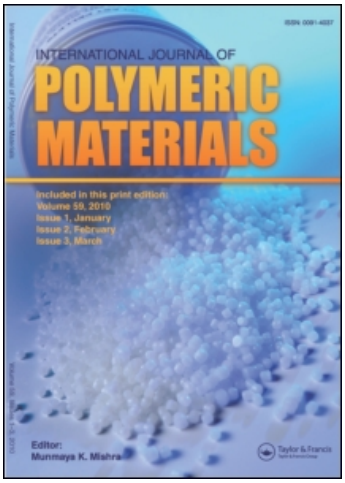


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# Study of Optical and Structure Properties of Polyester (PET) and Copolyester (PETG) Fibers by Interferometry

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# Study of Optical and Structure Properties of Polyester (PET) and Copolyester (PETG) Fibers by Interferometry

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*In this article results on the influence of drawing on the optical and structure properties of polyester (polyethylene terephthalate) and copolyester (glycol-modified polyethylene terephthalate) (4 wt% of ethylene glycol) fibers are reported. Refractive indices, intrinsic birefringence, mean polarizability per unit volume, optical orientation function, density, volume fraction of crystalline material, weight fraction crystallinity, and volume fraction of amorphous and work per chain of these fibers have been evaluated at different draw ratios. A comparison of optical and structure parameters for copolyester fiber and referenced monopolyester fiber has been done. This study has been carried out by means of computerized double-refracting Pluta interference microscope combined with opto-mechanical devices. Microinterferograms and relationships between the parameters obtained are given for illustration.*



**Keywords:** automatic fringe analysis, copolyester fiber, interferometry, optical, opto-mechanical, polyester, refractive indices, structural

## INTRODUCTION

The Poly(ethylene terephthalate) (PET) is used industrially as an inherently semicrystalline polymer, whereas the copolyester (PETG)

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is an uncrystallizable (amorphous) polymer. PET and PETG are found in numerous applications in the form of films and fibers of different thickness and orientation. PET is also one of the most studied polymers because the recrystallization of the glassy amorphous material could be obtained by drawing, which gives an anisotropic structure [1]. Consequently, PET and PETG fibers have been the focus of a great deal of research for many years.

Oriented PET and PETG can be produced using many techniques including tensile drawing, roll drawing, blow molding, and solid state extrusion [2]. Each of these processes has particular applications and advantages, relating to either the final properties or the economics of production. It is thus of primary importance to have fibers with improved mechanical properties able to impart to composite materials adequate strength, dimensional stability, toughness, and impact strength. Mechanical properties can be improved by a modification of the chemical structure of the fiber or by a modification of the physical structure without changing the basic composition. The improvement of mechanical properties of polymeric fibers depends on the control of three main parameters: the molecular mass of the polymer, the structural regularity of the molecule, and the drawing conditions.

Drawing is one of the most common methods for changing the physical properties of a polymeric material to strengthen it during processing. The main effect of drawing is the orientation of the chains and super molecular structures.

Microinterferometry has been widely employed for non-destructive testing in the polymeric fiber field [3–4]. The interferometry based on Biolar polarizing interference (PI) system has been recognized to be a useful tool for the measurement of the optical properties of polymeric fibers [3].

In this work, the opto-mechanical device is combined with computerized double-refracting Pluta interference microscope [5–6] for studying the effect of drawing on the optical and the structure properties of the polyester (PET) and copolyester (PETG) (glycol-modified PET, 4 wt% of ethylene glycol) fibers. The optical and structural parameters, like directional refractive index, refractive index profile, birefringence, mean polarizability per unit volume, orientation function, the fiber density, the volume fraction of crystalline material, the weight fraction crystallinity, the volume fraction of amorphous, and the average work per chain can be calculated. The tested sample is prepared by Kafr El-Dawar factory for production of polyester, Egypt.

## THEORY

### Application of Two-Beam Interference Microscope to Fibers

In general, the intensity distribution observed in the exit plane of two-beam interference microscope can be described as follows:

$$I = I_T + I_c \cos[\Delta(x, y) - \Phi] \quad (1)$$

where $I_T$ is the background intensity, $I_c$ is the interference pattern amplitude governing the image contrast, $\Delta(x, y)$ is the phase change introduced by the investigated object, and $\Phi$ is the bias, that is the phase difference between the interfringe wavefronts. When $\Phi$ is a constant, the observed field is homogenous (in the absence of $\Delta(x, y)$; empty field). When $\Phi$ is a function of space coordinates, fringe appears, that will be perturbed by the presence of the object introduced phase retardation $\Delta(x, y)$. From the form of Eq. 1, it follows that the intensity I in any interferogram is a periodic function of the bias $\Phi$ with $2\pi$ period. The phase shift of a probing beam is detected as fringe shift and the refractive index profile is calculated from fringe shift distribution. The optical path length "$\delta(x, y)$" covered by the illumination wave while traversing the object under study can be given by

$$\delta(x, y) = \frac{\lambda}{2\pi}\Delta(x, y) \quad (2)$$

where $\lambda$ is the light wavelength. The optical path difference ($\delta$) produced by the birefringent fiber is given by:

$$\delta^i = \pm t\,(n^i - n_L) = \pm\frac{Z^i\lambda}{b}, \quad (3)$$

where i denotes the state of polarization of the light used (||, parallel or $\perp$, perpendicular to the fiber axis under study), $n^i$ is the mean directional refractive index, $n_L$ is the refractive index of the immersion liquid, t is the thickness of the fiber, b is the interfringe spacing, and Z is the fringe deflection in the fiber image.

The mean refractive indices ($n^{||}$ and $n^{\perp}$) and the fiber birefringence ($\Delta n = n^{||} - n^{\perp}$) can be determined using Eq. 3.

### The Intrinsic Birefringence $\Delta n_{max}$ of Partially Oriented Fibers

Computerized Pluta microscope attached with a drawing device is used for determination of the intrinsic (maximum observable) birefringence $\Delta n_{max}$ at complete orientation of assembly for the fiber. For this purpose, the orientation parameter of the fiber at any given value of

the draw ratio is determined using mathematical expression depending on the theory given by Ward [7]:

$$\Delta n_{max} = 2\Delta n \left[ \frac{3}{1 - K^2} - \frac{3K \cos^{-1} K}{(1 - K^2)^{3/2}} - 1 \right]^{(-1)}, \quad (4)$$

where $\Delta n = n^{\parallel} - n^{\perp}$ is the birefringence of the fiber and $K = D^{-3/2}$; D is the draw ratio of the fiber.

## Determination of the Unknown Initial Value of Draw Ratio

The unknown initial value of draw ratio ($D_i$) for a given sample of fiber is determined. The refractive indices of the fiber are determined using the drawing device at several values of the draw ratios as $(D_i,\ D_i + R_1, D_i + R_2, \ldots\ldots, D_i + R_s)$, where $R_s$ is the increment in the experimental draw ratio and $s = 1, 2, 3, \ldots$. Plotting the relation between the draw ratio R versus the refractive indices ($n^{\parallel}$ and $n^{\perp}$), one gets a straight line represented by the following equations [8]:

$$n^{\parallel} = C_1 + B_1(D_i + R_s), \quad (5a)$$

$$n^{\perp} = C_2 - B_2(D_i + R_s), \quad (5b)$$

where $C_1, C_2, B_1,$ and $B_2$ are constants that can be estimated experimentally. The initial value of draw ratio ($D_i$) can be determined by solving Eq. 5.



## Some Structural Properties

### *The Optical Orientation Function*

The optical orientation function for tested fibers is calculated using the following relation [9]:

$$f(\theta) = (1 + a)f_\Delta - af_\Delta^2, \quad (6)$$

where $f_\Delta$ is an approximated value of $f(\theta)$ given by the following equation [10]:

$$f_\Delta = \frac{\Delta n}{\Delta n_{max}}, \quad (7)$$

where $\Delta n_{max}$ is the intrinsic birefringence $\Delta n_{max}$ of partially oriented fibers [9], $\Delta n$ is the measured mean birefringence, and:

$$a = \frac{2(n^{\parallel} n^{\perp})^2}{n_v{}^3(n^{\parallel} + n^{\perp})} - 1, \quad (8)$$

In which $n_v$ is the virtual refractive index as defined by the following equation:

$$n_v = \sqrt{1 + \frac{3[(n^{\parallel})^2 - 1][(n^{\perp})^2 - 1]}{[(n^{\perp})^2 - 1] + 2[(n^{\parallel})^2 - 1]}} \tag{9}$$

### Average Work per Chain of Fibers

The average work W for a collection of chains depend on the distribution of chain-end distances, and is obtained by the following equation [11]:

$$W = \frac{3K_B^T}{2}\left[\frac{1}{3}(D^2 - D^{-1}) + (D^{-1} - 1)\right], \tag{10}$$

where D is the draw ratio of the fiber, $K_B$ is the Boltzmann's constant and T is the absolute temperature.



### Density

The densities of the samples were estimated with the average refractive index obtained by a two-beam interferometric technique. For PET fiber, de Vries and coworkers found a linear relation between the fiber density (d) and the average refractive index as follows [12]:

$$d = 4.047\ \frac{n_{iso}^2 - 1}{n_{iso}^2 + 2}\quad (g\ cm^{-3}) \tag{11}$$

This relation is independent of the degree of crystallinity and the level of orientation. The average refractive index as the isotropic refractive index ($n_{iso}$) can be calculated using the following equation [13]:

$$n_{iso} = \frac{n^{\parallel} + 2n^{\perp}}{3} \tag{12}$$

### Crystallinity

The volume fraction of crystalline material $\chi_v$ and the weight fraction crystallinity ($\chi_w$) are calculated from the density data with the following equations [14]:

$$\chi_v = \frac{d - d_a}{d_c - d_a}, \tag{13a}$$

$$\chi_w = \frac{d - d_a}{d_c - d_a} \cdot \frac{d_c}{d_a}, \tag{13b}$$

where the crystallinity in determined in relation to 100% amorphous ($d_a$) and 100% crystalline ($d_c$) polymer densities, $d_a = 1.331\,g\ cm^{-3}$ and $d_c = 1.455\,g\,cm^{-3}$ [15]. The volume fraction of amorphous material

was determined by the following relation:

$$1 - \chi_v = 1 - \frac{d - d_a}{d_c - d_a} \tag{14}$$

## Measuring Technique

The measuring technique (Figure 1) consists of: variable-wavefront shear birefracting microinterferometer (Pluta polarizing interference

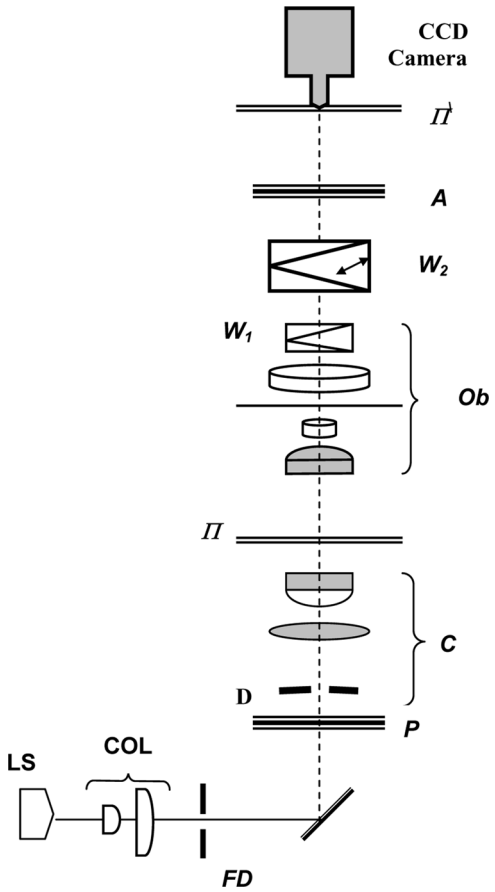


**FIGURE 1** Schematic diagrams: (a) Experimental setup of the Pluta polarizing interference microscope (Biolar PI): LS—light source, Col—collimator, FD—field diaphragm, P—polarizer, D—regular slit diaphragm, Π—object plane, $W_1$—rotatable Wollaston prism, $W_2$—tube Wollaston prism, A—analyzer, Π—image plane.

microscope Biolar PI) for transmitted light, mechanical (drawing) device, CCD camera, and PC computer. The output field of the microscope is scanned by CCD camera and the interference image is automatically captured and processed.

## RESULTS AND DISCUSSION

### Influence of Drawing on the Optical and Structural Properties of PET and PETG Fibers

Polyester (PET) and copolyester (PETG) are widely used for products such as drawn fibers. Their commercial success is principally due to their abilities to crystallize at large draw ratios during warm deformation processing. The imparted crystallinity increases stiffness and strength, improves dimensional stability, and increases density [15]. The computerized double-refracting Pluta interference microscope combined with a fiber-drawing instrument designed by Hamza and his co-workers [16] is employed for studying the opto-mechanical properties of PET and PETG fibers. PET and PETG fibers of initial draw ratio ($D_i$) are drawn to several values of low draw ratios ($D_i,\ D_i + R_1, D_i + R_2, \ldots\ldots, D_i + R_s$; $R_s$ is the increment of the draw ratio with respect to initial draw ratio $D_i$). These fibers are placed on the microscope slide and sited in a suitable position inside the mechanical device and immersed in a certain liquid ($n_L = 1.604$ at temperature 20°C). This mechanical device is attached to the Pluta interference microscope. The interference microscope is adapted to differentially sheared (duplicated) images configuration. When the fiber is drawn the draw ratio of the fiber can be varied gradually. Therefore the fringe deflection ($Z^i$) in the image of fiber and its thickness are changed. The microinterferograms at these draw ratios can be captured by RGB CCD camera and analyzed using a prepared software program.

Figures 2 and 3 demonstrate a set of microinterferograms that represent the duplicated images of the PET and PETG fibers at different

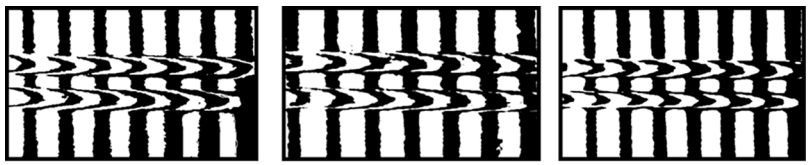


**FIGURE 2** Microinterferograms of drawing PET fiber at ratios D, D + 0.0444, D + 0.1332, and D + 0.2665, respectively.

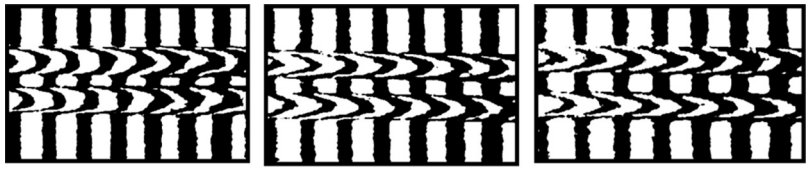


**FIGURE 3** Microinterferograms of drawing copolyster (PETG) fiber at ratios D, D + 0.0444, D + 0.1332, and D + 0.2665, respectively.

draw ratios ($D_i$, $D_i + 0.0444$, $D_i + 0.1332$, and $D_i + 0.2665$, respectively). A monochromatic light of wavelength 546 nm is used.

Using these optical experimental data and solving Eq. 5 the constants $C_1$, $C_2$, $B_1$, $B_2$, and initial draw ratio ($D_i$) are estimated and given in Table 1. Also, the initial draw ratio can be estimated using the extrapolation of the straight lines of Figure 4. The obtained interception was found at a point (actual draw ratio = 1), which corresponds to the isotropic refractive index ($n_{iso} = 1.5950$) and ($n_{iso} = 1.5935$) for PET and PETG fibers, respectively. By transferring the axes to the points (1, 1.5855) and (1, 1.5831) for both graphs, the initial draw ratios are found to be ($D_i = 1.807$) and ($D_i = 1.845$) for PET and PETG fibers, respectively.

Having the initial and suggested draw ratios the actual draw ratios can be calculated. Therefore the mean directional refractive indices ($n^{||}$ and $n^{\perp}$) and the birefringence ($\Delta n = n^{||} - n^{\perp}$) of PET and PETG fibers is plotted against the obtained actual draw ratio as shown in Figures 5 and 6.

These figures show that the refractive index $n^{\perp}$ of both PET and PETG fibers decreases with draw ration whereas the refractive index $n^{||}$ in the direction of the drawing increases rapidly with the draw ratio for compensating the decrease in $n^{\perp}$ to give a net increase in the birefringence ($\Delta n$). As expected, the optical parameters are sensitive to draw ratio and the change in these parameters of PET and PETG fibers due to this low drawing have the same behavior.

**TABLE 1** The Coefficients of the Lines in Figure 4 and the Initial Draw Ratio for PET and PETG Fibers

| | $C_1$ | $C_2$ | $B_1$ | $B_2$ | $D_i$ |
|---|---|---|---|---|---|
| PET | 1.5711 | 1.6072 | 0.116 | −0.0713 | 1.807 |
| PETG | 1.5805 | 1.6049 | 0.094 | −0.0641 | 1.845 |

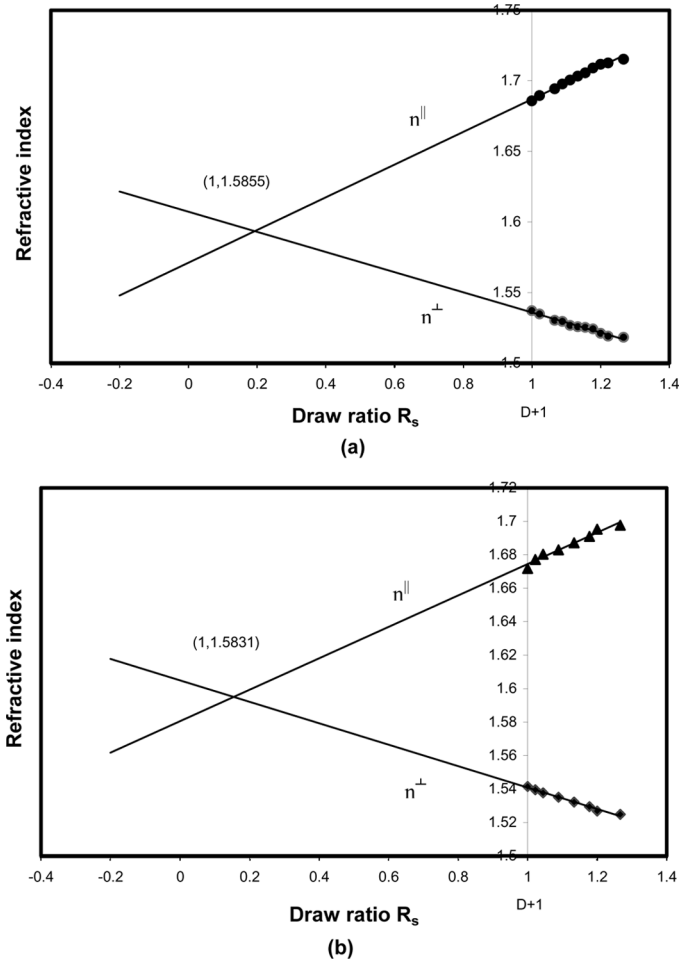


**FIGURE 4** The relation between the draw ratio R and the refractive indices ($n^{\|}$ and $n^{\perp}$) for (a) PET fiber and (b) PETG.

The average values of the intrinsic birefringence $\Delta n_{max}$ of PET and PETG fibers can be calculated using Eq. 4 and found to be 0.3759 and 0.3137, respectively.

The actual measurable characteristic of the polymer fiber is $\langle f(\theta) \rangle$ where $\theta$ is the angle between the statistical segment and the direction of the force inducing orientation; the brackets denote an average. The optical orientation functions of PET and PETG fiber can be calculated using Eq. 6 at different values of draw ratios. Figure 7 shows the

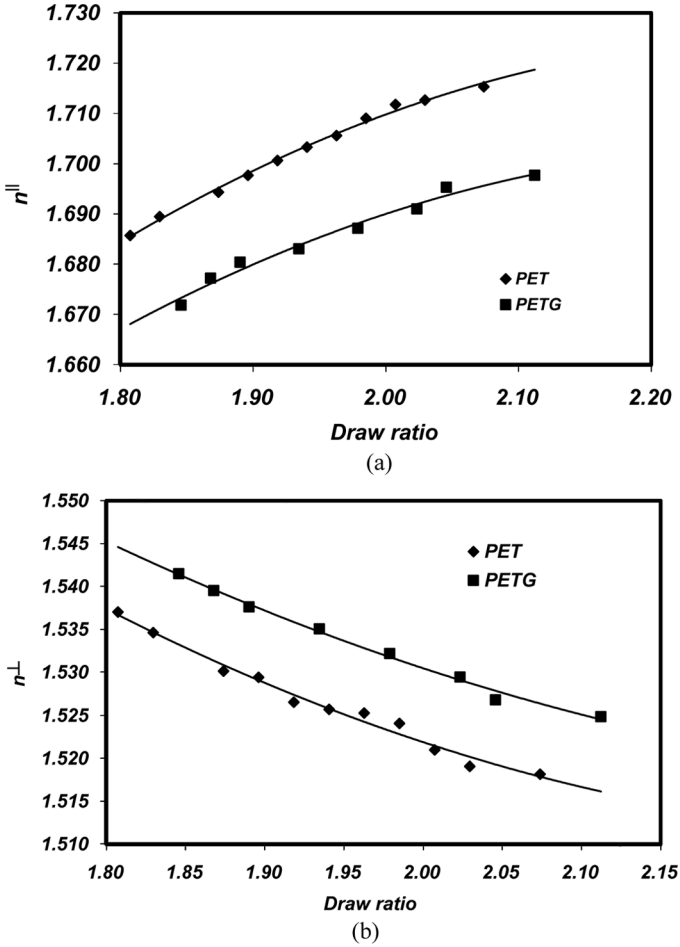


**FIGURE 5** The effect of drawing on the parallel (a) and perpendicular (b) refractive indices for PET and PETG fibers.

relationship between the optical orientation function of both PET and PETG fibers and draw ratio. This implies that the orientation function increased when the drawing increased.

Using Eq. 10 the average work per chain W of PET and PETG fibers is calculated and represented versus the birefringence Δn at room temperature as given in Figure 8. This relation represented by straight line as shown in the following suggested formula:

$$\Delta n = K_1 W + K_2 \tag{15}$$

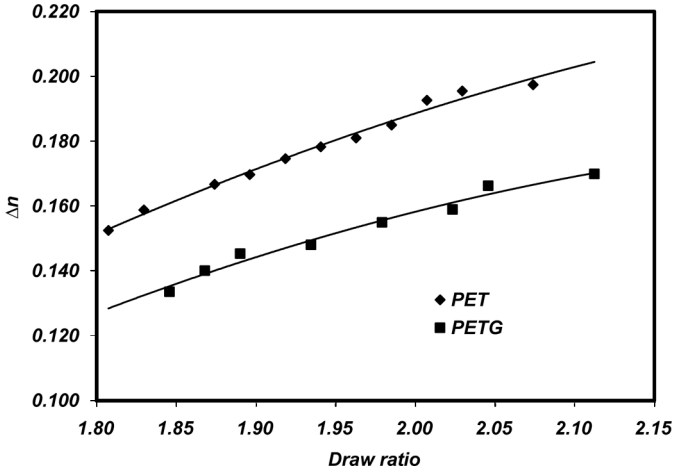


**FIGURE 6** The relation between the change in the birefringence and draw ratio of PET and PETG fibers.



with the constants $K_1 = 3 \times 10^{19}$ $J^{-1}$, $K_2 = 0.0831$ and $K_1 = 2 \times 10^{19}$ $J^{-1}$, $K_2 = 0.0796$ for PET and PETG fibers, respectively. According to Eq. 10 and 15, it is clear that the increase in the draw ratio leads to the increase in each of the stress of the fiber, the work done on the polymeric chain W, and finally the birefringence Δn.

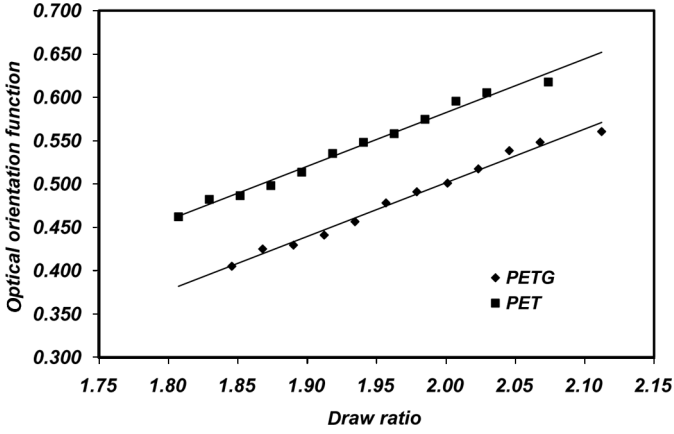


**FIGURE 7** Change of the optical orientation function with the actual draw ratio for PET and PETG fibers.

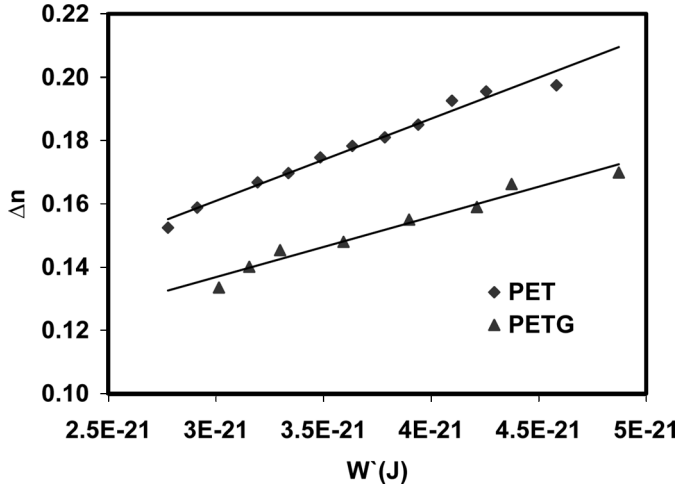


**FIGURE 8** The average work per chain W versus the birefringence (Δn) for PET and PETG fibers at room temperature.



The fiber density (d), the volume fraction of crystallinity material $\chi_v$, the weight fraction crystallinity $\chi_\omega$, and the volume fraction of amorphous $(1 - \chi_v)$ of PET and PETG fibers with drawing can be calculated as given in Tables 2 and 3 using Eq. 11, 13, and 14, respectively, at room temperature.

In general, at room temperature, the refractive indices ($n^{||}$ and $n^{\perp}$), the birefringence (Δn), the orientation function ($\langle f(\theta) \rangle$), and the average work per chain (W) of PET and PETG fibers increase as the draw

**TABLE 2** Structural Parameters of PET Fiber

| Draw ratio | $n_{iso}$ | d (g $cm^{-3}$) | $\chi_\omega$ | $\chi_\omega$ | $1 - \chi_v$ |
|---|---|---|---|---|---|
| 1.8073 | 1.5865 | 1.359314 | 22.83398 | 26.48811 | 77.16602 |
| 1.8295 | 1.5862 | 1.358665 | 22.31061 | 25.88098 | 77.68939 |
| 1.8517 | 1.5859 | 1.358105 | 21.85874 | 25.3568 | 78.14126 |
| 1.8739 | 1.5848 | 1.356055 | 20.20553 | 23.43903 | 79.79447 |
| 1.8961 | 1.5855 | 1.35727 | 21.18544 | 24.57575 | 78.81456 |
| 1.9184 | 1.5845 | 1.355465 | 19.73002 | 22.88742 | 80.26998 |
| 1.9406 | 1.5849 | 1.356121 | 20.25861 | 23.50059 | 79.74139 |
| 1.9628 | 1.5853 | 1.357028 | 20.99026 | 24.34933 | 79.00974 |
| 1.985 | 1.5857 | 1.357651 | 21.49314 | 24.93269 | 78.50686 |
| 2.0073 | 1.5845 | 1.35548 | 19.74188 | 22.90118 | 80.25812 |
| 2.0295 | 1.5835 | 1.353616 | 18.23905 | 21.15785 | 81.76095 |
| 2.0739 | 1.5838 | 1.354166 | 18.6826 | 21.67237 | 81.3174 |

**TABLE 3** Structural Parameters of PETG Fiber

| Draw ratio | $n_{iso}$ | d | $\chi_{\varpi}$ | $\chi_{\omega}$ | $1-\chi_{v}$ |
|---|---|---|---|---|---|
| 1.8457 | 1.5849 | 1.356186 | 20.31151 | 23.56196 | 79.68849 |
| 1.8679 | 1.5854 | 1.357086 | 21.03678 | 24.4033 | 78.96322 |
| 1.8901 | 1.5852 | 1.356707 | 20.73185 | 24.04957 | 79.26815 |
| 1.9123 | 1.5848 | 1.356107 | 20.24743 | 23.48762 | 79.75257 |
| 1.9345 | 1.5843 | 1.355162 | 19.48534 | 22.60358 | 80.51466 |
| 1.9568 | 1.5845 | 1.355385 | 19.66495 | 22.81194 | 80.33505 |
| 1.979 | 1.5838 | 1.3541 | 18.62872 | 21.60988 | 81.37128 |
| 2.0012 | 1.5821 | 1.350923 | 16.0667 | 18.63785 | 83.9333 |
| 2.0234 | 1.5832 | 1.35308 | 17.80643 | 20.656 | 82.19357 |
| 2.0457 | 1.5829 | 1.352463 | 17.30892 | 20.07887 | 82.69108 |
| 2.0679 | 1.5826 | 1.351898 | 16.85285 | 19.54981 | 83.14715 |
| 2.1123 | 1.5824 | 1.351478 | 16.5148 | 19.15767 | 83.4852 |



ratio increases. However, $n^{||}$, $\Delta n$, W, d, $\chi_v$, and $\chi_\omega$ of PET fibers are higher than that of PETG fibers. This confirms that the PET fiber acts as semicrystallide polymer, whereas the PETG is an uncrystallizable (amorphous) polymer; that is, the copolyester (PETG) chain is less regular than PET chain and as result has less crystallinity.

## CONCLUSION

- The change in the optical properties of PET fibers induced by (4 wt%) polyethylene glycol (PETG) is detected from results obtained from PET and PETG fibers ($n^{||}$, $n^{\perp}$, $\Delta n$, $\Delta n_{max}$).
- The method described for determining the initial value of the fiber draw ratio helps in the calculation of intrinsic birefringence ($\Delta n_{max}$) for any given fiber.
- The Biolar polarizing interference (PI) system has become the most secure, most precise, and quickest method.
- To minimize any uncertainty of measurements of the fiber optical parameters using the polarizing microscope, it is necessary to couple this microscope to the image processing system via a CCD camera.
- The orientation parameter ($I_\theta$) for both PET and PETG fibers increases with drawing. This means that crystallites and amorphous chains that are initially randomly arranged are re-oriented with their chain axes along the drawing direction.
- The intrinsic birefringence ($\Delta n_{max}$) achieved under the used mechanism and conditions of drawing was 0.3759 and 0.3137 for normal and modified polyester, respectively.

- Generally, it is worth noting that the optical and structure parameters of polyester (PET) fiber are higher in comparison to the copolyester (PETG) fiber. This is due to the fact that the copolyester chain is less regular than polyester chain and hence has less crystallinity.

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