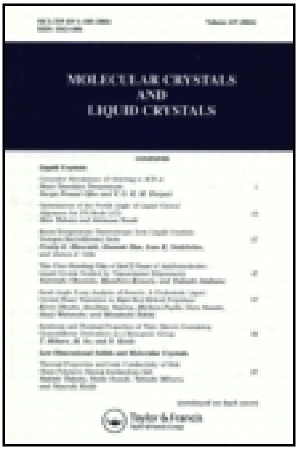

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The First Normal-Stress Difference in Shear of Nematic Liquid Crystals

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The First Normal-Stress Difference in Shear of Nematic Liquid Crystals

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The first normal-stress difference in shear flow of low-molecular-weight nematics is calculated from the Leslie-Ericksen theory. It is shown that, depending on the boundary orientation, the stress difference is either always negative, always positive or changes from negative to positive with increasing shear stress. This investigation was prompted by recent experimental studies demonstrating negative values of the first normal-stress difference for polymeric liquid crystals.

1 INTRODUCTION

It has recently been reported by Kiss and Porter[1] that, at certain shear rates, negative values of the first normal-stress difference are observed for certain polymeric liquid crystals (concentrated solutions of poly (γ-benzylglutamate)). This result is certainly unexpected, since all non-mesomorphic polymeric liquids are found to have a positive first normal-stress difference.[2] One consequence of the negative normal-stress difference is that mesomorphic polymers can exhibit negative dieswell.

To my knowledge, no explanation of this negative normal-stress difference has been given. There is, therefore, good reason to enquire what sign the first normal-stress difference will have in a low-molecular-weight nematic when this is sheared. Of course, there is no suggestion that the Leslie-Ericksen continuum theory[3,4] for low-molecular-weight nematics is adequate for describing the flow behavior of polymeric liquid crystals. Nevertheless, it is conceivable that some shared mesomorphic characteristics of both systems might give rise to similar normal-stress behavior.

We discuss nematics that orient in shear ($\alpha_3 < 0$) and consider shear flow between parallel plates, with the orientation lying in the plane of shear and strongly anchored on the boundaries. It is found that, depending on the boundary orientation, the first normal-stress difference is either always negative, always positive, or changes from negative to positive with increasing shear stress. Further, the normal-stress difference is of the same order of magnitude as the shear stress.

2 ANALYSIS

A nematic liquid crystal is confined between infinite parallel plates a distance $2h$ apart, one of which is moved with constant speed V parallel to the plates and the other with the same speed V in the opposite direction. The flow predicted by the Leslie-Ericksen theory[3,4] has been studied in some detail by MacSithigh and Currie[5-7] whose analysis we use subsequently. In Cartesian axes, with the origin half-way between the plates, y-axis normal to the plates and the x-axis parallel to the motion of the plates, the velocity vector $\mathbf{v}$ and the orientation director $\mathbf{n}$ are assumed to have the following components:

$$v_x = u(y), \quad v_y = v_z = 0,$$
$$n_x = \cos\theta,\ n_y = \sin\theta,\ n_z = 0,\ \theta = \theta(y). \tag{1}$$

For this flow, the first normal-stress difference $\sigma_{11} - \sigma_{22}$ is found from the Leslie-Ericksen theory[4] to be given by

$$\sigma_{11} - \sigma_{22} = f(\theta)(\theta')^2 + u' \sin\theta\cos\theta\{\alpha_1(\cos^2\theta - \sin^2\theta) - \alpha_2 - \alpha_3\} \tag{2}$$

where $\alpha_1 \ldots \alpha_6$ are the viscosity coefficients[4] (denoted by $\mu_1 \ldots \mu_6$ in Refs. 5, 6, 7) and

$$f(\theta) = K_1\cos^2\theta + K_3\sin^2\theta, \tag{3}$$

K_1 and K_3 being the splay and bending free energy coefficients.[3,4] However, in shear flow (Ref. 7, Eqs. 2.3 and 2.8)

$$u' = c/g(\theta), \tag{4}$$

$$f(\theta)(\theta')^2 = c\{B + \int^{\theta}(\gamma_1 + \gamma_2\cos 2\phi)/g(\phi)d\phi\}, \tag{5}$$

where c is the shear stress applied to the plates, B is an integration constant and

$$2g(\theta) = 2\alpha_1\sin^2\theta\cos^2\theta + (\alpha_5 - \alpha_2)\sin^2\theta + (\alpha_3 + \alpha_6)\cos^2\theta + \alpha_4,$$
$$\gamma_1 = \alpha_3 - \alpha_2,\ \gamma_2 = \alpha_6 - \alpha_5. \tag{6}$$

To determine the constant B, additional assumptions must be made about the nature of the solution for θ. It is assumed that the orientation is strongly anchored at the plates, at angle θ_1 (defined only up to a multiple of π by (1)). However, there are still infinitely-many possible solutions of Eq. (5). These solutions are discussed in detail by Currie and MacSithigh.[6] Considerations of stability and dissipation lead them to suggest that, at sufficiently high shear stress c, the solution for θ will be symmetric in y and monotonic for $0 \leq y \leq h$, attaining a maximum (or minimum) θ_m at $y = 0$. This type of solution satisfies either

$$\theta_c < \theta_1 (\text{mod } \pi) \leq \theta(y) \leq \theta_m \leq \theta_o, \tag{7}$$

(if $\theta_c < \theta_1(\text{mod } \pi) \leq \theta_o$) or

$$\theta_o \leq \theta_m \leq \theta(y) \leq \theta_1(\text{mod } \pi) \leq \pi + \theta_c, \tag{8}$$

(if $\theta_o \leq \theta_1(\text{mod } \pi) \leq \pi + \theta_c$) where $\theta_o = \cos^{-1}(-\gamma_1/\gamma_2)/2$ is the Leslie angle and θ_c is the critical angle introduced by Currie and MacSithigh,[6] which satisfies

$$\int_{\theta_c}^{\theta_o} (\gamma_1 + \gamma_2 \cos 2\phi)/g(\phi) d\phi = 0. \tag{9}$$

However, if the shear stress c is low, and $\theta_c < \theta_1(\text{mod } \pi) < -\theta_o$, then a solution of the above type is not possible, and it is not clear what type of solution will be adopted by the material. Since our interest here is principally in the normal-stress difference, we avoid this difficulty by restricting θ_1 to the range

$$-\theta_o \leq \theta_1 \leq \theta_c \tag{10}$$

for which solutions of the type (7) and (8) are always possible, and seem intuitively the most likely.

Since $\theta' = 0$ when $\theta = \theta_m$, it follows from (2), (4) and (5) that on the plates (where $\theta = \theta_1$) the first normal-stress-difference is given by

$$(\sigma_{11} - \sigma_{22})/c = \sin \theta_1 \cos \theta_1 \{\alpha_1(\sin^2 \theta_1 - \cos^2 \theta_1) - \alpha_2 - \alpha_3\}/g(\theta_1) + \int_{\theta_m}^{\theta_1} (\gamma_1 + \gamma_2 \cos 2\phi)/g(\phi) d\phi \tag{11}$$

We assume Parodi's relation[8-10] $\alpha_6 = \alpha_5 + \alpha_3 - \alpha_2$. Then (11) can be integrated to give

$$(\sigma_{11} - \sigma_{22})/c = \sin \theta_1 \cos \theta_1 \{\alpha_1(\cos^2 \theta_1 - \sin^2 \theta_1) - \alpha_2 - \alpha_3\}/g(\theta_1) + (2/\delta)[(\alpha_2\beta_2 + \alpha_3/\beta_2) \tan^{-1}(\tan \theta/\beta_2) - (\alpha_2\beta_1 + \alpha_3/\beta_1) \tan^{-1}(\tan \theta/\beta_1)]_{\theta_m}^{\theta_1}, \tag{12}$$

where

$$\begin{aligned} \beta_1^2 &= (\alpha_4 + \alpha_5 + \alpha_3 + \alpha_1 + \delta)/(\alpha_4 + \alpha_5 - \alpha_2), \\ \beta_2^2 &= (\alpha_4 + \alpha_5 + \alpha_3 + \alpha_1 - \delta)/(\alpha_4 + \alpha_5 - \alpha_2), \\ \delta^2 &= (\alpha_4 + \alpha_5 + \alpha_3 + \alpha_1)^2 - (\alpha_4 + \alpha_5 + \alpha_2 + 2\alpha_3)(\alpha_4 + \alpha_5 - \alpha_2) \end{aligned} \tag{13}$$

The value of the angle θ_m depends on the shear stress c. At zero shear stress, $\theta_m = \theta_1$, the boundary orientation. As the shear stress increases θ_m moves monotonically[5] towards the Leslie angle θ_o. Thus, the variation of $(\sigma_{11} - \sigma_{22})/c$ with θ_m indicates directly how the normal-stress-difference varies with increasing shear stress.

In Figure 1, $(\sigma_{11} - \sigma_{22})/c$ is plotted against $(\theta_m - \theta_1)/(\theta_o - \theta_1)$ for various boundary angles θ_1. Viscosity values are taken to be those suggested for MBBA by Tseng *et al.*:[11] $\alpha_1 = 0.0043$, $\alpha_2 = -0.0069$, $\alpha_3 = -0.0002$, $\alpha_4 = 0.0068$, $\alpha_5 = 0.0047\,(Ns/m)$, $\theta_0 = 9.7°$, $\theta_c = -21.1°$. Similar results are found for PAA using viscosity values reported by the Orsay Group.[12] As fol-

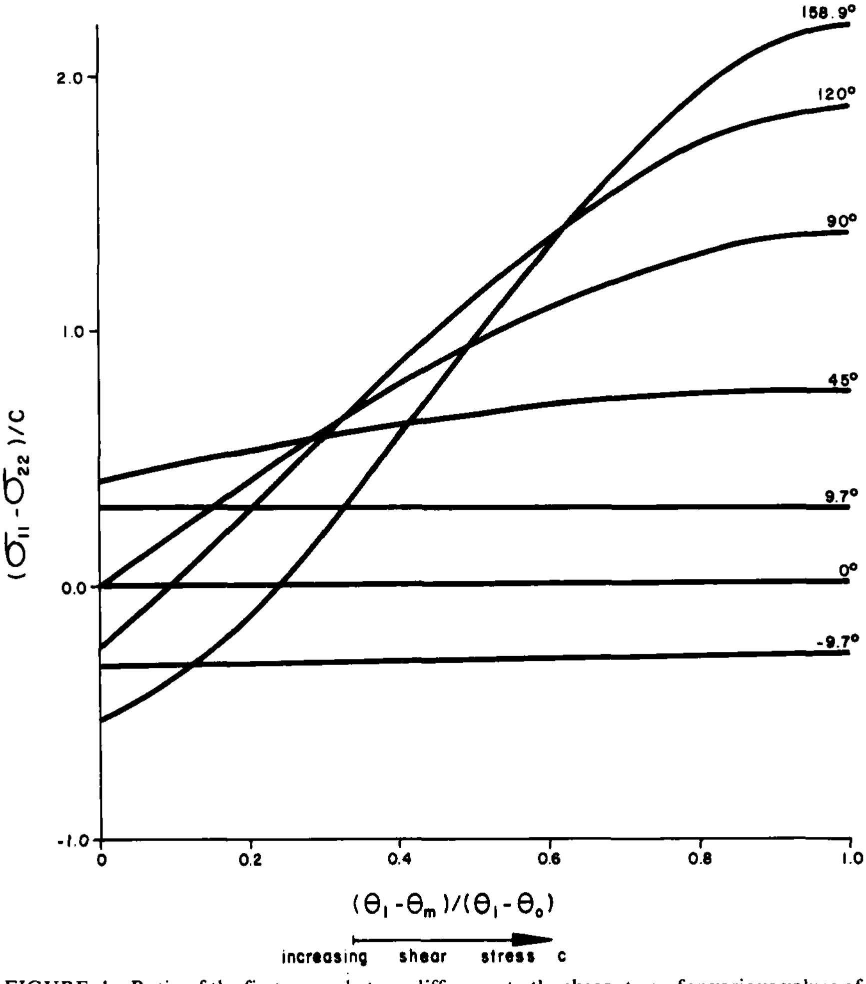

FIGURE 1 Ratio of the first normal-stress difference to the shear stress, for various values of the boundary orientation θ_1. Viscosity values for MBBA. $\theta_o = 9.7°$, $\theta_c = 21.1°$.

lows directly from Eq. (12), negative values of the normal-stress difference are found at very low shear stress ($\theta_m = \theta_1$) if $\sin 2\theta_1 < 0$, i.e. for the ranges $-\theta_0 < \theta_1 < 0$, $\pi/2 < \theta_1 < \pi + \theta_c$. As the shear stress increases, the normal-stress difference increases monotonically, and is positive for large values of the shear stress ($\theta_m = \theta_0$), except for boundary angles θ_1 in the range $-\theta_0 < \theta_1 < -0.65°$.

We note that the normal-stress difference is of the same order of magnitude as the shear stress c, and can be twice as large as c.

CONCLUSIONS

It has been shown that, for low-molecular-weight nematics that orient in shear ($\alpha_3 < 0$), the first normal-stress difference has the following properties:

(i) it is of the same order of magnitude as the shear stress, and can be up to twice as large.

(ii) it increases monotonically with the shear stress.

(iii) it can be always negative, always positive or change from negative to positive with increasing shear stress, depending on the boundary orientation.

Thus, a negative normal-stress difference, as observed by Kiss and Porter[1] for polymeric liquid crystals, can also occur in low-molecular-weight nematics. However, the sign of the normal-stress difference depends on the orienting effect of the boundary, an effect that does not seem to have been studied for polymeric liquid crystals. Moreover, the change from positive normal-stress difference, to negative and then back to positive (with increasing shear stress) observed by Kiss and Porter, is not predicted for low-molecular-weight nematics.

We conclude that orientational effects can produce negative first normal-stress differences in low-molecular weight nematics in simple shearing flow. The analysis given here will carry over to nearly all other viscometric flows.[13] However, for cone-and-plate flow (the geometry used by Kiss and Porter[1]) there has as yet been no solution found for nematics using the Leslie-Ericksen theory, so it cannot automatically be assumed that the results found here apply directly to that flow.

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References

1. G. Kiss and R. S. Porter, *J. Polym. Sci.: Polym. Symp.*, **65**, 193 (1978).
2. R. B. Bird, R. C. Armstrong and O. Hassager, *Dynamics of Polymeric Liquids* Vol. 1 (Wiley, New York, 1977) p. 143.

3. J. L. Ericksen, *Adv. Liq. Cryst.*, **2**, 233 (1976).
4. F. M. Leslie, *Adv. Liq. Cryst.*, **4**, 1 (1979).
5. G. P. MacSithigh and P. K. Currie, *J. Phys.*, D **10**, 1471 (1977).
6. P. K. Currie and G. P. MacSithigh, *Q. J. Mech. Appl. Math.*, **32**, 499 (1979).
7. P. K. Currie, *Rheol. Acta*, **16**, 205 (1977).
8. O. Parodi, *J. Phys. (Paris)*, **31**, 581 (1970).
9. P. K. Currie, *Solid State Comm.*, **12**, 31 (1973).
10. P. K. Currie, *Mol. Cryst. Liq. Cryst.*, **28**, 335 (1974).
11. H. C. Tseng, D. L. Silver and B. A. Finlayson, *Phys. Fluids*, **15**, 1213 (1972).
12. Orsay Liquid Crystal Group, *Mol. Cryst. Liq. Cryst.*, **13**, 187 (1971).
13. P. K. Currie, *J. de Physique*, **40**, 501 (1979).