Determination of the Birefringence of an Ideal Polyester Fiber (PET) with Regard to the Intermolecular Interactions

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Synopsis

Method of birefringence determination for an ideal polyester fiber has been described with regard to the interactions of surrounding macromolecules, $\Delta n = 0.2360$. Obtained results have been discussed in view of information given in the literature.

INTRODUCTION

The birefringence of ideal fibers was often determined by means of the theoretical method,[1] which is based on the Lorentz–Lorenz equation.[2] This equation connects the mean refractive index of material with the electric polarizability of its molecules. The birefringence value calculated in this way corresponds to birefringence of a fictitious polymeric monocrystal in which no interactions take place between neighboring macromolecular chains.

In Ref. 3 a new approach to calculation of birefringence of an ideal fiber has been presented. This new method is also based on the Lorentz–Lorenz equation; however, it takes into consideration the dipole interactions between neighboring macromolecules and the type of crystal structure of the fiber. In this paper the birefringence of an ideal PET fiber was calculated by means of the mentioned above method.

DESCRIPTION OF THE METHOD

A general assumption undertaken in the theoretical method used in many articles[1] is that for polymer crystals the Lorentz–Lorenz equation is valid:

$$(n^2 - 1)/(n^2 + 2) = \tfrac{4}{3}\pi N \alpha \tag{1}$$

where N = number of molecules in a volume unit, α = polarizability of a molecule, and n = refractive index. Such an assumption is incorrect because the Lorentz–Lorenz equation was derived only for some specific types of crystal structures. The introductory considerations which led Lorentz to eq. (1) were calculations of an electric field influencing a molecule of a dielectric medium. To solve this problem, Lorentz cut out a hypothetical spherical cavity around the considered molecules—called the Lorentz sphere.[2]

The local electric field in the place of considered molecule can be described as follows:

$$\mathbf{E}_{\mathrm{loc}} = \mathbf{E} + \mathbf{E}_k + \mathbf{E}_w \qquad (2)$$

Here $\mathbf{E}_k$ = the field which originates from polarizational charges induced on the inner surface of the spherical cavity. Lorentz has demonstrated that $\mathbf{E}_k = \frac{4}{3}\pi\mathbf{P}$, where $\mathbf{P}$ is the polarization vector of the dielectric. $\mathbf{E}_w$ = electric field influencing the molecule from the neighborhood's side which are present in the Lorentz spherical cavity. This field is essentially dependent on the type of the crystal structure of the dielectric. $\mathbf{E}$ = macroscopic field in the dielectric volume. Lorentz himself employed only with structures of the regular or spherical symmetry. In such a structures $\mathbf{E}_w = 0$ because of the compensation of the neighbor interactions on the considered molecule. Thus

$$\mathbf{E}_{\mathrm{loc}} = \mathbf{E} + \mathbf{E}_k = \mathbf{E} + \frac{4}{3}\pi\mathbf{P} \qquad (2')$$

This relationship is the foundation for further considerations, which lead to the eq. (1).

Most of the fiber-forming polymers crystallize in crystallographic systems that differ considerably from the regular or spherical systems. The application of Lorentz–Lorenz equation to calculations of birefringence of polymers would be possible only after its modification regarding the specific crystal structure. It is connected with the necessity of formulating the expression (2) for the local field $\mathbf{E}_{\mathrm{loc}}$ with regard to $\mathbf{E}_w$—the field characteristic of the specific type of fiber. To determine the field $\mathbf{E}_w$, a simplified model of the macromolecule chain was taken. The following assumptions were made:

(1) The macromolecule chains are homogeneous with respect to their polarizability on their whole length.

(2) The additivity of polarizability formulated by Denbigh[4] is true to the macromolecule chains.

(3) The polarizability of chain fragment with length (dl) represents a polarizability tensor $[\alpha]dl$. The tensor $[\alpha]$ is a polarizability per unit length tensor. The dipole moment for a length unit satisfies the condition

$$\mathbf{m} = [\alpha] \cdot \mathbf{E}_{\mathrm{loc}} \qquad (3)$$

The average polarizability per unit length (the elements of tensor $[\alpha]$) for a specified polymer can be calculated if one knows the macromolecule polarizability and the identity period in the direction of the fiber axis.

(4) The ellipsoid of polarizability connected with any macromolecule fragment is axially symmetric. Its axis of symmetry coincides with the axis of macromolecule chain.

(5) All macromolecule chains which form an ideal fiber are parallel to the fiber axis.

(6) The macromolecule chains are very long compared with the distances to one another. (The degree of polymerization is very high.)

(7) Each macromolecule and any selected fragment of macromolecule has an identical environment.

Based on the assumptions listed above, the general formulas for electric field $\mathbf{E}_w$ and polarization vector $\mathbf{P}$ were derived. Then, obtained formulas were substituted to the equation:

$$\mathbf{m} = [\alpha](\mathbf{E} + \frac{4}{3}\pi\mathbf{P} + \mathbf{E}_w) \qquad (4)$$

After some further transformations of eq. (4) (which are detaily presented in Ref. 3) one obtains the direct connection between the dipole moment **m** and the mean macroscopic field in the volume of polymer **E**

$$\mathbf{m} = [a'] \cdot \mathbf{E} \tag{5}$$

Here there has been adopted a coordinate system such that the matrix $[a']$ in it has expressions other than zero only diagonally. The elements of the tensor $[a']$ are

$$a'_{xx} = \alpha_\perp / (C - \alpha_\perp \sqrt{A^2 + B^2})$$
$$a'_{yy} = \alpha_\perp / (C + \alpha_\perp \sqrt{A^2 + B^2}) \tag{6}$$
$$a'_{zz} = \alpha_\parallel / D$$

The factors A, B, C, D are determined by the crystal structure of the fiber considered.[3] $\alpha_\perp$ and $\alpha_\parallel$ are the elements of the matrix $[\alpha]$, where $\alpha_\perp$ = the average polarizability per unit length in the direction perpendicular to the macromolecular chain axis and $\alpha_\parallel$ = the average polarizability per unit length in the direction of the macromolecular chain axis. The factors $a'_{xx}, a'_{yy}, a'_{zz}$ calculated in that way define the effective polarizability per unit length of the macromolecule. As they arise from the expressions (6), they are dependent both on the crystal structure of the polymer considered and on the polarizability of the macromolecule separated from the environment $(\alpha_\perp, \alpha_\parallel)$.

Having eqs. (6), it is possible to find easily the values of refractive indices of the ideal fiber. To achieve this purpose, the following connections were used:

$$\epsilon_i = 1 + 4\pi (P_i / E_i) \tag{7}$$

and

$$\epsilon_i = n_i^2 \tag{8}$$

where $i = x, y, z$. The resulting formulas for the refractive indices of the fiber considered are

$$n_x = \sqrt{1 + (4\pi/s)ka'_{xx}}$$
$$n_y = \sqrt{1 + (4\pi/s)ka'_{yy}} \tag{9}$$
$$n_z = \sqrt{1 + (4\pi/s)ka'_{zz}}$$

where $s = |\mathbf{a}| \, |\mathbf{b}| \sin \gamma$, $\mathbf{a}, \mathbf{b}$ = translation vectors of a plane lattice, which is made by cutting the ideal fiber with a plane perpendicular to its axis, γ = the angle between $\mathbf{a}$ and $\mathbf{b}$, and k = number of the macromolecular chains connected with the prism of the base s. The side edges of this prism are parallel to the fiber axis. The z-axes of the coordinate systems coincide with the axes of the fiber. Based on the refractive indices calculated from (9), one obtains the birefringence of the ideal fiber from

$$\Delta n = n'_z - \tfrac{1}{2}(n_x + n_y) \tag{10}$$

Because in the described method a simplified and static model of the polymer macromolecule was adopted and since only the dipole interactions were taken into account, the results obtained will only be approximations of the real values.

It seems, however, that such an approximation will be closer to the reality then those obtained with the method used till now.

DETERMINATION OF THE POLARIZABILITY OF PET MONOMER UNIT

The applications of the described formulas for refractive indices and bire-fringence in an individual fiber type must be preceded by the calculations of the values of the factors $a'_{xx}, a'_{yy}, a'_{zz}$. These factors are connected through eqs. (6) with the polarizability per length unit of the macromolecule $\alpha_\perp$, $\alpha_\parallel$, and with the structural factors A,B,C,D. The general formulas and the method of calculation of A,B,C,D are presented in Ref. 3. The values of $\alpha_\perp$ and $\alpha_\parallel$ are calculated from the definitions:

$$\alpha_\perp = A_p/L, \quad \alpha_\parallel = A_z/L$$

where L = the length of identity period along the axis of the macromolecule chain, A_z = polarizability of the macromolecule with the length L in the direction parallel to its axis, and A_p = average polarizability of the same macromolecule in the direction perpendicular to its axis. The polarizabilities A_p, A_z can be calculated using the method presented by Keedy et al.[1]

The polarizability of a macromolecule consisting of "n" bonds forming the angles φ_i with direction of the field $\mathbf{E}$ can be determined from the following expressions:

$$A_p = \sum_{i=1}^{n} a_{pi} = \sum_{i=1}^{n} \tfrac{1}{2}[a_{\parallel i} + 2a_{\perp i} - a_{zi}]$$

$$A_z = \sum_{i=1}^{n} a_{zi} = \sum_{i=1}^{n} [(a_{\parallel i} - a_{\perp i}) \cos^2 \varphi_i + a_{\perp i}]$$

(11)

where $a_{\parallel i}$ = polarizability of the ith bond along its axis, $a_{\perp i}$ = polarizability of the ith bond perpendicular to its axis, a_{pi} = polarizability of the ith bond per-

TABLE I
Coordinates of Atoms of PET Chain in Triclinic System According to Bunn and Daubeny[5]

Position of atom	x (Å)	y (Å)	z (Å)
C_1	0.21	−0.51	0.43
O_1	0.24	0.00	1.88
C_2	0.00	−1.08	2.55
O_2	−0.45	−2.47	1.83
C_3	0.00	−0.47	3.99
C_4	0.46	1.01	4.71
C_5	−0.53	−1.53	4.66
C_6	0.53	1.53	6.095
C_7	−0.46	−1.01	6.042
C_8	0.00	0.47	6.76
C_9	0.00	1.08	8.20
O_3	0.45	2.47	8.92
O_4	−0.24	0.00	8.87
C_{10}	−0.21	0.51	10.32

pendicular to the direction of the field $\mathbf{E}$, a_{zi} = polarizability of ith bond along the direction of the field $\mathbf{E}$. During determination of an ideal fiber birefringence it has been assumed that the field $\mathbf{E}$ is directed along the fiber axis, i.e., in accordance with the macromolecule axis. So, the PET macromolecule polarizability should be calculated in the direction of its axis and in the plane perpendicular to that one.

Data given by Daubeny, Bunn, and Brown[5] will be very helpful here because they determine the coordinates of atoms in the unit cell of triclinic system (Table I) and a configuration of the monomeric group of the PET macromolecule (Figure 1).[6] The z-axis (Fig. 1) overlaps with that of the macromolecule and passes through the center of the benzene ring, being simultaneously the center of symmetry of the chain. As results from eqs. (11), the values of A_p and A_z can be calculated when the angles φ_i between the bonds and the direction of the field are known. Let us consider an arbitrary bond with length "r" in triclinic system with geometry being in accordance with that of unit cell PET, where the z-axis coincides with that of macromolecule whereas the origin of coordinates coincides with that of the bond. Let us also assume that in this system the angles between
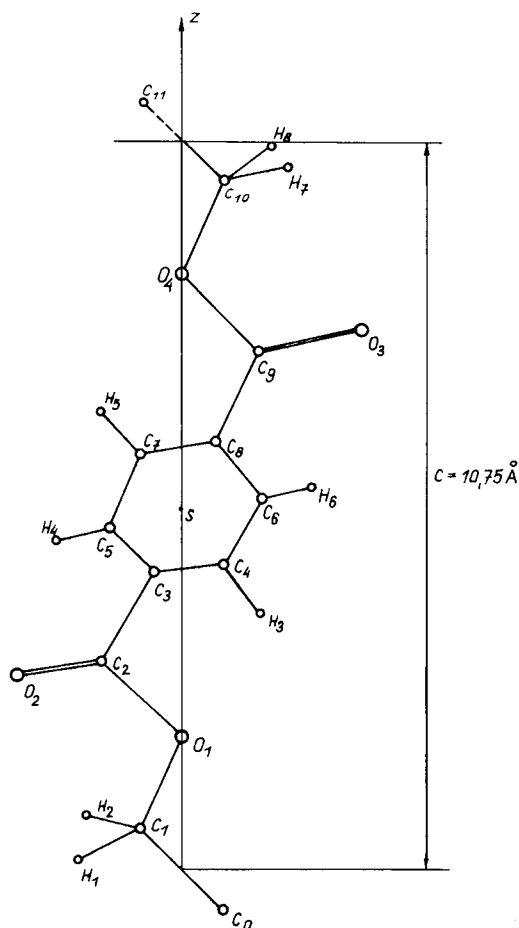

Fig. 1. Structure of macromolecule according to Ref. 5.

TABLE II

Polarizability Values a_z (in Direction of Chain Axis) and a_p (in Plane Perpendicular to Chain Axis) for PET Bonds

| Bond type	Bond length according to Ref. 5	φ (°)	$\cos \varphi$	$a_\parallel \times 10^{25}$ (cm³) (Ref. 4)	$a_\perp \times 10^{25}$ (cm³) (Ref. 4)	$a_z \times 10^{25}$ (cm³)	$a_p \times 10^{25}$ (cm³)
C_1—O_1	1.44	19.90	0.9402688	14.74	1.61	12.92	2.52
O_1—C_2	1.34	47.43	0.6764847	11.67	3.14	7.04	5.45
C_2—O_2	1.27	104.74	-0.2544469	19.9	7.5	8.30	13.30
C_2—C_3	1.49	23.95	0.9138702	18.8	0.2	15.73	1.73
C_3—$C_4(ar)^a$	1.35	76.65	0.2309486	22.5	4.8	5.74	13.18
C_3—$C_5(ar)^a$	1.35	44.61	0.7118893	22.5	4.8	13.77	9.16
C_4—$C_6(ar)^a$	1.35	20.93	0.93400	22.5	4.8	20.24	5.93
C_7—$C_8(ar)^a$	1.35	76.65	0.2309486	22.5	4.8	5.74	13.18
C_6—$C_8(ar)^a$	1.35	44.48	0.7135242	22.5	4.8	13.81	9.14
C_5—$C_7(ar)^a$	1.35	20.985	0.9336744	22.5	4.8	20.23	5.93
C_8—C_9	1.49	23.95	0.9138702	18.8	0.2	15.73	1.73
C_9—O_3	1.27	75.26	0.2544469	19.9	7.5	8.30	13.30
C_9—O_4	1.34	47.43	0.6764847	11.67	3.14	7.04	5.45
O_4—C_{10}	1.44	19.90	0.9402688	14.74	1.61	12.92	2.52
C_0—C_1	1.49	56.30	0.5548374	18.8	0.2	5.93	6.64
C_1—H_1	1.09	102.48	-0.2161169	7.9	5.8	5.90	6.80
C_1—H_2	1.09	102.48	-0.2161169	7.8	5.8	5.90	6.80
C_{10}—H_7	1.09	77.52	0.2161169	7.9	5.8	5.90	6.80
C_{10}—H_8	1.09	77.52	0.2161169	7.9	5.8	5.90	6.80
C_4—H_3	1.09	140.28	-0.7692058	7.9	5.8	7.04	6.23
C_7—H_5	1.09	39.72	0.7692058	7.9	5.8	7.04	6.23
C_5—H_4	1.09	99.525	-0.16554813	7.9	5.8	5.86	6.82
C_6—H_6	1.09	80.475	0.1654813	7.9	5.8	5.86	6.82

[a] ar = aromatic bond.
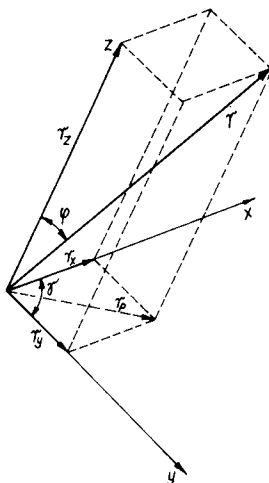
Fig. 2. PET unit cell according to Ref. 5: Angles between system axes are the same as these between cell PET edges; r = bond length.

the axes are the same as those between the edges of unit cell PET (Fig. 2). It results from the figure that

$$r_p^2 = r_x^2 + r_y^2 + 2r_x r_y \cos \gamma$$

$$r_p^2 = r^2 + r_z^2 - 2rr_z \cos \varphi$$

Hence

$$\cos \varphi = (r^2 + r_z^2 - r_p^2)/2rr_z$$
$$\cos \varphi = [r^2 + r_z^2 - (r_x^2 + r_y^2 + 2r_x r_y \cos \gamma)]/2rr_z \tag{12}$$

Knowing the coordinates of the bond, r_x, r_y, r_z, and its length r, the angle φ between this bond and z-axis can be calculated from (12), assuming that $\gamma = 112°$.[5,6] The results of calculations of bond polarizabilities in direction of the PET chain axis a_z and in the plane perpendicular to it, a_p, are presented in Table II. In these calculations the bond polarizabilities values $a_\parallel$ and $a_\perp$ given by Denbigh[4] were used.

In order to determine the values $a_\parallel$ and $a_\perp$ for the bond C—O that were not given by Denbigh, approximate dependence between bond length and its polarizability as suggested by Denbigh[4] has been used. Because of the fact that in Ref. 5 there is no data that refer to the coordinates of hydrogen atoms, the method of calculation of the angle between the bonds C—H and the macromolecule PET axis should be explained. This problem has been considered separately for hydrogen atoms in benzene ring of the monomer unit of PET and for those of H_1, H_2, H_7, H_8 (Fig. 1).

In the first case it has been assumed that:

(a) hydrogen atoms are situated in the same plane as the carbon atoms in benzene ring,

(b) the benzene ring is a regular hexagon of 1.35 Å side,

(c) hydrogen atoms are situated on extensions of hexagonal ring diagonals (Fig. 3).
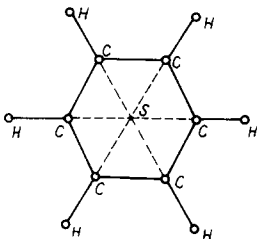
Fig. 3. Explained in the text.

It results from the above mentioned that the angles of inclination φ between bonds C—H and the chain axis must be the same as between the segments SC (Fig. 1) which corresponds to them. Knowing coordinates of the point S and of adequate carbon atoms, angles φ have been determined for SC segments and by the way also for C—H bonds that are their extensions.

In the second case, according to the data given by Williams and Flory,[7] it has been assumed that the angle $\angle CCH$ is 109°. This value was assumed for the following angles: $\angle C_0C_1H_1$, $\angle C_0C_1H_2$, $\angle C_{11}C_{10}H_7$, $\angle C_{11}H_{10}H_8$ (Fig. 1).

The angles $\angle H_1C_1H_2$ and $\angle H_7C_{10}H_8$ were assumed, according to Ref. 8 as equal to 109.5°. Calculations of angles φ between C—H bonds and the PET macromolecule axis were presented as an example for the group of bonds C_1—H_1 and C_1—H_2. For that let us consider a rectangular coordinate system whose center is the carbon atom C_1 (Fig. 4). The bond C_0—C_1 is situated in the plane $Y'Z'$ of the system and the axis Y' coincides with bisector of angle $\angle H_1C_1H_2$. Having assumed that these bonds are vectors with the origins in the center of coordinate system, we can establish as follows:
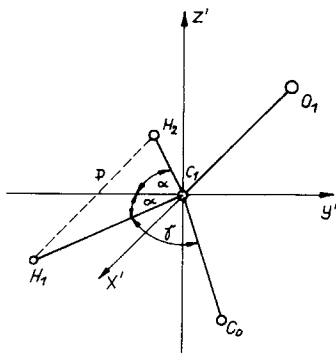
$$\mathbf{r}_{C_0C_1} \cdot \mathbf{r}_{C_1H_1} = r_{C_0C_1} \cdot r_{C_1H_1} \cdot \cos \gamma \qquad (13)$$

and besides

$$\mathbf{r}_{C_0C_1} \cdot \mathbf{r}_{C_1H_1} = r_{C_0C_1} \cdot \cos \angle C_0C_1P \cdot r_{C_1H_1} \cdot \cos \alpha \qquad (14)$$

Comparing both the above equations, we obtain

$$\cos \gamma = \cos \angle C_0C_1P \cdot \cos \alpha$$

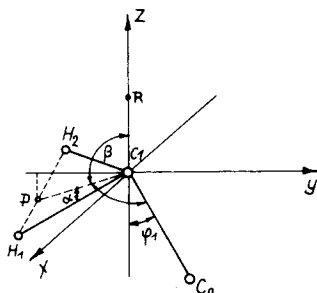Fig. 4. Auxiliary rectangular system $\gamma = 109°$, $\alpha = 54.75°$.

Fig. 5. System axis z coincides with that of macromolecule chain; P,R = geometrical auxiliary points.

Hence

$$\cos \sphericalangle C_0 C_1 P = \cos \gamma / \cos \alpha$$

and, assuming that $\gamma = 109°$ and $\alpha = 54.75°$, we obtain that the angle

$$\sphericalangle C_0 C_1 P = 124.3°$$

It is known from earlier calculations that the bond C_0—C_1 forms with chain axis the angle $\varphi_1 = 56.3°$. Knowing the values of angle φ_1 and of $\sphericalangle C_0 C_1 P$, the angle between the bisector of $\sphericalangle H_1 C_1 H_2$ and the chain axis can be determined. Assuming (Fig. 5) that the axis of system coincides with the macromolecule axis, we obtain the following:

$$(\sphericalangle C_0 C_1 P - \varphi_1) + \beta = 180°$$

Hence

$$\beta = 180° - \sphericalangle C_0 C_1 P + \varphi_1 = 112°$$

Considering the bond C_1—H_1 and the segment $C_1 R$ (Fig. 5) as two vectors with origins in the center of the coordinate system, we obtain

$$\mathbf{r}_{C_1 H_1} \cdot \mathbf{r}_{C_1 R} = r_{C_1 H_1} \cdot r_{C_1 R} \cdot \cos \sphericalangle H_1 C_1 R \tag{15}$$

$$\mathbf{r}_{C_1 H_1} \cdot \mathbf{r}_{C_1 R} = r_{C_1 R} \cdot r_{C_1 H_1} \cdot \cos \alpha \cos \beta \tag{16}$$

Comparing relations (15) and (16), we obtain

$$\cos \sphericalangle H_1 C_1 R = \cos \alpha \cos \beta, \quad \sphericalangle H_1 C_1 R = 102.5° \tag{17}$$

The angle $\sphericalangle H_1 C_1 R$ is the sought for angle φ between the C_1—H_1 bond and the PET chain axis. The angle φ for the C_1—H_2 bond, according to what was assumed before, has the same value.

As has been mentioned above, the values of A_p and A_z for the PET monomer unit were calculated using two sources of data: the values of transversal $(a_\perp)$ and longitudinal $(a_\parallel)$ polarizabilities of the bonds given by Denbigh[4]; the coordinates of atoms in the PET unit cell given by Daubeny, Bunn, and Brown.[5] However, the lengths of some bonds given by Denbigh differ slightly from the lengths of the some bonds given by Daubeny et al.[5]—compare Tables II and III. This disagreement can cause some small errors in calculations of A_p and A_z, but it seems that they are omittable considering the simplified model the macromolecular chain which has been assumed in the calculation of birefringence.

TABLE III

Bond type	Bond length (Å)
C—O	1.43
C=O	1.24
C—C	1.54
C—C (ar)	1.39

RESULTS AND DISCUSSION

Based on the data listed in Table II, polarizabilities, longitudinal A_z = 222.86 $\times$ 10^{-25} cm^3 and transverse, A_p = 162.48 $\times$ 10^{-25} cm^3, have been calculated. Than, using the formulas (6), (9), and (10) the values of refractive indices and birefringence of the ideal PET fiber were calculated.

Calculations were carried out taking into account larger and larger range of intermolecular interactions. Results are presented in the Table IV. The number N means here the number of succeeding surroundings of considered macromolecule, the influence of which is taken into account during the calculation. The term "surrounding" means a group of macromolecules on the perimeter of the parallelogram in the center of which the considered chain is situated. The first surrounding is formed by macromolecules on the perimeter of parallelogram with sides: $2a$ and $2b$ long (a,b = translation vectors of plane lattice which is made by cutting the ideal fiber with a plane perpendicular to its axis), whereas the Nth one—on the perimeter of the parallelogram with sides $2Na$ and $2Nb$ long.

TABLE IV
Results of Ideal Birefringence Determination for PET Fiber with Regard to Interactions of Surrounding Macromolecules

N	n_x	n_y	$n_p = (n_x + n_y/2)$	n_z	Δn
1	1.67356	1.44013	1.55685	1.79292	0.236077
2	1.67409	1.43991	1.55700	1.79292	0.235921
3	1.67432	1.43982	1.55707	1.79292	0.235852
4	1.67466	1.43968	1.55717	1.79292	0.235754
5	1.67483	1.43961	1.55722	1.79292	0.235704
6	1.67492	1.43957	1.55725	1.79292	0.235675
7	1.67499	1.43955	1.55727	1.79292	0.235657
8	1.67503	1.43953	1.55728	1.79292	0.234645
9	1.67506	1.43952	1.55729	1.79292	0.235636
10	1.67508	1.43951	1.55729	1.79292	0.235630
11	1.67509	1.43950	1.55730	1.79292	0.235625
12	1.67511	1.43950	1.55730	1.79292	0.235622
13	1.65712	1.43949	1.55730	1.79292	0.235619
14	1.67512	1.43949	1.55731	1.79292	0.235617
15	1.67513	1.43949	1.55731	1.79292	0.235615
16	1.67513	1.43949	1.55731	1.79292	0.235613
17	1.67514	1.43948	1.55731	1.79292	0.235612
18	1.67514	1.43948	1.55731	1.79292	0.235611
19	1.67515	1.43948	1.55731	1.79292	0.235610
20	1.67515	1.43948	1.55731	1.79292	0.235609

Simultaneously, for comparison of results, the calculations of PET fiber birefringence without regard to interactions has been carried out. This calculation has been realized by means of the method used in Refs. 1. One obtains $\Delta n = 0.2614$.

From comparison of the values presented in Table IV with that above it arises that the interactions of the nearest neighbors—the first surrounding—had the greatest influence on birefringence value. The influence of the next surroundings on the result is nearly omittable considering attainable measuring accuracy of the birefringence. Therefore, one can assume that the birefringence of the ideal PET fiber with regard to intermolecular interactions is equal to

$$\Delta n = 1.7929 - 1.5568 = 0.2360$$

The difference between the birefringence values calculated by means of these two methods is significant and equal to 0.0254. The value of birefringence presented in the actual paper differs also from the values which can be found in the literature. Dumbleton,[9] using Stein's equation[10] and data of sonic method,[11] suggests for polyester $\Delta n = 0.275$. Wiśniewska[12] using the sonic data obtains that $\Delta n = 0.267$.

The results mentioned above are obtained by means of the extrapolational methods which have used the experimental results. They are close to the result obtained by the authors without regarding to interactions ($\Delta n = 0.2614$).

Intermolecular interactions effect in a very important way on the optical properties of the polymer fibers, so it seems that the value of birefringence calculated by means of the method which takes into account this interactions

$$\Delta n = 0.2360$$

is more correct than the others.

It should be noticed that obtained results are considerably dependent on the assumed values of bonds polarizabilities. In the literature one can find the sets of values of bond polarizabilities which differ considerably, for example, data of Denbigh and Bunn.

Calculations of birefringence presented in this paper were carried out using the data of Denbigh, considering the conclusions suggested by Keedy et al.[1]

The authors can deliver on request the computer program for the calculation of birefringence by means of the method described in this paper.

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