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D. V. Shmeliova[1], E. P. Pozhidaev[1,2], S. S. Kharlamov[1], S. V. Pasechnik[1], V. A. Barbashov[2], V. G. Chigrinov[3]

CAPILLARY FLOW AND SHEAR VISCOSITY OF FERROELECTRIC LIQUID CRYSTAL

[1] MIREA – Russian Technological University, Problem Laboratory of Molecular Acoustics,
78 Vernadsky Av., Moscow, 119454, Russia. E-mail: s-p-a-s-m@mail.ru
[2]P. N. Lebedev Physical Institute, 53 Leninsky Pr., Moscow, 119991, Russia.
E-mail: epozhidaev@mail.ru
[3]Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong.
E-mail: eechigr@ust.hk

Experimentally shown that the shear flow of a ferroelectric smectic C liquid crystal (FLC) in flat capillary with homeotropic boundary conditions can be described in the framework of the Newtonian fluids theory if the pitch p_0 of the FLC helix is much less than the capillary gap. The motion of the contact line during the filling of the capillary was recorded in the experiment and compared with the theory, which made it possible to estimate the shear viscosity coefficient $\eta \approx 0.5$ Pa·s, which turned out to be much larger than the rotational viscosity coefficient γ_φ. In the capillary filling process, the principal optical axis of the FLC helical structure was oriented mainly perpendicular to the substrates, and the smectic layers were parallel to the substrates. At the same time, narrow dislocation bands arose along the flow direction. The principal optical axis deviated from the normal to the substrates at different angles within the dislocation bands. The total area occupied by dislocations does not exceed 5 % of the flow area; therefore, dislocations do not have a significant effect on the rheological behavior of the smectic C* phase.*

Key words: *ferroelectric liquid crystal, capillary shear flow, shear viscosity.*

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Д. В. Шмелева[1], Е. П. Пожидаев[1,2], С. С. Харламов[1], С. В. Пасечник[1], В. А. Барбашов[2], В. Г. Чигринов[3]

КАПИЛЛЯРНОЕ ТЕЧЕНИЕ И СДВИГОВАЯ ВЯЗКОСТЬ СЕГНЕТОЭЛЕКТРИЧЕСКОГО ЖИДКОГО КРИСТАЛЛА

[1]МИРЭА – Российский технологический университет, Проблемная лаборатория молекулярной акустики,
пр. Вернадского, д. 78, 119454 Москва, Россия. E-mail: s-p-a-s-m@mail.ru
[2]Физический институт им. П. Н. Лебедева РАН,
Ленинский пр., д. 53, 119991 Москва, Россия. E-mail: epozhidaev@mail.ru
[3]Гонконгский университет науки и технологии,
Залив Чистой Воды, Коулун, Гонконг. E-mail: eechigr@ust.hk

Экспериментально показано, что сдвиговый поток сегнетоэлектрического смектического C жидкого кристалла (СЖК) в плоском капилляре с гомеотропными граничными условиями описывается в рамках теории капиллярного потока ньютоновских жидкостей, если шаг p_0 спирали СЖК много меньше капиллярного зазора. Движение контактной линии при заполнении капилляра регистрировалось в эксперименте и сравнивалось с теорией, что позволило оценить величину коэффициента сдвиговой вязкости $\eta \approx 0,5$ Pa·s, который оказался много больше коэффициента вращательной вязкости γ_φ.*

В процессе заполнения капилляра главная оптическая ось спиральной структуры СЖК ориентировалась в основном перпендикулярно подложкам, а смектические слои – параллельно подложкам. Вместе с тем в направлении потока формировались узкие полосы дислокаций, в пределах которых главная оптическая ось отклонялась от нормали к подложкам на различные углы. Суммарная площадь, занимаемая дислокациями, не превышает 5 % от площади потока, поэтому дислокации не оказывают существенного влияния на реологическое поведение смектической С фазы.*

Ключевые слова: *сегнетоэлектрический жидкий кристалл, капиллярное сдвиговое течение, сдвиговая вязкость.*

Introduction

Among different types of liquid crystal (LC) phases, nematic liquid crystals (NLC) have found outstanding application in modern display industry [1] due to unique combination of strong electrical and optical anisotropy resulting from long ranged orientational order and high fluidity of NLC, connected with fluid-like translational motion of molecules [2]. In the hydrodynamic theory of NLC Leslie-Ericksen the unit vector (director $\boldsymbol{n}$), correspondent to the overall orientation of long molecular axes, is introduced as a new hydrodynamic parameter additional to the velocity $\boldsymbol{v}$ and pressure p used in traditional hydrodynamics of isotropic liquids. As a result, the equations of nematodynamics include a number of specific material parameters (the six Leslie coefficients α_i) describing the energy dissipation due to translational and orientational molecular motions and the connection between the velocity $\boldsymbol{v}(\boldsymbol{r},t)$ and director $\boldsymbol{n}(\boldsymbol{r},t)$ fields [3]. Such connection is responsible for a number of specific linear and nonlinear phenomena in LC flows [2]. In particular, it results in non-Newtonian character of shear flows of NLC [2, 4] which leads, in general, to dependences of the apparent shear viscosity coefficient on the shear rate and capillaries' dimensions [5]. Such behavior became simpler at stabilization of the orientation structure of NLC due to the action of strong magnetic (electric) fields [6] or internal surfaces of capillaries [7]. In this case, nematics flow as conventional Newtonian liquids [3] with the shear viscosity dependent on the orientation of a director relatively to the direction of the flow velocity and velocity gradient. The difference between maximal and minimal values of the shear viscosity coefficients, referred as two Miesovich viscosities η_1 and η_2, in most NLC is roughly equal to the rotational viscosity coefficient γ_1, which describes the energy dissipation due to a rotation of a director [2]. The information on the rotational and anisotropic shear viscosities is widely used at calculations of operating times of liquid crystal displays (LCD) and other LC devices for photonics [2, 8]. It is of importance that the relatively long (more than 1 ms) operation times of NLC based devices essentially restrict the further application of liquid crystals.

One of the most attractive ways to get fast LC devices is through the use of ferroelectric liquid crystals (FLC) showing microsecond operating times at low electric voltages [9, 10, 11]. Additionally to the orientational order, such materials are characterized by one-dimensional positional order, describing the distribution of molecular centers of mass in equidistant smectic planes and spontaneous polarization of LC media. In general, twenty viscous-like parameters, similar to Leslie's coefficients are introduced to describe the energy losses due to orientational and translational molecular motion and connection between them [13]. In the simplified version the azimuth rotation of a director in FLC under electric fields, which does not involve the layer structure's distortions, is traditionally described using only one coefficient of a rotational viscosity γ_φ with the same order of magnitude as the similar parameter in a nematic phase [9]. Nevertheless, the mentioned above connection between orientational and translational types of motion may play the essential role in practical applications of FLCs. In particular, this connection is responsible for irreversible distortions of layered structure under mechanical stresses, known as shock problem [10]. Solution of the shock problem [11, 12] plays the key role in elaboration of fast FLC displays, since shock-free FLCs can flow inside display cells under mechanical stress similar to NLCs, without any damage of the alignment quality.

In this paper we describe the first experimental investigation of the shock-free FLC under specific capillary flow, which provides the estimation of the apparent shear viscosity coefficient.

Experimental

In our experiments, we used sandwich like LC cells of a constant gap h with two open opposite edges, which is needed to realize a laminar plane flow with a velocity profile in XZ plane, as it is shown in Fig. 1. A typical LC cell consisted of the two glass plates (of thickness 1.1 mm) coated with ITO transparent electrodes. The inner surfaces of the plates were thoroughly cleaned to establish strongly water-wet glass surfaces and spin coated with 5 % chromolane's (chrome stearoyl chloride) solution in isopropanol. Afterwards the plates were baked to provide a thin nanometer layer of a chromolan, needed to get a homeotropic surface orientation. At the last stage, the plates were stacked with the help of striped teflon spacers and fixed with glue to form the flat channels of the gap h (23 ± 1 μm) width A (1 cm) and length L_{total} (1.8 cm). The prepared LC cell was placed between crossed polarizers, oriented at the angle 45° relatively to the flow direction. The schema of experimental set-up and experimental geometry for shear flow in FLC are shown in Fig. 1.

The capillary flow of FLC through the plane channel, which arose after placing LC on the one open edge of LC cell, was registered by a digital camera. All measurements were performed at room temperature $T = 25 \pm 0{,}5$ °C.

In our experiments, we used the shock-free FLC-595 developed at mixing of nematic host and chiral non-mesogenic guest according to the approach proposed in [12]. Temperature dependencies of the helix pitch (p_0), polar angle (θ_0), rotational viscosity (γ_φ) and normalized rotational viscosity (γ^*) of this FLC are presented in Fig. 2.

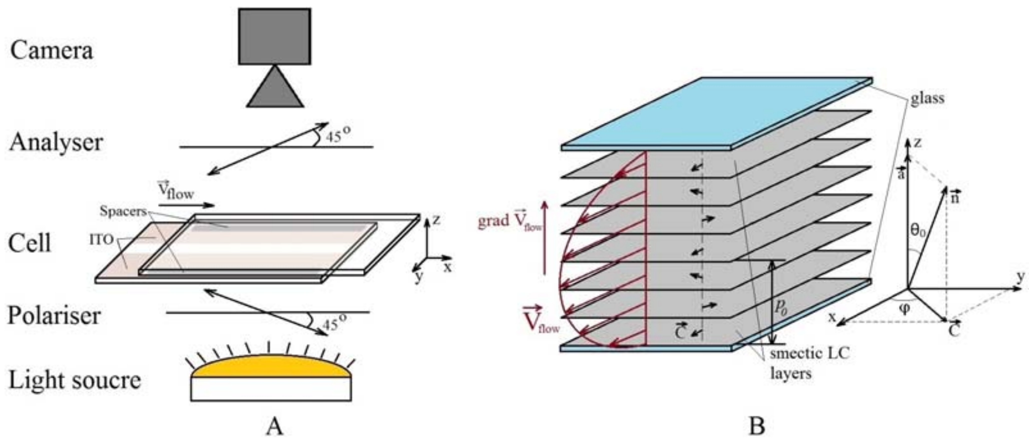

Fig. 1. A – Experimental set-up, *B* – Experimental geometry for shear flow in FLC, where p_0 – helix pitch, θ_0 – polar angle, φ – azimuth angle

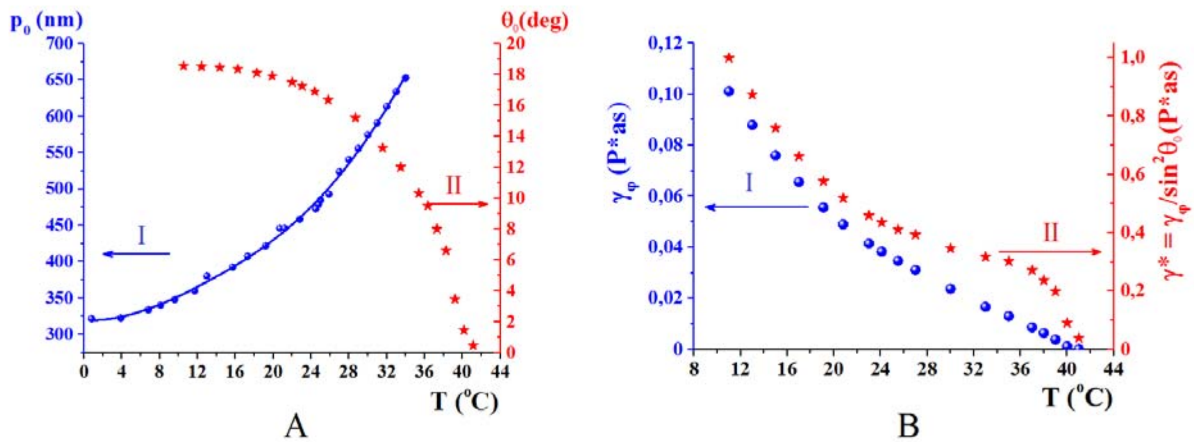

Fig. 2. A – Temperature dependences of the helix pitch (I) and polar angle (II) for FLC-595, *B* – Temperature dependences of rotational viscosity γ_φ (I) and normalized viscosity γ^* for FLC-595

Results and discussion

The instant images of the cell are shown in Fig. 3. The capillary forces, acting on FLC produced a motion of a contact line LC along the capillary which becomes slower due to viscosity forces.

Processing of the instant digital images made it possible to get information about displacement $L(t)$ of a contact line as a function of time. This function is shown in Fig. 4.

To explain the obtained results one can mention numerous investigations of capillary flows of isotropic liquids through capillaries of different cross section (see, for example [13]).

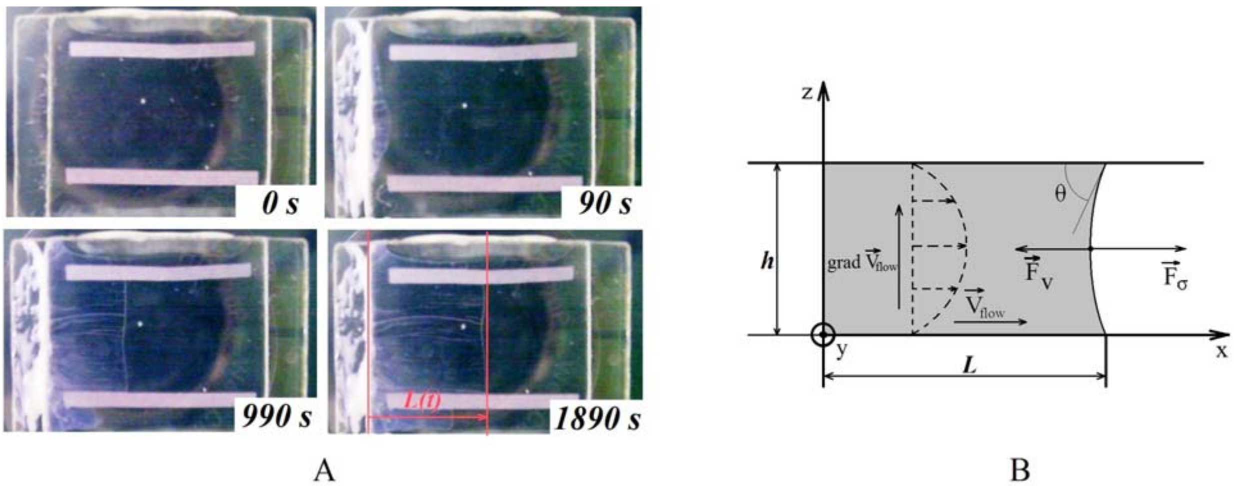

Fig. 3. A – The images of LC cell in crossed polarizers at different times (t).
B – Scheme illustrating the capillary flow of FLC under the action of capillary and viscous forces

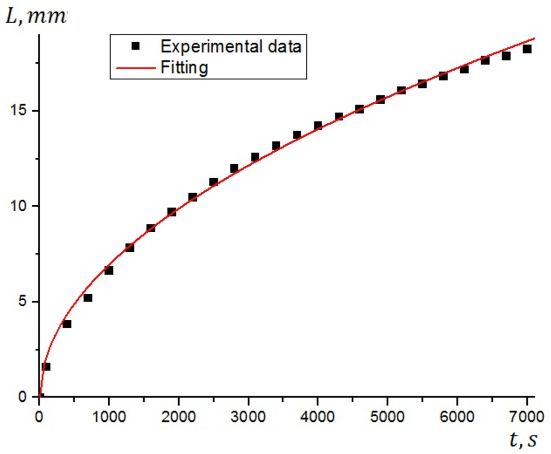

Fig. 4. The time dependence of a contact line displacement $L(t)$

In general, this phenomenon involves absorption of molecules of liquids by the solid walls of capillary which results in arising of a menisque of a definite curvature (characterized by a contact angle), moving along the capillary. Such processes are often described in terms of motion of a contact line. In general case a contact angle also depends on time, which makes the problem to be rather complicated even for the case of isotropic Newtonian liquids [13]. Nevertheless, the reasonable approximations, mentioned below, can be used to obtain rather simple analytic expressions, applicable for a description of our experimental results.

Firstly, we will consider FLC as a conventional Newtonian liquid with a constant value of the shear viscosity coefficient η, which depends on LC configuration. Obviously, such approximation is valid in the case of negligible influence of a flow on the initial structure of FLC, stabilized by surfaces. Indeed, analysis of microscopic images, presented in Fig. 5 show, that the capillary flow induced local changes in the initial orientation of FLC, which was manifested by a number of long living thin stripes of different internal structure, oriented presumably along the flow direction.

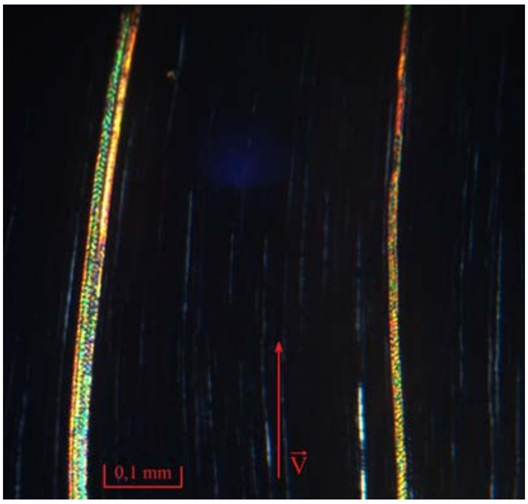

Fig. 5. Microscopic images of the cell with flow induced defects in crossed polarizers oriented at 45° relatively to the flow direction

Nevertheless, the space, occupied by such stripes is relatively small and one can assume that they play only the minor influence on the total capillary flow.

Secondly, we consider the contact angle as constant value, which is time-independent. It holds at small values of a capillary number [13]:

$$Ca = \frac{\eta v}{\sigma} << 1, \tag{1}$$

where $v = dl/dt$ – the velocity of a contact line, σ – a surface tension coefficient.

Using typical value σ = 0.03 $N.m^{-1}$ for liquid crystals [14] and registered values of $v \leq 10^{-4} m.s^{-1}$ at the initial stage of capillary flow, we conclude, that the inequality (1) is valid for η < 10 Pa s.

At these approximations, it is possible to describe a slow motion of a contact line along a horizontal capillary by taking into account only the capillary force F_σ and the viscos force F_v, acting on the sample of LC (Fig. 3, *B*). The correspondent equation of a balance for pressures can be written as:

$$P_\sigma = P_v, \tag{2}$$

where

$$P_\sigma = \frac{2\sigma\cos\theta}{h} \tag{3}$$

a capillary pressure,

$$P_v = (12\eta L/h^2)(dL/dt) \tag{4}$$

the pressure drop induced by viscosity losses at laminar flow of a liquid through the plane capillary, η – the shear viscosity of a liquid. Expression (4) is obtained by using the well-known formula for the flow rate $Q = dV/dt$ of a steady Poiseuille flow through a capillary of a gap h, width *A*, and length *L*:

$$Q = [(h^3 A)/(12\eta)](P_v/L) \tag{5}$$

By inserting (3) and (4) into expression (2) it is simple to get the next differential equation for $L(t)$:

$$L(dL/dt) = h\sigma\cos\theta/(6\eta) \tag{6}$$

The solution of the equation (5) results in the next simple expression for the time dependence of *L*

$$L^2(t) = [(h\sigma\cos\theta)/(3\eta)]t \tag{7}$$

It is worthwhile to notice that this simple expression is also approximately valid for the first stage of a rise of liquids in vertically oriented capillaries, when the gravity force is essentially smaller than the capillary and viscous forces [10].

The result of comparison of experimental data with the dependence (7), shown in Fig. 6 confirms the approximations, used above. It means, that the combination of material parameters $m = [(\sigma\ h\ \cos\theta)/(3\eta)]$ does not depend on time and can be determined from the slope of the strait line in Fig. 6. In our case $m = 6 \cdot 10^{-8}\ m^2c^{-1}$.

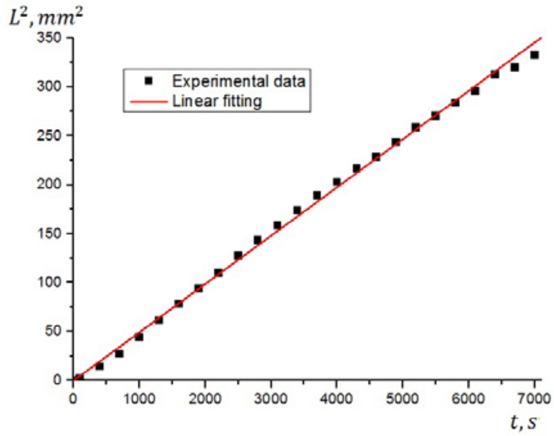

Fig. 6. Comparison between experimental data and theoretical dependence (7) shown by a solid line

To get numerical estimates for the shear viscosity coefficient, we fulfilled the additional experiment on a capillary rise of FLC in the vertically aligned cylindrical capillary of a radius *R* = 125 μm. Preliminary, the capillary was filled with 0.5 % solution of a chromolane in isopropanol. Afterwards the solvent was deleted due to the air blowing and evaporation. It resulted in homeotropic boundary conditions at the inner surface of a capillary similar to those described above for the plane capillary. The usage of the well-known expression for the height H of a capillary rise:

$$H = (2\sigma\cos\theta)/(\rho g R) \tag{8}$$

(ρ – LC density), made it possible to calculate the combination (σ cosθ) which was equal to $3.0 \cdot 10^{-3}$N.m^{-1} at experimentally measured value H = 4 mm. Inserting this value into equation (7) leads to the value of effective shear viscosity η = 0.47 Pa s, which has the same order of magnitude as the maximal shear viscosity of 5CB (η_1 = 0.105 Pa s [3]). At the same time, the effective shear viscosity exceeds the rotational viscosity $\gamma_\varphi \approx 0.02$ Pa s, correspondent to a motion of C – director, by more than an order, which is quite different in comparison with the mentioned above relation between two types of viscosities in nematics. It can be explained by the well-known dependence of γ_φ on the polar angle θ_0 [15]:

$$\gamma_\varphi = \gamma^* \sin^2\theta_0, \tag{9}$$

where γ^* – the parameter, describing losses at a motion of a director ***n*** along the cone surface, which does not critically depend on temperature and it is suitable for comparison with the rotational viscosity in a nematic phase [15].

In our case θ_0 = 17° (Fig. 2, *A*) and the shear viscosity η only slightly exceeds the normalized rotational viscosity γ^* = 0.41 Pa s at the same temperature (Fig. 2, *B*). It is similar to the case of nematics and can be explained by small value of the polar angle θ_0 for different values of the azimuth angle φ. It means, that the flow geometry, realized in our experiments is close to the principal geometry for the nematic's flow with the maximal value of the shear viscosity. The letter parameter also slightly exceeds the rotational viscosity of NLC, as it was mentioned above.

Finally, we have to point out, that the flow geometry, shown in Fig. 1, *B* and realized in our experiments resembles the simplest geometry for achiral smectic C-phase with the value φ independent on z – coordinate. In the latter case the shear viscosity is defined by the next expression a [13]:

$$\eta = \eta_1 \sin^2\varphi + \eta_2 \cos^2\varphi, \tag{10}$$

where η_1 and η_2 – are two (from seven) principal viscosities of smectic C-phase.

In the case of FLC, we can only approximately apply expression (10) to get:

$$\begin{aligned}\eta &= \eta_1 < \sin^2\varphi > + \eta_2 < \cos^2\varphi > \\ &= (\eta_1 + \eta_2)/2\end{aligned} \tag{11}$$

So, our measurements provide the estimate of the above mentioned combination of the principal shear viscosities, entering into expression for a viscous part of the stress tensor of FLC.

Conclusion

We have studied for the first time the capillary flow of the ferroelectric liquid crystal in LC cell with homeotropic type of surface orientation. It was established, that a motion of the contact line, registered in the experiment, was described by a simple power low, which is valid for a conventionally Newtonian liquid with the shear viscosity independent on the shear rate. It made possible to estimate, for the first time, the value of the shear viscosity of FLC. Contrary to the case of nematics, the letter parameter essentially exceeds the rotational viscosity of FLC, traditionally used for description of C-director motion under action of electric field. At the same time, the normalized value the rotational viscosity is close to the estimated value of the shear viscosity, which is typical for the most nematics. The presented results confirmed the possibility to use the capillary flow as an effective tool for a study of anisotropic viscous properties of liquid crystals. The further investigations, which out of scope of this paper, are needed to establish the connection between the helix pitch and viscosities of FLC and to clarify the nature of the flow induced long living defects.

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