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SOUND PROPAGATION IN MAGNETIC FLUIDS BASED ON MINERAL OILS NEAR THE GLASS TRANSITION TEMPERATURE OF THE DISPERSION MEDIUM

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Abstract
Aim. The paper establishes the dependence of the influence of the concentration of the solid phase on the acoustic parameters of a magnetic fluid based on transformer oil in a wide temperature range, including the temperature close to the glass transition point of the dispersion medium.
Methodology. The research is based on methods of physical acoustics and the pulse method of variable distance under external temperature influence in particular.
Results. The temperature and concentration dependences of the density, velocity and absorption coefficient of ultrasonic waves are investigated. A comparison is performed with the main theoretical models and approaches. In the temperature range near the glass transition point of the dispersion medium, additional effects are observed that are not described in the literature and are inconsistent with the currently existing theories of sound propagation in dispersed systems with a large density difference between the liquid and solid phase.
Research implications. Scientific and practical interest is due to the fact that the study of non-magnetized ferromagnetic colloids with a high contrast of densities between phases near the glass transition point of the dispersion medium is relevant, since there is a lack of research in this temperature range and, moreover, additional effects associated with the displacement of the phase transition at high concentrations of the solid phase are possible.
Keywords: acoustic spectroscopy, nanomaterials, ferromagnetic colloids, magnetic fluid, dispersed systems.
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РАСПРОСТРАНЕНИЕ ЗВУКА В МАГНИТНЫХ ЖИДКОСТЯХ НА ОСНОВЕ МИНЕРАЛЬНЫХ МАСЕЛ ВБЛИЗИ ТЕМПЕРАТУРЫ СТЕКЛОВАНИЯ ДИСПЕРСИОННОЙ СРЕДЫ

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Аннотация

Цель. Целью данной работы является установление зависимости влияния концентрации твёрдой фазы на акустические параметры магнитной жидкости на основе трансформаторного масла в широком диапазоне температур, в том числе вблизи точки стеклования дисперсионной среды.

Процедура и методы. Проведённые в данной работе исследования основаны на методах физической акустики, в частности импульсном методе переменного расстояния при внешнем температурном воздействии.

Результаты. Исследованы температурные и концентрационные зависимости плотности, скорости и коэффициента поглощения ультразвуковых (далее – УЗ) волн. Проведено сравнение с основными теоретическими моделями и подходами. В области температур возле точки стеклования дисперсионной среды наблюдаются дополнительные эффекты, не описанные в литературных источниках и не согласующиеся с существующими на данный момент теориями распространения звука в дисперсных системах с большой разностью плотностей между жидкой и твердой фазой.

Теоретическая значимость. Научный и практический интерес представляет исследование ненамагниченных ферромагнитных коллоидов с высоким контрастом плотностей между фазами вблизи точки стеклования дисперсионной среды, так как в этой области температур наблюдается недостаточность исследований и, к тому же, возможны дополнительные эффекты, связанные со смещением фазового перехода на высоких концентрациях твердой фазы.

Ключевые слова: акустическая спектроскопия, наноматериалы, ферромагнитные коллоиды, магнитная жидкость, дисперсные системы.

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Introduction

By now, the behavior of ultrasonic waves in colloids and suspensions of nanoparticles has been studied in sufficient detail at room and high temperatures [1–5]. Non-magnetized magnetic liquids with nanoscale ferroparticles can be considered as an ordinary colloid, since in the absence of external magnetic fields, structural mechanisms will be absent. This means that their acoustic studies could be compared with those of a similar concentration range of colloids, for example [6–13].

It is also interesting to study colloids containing rather high volume concentrations of solid particles in the region of low temperatures close to the glass transition point of the dispersion medium. This interest is conditioned not only by the insufficiency of studies of these substances under these conditions, but also by the possibility of considering the liquid-solid phase transition from the side of acoustic quantities. The low operating frequency of the studies was chosen due to the possibility of considering the liquid-glass phase transition on a macroscopic scale, which will allow us to judge the internal changes in the concentrated colloid during further studies.

Thus, the purpose of this work is to study the concentration series of magnetic fluids (hereinafter referred to as MFs), based on transformer oil near the glass transition temperature of the dispersion medium at a low operating frequency and analyze the results obtained.

Materials and methods of research

The object of research, as mentioned above, is a magnetic fluid based on transformer oil (hereinafter referred to as MFTO), represented by a concentration series [0.2%, 0.5%, 1%, 2%, 5%, and 10% volume concentrations of ferroparticles of magnetite with an average size of D = 15nm (see Fig. 1)]. Ferroparticles of magnetite are stabilized by oleic acid, which is a surfactant in this dispersed system.

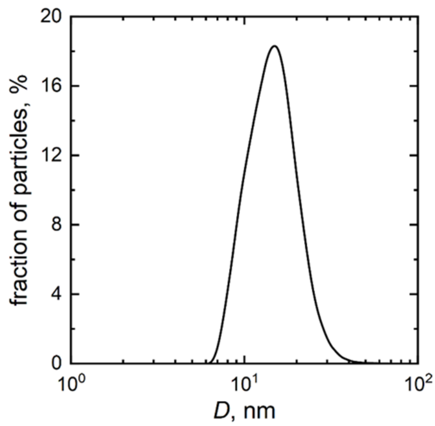

Fig. 1 / Рис. 1. Quantitative distribution of MFTO particle sizes based on the dynamic light scattering method / Количественное распределение размеров частиц MFTO на основе метода динамического рассеяния света

Source: compiled by the authors.

The principle of measuring acoustic parameters of MFTO (propagation velocity and absorption coefficient of ultrasonic waves is based on the pulse method of variable distance, the study was conducted at a frequency f = 3,65MHz, the amplitude of the signal U = 20V, the period between packets t = 150us. The transducers are made of lithium niobate ferroelectric ($LiNbO_3$) with a two-component coating consisting of chromium, which has good adhesive properties, and nickel, selected due to weak chemical activity to prevent interaction with the test sample. The calculated error of the measurement data does not exceed 1% for speed and 5–7% for the sound absorption coefficient. The external temperature impact is implemented using a thermostatic chamber with an accuracy of temperature setting up to 0.1 K, and the MFTO density was studied using a pycnometer having a nominal volume of 5 ml. To obtain the dependence of the type $\rho(T)$, a pycnometer with an MFTO sample was placed into a thermostatic chamber for a period of 10 min, after which measurements were made on laboratory scales GOSMETER VL-220M, having a measurement accuracy up to 10^{-7} kg.

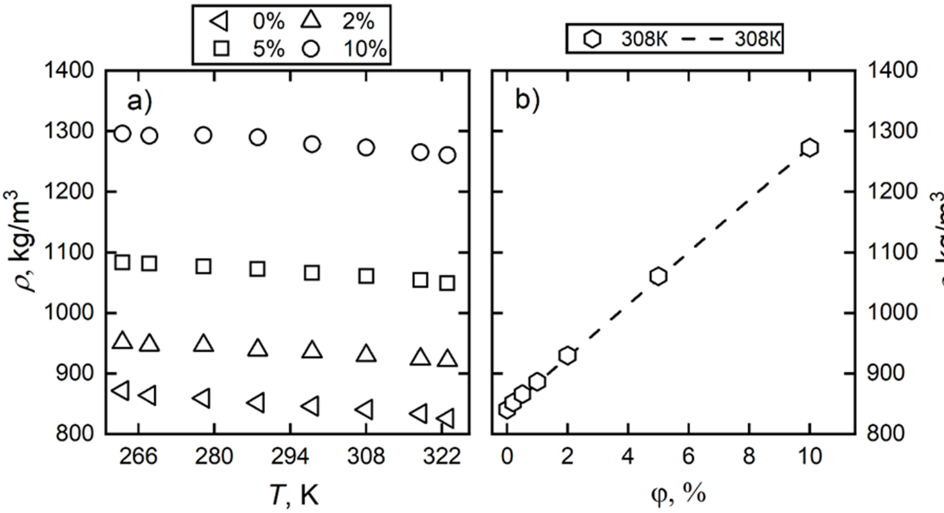

Fig.2 / Рис.2. *a* – Temperature dependence of the density $\boldsymbol{\rho(T)}$ in MFTO, and *b* – concentration dependence of $\boldsymbol{\rho(\varphi)}$ in MFTO: the straight line is $\langle\boldsymbol{\rho}\rangle(\boldsymbol{\varphi})$ according to formula (1) / *a* – температурная зависимость плотности $\boldsymbol{\rho(T)}$ MFTO, *b* – концентрационная зависимость $\boldsymbol{\rho(\varphi)}$ MFTO, прямая - $\boldsymbol{\rho}_{\text{теор}}(\boldsymbol{\varphi})$ по формуле (1)

Source: compiled by the authors.

The results of measuring the density of MFTO are shown in Fig. 2. The coincidence of experimental data with theoretical calculations based on the additivity model (1) allows us to judge the reliability of the concentrations provided by φ and apply other conclusions from continuum mechanics to the calculations of acoustic parameters [14]

$$\langle\rho\rangle = (1-\varphi)\rho_1 + \varphi\rho_2, \tag{1}$$

where $\langle\rho\rangle$ is the MF density, φ is the concentration of the solid phase in MFs, ρ_1 is the density of pure transformer oil, and ρ_2 is the density of magnetite.

Experimental results

Based on the calculations of (1) to MFTO, as mentioned above, it is possible to use other insights from continuum mechanics, in particular the calculation of the coefficient of adiabatic compressibility $\langle\beta\rangle$

$$\langle\beta\rangle = (1-\varphi)\beta_1 + \varphi\beta_2,$$

and, according to the known $\langle\beta\rangle$ and $\langle\rho\rangle$, calculate the Laplacian velocity of sound

$$c_0 = (\langle\rho\rangle\langle\beta\rangle)^{-\frac{1}{2}},$$

and the main amendments to c_0 introduced by Ritov and Isakovich [14; 15]

$$\Delta c_T = \frac{\varphi}{2} T {c_0}^3 \langle\rho\rangle\, \rho_2 \mathrm{C}_{p2} \left(\frac{\alpha_1}{\rho_1 \mathrm{C}_{p1}} - \frac{\alpha_2}{\rho_2 \mathrm{C}_{p2}}\right)^2,$$

$$\Delta c_\eta = c_л \frac{a\xi\sqrt{\xi}(1+b\sqrt{\xi})}{(1+\sqrt{\xi})^2 + \xi(1+b\sqrt{\xi})^2 - a\xi\sqrt{\xi}(1+b\sqrt{\xi})},$$

where C_{p1} and C_{p2} are the specific heat capacities of the liquid and solid phase at $p = const$, α_1 and α_2 are the coefficients of thermal expansion of the liquid and solid phase, $\xi = \frac{\omega\rho}{\eta} R^2$, $a = \frac{2}{9}\varphi\left(\frac{\rho_2-\rho_1}{\rho_1}\right)^2$, and $b = \frac{2}{9}\left(\frac{\rho_1-2\rho_2}{\rho_1}\right)$.

Thus, the final calculation of the values of the propagation velocity of ultrasonic waves will be presented as

$$c_{theory} = c_0 + \Delta c_\eta + \Delta c_T. \tag{2}$$

The results of calculations (2) are shown in Fig. 3.

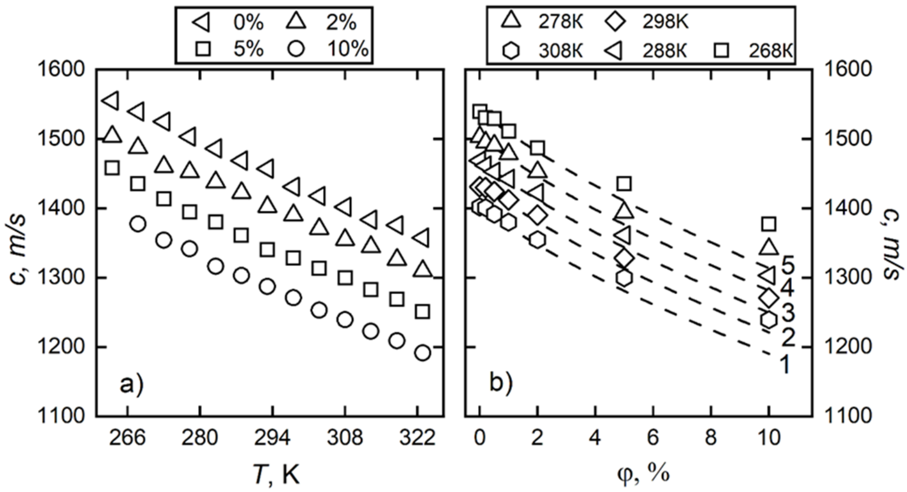

Fig. 3 / Рис. 3. *a* – Temperature dependence of the sound speed $\boldsymbol{c(T)}$ in MFTO, and *b* – concentration dependence $\boldsymbol{c(\varphi)}$ in MFTO: straight lines are $\boldsymbol{c_{theory}(\varphi)}$ according to formula (2), 1 – 308K, 2 – 298K, 3 – 288K, 4 – 278K, 5 – 268K / *а* – температурная зависимость скорости звука $\boldsymbol{c(T)}$ в MFTO, б – концентрационная зависимость $\boldsymbol{c(\varphi)}$ в MFTO, прямые – $\boldsymbol{c}_{\text{теор}}\boldsymbol{(\varphi)}$ по формуле (2), 1 – 308K, 2 – 298K, 3 – 288K, 4 – 278K, 5 – 268K

Source: compiled by the authors.

Similar ideas are used in the calculations of the absorption coefficient of ultrasonic waves with the difference that the dispersion in the case of α has a much greater [16] manifestation. Otherwise, the proper absorption of α_{int} is calculated by the formula

$$\alpha_{int} = \omega^2 \frac{\frac{4}{3}\eta + \zeta}{2\rho {c_0}^3},$$

where η is the shear, and ζ is the bulk viscosity of the carrier fluid. Corrections are also added to its own absorption: thermal

$$\alpha_T = \frac{R^2\omega^2}{6\chi_1}\varphi T c_0 \rho {\rho_1}^2 {C_p}^2 (\chi_1 {\chi_2}^{-1} + 0{,}2)\left(\frac{\beta_1}{\rho_1 C_{p1}} - \frac{\beta_1}{\rho_2 C_{p2}}\right)^2,$$

where χ_1 and χ_2 are the thermal conductivity of the dispersion medium and the solid phase, respectively, and the viscosity

$$\alpha_\eta = \frac{\omega}{c_0}\frac{a\xi\left(1+\sqrt{\xi}\right)}{\left(1+\sqrt{\xi}\right)^2 + \xi\left(1+b\sqrt{\xi}\right)^2},$$

and the final expression is represented as

$$\alpha_{theory} = \alpha_{int} + \alpha_\eta + \alpha_T. \qquad (3)$$

The results of theoretical calculations (3) are presented in Fig. 4.

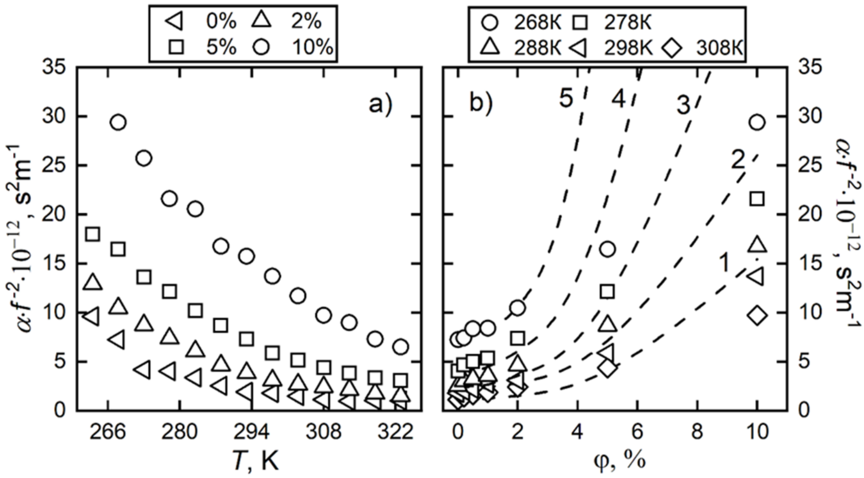

Fig. 4 / Рис. 4. *a* – Temperature dependence of the sound absorption coefficient $\frac{\alpha}{f^2}(\boldsymbol{T})$, and *b* – concentration dependence $\frac{\alpha}{f^2}(\boldsymbol{\varphi})$: lines are $\frac{\alpha_{theory}}{f^2}(\boldsymbol{\varphi})$, 1 – 308K, 2 – 298K, 3 – 288K, 4 – 278K, 5 – 268K / *a* – температурная зависимость коэффициента поглощения звука $\frac{\alpha}{f^2}(\boldsymbol{T})$, *b* – концентрационная зависимость $\frac{\alpha}{f^2}(\boldsymbol{\varphi})$, прямые – $\frac{\alpha_{theory}}{f^2}(\boldsymbol{\varphi})$, 1 – 308К, 2 – 298К, 3 – 288К, 4 – 278К, 5 – 268К

Source: compiled by the authors.

In the case of the speed of sound, Δc_η does not exceed the calculated error throughout the temperature and concentration range of measurements, and Δc_T begins to make a significant contribution, increasing with decreasing temperature, only at MFTO concentrations φ more than 2%. A similar behavior is typical for α_{theory}.

Conclusions

The acoustic values of c_{theory} and α_{theory} depend on the inertial properties of the dispersed system in which ultrasonic waves propagate. The viscous and thermal mechanisms are taken into account by introducing corrections to the initial values of c_0 and α_{int}.

The general type of interactions of sound waves with colloidal systems in the form of magnetic fluids is determined not only by the carrier medium, but also by the concentration of the solid phase in the dispersed system. In the case of small concentrations of magnetic fluid based on transformer oil, the acoustic parameters satisfy the classical descriptions of energy dissipation due to viscous and thermal waves at the phase interfaces. However, with an increase in the concentration of the solid phase, there is a discrepancy between theoretical calculations and experimental data due to the occurrence of additional absorption factors of ultrasonic waves in this system, which increase with the approach to the glass transition point of the dispersion medium.

It is noteworthy that the discrepancy between theoretical data and experimental results increases not only with an increase in the concentration of the solid phase, but also with an approach to the glass transition temperature of the dispersion medium. These effects can be justified by the displacement of the phase transition point of the MFs with increase in the concentration of the solid phase.

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