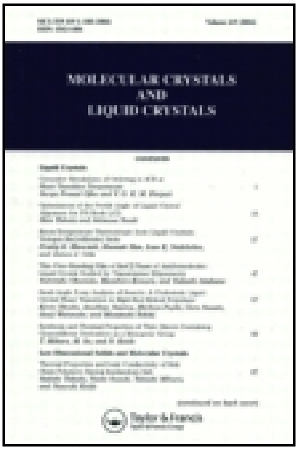

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Investigation of the Temperature Dependence of the Pitch of a Cholesteric Liquid Crystal

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Investigation of the Temperature Dependence of the Pitch of a Cholesteric Liquid Crystal

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The pitch and the refractive index of a cholesteryl chloride—cholesteryl nonanoate mixture as a function of temperature have been measured separately by using selective reflections and total reflections. The magnitude of the influence on the pitch due to the temperature dependence of the refractive index and the density has been determined.

1 INTRODUCTION

Selective reflection and other characteristic optical properties of cholesteric liquid crystals can be explained in terms of Bragg-like reflections caused by their internal helical structure.[1] The mean refractive index n and the pitch P of the helical arrangement are the basic optical parameters which characterize cholesteric liquid crystals. The wavelength of selective reflection for normal incidence, λ_0, is given by

$$\lambda_0 = n \times P \tag{1}$$

$$= n \times l \times \frac{2\pi}{\theta}, \tag{2}$$

where l is the interlayer distance and θ is the twist angle between the successive layers.

In many cholesteric materials λ_0 is very temperature sensitive and this property makes liquid crystals quite useful in thermography. From the measurement of the temperature dependence of λ_0,[1–5] of the density,[6–9] and of the refractive index,[10–14] it is known that the primary contribution to the temperature dependence of λ_0 is that of the twist angle θ. There are several theories[15,16] and phenomenological treatments[17] of this subject. Temperature changes in the refractive index and the density are related to those of λ_0. However, the magnitude of these small effects have not actually been determined.

The main purposes of the present investigation are to measure the temperature dependence of λ_0, the refractive index and the density of a mixture composed of cholesteryl chloride (CC) and cholesteryl nonanoate (CN), and to determine quantitatively the degree to which the temperature dependence of the refractive index and the density effects.

2 SAMPLE PREPARATION

A mixed crystal system composed of CC and CN is chosen for this investigation because it exists in a cholesteric state in the wide temperature range (15°C–65°C). The purity of these two component materials, which are obtained commercially and used without further purification, is higher than 98 %. The sample consists of molar percentages of CC(79 %) and CN(21 %). It is prepared by heating the materials to 100°C, mixing them mechanically without solvent, and allowing it to cool slowly to room temperature. Then it is kept in a desiccator.

3 TEMPERATURE DEPENDENCE OF THE DENSITY

The density of the sample was measured with an arrangement shown in Figure 1. A dilatometer made of Pyrex glass was cleaned with chromic acid

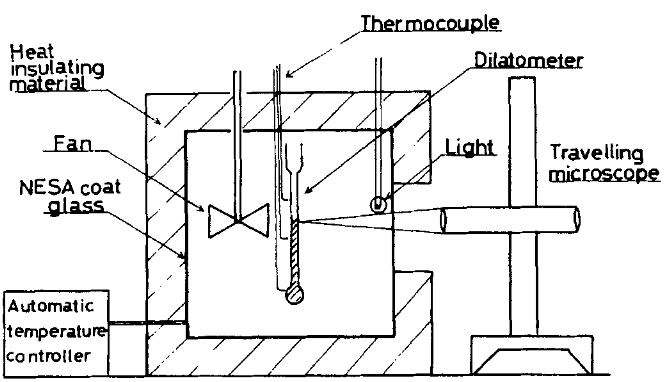

FIGURE 1 Experimental arrangement for density measurement.

and acetone and allowed to dry in a desiccator. The dilatometer was calibrated by distilled water. To read the scale, a travelling microscope whose resolution was 0.01 mm was used. The chief systematic error which limits the accuracy of the density determination is the mixed air in the sample. Therefore the sample was heated into the liquid state and the air in it was removed by a vacuum pump. Next, the sample in the liquid state was put into the dilatometer. Then the dilatometer was set in a thermobath whose temperature was controlled within the accuracy of 0.2°C. The temperature of the thermobath was changed by 2.5°C decrements from 78°C to 28°C, but in the vicinity of the transition point from liquid crystal state to liquid state it was changed every 0.4°C. Figure 2 shows the temperature dependence of the density of the sample. The transition temperature was (64.6 ± 0.2)°C for this sample.

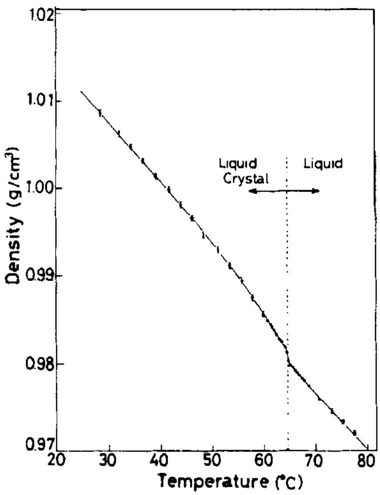

FIGURE 2 Density versus temperature.

4 TEMPERATURE DEPENDENCE OF THE REFRACTIVE INDEX AND THE PITCH

A thin cholesteric film was sandwiched between a pair of semi-cylindrical prisms as shown in Figure 3. The refractive index of the prism made of SF-5 glass was greater than that of cholesteric film. A 10μm-thick Mylar film was used as a spacer. The spiral axis of the cholesteric film was arranged to be perpendicular to the boundary. To obtain such a state, the shearing force was imposed on a cholesteric film by sliding the two prisms back and forth. This sample cell was set in the thermobath as shown in Figure 4. The temperature of the thermobath was controlled within the accuracy of 0.1°C.

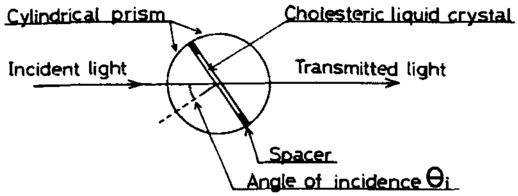

FIGURE 3 Constitution of the sample cell.

It has been known[18] that for a sandwich cell of a cholesteric liquid crystal, total reflection takes place at the incident angle

$$\theta_t = \sin^{-1} \frac{n}{n_g} \tag{3}$$

for perpendicular polarization, and at the incident angle

$$\theta_{3,t} = \sin^{-1} \frac{n_3}{n_g} \tag{4}$$

for parallel polarization. In Eqs. (3) and (4), n_g is the refractive index of the prism. Refractive indices n and n_3 are given by

$$n = \left(\frac{\varepsilon_p + \varepsilon_n}{2}\right)^{1/2}, n_3 = \varepsilon_n^{1/2} \tag{5}$$

where ε_p and ε_n are the components of the dielectric tensor parallel and perpendicular to the long axis of a molecule respectively. By measuring θ_t

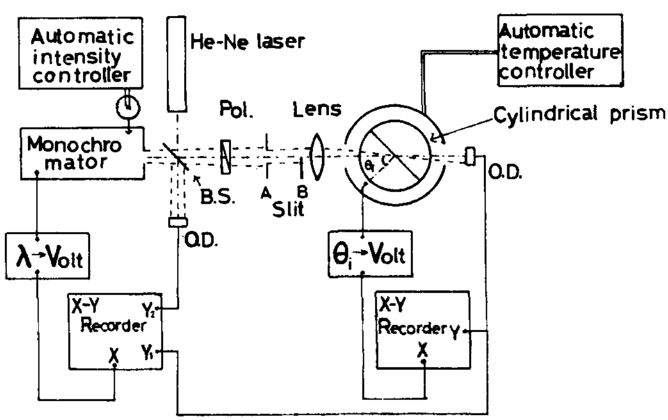

FIGURE 4 Experimental arrangement for determining refractive indices and pitch. Pol.: polarizer. O.D.: silicon photo-diode. B.S.: beam splitter.

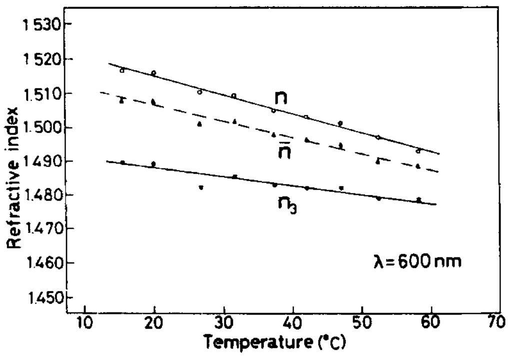

FIGURE 5 Refractive indices versus temperature for 600 nm.

and $\theta_{3,t}$, n and n_3 were determined. Figure 5 shows the temperature dependence of the refractive index of the sample for $\lambda = 600$ nm. There $\bar{n}$ is given by

$$\bar{n} = \left(\frac{2n^2 + n_3^2}{3}\right)^{1/2}. \tag{6}$$

Temperature coefficients of n and n_3 are $-5.7 \times 10^{-4}/°C$ and $-2.6 \times 10^{-4}/°C$ respectively. Figure 6 shows the dispersion curves of the refractive index at 37.5°C. Abbe's numbers of n and n_3 are 23 and 24 respectively.

The pitch was determined with following procedures.[19] First, the incident angle of the light beam is set at $\theta_i = \theta$ in Figure 4. Next, the wavelength of the incident light is varied, then at a certain wavelength λ_B the Bragg-like reflection occurs and the intensity of the transmitted light has a sharp dip.

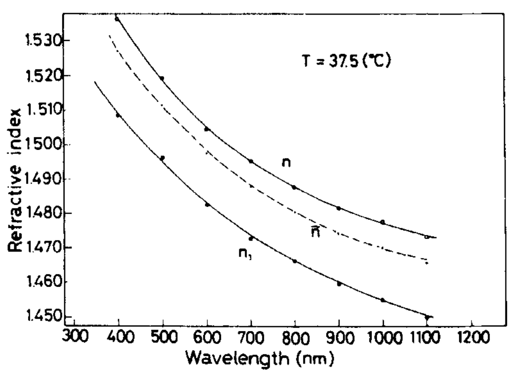

FIGURE 6 Refractive indices versus wavelength at 37.5°C.

Then the pitch P can be determined by[1]

$$P = \lambda_B \times (n^2(\lambda_B) - n_g^2(\lambda_B)\sin^2\theta)^{-1/2}$$

$$= \frac{\lambda_B}{n(\lambda_B)\cos\varphi}, \tag{7}$$

where $n(\lambda_B)$ and $n_g(\lambda_B)$ are the refractive indices of the cholesteric film and the prism at the wavelength λ_B respectively, and φ is the refracted angle in the cholesteric film. For the normal incidence ($\theta_i = 0$), Eq. (7) reduces to

$$P = \frac{\lambda_0}{n(\lambda_0)}. \tag{8}$$

The spectral response range of our optical detector (silicon photo-diode) was 400 nm–1100 nm. If λ_0 is within this range the pitch can be determined easily from Eq. (8). The temperature dependence of the pitch P and the wavelength of selective reflection λ_0 is shown in Figure 7. When the temperature was above 35°C the pitch was obtained from Eq. (8) and below 35°C from Eq. (7). The pitch decreases monotonically from 1 μm to 0.5 μm as the temperature rises.

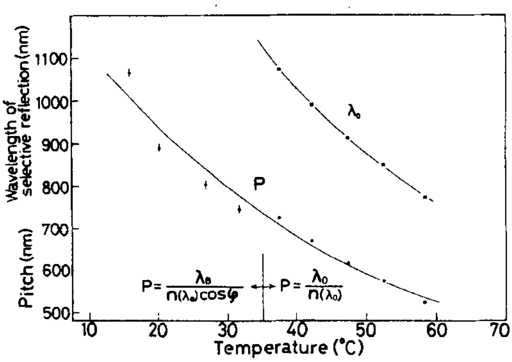

FIGURE 7 Pitch and wavelength of selective reflection versus temperature.

5 DISCUSSION

To discuss the influence of the temperature dependence of the refractive index and the density on that of λ_0, we consider the temperature coefficients of λ_0, the refractive index n, the interlayer distance l and the twist angle θ. By differentiating Eq. (2) with respect to temperature T, we obtain

$$\frac{1}{\lambda_0}\frac{d\lambda_0}{dT} = \frac{1}{n}\frac{dn}{dT} + \frac{1}{l}\frac{dl}{dT} - \frac{1}{\theta}\frac{d\theta}{dT}. \tag{9}$$

The temperature coefficients of λ_0, P and n at 40°C, 50°C and 60°C are listed in the first, the second and the third rows of Table I. In the fourth row of Table I, the temperature coefficients of l are listed. It is assumed that the temperature coefficient of l is equal to the coefficient of linear expansion of the sample. The coefficient of linear expansion is obtained from Figure 2 on the assumption that cubic expansion is isotropic.

Table I shows that the temperature coefficient of λ_0 is very much greater than that of n and l. It is therefore seen that the temperature change of λ_0 is caused mainly by that of θ. We note that the temperature coefficients of n and l are about $\frac{1}{70}$ and $\frac{1}{60}$ of that of λ_0, respectively. The signs of the temperature coefficient of n and l are the same and opposite to that of λ_0 respectively. For this sample, θ increases with increasing temperature, and consequently the pitch decreases and λ_0 decreases.

TABLE I

Temperature coefficients of wavelength of selective reflection λ_0, pitch P, refractive index n and interlayer distance l.

T (°C)	$\frac{1}{\lambda_0}\frac{d\lambda_0}{dT}$ ($\times 10^{-2}$/°C)	$\frac{1}{P}\frac{dP}{dT}$ ($\times 10^{-2}$/°C)	$\frac{1}{n}\frac{dn}{dT}$ ($\times 10^{-4}$/°C)	$\frac{1}{l}\frac{dl}{dT}$ ($\times 10^{-4}$/°C)
40	−1.7	−1.7	2	2.3
50	−1.4	−1.4	2	2.7
60	−1.4	−1.4	2	2.9

6 CONCLUSION

Temperature dependence of the pitch, the refractive index and the density was measured for a mixed cholesteric liquid crystal.

The change in magnitude of the wavelength of selective reflection for normal incidence due to the temperature dependence of the refractive index and interlayer distance was also determined.

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