RELAXATION OF VISCOUS STRESSES IN MAGNETORHEOLOGICAL SUSPENSIONS

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The kinetics of structural conversions in magnetic suspensions is important from the practical point of view as a factor capable of limiting the high-speed operation of devices based on the magnetorheological effect [1], and additionally, this factor has not been studied extensively. Viscosity η and magnetization M are the fundamental macroscopic characteristics of suspensions sensitive to structural changes. It is precisely the characteristic time of structure formation that determines the duration of the magnetic aftereffect in suspensions of multidomain particles of magnetically soft carbonyl iron, since the magnetization of the particles themselves is characterized by the time $\tau_0 \lesssim 10^{-6}$ [2]. In this case, in weak external fields H ($\approx$ to 10^3 A/m), according to the data of the experiment described in [3], the slow portion of the magnetization process amounts approximately to 50% of the equilibrium magnetization value. In the structuring process the magnetization of the suspension in a weak field is doubled. As the field increases, the contribution of the structuring to magnetization will obviously diminish; the reduction in the role of the mechanisms involved in the prolonged aftereffect with increased field strength is a general property of ferromagnetic materials [4]. The information provided by magnetic measurements relative to the state of the structure as the field grows is therefore diminished. Conversely, the effective viscosity of the suspension, regardless of field, is determined precisely by its structure. Indeed, the contribution of individual spherical magnetically soft particles (where they are magnetized in a shearing flux) to the effective viscosity amounts to $\approx\mu_0 H\tau_0$ and for typical values of M = 10^5 A/m, H = 10^5 A/m does not exceed 10^{-4} kg/m·sec.

The study which we carried out into the viscous aftereffects of magnetic suspensions of carbonyl iron in hydraulic aviation oil thickened to impart sedimentation stability to the suspension allowed us to establish the fact that in addition to the rapid process of structure formation characterized under typical conditions by a time scale of 10^{-4}—10^{-3}sec, a slow process takes place within a suspension in a state of motion: over tens of seconds the effective viscosity changes by as much as 10%. Quantitative estimates and the simplest modeling of the dynamic structure allow us to draw the conclusion that the vast portion of the viscous aftereffect is associated with the formation of the primary structure through the approach of the particles to each other under the forces of magnetic interaction. However, the slow process is associated with the transition of the primary state of the suspension, as a result of hydrodynamic mixing, to a state of dynamic equilibrium, with external nonequilibrium conditions.

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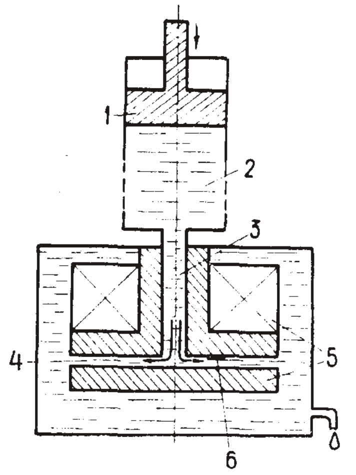

Fig. 1. Diagram of installation to record the fast portion of the viscous aftereffect in the hydraulic circuit. Explanations in text.

The Fast Portion of Viscous Aftereffect. The time constant of traditional viscosimeters amounts approximately to 0.5 sec, so that they are not suited to study fast aggregation processes. We recorded the fast portion of the aftereffect on the basis of the transient response within the hydraulic circuit, as illustrated schematically in Fig. 1. Piston 1, moving at a constant velocity in hydraulic cylinder 2 of diameter 0.05 m and length 0.7 m, moves the suspension through channel 3, which has a diameter of 0.01 m and length 0.2 m, to slit gap 4 between the poles of electromagnet 5, fabricated out of high-frequency ferrite, and it is drained off into a vessel (not shown here). The pole width is 0.05 m and the slit width is 0.001 m. Piezoelectric sensor 6, connected to an amplifier with a high-impedance input with a time constant of 2 sec, measures the change in pressure which arises on application of the current to the windings of the electromagnet. Calibration shows that the sensor exhibits a linear amplitude-frequency characteristic in the frequency range 0.5—10^5 Hz. In the steady-state regime the pressure at the inlet to the slit is proportional to the rate at which the suspension flows and to the hydraulic drag produced by the slit, the latter proportional to the effective viscosity of the suspension. The typical transient response recorded by the pressure sensor after the sudden application of the magnetic field is illustrated by the oscillograms in Fig. 2.

Figure 2a shows oscillograms of the pulses in pressure H and the oscillogram for the change in the field (for a single field value of 100 kA/m). We see that the time of field growth in the inductor amounts approximately to 50 μsec, which enables us to examine the transient responses in the hydraulic conduit in the frequency range below 10^3 Hz. As we can see, the pressure begins to increase after a slight delay lasting about 0.3 msec and the monotonic increase in pressure becomes oscillational in weak fields as the intensity of the field increases. Figure 2b shows oscillograms for the change in the pressure within the hydraulic conduit where the fluid is acted upon by a field pulse of H = 100 kA/m. The span of the oscillations increases in conjunction with the flow rate. The fluctuations in pressure are associated, obviously, with the finite velocity at which the perturbations are propagated through the fluid. Indeed, the period of the oscillations, with a magnitude of about 1 msec (see Fig. 2c), is in agreement with the time estimate required for the perturbations to pass through a cylinder of length 1 m at a speed of sound 10^3 m/sec. The pressure fluctuations give evidence to the effect that the characteristic time τ_η of the viscous aftereffect is smaller than the perturbation-propagation time t′ in the system under consideration. The absence of oscillations in weak fields may indicate that we are dealing here with an inverse relationship: $\tau_\eta > t'$. Let us also note that the abscissa of the first maximum in pressure is shifted in the direction of shorter times as field strength is increased; however, as the field strength is increased it ceases to change. This also indicates that the characteristic time of the viscous aftereffect depends on the field and under these conditions changes from $\tau_\eta > t'$ to $\tau_\eta < t'$ as field strength increases.

We also examined the viscous aftereffect under conditions of Couette flow. The nonmagnetic plate was shifted in these experiments at a fixed velocity through the gap formed by the ferrite tips of the electromagnet, where the gap was filled with the suspension. The increment in viscous stresses in the sudden actuation of the field was determined by means of a piezoelectric sensor. The duration of the transient response in the inductor of the magnetic field amounted to 50 μsec. The typical process of viscous aftereffect, recorded by the piezoelectric sensor after actuation of the field, is shown in the oscillogram (Fig. 3) for H = 100 kA/m. As in the hydraulic conduit, it is characterized by a time delay of ~10^{-4} sec. The increment in pressure occurs within ~$2\cdot10^{-4}$ sec. This is smaller than the period of mechanical oscillations within the measurement system (~$3\cdot10^{-4}$ sec). Thus, the velocity of the viscous response of the magnetic suspension, recorded in these experiments, exceeds the speed with which the signal is propagated through the structural elements. The cited results make it possible to formulate the following concept dealing with the fast

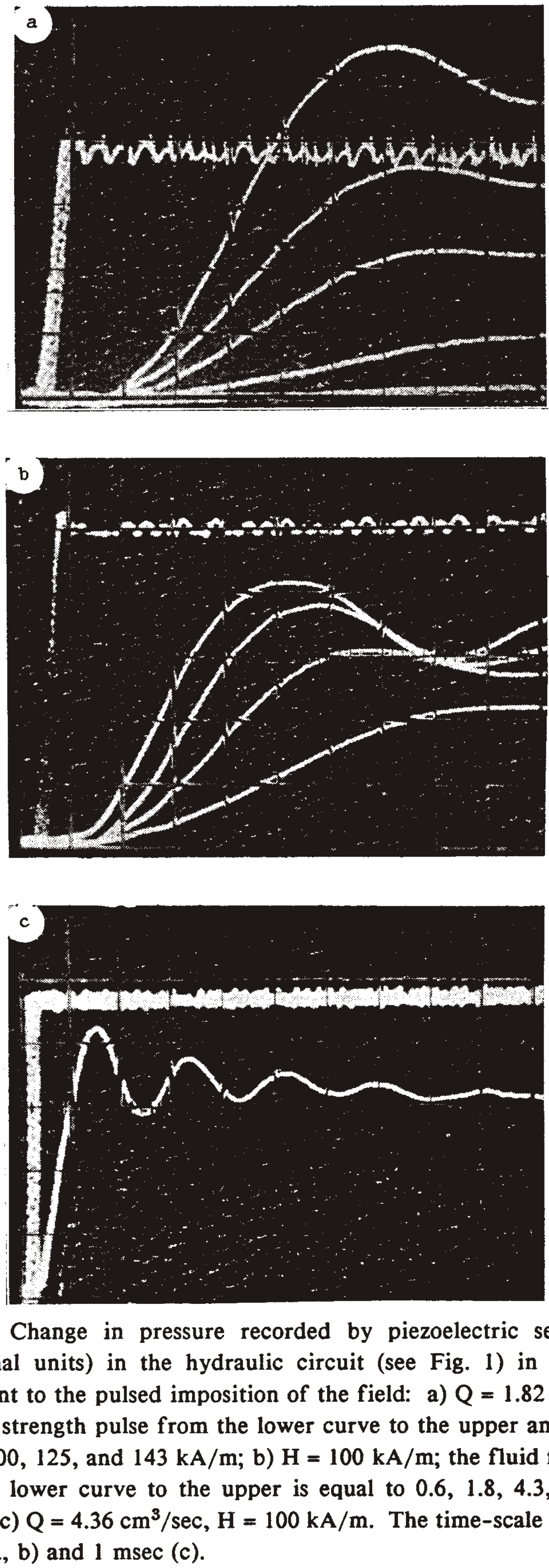

Fig. 2. Change in pressure recorded by piezoelectric sensor (in conditional units) in the hydraulic circuit (see Fig. 1) in the time subsequent to the pulsed imposition of the field: a) Q = 1.82 cm^3/sec; the field strength pulse from the lower curve to the upper amounts to 50, 75, 100, 125, and 143 kA/m; b) H = 100 kA/m; the fluid flow rate from the lower curve to the upper is equal to 0.6, 1.8, 4.3, and 9.0 cm^3/sec; c) Q = 4.36 cm^3/sec, H = 100 kA/m. The time-scale divisions are 0.2 (a, b) and 1 msec (c).

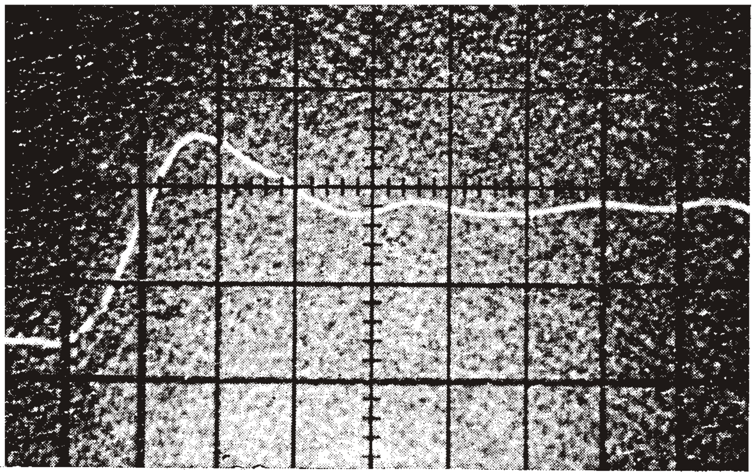

Fig. 3. Change in pressure recorded by the piezoelectric sensor (in conditional units) within the time governed by the increment in viscous stresses in the Couette flow after the jumpwise actuation of the field. The time-scale division is 0.1 msec.

component of the suspension structuring process. Since the magnetization of individual magnetically soft particles, as is demonstrated above, provides no marked contribution to the effective viscosity, the observed delay is apparently defined by the time required for the appearance of the smallest, i.e., paired, aggregates. We will evaluate it in terms of the time required to cause the particles found, at the instant of field actuation, at a distance r_0 from each other, averaged for the given concentration, to approach each other. If the particle radius is a, then n (the number of particles per unit volume) is associated with the volumetric concentration φ by the relationship $n = 3\varphi/(4\pi a^3)$, while $r_0 = n^{-1/3} = a(4\pi/3\varphi)^{1/3}$. The nearing of the particles to each other is determined by the equation for the balance of forces between interparticle interaction and viscous friction. In the dipole approximation (here the mutual perturbation of particle magnetization is neglected) the force of interaction between particles exhibiting magnetic moment m_0 and positioned in the direction of the field is $6\mu_0 m_0^2/r^4$. Using the Stokes formula $f = 6\pi\eta_0 a d(r/2)dt$ for the viscous force, we will write this equation of particle approach in the form

$$2\mu_0 m_0^2/r^4 + \pi\eta_0 a\, dr/dt = 0.$$

Here η_0 is the viscosity of the carrier fluid and r is the distance between the particles. The solution of this equation, with the initial condition $r(0) = r_0$, is

$$r^5 = r_0^5 - (10\mu_0 m_0^2/\pi\eta_0 a)\, t.$$

From this, for the time $\tau_p = t(2a)$ of pair approach it follows that

$$\tau_p = (\pi\eta_0 a/10\mu_0 m_0^2)\,[r_0^5 - (2a)^5].$$

For moderate concentrations, the second term in the brackets is considerably smaller here than the first, so that we have the formula

$$\tau_p = \frac{\eta_0 a^6 \pi^{8/3}}{10\mu_0 m_0^2}\left(\frac{4}{3\varphi}\right)^{5/3}$$

In weak magnetic fields, when we take into consideration the magnetization factor for a sphere, the particle moment is $m_0 = 4\pi a^3 \chi_0 H/[3(1 + \chi_0/3)]$. Since the initial susceptibility χ_0 of the particle material is considerably larger than unity, we have the possibility here of $\chi_0 \to \infty$, which leads to $m_0 = 4\pi a^3 H$. In a strong field ($H \gg M_s$; M_s is the saturation magnetization of the particle) $m_0 = (4\pi/3) M_s a^3$. Thus, for the structural delay-time asymptotes in weak and strong fields we have, respectively,

$$\tau_p = \begin{cases} 0{,}02\eta_0/(\mu_0 H^2 \varphi^{5/3}) & (H \ll M_s); \\ 0{,}2\eta_0/(\mu_0 M_s^2 \varphi^{5/3}) & (H \gg M_s). \end{cases} \quad (1)$$

Assuming the $\eta_0 = 1$, $\varphi = 0.1$, and H = 10 kA/m, according to (1) we have the estimate $\tau_p = 3 \cdot 10^{-4}$ sec. This is precisely the order of magnitude for the delay observed experimentally. The time τ_{st} needed to form the developed primary structure is defined as the time needed to propagate a chain of particles through the entire volume of the suspension. If its linear dimension is represented by l, then there will be $l/2a$ particles in the chain. If the growth of the chain is represented as a series of p doublings of its length, we can then write $\tau_{st} = p\tau_p$. The quantity p is found from the equation $2^p = l/2a$, from which we have $p = \log(l/2a)/\log 2$. In our case $l = 10^{-3}$ m, $a = 3 \cdot 10^{-6}$ m, and this gives us p = 7, so that for the appearance of the developed primary structure we need a period of time that is greater by an order of magnitude than the time τ_p required for the formation of the pair, i.e., the delay time. It is precisely this that is observed experimentally, with the establishment of pressure monotonic in nature (see Fig. 2).

The Slow Portion of the Viscous Aftereffect. The estimates of the characteristic time of the fast aftereffect, such as those presented above, were derived for a quiescent suspension. As is well known, the shearing flow promotes coagulation by causing the particles to collide (see [5]). Based on the Smolukhovskii formula, the number of encounters between noninteracting spherical particles per unit volume per unit time is estimated as follows ($\dot{\gamma}$ is the shearing velocity)

$$N = (32/3) n^2 \dot{\gamma} a^3.$$

Consequently, $\approx \dot{\gamma}\varphi$ collisions per second are encountered by a single particle, so that the characteristic time of hydrodynamic particle approach is given by

$$\tau_\eta = 1/\dot{\gamma}\varphi. \quad (2)$$

For typical values of $\dot{\gamma} = 10^2$ sec^{-1}, $\varphi = 0.1$, we have $\tau_\eta = 10^{-1}$ sec. This is considerably greater than the characteristic time τ_{st} for the appearance of structure ascribed to magnetic forces. However, in weak fields (H ≤ 10 kA/m) and for large shearing velocities ($\dot{\gamma} \gtrsim 10^4$ sec^{-1}) or considerable viscosities the times τ_η and τ_{st} may become comparable. The magnitude of the ratio

$$k = \tau_{st}/\tau_\eta = 0{,}02 p \dot{\gamma} \eta / \mu_0 H^2 \varphi^{2/3} \quad (3)$$

must play an important role in the viscous aftereffect of the suspension. If $k \ll 1$, then as the field is actuated the essentially steady initial structure examined above can be formed within the suspension, and it is then transformed within the flow into a new state of dynamic equilibrium. Conversely, if $k \gg 1$, the dynamic structure will be formed out of particles from the dispersion phase, said particles uniformly distributed through the volume of the fluid.

How are we to describe the nature of the viscous aftereffect, measured in tens of seconds? We should take note of the fact that existing concepts as to the nature of a dynamic structure in magnetic suspensions are rather meager. These concepts are based on models of a "quasihard" chain or ellipsoidal elongated structural element, whose effective parameters are defined by the balance of magnetic and hydrodynamic forces. A method of direct numerical investigation into the behavior of an ensemble of interacting magnetic particles suspended in a viscous medium was first used in [6] to study the processes in a dynamic structure of magnetic suspensions. From the simplest examples of two particles in a rotating field, dealt with in [6], already we encounter a need to reexamine existing concepts as to the dynamic structure of the suspensions: in an ensemble of interacting particles under nonsteady conditions, in combination with its macroscopic displacement (rotation), we find complex relative displacements of particles (oscillations). It is important that up to one-half of all of the dissipated energy be expended to maintain the internal motions in the particle associations.

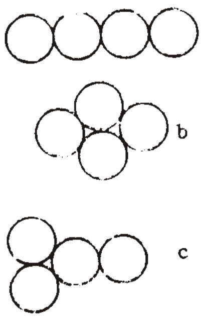

Fig. 4

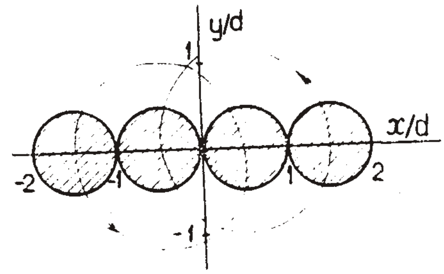

Fig. 5

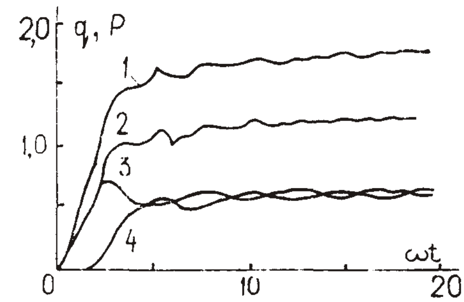

Fig. 6

Fig. 4. Equilibrium structures of four magnetically soft particles in a strong magnetic field. Explanations in text.

Fig. 5. Trajectories associating four magnetic particles in a rotating field.

Fig. 6. The process of establishing the force moment q (1) and energy dissipation P (2) and its rotational P_ω (3) and oscillatory P_r (4) components for the association of four particles in the case of a rotating field with $\omega/\omega_* = 0.7$.

The study of an association consisting of four particles in a rotating field, said study carried out under the same assumptions as in [6] (i.e., the magnetic moment is fixed by the field and exhibits no orientational relation to the body of the particle, with the viscous force for each particle determined without consideration of other particles, on the basis of the Stokes formula), allows us to formulate certain additional concepts as to the nature of the dynamic structure. The steady-state particle-association trajectory depends on the initial positions of the particles, and here we are dealing with several types of trajectories, each of which is associated with one of the possible configurations of an equilibrium cluster, these configurations specified as the initial state (Fig. 4). Each of these makes a contribution that is different from the others to energy dissipation and torque.

In qualitative terms, the motion of an association of particles under the initial conditions (see Fig. 4a) is characterized by the following. At low frequencies, the chain is in rotation synchronously with the field, bending all the more markedly as the frequency increases all the way to destruction. The formed "cloud" of particles moves in complex fashion. It is interesting to note that the paired aggregate in an association of four particles, after destruction of the chain, is unstable: the destruction of a large aggregate does not result in the appearance of smaller stable aggregates. Figure 5 shows the trajectory of a chain made up of four particles, destroyed at a frequency of $\omega = 0.3\omega^*$, where $\omega^* = \pi M_s/18\eta$ is the limit frequency for the rotation of the particle pair synchronous with the field. The motion is illustrated here within a coordinate system rotating together with the field, the latter being directed along the x axis and rotating in a clockwise direction (d = 2a is the particle diameter).

If the field frequency is suddenly changed during the process in which the chain is destroyed in rotation, cluster formation becomes possible (see Fig. 4b). The cluster turns out to be stable at all velocities of rotation. At low frequencies it rotates synchronously with the field and as it attains a critical frequency it becomes asynchronous. Here, radial oscillations arise within the cluster, during the course of which pairs of two opposite particles appear alternately at the center.

The above allows us to draw the conclusion that stable formations do not arise in the suspension. The dynamic state is apparently characterized by the presence of a spectrum of temporary associations interchanging particles. In this case, even in an ensemble consisting of four particles, the time required to reach a steady-state trajectory after destruction of the initial equilibrium structure (of the chain) is quite great, i.e., it exceeds by an order of magnitude the time required to form this given structure. This is illustrated in Fig. 6, which shows the relationships between the rate of total energy dissipation P, its rotational and oscillatory components P_{ω} and P_r, as well as the resulting moment of forces q, averaged over the trajectories of motion during the time that the field is active. The quantity ω_*^{-1} serves as a scale of time, and it is approximately equal to the characteristic time for particle-pair approach when $\varphi = 0.1$. The dimensions P and q are the maximum values for the corresponding quantities at the limit of synchronous pair rotation (see [6]). Attention should be drawn to the fact that rotational dissipation diminishes from the onset of chain destruction; however, total dissipation and the moment of forces continue to increase as a consequence of oscillational dissipation.

Direct observation of structuring in the thin layer of a suspension where a field is quickly applied and subsequently repeatedly reversed provides an excellent idea as to the dynamic processes that occur within magnetic suspensions. The resulting chains are neither strictly linear nor of the single-particle variety. There are constrictions and branchings (jumpers). The linear ordering is all the more pronounced, the greater the field intensity. In weak fields, when the susceptibility of the particle material is at its maximum, the formation of bunched strands rather than of chains becomes useful as a consequence of magnetic reflection. The appearance of branching, as is easily seen, is facilitated by the larger of the particles, whose number, however, is small. Such particles serve as structural centers and are able to attract a number of smaller particles to themselves, and to each of these a chain may become attached, producing a branch or jumper. In the course of multiple field reversal the original "porous" structure becomes increasingly dense, the branches disappear, and strictly linear dense bunched strands appear.

This indicates that the initial structure is not the most advantageous from the standpoint of energy. It is quite clear that it may become such not only as a consequence of the effect of the variable field, but also in hydrodynamic fashion. This may be one of the reasons for the observed increment in viscous stresses during the process of motion. The concepts formulated here allow us to point to two possible mechanisms for the prolonged viscous aftereffect.

One of these is related to the presence of a broad spectrum of associated particle states under dynamic conditions, as well as with the magnitude of the distance separating the dynamic equilibrium spectrum of states from the original. In order to cover this distance a single collision between the elements of the primary structure is inadequate. If we adopt the above-indicated value from (2) as the characteristic association "collision" time, then to achieve agreement with the experimentally observed viscous-aftereffect times we need thousands of "collisions" for each association. There is no doubt that a tremendous number of intermediate states exists between the initial state of the structural element and its dynamic equilibrium state. It is most probable that the motion of the association in the space of states proceeds gradually. The polydispersion of the particles may also play an important role. Dynamic equilibrium is established only when the largest of the particles interact with each other an adequate number of times. If we have one such particle for every thousand small particles, the characteristic time of hydrodynamic approach for these particles is comparable to the observed time for the slow viscous aftereffect.

Let us examine the experimental results. We used two types of rotating viscosimeters: the "Rotovisco" bell type and the "cone-plate" type. The measurement unit of the viscosimeter was positioned within the high-frequency inductor of a radial (for the "Rotovisco") or of a uniform vertical (for the "cone-plate" type) magnetic field. By means of these viscosimeters it is possible to record changes in viscous stresses for durations of no less than 0.5 sec; the characteristic time for the growth in the front of the field strength amounted to 0.04 sec. The time change for the viscous stresses σ in the bell-type viscosimeter, starting at the instant t = 0 of field actuation, is shown in Fig. 7. The curves have been obtained for a shearing velocity of $\dot{\gamma} = 170\ \sec^{-1}$ for various values of H. With the "cone-plate" viscosimeter we obtained similar results. As we can see, following the sudden increase in viscosity where the field is actuated within ~1

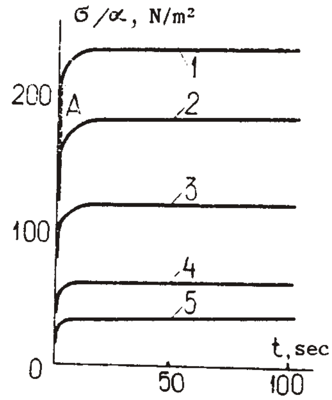

Fig. 7

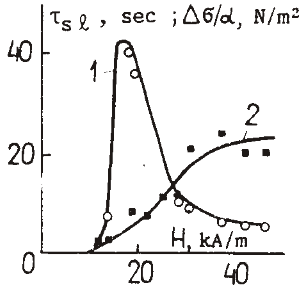

Fig. 8

Fig. 7. Increase in viscous stresses from the instant at which the field is actuated, in a bell-type viscosimeter with H = 45 (1), 39 (2), 34.4 (3), 24.8 (4), and 18.4 kA/m (5). $\dot{\gamma} = 170\ \text{sec}^{-1}$; $\alpha = 1.75$.

Fig. 8. Characteristic time τ_{sl} of the slow portion of the viscous aftereffect (1) and the slow portion of the viscous stresses $\Delta\sigma$ (with $\alpha = 1.75$) (2) as functions of H for $\dot{\gamma} = 170\ \text{sec}^{-1}$.

sec, a fact that is associated with the inertia of the instrumentation, there follows a rather prolonged period during which the viscous stresses change by as much as 10% relative to the initial value.

The slow portion of the viscous stresses increases virtually exponentially. To determine its magnitude and the characteristic time, we resorted to the following procedure. The problem involves the fact that it is possible, on the measured curve $\sigma(t)$ (see Fig. 7), to determine only very conditionally the coordinates of the point where the fast evolution of the viscous stresses ends and where the slow part begins. Having chosen some point A somewhat below entry into the slow segment of curve $\sigma(t)$, we make the assumption that the subsequent course of this relationship will be described by the formula

$$\sigma=\sigma_A+\sigma'\{1-\exp[-(t-t_A)/\tau']\}+\Delta\sigma\{1-\exp[-(t-t_A)/\tau_{sl}]\}. \tag{4}$$

It is the function of the second term in this formula to correct the choice for the slow component of the viscous stresses, characterized by the quantities $\Delta\sigma$ and τ_{sl}. Indeed, the values of the shorter of the relaxation times τ', obtained through approximation of the experimental curves with relationship (4), amounted to 1 sec, which coincides with the time constant of the instrument. Figure 8 shows the relationship between the derived values of the quantities $\Delta\sigma$, τ_{sl} and H. As we can see, the magnitude of the slow portion of the viscous stresses increases monotonically with the field, whereas the characteristic time of this process, depending on field intensity, exhibits a pronounced maximum at $H \approx 15$ kA/m. It turns out that in this field the ratio of the structuring time τ_{st} to the hydrodynamic time τ_η is close to unity. Indeed, assuming in (3) that $P = 7$, $\eta = 1$, $\dot{\gamma} = 170$, $H = 15$ kA/m, $\varphi = 0.1$, we have the estimate $k = 0.4$. It may therefore be concluded that the most prolonged process of viscous aftereffect occurs when the hydrodynamic time and the structuring time are comparable. This must be taken into consideration in the design of magnetorheological equipment.

LITERATURE CITED

1. Z. P. Shul'man and V. I. Kordonskii, *The Magnetorheological Effect* [in Russian], Minsk (1982).
2. I. S. Tolmasskii, *Carbonyl Ferromagnetics* [in Russian], Moscow (1976).
3. Z. P. Shul'man, V. I. Kordonskii, I. V. Prokhorov, and B. M. Khusid, "Magnetic relaxation in ferrosuspensions," *Magn. Gidrodin.*, No. 3 18—24 (1985).
4. R. Bozart, *Ferromagnetism* [Russian translation], Moscow (1962).
5. V. G. Levich, *Physicochemical Hydrodynamics* [in Russian], Moscow (1959).
6. B. É. Kashevskii and A. L. Novikova, "The paired interaction of particles in a magnetic suspension under the action of a rotating field," *Magn. Gidrodin.*, No. 3, 33—38 (1989).