

## DEPENDENCES OF THE MODULI OF ELASTICITY OF MAGNETIC FLUIDS ON THE PARAMETERS OF STATE

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On the basis of dynamic expressions obtained earlier for the moduli of volume,  $K(\omega)$ , and shear,  $\mu(\omega)$ , elasticity and making use of a magnetic fluid composed of  $\text{Fe}_3\text{O}_4$  magnetic particles in kerosene as an example, the numerical analysis of the dependences of those characteristics on the fluid state parameters has been carried out. The characters of the dependences for the relaxation volume,  $K_r$ , and shear,  $\mu$ , moduli of elasticity on the concentration, density, and saturation magnetization were shown to be identical, namely, the growth of each of those parameters is accompanied by the growth of  $K_r$  and  $\mu$ . The temperature growth gives rise to a linear decrease of both  $K_r$  and  $\mu$ , whereas the growth of the magnetic field strength gradient to their linear increase. The results obtained agree with the experimental ones and confirm that the structural relaxation affects viscoelastic properties of magnetic fluids.

Elastic properties of a liquid are characterized by the response functions of its density to an external baric influence, i.e. by the adiabatic and isothermal compressibilities. Knowing the liquid density and the ultrasound speed in it, one can determine the static adiabatic compressibility  $\beta_s$  [1].

A number of works [2–4] were devoted to studying the dependence of the modulus of elasticity of magnetic fluids (MFs) on the state parameters. The calculations of the adiabatic compressibility carried out in work [2] revealed that pure kerosene is characterized by maximal compressibility. At high concentrations ( $c = 9.27\%$ ), the compressibility decreases and reaches the minimal value. At low pressures, the compressibilities of kerosene and a kerosene-based MF monotonously increase. In work [3], the temperature and concentration dependences of the adiabatic compressibility of the transformer-oil- and polysilicane-based MFs were experimentally studied. The value of the ratio  $\beta_s/\Delta c$ , where  $\Delta c$  is a variation of the solid particle concentration, was found to decrease with the temperature growth. In work [4], the role of the adiabatic compressibility of a stabilizing substance – oleic acid – in the formation of the MF compressibility value was estimated. According to the results of corresponding calculations, the adiabatic compressibilities of both the stabilizing substance and the disperse medium of the fluids under consideration

increase with the temperature growth, but their ratio remains constant within the temperature interval  $299 \div 353$  K.

Hence, the analysis of the available results of experimental and theoretical researches dealing with the elastic properties of MFs, which was made above, demonstrates that the regular studies of the dependences of the moduli of elasticity on the fluid state parameters are not sufficient for today, i.e. there is no microscopic theory which would predict the character of the dependences of the moduli of elasticity on the state parameters and the influence of an external magnetic field on them. However, MFs play the role of viscoelastic media in various devices and mechanisms functioning under the conditions of enhanced pressures and temperatures [5].

In this connection, the aim of this work is to carry out a numerical study for the dependences of the moduli of elasticity on the MF state parameters and to find how an external magnetic field affects them. This study is based on the dynamic expressions for the volume,  $K(\omega)$ , and shear,  $\mu(\omega)$ , moduli of elasticity which were obtained earlier in the framework of the molecular-kinetic theory [6]. The expressions for  $K(\omega)$  and  $\mu(\omega)$  look like

$$\begin{aligned}
 K(\omega) &= K_s + \frac{2\pi n^2 \sigma^3 \omega}{3} \int_0^\infty dr r^3 \frac{d\Phi}{dr} \int_0^r G_2(r, r_1, \omega) \times \\
 &\times \left[ \varphi(r_1) - \frac{\mu_0}{\beta} (\vec{M} \vec{\nabla}) \left( \frac{\partial H}{\partial \vartheta} \right)_{\rho, T} \left( \frac{\partial g_0(r_1)}{\partial r_1} \right) r_1 \right] d\vec{r}_1, \quad (1) \\
 \mu(\omega) &= \frac{nkT(\omega\tau_1)^2}{1 + (\omega\tau_1)^2} + \frac{2\pi n^2 \sigma^3 \omega}{15} \int_0^\infty dr r^3 \frac{d\Phi}{dr} \times \\
 &\times \int_0^r G_2(r, r_1, \omega) \frac{\partial g_0(r_1)}{\partial r_1} r_1 \times
 \end{aligned}$$

$$\times \left[ 1 - \frac{5\mu_0}{2\beta} (\vec{M}\vec{\nabla}) \left( \frac{\partial H}{\partial \vartheta} \right)_{\rho, T} \right] d\vec{r}_1, \quad (2)$$

where

$$K_s = n \left( \frac{\partial \rho}{\partial n} \right)_T + \frac{T}{nC_v} \left( \frac{\partial \rho}{\partial T} \right)_T$$

is the adiabatic modulus of elasticity,  $C_v$  the specific heat at constant volume,

$$G_2(r, r_1, \omega) = -\frac{\tau_0}{2} \left( \frac{2}{\omega\tau_0} \right)^{1/2} \left[ (\sin \varphi_1 + \cos \varphi_1) \times \right.$$

$$\left. \times \exp(-\varphi_1) - (\sin \varphi_2 + \cos \varphi_2) \exp(-\varphi_2) \right]$$

the Fourier transform of the fundamental solution of Smoluchowski equation,

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

$$\varphi(r_1) = 2ng_0(r_1) \left\{ 1 + \frac{1}{6} \frac{\partial \ln g_0(r_1)}{\partial \ln r_1} - \right.$$

$$\left. - \frac{1}{2} \left[ n \left( \frac{\partial g_0(r_1)}{\partial r_1} \right)_T + \gamma T \left( \frac{\partial g_0(r_1)}{\partial T} \right)_n \right] \right\},$$

$\gamma = (mC_v)^{-1} (\partial p / \partial T)_n$ ,  $\Phi(|\vec{r}|)$  is the potential of particle-particle interaction,  $g_0(|\vec{r}|, n, T)$  the equilibrium particle radial distribution function,  $k$  the Boltzmann constant,  $n$  the particle concentration,  $T$  the absolute temperature,  $\sigma$  the diameter of magnetic particles;  $\mu_0$  the magnetic permeability of vacuum;  $\beta$  the friction coefficient;  $\vec{M}$  the magnetization vector;  $H$  the magnetic field strength,  $\rho$  the MF density,  $\tau_1 = m/(2\beta)$  is the time of translational relaxation for the viscous stress tensor,  $\tau_0 = \omega_0^{-1} = \beta\sigma^2/(2kT)$  is the phenomenological time of structural relaxation, and  $\omega$  is the external process frequency.

As is seen from expressions (1) and (2), the dynamic moduli of elasticity demonstrate a complicated dependence on  $\Phi(|\vec{r}|)$  and  $g_0(|\vec{r}|, n, T)$ , the explicit form of which being determined by the specific choice of the MF model. According to the MF model proposed in work [7], the potential of particle-particle interaction, provided that there is an external magnetic field of the strength  $H$ , can be written down as follows:

$$\Phi(\vec{r}_{ij}) = \Phi^H(\vec{u}) + \Phi^S(\vec{r}_{ij}), \quad (3)$$

where:  $\Phi^H(\vec{u}_j) = -kTh(\vec{u}\vec{H})/H$  is the potential energy of interaction between the  $j$ -th particle and the magnetic field  $\vec{H}$  directed in parallel to the displacement vector  $\vec{u}$ ;  $h = \mu_0 mH/(kT)$  is the Langevin parameter,  $m$  the magnetic moment of the particle,  $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$  is the vector of relative displacement of the  $i$ -th and  $j$ -th particles, and  $\Phi^S(r) = 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6]$  is the Lennard-Jones potential.

According to work [8], in the spherically symmetric case, we seek the radial function of particle distribution in the form

$$g(r, n, T) = y(r, \rho^*) \exp(-\Phi(r)/kT), \quad (4)$$

where  $y(r, \rho^*)$  is the binary distribution function of two cavities. Taking the complexity of expressions (1) and (2) into account, the expression found by Carnahan and Starling,

$$y(\rho^*) = \frac{(2 - \rho^*)}{2(1 - \rho^*)^3}, \quad (5)$$

is taken as a contact value for  $y(r, \rho^*)$ . Here,  $\rho^* = \frac{\pi}{6} n\sigma^3 = \frac{\pi}{6} \rho \frac{N_0 \sigma^3}{M}$  is the reduced MF density,  $N_0$  the Avogadro constant, and  $M$  the molar mass.

Taking Eqs. (3)–(5) into account, expressions (1) and (2) can be written down in a reduced form:

$$K(\omega) = K_s + \frac{2\pi n^2 \sigma^3 kT}{9} \left[ \left( -\frac{1}{3} + \frac{\mu_0 M_s \tau_0 |\vec{\nabla} H|}{\beta l} \right) \times \right. \\ \left. \times I_1 - 2I_2 + I_3 + \gamma I_4 \right], \quad (6)$$

$$\mu(\omega) = \mu_k(\omega) - \frac{2\pi n^2 \sigma^3 kT \omega}{45} \left( 1 + \frac{5\mu_0 M_s |\vec{\nabla} H| \tau_0}{2\beta l} \right) I_1, \quad (7)$$

where

$$J_1 = \int_0^\infty dr D^*(r) \int_0^r G_2(r, r_1 \omega) D^*(r_1) g(r_1) dr_1,$$

$$J_2 = \int_0^\infty dr D^*(r) \int_0^r G_2(r, r_1 \omega) g(r_1) r_1 dr_1,$$

$$J_3 = \int_0^\infty dr D^*(r) \int_0^r G_2(r, r_1 \omega) y_2(\rho^*) g(r_1) r_1 dr_1,$$

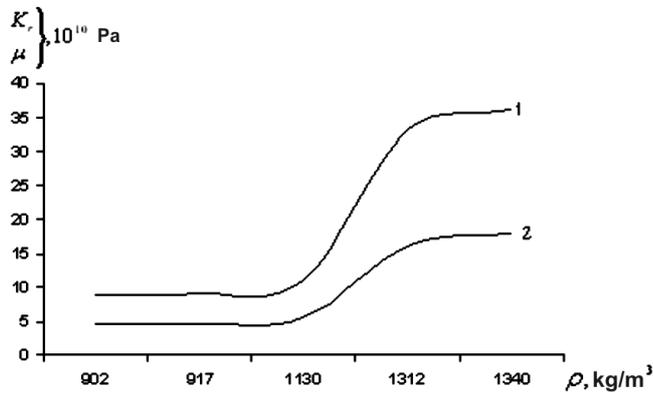


Fig. 1. Dependences of the  $K_r$  (1) and  $\mu$  (2) moduli of elasticity on the density  $\rho$

$$J_4 = \int_0^\infty dr D^*(r) \int_0^r G_2(r, r_1 \omega) [-\Phi^{*L-D}(r_1) +$$

$$+\Phi^{*H}(r_1)] g(r_1) r_1 dr_1,$$

$$\Phi^{*L-D}(r) = \frac{\Phi^{L-D}(r)}{kT} = L^* (r^{-12} - r^{-6}),$$

$$\Phi^{*H}(r) = \frac{\Phi^H(r)}{kT} = -\frac{\mu_0 m |\vec{\nabla} H| l}{kT},$$

$$D^*(r) = 6L^* (2r^{-6} - 1) r^{-5},$$

$$y_2(\rho^*) = \frac{(5 - 2\rho^*) \rho^*}{(1 - \rho^*)(2 - \rho^*)},$$

$L^* = 4\varepsilon/(kT)$ ,  $\mu_k(\omega^*) = \frac{nkT\omega^*}{1+\omega^{*2}}$  is the kinetic part of the shear modulus of MF elasticity,  $\omega^* = \omega\tau_1$  is the reduced frequency,  $\tau_1 = 10^{-12}$  s, and  $l = 10^{-2}$  m is the characteristic dimension of the system.

The value of  $\varepsilon$  is the depth of the potential well  $\Phi(|\vec{r}|)$  and, hence, characterizes the intensity of intermolecular forces. According to the results of work [9], in the case where there is an aligning magnetic field, the depth of the potential well is  $\varepsilon = 0.9kT$ . In MFs with a higher quality, the diameter of solid particles  $\sigma$  is scattered within the limits  $0.3 \div 10$  nm, their concentration is  $n = 10^{16} \div 10^{18}$  cm $^{-3}$ , and the magnetic moment  $m$  of a particle is about  $10^{-19}$  J/T [9].

Making use of expressions (6) and (7), where, for the specific values of the parameters, there were

selected those for a MF on the basis of kerosene and Fe $_3$ O $_4$  magnetic particles, numerical calculations of the dependences of the relaxation volume,  $K_r = K(\omega) - K_s$ , and shear,  $\mu$ , moduli of elasticity on the MF state parameters were carried out for the fixed value of the reduced frequency  $\omega^* = 1$ , which corresponds to the actual frequency  $\omega = 10^{12}$  Hz. The values for other calculation parameters – the concentration  $c$ , the density  $\rho$ , and the saturation magnetization  $M_s$  – were taken from work [9]. The results of calculations concerning the dependences of the isofrequency relaxation volume,  $K_r$ , and shear,  $\mu$ , moduli of elasticity on the density, concentration, and saturation magnetization at  $T = 298$  K and  $|\vec{\nabla} H| = 50$  A/m $^2$  are given in the table.

As is evident from the table, provided that the values of the frequency, the temperature, and the gradient  $|\vec{\nabla} H|$  are fixed, the values of  $K_r$  and  $\mu$  grow with the increase of the density, concentration, and saturation magnetization, whereas their ratio remains constant ( $K_r/\mu = 2$ ). These results, being in satisfactory agreement with the results of experimental works [2–4], confirm that the account of the influence of structural relaxation on the MF viscoelastic properties was made correctly. Hence, the dependences of the relaxation volume,  $K_r$ , and shear,  $\mu$ , moduli of MF elasticity on the density, concentration, and saturation magnetization possess the same character.

In Fig. 1, the dependences of the dynamic relaxation volume,  $K_r$ , and shear,  $\mu$ , moduli of elasticity on the fluid density at  $T = 298$  K,  $\omega^* = 1$ , and  $|\vec{\nabla} H| = 50$  A/m $^2$  are depicted. The figure demonstrates that those dependences increase nonlinearly with the growth of the MF density [2–4]. In Fig. 2, the dependences of  $K_r$  and  $\mu$  on the magnitude of saturation magnetization at  $T = 298$  K,  $\omega^* = 1$ , and  $|\vec{\nabla} H| = 50$  A/m $^2$  are exhibited. These dependences are also nonlinear functions of  $M_s$ . At last, in Fig. 3, the isofrequency concentration dependences of  $K_r/K_s$  and  $\mu/\mu_0$  at  $T = 298$  K,  $\omega^* = 1$ , and  $|\vec{\nabla} H| = 50$  A/m $^2$  are shown. The increase of the concentration gives rise to the nonlinear growth of both  $K_r/K_s$  and  $\mu/\mu_0$ , which is in qualitative agreement with experimental data [2–4].

$\rho$ , kg/m $^3$	$c$ , %	$M_s$ , 10 $^3$ A/m	$\mu$ , 10 $^{10}$ Pa	$K_r$ , 10 $^{10}$ Pa
902	2.44	8.1	4.45	8.90
907	2.87	8.3	4.50	9.07
1130	7.90	10.4	5.50	11.00
1312	11.20	31.2	16.00	33.00
1340	12.8	34.2	18.00	36.00

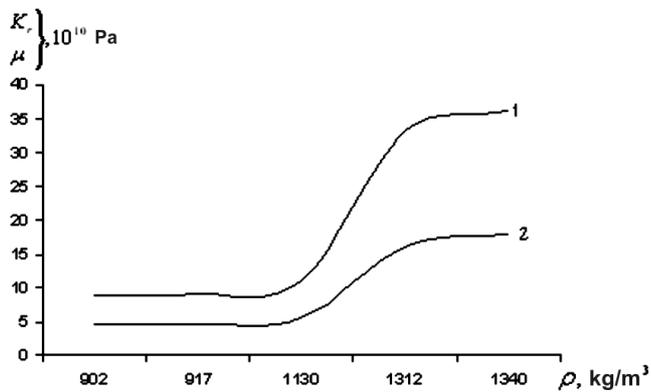


Fig. 2. Dependences of the  $K_r$  (1) and  $\mu$  (2) moduli of elasticity on the saturation magnetization  $M_s$

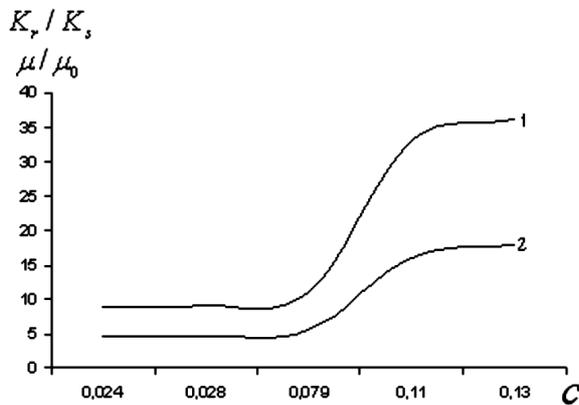


Fig. 3. Dependences of the  $K_r/K_s$ - (1) and  $\mu_s/\mu_0$ -ratios (2) on the concentration  $c$

In Fig. 4, the temperature dependences of the isofrequency relaxation volume,  $K_r$ , and shear,  $\mu$ , moduli of elasticity at  $T = 298$  K,  $\rho = 1340$  kg/m<sup>3</sup>,  $\omega^* = 1$ , and  $|\vec{\nabla}H| = 50$  A/m<sup>2</sup> are given. Here, both  $K_r$  and  $\mu$  decrease with the temperature growth [2].

In Fig. 5, the dependences of the relaxation volume,  $K_r$ , and shear,  $\mu$ , moduli of MF elasticity on the gradient of magnetic field strength at  $T = 298$  K,  $\omega^* = 1$ , and  $\rho = 1340$  kg/m<sup>3</sup> are depicted. With increase in the gradient of the magnetic field strength, both  $K_r$  and  $\mu$  increase linearly.

Thus, the results of numerical calculations for the quantities  $K_r$  and  $\mu$ , which were presented above, as well as their comparison with available experimental data, evidence for the correctness of the account made for the contribution of various internal relaxation processes running in MFs – in particular, structural relaxation – to the viscoelastic MF properties.

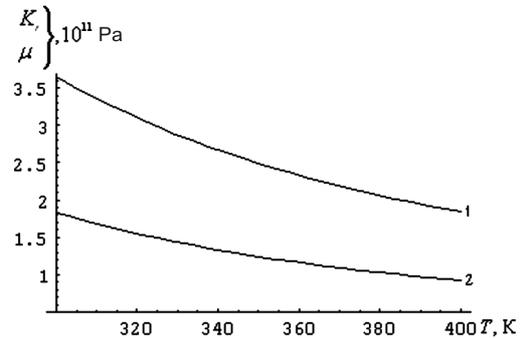


Fig. 4. Dependences of the  $K_r$  (1) and  $\mu$  (2) moduli of elasticity on the temperature  $T$

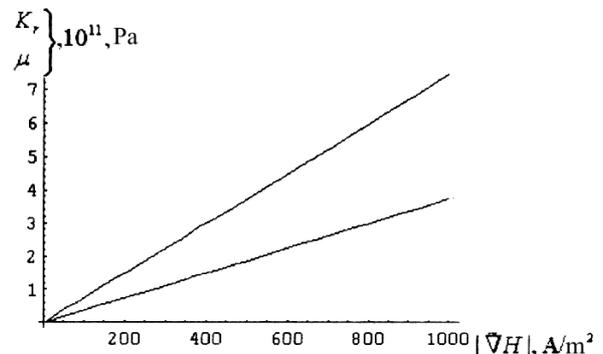


Fig. 5. Dependences of the  $K_r$  (1) and  $\mu$  (2) moduli of elasticity on the magnetic field gradient  $|\vec{\nabla}H|$

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ЗАЛЕЖНІСТЬ МОДУЛІВ ПРУЖНОСТІ МАГНІТНИХ  
РІДИН ВІД ПАРАМЕТРІВ СТАНУ

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Резюме

На основі раніше отриманих динамічних виразів для об'ємного  $K(\omega)$  та зсувного  $\mu(\omega)$  модулів пружності на прикладі магнітної рідини на основі гасу і магнітних частинок проведено чи-

сельне дослідження їх залежностей від параметрів стану. Показано, що характер залежності релаксаційного об'ємного  $K_r$  та зсувного  $\mu$  модулів пружності від концентрації, густини і намагніченості насичення однаковий, а саме з ростом останніх модулів  $K_r$  і  $\mu$  також зростають. Зі збільшенням температури величини  $K_r$  і  $\mu$  зменшуються лінійно, а з ростом градієнта напруженості магнітного поля – лінійно зростають. Отримані результати узгоджуються з результатами експериментальних робіт і підтверджують вплив структурної релаксації на в'язкопружні властивості магнітних рідин.