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THEORETICAL PREREQUISITES FOR ONE MAGNETOOPTICAL METHOD OF MEASUREMENT OF THE VISCOSITY OF A FLUID

S. B. Kashevskii UDC 532.13

The viscosimetry method which is suitable for studying the kinetics of viscosity of a polymer solution in a drying film and is based on the measurement of the dynamics of light transmission of a two-layer system (excited by magnetic-field pulses and formed from sedimentation of noncolloidal ferromagnetic particles suspended in the fluid) has been substantiated. Optimum conditions for its implementation have been determined by computer modeling of the collective behavior of a system of particles.

Introduction. Investigation of the rheological properties of polymer solutions is of manifold theoretical and practical interest [1]. Indirect viscosimetric methods the basis for which is the measurement of the velocity (limited by the viscosity of a carrier medium) of optical or magnetic response of a system of ferroparticles distributed in the volume of a polymer matrix have been proposed along with traditional methods [2–4]. The need for information on the kinetics of matrix viscosity in the process of electromagnetic formation of conducting films from a polymer composition filled with nickel particles was directly responsible for the present investigation [5]. The method in question is based on the measurement of the time dependence of the light transmission of a layer filled with ferroparticles under pulsed action of a magnetic field which is transverse to the layer. Brightening along the field lines is attributable to the formation of particle chains. If the properties of the particle and the field strength are fixed, the value of the effect and the rate of its rise depend on the viscosity of the matrix and the concentration of the filler. Not only does the viscosity of the solution change in the process of drying but the concentration of the particles and their spatial distribution due to sedimentation change also. To eliminate concentration changes one should carry out magnetooptical measurements after sedimentation of the particles in a hermetically closed layer. In practical implementation of the method considered, some questions arise, namely: what are the form of the initial response of the system to a sudden application of the field and the sensitivity of the response to the degree of filling (coverage) of the bottom of the cell with particles and to the sedimentation level? To answer them and to substantiate the optimum parameters of the method we have performed computer modeling of the entire process, including the settling of particles in the free state and their aggregation and the brightening of the layer in sudden application of the field.

Computer Modeling. Let us consider a portion of the layer of thickness h (several particle diameters) the foundation of which is a square with side L_x (in particle diameters). The height of the layer must satisfy the condition that the particles do not reach the upper boundary of the layer on the modeled initial portion of the process of formation of the structure. The initial state of dispersion in the separated volume is characterized by a uniform particle distribution and is modeled by a random number generator. The number of particles thrown into the layer N is prescribed by the relation

$$N = \mathrm{round}\,(sL_x^2)\,, \tag{1}$$

where the parameter s characterizes the degree of filling of the foundation upon sedimentation in relation to a dense square packing. Modeling the behavior of the ensemble of particles, we disregard the effect of their hydrodynamic interaction and the inertia force.

A. V. Luikov Heat and Mass Transfer Institute, National Academy of Sciences of Belarus, Minsk, Belarus; email: bekas@itmo.by. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 76, No. 2, pp. 9–11, March–April, 2003. Original article submitted October 4, 2002.

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We take into account the sedimentation (equal for all the particles) $\mathbf{F}^{\mathrm{g}}$ and magnetic $\mathbf{F}_\alpha^{\mathrm{m}}$ forces (the subscript α denotes the particle number). To allow for the impermeability of the particles and the boundaries of the region we introduce the contact-interaction force $\mathbf{F}_\alpha^{\mathrm{c}}$. The motion of each particle is described by the equation

$$d\mathbf{R}_\alpha / dt = (1/6\pi\eta\alpha)\,(\mathbf{F}_\alpha^{\mathrm{m}} + \mathbf{F}^{\mathrm{g}} + \mathbf{F}_\alpha^{\mathrm{c}})\ .$$

The magnetic interaction between particles in the magnetic field $\mathbf{H}$ is described in the dipole approximation

$$\mathbf{F}_\alpha^{\mathrm{m}} = \frac{2}{3}(\pi M a)^2\,\mathbf{f}_\alpha^{\mathrm{m}}\ ,\quad \mathbf{f}_\alpha^{\mathrm{m}} = \sum_{\beta\neq\alpha}\mathbf{f}_{\alpha\beta}^{\mathrm{m}}\ ,\quad \mathbf{f}_{\alpha\beta}^{\mathrm{m}} = \frac{1}{2r_{\alpha\beta}^4}\left[\mathbf{n}_{\beta\alpha} + 2\mathbf{h}\,(\mathbf{n}_{\beta\alpha}\mathbf{h}) - 5\mathbf{n}_{\beta\alpha}\,(\mathbf{n}_{\beta\alpha}\mathbf{h})^2\right],$$

where $\mathbf{r}_{\alpha\beta} = \mathbf{R}_{\alpha\beta}/(2a)$, $\mathbf{R}_{\alpha\beta} = \mathbf{R}_\beta - \mathbf{R}_\alpha$, $\mathbf{n}_{\beta\alpha} = \mathbf{R}_{\beta\alpha}/R_{\beta\alpha}$, and $\mathbf{h} = \mathbf{H}/H$ (the subscript β is the particle number and H is the absolute value of the field). The sedimentation force has the form $\mathbf{F}^{\mathrm{g}} = (4/3)\pi a^3\Delta\rho g$. The accuracy in description of the contact interaction of particles is of no importance in our case and the corresponding force is prescribed by the relation

$$\mathbf{F}_{\beta\alpha}^{\mathrm{c}} = B\left[\left(\frac{\Delta}{R_{\alpha\beta} - 2a}\right)^2 - 1\right]\mathbf{n}_{\beta\alpha}\quad (R_{\alpha\beta} < 2a + \Delta)\ ,\quad \mathbf{F}_{\beta\alpha}^{\mathrm{c}} = 0\quad (R_{\alpha\beta} \geq 1 + \Delta)\ ,$$

where the parameters B and Δ have a formal character and are selected in numerical experiments. In actual practice, the value of B is several units of the force of the maximum dipole–dipole attraction of a pair of particles in contact while the value of Δ is several hundredths of the particle diameter. Analogously we prescribe the force keeping the particles from going out of the volume under study.

By employing the scales of the distance $2a$ and the time $t^* = 18\eta/\pi M^2$ and the notation $\tau = t/t^*$, $r = R/2a$, and $\delta = \Delta/2a$, we describe the dynamics of the ensemble by the system of dimensionless equations ($\alpha = 1, 2, ..., N$)

$$\frac{d\mathbf{r}_\alpha}{d\tau} = \mathbf{f}_\alpha^{\mathrm{m}} + \mathbf{f}_\alpha^{\mathrm{c}} - \mathbf{k}f^{\mathrm{g}}\ , \tag{2}$$

where $f^{\mathrm{g}} = 2a\Delta\rho g/(\pi M^2)$ is the ratio of the sedimentation force to the force of magnetic interaction of the particles in contact. Let us evaluate this quantity. Setting $a = 5\cdot10^{-4}$ cm, $\Delta\rho = 7$ g/cm^2, and $M = 50$ G, we find the value $f \approx 10^{-3}$.

The characteristic settling time of particles is determined as the time it takes the particle at the upper boundary of the layer to settle:

$$\tau^* = (f^{\mathrm{g}})^{-1}\,(h - 1)\ . \tag{3}$$

We describe light transmission in the approximation of geometrical optics, assuming that the polymer solution is optically transparent and a part of the luminous flux which meets not a single particle on its way is transmitted by the layer. The total intensity of the light incident on the layer considered is equal to I_0, while the total intensity of the light transmitted by the dispersion layer and arriving at the detector is equal to $I = k_{\mathrm{t}}I_0$. The electric signal produced by the detector is $U = U(I)$. The change in the signal because of the change in the intensity of the transmitted light under the action of the field (the intensity change is weak) is determined by the relation

$$U(t) = U(0) + \left.\frac{dU}{dI}\right|_{t=0}[I(t) - I(0)]\ .$$

The rate of rise of the signal is in proportion to the rate of rise of the light-transmission coefficient:

$$\frac{dU}{dt} = \alpha\frac{dI}{dt} = \alpha I_0\frac{dk_{\mathrm{t}}}{dt}\ . \tag{4}$$

TABLE 1. Influence of the Relative Settling Time of Particles τ_s/τ^* and the Degree of Filling of the Cell's Bottom with Particles on the Rate of Rise of the Light Transmission φ under the Action of the Magnetic Field

τ_s/τ^*	s				
	0.50	0.75	1.00	1.25	1.50
0.75	0.056	0.118	0.170	0.148	0.187
1.00	0.028	0.092	0.109	0.123	0.140
1.25	0.013	0.042	0.107	0.118	0.117
1.50	0.005	0.020	0.110	0.117	0.116

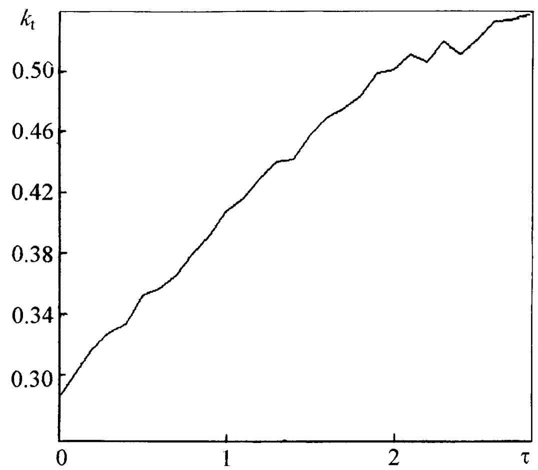

Fig. 1. Variation in the light-transmission coefficient with time for the value of the filling coefficient $s = 1$ and the settling time τ_s/τ^* (result of numerical modeling).

To determine the light-transmission coefficient in the numerical experiments we "throw," in a random manner, a rather large number of n perpendicular straight lines through the dispersion layer, compute the number of straight lines n' passing by all the particles in the layer, and set $k_t = n'/n$.

The numerical experiments have been carried out for the cell with dimensions $L_x = 30$ and $h = 10$. The order of the calculations is as follows. For the selected value of the parameter of filling s we scatter N particles (1) in the cell volume using the random number generator. Next, when the magnetic field is switched off, we switch on the process of settling of the particles during a certain period τ_s. Then, upon switching on the vertical magnetic field, we model the process of structurization of the particles, computing the light-transmission coefficient at successive time intervals and monitoring the height of ascent of the particles to the upper boundary of the layer.

Figure 1 gives as an example the curve of variation of the light-transmission coefficient with time from the instant of switching on of the field for the value of the filling coefficient $s = 1$ and the settling time $\tau_s = \tau^*$. As we see, the initial portion of the curve is linear in character:

$$k_t = k_0 + \varphi\tau \, . \tag{5}$$

The slope φ represents the rate of rise of the light transmission and is determined by the least-squares method. With account for (5) and $\tau = (\pi M^2/18\eta)/t$ and for the relation $M = (3/4\pi)H$, which holds in rather small fields, formula (4) for the electric signal of the photodetector takes the form

$$\frac{dU}{dt} = \frac{\gamma}{\eta} \, , \quad \gamma = \frac{H^2}{32\pi} \alpha\varphi I_0 \, . \tag{6}$$

It follows that for fixed intensity of the light source I_0, degree of filling of the layer s, and field strength H the slope of the initial portion of the dependence of the signal of the light detector on the time of switching on of the field is in inverse portion to the viscosity.

We performed numerical experiments with the aim of studying the influence of the degree of filling s and the settling time τ_s on the quantity recorded. The results are summarized in Table 1.

CONCLUSIONS

An analysis of the numerical data enables us to draw some important conclusions. In the case of a low content of particles ($s < 1$), the response of the system to the switched-on field (quantity φ) is not stabilized with an increase in the sedimentation time to $1.5\tau^*$. Moreover, the response is delayed in the numerical experiments. Conversely, in the case $s > 1$, the response is stabilized after a time of $1.25\tau^*$ and the delay is absent. These facts are attributed to the features of the final stage of formation of an equilibrium sedimentation layer, which is associated with the resorption of accidental clusters of particles at the cell's bottom. In the case $s < 1$, the clusters disappear totally and sedimentation equilibrium corresponds to a single layer of particles. The state of the latter possesses the property of unstable equilibrium in the perpendicular magnetic field, which explains the occurrence of a delay. When $s > 1$, a part of the particles is left, upon sedimentation, in the second layer; this state in the field is unstable and ensures an instantaneous response of the system to the switching on the field. Thus, one should select the values $s = 1.25$ and $\tau_s = 1.25\tau^*$ in the experiment. We particularly emphasize that the maximum displacement of the particles to the upper boundary over the period of action of the field $\tau = 1$ does not exceed 1 diameter.

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NOTATION

a, particle radius; g, free-fall acceleration; $\mathbf{k}$, unit vector in opposition to the force of gravity; k, coefficient; M, particle magnetization; $\mathbf{R}$, radius vector; R, its absolute value; t, time; α, instrument constant; η, viscosity; ρ, density; $\Delta\rho$, difference of the densities of the particle and the fluid. Subscripts: c, contact; g, gravitational; m, magnetic; s, sedimentation; t, transmission; 0, initial state.

REFERENCES

1. A. Ya. Malkin and A. E. Chalykh, *Diffusion and Viscosity of Polymers. Measurement Techniques* [in Russian], Moscow (1979).
2. J.-C. Bacri, J. Dumas, D. Gorse, R. Perzynski, and D. Salin, *J. Physique Lett.*, **46**, 1199–1205 (1985).
3. Yu. L. Raikher and V. V. Rusakov, *Kolloid. Zh.*, **63**, No. 5, 665–673 (2001).
4. S. R. Gorodkin, É. A. Zal'tsgendler, V. I. Kordonskii, I. V. Prokhorov, B. M. Khusid, and Z. P. Shul'man, *Method for Measuring Fluid Viscosity*, USSR Inventor's Certificate No. 1394107, Byull. No. 17 (1988).
5. B. É. Kashevskii, I. V. Prokhorov, V. A. Kuz'min, and A. A. Rusalovich, *Dokl. Nats. Akad. Nauk Belarusi*, **46**, No. 2, 119–121 (2002).