

## Refractive Index Ellipsoids of a Polycarbonate Magneto Optical Memory Disk Substrate

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To investigate the generating mechanism of optical anisotropy in a polycarbonate (PC) magneto optical memory disk substrate, refractive index ellipsoids were measured for as-molded and annealed disk substrates. The shape of the refractive index ellipsoid for the as-molded disk substrate was muffin-shaped; that is, the thickness direction was substantially smaller than the other directions. Even after an annealing process to remove residual stress in the disk substrate, the shape of the ellipsoids remained anisotropic and muffin-shaped. This fact suggests that the molecular orientation of constructive units with anisotropic refractive indices is mainly responsible for the optical anisotropy in this PC substrate.

**KEYWORDS:** magneto optical memory disk, refractive index ellipsoid, polycarbonate, annealing, retardation

### §1. Introduction

As a magneto optical (MO) disk substrate material, polycarbonate (PC) is thought to be most suitable because of its excellent thermal, mechanical and low-hygroscopic properties. Although PC has superior injection molding characteristics, an injection molded disk substrate shows large retardation.<sup>1-3)</sup>

Since the MO readout is accomplished by detecting a slight rotation in the polarization of a linearly polarized beam reflected off the recording media, retardation of the disk substrate degrades the performance of the MO readout.<sup>4-6)</sup> Although some attempts have been made to reduce the amount of retardation by improving the injection molding process for a normal incident beam,<sup>1-3,8,9)</sup> or by analyzing the refractive index ellipsoid for oblique rays,<sup>7,8)</sup> a clear means of solving this problem has not been obtained yet, probably because of the lack of consideration for the direction of birefringence.

In this study, to determine the appropriate method of lowering the retardation in PC disk substrate, we measured refractive index ellipsoids of as-molded and annealed PC disk substrates and investigated the generating mechanism of large retardation for oblique rays.

### §2. Experimental

We constructed a rotating analyzer birefringence measuring system for this study. The block diagram of this system and the directions of the principal axes in constructive optical elements are shown in Fig. 1. The retardation  $\Delta$  caused by the difference in the principal values of the refractive index ellipse on the plane normal to the beam direction and the angle  $\theta$  between the major axis of this ellipse and the polarized axis of the incident beam on that plane was measured. By applying Jones matrix calculation for this system,  $\Delta$  and  $\theta$  are obtained from the rotation angles of the polarized axis of the analyzer  $\alpha$  and corresponding transmitted light intensities  $I(\alpha)$  as

$$\theta = \tan^{-1} (S_1/S_2)/2$$

$$\Delta = \sin^{-1} (-S_1/(S_0 \cdot \sin 2\theta))$$

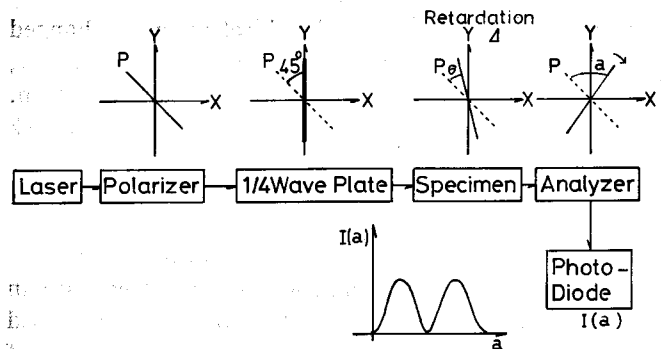


Fig. 1. Constructions of the birefringence measuring system and principal axes directions of each optical elements. Transmitted light intensities are measured as the function of  $\alpha$  (direction of polarized axis in analyzer).

where

$$S_0 = \sum_{\alpha=0}^{180} I(\alpha)/180$$

$$S_1 = \sum_{\alpha=0}^{180} I(\alpha) \cdot \cos 2\alpha/90$$

$$S_2 = \sum_{\alpha=0}^{180} I(\alpha) \cdot \sin 2\alpha/90$$

As PC disk substrates, we used Idemitsu OD3 disk substrates (diameter, 130 mm; thickness, 1.2 mm; Idemitsu-Petrochemical Co., Ltd.).

The refractive index ellipsoids for the disk substrates were obtained from the data of the birefringence measured with incident beams at different directions, as depicted in Fig. 2. We took the X-, Y-, and Z-axes as the radial, tangential, and thickness directions respectively, and defined the direction of the incident beam by a combination of incident and rotation angles, which, respectively, show the discrepancy from the Z-axis, and rotation around the Z-axis. (The rotation angle setted 0 when the incident beam was on the XZ-plane.)

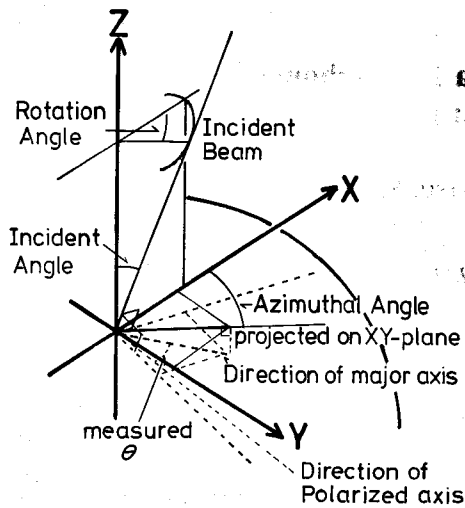


Fig. 2. Definition of beam directions and directions of principal axes.

### §3. Results and Discussion

In the disk substrate, the birefringence changed substantially in the radial direction. On the contrary, there was no obvious change in the tangential direction. Hence, we measured refractive index ellipsoids of disk substrates at different distances of 25, 35, 45, 55 mm from the center along one radial direction.

Typical results of the retardation at 45 mm are shown in Fig. 3. A very small amount of retardation was observed when the incident angle was  $0^\circ$ . However, with an increase in the incident angle, the retardation increased rapidly indicating the existence of a large anisotropy of the refractive index ellipsoid in the Z direction. From this result, two cases, that is, the value in the Z direction is either smaller or larger than that in the X, or Y direction, were suggested as a possible ellipsoid shape. To determine the true ellipsoid shape, it is necessary to consider the directions of the major axes in the refractive index ellipsoids responsible for these retardations.

As shown in Fig. 2, the azimuthal angle was determined as the angle formed by the X-axis and projected direction of the major axis on the XY-plane, it was calculated from the measured angle  $\theta$ , taking the direction of the polarized axis of the incident beam into consideration. Typical results of the azimuthal angle corresponding to Fig. 3 are shown in Fig. 4. For convenience,

we employed the approximated value of 1.5800\* as the lowest value of the refractive index, and obtained the refractive index ellipsoid as that which satisfied Fig. 3 and Fig. 4 and their equivalent.

The typical shape of a refractive index ellipsoid is shown in Fig. 5 where the difference is emphasized. The ellipsoid is muffin-shaped, where the two principal values in the XY-plane are almost the same, while the third principal value in the thickness direction is substantially smaller than the others. Numerical values of the differences in these principal values,  $\Delta n_{xz}$  and  $\Delta n_{xy}$ , are shown in Table I.

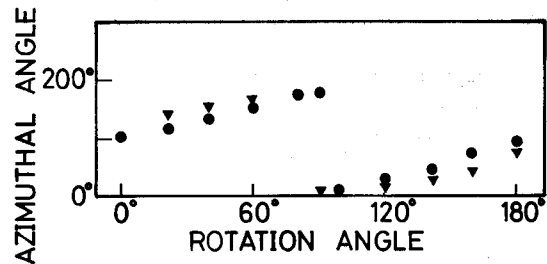


Fig. 4. Typical results of azimuthal angles (angles between projected directions of the principal axes on the XY-plane and X-axis) corresponding to Fig. 3. The results at the incident angle of  $0^\circ$  and  $20^\circ$  are overlapped with the data at  $30^\circ$ .

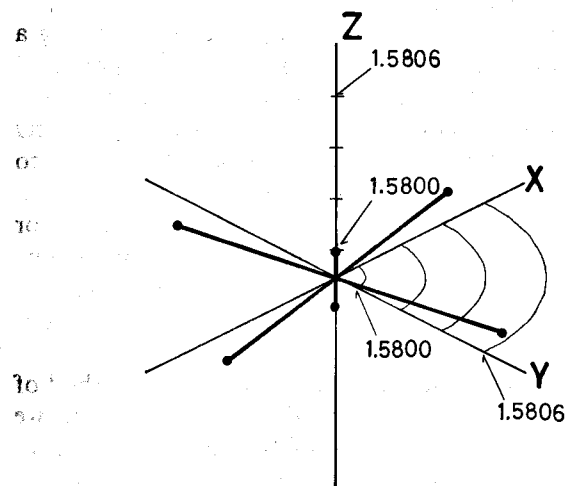


Fig. 5. Typical shape of refractive index ellipsoids (obtained at 45 mm), where the data are illustrated to emphasize the differences, and expressed by principal axes and principal values.

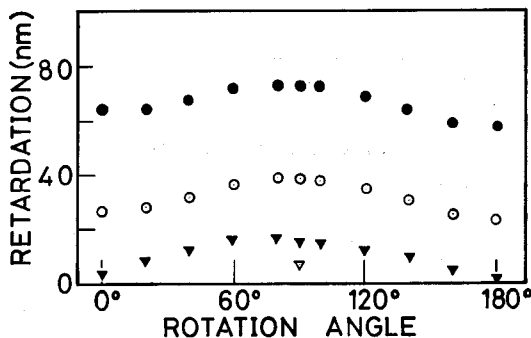


Fig. 3. Typical results of retardations obtained at 45 mm (single pass). Each symbol indicates the data at different incident angles ( $\nabla$   $0^\circ$ ;  $\blacktriangledown$   $10^\circ$ ;  $\circ$   $20^\circ$ ;  $\bullet$   $30^\circ$ ).

Table I. Differences of principal values in refractive index ellipsoids.

	Distance from center of disk (mm)			
	25	35	45	55
(as-molded)				
$\Delta n_{xz} \times 10^4$	5.3	5.7	5.7	5.8
$\Delta n_{xy} \times 10^4$	0.1	0.1	0.1	0.2
(annealed)				
$\Delta n_{xz} \times 10^4$	4.8	4.5	4.1	4.3
$\Delta n_{xy} \times 10^4$	0.3	0.3	0.2	0.1

\*The value of 1.579 was obtained for a hot-pressed film specimen by a prism coupler method as an average refractive index.

As origins of anisotropies of refractive indices in polymer solids, two factors are thought to be responsible: that is, molecular orientation and residual stress. For an as-molded disk substrate, both of these two factors must be involved. Therefore, by performing an annealing procedure on the as-molded sample, we tried to remove the influence of residual stress and evaluate the contribution of the molecular orientation.

In order to determine the annealing temperature, we first measured the temperature dependence of the dynamic modulus of this material. Figure 6 shows the elastic storage and loss moduli of a hot-pressed film sample measured at 3.5 Hz by Rheovibron DDV-II-EA (Toyo Baldwin). Rapid decrease of the storage modulus and the corresponding rapid increase of the loss modulus were observed around 125°C, suggesting that rather large scale molecular motions begin to occur at this temperature region. Hence, we employed 125°C as the annealing temperature.

Figure 7 shows the annealing time dependence of the retardation observed at 125°C (incident angle; 30°, rotation angle; 0°). For complete removal of the residual stress, an annealing time of 30 hours was suggested to be necessary. Thus, we employed the annealing condition of 30 hours at 125°C in the following experiments.

The results for samples received, such as the annealing procedure, revealed that shapes of refractive index ellip-

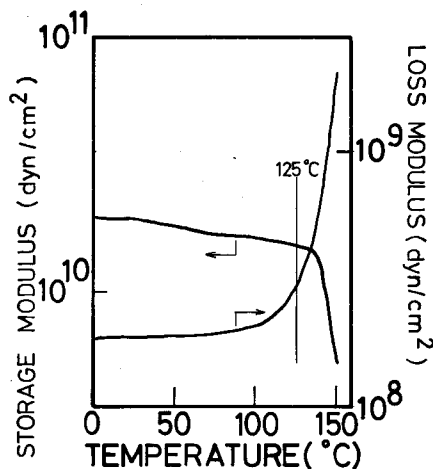


Fig. 6. Temperature dependence of dynamic storage and loss moduli of a hot-pressed film specimen measured at 3.5 Hz.

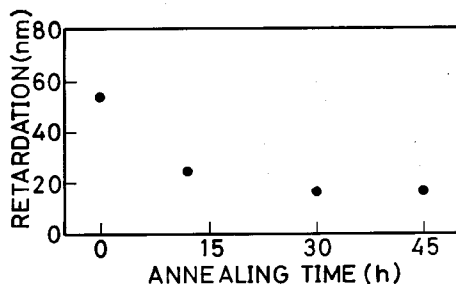


Fig. 7. Annealing time dependence of retardation (rotation angle 0° and incident angle 30°). For annealing times longer than 30 hours retardations fall at a constant value, suggesting that complete removal of the residual stress is achieved.

soids of the annealed disk substrate are still similar to those of as-molded disk substrates, and the principal value in the thickness direction is still substantially smaller than those in other directions. Numerical values of the differences of these principal values,  $\Delta n_{xz}$  and  $\Delta n_{xy}$ , are shown in Table I.

Figure 8 shows the directions of principal axes of refractive index ellipses in the XY-plane before and after annealing. After annealing, the directions of the principal axes lined up along the X and Y axes, as previously observed by Iwasawa and Funakoshi,<sup>8)</sup> suggesting that the molecular orientation of the disk substrate is similar to that for the sample biaxially elongated in the radial and tangential directions. This is supported further by the consideration of molten polymer flow in the mold cavity.<sup>3)</sup> Almost the same levels of molecular orientation in the radial and tangential directions result in very small vertical retardation, but relatively large oblique retardation occurs. It should be noted here that the degrees of orientation considered are very small in absolute value.

As seen above, the anisotropy of the refractive index, even in the as-molded disk substrate, is thought to originate mainly from the molecular orientation. Since the anisotropy of refractive indices in polymer materials caused by molecular orientations can be expressed as a product of the degree of orientation of the constructive units and their refractive index anisotropies, the decrease of either or both of these two factors should be attempted to reduce the total refractive anisotropy in PC disk substrate.

Although the suppression of the degree of orientation has been achieved by the development of injection molding techniques,<sup>1-3,9)</sup> the change of molecular weight distributions<sup>7,10)</sup> or the alteration of end groups in polymer chains,<sup>11)</sup> the suppression of the refractive index anisotropy of the disk substrate has already reached the maximum limitation in polycarbonate (PC).<sup>4-6)</sup> Therefore, to obtain a disk substrate with much lower refractive index anisotropy, i.e., to decrease the retardation for oblique rays, the use of a polymer which has a low refractive index anisotropy in constructive units is necessary.

However, contributions of residual stress to the refractive index anisotropy is estimated to be about 1/5 of the

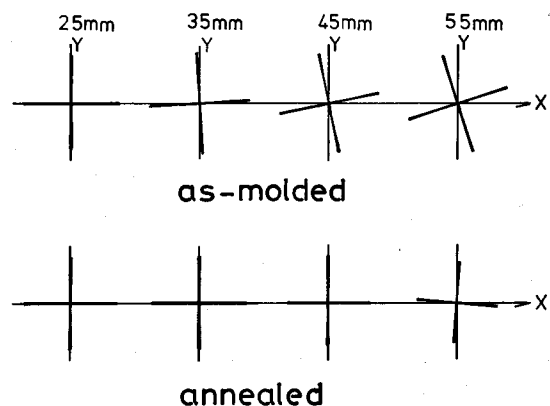


Fig. 8. Directions of the principal axes of refractive index ellipses on the XY plane obtained for as-molded and annealed samples.

total anisotropy by measuring and comparing the values for as-molded and annealed disk substrates, and as shown in Fig. 8 this contribution causes the change in the directions of the principal axes in the  $XY$ -plane. Moreover, the changes in the principal axis directions of birefringence along the tangential directions cause high-frequency noises in the MO readout. Hence this contribution must also be decreased. The anisotropy of the refractive index of the polymer solid caused by residual stress is expressed as the product of the stress and stress-optical coefficient of materials. Therefore, to decrease the contribution of the residual stress, the use of a polymer which has low stress-optical coefficient is also necessary.

As discussed above, future development of a modified polycarbonate which maintains the present advantages of polycarbonate and has a low anisotropy of the refractive index in the constructive units and a low stress-optical coefficient is expected.

#### §4. Conclusions

The shape of the refractive index ellipsoids of a polycarbonate MO disk substrate was muffin-shaped, that is, the values in the radial and tangential directions were almost the same, while that in the thickness direction was substantially smaller. From the results of the annealing experiment, it was suggested that these

anisotropies originated mainly from the molecular orientation. Therefore, a modified polycarbonate with a low anisotropy of the refractive index in the constructive units and a low stress-optical coefficient should be developed for a much higher performance MO memory disk substrate.

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