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Determination of Polyolefin Film Properties from Refractive Index Measurements. II. Birefringence

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Synopsis

The use of refractive index data for studying changes in oriented polyolefin films has been demonstrated. Application of this technique to oriented polypropylene and polyethylene films showed that the birefringence is a linear function of the refractive index in the direction of maximum orientation. By utilizing refractive index, density, and crystallinity data the crystalline refractive indices n_c and $(n_a + n_b)/2$ and polarizabilities $\alpha_{\|}$ and $\alpha_{\perp}$ could be estimated for polypropylene and polyethylene.

INTRODUCTION

When plastic films are oriented, the deformation of the film is frequently characterized in terms of a change in birefringence.[1-7] Three birefringence values can be defined, depending on the direction of light rays passing through the film. If the light is normal to the film plane, the birefringence will be given by $(n_x - n_y)$, where n_x is the refractive index parallel to the direction of orientation in the plane of the film, and n_y is the index perpendicular to the direction of orientation in the plane of the film. If the light is parallel to the plane of the film, two birefringence values, $n_y - n_z$ and $n_x - n_z$, can be measured, the term n_z being the refractive index of the film along the normal to the film plane. In this paper we shall be primarily concerned with $n_x - n_y$, since this is the birefringence most frequently measured in the film industry.

Uniaxial orientation results in an increase of birefringence, or difference between the refractive indices parallel (n_x) and perpendicular (n_y) to the direction of orientation. With balanced biaxial orientation there is no increase in the difference $n_x - n_y$, although both n_x and n_y increase during orientation. It is a general practice to measure the birefringence directly[8] by determining the retardation of light passing through a film. In addition to being time-consuming, this procedure gives no information concerning the change of refractive index in the film being studied.

In an earlier paper[9] a technique was described for rapidly and accurately determining n_x, n_y, and n_z, the refractive indices along the major axis of

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polymer films. From these data it was possible to determine three birefringence values ($n_x - n_y$, $n_x - n_z$, and $n_y - n_z$), the film density, and crystallinity, if one assumed the additivity of crystalline and amorphous specific volumes.[10,11] It is the purpose of this paper to further demonstrate the utility of applying refractometric techniques to the study of polymer film properties by describing the results of an investigation of the effect of orientation on the refractive index of polypropylene and polyethylene films and the relation of birefringence to refractive index in these films.

EXPERIMENTAL

Preparation of Film Samples

Oriented films having different draw ratios were prepared by uniaxially and biaxially orienting 10-mil polypropylene sheet at 300°F. on a T. M. Long film stretcher. The biaxial orientation was done simultaneously, sequentially, and by a combination of simultaneous and sequential drawing. Films prepared at 300°F. had average densities of 0.9053–0.9069 g./cm.[3] Oriented polypropylene films of lower density were prepared by uniaxially orienting quenched 1-mil film at room temperature and then annealing at 200°F. for periods of up to 20 min., to increase the density to a desired level.

Oriented polyethylene films were prepared by uniaxially orienting a 1-mil low-density polyethylene film at room temperature.

Measurement of Refractive Indices

The refractive indices n_x, n_y, and n_z along the major axes of the films were measured using a Bausch and Lomb Abbe-32 refractometer.[9] The n_x value corresponds to the machine direction (M.D.), or direction of maximum orientation, the n_y value corresponds to the transverse direction (T.D.), or refractive index perpendicular to n_x, and the n_z value corresponds to the refractive index in a direction normal to the plane of the film.

To measure n_x and n_z the film sample was mounted on the refracting prism with the x axis (M.D.) perpendicular to the long axis of the refracting prism. A polarizing eyepiece was attached to the refractometer and positioned so that the privileged direction (P.D.) of the polarizer was parallel to the line separating the fields of the refractometer. The refractive index measured was n_x. The polarizer was then rotated 90°, so that the P.D. was perpendicular to the field line; the refractive index measured was n_z. After the determination of n_x and n_z the film sample was remounted on the refracting prism with its machine direction parallel to the long axis of the prism. With the P.D. of the polarizer parallel to the field line the refractive index measured was n_y, and with the P.D. perpendicular to the field line the refractive index n_z was measured again. The birefringence $n_x - n_y$ was the difference between the refractive indices n_x and n_y. In a similar manner the birefringences $n_y - n_z$ and $n_x - n_z$ could be determined by using the refractive indices n_x, n_y, and n_z. Stein[12] has described a procedure for measuring $n_y - n_z$ and $n_x - n_z$ by measuring the retardation of light passing through tilted film samples. The procedure described above eliminates the need for special tilting apparatus and allows more rapid measurement of $n_y - n_z$ and $n_x - n_z$.

Experiments with Abbe refractometers made by different manufacturers indicated that the sharpness of the line between the dark and light portions of the refractometer field is dependent on the type of instrument and method of mounting of the specimen in each instrument. On the Bausch and Lomb Abbe-3L refractometer the sharpest line was obtained with transmitted light, when the film sample was attached to the refracting prism without a contacting liquid, or for films having rough surfaces with only a trace of contacting liquid. Sufficient contacting liquid was then placed between the illuminating prism and sample, to give good light transmission. Any inert liquid having a higher index of refraction than the film sample can be used as the contact liquid. Methyl salicylate was used for this work. The sharpness of the line with the above-described mounting technique was equivalent to the sharpness obtained with a liquid sample.

In addition to being applicable to one-component polymer films, the refractive index technique can sometimes be effectively applied to the study of two-component film laminates. It has been found that for some laminates the critical lines for each layer are visible in the refractometer field, allowing the refractive indices and densities of each layer to be determined simultaneously. Studies of this refraction characteristic showed that observation of two critical lines is most likely when the refractive indices and proportions of the layers are similar. When the intensity of the critical line produced by the low refractive index layer is high, the critical line of the high refractive index layer will be obscured, to the point of not being visible. Such was the case in measurements of the refractive indices (Appendix Table IV) of a polypropylene–polyethylene laminate used in this work. Because of the large refractive index difference between the two oriented layers only the critical line for the polypropylene was visible. The establishment of two critical lines for studying this effect is readily accomplished in the laboratory by mounting single-component polypropylene and polyethylene films together on the refractometer prism.

Density

The densities of the films were calculated from the average refractive index,

$$\bar{n} = {}^1/_3(n_x + n_y + n_z) \tag{1}$$

by using

$$\rho = (\bar{n} - 0.9374)/0.6273 \tag{2}$$

which is the equation giving the polypropylene and polyethylene density as a function of refractive index.[9] The refractive index data measured on the films used in this work are given in Appendix Tables I–IV.

Appendix Table I
Refractive Index Data for Polypropylene Oriented at 300°F.

Type of orientation[a]	n_x	n_y	n_z	Δ	$\bar{n}$	Elongation, % X	Elongation, % Y
S	1.5095	1.5089	1.4980	0.0006	1.5055	472	464
S, SE	1.5096	1.5069	1.4989	0.0027	1.5051	432	356
S	1.5112	1.5058	1.4989	0.0054	1.5053	376	304
S, SE	1.5127	1.5048	1.4976	0.0079	1.5050	588	400
S	1.5127	1.5044	1.4991	0.0083	1.5054	404	220
S	1.5137	1.5052	1.4975	0.0085	1.5054	540	384
SE	1.5132	1.5044	1.4986	0.0088	1.5054	404	200
S, SE	1.5150	1.5035	1.4979	0.0115	1.5055	556	308
U	1.5129	1.5014	1.5019	0.0115	1.5053	292	—
S	1.5154	1.5031	1.4979	0.0123	1.5051	548	292
SE	1.5169	1.5018	1.4969	0.0151	1.5053	604	368
S	1.5084	1.5082	1.4975	0.0002	1.5047	—	—
SE	1.5098	1.5076	1.4974	0.0032	1.5049	404	340
S	1.5106	1.5064	1.4969	0.0042	1.5046	528	472
S	1.5114	1.5049	1.4970	0.0065	1.5044	480	360
SE	1.5132	1.5039	1.4971	0.0093	1.5047	384	333
S	1.5159	1.5007	1.4975	0.0152	1.5047	492	204
S	1.5181	1.4996	1.4968	0.0185	1.5048	580	216
U	1.5189	1.5001	1.5013	0.0188	1.5048	512	—
S, SE	1.5096	1.5065	1.4960	0.0031	1.5040	560	496
SE	1.5141	1.5016	1.4953	0.0125	1.5037	524	344
SE	1.5150	1.5020	1.4959	0.0130	1.5043	548	420
SE	1.5164	1.5007	1.4955	0.0157	1.5042	580	268
S	1.5081	1.5064	1.5039	0.0017	1.5061	—	—
S	1.5098	1.5080	1.5010	0.0018	1.5063	—	—
S	1.5100	1.5055	1.5030	0.0045	1.5062	—	—
U	1.5100	1.5057	1.5041	0.0043	1.5066	—	—
U	1.5111	1.5039	1.5031	0.0072	1.5060	—	—
U	1.5156	1.5023	1.5019	0.0133	1.5066	—	—
U	1.5134	1.5039	1.5032	0.0095	1.5068	—	—
U	1.5146	1.5021	1.5020	0.0125	1.5062	—	—
U	1.5195	1.4998	1.4987	0.0197	1.5060	—	—
S	1.5107	1.5050	1.5035	0.0057	1.5064	288	184
U	1.5110	1.5036	1.5046	0.0074	1.5064	224	—
U	1.5232	1.4976	1.4978	0.0256	1.5062	528	—

[a] *U* = uniaxial; S = simultaneous; SE = sequential; S, SE = simultaneous followed by sequential.

RESULTS AND DISCUSSION

One of the major advantages of characterizing oriented films in terms both of the refractive indices n_x, n_y, and n_z, and of birefringence is the improved differentiation of orientation effects in the films. Since different stretching histories frequently result in the same birefringence, knowledge of the refractive indices yields additional data that help distinguish among

Appendix Table II

Refractive Index Data for Polypropylene Oriented at Room Temperature

Type of orientation	n_x	n_y	n_z	Δ	$\bar{n}$
U	1.5056	1.4844	1.4829	0.0212	1.4909
U	1.5071	1.4835	1.4830	0.0236	1.4912
U	1.5076	1.4837	1.4805	0.0239	1.4906
U	1.5079	1.4825	1.4815	0.0254	1.4906
U	1.5080	1.4824	1.4817	0.0256	1.4907
U	1.5085	1.4826	1.4815	0.0259	1.4908
U	1.5083	1.4825	1.4815	0.0258	1.4908
U	1.5086	1.4823	1.4810	0.0263	1.4906
U	1.5100	1.4815	1.4805	0.0285	1.4907
U	1.5071	1.4865	1.4860	0.0206	1.4932
U	1.5084	1.4856	1.4851	0.0228	1.4930
U	1.5088	1.4858	1.4853	0.0230	1.4933
U	1.5125	1.4833	1.4836	0.0292	1.4931
U	1.5144	1.4910	1.4902	0.0234	1.4985
U	1.5154	1.4905	1.4892	0.0249	1.4984
U	1.5148	1.4897	1.4897	0.0251	1.4980
U	1.5128	1.4895	1.4895	0.0233	1.4972
U	1.5138	1.4892	1.4885	0.0246	1.4972

Appendix Table III

Refractive Index Data for Polyethylene Uniaxially Oriented at Room Temperature

n_x	n_y	n_z	Δ	$\bar{n}$	Elongation, %
1.5182	1.5176	1.5147	0.0006	1.5168	0
1.5187	1.5160	1.5130	0.0027	1.5159	—
1.5190	1.5154	1.5135	0.0036	1.5159	—
1.5202	1.5146	1.5124	0.0056	1.5154	12.8
1.5229	1.5125	1.5105	0.0104	1.5153	24.0
1.5238	1.5114	1.5093	0.0124	1.5148	—
1.5251	1.5111	1.5092	0.0140	1.5151	—
1.5258	1.5107	1.5092	0.0151	1.5152	32.0
1.5266	1.5107	1.5085	0.0159	1.5152	—
1.5275	1.5094	1.5079	0.0181	1.5149	46.4
1.5320	1.5080	1.5057	0.0240	1.5152	—
1.5314	1.5072	1.5054	0.0242	1.5147	78.4
1.5343	1.5065	1.5047	0.0278	1.5152	—
1.5353	1.5054	1.5039	0.0299	1.5149	—
1.5374	1.5042	1.5029	0.0332	1.5148	193.6
1.5408	1.5023	1.5017	0.0385	1.5148	276.8
1.5345	1.5061	1.5044	0.0304	1.5150	123.2
1.5386	1.5030	1.5020	0.0356	1.5148	220.8
1.5448	1.5010	1.5000	0.0438	1.5152	412.8

Appendix Table IV

Refractive Index Data for Polypropylene Layer of Polypropylene–Polyethylene Laminate Uniaxially Oriented at Room Temperature

n_x	n_y	n_z	Δ	$\bar{n}$	Elongation, %
1.4933	1.4919	1.4915	0.0014	1.4922	8.2
1.4938	1.4904	1.4903	0.0034	1.4915	19.7
1.4945	1.4902	1.4894	0.0043	1.4914	34.4
1.4956	1.4895	1.4892	0.0061	1.4914	40.9
1.4961	1.4885	1.4885	0.0076	1.4910	55.8
1.4987	1.4868	1.4863	0.0119	1.4906	83.6
1.5018	1.4855	1.4855	0.0163	1.4909	113
1.5024	1.4850	1.4850	0.0174	1.4908	129
1.5046	1.4841	1.4840	0.0205	1.4909	162
1.5045	1.4833	1.4833	0.0212	1.4904	179
1.5079	1.4817	1.4817	0.0262	1.4904	261
1.5085	1.4810	1.4810	0.0275	1.4902	275
1.5107	1.4812	1.4812	0.0295	1.4910	343

oriented films. Examination of the data in Table I shows the type of differences that can be obtained. Comparing the first two films, it can be seen that, although both the uniaxially and biaxially oriented samples had a birefringence of 0.0115, the refractive indices of the two films were different. The second group of films, which were biaxially oriented, both had similar birefringence values, of 0.0123 and 0.0125, but again had different refractive indices.

TABLE I

Comparison of the Properties of Films Having the Same Birefringence

Type of orientation	Elongation, %		Birefringence	Refractive index			Density, g./cm.³
	X axis	Y axis		n_x	n_y	n_z	
Uniaxial: 1 × 3	292	−8.0	0.0115	1.5129	1.5014	1.5019	0.9053
Biaxial: simultaneous (3 × 3) + sequential to ×6 in one direction	556	308	0.0115	1.5150	1.5035	1.4979	0.9056
Biaxial: simultaneous (3 × 6)	548	292	0.0123	1.5154	1.5031	1.4979	0.9050
Biaxial: sequential (2 × 5)	524	344	0.0125	1.5141	1.5016	1.4953	0.9028

Additional information can be obtained by using eq. (2) to convert the average refractive index of the film to a density value. From the data in Table I it can be seen that various density–birefringence combinations can be obtained for films having different stretching histories. The first two films had similar density and birefringence values, while the second two films had the same birefringence but different densities.

Determination of the birefringence from refractive index data, instead of by direct measurement, allows one to determine directly $n_x - n_y$, $n_x - n_z$, and $n_y - n_z$. That good results can be obtained from the refractive index measurement is indicated by the data plotted in Figure 1. The birefringence, which was determined from refractive indices, is plotted as a function of elongation. It can be seen that the polyethylene curve was in excellent agreement with the data of Stein and Norris,[8] who measured the birefringence directly.

It is apparent, therefore, that by measuring the refractive indices of oriented films sufficient data are also being generated for determining the birefringence ($n_x - n_y$, $n_x - n_z$, and $n_y - n_z$) and density of the film. Collection of these data thus affords the option of characterizing oriented films in

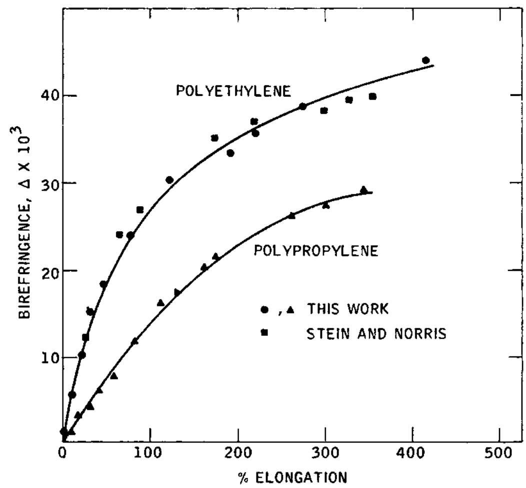

Fig. 1. Increase of birefringence with increasing elongation.

terms of three birefringence values, three refractive indices, and the film density.

Birefringence and Refractive Index in Polypropylene

When the birefringence of oriented polypropylene films is plotted as a function of n_x, the refractive index in the machine direction, or direction of major orientation, a set of parallel straight lines is obtained. All the points falling on a given line correspond to films having the same density and average refractive index. This result is shown in Figure 2. The data forming the line $\bar{n}$ = 1.5053–1.5063 were obtained from polypropylene films oriented uniaxially and biaxially at elevated temperatures. The data forming the other lines of Figure 2 were obtained from uniaxially cold-drawn polypropylene films. Because of the neckdown tendency of polypropylene it was not possible to obtain birefringence values lower than 0.020 from single-component polypropylene films. The low birefringence values shown on the line $\bar{n} = 1.4909$ of Figure 2 and plotted in Figure 1 were obtained by measuring the refractive index of the polypropylene layer of a uniaxially oriented

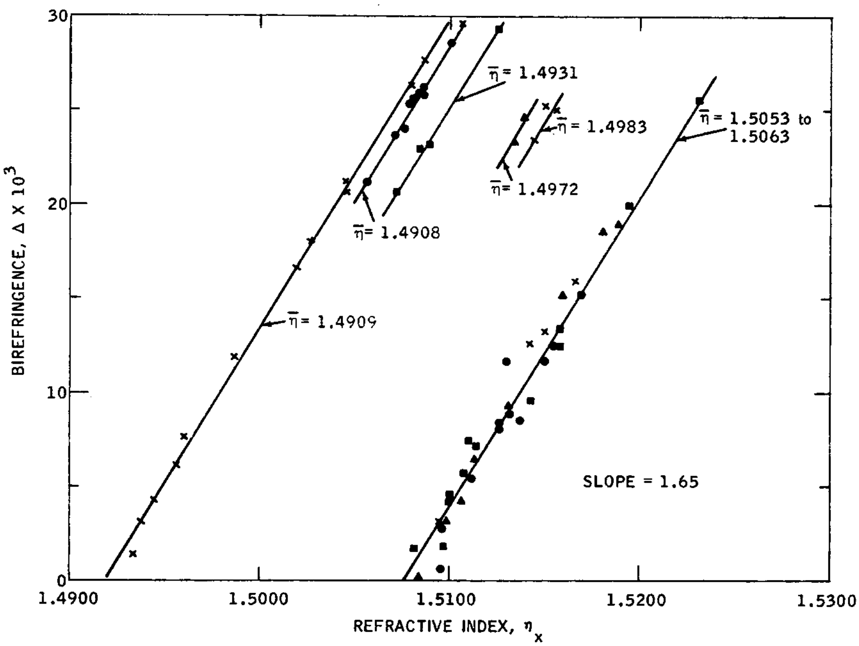

Fig. 2. Relation between birefringence and refractive index n_x in oriented polypropylene.

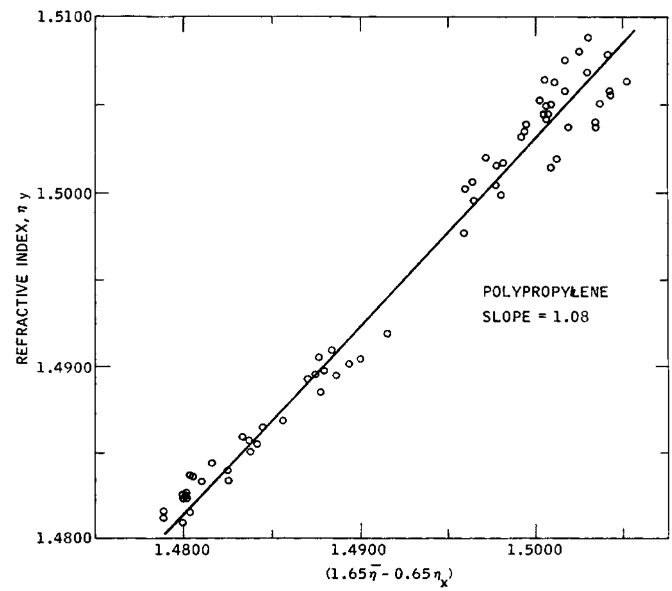

Fig. 3. Change of refractive index n_y as a function of $\bar{n}$ and n_x in oriented polypropylene.

polypropylene–polyethylene laminate. Orientation of the laminate gave enough support to the polypropylene to eliminate neckdown.

From Figure 2 it is seen that

$$\Delta = n_x - n_y = Kn_x + B \tag{3}$$

When $\Delta = 0$, then $B = -Kn_0$, giving the ordinate intercept in terms of K the slope of the lines in Figure 2 and the abscissa intercept n_0 at $\Delta = 0$. Equation (3) is used in the form

$$\Delta = n_x - n_y = Kn_x - Kn_0 \tag{4}$$

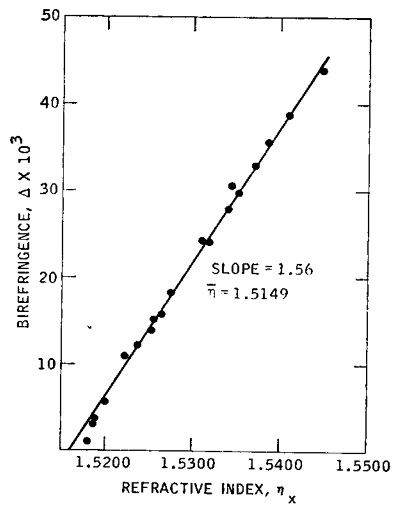

Fig. 4. Relation between birefringence and refractive index n_x in uniaxially oriented polyethylene.

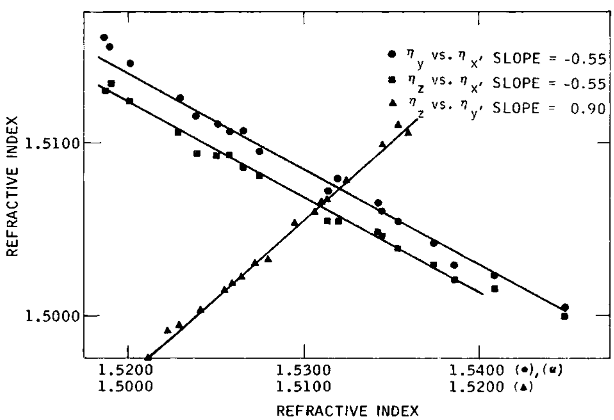

Fig. 5. Relation between n_x, n_y, and n_z in uniaxially oriented polyethylene.

The term n_0 is equal to n, the average refractive index of the film, plus a constant c representing a small positive displacement of the intercept. The average value of c was 0.0017 for polypropylene and 0.0010 for polyethylene.

Because Δ is a linear function of n_x, the refractive index n_y will also be linearly dependent on n_x. Rearranging eq. (4) and substituting K values from Figure 2 yields an equation which gives the refractive index n_y in terms of $\bar{n}_x$, $\bar{n}$, and c:

$$n_y = -0.65n_x + 1.65(\bar{n} + c) \qquad (5)$$

A plot of the measured n_y as a function of n_x and $\bar{n}$ should yield a single straight line, which includes all of the n_y data irrespective of polymer density. In Figure 3, which is a graph of n_y as a function of $(1.65\bar{n} - 0.65n_x)$, it can be seen that the data (n_y of Appendix Tables I, II, and IV) do fall close to a single line.

In Figure 4 are plotted data showing that the birefringence and refractive index in polyethylene are also linearly related. From these data it is found that

$$n_y = -0.56n_x + 1.56(\bar{n} + c) \qquad (5a)$$

The relations among the various refractive indices (Appendix Table III) in polyethylene are graphed in Figure 5.

Refractive Indices in Polypropylene Crystal

By utilizing eqs. (4), (5), and (5a) relating Δ, n_x, and n_y it is possible to estimate both the refractive index $n_{c,x}$ parallel to the c axis of the polypropylene crystal and the average of the refractive indices along the a and b crystal axes, i.e. $(n_a + n_b)/2$, which was assumed to be equal to $(n_y + n_z)/2$.

By replacing Kn_0 [eq. (4)] with the intercept value for crystalline polypropylene an equation relating the birefringence and refractive index n_x in the polymer crystal is obtained. Conversion of the crystalline density, 0.9360 g./cm.[3,13,14] into the average refractive index [eq. (2)] gave a value of Kn_0 equal to (1.65)(1.5245 − 0.0017). Equation (4) then has the form

$$\Delta_c = 1.65n_x - 2.5182 \qquad (6)$$

for the crystalline polymer.

When $\Delta_c = \Delta_c^0$ where Δ_c^0 is the crystalline birefringence, the refractive index n_x will equal n_c, the crystalline refractive index along the c axis of the crystal. Substituting in eq. (6) the value $\Delta_c^0 = 0.033$, reported by Samuels,[15] one obtains 1.546 as the refractive index along the c axis of the polypropylene crystal. Solution of eq. (5) with $n_{c,x} = 1.546$ and $n = 1.5254$ gave a refractive index of 1.513 for n_y. From eq. (1) a value of 1.514 was obtained for n_z. In Table II are listed polypropylene crystalline refractive indices as well as polarizabilities, which were calculated by substituting $n_{c,x}$ and $(n_y + n_z)/2$ in the Lorentz-Lorenz equation. It can be seen that the polarizability difference reported in this paper agrees well with values reported by Samuels[15] and Tsvetkov.[16]

TABLE II
Refractive Indices and Polarizabilities of Polypropylene and Polyethylene Crystals

Polymer	Assumed cryst. dens., g./cm.^{3}	Refractive index of crystal		Polarizability per cm.^{3}		
		$n_{c,x}$	$\left(\frac{n_a + n_b}{2}\right)$	$(\alpha_{\parallel})(\alpha_{c,x})$	$\left(\frac{\alpha_a + \alpha_b}{2}\right)$	$\alpha_{\parallel} - \alpha_{\perp}$
Polypropylene						
This work	0.936[b]	1.546	1.514	0.0756	0.0718	0.0038
Tsvetkov[16]	—	—	—	—	—	0.0035
Samuels[15]	0.936	—	—	—	—	—
Polyethylene						
This work	0.954[c]	1.573	1.517	0.0786	0.0722	0.0064
	1.00[b]	1.602	1.546	0.0819	0.0756	0.0063
Bunn and de Daubeny[17]	0.954	1.575[e]	1.517[e]	—	—	—
Stein and Norris[8]	0.954	—	—	0.0790[f]	0.0723	0.0067

[a] Assumed to be equal to $(n_y + n_z)/2$.
[b] Natta et al.[13,14]
[c] Bunn and de Daubeny.[17]
[d] Nichols.[18]
[e] Values reported for $C_{36}H_{74}$ by Bunn and de Dauveny.[17]
[f] Calculated by Stein and Norris, using refractive index data of Bunn and de Daubeny.[17]

Refractive Indices in the Polyethylene Crystal

Since the total birefringence of polyethylene is also a linear function of the refractive index n_x (see Fig. 4), the crystalline refractive indices and polarizabilities can be determined for this polymer in the same manner as that used for polypropylene.

From the equation reported by Stein and Norris,[8]

$$\Delta_c = 0.0286(3 \cos^2 \epsilon - 1)$$

a value of $\Delta_c = \Delta_c^0 = 0.0576$ is obtained when $\cos^2 \epsilon = 1$, that is, when the crystal c axis is oriented parallel to the direction of orientation.

The refractive index $n_{c,x}$ was then determined from eq. (4) with $\Delta = 0.0572$ and $K = 1.56$ and the intercept Kn_0 determined with $c = 0.0010$. Two values of $n_{c,x}$ were calculated from assumed crystal densities of 0.954 g./cm.^{3} ($\bar{n} = 1.5360$), given by Bunn and de Daubeny;[17] and 1.00 g./cm.^{3} ($\bar{n} = 1.5647$), reported by Nichols.[18] The refractive index n_y was calculated from eq. (5a). The value of n_z was again obtained with eq. (1). The resulting polyethylene refractive indices $n_{c,x}$ and $(n_a + n_b)/2$ and also polarizability data determined by substituting the refractive indices in the Lorentz-Lorenz equation are shown in Table II. It can be seen that these data, which were obtained by extrapolating birefringence measurements, are in good agreement with similar data reported in the literature when a polyethylene crystalline density of 0.954 g./cm.^{3} is used.[17] When the crystalline density was taken as 1.00 g./cm.^{3},[18] the calculated refractive index and polarizability values were greater than the reported literature data.

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