустичних параметрів рідин N_2, O_2, CH_4 в широкому інтервалі зміни густини і температури.