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Frequency dependence of the electrorheological effect in the nematic phase of pentyl cyanobiphenyl

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The effect of an electric field on the viscosity, the electrorheological (ER) effect, is studied as a function of the frequency of the electric field for the nematic phase of 4-*n*-pentyl-4′-cyanobiphenyl (5CB). In the frequency region between 10 Hz and 1 kHz, a gradual change of the ER effect is observed, with its behaviour depending on the shear rate and the amplitude of the electric field. On the basis of a calculation of the orientational motion of the director and its effect on the viscosity, the observed frequency dependence is suggested to occur as a result of an orientational relaxation of the director.

1. Introduction

The application of an electric field of a few kV mm^{-1} induces a viscosity change in some colloidal suspensions and liquid crystals. This phenomenon is called the electrorheological (ER) effect and has been extensively studied both from the engineering and the scientific points of view [1–3]. In liquid crystals, some studies have been performed on various liquid crystalline phases [4–10], for which the effect in the nematic (N) phase has been well clarified [4–9]. When an electric field is applied, a nematic liquid crystal having a positive ε_a (dielectric anisotropy) shows an increase in viscosity [4–7, 9], while a decrease is observed for those having a negative ε_a [8]. In 5CB, which has a positive ε_a, an increase in viscosity is observed, accompanied by a change of the fluidity from a Newtonian to a non-Newtonian and again to a Newtonian flow with increase in the amplitude of the electric field [7, 9]. This result is successfully interpreted on the basis of the Leslie–Ericksen theory; the application of the electric field induces an orientational change of the director, leading to the change in viscosity [9].

When the ER effect is measured under an a.c. field, a frequency dependence for the effect is observed. In colloidal suspensions, the ER effect decreases with frequency and disappears at high frequencies. This is understood to occur as a result of the relaxation of the interfacial polarization induced between the particle and the liquid [11, 12]. On the other hand, the frequency dependence of the ER effect in liquid crystals is not well understood. In the present study, the ER effect in the nematic phase of 5CB is measured as a function of electric field frequency, with the finding that some characteristic frequency dependence of the ER effect is observed between 10 Hz and 1 kHz. This result is discussed in terms of the orientational motion of the director, indicating that the frequency dependence is caused by orientational relaxation of the director.

2. Experimental

The liquid crystal 5CB, which exhibits the nematic phase between 295.2 and 308.5 K, was obtained from Merck Ltd, UK, and was used without further purification. The measurement of the ER effect was made at a constant temperature, 300 K, in the N phase. The rheological properties were measured with a home-made viscometer of a double cylinder type in the shear rate region between 69.5 and 1318 s^{-1}. The electric field of a few kV mm^{-1} was generated by applying a high voltage to the gap (1 mm) between the inner and the outer cylinders, making the application of the electric field perpendicular to both the velocity and the velocity gradient directions. For the supply of high voltages, a variable frequency a.c. power source (CVFT1-50HVP2, Tokyo Seiden Co., Ltd, Japan), which operates with an amplitude of up to 5 kV rms and a frequency from 10 Hz to 5 kHz, was used. Throughout this report, the amplitude of the electric field is expressed in rms.

3. Results

As has already been clarified [7, 9], when an electric field is applied, the viscosity of 5CB at 300 K increases

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and saturates at high field with its magnitude about four times that in the absence of the electric field. The viscosity change is accompanied by a change of fluidity: the flow in the absence of the field is Newtonian, which changes to non-Newtonian at medium field and back to Newtonian at high field. These results are successfully interpreted on the basis of Leslie–Ericksen theory [13, 14]: the director in the absence of the electric field aligns near the velocity direction with the flow alignment angle θ given by $\tan^{-1}(\alpha_2/\alpha_3)^{1/2}$, which changes its direction parallel to the electric field at a high enough field, thus leading to the viscosity increase [9].

When measuring the frequency dependence of the ER effect, a characteristic behaviour is observed. Figure 1 shows the result at 300 K in the N phase, where the data measured at a shear rate of 988.5 s^{-1} are given. This figure shows that the frequency dependence changes with the amplitude of the electric field. Below 0.5 kV mm^{-1}, the shear stress gradually decreases with frequency, while above 1.0 kV mm^{-1} it increases with frequency and saturates around a few kHz with the increment of the shear stress being smaller at higher electric fields. The frequency dependence of the shear stress when varying the shear rate is also characteristic. As shown in figure 2, no frequency dependence is observed at lower shear rates, while at higher shear rates an increase of the shear stress is observed with increased frequency of the electric field. Such a frequency dependence in the N phase is different from that in ER suspensions, where the ER effect decreases with frequency and disappears at high frequencies. For example in the zeolite/silicone oil system the ER effect disappears around a few kHz, which has been interpreted to be caused by the relaxation of the interfacial polarization induced by the electric field [12].

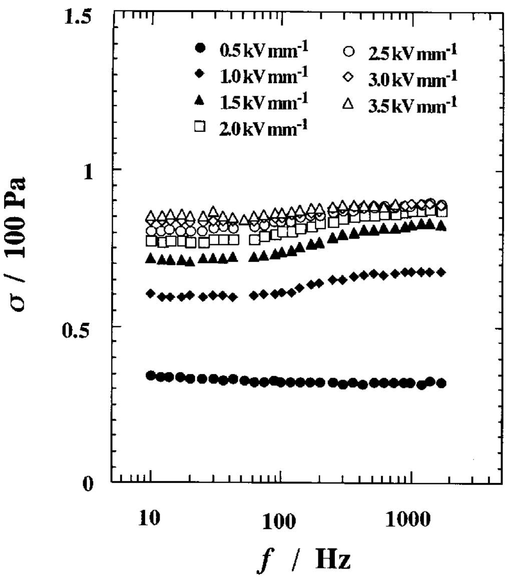

Figure 1. Frequency (f) dependence of the shear stress (σ), measured at a shear rate of 988.5 s^{-1}, at various amplitudes of the electric field. The measurements is made at 300 K in the N phase.

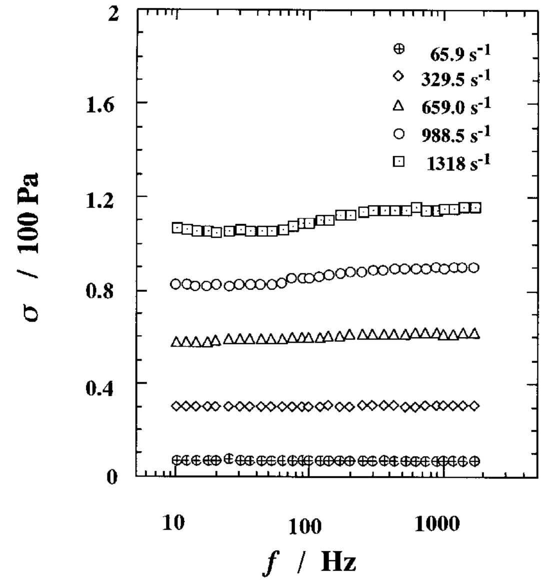

Figure 2. Frequency (f) dependence of the shear stress (σ), when an electric field of 3 kV mm^{-1} rms is applied, at various shear rates. The measurement is made at 300 K in the N phase.

4. Discussion

For discussion of the frequency dependence of the ER effect, we use the coordinate system given in figure 3, where the directions of the director, the shear deformation and the electric field are shown.

Since the observed frequency dependence of the ER effect is expected to be associated with the orientational motion of the director, its motion under the shear and the electric fields is considered on the basis of a balance of the torque exerted on the director. As is well known, the torque balance equation is given by [15]:

$$\Gamma_{m} + \Gamma_{r} + \Gamma_{v} + \Gamma_{e} + \Gamma_{el} = 0 \qquad (1)$$

where $\Gamma_m = I\partial^2\theta/\partial t^2$, $\Gamma_r = \gamma_1 \partial\theta/\partial t$, $\Gamma_v = (\alpha_3 \sin^2\theta - \alpha_2 \cos^2\theta)\dot{\gamma}$, $\Gamma_e = h(\theta)\partial^2\theta/\partial z^2 + h'(\theta)/2(\partial\theta/\mathrm{d}z)^2$, and $\Gamma_{el} = -1/2\varepsilon_a E^2 \sin 2\theta$

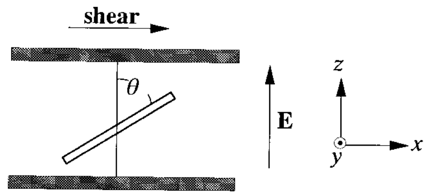

Figure 3. A coordinate system specifying the directions of the director, the shear deformation, and the electric field. θ is the orientational angle of the director; a two dimensional alignment of the director in the zx-plane is supposed.

are torques associated with the moment of inertia, the rotation, the shearing, the elastic distortion and the electric interactions, respectively. Here I is a moment of inertia, $\gamma_1 = \alpha_3 - \alpha_2$ (α_2 and α_3 are Leslie coefficients), $h(q) = K_1 \sin^2 \theta + K_3 \cos^2 \theta$ with K_1 and K_3 being splay and bend elastic constants, ε_a is the dielectric anisotropy, and E is the electric field ($\sqrt{2}E_0 \cos \omega t$). Of these terms Γ_m and Γ_e can be neglected owing to their small contribution [15, 16]. Furthermore if there occurs a flow alignment of the director, i.e. $\partial\theta/\partial t = 0$, the rotational term Γ_r becomes zero and the torque balance equation reduces to $\Gamma_v + \Gamma_{el} = 0$. This leads to equation (2), which determines an equilibrium orientational angle θ_e under the shear and the electric fields:

$$\tan \theta_e = 1/2\{\varepsilon_a E^2/(\alpha_3 \dot{\gamma}) + [(\varepsilon_a E^2/\alpha_3 \dot{\gamma})^2 + 4\alpha_2/\alpha_3]^{1/2}\}. \tag{2}$$

Using equation (2), the time variation of θ_e under an electric field of $E = \sqrt{2}E_0 \cos(2\pi t/t_p)$ is calculated, and the result is given in figure 4. For this calculation, the Leslie coefficients, $\alpha_2 = -70$, $\alpha_3 = -3.8$, $\alpha_4 = 71$, $\alpha_5 = 52$ and $\alpha_6 = -28$ mPa s, and the dielectric anisotropy $\varepsilon_a = 10.8$ at 300 K [17] are used. When $E = 0$ the equilibrium angle θ_e is 77°, which is derived from $\theta = \tan^{-1}(\alpha_2/\alpha_3)^{1/2}$, and it becomes smaller at higher fields with its behaviour depending on the amplitude of the electric field. The period of θ_e becomes one-half the period t_p of the electric field owing to the E^2 dependence of the electric torque Γ_{el}.

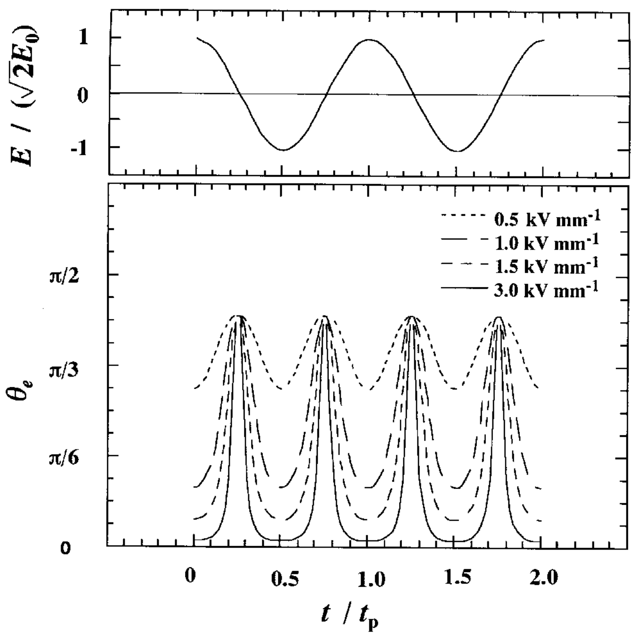

Figure 4. Calculated time dependence of the equilibrium orientational angle θ_e induced by the shear ($\dot{\gamma} = 988.5$ s^{-1}) and the electric fields. Time variation of the electric field, $\sqrt{2}E_0 \cos(2\pi t/t_p)$, is also given. t_p is the period of the electric field.

In the time variation of θ_e shown in figure 4, the relaxation effect of the orientational motion of the director is not taken into account. As has been shown [15], the relaxation time can be derived from the torque balance equation:

$$-\gamma_1 \partial\theta/\partial t = -(\alpha_3 \sin 2\theta - \alpha_2 \cos 2\theta)\dot{\gamma} + 1/2\varepsilon_a E^2 \sin 2\theta. \tag{3}$$

If we expand equation (3) around an angle θ_0, i.e. $\theta = \delta\theta + \theta_0$, we get

$$\begin{aligned} -\gamma_1 \partial(\delta\theta)/\partial t = {} & [-(\alpha_3 + \alpha_2)\dot{\gamma} \sin 2\theta_0 + \varepsilon_a E^2 \cos \theta_0]\delta\theta \\ & - (\alpha_3 \sin^2 \theta_0 - \alpha_2 \cos^2 \theta_0)\dot{\gamma} \\ & + 1/2\varepsilon_a E^2 \sin 2\theta_0. \end{aligned} \tag{4}$$

In the right hand side of the equation (4), the first term is dependent on $\delta\theta$ while the second and third terms are not, leading to an orientational relaxation time given by

$$\tau = \gamma_1/[-(\alpha_3 + \alpha_2)\dot{\gamma} \sin 2\theta_0 + \varepsilon_a E^2 \cos 2\theta_0]. \tag{5}$$

This equation shows that the relaxation time depends not only on the viscosity coefficients α_2, α_3 and γ_1, and the dielectric anisotropy ε_a but also on $\dot{\gamma}$ and E. For example, at $E = 2$ kV mm^{-1} and $\dot{\gamma} = 988.5$ s^{-1}, τ is about 1 ms.

When θ_0 deviates from θ_e, θ_0 tends to approach θ_e with a relaxation time τ given by equation (5). This effect is incorporated into the time dependence of the orientational motion of the director; an orientational angle θ_r, which includes the relaxation effect, is calculated at $E_0 = 3$ kV mm^{-1} and $\dot{\gamma} = 988.5$ s^{-1} (figure 5). Here behaviour at electric field frequencies of 10, 100 and 1000 Hz, which correspond to periods of 100, 10 and 1 ms, respectively, is shown. At low frequency, e.g. 10 Hz, the time variation of θ_r is similar to θ_e (figure 4), indicating that the relaxation effect is too small to affect the time dependence of θ. On the other hand, at the high frequency of 1000 Hz, the amplitude and phase of θ_r are largely modified from θ_e, suggesting that the orientational motion of the director is affected by the relaxation effect. The shear stress depends on the orientational angle and the shear rate [13–15]:

$$\sigma = 1/2[(\alpha_3 + \alpha_6)\sin^2 \theta_r + (\alpha_5 - \alpha_2)\cos^2 \theta_r + \alpha_4]\dot{\gamma}. \tag{6}$$

Inserting θ_r into equation (6), we get the time dependence of the shear stress. The calculated result thus obtained at a shear rate of 988.5 s^{-1} is depicted in figure 6.

In the measurement of the steady viscosity, it should be noted that we are measuring a time averaged shear

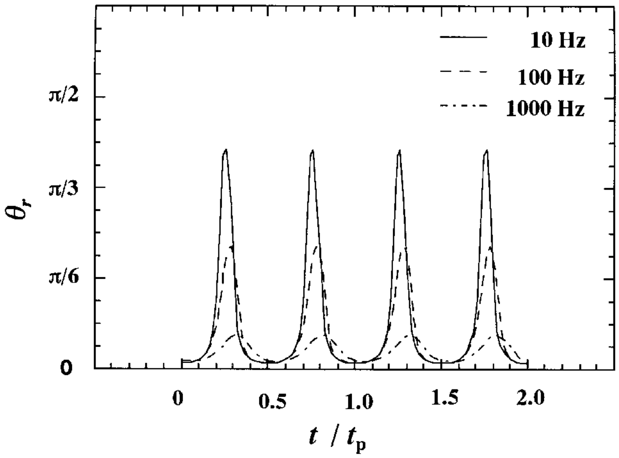

Figure 5. Calculated time dependence of the orientational angle θ_r induced by the shear ($\dot{\gamma} = 988.5\ s^{-1}$) and the electric field, $\sqrt{2}E_0 \cos(2\pi t/t_p)$, $E_0 = 3\,kV\,mm^{-1}$. The relaxation time is incorporated for the calculation of θ_r. At higher frequency, e.g. 1000 Hz, the relaxation effect can be recognized.

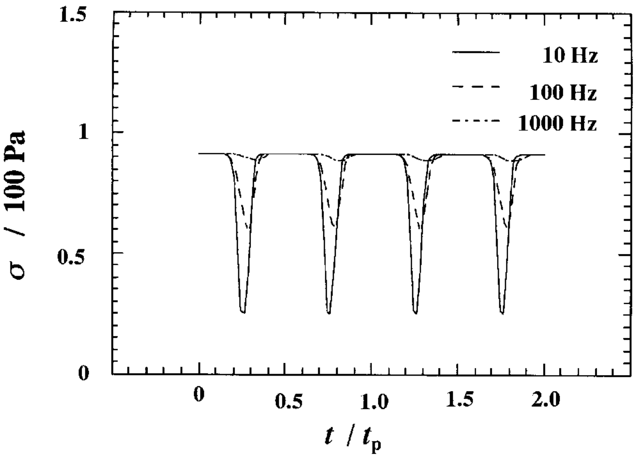

Figure 6. Calculated time dependence of the shear stress σ induced by the shear deformation ($\dot{\gamma} = 988.5\ s^{-1}$) and the electric field, $\sqrt{2}E_0 \cos(2\pi t/t_p)$, $E_0 = 3\,kV\,mm^{-1}$.

stress $\langle\sigma\rangle$. The calculation of $\langle\sigma\rangle$ is therefore made by varying the frequency of the electric field from 10 to 1000 Hz. The result is given in figure 7, showing that the calculated result as a function of the amplitude of the electric field is in good agreement with the experimental result (figure 1). This agreement indicates that the frequency dependence of the ER effect in the N phase is caused by the relaxation effect of the orientational motion of the director. This is further supported from the calculated shear rate dependence (figure 8), which also successfully replicates the experimental result (figure 2).

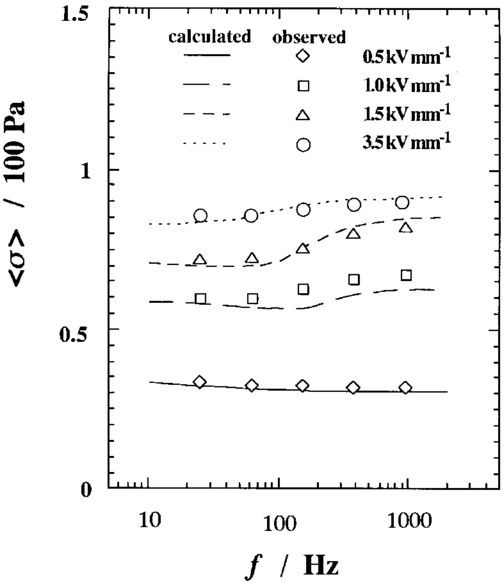

Figure 7. Calculated frequency dependence of the averaged shear stress, $\langle\sigma\rangle$, at various amplitudes of the electric field. The shear rate is $988.5\ s^{-1}$. Experimental results are specified by the various symbols indicated.

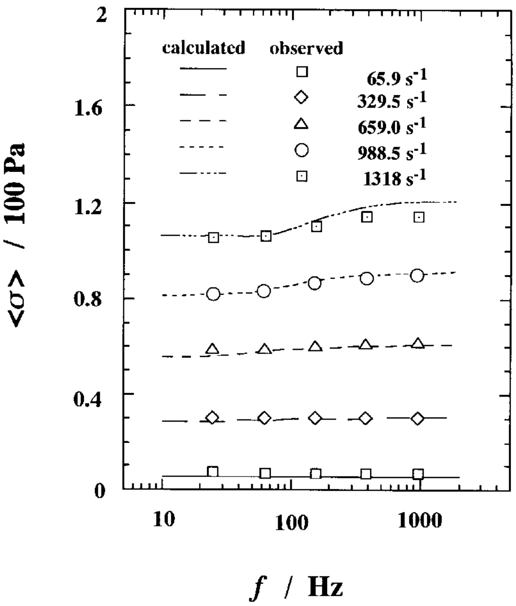

Figure 8. Calculated frequency dependence of the averaged shear stress, $\langle\sigma\rangle$, at various shear rates under an electric field of $3\,kV\,mm^{-1}$. Experimental results are specified by the various symbols indicated.

5. Summary and conclusion

Measurement of the ER effect has been made in the N phase of 5CB as a function of the frequency of the electric field, showing that the frequency dependence is different from that observed in colloidal suspensions. By including the relaxation effect of the orientational motion of the director, a calculation of the frequency dependence of the ER effect has been performed. The calculated result is in good agreement with experimental measurement, indicating that the observed frequency dependence is indeed caused by the relaxation of the orientational motion of the director.

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