Stress birefringence in compressible polymer fibres with irregular transverse sections

M. M. EL-TONSY

Physics Department, Faculty of Science, Mansoura University, Mansoura, Egypt

From an interferometric point of view a mathematical formula is developed for computing the exact optical path variations associated with a deformed cross-section of a fibre due to the drawing process. Fizeau fringes in transmission crossing Perlon fibres are obtained to illustrate the experimental applicability of the new corrected formula. The correction term is expressed as a function of the material compressibility, applied stress and the draw ratio. The birefringence of Perlon fibres is changed mechanically according to two different mechanisms in successive ranges of draw ratio. The mechanical response of Perlon fibres is found to be dependent on the direction with respect to the fibre axis. Another term is suggested to describe the mechanical anisotropy of a polymer. Microinterferograms are given for illustration.

1. Introduction

The utility of polymers in fibre form, both as textile materials and as reinforcing materials in various structures, is related to two interesting characteristics of polymeric fibres – the uniaxial orientation of the polymer molecule along the fibre axis, and their high surface-to-volume ratio.

The first characteristic in fibre orientation imparts the anisotropic character of the polymer to the fibre and controls the bulk properties of the fibre. These properties are tensile strength, tensile modulus, bending modulus, elongation, creep rate, moisture regain, melting point and thermal conductivity. The other characteristic is the high surface-to-volume ratio found in fibres, compared to bulk polymer. The surfaceto-volume ratio, of course, increases as the fibre thickness decreases [1].

Optical anisotropy produced in fibres by stretching gives valuable information for characterization of these fibres on the molecular level [2–8].

In recent years, interferometric determinations were used to determine the optical properties of both natural and man-made fibres. In the case of fibres with irregular transverse sections, the refractive indices and birefringence can be obtained interferometrically by using both two-beam and multiple-beam techniques [9-13]. The previously used relation [13]

$$n_{\rm a} = n_{\rm L} + (\lambda/2h) (F/A) \qquad (1)$$

where n_a is the mean refractive index of the fibre material, n_L the refractive index of the immersion liquid, F the area enclosed under the fringe shift, h the fringe interspacing, λ the wavelength of the monochromatic light used, and A the fibre cross-sectional area (magnification is taken into account), is inapplicable to fibres with irregular, or even regular, crosssections when they undergo a dynamical drawing process because of the undetectable deformation in both shape and area of the fibre cross-section. In the present work, a correction is introduced into Equation 1 in order to take into account the deformation produced in the cross-section of the fibre during the drawing process.

2. Theory

In order to calculate the deformed area of a fibre cross-section due to the drawing process, let V, A and l be volume, cross-sectional area and length, respectively, of the undrawn sample. If the material has a compressibility, K, and is drawn under tensile stress, P, deformation will appear. Let ΔV , ΔA and Δl be the produced changes in volume, cross-sectional area and length, respectively. Because the compressibility is defined as [14]

 $K = \Delta V/PV$

i.e.

$$KPV = (A - \Delta A) (l + \Delta l) - V$$
 (3)

then

$$(A - \Delta A) = A(PK + 1)/R \qquad (4)$$

where

$$R = (l + \Delta l)/l = \text{draw ratio}$$
(5)

Equation 4 gives the actual deformed cross-sectional area, $(A - \Delta A)$, of the fibre in terms of drawing stress, material compressibility and the produced draw ratio. Therefore, the drawing mechanical system should offer a direct measurement for drawing stress and draw ratio simultaneously, while the material compressibility should be independently determined.

In a special case, fibres made from rigid polymers possess a weak compressibility and have a short elongation at break. For such fibres, one may consider, as an approximation, that $PK \ll 1$, hence Equation 4 may be reduced to the form

$$(A - \Delta A) = A/R \tag{6}$$

Equation 6 can be applied with sufficient accuracy to

(2)



Figure 1 The modified stress-strain device. B, Spring balance; h_1 , h_2 , h_3 and h_4 , frictionless wheels; P, pointer; S, millimeter scale; F, optical plate; J, jig; e, threaded rod.

fibres whose maximum draw ratio at break does not exceed 1.6, at which the expected error in measuring the deformed area is below 2.3% of the undrawn (undeformed) cross-sectional area.

3. Results and discussion

The optical apparatus for producing multiple-beam Fizeau fringes in transmission for measuring the optomechanical behaviour of fibres was described in detail previously [8]. Fig. 1 shows the design of the previously used stress-strain device. Drawing the Perlon fibres shows that R (at break) = 1.422, i.e. $R_{max} < 1.6$. So, introducing Equation 6 into Equation 1 gives

$$n_{\rm a}^{||} = n_{\rm L} + (\lambda/2h) \left(F^{||}R/A\right)$$
 (7)

with an analogous formula for n_a^{\perp} .

Equation 7 can be applied, fairly, to interferograms of a Perlon fibre undergoing a successive deformation during the drawing process.

The undrawn cross-sectional area, A, of Perlon fibres is obtained optically, in Fig. 2, and it is found that $A = 708 \,\mu\text{m}^2$.

Fig. 3a to c are microinterferograms of multiplebeam Fizeau fringes in transmission for a Perlon fibre at draw ratios 1.044, 1.089 and 1.133, respectively. Monochromatic light of wavelength 546.1 nm vibrating parallel to the fibre axis was used. Using Equation 7, Fig. 4 shows the behaviour of $n_a^{||}$ for Perlon fibres when the draw ratio increases. It is clear that $n_a^{||}$ of Perlon fibres possesses a steady value as the draw ratio reaches 1.4 at 31 °C.

Fig. 5a to c are microinterferograms of multiplebeam Fizeau fringes in transmission for a Perlon fibre at draw ratios 1, 1.157 and 1.225, respectively. Monochromatic light of wavelength 546.1 nm and vibrating perpendicular to the fibre axis was used.



Figure 2 The optical view of a cross-section of undrawn Perlon fibres.

Fig. 6 shows the behaviour of n_a^{\perp} for Perlon fibres with the dynamic change of the draw ratio. It is clear that n_a^{\perp} is rapidly reduced to a steady value as the draw ratio reaches 1.4 at 31 °C.

The obtained values of n_a^{\parallel} and n_a^{\perp} are used to determine the birefringence $\Delta n_a = n_a^{\parallel} - n_a^{\perp}$ for Perlon fibres. Fig. 7 shows the variation of the birefringence Δn_a of Perlon fibres when they are drawn upwards. It

Apex of interferometer







Figure 3 Microinterferograms of multiple-beam Fizeau fringes in transmission crossing a Perlon fibre using monochromatic light vibrating parallel to the fibre axis at draw ratios (a) 1.044, (b) 1.089 and (c) 1.133.



Figure 4 The behaviour of $n_a^{||}$ on increasing the draw ratio for a Perlon fibre ($\lambda = 546.1$ nm, T = 31 °C).

is noticed that $\Delta n_a(R)$ changes with increased slope up to R = 1.2 and then the slope decreases gradually until the cut-off strain. The contrasting behaviour for slope of the function $\Delta n_a(R)$ may indicate an alternation in the alignment mechanism of the molecular chains.



Figure 5 Microinterferograms of multiple-beam Fizeau fringes in transmission crossing a Perlon fibre using monochromatic light vibrating perpendicular to the fibre axis at draw ratios (a) 1, (b) 1.157 and (c) 1.225.



Figure 6 The behaviour of n_a^{\perp} on increasing the draw ratio for a Perlon fibre ($\lambda = 546.1 \text{ nm}, T = 31 \,^{\circ}\text{C}$).



Figure 7 The variation of birefringence of Perlon fibres on increasing the draw ratio.

With respect to Figs 4 and 6, the maximum rates of variation of n_a^{\parallel} and n_a^{\perp} with increasing draw ratio R are $(dn_a^{\parallel}/dR) = 0.0245$ and $(dn_a^{\perp}/dR) = -0.0375$. The change in refractive index of a polymer, along a particular direction, corresponds to a similar change in density of structure along the same direction [15]. Thus the transverse response of molecular structure, in Perlon fibres, for the applied axial stress is, numerically, greater than its longitudinal response. In other words, one may consider that the mechanical chain flexibility is higher in the transverse direction than in the axial direction for Perlon fibres. Therefore, it is suggested that the factor $\Delta Y = (dn_a^{\parallel}/dR) + (dn_a^{\perp}/dR)$ indicates the mechanical anisotropy of a polymer.

Hannes [16] showed that the density of a fibre material is related to the change in the isotropic refractive index, δn_{iso} , where

$$n_{\rm iso} = (n^{||} + 2n^{\perp})/3$$
 (8)

Fig. 8 shows the variation of n_{iso} for Perlon fibres on increasing the draw ratio. From this curve one can find δn_{iso}

$$\delta n_{\rm iso}(R) = n_{\rm iso}(1) - n_{\rm iso}(R) \tag{9}$$

where $n_{iso}(1)$ and $n_{iso}(R)$ are the isotropic refractive



Figure 8 The variation of the isotropic refractive index, n_{iso} , of Perlon fibres undergoing dynamic drawing.

indices for the undrawn (R = 1) and drawn samples, respectively. Fig. 8 and Equation 9 show that $\delta n_{iso}(R)$ is always positive for Perlon, which means that the density of the fibre medium decreases as the draw ratio increases. Also it is clear that δn_{iso} possesses a maximum at R = 1.28, i.e. the molecular package of the material reaches a minimum at this draw ratio and then it increases again. Such behaviour of the molecular chain package may provide useful information about the shape of the polymer chains.

4. Conclusions

Most synthetic fibres are drawn several times their extruded length to give satisfactory properties. In some polymers the undrawn fibre is non-crystalline, and the drawing promoting orientation of the molecules is followed by crystallization [17]. In this work, the measuring technique, based on the multiple-beam Fizeau fringes, was corrected to take into account the produced deformations by drawing a fibre in order to determine fair results for the opto-mechanical properties of polymer fibres.

From the measurements carried out for Perlon fibres, the following conclusions were drawn.

1. The deformed cross-sectional area of a fibre during a drawing process was calculated in terms of the material compressibility and the applied tensile stress.

2. The used stress-strain device [8], Fig. 1, is recommended as a more suitable device for the determination of the opto-mechanical properties of fibres.

3. As the draw ratio increases the double refraction of Perlon fibres increases. This means that the inherent optical anisotropy of the chain-like macromolecules forming the preferred axial orientation of the molecular chains that constitute the fibre, increases.

4. It is clear that birefringence increases on drawing a fibre by two different mechanisms: (a) fast increase in $n^{||}$ while n^{\perp} is slowly decreasing; (b) fast decrease in n^{\perp} while $n^{||}$ is slowly increasing. Birefringence of Perlon fibres is found to increase on drawing, according to the two mechanisms in successive ranges of the draw ratio, Fig. 7.

5. The study of the rate of change of $n_a^{||}$ and n_a^{\perp} , with respect to the draw ratio, clarifies that the mechanical properties of the structure in a direction perpendicular

to the fibre axis differ from those in an axial direction, which is expected for an anisotropic medium.

6. The suggested factor $\Delta Y = (dn^{\parallel}/dR) + (dn^{\perp}/dR)$ clarifies the mechanical anisotropy of polymer fibres.

7. Changes in the isotropic refractive index δn_{iso} of Perlon fibres due to drawing indicate that the density of the medium decreases to a minimum and then increases before the cut off.

8. The higher the orientation, the more mutually parallel are the molecules, and the smaller is the average angle formed by them with the fibre axis [18].

9. The microinterferograms clearly identify differences in optical path variations in undrawn and drawn fibres.

From the above conclusions, the correction of the interferometric technique together with the modified stress-strain device provide a more accurate optomechanical view for polymer fibres, which is important not only for the fibre producers but also for physicists who are interested with the structure modelling of polymers. It appears that the analysis of Fizeau fringes crossing fibres of deformed cross-section requires further study going beyond the assumption that the fibre is a single medium.

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