Investigation of the rheological properties of liquid crystals by means of acoustic flows

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Acoustic flows that arise in a non-Newtonian liquid during propagation of a damped plane wave are studied. An expression is obtained which connects the location of the maximum velocity of the acoustic flow on the axis of the radiator with the Maxwell relaxation time. The relaxation time in MBBA liquid crystals is determined from the experimentally observed configuration of the acoustic flows.

Matter in the liquid crystal state occupies an intermediate position between a solid crystal and a liquid, and exhibits both elasticity and viscosity under the action of mechanical stresses. As Ostwald has demonstrated,[1] at $\omega\tau \sim 1$ the flow of liquid crystal material does not obey Newton's law, i.e., the ordinary equations of hydrodynamics are not applicable in these media without account of corrections for relaxation effects.

One of the best known methods for determining the relaxation time is based on measurement of the velocity dispersion of acoustic waves or the change in the absorption coefficient over a range of ultrasonic frequencies.[2] Such a type of study has been carried out in nematic crystals and described in a number of papers.[3-6] In the present work, a new method is presented for the estimate of the relaxation time in non-Newtonian liquids, in particular, liquid crystals, from the parameters of acoustic flows; the equations of motion of a rheological liquid are taken as the initial equations of motion. These are valid in the range of frequencies which satisfy the condition $\omega\tau \leq 1$.

The equations which describe acoustic flow in non-Newtonian liquids are of the form[7]

$$\frac{\partial \mathbf{U}}{\partial t}+(\mathbf{U}\nabla)\mathbf{U}=-\frac{\nabla P}{\rho_0}+\alpha_0\frac{q^2\mathbf{n}E_0}{\rho_0}+\frac{\eta(0)}{\rho_0}\nabla^2\mathbf{U}+\frac{^1/_3\eta(0)+\zeta(0)}{\rho_0}\nabla(\nabla\mathbf{U}),\quad \nabla[U+V_0\rho_a/\rho_0]=0, \tag{1}$$

where U is the velocity of acoustic flow, E_0 the energy density in the sound wave, n a unit vector in the direction of the wave, t the time, ω the frequency, $\eta(0)$ and $\zeta(0)$ the shear and bulk viscosities in the Newtonian liquid, V_0 the oscillation velocity, P the pressure, ρ_0 the density of the medium at rest, c the velocity of sound in the non-Newtonian liquid, and α_0 the sound absorption coefficient.

The following conditions are satisfied in the experiment: 1) $V_0/c_0 < 1$; 2) $U/c_0 < 1$; 3) $\mathrm{Re} = Ua/\gamma(0) < 1$; 4) the time of observation exceeds the time of establishment of the acoustic flow $t_{est} = a^2/\gamma(0)$.[8] (Here and below, $\gamma(0)$ is the kinematic viscosity of the medium and a is the radius of the radiator, which has the form of a disc.) Under such conditions, the set of equations (1) which describes the acoustic flow in the rheological liquid takes the form

$$-\nabla P+\alpha_0 E_0=\eta(0)\nabla^2\mathbf{U},\quad \nabla U=0; \tag{2}$$

$$\alpha_0=\frac{\omega^2\tau(c_\infty^2-c_0^2)(1+\omega^2\tau^2)^{1/2}}{2c_0^3(1+\omega^2\tau^2c_\infty^2/c_0^2)^{3/2}}, \tag{3}$$

where c_0 and c_∞ are the sound velocities as $\omega \to 0$ and $\omega \to \infty$, respectively.

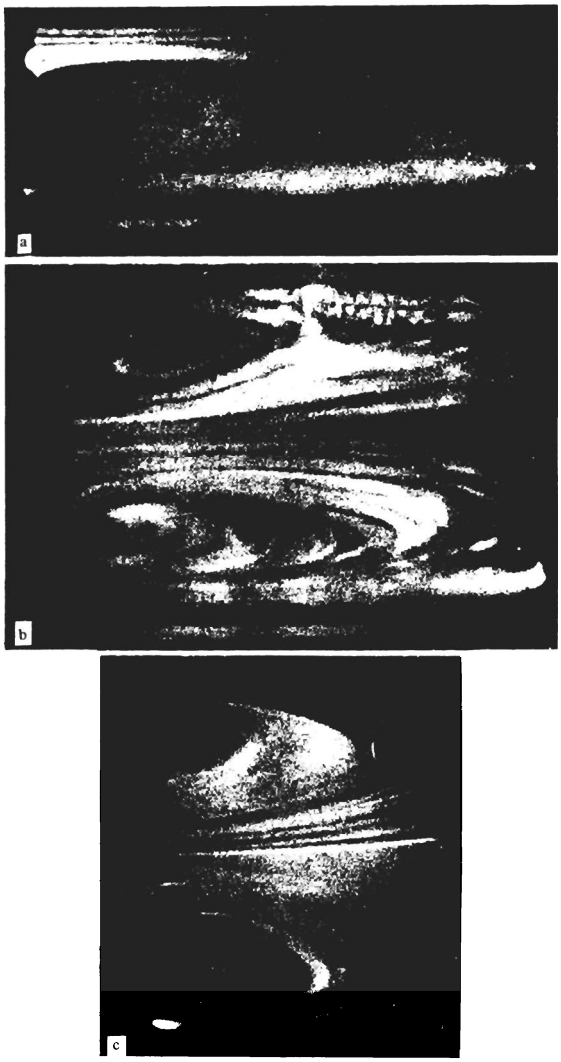
As shown in [9], the velocity distribution of the acoustic flow in a Newtonian liquid, due to a traveling wave and under the condition $\alpha_0 l > 1$, has a maximum located at a distance

$$x_m = 1/2\alpha_0 \tag{4}$$

from the radiator. We note that a non-reflecting boundary is located in the plane $x = l$. Since the equations of flow in a non-Newtonian liquid are identical in form with the equations of flow in an ordinary Newtonian liquid, we can state that a similar distribution of the flow velocities exists in the rheological liquid, but in this case the absorption coefficient, on the value of which the location of the velocity maximum depends, is determined by the expression (3). Thus, by knowing the configuration of the acoustic flow, we can determine the value of the sound absorption coefficient in the rheological liquid from the position of the velocity maximum on the axis of the acoustic beam (the x_m coordinate); then, with the help of the expression (3), we can calculate the relaxation time.

The proposed method was used for the determination of the relaxation time in methoxybenzylidenebutylaniline (MBBA) in the temperature range of the existence of the mesomorphic state (21–41°C). A cell in the shape of a parallelepiped (6.6×1.8×3 cm) was divided into two sections by a thin, sound-transparent film. The radiator (a disc of diameter 0.6 cm) was located at one end of the cell adjoining the compartment which was filled with the liquid crystal. The second compartment of the cell contained a solution of glycerin in water. The temperature of the sample was controlled with a water jacket surrounding the cell. Observation and photography of the field of flow were carried out from above. The field of view was determined by a beam of light through a slit, and the light beam was directed perpendicular to the acoustic beam and intersected it at the focus of the photographic system.

Observation of the acoustic flow in a liquid crystal was achieved by making use of the fact that the part of the sample which directly abuts the surface of the radiator was heated upon operation of the latter, and the material in the thin, near-surface layer underwent transition to the isotropic state. Outside this layer, the temperature of the sample in the cell was maintained constant at 31°C during the photography. Since the MBBA crystal is transparent in the isotropic phase, and scatters the light in the nematic phase, the separation boundary could easily be distinguished. The material in the isotropic state, dragged along by the acoustic flow in the nematic phase, moves in the nematic phase in the form of a thin transparent jet and "draws" a stream line in the latter.



Picture of the established acoustic flow in an MBBA crystal at a temperature of 31°C and at the frequencies: a) 2.1 MHz; b) 2.1 Mhz (magnified 4.5 times): c) 6.3 MHz (magnified 4.8 times). The source is located to the left, the photography is from above. The stream lines are produced by movement of the isotropic phase.

Figure (a) shows a photograph of the establishment of acoustic flow in an MBBA crystal at a frequency of 2.1 MHz (top view). The geometry of the experiment is such that the radiator lies to the left. The light lines on the dark background, which are identified with the stream lines, are produced by motion of the transparent isotropic phase. The drawing also shows the photograph of the established flow in the nematic phase of MBBA at frequencies of 2.1 (b) and 6.3 (c) MHz. The range of values of x_m within which the maximum of the flow velocity on the axis of the sound beam is localized is estimated from the density of the lines. The relaxation times in the nematic phase of MBBA were then calculated from these data with the help of Eqs. (3) and (4) (see the table).

Some divergence of the values of τ at two frequencies is evidently due to the degree of satisfaction of the condition $\alpha_0 l > 1$ (l is the length of the cell), which reflects the influence of the wall on the configuration of the flow, the validity of which is assumed in the theoretical representations of the distribution of the velocities of acoustic flows in Newtonian liquids used by us.

ω, MHz	T, °C	x_m, cm	τ, sec
2.1	31	0.98—1.08	$(8.7-9.5)\cdot 10^{-9}$
6.3	31	0.4—0.69	$(1.5-3)\cdot 10^{-9}$

The values of τ in the nematic phase of MBBA, given in the table, correlate in order of magnitude with values obtained by other methods.[3-6] Thus, from the data of Eden et al.[6] in the temperature range 23–77°C, the quantity τ varied within the range 2×10^{-8}–5×10^{-9} sec. At a temperature of 22°C, the time of structural relaxation in MBBA amounts to 2×10^{-8} sec.[4]

We can thus state that the proposed method of determination of the relaxation time in rheological media from the configuration of acoustic flows allows us to estimate quickly and accurately the order of the quantities studied. The merit of the method lies in the simplicity of the arrangement of the acoustic experiment, which does not require complex and cumbersome apparatus. The method also possesses the advantage that it does not require measurements of the absolute values of the flow velocity: to estimate the order of magnitude of the relaxation times, it suffices to establish only the relative location of the stream lines of the acoustic flow. In those cases in which this is necessary, the accuracy of the method can be appreciably increased by measuring the distribution of the absolute values of the flow velocity on the axis of the sound beam. If the medium studied permits the introduction of foreign impurities, the observation of the acoustic flows can be accomplished by ordinary methods.

It should be noted that the agreement of the data obtained by the proposed method with the results of other experiments gives grounds for assuming that the theoretical model of the medium used by us, based on a linear rheological equation,[7] is valid in the considered frequency range.

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65