A COMPARATIVE STUDY OF THREE METHODS USED FOR THE MEASUREMENT OF REFRACTIVE INDICES AND BIREFRINGENCE OF POLYMER FIBRES

By A. A. Hamza, I. M. Fouda and A. H. Hashish

Physics Department, Faculty of Science, Mansoura University*

(Received October 3, 1979; revised version received February 21, 1980)

Three different methods namely, the Becke-line method, the application of multiple-beam Fizeau fringes in transmission and the Pluta polarizing interference microscope have been used to measure the refractive indices for light vibrating parallel and perpendicular to the axis of samples of polyvinyl alcohol fibres. The birefringence of these fibres were also determined. The applicability and advantages of each method have been discussed from the point of view of fibre microscopy. Illustrations were made using photomicrographs.

1. *Introduction*

The optical properties of fibres like refractive index and birefringence are useful source of information about their structure. Measurements of the refractive indices of each layer of the fibre give an indication to the density of packing of molecules. Birefringence provides a measure of the degree of orientation of molecular chains.

The Becke-line method [1] was used to measure the refractive indices and birefringence of different fibres. Both the double beam and multiple-beam interference techniques have been used by Faust [2–4] in order to determine refractive index variations within optically heterogeneous specimens. Some papers [5–8] present the usefulness of the interference microscopy in the measurement of the optical properties of polymeric fibres.

Barakat and Hindeleh [9] used multiple-beam interferometric methods for determination of refractive indices and birefringence of normal viscose rayon fibres. Barakat and El-Hennawi [10] used these methods for the studying of the optical properties of Acrilic fibres.

Pluta [11, 12] developed and described the use of a double refracting interference microscope with variable amount and direction of wave front shear for the study of synthetic polymer fibres. Using the Pluta microscope, Hamza and Sikorski [13] studied the optical anisotropy of poly (p-phenylene terphthalamide) fibres.

* Address: Physics Department, Faculty of Sciences, Mansoura University, Mansoura, Egypt.

In this work, three different methods have been used to measured the refractive indices and birefringence of polyvinyl alcohol (P.V.A.) fibres. These are:

1. The Becke-line method, to measure the refractive index of the outer layer of the fibre for light vibrating parallel and perpendicular to its axis.

2. The application of multiple-beam Fizeau fringes in transmission to measure the mean refractive index, birefringence and the variation of the refractive index along the axis of the fibre.

3. The Pluta microscope [11, 12] to measure the mean refractive indices of fibres for light vibrating parallel and perpendicular to the fibre axis. The birefringence was calculated indirectly from the refractive indices and was measured directly from a non-duplicated image of the fibre.

Applicability and advantages of every method have been studied.

2. *Experimental procedure and results*

Determination of refractive indices and birefringence of the skin of P.V.A. fibres by the Becke-line method

The Becke-line method was discussed elsewhere [1]. In this work a series of liquids with refractive index increasing in steps of approximately 0.0004 was prepared by mixing different volumes of α-bromonaphthalene ($n_D = 1.658$ at 20°C) and liquid paraffin

TABLE 1

Refractive indices and birefringence of P.V.A. fibre's skin measured by the Becke-line method*

$n_s^{\|\|}$	$n_s^{\perp}$	Δ_s
1.581	1.543	0.038

* This method is repeated with known liquids until the nearest approximation to the refractive index is obtained. By this method, values can be measured with an accuracy of ± 0.0005 [14].

($n_D = 1.481$ at 20°C). Table I gives the refractive indices $n_s^{||}$ and $n_s^{\perp}$ of P.V.A. fibre's skin and the birefringence Δn_s measured by the Becke-line method at 27°C by using monochromatic light of wavelength 546.1 nm.

Measurement of the mean refractive indices and birefringence fibres using multiple-beam Fizeau fringes in transmission

The optical set-up for forming Fizeau fringes in transmission were used. The wedge interferometer consists of two circular optical flats 3.5 cm in diameter, 0.7 cm thick, and flat to ± 0.01 micron. The inner surface of each flat was coated with a silver layer of reflec-

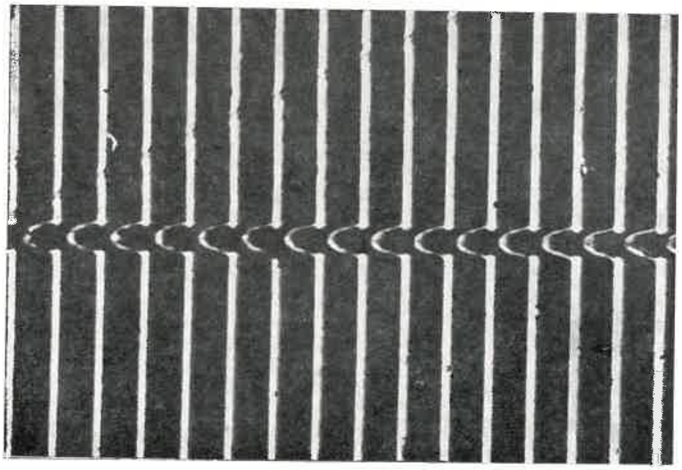


Plate 1 shows multiple-beam Fizeau fringes in transmission for light vibrating perpendicular to the fibre axis for a sample of P.V.A. fibre

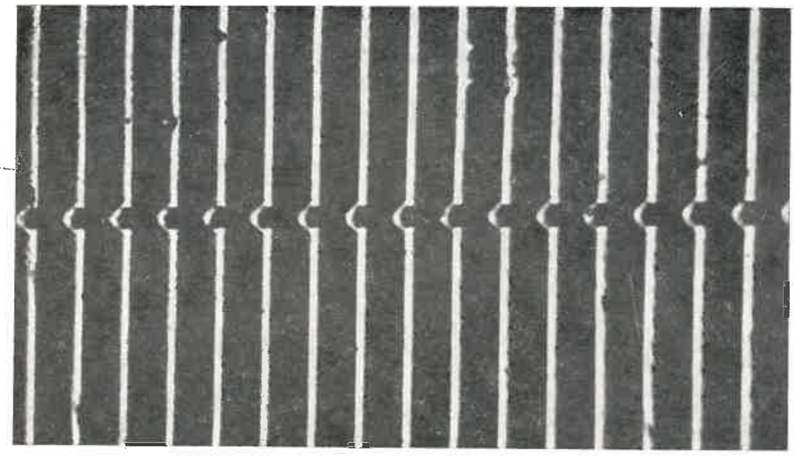
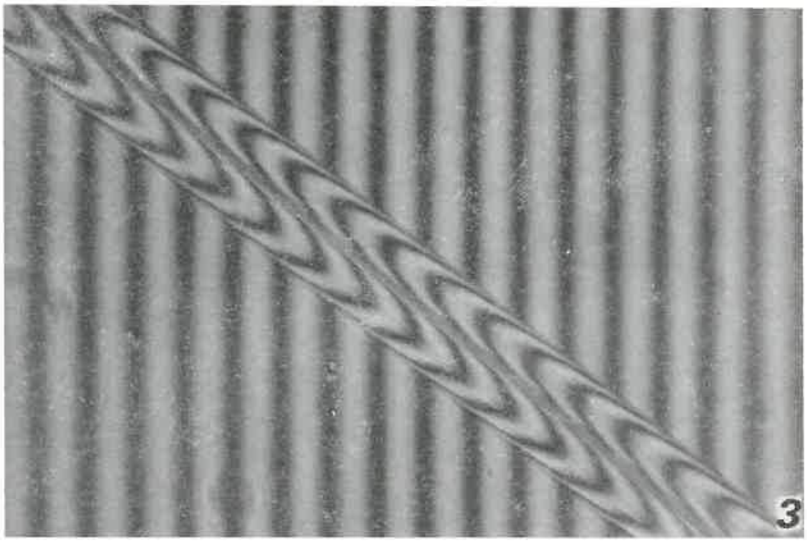
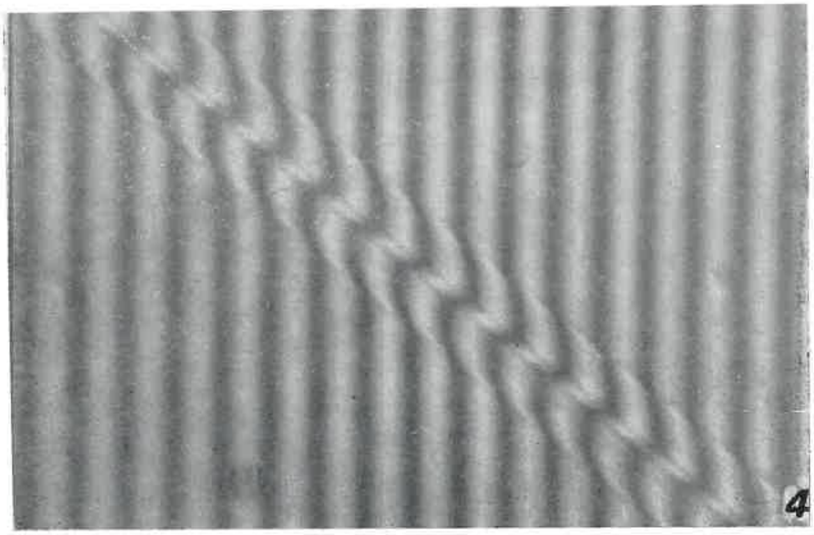


Plate 2 shows multiple-beam Fizeau Fringes in transmission for light vibrating perpendicular to the fibre axis of another sample of P.V.A. fibre





Plates 3, 4 show duplicated images of P.V.A. fibres

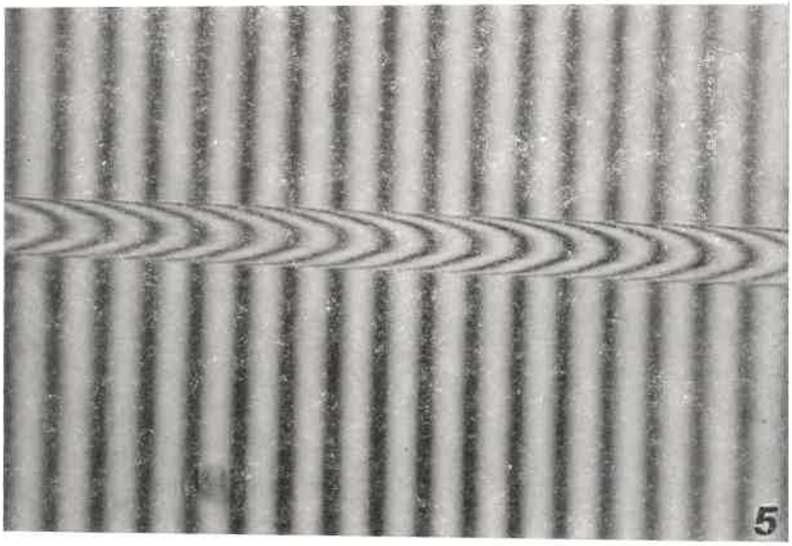


Plate 5 shows differentially sheared image of P.V.A. fibre tivity more than 75% with transmission of about 22%. The two flats were put in a special "Jig".

The silver coat was prepared by thermal evaporation of spec-pure silver in a vacuum better than 10^{-4} Torr. A drop of liquid with refractive index close to that of the fibre's skin was put on the silvered face of the lower optical flat. Mixtures of α-bromonaphthalene and paraffin were used as immersion liquids. The fibre was immersed in the liquid and the upper optical flat was then introduced to form the interferometer. The "Jig" was so adjusted such that the liquid fringes were normal to the fibre. A polaroid was inserted in the path of the monochromatic beam to allow the incident beam of light to vibrate either parallel or perpendicular to the fibre axis. As the fringes crossed the fibre they suffered a shift whose amount and shape depends on the difference between the refractive index of the liquid n_L and the mean refractive index of the fibre n_a.

By determining the value of the fringe shift dz as a fraction of an order separation Δz between two consecutive straightline fringes, the value of the mean refractive index of the fibre was calculated from the relation

$$\frac{dz}{\Delta z} \cdot \frac{\lambda}{2t_f} = (n_a - n_L), \tag{1}$$

where t_f — the fibre thickness, and λ — the wavelength of light.

Plate 1 shows multiple-beam Fizeau fringes in transmission for light, of wavelength 546.1 nm, vibrating perpendicular to the fibre axis for a sample of P.V.A. fibre.

The average value of the mean refractive index for light vibrating parallel to the P.V.A. fibre axis was found to be 1.576 at 20°C, whilst, that for light vibrating perpendicular to the fibre axis was found to be 1.543 at the same temperature. Therefore, the mean birefringence Δn_a will be equal to 0.033.

Variation of refractive index along the axis of P.V.A. fibres

Plate 2 shows multiple-beam Fizeau fringes in transmission for light vibrating perpendicular to the fibre axis for a P.V.A. fibre sample. n_L being 1.5415 at 26.5°C and $\lambda = 546.1$ nm. The shift was found to be towards the axes of the wedge, i.e. $n_L > n_a^{\perp}$.

It was found that there were no observable changes of the refractive index along the axis of the fibre, as indicated in Table II.

TABLE II

Refractive indices along the fibre axis

The distance in microns from certain point	$n_a^{\perp}$
0.00	1.539
8.00	1.539
17.54	1.539
29.13	1.539
40.30	1.539
45.64	1.539
50.00	1.539

Measurement of the mean refractive indices and birefringence of fibres using the Pluta microscope

This microscope incorporates a polarising interference system consisting of a birefringent prism (a special kind of the Wollaston prism), polarizer, analyser and a slit diaphragm.

The function of the birefringent prism consists basically in the splitting (bifurcating) of incident light beam into two beams viz. an ordinary and an extraordinary beam, and in producing an adequate phase shift between light beams so split.

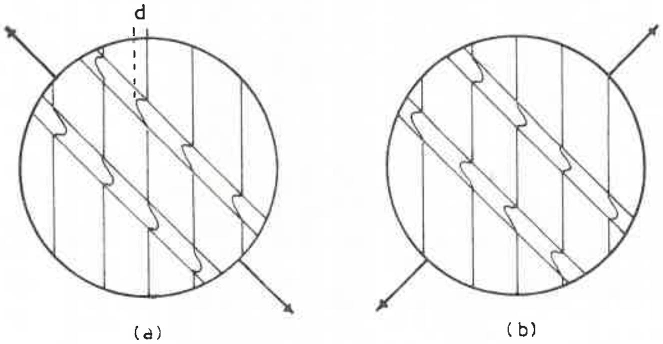


Fig. 1. Totally duplicated images of the fibre with light vibrating (a) parallel (b) perpendicular to the fibre axis

If the fibre is immersed in a liquid having refractive index of a value n_L which lie between the values of the refractive indices of the fibre $n^{||}$ and $n_{\perp}$, the direction of the fringe displacement becomes reversed as shown in Figs. 1a and b. The optical path difference Φ can be written as:

$$\Phi^{\perp} = \frac{d^{\perp}}{h'} \cdot \lambda = (n^{\perp} + n_L)t, \tag{2}$$

and

$$\Phi^{||} = \frac{d^{||}}{h'} \cdot \lambda = (n^{||} - n_L)t, \tag{3}$$

where d is a fringe shift inside the fibre (deviation of the interference fringes), h' — inter-fringe spacing, λ — applied light wavelength, n — refractive index of the fibre, n_L — refractive index of the immersion liquid, t — fibre thickness. Therefore,

$$n^{||} = \frac{d^{||}}{h'} \cdot \frac{\lambda}{t} + n_L, \tag{4}$$

$$n^{\perp} = \frac{d^{\perp}}{h'} \cdot \frac{\lambda}{t} + n_L, \tag{5}$$

and

$$\Delta n = n'' - n^{\perp} = \left(\frac{d^{||} - d^{\perp}}{h}\right) \frac{\lambda}{t}. \tag{6}$$

The direction of the fringe displacement, across the fibre image depends on the relative values of the refractive indices of the fibre and that of the immersion liquid [13].

The birefringence, Δn, could be measured directly from the differentially sheared (non-duplicated) image as indicated in Figs. 2a and b. It gives also an indication about

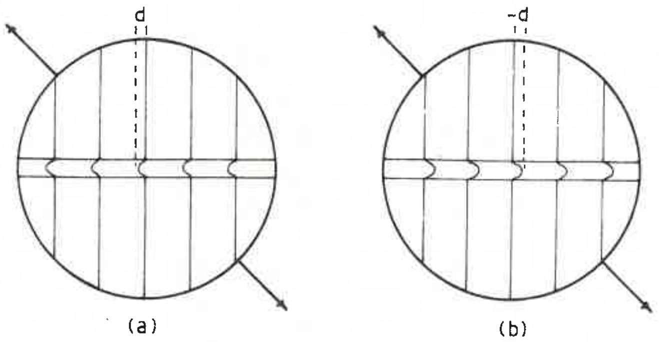


Fig. 2. Differentially sheared images of a fibre of (a) positive birefringence and (b) negative birefringence

the sign of the birefringence. The optical path difference in this case can be written as follows:

$$\Phi = \frac{d}{h'} \cdot \lambda, \tag{7}$$

where λ is the wavelength of light. Therefore,

$$\Delta n = \frac{\Phi}{t} = \frac{d}{h'} \cdot \frac{\lambda}{t}, \tag{8}$$

where t is the fibre thickness.

Plates 3,4 and 5 show typical microinterferograms obtained with P.V.A. fibres.

TABLE III

The measured values of refractive indices and birefringence of P.V.A. fibres using the Pluta microscope

n_L	λ* [nm]	t [μm]	$\frac{d^{\parallel}}{h'}$	$\frac{d^{\perp}}{h'}$	$\frac{d}{h'}$	$n^{\parallel}$	$n^{\perp}$	$n^{\parallel}-n^{\perp}$	Δn from differentially sheared image
1.536	550	27.7	$\frac{0.29}{0.19}$	$\frac{0.07}{0.19}$	$\frac{0.36}{0.19}$	1.566	1.529	0.037	0.037

* Taken as an average of white light.

The mean values for the refractive indices $n^{||}$ and $n^{\perp}$ and birefringence, Δn, obtained on twenty fibres using the Pluta microscope are shown in Table III. This table gives also the values of the birefringence calculated directly from the differentially sheared (single) image in the Pluta microscope.

3. *Discussion*

For homogeneous fibres, the Becke-line method and the interferometric methods (application of multiple-beam Fizeau fringes in transmission and the Pluta microscope) can be used to measure their refractive indices, whilst for heterogeneous fibres the interferometric methods provide the more appropriate information. Also the interferometric method enables one to measure the refractive indices for light vibrating parallel and perpendicular to the fibre axis on the same fibre, whilst in the Becke line method, the two refractive indices cannot be measured on the same fibre.

The interferometric methods enable one to measure the variation of refractive index along the axis of the fibre and the refractive index of every layer of heterogeneous fibres. The Becke-line method cannot be used to do so.

When comparing between the method of application of multiple-beam Fizeau fringes and the Pluta microscope, the latter presents a quick and easy method to measure the mean refractive indices and the mean birefringence without the preparation of an interferometer for each measurement. It is capable of giving both the uniform and fringe interference fields with continuously varied amount and direction of lateral image duplication. The Pluta microscope offers a particularly reliable and convenient method of tracing and measuring the magnitude of the fringe displacement within the fibre image, by transverse traverse of the (normal) Wollaston prism by means of a micrometer [12], without any movement of the sample. This is useful in the case where there is a big difference between the refractive indices of the fibre and the immersion liquid.

Multiple-beam fringes are often extremely sharp. Under good conditions the multiple--beam system is much more sensitive than the two beam one. However, since the light passes through the specimen many times, the phase relationships within the object are not accurately portrayed as with the two-beam system. The magnitude of this phase distortion will depend upon the fineness of the structure within the object and upon the thickness of the object.

The accuracy and limitations of the method of multiple-beam fringes is as follows:

a) A fibre of a thickness less than about 0.1 mm can be studied.

b) An accuracy in the measurements of refractive indices of within ± 0.0001 is possible under favourable circumstances.

c) A small interferometric gap ensures that the image faithfully portrays the structure of the specimen.

d) Any irregularities in the surfaces of the interferometer plates will lead to their own fringe displacements.

Concerning the use of the Pluta microscope, accuracy of the measurement of the optical difference, with a fringe field (normal Wollaston Prism), is about 0.05. Therefore, the error in the assessment of the value of refractive indices and birefringence cannot be better than 0.003–0.001; as in our case where the diameter of the fibre is about 30 μm, and determined with an accuracy of about 1 μm [12].

The authors would like to thank Prof. Dr. Y. K. M. Badawy and Prof. Dr. M. D. Migahed, Physics Department, Mansoura University for their useful discussions. One of us (A.A.H.) wishes to thank Dr. J. Sikorski, Textile Physics Laboratory, University of Leeds, for his great help and useful discussions.

REFERENCES

[1] R. C. Faust, *Physical Methods of Investigating Textiles*, edited by R. Meredith and J. W. S. Hearle, Textile Book Publishers, Inc., New York 1959, p. 320.
[2] R. C. Faust, *Proc. Phys. Soc.* **B55**, 48 (1952).
[3] R. C. Faust, *Proc. Phys. Soc.* **B67**, 138 (1954).
[4] R. C. Faust, *Quart. J. Micr. Sci.* **97**, 569 (1956).
[5] A. N. J. Heyn, *Text. Res. J.* **27**, 449 (1957).
[6] V. G. Peck, W. L. Carter, *Text. Res. J.* **34**, 40 (1964).
[7] F. D. Poling, N. R. S. Hollis, *J. Microsc.* **16**, 201 (1968).
[8] S. C. Simmens, *J. Microsc.* **89**, 291 (1969).
[9] N. Barakat, A. M. Hindeleh, *Text. Res. J.* **34**, 581 (1964).
[10] N. Barakat, H. A. El-Hennawi, Part 1, *Text. Res. J.* **41**, 391 (1971).
[11] M. Pluta, *Optica Acta* **18**, 661 (1971).
[12] M. Pluta, *J. Microsc.* **96**, 309 (1972).
[13] A. A. Hamza, J. Sikorski, *J. Microsc.* **113**, 15 (1978).
[14] F. Happey, *Applied Fibre Science*, Vol. 1, Academic Press, London 1978, p. 69–155.