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LETTER TO THE EDITOR

The shear viscosity minimum of freely flowing nematic liquid crystals

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Abstract

The analysis of temperature dependence of the shear viscosity of freely flowing nematic liquid crystals shows that the anisotropic fluid adopts such a manner of flow, which corresponds to the minimum of its viscosity at given conditions. We postulate that the principle is of general significance, i.e. it concerns any fluid in the flow.

The momentum transport in nematic liquid crystals shows an anisotropy as it depends on the mutual orientations of the macroscopic molecular ordering (the director n), the flow velocity (v) and the velocity gradient ($\text{grad}v$). In 1946 Mięśowicz defined three principal shear viscosity coefficients of nematics [1], which can be measured in three different experiments, sketched in figure 1. In a typical experiment a magnetic field is used for molecular alignment in the nematic sample. Intuition suggests that for mesogenic elongated molecules, the lowest resistance to the nematic flow, i.e. the lowest viscosity value, should be found in the experiment presented in figure 1(b). Indeed, when the orienting magnetic field, i.e. the director n , is parallel to the velocity of the nematic flow, the lowest viscosity (η_2) is recorded. Among the two remaining viscosities, η_1 has the highest value while η_3 is close to the viscosity measured in the isotropic phase of the compound. If one excludes the temperature range in the proximity of the clearing point, the temperature dependences of the shear viscosities $\eta_1(T)$, $\eta_2(T)$ and $\eta_3(T)$ are roughly parallel to each other [2–4].

This relatively simple picture of the viscosity of nematic liquid crystals is disturbed for the compounds exhibiting the transition to the smectic A phase [4–6]. Then, with decreasing temperature, the viscosity η_2 shows a strong increase and goes to infinity at the nematic to smectic A transition. The η_1 and η_3 viscosities are almost unaffected by the transition. At a temperature a few degrees before the transition to the smectic A phase, the viscosities η_2 and η_3 interchange their roles and then the lowest nematic viscosity corresponds to the flow depicted in figure 1(c). This experimental fact is crucial for the present work.

The presmectic behaviour of η_2 viscosity is due to the formation of precursors of smectic planes in which the molecular mobility, such as in figure 1(b), is restricted. Recently, the main features of the presmectic behaviour of the nematic viscosity coefficients were successfully

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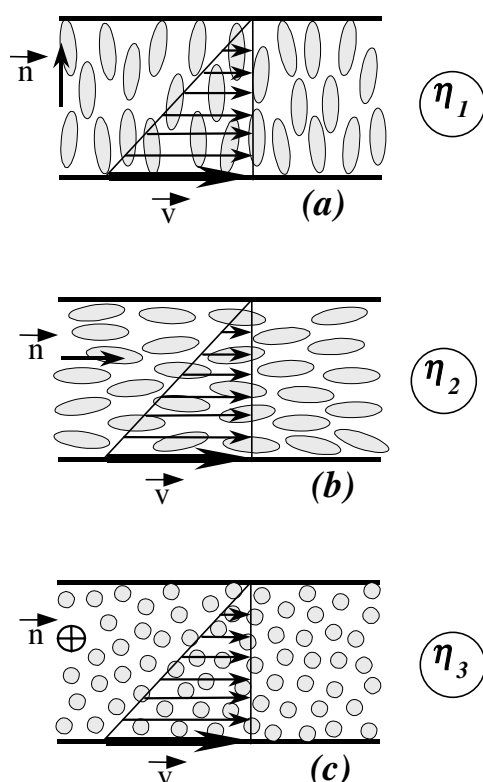


Figure 1. The experimental conditions for measurements of the three Mięslowicz shear viscosity coefficients of nematic liquid crystals: (a) $n \perp v$, $n \parallel \text{grad}v$; (b) $n \parallel v$, $n \perp \text{grad}v$; (c) $n \perp v$, $n \perp \text{grad}v$.

simulated by Bennette and Hess with the use of nonequilibrium molecular dynamics methods [7].

The above introductory remarks concerned the viscosity of the nematic liquid crystals oriented with external forces. Now, let us consider the behaviour of the nematics in a *flow without external ordering forces* i.e. the nematic in a free flow. Such a flow shows some peculiarities, which are worthy of notice.

The simplest mesogenic compound exhibits isotropic and nematic phases. Then, for a freely flowing compound the transition from the isotropic to nematic phase manifests itself in a strong decrease of the shear viscosity and, as numerous experiments show, if one neglects the temperature region just after the transition, this viscosity is very close to η_2 Mięslowicz coefficient in the whole temperature range of the nematic phase. In the literature this phenomenon is known as a nematic flow alignment [2, 8–12] suggesting that the nematic elongated molecules in a free flow should align as in figure 1(b) because it is ‘natural and self-evident’. Indeed, it always occurs for the above mentioned compounds exhibiting the nematic phase only. However, as we will see later, due to the presmectic effects, a free flow of nematic liquid crystals can also lead to quite different molecular alignment than that in figure 1(b).

In the 1970s the investigation of the viscous properties of nematic liquid crystals by measurement of the viscosity of a freely flowing substance were quite popular because of the above mentioned convergence of the results with the η_2 viscosity and because of the simplicity

of the experiment. However, later on the experiments showed that the viscosity of certain freely flowing nematic liquid crystals behave quite differently from η_2 and, for example, the Arrhenius dependence $\ln \eta \sim 1/T$ was strongly nonlinear or the evaluated viscosity activation energy was excessively high. Finally, the studies of the viscosity of the freely flowing nematic liquid crystals became unfeasible [2, 3].

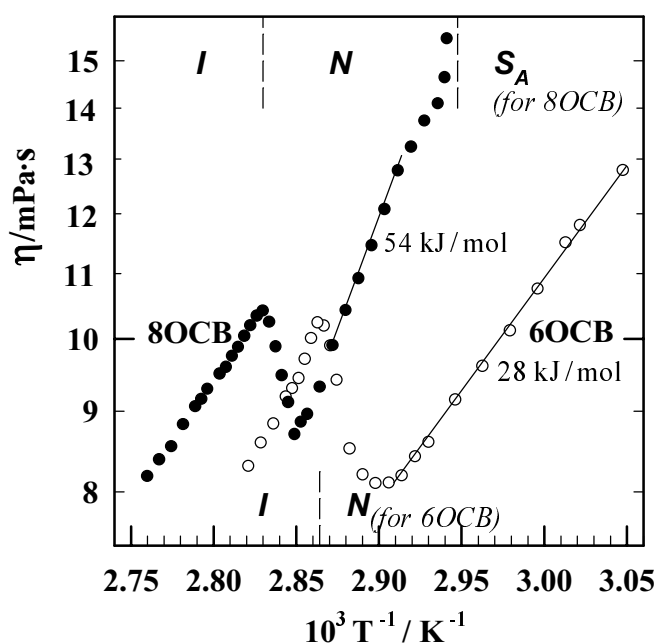


Figure 2. Temperature dependence of the shear viscosity measured for two freely flowing mesogens: *n*-hexyloxycyanobiphenyl (6OCB) and *n*-octyloxycyanobiphenyl (8OCB). The values of the viscosity activation energy are given in the picture.

The experimental results presented in figure 2 demonstrate the above mentioned problem. The figure depicts the temperature dependence of the shear viscosity measured for two mesogenic substances in a free flow. The first substance, *n*-hexyloxycyanobiphenyl ($C_6H_{13}O-Ph-Ph-C\equiv N$, 6OCB), has only the isotropic and nematic phases with the transition between them at 76 °C. The second substance is *n*-octyloxycyanobiphenyl ($C_8H_{17}O-Ph-Ph-C\equiv N$, 8OCB), with the isotropic, nematic and smectic *A* phases. The isotropic to nematic phase transition occurs at 80 °C and the nematic to smectic *A* transition at 66 °C.

The viscosity was determined with a Haake viscometer Rotovisco RV 20 with the measuring system CV 100, consisting of a rotary beaker filled with the studied liquid and a cylindrical sensor (Mooney–Ewart type (ME15)), placed in the centre of the beaker. The liquid gap was 0.5 mm. The accuracy of the viscosity determination was 0.5%. In the available range of shear rates (30–300 s^{-1}) the liquids studied show a Newtonian behaviour both in isotropic and nematic phases. The temperature of the sample was controlled with a Haake F3 thermostat with an accuracy of ± 0.1 °C.

As seen in figure 2, the difference in temperature dependence of the viscosity of both compounds in the nematic phase is significant. In spite of the lack in the literature of experimental data on the Mięśowicz viscosity coefficients for 6OCB, the results presented in figure 2 show that the viscosity of freely flowing nematic 6OCB behaves in a very similar

way to the Mięśowicz η_2 viscosity coefficient of other nematics [2, 3]. The activation energy resulting from the temperature dependence of the viscosity of 6OCB is equal to 28 kJ mol^{-1} , the typical value for the nematic liquid crystals [13].

The analysis of the temperature dependence of the viscosity measured in the nematic phase of 8OCB is not as simple as in the case of 6OCB. As a matter of fact, the viscosity of 8OCB shows certain features of the η_2 Mięśowicz viscosity coefficient, however, with some distortion in the region just after the transition to the nematic phase, with unexpectedly strong temperature dependence and a very high value of the activation energy (54 kJ mol^{-1}) and with some irregularities beginning at about 70°C . Indeed, these anomalies can discourage one from studying the viscosity of freely flowing nematic liquid crystals.

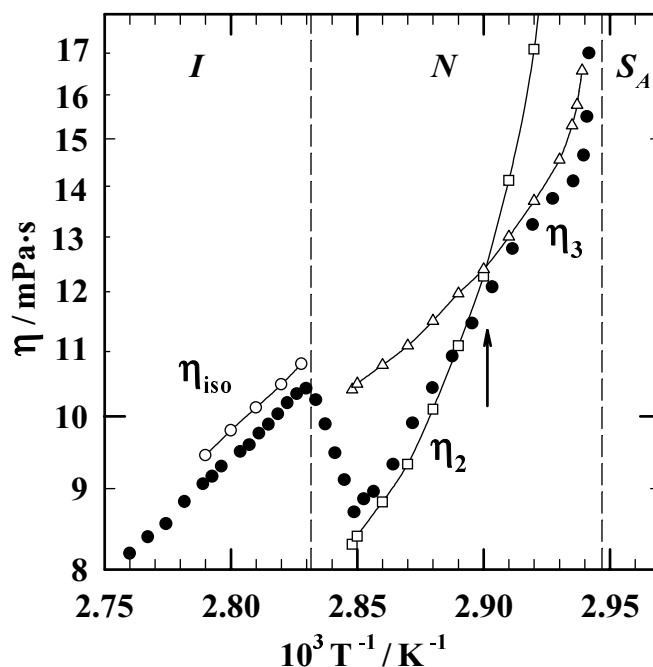


Figure 3. Temperature dependence of the shear viscosity of 8OCB measured in the free flow (full points) and the η_2 and η_3 Mięśowicz viscosities measured by Graf *et al* [4] for the sample oriented with magnetic field (open points).

The elucidation of these anomalies is possible due to the measurements of the Mięśowicz viscosity coefficients which were expertly carried out by Graf *et al* for 8OCB [4]. In figure 3 the results obtained by us for freely flowing 8OCB (full points) are shown together with the results of Graf *et al* (open points linked by the lines) obtained with the use of a magnetic field for the nematic alignment as in figures 1(b) and 1(c), respectively. Although the orientational molecular alignment at the walls cannot be neglected (especially in the temperature region just before the transition to the nematic phase), the results presented in figure 3 show evidence that at about 71°C the freely flowing nematic 8OCB changes its manner of flow from that presented in figure 1(b) to that presented in figure 1(c).

We believe that this observation could have wide significance and that the results presented here only illustrate a general principle which can be formulated as follows: *a free fluid adopts such a manner (or way) of flow, that corresponds to the minimum of its viscosity at given conditions.*

According to this principle a free flow of nematic liquid crystals is followed by the molecular alignment as in figure 1(b) only in the circumstances when the η_2 is the lowest possible viscosity. As a matter of fact, it always happens for the nematic liquid crystals in the absence of the pretransitional effects. In the presmectic region where the η_3 viscosity is the lowest viscosity, the nematic free flow is followed by the molecular ordering corresponding to figure 1(c).

In the case of isotropic fluids *the principle of the viscosity minimum* should be especially useful in the description of the flow of the fluids in the presence of the viscosity gradient which can be caused, for example, by the temperature gradient or by mixing of two flowing fluids of essentially different viscosities.

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