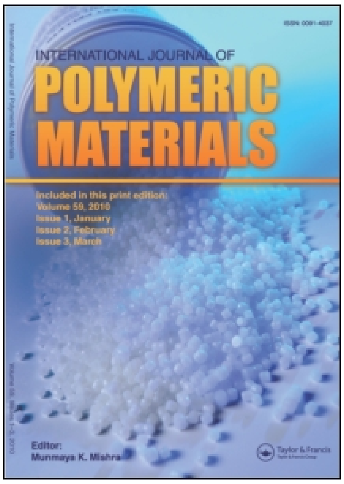

International Journal of Polymeric Materials
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713647664

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H. M. Shabana[a]; A. Abdul-jaleel[b]
[a] Applied Science Dept., College of Technological Studies, Shuwaikh, Kuwait [b] Kuwait Institute for Scientific Research, Safat, Kuwait

To cite this Article Shabana, H. M. and Abdul-jaleel, A.(2005) 'Optical Properties of Highly Birefringent Polymer Films Using Simple Techniques', International Journal of Polymeric Materials, 54: 11, 1009 — 1018
To link to this Article: DOI: 10.1080/009140390517722
URL: http://dx.doi.org/10.1080/009140390517722

International Journal of Polymeric Materials, 54:1009–1018, 2005

ISSN: 0091-4037 print/1563-5333 online
DOI: 10.1080/009140390517722

Optical Properties of Highly Birefringent Polymer Films Using Simple Techniques

H. M. Shabana
Applied Science Dept., College of Technological Studies, Shuwaikh, Kuwait

A. Abdul-jaleel
Kuwait Institute for Scientific Research, Safat, Kuwait

Two optical systems are used for characterizing free-standing highly birefringent polymer films. The first system is the Michelson interferometer and is utilized, in the presence of a polarizer, to determine the refractive indices in the two principal vibration directions along and across the axis of the sample, and the birefringence. The second system directly determines the birefringence. It is based on measurements of the optical path difference and the light intensity before and after striking the sample. The results show that the obtained values of birefringence, by the two methods, are in good agreement with each other. Certain optical parameters related to the measured values of refractive indices and birefringence are also calculated. The used methods are recommended for investigating thick and highly birefringent materials, where the conventional methods face certain problems such as the large number of interference fringe order and the highly refractive indices of the used immersion liquids.

Keywords: birefringent polymer films, interferometer, refractive indices, birefringence, optical properties

INTRODUCTION

It is very important to measure the optical properties of films in the fields of optics, optoelectronics, and microelectronics. The refractive indices, in the two orthogonal vibration directions, and the related optical parameters are important for the characterization of

Received 22 July 2004; in final form 26 July 2004.
The author thanks Prof. M. A. Mabrouk for a useful discussion.
Address correspondence to H. M. Shabana, Applied Science Dept., College of Technological Studies, PO Box 42325, Shuwaikh 70654, Kuwait. E-mail: hmshabana@hotmail.com

anisotropic polymer films. The methods of film fabrication and treatment help to a great extent in the variation of the obtained properties and optimizing materials of high industrial interest [1].

The problem of investigating refractive indices, birefringence, thickness, and certain other parameters was discussed in many publications using several experimental tools. Among the variety of these tools, interferometry has proved to be one of the most accurate and is widely used in both research and industry [2, 3].

There are many physical applications of the principles of interferometry. Some of these are only of historical significance, whereas others are now being used extensively [4]. All interferometric techniques are mainly used to measure precisely the optical path difference over the entire visible spectrum from which the thickness, refractive indices, birefringence, and other optical parameters can be determined. They also determine correctly the interference order in the image of the examined object. The highly coherent quasimonochromatic light obtained from the laser source has made it particularly easy to create new interferometric configurations. The Michelson interferometer is a widely used instrument for measuring the wavelengths of various light sources, for using the wavelength of a known source to measure extremely small distances, and for investigating optical media.

The values of thickness and refractive indices of polymer films have been determined by many authors [5–15]. Goodman [5] used IR spectrophotometers to estimate the film thickness and refractive index from the optical interference fringe patterns. Carreno et al. [6] used spectrophotometric method for the measurements of thickness and refractive index. The optical parameters of thin films by *p*-polarized laser beams were investigated by Liu et al. [7], whereas in analytical method was used by Zheng and Kikuchi [8] for the same purpose. Lloyd's interferometer was applied by Hamza et al. [9] to determine the same parameters of thin films with different thickness. Russo [10] presented an alternative method for the determination of birefringence in stretched polymeric films. Another attempt has been done by Nasr [11] to estimate the birefringence of thin photoactive polymer films. Medhat et al. [12] obtained fringes of equal tangential inclination by curved-induced birefringence. Recently, Sadik et al. used a general interferometric approach based on variable incident angle for refractive index and thickness measurement of birefringent single-medium [13] and multi-medium objects [14]. Shabana [15] determined the thickness and refractive index of isotropic polypropylene films using different interferometric techniques. Comparison between the obtained results was given.

In the present work an experimental set-up for the determination of refractive indices and birefringence of free-standing oriented polymer sheets is introduced. Another optical technique is also used to determine the birefringence directly. The optical principles and the mathematical formulae are derived. Certain optical parameters such as isotropic refractive index, polarizability per unit volume, and surface reflectivity are calculated. Comparison between the obtained values of birefringence by the two methods is also given.

THEORETICAL CONSIDERATIONS

The refractive index can be determined from the analysis of the obtained fringe patterns depending on the basic equations [5]

$$2n_f t_f \cos\varphi = m\lambda \tag{1a}$$

$$2n_f t_f \cos\varphi = (m + 1/2)\lambda \tag{1b}$$

Equations 1a and b, give the locations of the fringe maxima and minima, where n_f is the refractive index of the film, t_f is the film thickness, ϕ is the refraction, m is the fringe order, and λ is the wavelength of the used monochromatic light. According to Eq. 1 and the applied interferometric technique, one can estimate the required equations for the refractive indices of the films. In a Michelson interferometer it is necessary to slowly vary the length of the film through which the interference beam passes. A greater length of film is obtained as it is rotated. The change in path length is related to the measured fringe transmissions by the following equation:

$$2n_a d_a(\theta) + 2n_f t_f(\theta) = m\lambda \tag{2}$$

where m is the collected number of fringe transmissions, λ is the wavelength of light used, n_a and n_f are the refractive indices of air and film, respectively, $d_a(\theta)$ is the distance traveled in air, $t_f(\theta)$ is the film thickness, and θ is the angle of rotation of the film. Eq. 2 can be rearranged and then used together with some basic mathematics to calculate the refractive index of the investigated film as follows [16]:

$$n_f = \frac{(2t_f - m\lambda)(1 - \cos\theta)}{2t_f(1 - \cos\theta) - m\lambda} \tag{3}$$

Eq. 3 can be applied to determine the refractive indices $n_f^{||}$ and $n_f^{\perp}$ if a polarizer is introduced in the optical path between the laser source and the beam splitter. The collected number m changes according to the polarization direction, parallel or perpendicular to the axis of the examined film.

The direct determination of birefringence in this article is based on recording the intensity ratio I/I_o, where I and I_o are the intensity of light reaching the power meter and that just before striking the sample, respectively. When the polarized light strikes the sample, and the plane of polarization makes an angle of 45 with the axis of the sample, the electric vector can be resolved into parallel and perpendicular components. The two waves are passing through the sample with different velocities. As a result, a phase change in the emerging light is observed. This change of phase (δ) is directly proportional to the intensity of light I as follow

$$I/I_o = \sin^2(\delta/2) \tag{4}$$

The phase difference δ is related to the optical path difference, $t_f\left(n_f^{\|} - n_f^{\perp}\right)$, via the well-known relation

$$\delta = \frac{2\pi}{\lambda} t_f\left(n_f^{\|} - n_f^{\perp}\right) \tag{5}$$

Because the birefringence Δn_f is taken as the difference between $n_f^{\|}$ and $n_f^{\perp}$, it can then be given from the combination of Eqs. 4 and 5, as follows:

$$\Delta n_f = \frac{\lambda}{\pi t_f} \sin^{-1}(I/I_o)^{1/2} \tag{6}$$

The obtained values for $n_f^{\|}$ and $n_f^{\perp}$ are used for calculating the following optical parameters. For isotropic refractive index the following equation is used:

$$n_{iso} = \left(n_f^{\|} + 2n_f^{\perp}\right)/3 \tag{7}$$

The polarizability per unit volume, $P^{\|}$, is estimated from the relation

$$P^{\|} = \frac{3}{4\pi}\frac{\left(n_{\|}^2 - 1\right)}{\left(n_{\|}^2 + 2\right)} \tag{8}$$

And an analogous equation for $P^{\perp}$.

The surface reflectivity of a polymer for light at normal incidence can be estimated from Fresnel equations and knowledge of the mean refractive index $\bar{n}_f$ [1]. Thus, the percentage reflection R (in air) is given by

$$R = \left(\frac{n_f - 1}{n_f + 1}\right)^2 \times 100 \tag{9}$$

EXPERIMENTAL TECHNIQUES AND RESULTS

Experimental Techniques

Two optical systems are used in this study. Both of them are illuminated by a helium-neon laser source, and are described in what follows.

Set-Up of Michelson Interferometer

The well-known Michelson interferometer provides a simple configuration. It is schematically shown in Figure 1. For accurate optical alignment, the components should be carefully arranged. Prior to setting up the experiment, the laser is warmed up to eliminate any possible fringe or intensity variations. A laser source of wavelength 632.8 nm

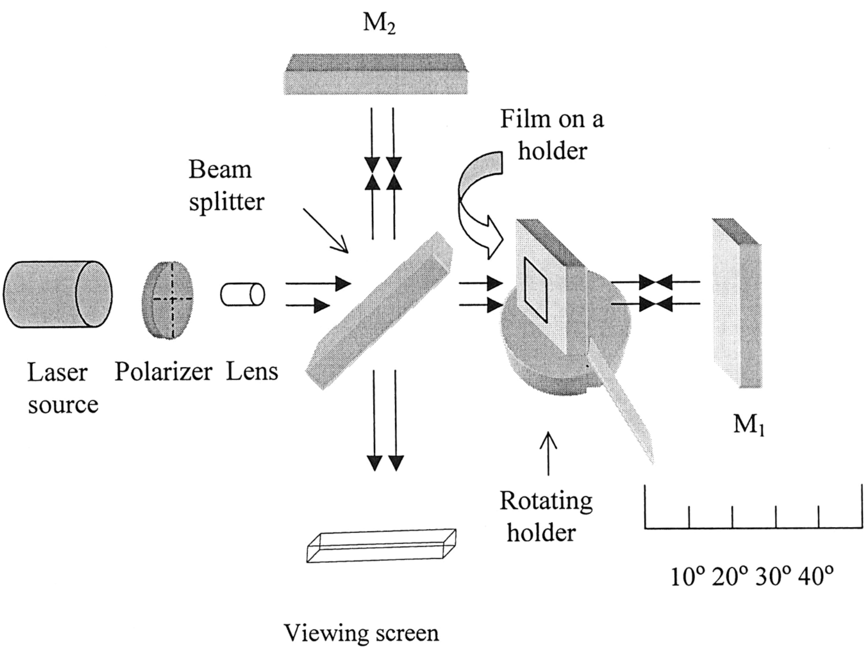

FIGURE 1 Schematic representation of the Michelson interferometer for determining refractive indices and birefringence of anisotropic polymers.

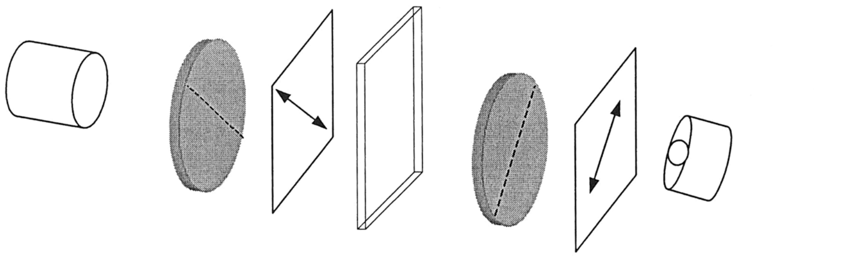

FIGURE 2 Optical set-up for direct measurement of birefringence.

is used and a polarizer is inserted in front of it to enable the determination of both refractive indices $n_f^{||}$ and $n_f^{\perp}$. The sample is then mounted, in the path of the transmitted beam, on a rotational component holder so that it is exactly perpendicular to the optical path. The light rays spread out when 18 mm convex lens is placed between the polarizer and the beam-splitter. The inter-fringe wave fronts have equal amplitude and frequency but they differ in phase. The counted number of fringe transmissions changes by rotating the holder.

Set-Up for a Direct Measurement of Birefringence

A schematic representation of this system is shown in Figure 2. The unpolarized light is incident on an ideal linear polarizer. This polarizer makes an angle of 45° with the incident beam of light. A second identical ideal polarizer, or analyzer, is introduced in opposite direction to the first. The irradiance I that reaches the detector equals zero. The value of I changes to positive reading when in anisotropic sample is introduced between the two polarizers, whereas the maximum irradiance I_o is taken, by the detector, just before striking the sample. By the knowledge of the wavelength of the used source of light and the sample thickness, the birefringence Δn_f can then be calculated.

EXPERIMENTAL RESULTS

Refractive Indices and Birefringence Measurements

The Michelson interferometer has been devised for estimating the refractive indices of free-standing polymer films. The method requires that the film under investigation must neither scatter nor absorb the light passing through it. Two different polymer sheets of different materials and having different thickness were used in this study. The two interfering beams of light are initially in phase because they split from the same initial laser beam. The optical path length of one of the interfering beams changes as a result of the rotation of the sample holder. As a result, the number of times the fringe pattern restored to its original state is counted. After introducing the sample in the light path, it is not necessary that the interference pattern be perfectly symmetrical or sharp. As long as the minima and maxima can be clearly distinguished accurate measurements can be made. It is also very important to make sure that the counted number of fringe transmissions is absolutely correct. This can be done by centering the interference pattern on a viewing screen and by selecting a reference line on a millimeter scale at the boundary between the bright and the dark fringes. When the wavelength and the thickness of the examined samples are known, the values of the refractive indices are calculated. The thickness t_f of each of the examined samples was measured using a digital micrometer (see Table 1). Each of the refractive indices $n_f^{\|}$ and $n_f^{\perp}$ for each sample was calculated, using Eq. 3, at different angles of rotation, and then the average was taken in order to minimize errors. Changing the polarization direction to the position normal to the sample axis, and by repeating the experiment at several angles of incidence, the refractive index $n_f^{\perp}$ of each sample was calculated. Examples of the obtained circular interference fringe patterns for

TABLE 1 Values of Thickness t_f, Angle of Rotation θ, Collected Number of Minima m, and Refractive Indices $n_f^{\|}$ and $n_f^{\perp}$

		Light vibrating parallel to the sample axis			Light vibrating perpendicular to the sample axis		
Sample	t_f (μm)	θ	m	$n_f^{\|}$	θ	m	$n_f^{\perp}$
1	191	10.7	4	1.6100	13.2	5	1.5513
		15	8	1.6147	15.5	8	1.5524
		16.8	10	1.6092	18.1	11	1.5539
		19.8	14	1.6073	21.7	16	1.5551
		$n_f^{\|}$ (average) = 1.6103			$n_f^{\perp}$ (average) = 1.5532		
2	188	13.5	6	1.5600	13.9	6	1.5108
		15.6	8	1.5548	16	8	1.5121
		18.2	11	1.5585	17.9	10	1.5070
		21.8	16	1.5609	22.5	16	1.5057
		$n_f^{\|}$ (average) = 1.5585			$n_f^{\perp}$ (average) = 1.5089		

TABLE 2 Values of Refractive Index Difference $n_f^{||} - n_f^{\perp}$ and birefringence Δn

Sample	Michelson method $(n_f^{||} - n_f^{\perp}) \times 10^{-3}$	The direct method $\Delta n \times 10^{-3}$
1	57.1	58
2	49.6	48.9

samples 1 and 2, respectively, were submitted to the editor but are not shown here.

Table 1 summarizes the values of the angle of rotation θ, the collected number of minima m and the corresponding values of refractive indices $n_f^{||}$ and $n_f^{\perp}$. The value of birefringence for each of the examined samples was taken as the difference between $n_f^{||}$ and $n_f^{\perp}$ and was given in Table 2.

This method of interference overcomes the problem of thick and highly birefringent materials where they can simply be investigated. Moreover, the samples have been investigated in air and with no need for immersion liquids.

Direct Measurements of Birefringence

The optical arrangement shown in Figure 2 was used for the direct determination of birefringence. The power meter is used to measure the intensities of light I and I_o. Several readings were recorded and their average was taken for greater accuracy. The birefringence Δn_f of each sample was then calculated using Eq. 6, and their values are given in Table 2. In comparison, the obtained values of birefringence, using both methods, are nearly equals. The difference between them may arise from the difficulty of investigating the same restricted area of the sample.

The values of the refractive indices $n_f^{||}$ and $n_f^{\perp}$ were utilized for calculating important optical parameters for the characterization of the polymer sheets. The isotropic refractive index n_{iso}, the polarizabilities

TABLE 3 Values of Isotropic Refractive Index n_{iso}, Polarizabilities per Unit Volume $P^{||}$, $P^{\perp}$, and Surface Reflectivity R

Sample	n_{iso}	$P^{||} \times 10^{-3}$	$P^{\perp} \times 10^{-3}$	R%
1	1.5722	82.79	76.41	5.08
2	1.5254	77.01	71.26	4.44

per unit volume $P^{\parallel}$ and $P^{\perp}$ and the surface reflectivity R were calculated using Eqs. 7, 8, and 9, respectively. The results of these optical parameters are given in Table 3.

CONCLUSION

The used optical systems have been employed in the characterization of oriented polymeric films via the determination of their optical properties. A Michelson interferometer was utilized, in the presence of a polarizer, to determine the refractive indices and birefringence of the examined samples. The arrangement of this system enables the determination of the optical parameters, where they can hardly be investigated by the conventional microscopic interferometers. Moreover, this method has the advantage of investigating thick specimens (>100 μm) and highly birefringent materials.

Another system was used to determine the birefringence directly. The used laser source has the advantage of getting accurate measurements because a restricted area of the sample can be selected for investigation. The difference between the obtained values of birefringence by the two used methods may arise from the difficulty of investigating the same restricted area of the sample.

As a result, the used optical systems have been established as making an important contribution to the investigation of the optical properties of oriented polymer sheets. They can, therefore, be considered as important technological techniques in the field of polymer science.

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