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The Influence of a Magnetic Field on the Elastic and Viscous Properties of Magnetoelastics

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Abstract—The effect of a uniform magnetic field on the viscous and elastic properties of new, rubber-elastic, magnetically guided polymeric materials (magnetoelastics) capable of substantial deformation in an external magnetic field was studied. It was found that the application of a magnetic field leads to a considerable rise in the Young modulus and in the viscosity coefficient of these materials. A model is proposed which describes the effect of magnetic field on the elasticity parameters. Based on the obtained experimental data, the influence of an external magnetic field and a magnetoelastic-stretching force F on the volume fraction of polymer regions contracted by interaction of magnetic particles is considered in terms of the proposed theoretical model.

INTRODUCTION

Elastic composite materials comprising different polymers and fillers with strong magnetic properties (known as magnetic elastomers or magnetoelastics [1]) are widely used as permanent magnets, cores, and holding or fastening elements that are capable of repeating the irregularities and curvature of ferromagnetic surfaces contacting with them.

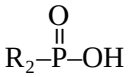
However, there are few works concerning the investigation of composite polymer materials that respond to a noticeable extent to an external magnetic field by changing their shape. Of these works, the studies on magnetic gels comprising chemically crosslinked polymers filled with magnetic particles ~10 nm in size are of primary interest [2–4]. In the cited works, it was shown that the shape of a magnetic gel in a nonuniform magnetic field depends on the strength and configuration of the field. Magnetically induced unidirectional deformations observed in this case were found to be as large as 40% of the sample size in the absence of the field. Studies on gelatin-based magnetic gels [5], fabrication of magnetic polyacrylamide hydrogels [6], and construction of the surface replica of a magnetic polymer composite required for magnetic control of wettability [7] may be also noted.

In this work, we studied the magnetic field effect on the elastic and viscous properties of new rubber-elastic magnetoguided polymer materials, the magnetoelastics [8–11]. The giant magnetodeformation effect observed in this materials, which consists in the deformation of a magnetoelastic exposed to an external magnetic field, not only exceeds considerably the deformations of piezo-, electro-, and magnetic materials but is also stronger than the magnetodeformation effects observed in gels.

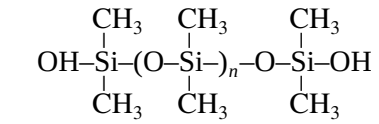
As an illustration, Fig. 1 shows photographs of a magnetoelastic sample.

EXPERIMENTAL

Magnetoelastics were prepared by dispersing ultrafine and coarser magnetic particles in a siloxane rubber oligomer in the presence of a surfactant and plasticizers. The organophosphorus surfactant of the general formula



was used to disperse and stabilize the dispersion of magnetic powders in the final polymer. The plasticizers were PMS-100 and PMS-500 polymethylsiloxanes. The dispersion was performed with a UZDN-A ultrasonic disperser. The basic structuring polymer in the materials of interest was a siloxane oligomer, which was prepared by hydrolysis of dichlorodimethylsilane to yield oligomers of the general formula



and the molecular mass of 10^4–10^5.

The network matrix structure was formed by reacting these oligomers with tetraethylsilane in the pres-

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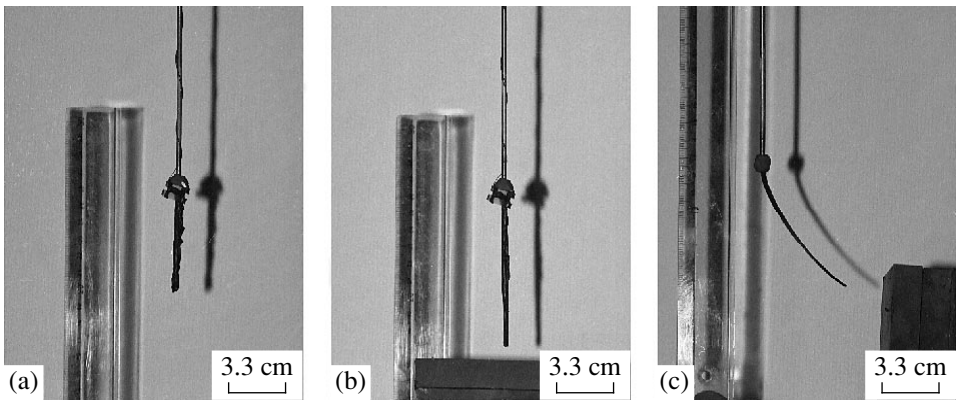

Fig. 1. Photographs of a magnetoelastic sample (a) in the absence of field and (b, c) in the field of a permanent magnet.

ence of butyltin caprylate as a catalyst. As a result, samples having high elasticity and a relatively large extent of volume filling of the polymer with magnetic particles were obtained.

An increase in the elasticity was achieved by adding a plasticizer and selecting the polymerization conditions for a magnetoelastic. In some respect, magnetoelastics are solid-state analogues of magnetorheological fluids [12] which display the magnetorheological effect, the dramatic change in mechanical properties (viscosity, plasticity, elasticity) of some suspensions by the action of a magnetic field. Figure 2 depicts the dependence of the Young modulus for magnetoelastics having a close mass fraction of iron particles (~30%) of ~2 μm size on the plasticizer mass fraction in the polymer matrix, which is defined by $\nu = m_{pl}/(m_{pl} + m_{pol})$, where m_{pl} is the mass of the plasticizer and m_{pol} is the polymer mass. We can see that, as the proportion of the plasticizer in the polymer composition increases, the Young modulus of magnetoelastics decreases. In this case, for the same value of ν, the increase in the mass fraction of magnetic particles in the magnetoelastic leads to an increase in the Young modulus.

Measurements on the samples exhibiting a strong magnetodeformation effect showed that their Young modulus is in the range 10^3–10^4 Pa, which is much smaller than that for the structuring (plasticizer-free) polymer, $E \sim 3.1 \times 10^5$ Pa.

Figure 3 depicts the schematic of the experimental setup used to study the effect of magnetic field on the elastic and viscous properties of magnetoelastics. To examine the elastic properties, the elongation of a cylindrical or ribbon sample placed in a uniform magnetic field H directed perpendicular to a stretching force F was measured depending on the strength of the force. After that, the dependence of the elastic stress $\sigma = F/S$ on the relative elongation of a sample $\Delta x = \Delta l/l_0$ (where $\Delta l = l - l_0$ is the sample elongation by the action of force F, l and l_0 are the length of the stretched and the unstretched sample, respectively; and S is the cross section of the sample) was calculated. Then, the Young modulus was determined from the plots of $\sigma(\Delta x)$ dependences found.

Note that when samples were placed in the magnetic field, an increase in their length was observed, which was as large as 20% of the original length for soft samples (having a small Young modulus). The results that are presented below have been calculated for the initial length l_0 corresponding to the equilibrium state of a sample (stretching force $F = 0$) in the presence or in the absence of magnetic field.

RESULTS AND DISCUSSION

It was found that the application of magnetic field leads to a substantial increase in the Young modulus. As an example, Fig. 3a shows the dependence of σ on Δx for a cylindrical magnetoelastic sample of 3.1 mm diameter with the mass concentration of the magnetic phase of 42% and the Young modulus $E = 6.9 \times 10^3$ Pa,

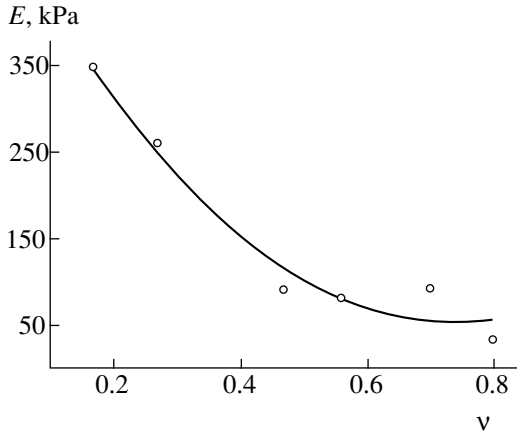

Fig. 2. Dependence of the Young modulus E on the plasticizer mass fraction ν in the polymer matrix for magnetoelastics with the mass fraction of 2-μm iron particles of about 30%.

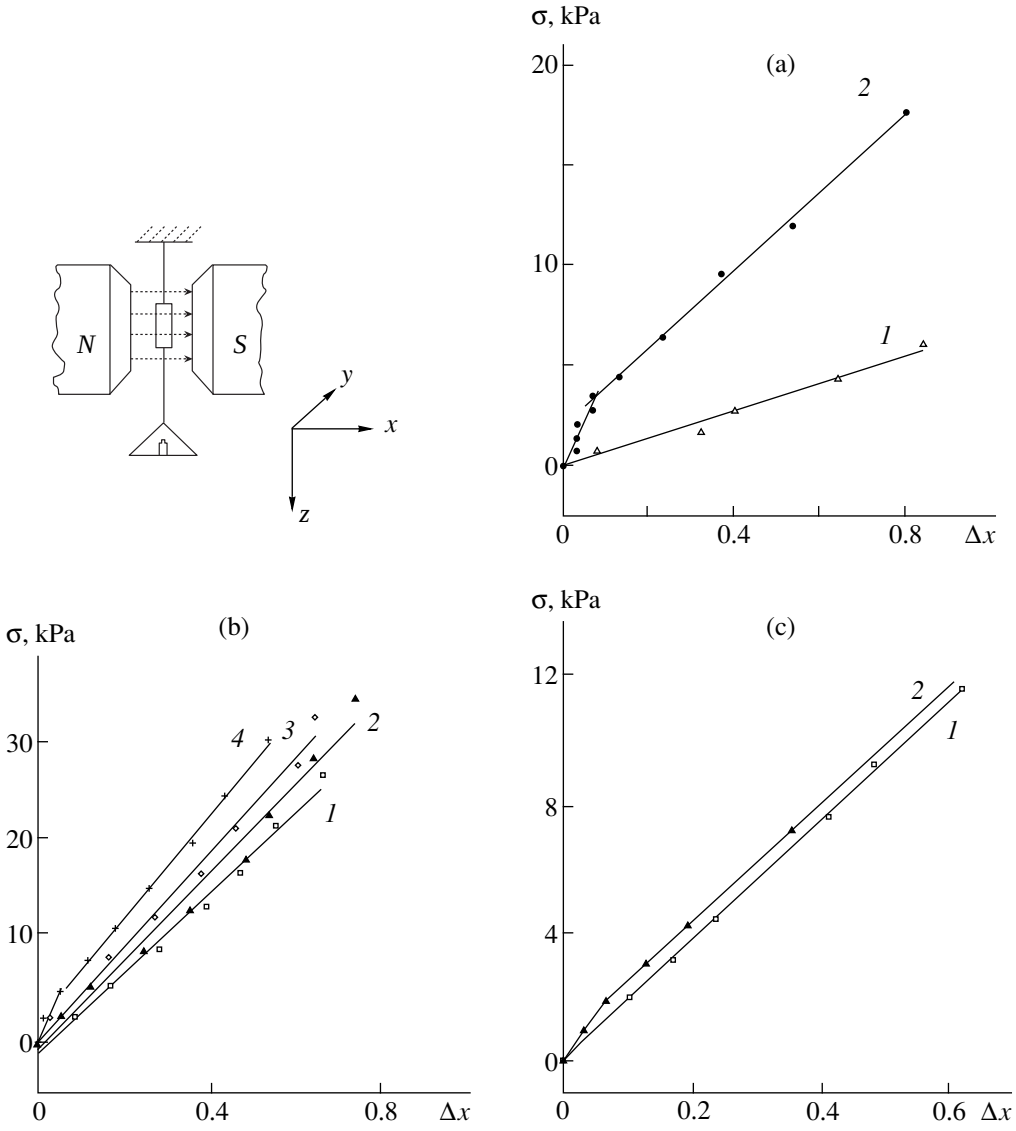

Fig. 3. Dependence of the elastic stress σ on the relative elongation Δ*x* for magnetoelastics with the magnetic-phase mass fraction of (a) 42, (b) 30 and (c) 24%, as measured in the absence of field and in different magnetic fields: *E* = (a) 6.9×10^3, (b) 3.8×10^4, or (c) 1.9×10^4 Pa. The insert shows the scheme of the experiment.

as measured in the absence of magnetic field (curve *1*). As the dispersed magnetic phase, iron particles of ~2 μm size were used. The dependence of the specific magnetization *j* of this magnetoelastic on the strength of the external magnetic field is linear in the fields lower than those of saturation magnetization (Fig. 4a).

As is seen from Fig. 3a, the dependence σ(Δ*x*) measured in the absence of the field shows the classical linear behavior. However, when the sample was placed in the magnetic field of *H* = 135.3 kA/m, the dependence σ(Δ*x*) became nonlinear, exhibiting two portions with the Young modulus different from the initial value (at *H* = 0), the modulus relevant to the initial stretching stage having the highest value.

Studying more "rigid" samples (with a higher Young modulus in the absence of the field) containing magnetic particles of the same size showed that the Young modulus also increases in value under exposure to the uniform magnetic field *H*, although the initial linear portion in the region of small (Δ*x*) is less pronounced and becomes noticeable only for the dependences measured at high field strengths *H*. As an example, Fig. 3b presents the dependences of σ on Δ*x* for a more rigid sample in the form of a $36 \times 3.6 \times 0.55$-mm strip cut from a magnetoelastic containing 30 wt % of iron particles of ~2 μm size and having the Young modulus $E = 3.8 \times 10^4$ Pa in the absence of magnetic field. The dependences were measured in the absence (curve *1*) and in the presence of a magnetic field of *H* = 58.1, 106.7,

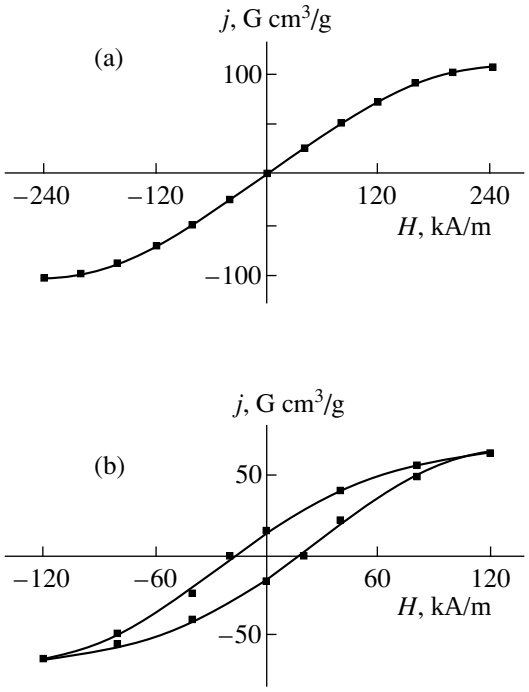

Fig. 4. Dependence of the specific magnetization j on the magnetic field strength H for magnetoelastics with iron particles of (a) 2 μm and (b) 110 Å size.

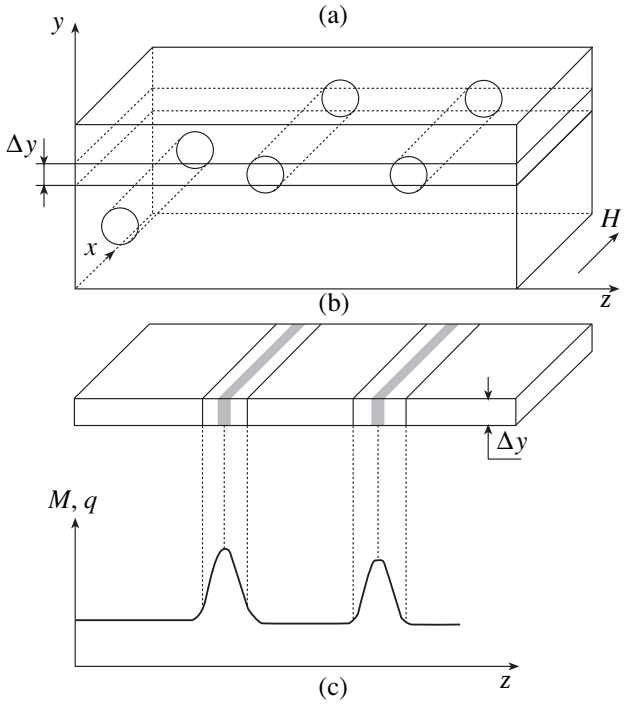

Fig. 5. Schematic representation of (a) the structure of a magnetoelastic placed in a uniform magnetic field, with cylindrical contracted regions of the polymer matrix; (b) thin layer of Δy thickness; and (c) magnetization and concentration distributions along the direction z. The shaded area shows the region with an increased concentration of magnetic particles that is most resistant to stretching.

or 135.3 kA/m (curves *2*, *3*, and *4*, respectively). It can be seen that the existence of two linear portions is displayed in the most pronounced way on the dependence measured at H = 135.3 kA/m. The pattern of the dependence of specific magnetization j on the external magnetic field strength resembles that for the dependence depicted in Fig. 4a.

In the study of samples with finer particles, two linear portions were also observed, the slope of the second portion being virtually the same as that of the curve for the dependence $\sigma(\Delta x)$ measured in the absence of magnetic field. As an example, Fig. 3c depicts the dependences of σ on Δx measured for a $28 \times 5.5 \times 1$-mm ribbon sample cut from a magnetoelastic containing 24 wt % of iron particles of 110 Å size and having the Young modulus $E = 1.9 \times 10^4$ Pa in the absence of magnetic field. The magnetic characteristics of this magnetoelastic are presented in Fig. 4b.

To explain the obtained experimental results, we considered a model that suggests the occurrence of internal structuring of magnetic particles when the sample is placed in the uniform magnetic field. The dipole–dipole interaction forces acting in the magnetic field drive the particles out of their initial positions, thus deforming the rubbery polymer matrix. The interaction energy of two magnetized particles is described by the following interaction potential [13]:

$$U_{12} = (\mu_0/(4\pi r^3))[\mathbf{m}_1\mathbf{m}_2 - 3(\mathbf{m}_1\mathbf{r})(\mathbf{m}_2\mathbf{r})/(r^2)], \quad (1)$$

where μ_0 is the magnetic permeability of the vacuum, $\mathbf{m}_1$ and $\mathbf{m}_2$ are the magnetic moments of the particles, and $\mathbf{r}$ is the vector connecting the centers of the particles. In the case of parallel magnetic moments in an orienting external magnetic field, the minimum of dipole–dipole interaction energy corresponds to aligning the particles along the same line.

The phenomenon of aligning magnetic particles in chains directed along magnetic field lines is well known in the physics and chemistry of magnetic powders, ferrovarnishes, and magnetic fluids and colloids. Magnetic particles occurring in a magnetoelastic and attracting one another also tend to form chains. However, the polymer matrix precludes their association, and cylindrical contracted-polymer regions stretched along the field lines and contracted by magnetic forces F_m from all sides are formed in the sample (see Fig. 5a which schematically shows the theoretically predicted structure of a rectangular-band sample of a magnetoelastic placed in a uniform magnetic field). The concentration of magnetic particles in these cylindrical regions is higher than in elongated regions, which is expressed by a rise in local magnetization and in the value of magnetic charges at the face ends of contracted cylindrical regions. We believe that it is the interaction of like poles emerged at the face ends of the contracted cylindrical regions which is responsible for the experimentally observed elongation of the magnetoelastic in the direction z perpendicular to the external magnetic

field. In high magnetic fields of the order of 500–600 kA/m, the repulsion forces between the cylindrical regions are so large in this case that a characteristic multiple-strand relief can appear on the sample surface parallel to the magnetic field. Figure 6 presents the photographs of the lateral surface of a cylindrical magnetoelastic sample containing 42 wt % of ~2-μm iron particles and having the Young modulus $E = 6.9 \times 10^3$ Pa, in the fields of $H = 16$ and 557.2 kA/m.

To simplify consideration of the elastic properties of such nonuniformly deformed materials, let us divide the sample shown in Fig. 5a into thin layers of Δy thickness each (Fig. 5b). We see that, in the selected layer, the contraction and elongation regions of the polymer matrix alternate along the z axis. If the distribution of cylindrical regions throughout the sample is homogeneous, the structure of all layers will be identical and the behavior of any of them will represent that of the sample as a whole.

Figure 5c schematically shows the dipole–dipole interaction-induced redistribution of concentration of magnetic particles and, hence, magnetization M of the magnetoelastic in the z direction.

When a unidirectional stretching force F is applied along the z axis, the equilibrium condition for originally elongated regions can be defined as the equality of the external stretching force F to the elastic force F_{el} that results from the stretching of these regions

$$F = F_{el}. \quad (2)$$

In the magnetoelastic regions originally contracted by magnetic forces, the magnetic contracting forces F_m acting upon the particles are counterbalanced by the elastic forces of the contracted ($F^*_{el, con}$) and elongated (F_{el}) polymer operating at the boundary of cylindrical regions on the side of contracted and elongated magnetoelastic regions. Note that the strength of the elastic contracting forces of the polymer matrix differs from that of the elastic stretching forces. For example, the compression modulus (compressibility) of rubber is the same as that of a liquid, of the order of 2×10^9 Pa. However, the Young modulus (about 10^6 Pa) is almost 2000-fold smaller and it is $\sim 2 \times 10^5$ times weaker than the Young modulus of a typical solid [14].

The condition of equilibrium at the boundary of originally contracted regions can be defined by

$$F_m = F_{el} + F^*_{el, con}. \quad (3)$$

Taking into account that relationship (2) suggests the elastic force F_{el} appearing in the stretched regions to be equal to the external applied force F, Eq. (3) can be rewritten as

$$F = F_m - F^*_{el, con}. \quad (4)$$

From Eq. (4), it follows that, as the applied force F increases, the elastic compression force $F^*_{el, con}$ must first decrease to zero and, after that, the originally contracted regions start to elongate. The equilibrium conditions in this case will be as follows:

$$F = F_m + F^*_{el, str}$$

or

$$F^*_{el, str} = F - F_m, \quad (5)$$

where $F^*_{el, str}$ is the elastic force of the magnetoelastic region that has been originally contracted but is currently stretched by the force F.

The total elongation of all originally stretched regions by the action of the applied force F can be given by

$$\Delta l_1 = F l_0 (1 - k_1)/(SE) \quad (6)$$

where k_1 is the volume fraction originally occupied by contracted regions in the layer Δy under consideration,

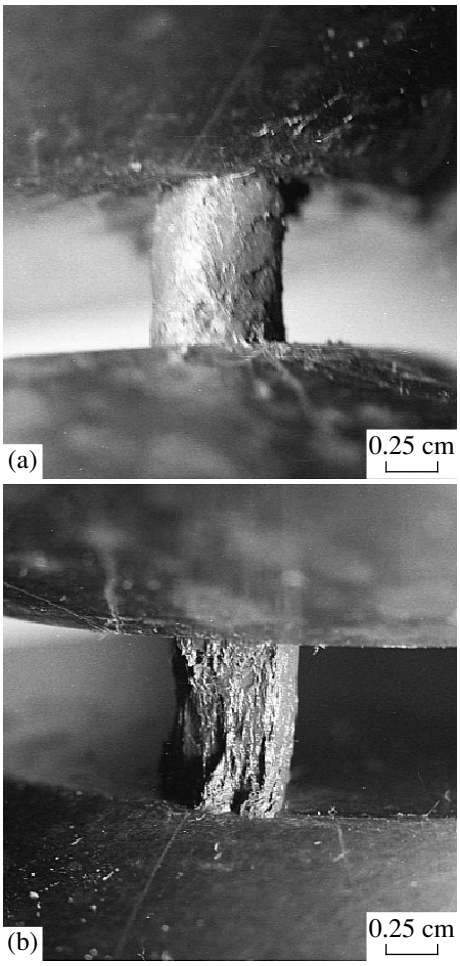

Fig. 6. Photographs of the lateral surface of a magnetoelastic sample in uniform magnetic fields of (a) 16 and (b) 557.2 kA/m.

S is the cross section of the layer, and E is the Young modulus of the stretched polymer.

The total elongation of all originally contracted regions before their transition from the contracted to the stretched state will be denoted by

$$\Delta l_2 = \varepsilon, \quad (7)$$

where ε is an infinitesimal due to low compressibility of the polymer in the cylindrical contracted regions under consideration.

The Young modulus E_1 experimentally measured in the region of the initial linear dependence for such a structure prior to the transition of the contracted regions to the stretched state can be represented as follows:

$$E_I = Fl_0/(S(\Delta l_1 + \Delta l_2)). \quad (8)$$

After the substitution of (6) and (7) in Eq. (8), we get

$$E_I = E/(1 - k_1). \quad (9)$$

From Eq. (9), it follows that the magnetic interaction resulting in the formation of the contracted polymer regions is also responsible for the increase in the Young modulus observed experimentally in the region of small displacements Δx. When the stretching force F (Eq. (5)) becomes greater than a certain polymer-contracting magnetic force F_m^*, the elongation of the originally contracted regions begins, which is manifested in a decrease in the experimentally measured Young modulus down to a value that may be represented as follows for the second linear portion of the dependence:

$$E_{II} = E(F - F_m^*[1 - F(k_1 - k)/(ES_0)]) / (F(1 - k) - F_m^*(1 - k_1)). \quad (10)$$

Here F_m^* is the magnetic force at which the originally contracted region begins to elongate and k is the volume fraction of contracted regions that still exist at a given stretching force F.

In the most general case, the complete degradation of the contracted polymer regions is attainable only in the absence of the external magnetic field. If $F \gg F_m^*$, the Young modulus E_{II} of the second linear part tends to the value

$$E_{II} = E/(1 - k), \quad (11)$$

which becomes equal to the Young modulus measured in the absence of the magnetic field, $E_{II} = E$, in the case of weak magnetic interaction of particles and complete stretching of contracted regions by the applied force F.

Equations (9) and (10) obtained for the two experimentally observed linear parts of the dependence of the Young modulus show that, in the initial stage of sample stretching when large contracted polymer regions exist in the magnetic field, the measured Young modulus is much greater than the original one observed under field-free conditions.

Then, as the applied stretching force F increases, the volume fraction of the contracted polymer decreases and we arrive to the second linear part of the dependence of σ on Δx characterized by a insignificant change in the volume fraction of the still contracted polymer that requires considerable efforts for its stretching. In this case, depending on whether it is possible to completely stretch the material in the second linear region, thus decreasing to a considerable extent the magnetic interaction forces, or there are the polymer regions that are still contracted by the magnetic interaction of the particles, the Young modulus E_{II} will either be equal to the modulus measured in the absence of the field or exceed it, respectively.

The results discussed above pertain to the stretching of a thin magnetoelastic layer of Δy thickness. To find the E_I and E_{II} values for the material of an arbitrary volume, it is necessary that the interaction between adjacent layers be additionally taken into account; fortunately, the simple model used in this work allows the elastic properties of different magnetoelastics to be qualitatively described and compared with one another.

If the values of Young modulus in the region of small deformations Δx in the presence and in the absence of field are known, it is possible to find the volume fraction of originally contracted polymer regions k_1, which allows speculations on the character of interaction of magnetic particles in a magnetoelastic.

We determined the contracted-polymer volume fractions k_1 for the samples whose $\sigma(\Delta x)$ dependences are shown in Fig. 3. For the magnetoelastics containing 42% (Young modulus in the absence of field $E = 6.9 \times 10^3$ Pa) and 30% by mass ($E = 3.8 \times 10^4$ Pa) of ~2-μm iron particles and 24 wt % of 110-Å iron particles ($E = 1.9 \times 10^4$ Pa), the volume fractions of the contracted polymer calculated by Eq. (9) are 0.85, 0.64, and 0.36, respectively.

As follows from the results obtained at an identical size of rather coarse magnetic particles (~2 μm), the higher the concentration of magnetic particles, the greater the initial volume fraction of a contracted polymer. As the stretching force F increases, the volume fraction of the contracted polymer decreases, and hence, the Young modulus measured in the magnetic field decreases down to a new value E_{II} (Eqs. (10) and (11)). Figure 7a presents the results of consideration of the dependence of the Young moduli E_I and E_{II} on the external uniform magnetic field H for a sample in the form of a $36 \times 3.6 \times 0.55$-mm^3 strip cut from the magnetoelastic containing 30 wt % of ~2-μm iron particles with the Young modulus $E = 3.8 \times 10^4$ Pa in the absence of magnetic field. The $\sigma(\Delta x)$ curves for this material, as measured in the fields of H = 58.1, 106.7, and 135.3 kA/m, are depicted in Fig. 3. As follows from the data presented in Fig. 7a, the Young modulus E_I of the magnetoelastic increases in a nonlinear manner with increasing the external magnetic field that structures

the material. The substantial increase in E_I is due to the rise in magnetization of multidomain iron particles and, consequently, the resultant enhancement of the dipole–dipole interaction. These factors also affect the similar dependence of E_{II} on H.

The dependences of the residual volume fraction k of contracted polymer regions on the strength of the structuring magnetic field for two different stretching forces F corresponding to the second linear part are shown in Fig. 7b, as calculated by Eq. (10). It is seen that the external magnetic field prevents the stretching of originally contracted regions of the polymer matrix, which is manifested in the rise in E_{II} with increasing the external magnetic field. The pattern of the dependence of the volume fraction k of contracted polymer regions on the stretching force F for two values of magnetic field is shown in Fig. 7c. It is seen that, the effect of the force F on k is not so profound in the second linear part of the $\sigma(\Delta x)$ curve and reduces to some decrease in the residual volume fraction of the contracted polymer.

The magnetoelastic with fine magnetic particles (~110 Å) exposed to the magnetic field of H = 135.3 kA/m has an insignificant volume fraction of the originally contracted polymer (k_1 = 0.36). In this case, as the stretching force F increases, almost complete stretching is achieved for the magnetoelastic, for which the Young modulus E_{II} is close to the modulus E in the absence of the field.

Such a behavior of the magnetoelastic seems to be due to weaker dipole–dipole interaction forces between finer magnetic particles having a smaller magnetic moment. Moreover, additional investigations showed that fine magnetic particles are weaker bound to the polymer matrix, which can also somewhat undermine the interaction of the magnetic subsystem with the elastic matrix.

The analysis of the free oscillations of a spring pendulum in which a cylindrical magnetoelastic sample was used as the spring showed that intense damping of oscillations was observed for some samples when a uniform magnetic field was applied across the sample. For example, the number of vibrations after displacement from the equilibrium position until complete damping decreased by 40% in the magnetic field of H = 58.1 kA/m for the pendulum made of the magnetoelastic comprising 2-μm magnetic iron particles with E = 4.2×10^3 Pa and having a length of ~40 mm and an oscillation period of 0.4 s. However, a similar pendulum with close geometric dimensions and the same oscillation period but made of the magnetoelastic having an iron particle size of 110 Å and the Young modulus $E = 1.6 \times 10^4$ Pa produced an almost equal number of vibrations before the decay at an equal displacement from the neutral position in the presence of a field of H = 58.1 kA/m and in the absence of the field.

The experimentally observed decrease in the number of vibrations of the pendulum before its full stopping as compared to that in the absence of magnetic field indicates an increase in the intrinsic viscosity of magnetoelastics in the magnetic field. The increment in viscosity correlates with the rise in the volume fraction of the contracted polymer in this case.

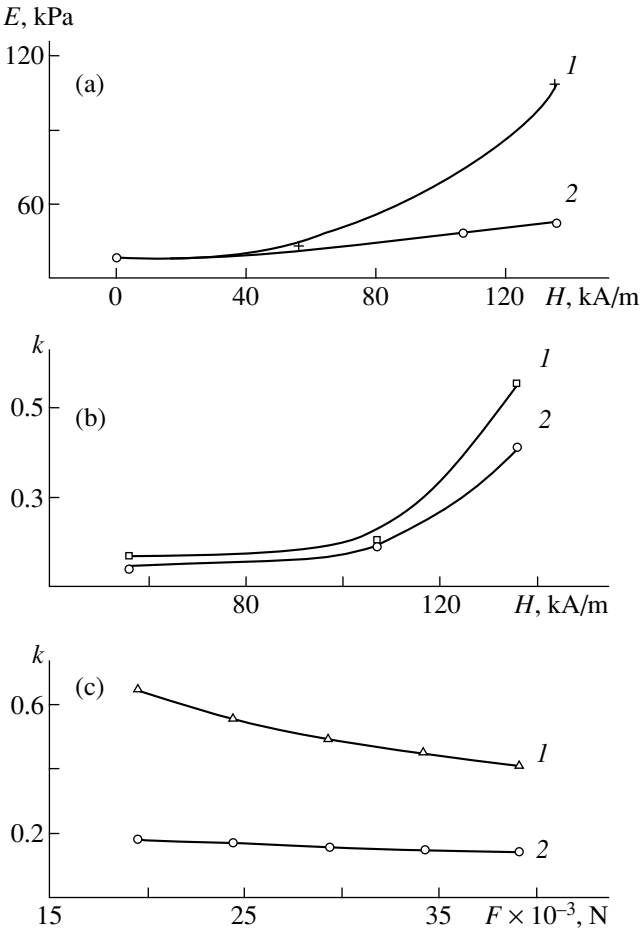

Fig. 7. Dependence of (a) the Young moduli (*1*) E_I and (*2*) E_{II} on the strength of external magnetic field. The volume fraction k of compressed regions as a function of (b) magnetic field at F = (*1*) 24×10^{-3} or (*2*) 30×10^{-3} N and (c) stretching force F in two different magnetic fields at H = (*1*) 135.3 and (*2*) 58.1 kA/m.

CONCLUSIONS

An external magnetic field was experimentally shown to have a strong effect on the viscous and elastic properties of magnetoelastics, and a model for describing the magnetic field effect on the elastic characteristics of these materials was proposed. In terms of this model, expressions were derived for the Young modulus in the regions of two experimentally observed linear parts of the dependence $\sigma(\Delta x)$ in a uniform magnetic field.

Based on the experimental data obtained, the influence of the external magnetic force and a force F

applied to stretch a magnetoelastic on the volume fraction of regions contracted by the magnetic interaction of particles was considered. The effect of increasing the viscosity by the external magnetic field was revealed for magnetoelastics with a large volume fraction of contracted polymer.

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