Influence of temperature on the optical and structural properties along the diameter of: I. Polymer fibres

1999 J. Phys.: Condens. Matter 11 5331

(http://iopscience.iop.org/0953-8984/11/27/309)

J. Phys.: Condens. Matter **11** (1999) 5331–5341. Printed in the UK PII: S0953-8984(99)98102-X

Influence of temperature on the optical and structural properties along the diameter of: I. Polymer fibres

A A Hamza, T Z N Sokkar, K A El-Farahaty and H M El-Dessouky
Physics Department, Faculty of Science, Mansoura University, Mansoura 35516, Egypt
E-mail: hamzaaa@idsc.gov.eg

Received 5 October 1998, in final form 13 April 1999

Abstract. A modified heating device connected to a wedge interferometer is designed to study the influence of temperature on the optical and structural properties along the diameter of undrawn polypropylene fibres. To determine the optical orientation function profile, a model that divides the circular cross section into very small circular zones is suggested. The refractive indices and optical orientation function of each zone are considered assuming there is a constant fully oriented birefringence throughout the fibre. This investigation provides useful information about the structural properties of polypropylene fibres and throws light on its opto-thermal behaviour. The investigation is carried out using multiple-beam interference Fizeau fringes in transmission. Microinterferograms are given for illustration.

1. Introduction

It is now well established that the principal indices of refraction and the birefringence of fibres are a manifestation of many of their structural characteristics. Therefore, measuring these optical properties provides useful information about the processability of the fibre.

In polymeric fibre studies, attempts are considered to relate the molecular structure of a fibre to its thermal properties which have a direct effect on the structural parameters such as refractive index, polarizability per unit volume and orientation of the chain molecules or orientation function that measures the average angle between the chain axis and the fibre axis [1]. The orientation function for partially oriented fibres is evaluated by Kühn and Grün [2], followed by Ward [3] to obtain the relationship between the measured birefringence B and the maximum birefringence B_{max} of the ideal fibre [1–4].

The properties of polymer used in fibre reinforcement in tyres or belts or both exhibit different values when they are measured parallel and perpendicular to the fibre axis, e.g. thermal expansion and thermal conductivity [4].

Both double-beam and multiple-beam interferometries were applied to the measurement of the refractive indices and birefringence of both natural and synthetic fibres. These fibres are with regular or irregular transverse sections and of homogeneous or heterogeneous structure [5–12].

The two-beam interference microscope and multiple-beam Fizeau interference method are reviewed elsewhere [13]. Barakat and Hamza [14] made an extensive study of the interferometry of the fibrous materials. They considered in great detail the interferometric methods for the studying of optical, synthetic and natural fibres.

0953-8984/99/275331+11$30.00

Recently, Hamza *et al* [15] derived a mathematical expression for measuring the refractive index profile of the homogeneous fibres considering the refraction of the incident beam inside the fibre. They extended this expression for a skin–core fibre [16]. Also a mathematical expression for the refractive index profile of graded index fibres has been obtained by Hamza *et al* [17].

Investigation of the above mentioned optical properties of fibres at several different temperatures throws light on the opto-thermal behaviour of the fibre under study. Attempts have been made to study the effect of temperature on the refractive indices and birefringence of undrawn polypropylene fibres.

Faust [6] designed an opto-thermal device to study the variation of the refractive index of the fibre with variation of that of the surrounding liquid via the changing the temperature of the interferometric system instead of changing the wavelength of light used, for thick samples (more than 100 μm).

Hamza *et al* [18] studied the effect of temperature (15–31 $^\circ$C) on the optical properties of wool fibres, Fouda *et al* [19] on vestan (polyester) fibres and Hamza *et al* [20] on graded optical fibres, and they have suggested empirical relationships between refractive index, birefringence and temperature.

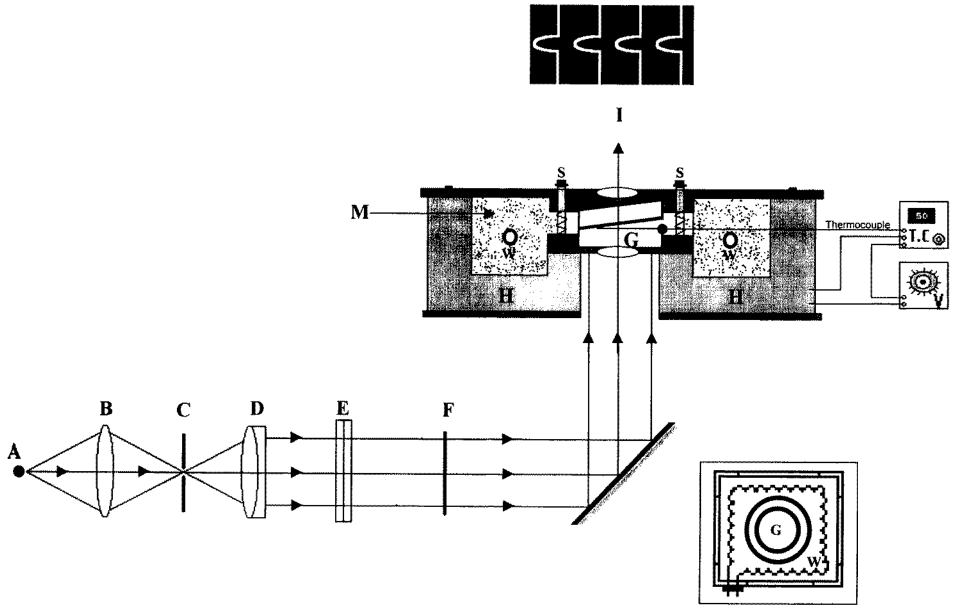
The present work reports a study of the variation of the principal refractive indices ($n^{\parallel}$ and $n^{\perp}$), the polarizability per unit volume ($p^{\parallel}$ and $p^{\perp}$), the birefringence B, the optical orientation function $f(\theta)$ and their profiles with temperature (from 20 up to 50 $^\circ$C) for undrawn polypropylene fibres. A model of circular zones that divide the multi-layer circular fibre cross section into circular zones of constant refractive indices and optical orientation function is assumed. For this purpose a modified heating device connected to a wedge interferometer is designed. The investigation was carried out using multiple-beam interference Fizeau fringes in transmission [16].

2. Experimental details

2.1. Design of the opto-thermal device

A Faust opto-thermal device [6] is modified to be suitable for measuring the variation of both the refractive indices ($n^{\parallel}$ and $n^{\perp}$) and the birefringence B of the fibre with temperature using multiple-beam Fizeau fringes in transmission as shown in figure 1(a). This modified device consists of a rectangular metallic chamber H (17.8 cm $\times$ 17.8 cm $\times$ 3 cm). At the centre of this chamber there is a wedge interferometer G. This interferometer consists of two circular optical flats 35 mm in diameter, 7 mm thickness and $\pm$0.01 mm flatness.

To produce multiple-beam Fizeau fringes in transmission, the inner surface of each flat is coated with a highly reflecting and partially transmitting silver layer. It reflectance is more than 75%, with transmittance of about 22%. The two-coated optical flats are then fixed in a special metallic jig to enable the adjustment of the interference fringes to cross the fibre perpendicular to its axis (cf [9]). There is an open circular window in the floor of the chamber and another in the corresponding roof for illuminating the interferometer. A heating coil W is built in a thermal refractory material M inside the lower part of the chamber, and surrounding the interferometer for raising the temperature of the system. The temperature is controlled by two components. One of them is a suitable temperature controller (T.C), which is added to modify the Faust device [6] and consists of a sensitive thermocouple and a digital recording element ranging from 0 up to 200 $^\circ$C with sensitivity $\pm$0.5 $^\circ$C. The other component is a varic V that has scaling range from 0 to 270 volts. The fibre under test is fixed with a suitable adhesive material at the edges of the lower optical flat. A drop of liquid was placed upon the



(a) (b)

Figure 1. (a) Cross section view of the opto-thermal device carried by the multiple-beam Fizeau system; it consists of H, the metallic chamber (heater), W, the heating wire, M, a thermal refractory material, G, the interferometer (jig), S, one of three screws, T.C, the temperature controller, and V, the varic (voltage). (b) Plane view of the heating device.

silvered face of the lower optical flat. At the wavelength (546.1 nm) used in the experiment and at room temperature the refractive index of the immersion liquid should be nearly the same as that of the tested fibre. Now the modified device can be transferred to the microscope stage without the roof. The interferometer, illuminated at normal incidence by a parallel beam of mercury green light, is then adjusted via means of three screws (S) which are added to the Faust device instead of the three wedged bars to form the sharpest fringes normal to the fibre axis. Then the device is effectively closed with the roof. The whole device is completely lagged by a thermal isolator to minimize the heat losses and at the same time to protect the microscope stage from heat. The interference pattern is recorded photographically at each value of the controlled temperature.

3. Theoretical considerations

3.1. Mean refractive indices ($n^{\parallel}$ and $n^{\perp}$) and birefringence B

The values of the mean refractive indices are calculated according to the analysis for the multiple-beam interference pattern [10]:

$$n^i = n_L + \frac{F^i \lambda}{2MAb} \tag{1}$$

where n_L is the refractive index of the immersion liquid, M is the magnification, A is the mean cross-sectional area of the fibre, b is the interfringe spacing, λ is the wavelength of monochromatic light used, i denotes the state of polarization of the light used ($\parallel$ parallel or $\perp$ perpendicular to the fibre axis) and F is the area enclosed under the fringe shift that is measured manually from the tracing of fringe shift profile. The uncertainty of δF along the fibre axis leads to uncertainty in measuring the refractive index δn that is given by

$$\delta n^i = \frac{\lambda}{2MAb}\delta F^i. \tag{2}$$

The mean birefringence of the fibre is given by

$$B = n^{\parallel} - n^{\perp} \tag{3}$$

where $n^{\parallel}$ and $n^{\perp}$ are the fibre mean refractive indices when using light vibrating parallel and perpendicular to the fibre axis, respectively.

3.2. Refractive index profile of the fibre

Hamza *et al* [17] calculated the refractive index profile of fibres taking into account the refraction of the incident beam by the fibre layers from the following recurrence relation:

$$\frac{\lambda Z_Q}{2b} = \sum_{j=1}^{Q-1} 2n_j[K_1 - K_2] + 2n_Q K_3 - n_0 K_4 \tag{4}$$

where K_1, K_2, K_3 and K_4 are given by

$$K_1 = \sqrt{[R-(j-1)a]^2 - (d_Q n_0/n_j)^2}$$
$$K_2 = \sqrt{(R-ja)^2 - (d_Q n_0/n_j)^2}$$
$$K_3 = \sqrt{[R-(Q-1)a]^2 - (d_Q n_0/n_Q)^2}$$
$$K_4 = \sqrt{R^2 - d_Q^2} + \sqrt{R^2 - X_Q^2}$$

where R is the fibre radius, a is the layer thickness ($a = R/Q$), Q is the number of layers, d_Q is the distance between the incident beam and the fibre centre, X_Q is the distance between the emerging beam and the fibre centre, Z_Q is the fringe shift corresponding to the point X_Q in the multiple-beam interference pattern, λ is the monochromatic wavelength and b is the fringe spacing.

3.3. The polarizability per unit volume

The values of mean polarizabilities per unit volume parallel ($p^{\parallel}$) and perpendicular ($p^{\perp}$) to the fibre axis are calculated using the Lorentz–Lorenz equation [21];

$$p^{\parallel} = \left(\frac{3}{4\pi}\right)\frac{n_{\parallel}^2 - 1}{n_{\parallel}^2 + 2} \tag{5}$$

and

$$p^{\perp} = \left(\frac{3}{4\pi}\right)\frac{n_{\perp}^2 - 1}{n_{\perp}^2 + 2} \tag{6}$$

where $p^{\parallel}$ and $p^{\perp}$ are previously mentioned.

3.4. The optical orientation function

Multiple-beam Fizeau fringes are used to study the effect of temperature on the molecular orientation of the polypropylene fibres. This orientation distribution function is given by [22, 23]

$$f(\theta) = (1+\chi) f_\Delta - \chi f_\Delta^2 \tag{7}$$

where χ is a constant and is given by

$$\chi = \frac{2n_\parallel^2 n_\perp^2}{n_v^3(n_\parallel + n_\perp)} - 1$$

and

$$f_\Delta = \frac{B}{B_{\text{max}}}$$

where n_v is the virtual refractive index, which nearly equal to the isotropic refractive index, and B and B_{max} are the measured and fully oriented birefringence, respectively; $B_{\text{max}} = 0.045$ for polypropylene fibres [22].

To determine the optical orientation profile of circular fibre cross section (optical orientation function along the fibre diameter), we assume a model of circular zones that divide the circular fibre cross section into very small circular zones. In this model the following assumptions must be taken into consideration; (i) each zone has constant refractive index and constant optical orientation function and (ii) the fibre has constant fully oriented birefringence along the fibre diameter.

4. Results and discussion

The investigated sample was undrawn polypropylene fibre. Monochromatic light of wavelength $\lambda = 546.1$ nm was used. The refractive index of the immersion liquid was 1.5020 at room-temperature 20 °C and its thermal coefficient is calculated to be -5×10^{-4} °C^{-1}.

Figures 2(a)–(c) are microinterferograms showing multiple-beam Fizeau fringes in transmission for a sample of polypropylene fibre at different temperatures, 29, 46 and 50 °C, respectively.

Figures 3(a)–(c) are microinterferograms for the same sample of fibre at temperatures 23, 26 and 50 °C, respectively, but for the other perpendicular component of the light vibration.

From these figures, it is obvious that if the temperature of the system is changed from room temperature to any elevated temperature, the fringe shifts in both directions are changed.

The mean refractive indices ($n^\parallel$ and $n^\perp$), the mean birefringence B and the mean polarizabilities ($p^\parallel$ and $p^\perp$) are calculated using equations (1), (3), (5) and (6) at different temperatures with the aid of the recorded interferograms. Also the uncertainty ($\delta n = \pm 0.000\,12$) is calculated from equation (2).

Figure 4 illustrates the variation of the mean refractive indices ($n^\parallel$ and $n^\perp$) and the mean birefringence B. It is found that the $n^\parallel$ and $n^\perp$ and B of the polypropylene fibre decrease linearly with increasing temperature T (°C) according to the following suggested relationship:

$$\alpha = \beta T + \gamma \tag{$*$}$$

where α means the refractive index (n_L or $n^\parallel$ and $n^\perp$), the birefringence B and the polarizability ($p^\parallel$ and $p^\perp$) of the fibre, β and γ are constants characterizing the opto-thermal properties of the fibre, since the parameter β represents the thermal coefficient for the optical properties n_L, $n^\parallel$, $n^\perp$, B, $p^\parallel$ and $p^\perp$. Both β and γ are tabulated in table 1.

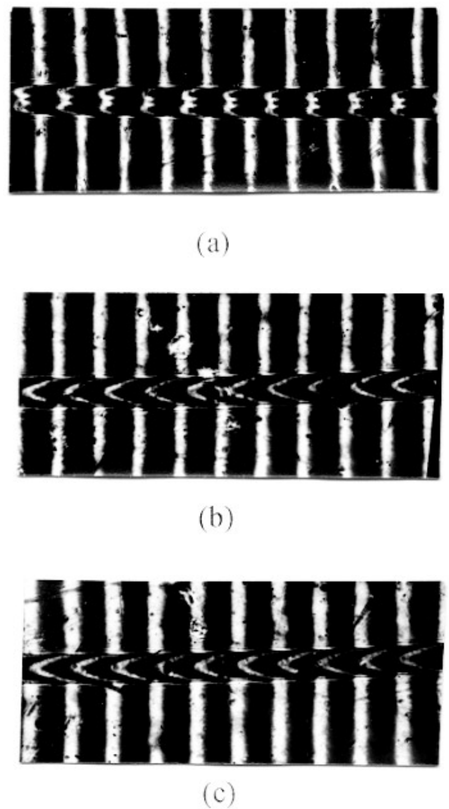


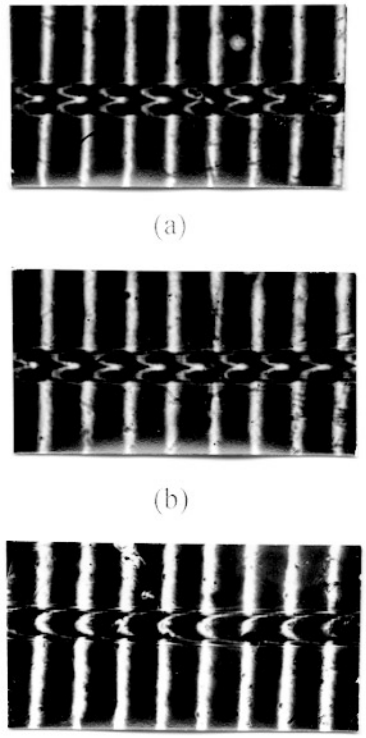
Figure 2. Microinterferograms showing the multiple-beam Fizeau fringes in transmission crossing the polypropylene fibre for light vibrating parallel to the fibre axis at $\lambda = 546.1$ nm at temperatures (a) 29, (b) 46 and (c) 50 °C.

Table 1. Constants β and γ for $n^{\parallel}$, $n^{\perp}$, B, $p^{\parallel}$ and $p^{\perp}$.

α	β	γ
$n^{\parallel}$	-9.4×10^{-4}	1.521 55
$n^{\perp}$	-9.0×10^{-4}	1.513 98
B	-4.0×10^{-5}	0.007 57
$p^{\parallel}$	-1.58×10^{-4}	0.094 78
$p^{\perp}$	-1.54×10^{-4}	0.093 80

The above optical properties $n^{\parallel}$, $n^{\perp}$, B, $p^{\parallel}$ and $p^{\perp}$ are mean optical parameters for polypropylene fibre. In order to obtain more useful information about the structural characteristics of polypropylene fibre, we study the influence of temperature on the optical and structural properties across the fibre diameter.

The refractive index profiles for $n^{\parallel}$ and $n^{\perp}$ are measured experimentally using a computer program based on the recurrence equation (3). Figures 5 and 6 show these profiles. It is obvious from these plots that the fibre under test has a skin–core structure. From the behaviour of these profiles it is found that these values across the fibre diameter decrease on increasing the temperature from room temperature up to 35 °C, but when the temperature increases gradually



(a) (b) (c)

Figure 3. Microinterferograms showing the multiple-beam Fizeau fringes in transmission crossing the polypropylene fibre for light vibrating perpendicular to the fibre axis at $\lambda = 546.1$ nm at temperatures (a) 23, (b) 26 and (c) 50 °C.

up to 50 °C these values are the same across the fibre diameter; this may suggest that the polypropylene fibre may be considered a homogeneous fibre at this temperature.

The analysis of the refractive index profiles ($n^{\parallel}$ and $n^{\perp}$) across the diameter, shown in figures 5 and 6, indicates that the birefringence ($n^{\parallel}$ and $n^{\perp}$) is negative in every zone for some temperatures. However in figure 4 it appears that the mean refractive index $n^{\parallel}$ is greater than $n^{\perp}$ at all temperatures, which gives positive birefringence. This discrepancy (between the observed mean birefringence which is positive and the values for the individual annuli which appeared generally to be negative) may be due to the very low values of birefringence for undrawn polypropylene fibre. Recently, we have been looking into this problem because it is probably relevant to measurements on all fibres of low birefringence.

The polarizability profiles for $p^{\parallel}$ and $p^{\perp}$ and the orientation function $f(\theta)$ is calculated from the values of previously measured refractive index and birefringence profiles and using the suggested zone model and equations (5)–(7). The polarizability and refractive index profiles are closely related, hence their profiles are similar. Figure 7 shows the orientation function profile at different temperatures from 20 up to 50 °C. It is clear that the influence of temperature on this function across the fibre diameter due to the increasing of temperature up to 34 °C is small. When the temperature increases up to 50 °C these values across the fibre diameter decrease and become nearly the same for one temperature. This result may lead us to conclude that the average angle between the chain axis and the fibre axis across the fibre diameter is

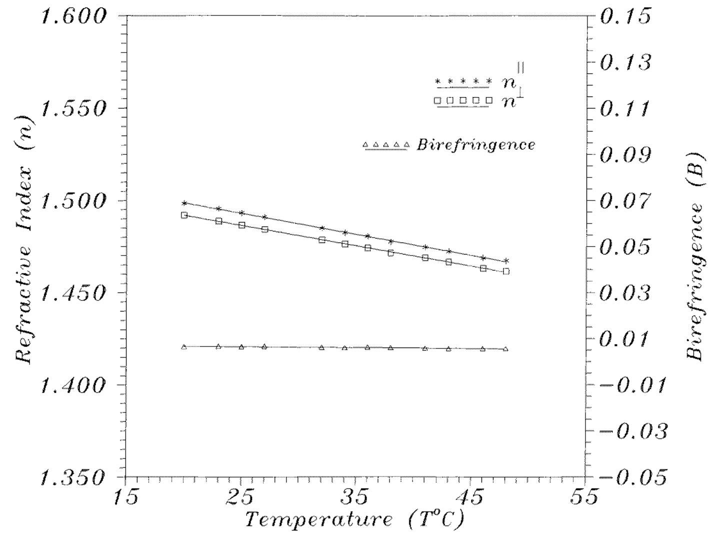


Figure 4. The relation between the temperature and the refractive indices $n^{\parallel}$, $n^{\perp}$ and birefringence B of polypropylene fibre.

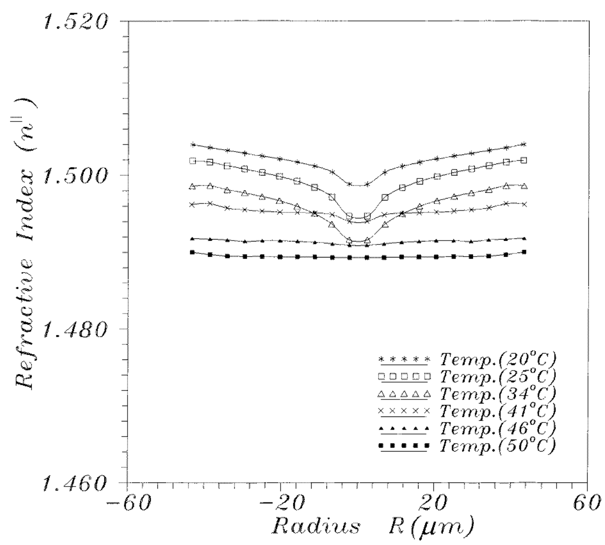


Figure 5. The refractive index profiles for $n^{\parallel}$ of polypropylene fibre at different temperatures.

increased. This means that the structural arrangement of molecules within the fibre is weakly arranged along the fibre axis. Hence the fibre may have weak mechanical properties at this temperature.

Figures 8 and 9 show the relation between temperature and refractive indices $n^{\parallel}$ and $n^{\perp}$, respectively. In each figure the value of refractive index is estimated at the middle of both of core and skin regions. It is obvious that the variation of refractive index with temperature in the core region is an exponential relationship (i.e. the core of the fibre has variable thermal coefficient), while this relationship in the skin region may be linear (i.e. the skin of the fibre

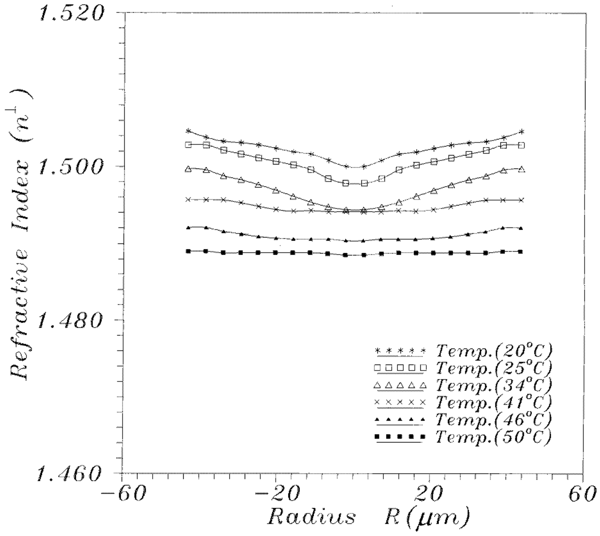


Figure 6. The refractive index profiles for $n^{\perp}$ of polypropylene fibre at different temperatures.

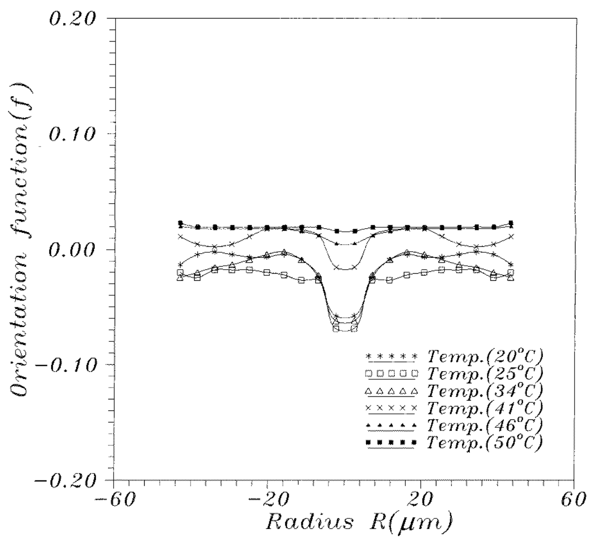


Figure 7. The orientation function $f(\theta)$ profile of polypropylene fibre at different temperatures.

has a constant thermal coefficient). The thermal coefficient is calculated in the skin region for both refractive indices from the figures 8 and 9 and is given as follows:

$$\mathrm{d}n^{\parallel}/\mathrm{d}T = -3.9 \times 10^{-4}\,^{\circ}\mathrm{C}^{-1} \qquad \mathrm{d}n^{\perp}/\mathrm{d}T = -3.7 \times 10^{-4}\,^{\circ}\mathrm{C}^{-1}.$$

5. Conclusion

The influence of temperature on optical and structural properties for undrawn polypropylene fibres is investigated. The variation of mean refractive indices, birefringence and polarizability

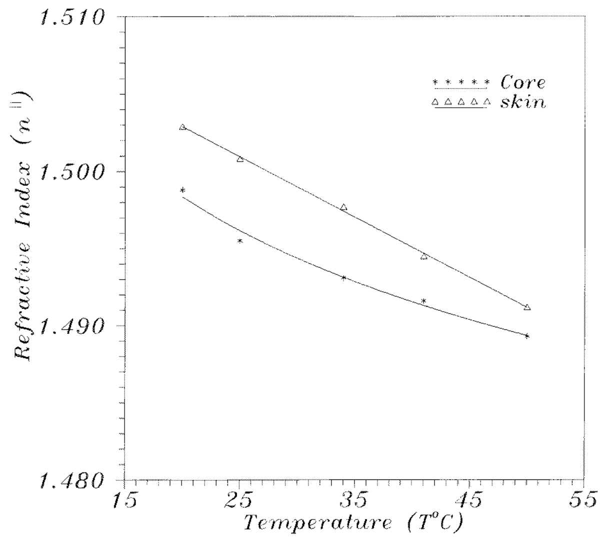


Figure 8. The relation between the temperature and the refractive index $n^{\parallel}$ at a point in the centre of both core and skin polypropylene fibre.

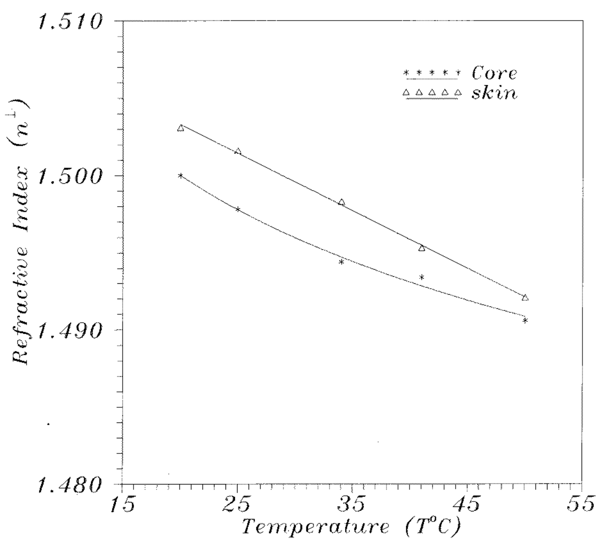


Figure 9. The relation between the temperature and the refractive index $n^{\perp}$ at a point in the centre of both core and skin of polypropylene fibre.

of the fibre due to relative temperature are detected. The results are in good agreement with the previous work [17, 18]. In order to obtain more useful information about the structural characteristics of the fibre, this investigation is carried out across the fibre diameter. A circular zone model is suggested to calculate the refractive index profile, birefringence profile and optical orientation function profile. Such information with increasing temperature may detect the structural variation of undrawn polypropylene fibre from the skin–core structure at room temperature to homogeneous fibre structure at nearly 50 °C. Also, it provides information about opto-thermal behaviour of the fibre in both core and skin regions.

In this work the undrawn polypropylene fibre is heated up to 50 °C via the designed heating device without any damage. This fibre may be heated up to more than 50 °C.

References

[1] Hartshorne N H and Stuart A 1970 4th edn (London: Arnold Co.) p 614
[2] Kuhn W and Grun F 1942 *Kolloid Z.* **101** 248
[3] Ward I M 1962 *Proc. Phys. Soc.* **80** 1176
[4] Fva R A 1980 *Polymers Part C: Physical Properties* (New York: Academic) p 137
[5] Faust R C 1952 *Proc. Phys. Soc.* B **65** 48
[6] Faust R C 1954 *Proc. Phys. Soc.* B **67** 138
[7] Simmens S C 1958 *Nature* **181** 1260
[8] Barakat N 1971 *Text. Res. J.* **41** 167
[9] Pluta M J 1972 *J. Microsc.* **96** 309
[10] Hamza A A, Sokkar T Z N and Kabeel M A 1985 *J. Phys. D: Appl. Phys.* **18** 1773
[11] Hamza A A, Sokkar T Z N and Kabeel M A 1985 *J. Phys. D: Appl. Phys.* **18** 2321
[12] Roche E G, Rubin P and Van Kavelaar R F 1987 *Text. Res. J.* **57** 371
[13] Hamza A A 1986 *J. Microsc.* **142** 35
[14] Barakat N and Hamza A A 1990 *Interferometry of Fibrous Materials* (Bristol: Hilger)
[15] Hamza A A, Sokkar T Z N and Ramadan W A 1992 *Pure Appl. Opt.* **1** 321
[16] Hamza A A, Sokkar T Z N, Ghander A M, Mabrouk M A and Ramadan W A 1994 *Pure Appl. Opt.* **3** 943
[17] Hamza A A, Sokkar T Z N, Ghander A M, Mabrouk M A and Ramadan W A 1995 *Pure Appl. Opt.* **4** 161
[18] Hamza A A, Fouda I M, El-Farahaty K A and El-Sayed K A 1988 *Acta. Phys. Pol.* A **73** 767
[19] Fouda I M, El-Tonsy M M and Oraby A H 1990 *J. Mater. Sci.* **25** 1416
[20] Hamza A A and Mabrouk M A 1991 *J. Mod. Opt.* **38** 97
[21] Lorentz H A 1952 *Ann. Phys., NY* **9** 641
[22] de Vries H 1979 *Colloid Polym. Sci.* **257** 226
[23] Bower D I 1981 *Polym. Sci. Polym. Phys.* **19** 93