FREQUENCY DISPERSION OF THE SHEAR AND BULK VISCOSITY COEFFICIENTS OF LIQUID ARGON AS FUNCTIONS OF STATE PARAMETERS

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Experimental and theoretical works studying the shear η_S and bulk η_V viscosity coefficients of simple liquids and the optimal choice of the intermolecular interaction potential $\Phi(|\mathbf{r}|)$ and the radial distribution function $g(|\mathbf{r}|)$ are briefly analyzed. The last are used to determine the friction coefficient β and the relaxation times τ and τ_0 as functions of the temperature T and the density ρ, as well as to numerically calculate the viscosity coefficients $\eta_S(\omega)$ and $\eta_V(\omega)$ in wide ranges of state parameters and frequencies. The obtained results are in satisfactory quantitative agreement with the theoretical and experimental data for liquid argon reported in the literature.

1. Introduction

Investigations of the structure and nonequilibrium properties of liquids are closely related to the study of the nature of internal relaxation processes and transport phenomena in them. The last are accompanied by dissipative phenomena described with the help of transport coefficients and corresponding elastic moduli. The most extensive experimental researches were performed for such static and dynamic transport characteristics as the coefficients of diffusion, shear viscosity, heat conduction, and electrical conduction, as well as the acoustic parameters such as the velocity and the absorption coefficient of sound waves in a wide range of thermodynamic state parameters and frequencies [1–3]. However, the bulk viscosity coefficient and the moduli of shear and bulk elasticities, as well as thermo- and electroelasticities, cannot be determined by direct experimental measurements. They are found indirectly by measuring other coefficients or physical parameters. For example, the bulk viscosity coefficient η_V is determined from measurements of the excess absorption of sound waves in liquids [4–8]. Due to uncertainties of measuring the sound absorption coefficient α, as well as errors in the used data on thermodynamic and kinetic properties of liquids (densityρ, velocity of sound C, heat capacities c_P and c_V, shear viscosity η_S, and thermal conduction λ), the accuracy of determination of η_V in liquids [4–6] is insufficient to obtain clear empiric regularities and to compare them with theoretical data.

The experimental values of η_V and η_S obtained in [7] for liquid argon at $\nu = 5 \times 10^6$ Hz were compared to the theoretical results [3] which are due mainly to two contributions: one of them is caused by the “hard” part of the interaction potential at the interparticle distance $r = \sigma$, while the other “structural” contribution is due to the long-range forces and is also related to the distortion of the pair distribution function at the distances $r > \sigma$. The experimental values of η_V and η_S decrease, by saturating at large densities ρ (low T and high P). Moreover, the temperature interval grows with the pressure P. The region, the function $\eta_V(T)_p$ decreases, is longer and sharper in the case of higher isobars ($P =$ const), which results in their intersection with one another and with the boundary curve $\eta_V(T)_H$.

It is shown that, at $T \geq 100$ K, the saturation curve of liquid argon manifests the dependence of α/ν^2 on the frequency ν, that is the frequency dispersion of the sound absorption is caused by the contributions of the kinetic viscosity coefficients, whose study is of high interest.

The authors of work [7] made an attempt to determine and analyze the interrelation between the characteristic relaxation times of the bulk and shear viscosities (τ_V and τ_S, respectively) and the equilibration times in the configuration and momentum parts of the phase space (τ_q and τ_p). The high-frequency expansions of the momentum flux autocorrelators in the Green–Kubo relations for the viscosity coefficients [9] yield the limit values of the bulk (K_0, K_∞) and the shear (μ_∞) elastic moduli used for the determination of the relaxation times τ_V and τ_S. The latter represent the decay times of the autocorrelators for the diagonal and non-diagonal elements of the momentum flux tensor in the exponential approximation. We now follow [10], by assuming that the autocorrelators can be expanded into the independently relaxing potential and kinetic parts and that the times τ_q and τ_p determine the exponential decay of each part. Then, according to [7], we obtain that the viscosity coefficients η_V and η_S can be expressed in terms of the same decay times τ_q and τ_p. The authors also tried to independently estimate the latter considering τ_q as the decay time of the mean force autocorrelator $\tau_q \approx \tau_{\langle |F^2| \rangle} \approx \frac{3\,k\,T}{\langle |F^2| \rangle}\,\beta$ and deriving τ_p from the definition of the self-diffusion coefficient under the assumption about the exponential time dependence of the velocity autocorrelator, $\tau_P \approx \tau_{\langle |v^2| \rangle} \approx m/\beta$. In this case, in order to determine τ_q and τ_p, one needs to know the dependence of the friction coefficient β of the liquid on the state parameters (temperature T, density ρ, and pressure P) that remains unclear till now.

The molecular dynamics method [11–14] was used to numerically calculate the frequency-dependent kinetic coefficients and the corresponding elastic moduli of simple liquids in a wide range of thermodynamic state parameters and frequencies. The low-frequency behavior of the transport coefficients of simple liquids is characterized by the “cuspidal point” singularity (i.e. asymptotics) proportional to $\omega^{1/2}$. Analyzing this singularity, one can see that the time autocorrelation function of microscopic stresses has “far tails” of the $t^{-3/2}$ type at large times and that the amplitude of this tail considerably exceeds the value following from theoretical estimates of the corresponding kinetic contributions.

The theoretical study of transport phenomena, elastic and acoustic properties of liquids based upon both model and molecular-kinetic theories was the subject of numerous works [1–3, 15–17], where analytical expressions for transport coefficients of liquids and their elastic and acoustic parameters were obtained with the use of various approximations. The results contain the contributions of internal relaxation processes and are given in terms of the potential of interaction between liquid structural units, the radial distribution function, and other molecular parameters of the medium.

In [18], the kinetic equations for the one- and two-particle distribution functions were used to obtain the analytical expressions for the dynamic shear $\eta_S(\omega)$ and bulk $\eta_V(\omega)$ viscosity coefficients and to perform numerical calculations for liquid argon in a wide frequency range. However, the value of the friction coefficient β was supposed constant in all these calculations ($\beta = 2.85 \times 10^{-13}$ kg/s), i.e., its dependence on the thermodynamic state parameters was not taken into account. The density dependence in the radial distribution function was also neglected, because we were interested in the frequency behavior of the kinetic coefficients. In order to improve the agreement between the theoretical calculations and the experimental results obtained for the coefficients β, η_S, and η_V, as well as for the relaxation times of the stress tensor in the momentum and configuration spaces, it is necessary to choose the real intermolecular interaction potential $\Phi\left(|\mathbf{r}|\right)$ and the radial distribution function $g\left(|\mathbf{r}|\right)$ in the optimal way, but this problem remains else unsolved.

In [19, 20], the friction coefficient β, the relaxation times τ and τ_0, and the shear viscosity coefficient $\eta_S(\omega)$ for liquid argon were theoretically calculated in wide temperature and density ranges by choosing the optimal form of two modified Lennard-Jones potentials and the Buckingham potential and by taking the temperature and density dependences of the radial distribution function into account. The obtained results were compared with the theoretical data reported in [3] and experimental results [7]. The analysis of the numerical calculations of the shear viscosity $\eta_S(\omega)$ shows that the most satisfactory agreement between the theoretical and experimental results is obtained with the modified intermolecular interaction potential for real liquids proposed by J. Rowlinson in [3], where the attraction potential was chosen almost twofold less than the r^{-6} coefficient in the Lennard-Jones potential. Based upon this optimal model, we will study the dynamic shear $\eta_S(\omega)$ and bulk $\eta_V(\omega)$ viscosity coefficients in wide temperature and density ranges, whose numerical values are borrowed from [7]. Moreover, the parameters present in the potential parts of the coefficients $\eta_S(\omega)$ and $\eta_V(\omega)$ (the friction coefficient β, the relaxation time of the stress tensor in the momentum space τ, and the phenomenological parameter τ_0 that represents an analog of the relaxation time of a diffusing molecule) are self-consistently determined on the basis of the given model as functions of ρ and T.

2. Choice of Model and Initial Expressions for Viscosity Coefficients

The initial expressions will be chosen as the following analytical relations for the dynamic shear $\eta_S(\omega)$ and bulk $\eta_V(\omega)$ viscosities coefficients obtained on the basis of the kinetic equations for the one- and two-particle distribution functions with regard for the space correlation of the density in the configuration space [17, 18]:

$$\eta_S(\omega)=\frac{n\,k\,T\,\tau}{1+(\omega\tau)^2}+\frac{2\,\pi\,n^2\,\sigma^3}{15}\int\limits_0^\infty dr\;r^3\,\frac{d\Phi\,(|\mathbf{r}|)}{dr}\times$$

$$\times\int\limits_{-\infty}^{+\infty}G_1(r,r_1,\omega)\,\frac{\partial g(|\mathbf{r}_1|)}{\partial r_1}\;r_1\,d\mathbf{r}_1\,, \qquad (1)$$

$$\eta_V(\omega)=\frac{2\pi\,n^2\,\sigma^3}{3}\int\limits_0^\infty dr\;\;r^3\,\frac{d\Phi\,(|\mathbf{r}|)}{dr}\times$$

$$\times\int\limits_{-\infty}^{+\infty}G_1(r,r_1,\omega)\;\varphi_0(|\mathbf{r}_1|)\;d\mathbf{r}_1, \qquad (2)$$

$$G_1(r,r_1,\omega)=\frac{\tau_0(2\,\omega\,\tau_0)^{-1/2}}{4\pi\,r\,r_1}\Big[e^{-\varphi_1}(\sin\varphi_1-\cos\varphi_1)-$$

$$-e^{-\varphi_2}(\sin\varphi_2-\cos\varphi_2)\Big], \qquad (3)$$

where

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

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

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

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

$\tau=m/(2\beta)$; $\tau_0=\beta\,\sigma^2\,/2kT$. Here, m, σ, $n=\frac{N}{V}$, $\mathbf{r}_{12}=\mathbf{q}_2-\mathbf{q}_1$, and $r=\mathbf{r}_{12}/\sigma$ are the mass, diameter, density, distance and reduced distance between the liquid particles, respectively, β is the friction coefficient, k is the Boltzmann constant, T is the absolute temperature, and ω is the cyclic frequency of the process.

Formulas (1)–(3) describe the dynamic behavior of the viscosity properties of liquids in a wide frequency range. The first (kinetic) term of the shear viscosity $\eta_S(\omega)$ takes into account the contribution of the translational relaxation, i.e. the relaxation of the viscous stress tensor in the momentum space, whose characteristic time is equal to $\tau=m/(2\beta)$. The frequency dependence of the viscosity coefficients $\eta_S(\omega)$ and $\eta_V(\omega)$ is mainly described by means of the space-time behavior of the fundamental solution (Green function) of the Smoluchowski equation for the binary density in the configuration space $G_1(r,r_1,\omega)$.

The function $G_1(r,r_1,\omega)$ is a complex mathematical expression and characterizes the recovery of the equilibrium structure in the configuration space according to the diffusion law, but it does not allow one to explicitly separate the characteristic relaxation time. The process of structural relaxation has a continuous time spectrum, so an analog of the time τ_q from [7] can be obtained only in the approximation of the exponential decay of relaxing flows. The phenomenological parameter τ_0 enters (3) in a complex way. The presence of the function $G_1(r,r_1,\omega)$ enables one to determine the values of $\eta_S(\omega)$ and $\eta_V(\omega)$ in the whole frequency range and the asymptotic behavior of these coefficients: they change as $\omega^{1/2}$ at low frequencies (which agrees with the results obtained within the molecular dynamics method [11–14]) and as ω^{-1} at high frequencies.

According to expressions (1)–(3), the coefficients $\eta_S(\omega)$ and $\eta_V(\omega)$ can be determined if the molecular parameters of the liquid such as the mass m, the particle diameter σ, and the potential well depth ε are known. However, the study of the viscosity coefficients $\eta_S(\omega)$ and $\eta_V(\omega)$ on the basis of the kinetic equations meets some difficulties concerning the friction coefficient β that cannot be determined in the framework of the considered theory. The right-hand sides of the kinetic equations that ensure their time irreversibility (in our case, the Fokker–Planck collision operator) describe the dissipative processes in liquids and include the friction coefficient β. In [3], the friction coefficient was derived by finding the autocorrelation momentum function and the mean force acting on a trial particle moving with constant velocity and had the form

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

where ρ is the liquid density, $\nabla^2=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial}{\partial r}\right)$ is the radial part of the Laplace operator, $\Phi\,(|\mathbf{r}\,|)$ is a modified

Table 1

T, R	ρ, kg/m^3	P, atm	η_S, mPa·s			η_V, mPa·s			η_V/η_S		
			[3]	[7]	this work	[3]	[7]	this work	[3]	[7]	this work
90	1380	1.32	0.171	0.235	0.239	0.252–0.116	0.156	0.231	1.474	0.663	0.967
128	1120	50	0.073	0.096	0.086	0.093	0.170	0.075	1.274	1.771	0.872
133.5	1120	100	0.073	0.084	0.087	0.097	0.165	0.077	1.329	1.964	0.885
185.5	1120	500	0.087–0.077	0.092	0.097	0.112	–	0.089	1.287	–	0.918

Lennard-Jones potential with a strong repulsion term such as the hard sphere potential, and r is the dimensionless mutual distance. Therefore, in order to study the nature and to obtain the dependences of the friction and viscosity coefficients on the state parameters (density, temperature, and pressure), one should choose the optimal intermolecular interaction potential $\Phi\left(|\mathbf{r}|\right)$ and radial distribution function g(r).

Following the results of works [19, 20], we accept the following initial expressions for the potential $\Phi\left(|\mathbf{r}|\right)$ and the function $g\left(|\mathbf{r}|\right)$:

$$\Phi\left(|\mathbf{r}|\right) = \begin{cases} \infty, & \text{at} \quad r < \sigma, \\ 4\varepsilon\left[\, r^{-12} - 0,5\, r^{-6}\right], & \text{at} \quad r \geq \sigma, \end{cases} \tag{5}$$

$$g\left(|\mathbf{r}|\right) = y\left(\rho*\right)\,\exp\,\left(-\Phi\left(|\mathbf{r}|\right)/kT\right), \tag{6}$$

where $y\left(\rho*\right) = (2-\rho*)/2(1-\rho*)^3$ is the Carnahan–Starling function, $\rho* = (\pi/6)\,N_0\,\sigma^3\rho/M$ is the reduced density, N_0 is the Avogadro number, M is the molar mass, and ρ is the liquid density.

Substituting formulas (5) and (6) into expressions (1)–(4), we obtain the following expressions used for the numerical calculation of the coefficients of friction β and viscosity $\eta_S(\omega)$ and $\eta_V(\omega)$:

$$\beta^2 \, = (192M\,\varepsilon/N_0\,\sigma^2)\,\rho^*\times$$

$$\times\int\limits_0^\infty r^{-6}\,\left(22\,r^{-6}\, - 2.5\right)\,g\left(r\right)\,dr, \tag{7}$$

$$\eta_S(\omega) = \,6\varepsilon\,\tau\rho * T^*/\pi\sigma^3(1+\omega*^2) -$$

$$-\frac{(24)^3}{5\pi}\,\frac{\varepsilon\,\tau_0}{\sigma^3}\,\left(\frac{\rho^{*2}}{T^*}\right)\int\limits_0^\infty dr\;\, r^{-5}\,(2r^{-6}-0.5)\times$$

$$\times\int\limits_0^r G_1^*\,(r,\, r_1,\omega) r_1^{-5}\,(2r_1^{-6}-0.5)\,g(r_1)\;dr_{\,1}\,, \tag{8}$$

$$\eta_V(\omega) = \,\frac{48}{\pi}\,\frac{\varepsilon\,\tau_0}{\sigma^3}\,{}_{\rho}\,{*}^2\;\,T\,{*}_{(}\,I_1 + I_2), \tag{9}$$

where

$$I_1 = -\frac{96}{T^{*2}}\,\int\limits_0^\infty dr\;\, r^{-5}\,(2r^{-6}-0.5)\times$$

$$\times\int\limits_0^r G_1^*\,(r,\, r_1,\omega)\; r_1^{-5}\,(2r_1^{-6}-0.5)\,g(r_1)\;dr_{\,1}\,,$$

$$I_2 = \frac{12}{T^*}\,\int\limits_0^\infty dr\;\, r^{-5}\,(2r^{-6}-0.5)\times$$

$$\times\int\limits_0^r G_1^*(r,\, r_1,\omega)\,[y_2(\rho^*)+\gamma\,\Phi^*(r_1)]\;\,g(r_1)\;r_{\,1}\;dr_{\,1}\,,$$

$$G_1^*(r,r_1,\omega) = \frac{4\pi\, r\, r_1}{\tau_0} G_1(r,r_1,\omega) =$$

$$= \frac{1}{\alpha}\,\left[e^{-\varphi_1}(\sin\varphi_1-\cos\varphi_1) - e^{-\varphi_2}(\sin\varphi_2-\cos\varphi_2)\right],$$

$$\gamma = \frac{c_P}{c_V}\; = \frac{K_S}{K_T}; \quad y_2(\rho^*) = \rho^*(5-2\rho^*)/(1-\rho^*)(2-\rho^*);$$

$$\alpha = \sqrt{2\omega\tau_0} = \sqrt{2\omega^*\frac{\tau_0}{\tau}};$$

$$\varphi_1 = \varphi_1(r,r_1,\omega) = \frac{\alpha}{2}(r-r_1)\,;$$

$$\varphi_2 = \varphi_2(r,r_1,\omega) = \frac{\alpha}{2}(r+r_1),$$

$T* \,= kT/\varepsilon$ is the reduced temperature, ε is the value of the potential $\Phi\left(|\mathbf{r}|\right)$ at the point $r = r_{\min}$, and $\omega* = \omega\tau = \omega\, m\,/2\beta$ is the reduced frequency.

Thus, the obtained analytical expressions (7)–(9) enable one to study theoretically and to perform numerical calculations of the kinetic shear $\eta_S(\omega)$ and bulk $\eta_V(\omega)$ viscosities of liquids as functions of the state parameters in a wide frequency range.

Table 2

T, K	ρ, kg/m^3	η_S, mPa·s		η_V, mPa·s			η_V/η_S		
		[7]	this work	[7]	$\langle\eta_V\rangle$, [7]	this work	[7]	$\langle\eta_V\rangle/\eta_S$, [7]	this work
86	1402	0.272	0.271	0.152	0.163	0.262	0.559	0.599	0.970
	1407	0.276	0.276	0.163	0.168	0.269	0.591	0.609	0.975
	1413	0.280	0.282	0.174	0.177	0.277	0.621	0.632	0.981
	1419	0.283	0.289	0.185	0.185	0.285	0.654	0.654	0.987
90	1377	0.235	0.236	0.153	0.161	0.228	0.651	0.685	0.964
	1383	0.239	0.241	0.155	0.162	0.234	0.649	0.678	0.970
	1390	0.242	0.248	0.168	0.171	0.242	0.694	0.707	0.977
	1396	0.245	0.254	0.180	0.182	0.249	0.735	0.743	0.983
	1405	0.250	0.262	0.193	0.193	0.260	0.772	0.772	0.992
	1418	0.255	0.276	0.211	0.211	0.277	0.827	0.827	1.005
100	1312	0.180	0.173	0.156	0.157	0.165	0.867	0.872	0.952
	1319	0.183	0.178	0.152	0.155	0.171	0.831	0.847	0.959
	1327	0.186	0.183	0.155	0.156	0.177	0.833	0.839	0.967
	1334	0.189	0.188	0.161	0.160	0.183	0.852	0.847	0.974
	1347	0.194	0.197	0.172	0.172	0.195	0.887	0.887	0.986
	1362	0.199	0.209	0.188	0.188	0.209	0.945	0.945	1.001
110	1240	0.144	0.130	0.167	0.154	0.121	1.160	1.069	0.930
	1248	0.148	0.134	0.160	0.150	0.125	1.081	1.014	0.938
	1258	0.151	0.139	0.149	0.144	0.131	0.987	0.954	0.948
	1268	0.155	0.144	0.151	0.150	0.138	0.974	0.968	0.958
	1286	0.162	0.153	0.166	0.159	0.150	1.025	0.981	0.976
	1303	0.169	0.163	0.175	0.175	0.162	1.036	1.036	0.993
120	1160	0.113	0.098	0.184	0.167	0.087	1.628	1.478	0.888
	1164	0.114	0.099	0.174	0.165	0.089	1.526	1.447	0.893
	1181	0.119	0.105	0.161	0.156	0.096	1.353	1.311	0.911
	1195	0.123	0.111	0.156	0.153	0.102	1.268	1.244	0.926
	1219	0.130	0.119	0.163	0.163	0.112	1.254	1.254	0.947
	1241	0.137	0.130	0.169	0.169	0.127	1.234	1.234	0.974
130	1065	0.086	0.072	0.191	0.189	0.059	2.221	2.198	0.811
	1092	0.088	0.079	0.179	0.175	0.067	2.034	1.989	0.844
	1113	0.092	0.085	0.165	0.161	0.074	1.793	1.750	0.869
	1149	0.099	0.096	0.163	0.163	0.087	1.646	1.646	0.910
	1175	0.105	0.104	0.164	0.164	0.098	1.562	1.562	0.938
135	1031	0.075	0.066	0.222	0.221	0.051	2.960	2.947	0.781
	1037	0.076	0.067	0.195	0.194	0.053	2.566	2.553	0.789
	1065	0.079	0.073	0.170	0.166	0.060	2.152	2.101	0.824
No estimates were made for the saturation line									
140	968	0.065	0.055	0.253	0.253	0.039	3.892	3.892	0.707
	1011	0.068	0.063	0.198	0.193	0.048	2.912	2.838	0.765
	1065	0.073	0.074	0.176	0.176	0.062	2.411	2.411	0.835
	1103	0.077	0.084	0.169	0.169	0.074	2.195	2.195	0.881

3. Results of Numerical Calculations

Relations (7)–(9) with regard for formulas (5) and (6) were used for numerical calculations of the shear $\eta_S(\omega)$ and bulk $\eta_V(\omega)$ viscosity coefficients of liquid argon as functions of the thermodynamic state parameters (ρ, T, and P) in a wide frequency range. The values of the temperature $T(86 \div 140$ K) and other

Table 3

T, K	ν^*	η_S, mPa·s					η_V, mPa·s				
	ρ, kg/m^3	[7]	10^{-6}	10^{-4}	10^{-2}	1	[7]	10^{-6}	10^{-4}	10^{-2}	1
86	1402	0.272	0.271	0.262	0.188	0.011	0.152	0.262	0.265	0.252	0.017
	1407	0.276	0.276	0.267	0.191	0.011	0.163	0.269	0.272	0.257	0.017
	1413	0.280	0.282	0.273	0.195	0.011	0.174	0.277	0.280	0.263	0.018
	1419	0.283	0.289	0.279	0.199	0.012	0.185	0.285	0.288	0.270	0.018
90	1377	0.235	0.236	0.229	0.171	0.011	0.153	0.228	0.231	0.225	0.017
	1383	0.239	0.241	0.235	0.174	0.011	0.155	0.234	0.237	0.231	0.017
	1390	0.242	0.248	0.241	0.178	0.011	0.168	0.242	0.245	0.237	0.017
	1396	0.245	0.254	0.246	0.182	0.012	0.180	0.249	0.252	0.243	0.018
	1405	0.250	0.262	0.255	0.187	0.012	0.193	0.260	0.263	0.251	0.018
	1418	0.255	0.276	0.268	0.195	0.012	0.211	0.277	0.280	0.264	0.018
100	1312	0.180	0.173	0.170	0.135	0.011	0.156	0.165	0.167	0.169	0.016
	1319	0.183	0.178	0.174	0.138	0.011	0.152	0.171	0.173	0.174	0.016
	1327	0.186	0.183	0.179	0.142	0.011	0.155	0.177	0.180	0.180	0.016
	1334	0.189	0.188	0.184	0.145	0.011	0.161	0.183	0.186	0.185	0.017
	1347	0.194	0.197	0.193	0.152	0.011	0.172	0.195	0.197	0.195	0.017
	1362	0.199	0.209	0.204	0.159	0.012	0.188	0.209	0.211	0.207	0.018
110	1240	0.144	0.130	0.128	0.107	0.010	0.167	0.121	0.122	0.125	0.014
	1248	0.148	0.134	0.132	0.109	0.010	0.160	0.125	0.127	0.129	0.015
	1258	0.151	0.139	0.136	0.113	0.010	0.149	0.131	0.133	0.134	0.015
	1268	0.155	0.144	0.141	0.117	0.010	0.151	0.138	0.139	0.140	0.015
	1286	0.162	0.153	0.151	0.124	0.011	0.166	0.150	0.151	0.151	0.016
	1303	0.169	0.163	0.160	0.131	0.011	0.175	0.162	0.164	0.161	0.017
120	1160	0.113	0.098	0.097	0.083	0.009	0.184	0.087	0.088	0.089	0.012
	1164	0.114	0.099	0.098	0.084	0.009	0.174	0.089	0.089	0.091	0.013
	1181	0.119	0.105	0.104	0.089	0.009	0.161	0.096	0.097	0.098	0.013
	1195	0.123	0.111	0.109	0.093	0.009	0.156	0.102	0.103	0.104	0.014
	1219	0.130	0.119	0.117	0.099	0.010	0.163	0.112	0.113	0.113	0.014
	1241	0.137	0.130	0.128	0.108	0.010	0.169	0.127	0.127	0.125	0.015
130	1065	0.086	0.072	0.072	0.064	0.007	0.191	0.059	0.059	0.060	0.010
	1092	0.088	0.079	0.078	0.069	0.008	0.179	0.067	0.067	0.067	0.011
	1113	0.092	0.085	0.084	0.073	0.008	0.165	0.074	0.074	0.074	0.011
	1149	0.099	0.096	0.094	0.082	0.009	0.163	0.087	0.087	0.086	0.013
	1175	0.105	0.104	0.103	0.089	0.009	0.164	0.098	0.098	0.096	0.013
135	1031	0.075	0.066	0.065	0.059	0.007	0.222	0.051	0.052	0.052	0.009
	1037	0.076	0.067	0.066	0.060	0.007	0.195	0.053	0.053	0.053	0.009
	1065	0.079	0.073	0.072	0.065	0.007	0.170	0.060	0.061	0.060	0.010
No estimates were made for the saturation line											
140	968	0.065	0.055	0.055	0.050	0.006	0.253	0.039	0.039	0.039	0.008
	1011	0.068	0.063	0.062	0.056	0.006	0.198	0.048	0.048	0.048	0.009
	1065	0.073	0.074	0.073	0.065	0.007	0.176	0.062	0.062	0.061	0.010
	1103	0.077	0.084	0.083	0.073	0.008	0.169	0.074	0.074	0.071	0.011

parameters were taken from [7]. When calculating τ, τ_0, $\eta_S(\omega)$, and $\eta_V(\omega)$, the friction coefficient β was determined according to (7) in the hard sphere approximation [20]. Table 1 presents the theoretical results obtained for the isofrequency ($\nu^* = 10^{-6}$, $\nu \sim 10^6$ Hz) and the shear $\eta_S(\omega)$ and bulk $\eta_V(\omega)$ viscosities, the theoretical data reported in [3], and the experimental results of Table 4 from [7] at various temperatures, densities, and pressures of liquid argon.

The calculated values of $\eta_S(\omega)$,$\eta_V(\omega)$, and the ratio η_V/η_S are in better agreement with the experimental results [7] than the theoretical data given in [3]. It is worth noting that, according to our calculations, the values of the ratio η_V/η_S as a function of ρ, T, and P lie in the interval $0.872 \div 0.967$. It is due to the correct account of the contributions of structural relaxation, as, according to [1,2], this interval is of the order of $0.5 \div 10$.

Table 2 shows the results of our theoretical calculations of $\eta_S(\omega)$, $\eta_V(\omega)$, and the ratio η_V/η_S for liquid argon at $\nu^* = 10^{-6}$ in the temperature interval $86 \leq T \leq 140$ K at various densities. Our results are compared to the experimental results and the values obtained for these coefficients and $\langle\eta_V\rangle$ in [7].

As follows from Table 2, the data obtained for $\eta_S(T)_\rho$ and $\eta_S(\rho)_T$ are in good quantitative agreement with the experimental results. One observes an increase of $\eta_S(\rho)_T$ for each isotherm and a decrease of $\eta_S(T)_\rho$ for the isochors. As for the values of $\eta_V(T)_\rho$ and $\eta_V(\rho)_T$, those obtained in this work and in experiments, as well as $\langle\eta_V(T)_\rho\rangle$ and $\langle\eta_V(\rho)_T\rangle$ (according to [7]), are in satisfactory agreement. It is worth noting that, at low temperatures, our data are somewhat overestimated, but the quantitative agreement is getting better with increase in the temperature. The ratios $\frac{\eta_V}{\eta_S}(T)$ and $\frac{\eta_V}{\eta_S}(\rho)$ in the whole temperature and density ranges lie within $0.707 \div 1.005$, whereas their experimental values lie in the interval $0.559 \div 3.892$. At low temperatures and large densities, the experimental and theoretical values of η_V/η_S coincide quantitatively. At high temperatures $T \geq 130$ K, the experimental ratios η_V/η_S grow due to an increase of η_V and a decrease of η_S, while the theoretical ratio η_V/η_S remains almost constant and equal to $0.707 \div 0.938$ in this temperature range, because both η_S and η_V decrease with rising the temperature in this region.

As was noted in [7], the saturation line for liquid argon manifests the dependence of α/ν^2 on ν at $T \geq 140$ K caused by the frequency dispersion of the transport coefficients. That is why we used relations (5)–(7) to investigate the frequency dependence of the shear and bulk viscosity coefficients in a wide range of reduced frequencies $\nu^* = 10^{-6} \div 1$ ($\nu \sim 10^6 \div 10^{12}$ Hz) at a fixed temperature T and a fixed density ρ. The results are presented in Table 3 and compared with the static experimental data on the dependences $\eta_S(\nu)$ and $\eta_V(\nu)$ obtained in [7] at $\nu = 5 \times 10^6$ Hz.

The theoretical results given in Table 3 show that the frequency dispersion region is located at $\nu^* = 10^{-6} \div 1$ ($\nu \sim 10^6 \div 10^{12}$ Hz), i.e. in a wide frequency range, which is due to the contribution of the structural relaxation. According to the general relaxation theory, this interval is approximately equal to 10^2 Hz according to the decay law of these coefficients $\sim \nu^{-2}$, whereas in our case, it is $\sim \nu^{-1}$.

Thus, the results of theoretical researches performed in the framework of the chosen model of the intermolecular interaction potential $\Phi(|\mathbf{r}|)$ and the radial distribution function $g(|\mathbf{r}|)$ are in satisfactory qualitative and quantitative agreement with the experimental data on the viscosity coefficients of liquid argon. Good agreement is observed at low temperatures and high densities of liquid argon. Consequently, this model allows one to study viscous, elastic, and acoustic properties of simple liquids in a wide range of the state parameters and in a vicinity of the triple point.

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ВИВЧЕННЯ ЧАСТОТНОЇ ДИСПЕРСІЇ КОЕФІЦІЄНТІВ ЗСУВНОЇ І ОБ'ЄМНОЇ В'ЯЗКОСТІ РІДКОГО АРГОНУ ЗАЛЕЖНО ВІД ПАРАМЕТРІВ СТАНУ

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Резюме

Наведено короткий аналіз експериментальних і теоретичних робіт по вивченню коефіцієнтів зсувної η_S та об'ємної η_V в'язкості простих рідин, а також оптимальний вибір потенціалу міжмолекулярної взаємодії $\Phi\left(|\mathbf{r}|\right)$ й радіальної функції розподілу $g\left(|\mathbf{r}|\right)$. Останні використовуються для визначення коефіцієнта тертя β і часу релаксації τ та τ_0, залежно від температури T і щільності ρ, а також проведення безлічі розрахунків коефіцієнтів в'язкості $\eta_S(\omega)$ і $\eta_V(\omega)$ у широкому інтервалі зміни параметрів стану і частот. Отримані результати знаходяться в кількісному задовільному узгодженні з теоретичними і експериментальними літературними даними для рідкого аргону.