Birefringent Characterization of Necking Phenomenon Along Cold-Drawn Polypropylene Fibers. I. Offline Drawing

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Received 1 August 2006; accepted 18 December 2006
DOI 10.1002/app.26101
Published online 2 April 2007 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: An automated two-beam interference Pluta microscope was used to measure the optical anisotropy or birefringence of polymer fibers. A sample of polypropylene (PP) fibers was used and cold-drawn to low draw ratios. The neck shoulder ''necking'' was micro-detected at draw ratio 1.3 and characterized using the measured thickness and birefringence along the drawn PP fiber. Birefringence profiles at different regions along deformed PP fiber were determined to confirm the effect of necking on these fibers. The influence of this phenomenon on the molecular orientation function of PP fibers was presented. The strong dependence of the birefringence on the thickness was formulated analytically at the necked zone of the drawn PP fiber. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 105: 757–764, 2007

Key words: interferometry; birefringence; necking; polypropylene; fibers

INTRODUCTION

The phenomenon of neck formation in polymers has attracted considerable attention. During the drawing process, an initial (unoriented, i.e. isotropic or undrawn) material is transformed into an oriented (anisotropic or drawn) one across a narrow transition region called ''neck.'' This necking comprises a localized marked reduction in cross section, which occurs under uniaxial deformation. It can be detected and characterized interferometerically along the fibers by measuring their birefringence, which is highly recommended in the field of textile and polymer technology.

Measurement of the optical anisotropy (birefringence) of the polymer fibers during uniaxial drawing is a useful task to characterize the deformation mechanisms of these man-made fibers. In the particular case of semicrystalline polymers, such as PP fibers, yielding and cold drawing contain two types of nonuniform deformation processes: the first one is the initiation of local necking and the other is the propagation of neck shoulders along the specimen. Both types result from the local instability of deformation but they are different in behavior.[1,2]

In both crystalline and amorphous polymer fibers, a reduction in nominal stress following the initial stress rise has been associated with inhomogeneous strain, or ''necking.'' Deformation involving neck propagation is often referred to as cold-drawing, although at high enough strain rates and in some crystalline polymers, necking can occur above the glass transition temperature (T_g). Necking is characterized by localized extension and corresponding decrease in cross-sectional area of some axial position along the sample.[3]

When a slender bar of a polymeric material in the form of, say, a fiber is subjected to an appropriate uniaxial tensile load or is stretched at an appropriate rate, the initial motion of homogeneous extension can evolve into a nonhomogeneous motion in which the fiber thins down over a short region along its length and forms a neck.[4] Necking in cold drawing is a smooth jump in cross-sectional area of long and thin bars (fibers), propagating with a constant speed. Necks in polymers are now commonly used in modern processing of polymer fibers. Qualitative structural models of necking have been intensively discussed in experimental papers. The most popular such model, proposed by Peterlin and Olf,[5] considered the folded chain blocks in the necking of semicrystalline polymers as being tilted, sheared or broken off the lamellae and becoming incorporated in the (amorphous) microfibrils.

The process of diffuse necking, in particular, has received much attention and was the object of some controversy over the parameters, which control its development. Considerable progress was made when several authors, such as G'Sell et al.,[6] proved

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Journal of Applied Polymer Science, Vol. 105, 757–764 (2007)

that the kinetics of neck formation and propagation could be modeled and predicted quantitatively from the constitutive equation of the material expressed in terms of true stress, strain, and strain rate. Nowadays, this concept is broadly acknowledged, and plastic instability phenomena are thus modeled in complex structures for industrial products by means of finite-element codes. This is the reason why there is such a large need for databases that give the intrinsic parameters of polymers up to large strains.

This work throws light on the interferometric detection and characterization of the necking phenomenon along PP fibers using two-beam Pluta microscope.[7,8] Along the neck shoulder of PP fibers, the thickness, birefringence, and orientation function have been evaluated. In addition the birefringence profile is determined at different regions along the deformed PP fiber. An empirical formula between birefringence and thickness in the neck region of PP fiber is suggested.

WHY THE NECKING OCCURS IN POLYMER FIBERS?

The formation of necking has attracted considerable attention, but it is not readily explained. This unusual behavior has been seen in different polymers such as PET, PBT, polyamide 66, PE, and PP. When necking is observed it is invariably associated with crystallization in the fiber, frequently occurring at a certain point just before crystallization is detected. However, crystallization in the fiber is itself not a sufficient condition to cause necking, as many examples exist of crystallization occurring in the fiber without noticeable neck formation. Some authors[9–11] have suggested that the heat due to drawing supplies enough energy to cause a local increase in temperature of the fiber, resulting in a decrease in local viscosity. Shimizu et al.[12] have suggested that the viscosity may decrease due to formation of an oriented mesomorphic phase.

Necking was first thought to be caused by the transformation of crystalline regions under the applied stress, until it was observed to also occur in amorphous polymers. Another early interpretation was that thermal softening due to localized heating initiated neck formation, but it was later demonstrated that necking could occur under certain isothermal conditions. Both these effects can be involved in the necking process, and will influence its propagation, but they are not necessarily responsible for its initiation. However, most materials are not perfectly homogenous and necking is the manifestation of instability in the yielding process due to defects at the molecular or macroscopic level, which are more compliant than the surrounding material and act as areas of stress concentration.[3]

Recently, Hamza et al.[2] reported that the necking in PP fibers occurred because of the mechanical weak bonds, the formation of voids or pores within the fiber structure, microscopic inhomogeneities and, a large extent of the oriented polymer molecules. Experimentally, during the cold drawing, the necking deformation appears at different regions along the fiber axis when the heating rate of these regions exceeds the limit of flowing temperature of the molecular chains. In cases where the cold-drawing process is associated with the release of quantity of heat that is equivalent to the work done on the fiber, a part of the heat leads to heating the fiber, and the other part is dissipated into the surrounding.

The probability of the necking phenomenon in polymer fibers increases by increasing each of the drawing rates and also increasing in the temperature of the surrounding more than T_g of fiber. Accordingly, the predication of necking phenomena may be mostly probable when: (i) decreasing the temperature of drawing or spinning processes, (ii) increasing the rate or speed of drawing process, and (iii) increasing diameter or thickness of the spun fibers.[13,14]

THEORETICAL CONCEPTS

The mean birefringence Δn of the fiber can be determined interferometerically by the following equations:[15]

$$\Delta n = \frac{\Delta Z \lambda}{bt} \tag{1}$$

where ΔZ is the fringe shift displacement, λ is the wavelength of the monochromatic light used, b is the interfringe spacing, and t is the fiber thickness.

The birefringence profile refers to the variation of birefringence across the fiber radius. This profile (taking into account the refraction of the incident beam by the fiber layers) can be determined using special software[16] based on the following equation;

$$\Delta n_Q = \left[\frac{1}{R-(Q-1)r}\right]\left[\frac{\lambda \Delta Z_Q}{2b} - r\sum_{j=1}^{j=Q-1} \Delta n_j\right] \tag{2}$$

where r is the layer thickness ($r = R/N$) (R is the fiber radius, N is the number of layers), Q is the layer number, and ΔZ_Q is the fringe shift corresponding to the Qth layer.

EXPERIMENTAL TECHNIQUE

The fiber-drawing device[17] was modified and attached to two-beam Pluta microscope as shown in the schematic diagram of Figure 1, in which, a sam-

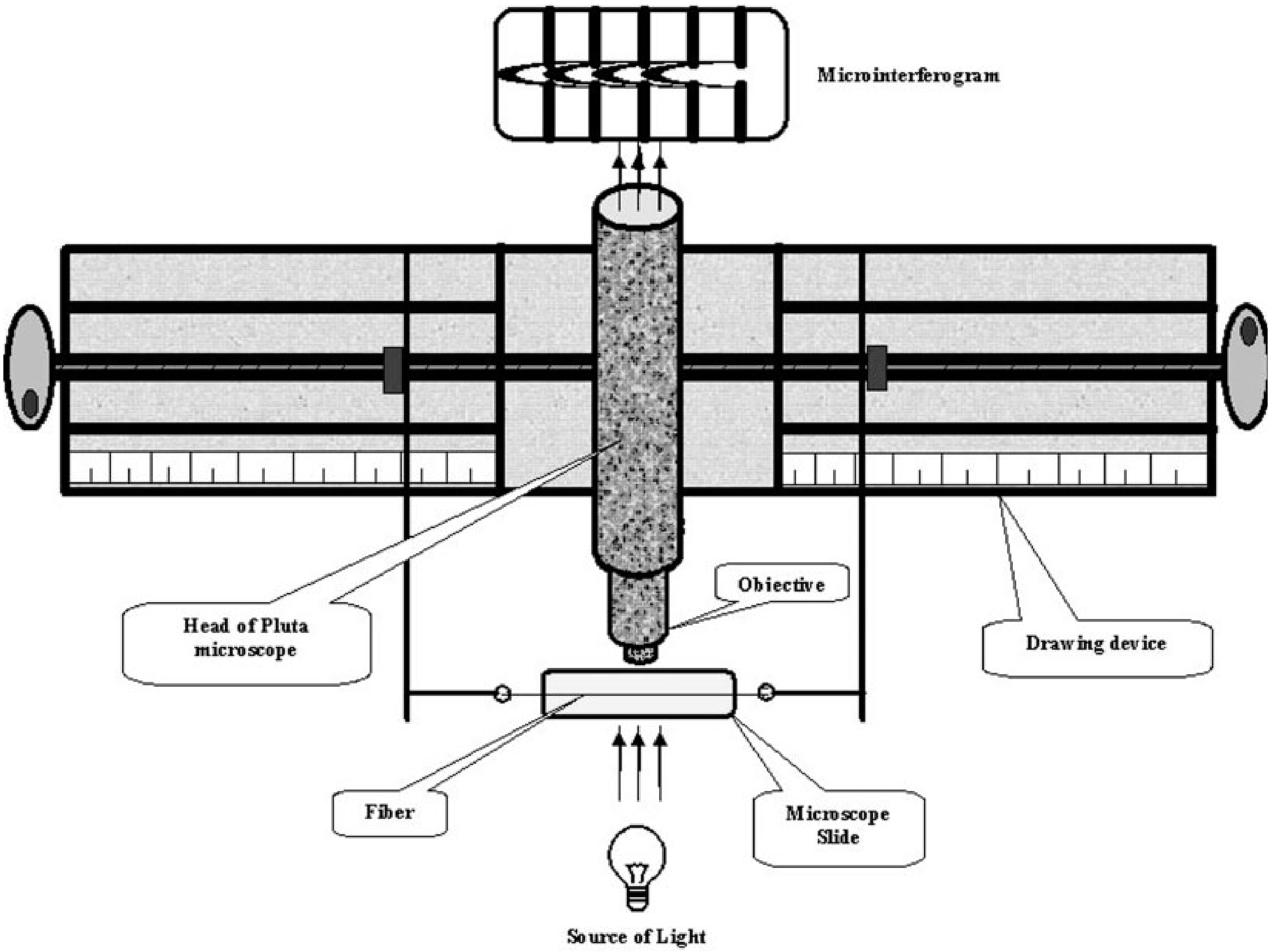


Figure 1 A modified schematic diagram showing the used experimental technique, which consists of Pluta microscope attached with the drawing device and CCD camera as an electronic interface between it and the computer.[17]

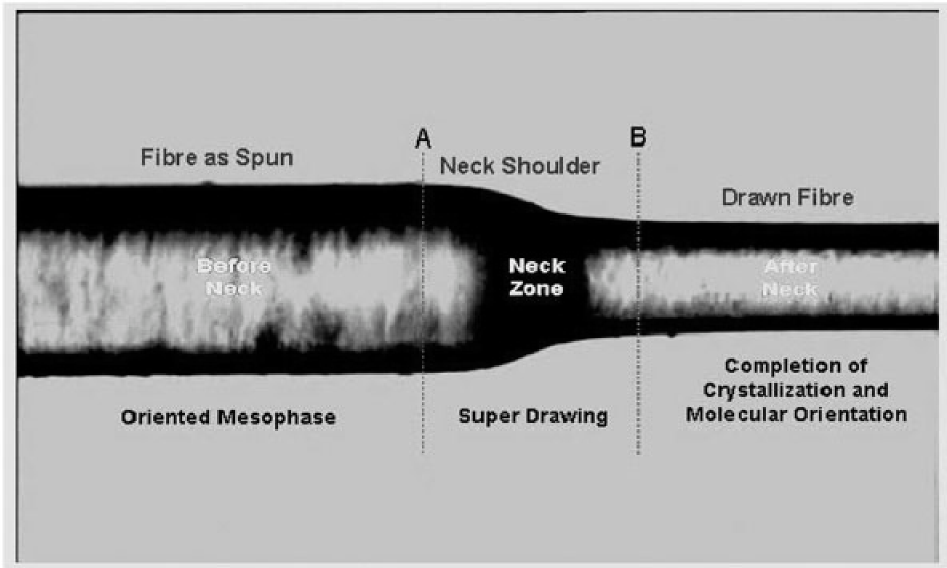
ple (10 cm in length) of undrawn PP fibers was fixed between the two clamps of the drawing device and mounted on the slide stage of Pluta microscope. At room temperature (25°C), the PP fiber was drawn with draw ratio around 1.3. Using monochromatic light of wavelength 546 nm and CCD camera connected between Pluta microscope and PC, microinterferograms are captured and recorded along the fiber axis.

RESULTS AND DISCUSSION

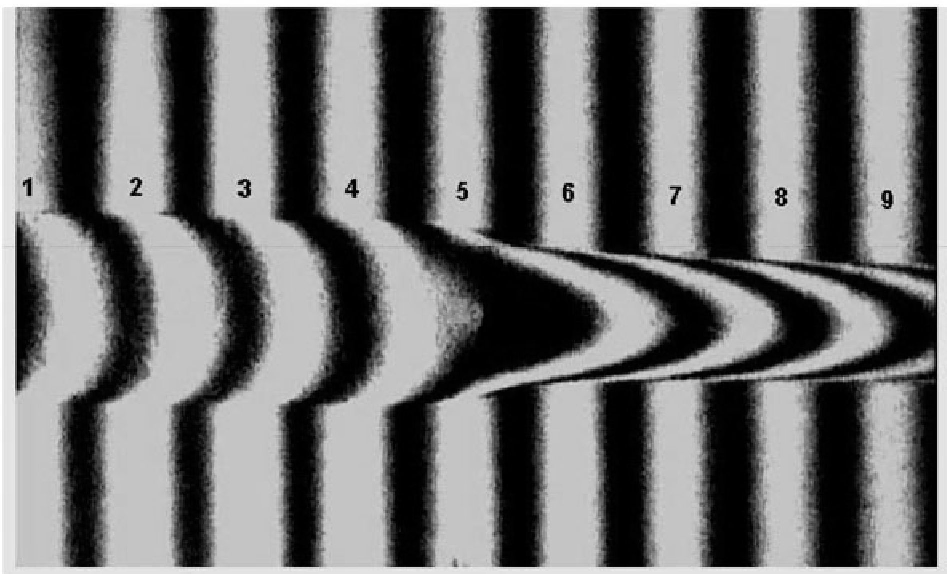
Two images of PP sample are obtained; one of them as shown in Figure 2(a) is photographed using the optical position of Pluta microscope to show the morphological deformation of the PP fiber (DR = 1.3). The second one is called ''microinterferogram'' as shown in Figure 2(b) photographed using the nonduplicated position of Pluta microscope to show the birefringence variation inside the deformed PP fiber.

The necking phenomenon usually occurs when a homogeneous polymeric fiber is stretched uniaxially.[18] In this case, the polymer fiber is not deformed homogeneously. Instead, two almost uniform sections occur in the sample: one being nearly equal to its initial thickness and another being considerably thinner in the cross-sectional dimensions. These sections are jointed by a relatively short transition (necking) zone that propagates with a constant speed along the fiber as a stepwise wave in the direction of the fiber's thick end. This is shown experimentally in PP fiber in Figure 2(a).

In Figure 2, the fiber image (a) is marked by points A and B to be easier for description and fringe shifts of the microinterferogram (b) are also marked by the numbers (1, 2, 3, . . . 9) so as to be easier for measuring the birefringence along the fiber axis. The fiber image [Fig. 2(a)] can be classified into three microscopic regions according to the morphological model of Shimizu et al.[9] and the molecular



(a)



(b)

Figure 2 Imaging of fiber of cold-drawn polypropylene (PP) fibers; (a) fiber image using the optical position of Pluta microscope and (b) fiber microinterferogram using the nonduplicated position of Pluta microscope.

model of Peterlin.[19] The ''as spun'' part of fiber (before neck) may be identified as ''oriented mesophase,'' the neck shoulder (neck zone) as ''super drawing'' and drawn fiber part (after neck) as ''completion of crystallization and molecular orientation.'' The microinterferogram shown in Figure 2(b) can be interpreted basically on this model as follows: (i) In the oriented mesophase, the displacements of fringe shifts (1, 2, and 3) inside the fiber are small, this means that the fiber is slightly drawn, has larger thickness and lower birefringence; (ii) in the super drawing (AB), the displacements of fringe shifts (4 and 5) inside the fiber are longer, this means that the fiber is super drawn, has gradient thickness and moderate birefringence; (iii) In the completion of crystallization and molecular orientation, the displacements of fringe shifts (6 and 7) inside the fiber are too longer, this means that the fiber is highly drawn, has smaller thickness and high birefringence.

Figure 3 shows the graphical representation of Figure 2 in which there are two graphs; the first one (squared-points graph) shows the variation of the

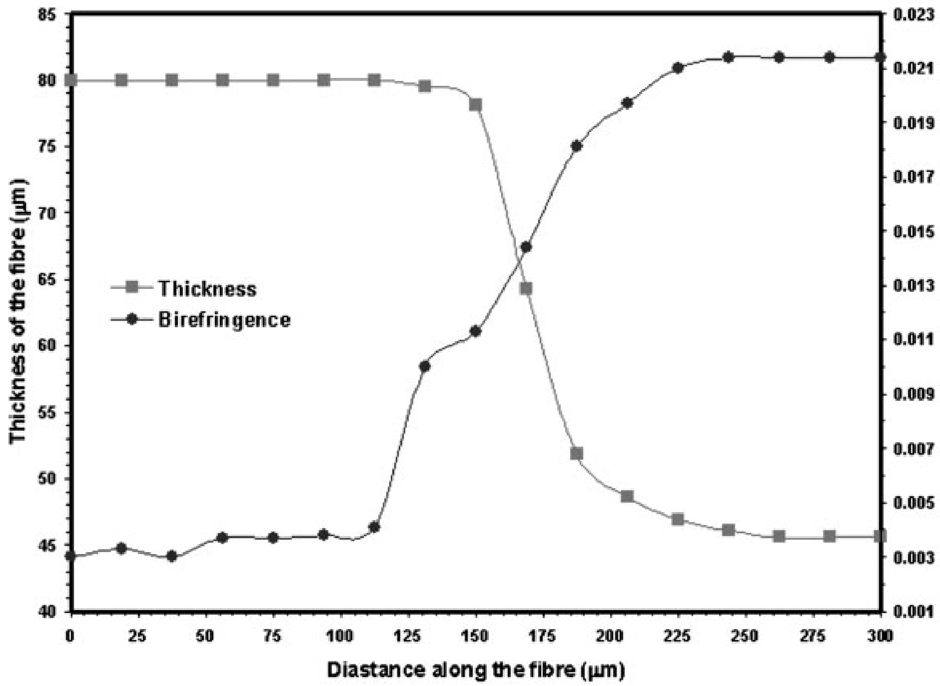


Figure 3 Variation of fiber thickness and birefringence along the axis of drawn PP fiber.

fiber thickness along the PP fiber axis and the second one (circled-points graph) shows the variation of the mean birefringence [calculated using eq. (1)] of PP fiber along the fiber axis. The thickness and birefringence of the fiber were measured from the fiber image (a) and microinterferogram (b) shown in Figure 2, respectively. It is clear the behavior of the squared-points graph reflects the thickness gradient of the deformed fiber, that is, the fiber thickness is constant until the distance 112.39 µm or point A [Fig. 2(a)] then slightly decreases until the distance 149.85 µm then suddenly drop to the distance 243.5 µm or point B [Fig. 2(a)] then becomes constant again. This significant draw down in the fiber thickness through the region AB [Fig. 2(a)] verifies the existence of ''necking phenomenon'' or ''neck shoulder'' at this region (AB) along the fiber. Also it is seen that the mean birefringence (circled-points graph) rises abruptly along the PP fiber, especially at the zone (AB) ranging from 112.39 µm to 243.5 µm [Fringe shifts 5 and 6 in Fig. 2(b)], at which the neck occurs.

The birefringence profile, which presents accurate measurements of the birefringence across the fiber radius, was determined via software program[16] based on eq. (2). Figure 4 shows four birefringence profiles of PP fiber at different positions or fringes (3, 4, 5, and 6) along the axis of this fiber. It is obvious that the first profile (Fringe 3) is flat, that is, the birefringence has constant values across the fiber radius, and this profile confirms that this part of the fiber still undrawn. The second one (Fringe 4) converges, that is, the birefringence has variable values across the fiber radius, and this profile confirms that this part of the fiber is slightly drawn. The third one (Fringe 5) seams to be an elongated convergence, that is, the birefringence shoots up and has significant rise in its values across the fiber radius, this profile confirms that this part of the fiber is super drawn or necked. The fourth profile (Fringe 6) seams to be more elongated, that is, the values of birefringence become high and vary gradually from the center to the outside of the fiber, which profile confirms that this part of the fiber is fully oriented or drawn. Now the necking phenomenon in PP fibers becomes more abrupt and more evident with a rapid draw down in the fiber thickness and rapid increase in the values of birefringence along and across the fiber axis.

An alignment of amorphous or crystalline chains during an extension or drawing deformation in polymer materials raises the birefringence that called ''orientation birefringence.'' This birefringence is the quantity that is most generally measured when characterizing the molecular orientation of the drawn fibers.[20] The relation between the oriented (observable) birefringence Δn and the intrinsic (maximum) birefringence Δn_o defines this orientation function as follows:[21,22]

$$F_\Delta = \frac{\Delta n}{\Delta n_o} \tag{3}$$

Using the calculated data of Δn and $\Delta n_o = 0.045$[24] of PP fibers into eq. (3), the orientation factor F_Δ is determined along the drawn PP fiber as shown in Figure 5. It is clear that this factor, which expresses

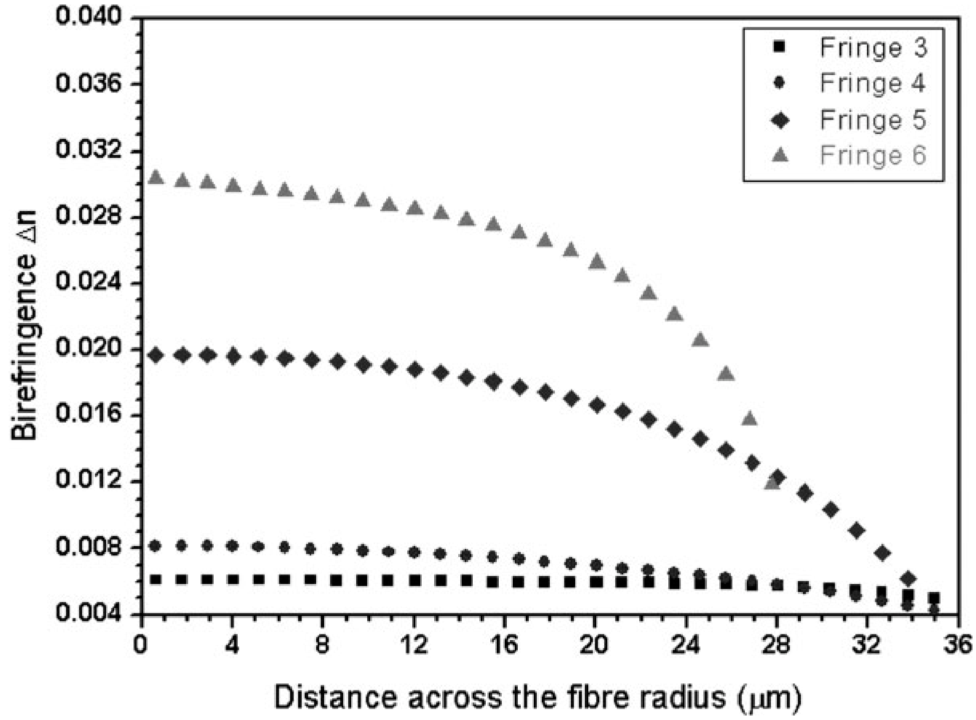


Figure 4 Variation of birefringence across the radius of drawn PP fiber (i.e. birefringence profile) at different positions (fringe shifts) along the fiber.

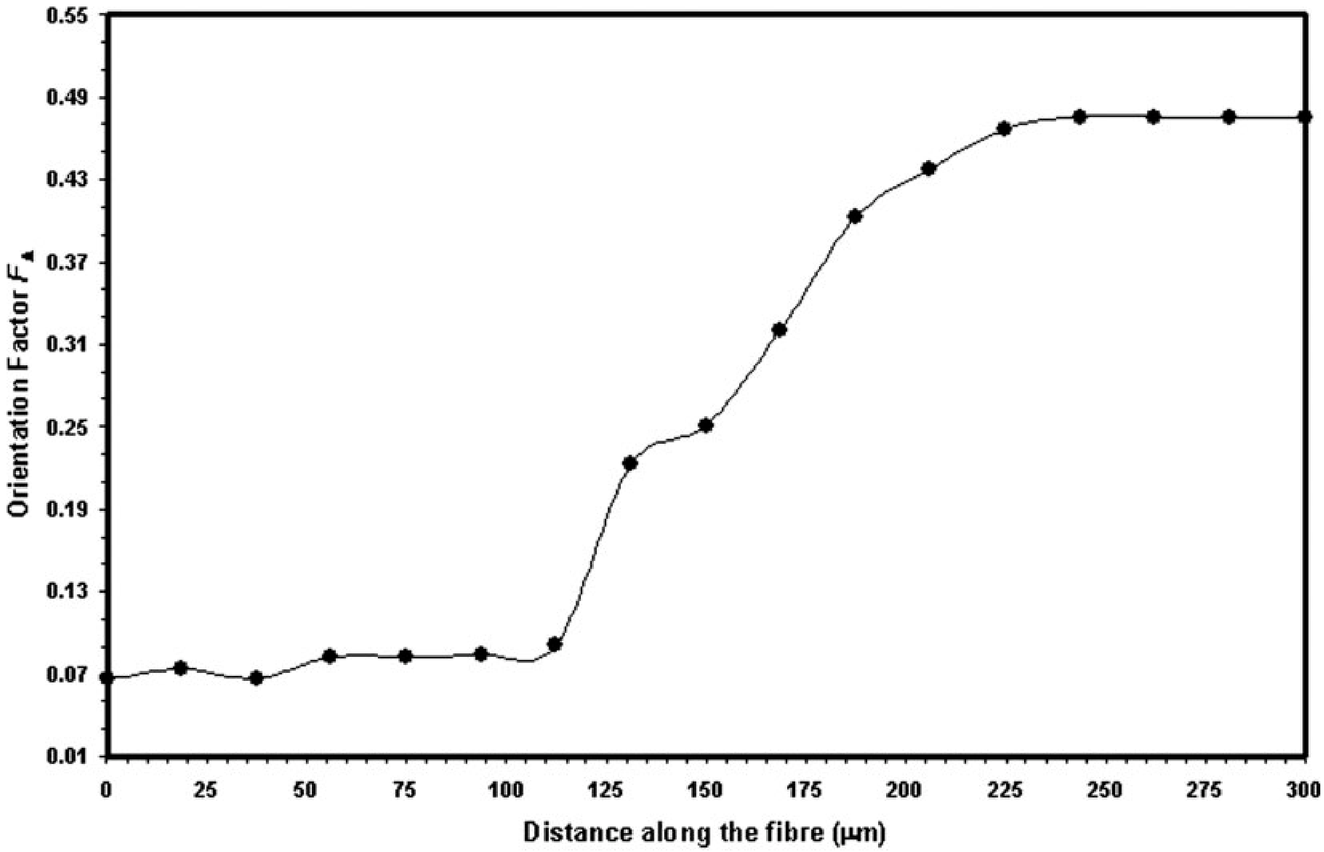


Figure 5 Variation of Herman's orientation factor along the axis of drawn PP fiber.

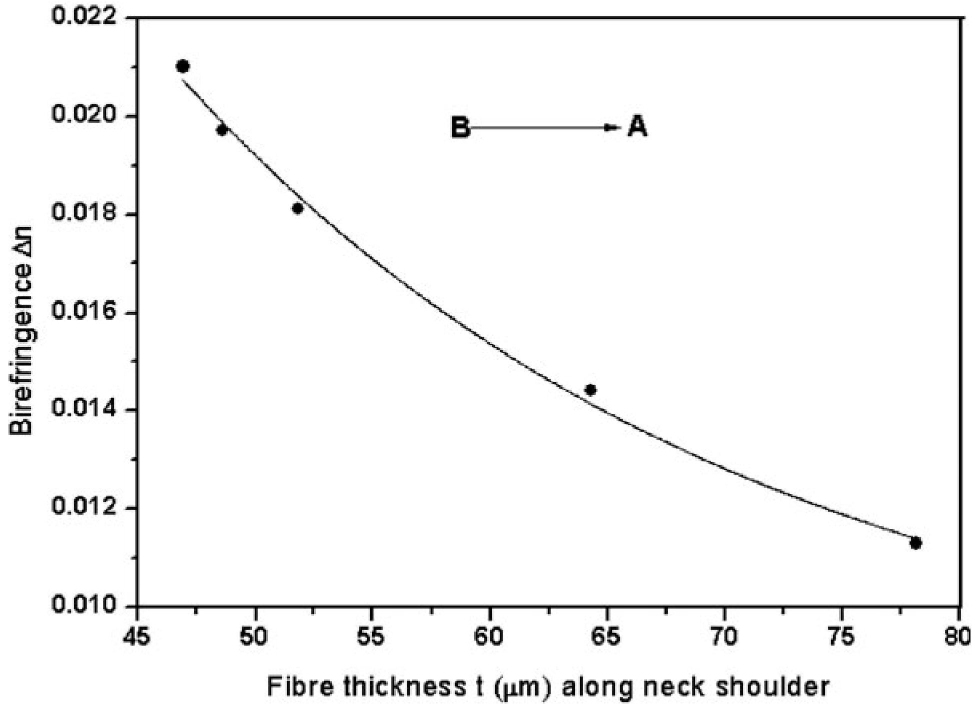


Figure 6 Variation of birefringence (Δn) against the thickness (t) along the neck shoulder inside PP fiber.

on the degree of orientation of molecules inside the fiber, have the same behavior of mean birefringence along the fiber. Also this confirms the effect of cold-drawn or necking deformation on the tested PP fiber.

According to the highly dependence of the birefringence of fibers on the fiber thickness, Figure 6 shows this relationship inside the neck region (AB) of PP fibers. The obtained graph represents the variation of thickness from end of neck shoulder B to its starting point A. The following equation is suggested to describe mathematically the relation between birefringence (Δn) and thickness (t) of the necked PP fibers.

$$\Delta n = \alpha + \beta \exp\left(-\frac{t}{\gamma}\right) \tag{4}$$

where $\alpha = 0.0078$, $\beta = 0.0888$, and $\gamma = 24.31$ are constants depending on the fiber material (PP). The accuracy in measuring the birefringence using the interference Pluta microscope is ± 0.001[7,8] and that for the thickness are ± 1 μm.[24] Now it is easier using this equation to know the birefringence at any point along the neck shoulder of the PP fibers.

CONCLUSION

In parallel to the well-known techniques such as SEM and X-ray, Interferometry still is a valuable tool for characterizing the structural deformation of polymer fibers. In comparison with the conventional methods, the used automated Pluta microscope provided with special software is considered as an imaging and analytical technique by which the necking phenomenon is well investigated.

In the course of cold drawing of polymer fibers, the necking deformation became an expected phenomenon. The birefringence of fiber is considered a micro-detector of the influence of necking deformation during the drawing of polymer fibers. The effect of necking is well confirmed by measuring the thickness, birefringence profile and orientation factor along the cold-drawn PP fiber. The suggested formula between the birefringence and thickness of the PP fiber is useful for the analytical interpretation of the neck shoulder.

Useful discussions with Prof. A. A. Hamza, Prof. T. Z. Sokkar, Prof. C. A. Lawrence, and Dr. M. Mahmoudi are grateful acknowledged.

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