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Optical properties of vinyon copolymer fibre

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Birefringence for vinyon fibres have been investigated. The principal refractive index parallel and perpendicular to the stretch direction were found. Double-beam and multiple-beam interferometry methods were applied. The double-beam technique was applied to the mean birefringence and the mean refractive indices for plane-polarized light parallel and perpendicular to the fibre axis. The diffraction of a He–Ne laser beam was used to measure the dimensional parameters, transverse sectional shape, and area of the fibre. The fibre area was used to calculate the principal refractive indices. The Becke-line method was used to measure the refractive indices of the outer layer of the fibre (skin) for polarized light parallel and perpendicular to the fibre axis. The results were found to be in good agreement with published data.

1. Introduction

The preparation of multifilament drawn yarn generally proceeds in three stages (1) spinning of the fibre from a melt (2) drawing the spun fibre, and (3) heat setting of the drawn fibre under tension. The spun fibre was produced by forcing a melt through a small die hole and keeping it under tension while it was air quenched. A combination of the normal forces in the swollen liquid at the die and the tension maintained during the air quench caused the resulting spun fibre to be oriented, as is attested by its birefringence. Birefringence is a measure of the total molecular orientation of a system. The refractive index, in turn, is a measure of the velocity of light in the medium and is related to the polarizability of the chains. Thus the final measured birefringence is a function of contributions from the polarizabilities of all the molecular units in the sample [1].

Thus optical properties of fibres can give a clear indication of the structural characteristics of these fibres. The values of refractive indices and birefringence for each layer of the fibre are useful in this respect. One of the serious problems in fibre science is the accurate measurement of these values.

Different techniques of double-beam and multiple-beam microinterferometry are given, together with their application in the investigation of fibre properties. The use of the polarizing microscope in this respect is also given [2]. In this paper double-beam and multiple-beam interferometric methods were applied to determine the mean refractive indices and birefringence for each layer of the vinyon fibre (a vinyl chloride-vinylacetate copolymer) having two layers (a core surrounded by a skin). Also the fibre diameter and the geometry of the transverse section were determined from the diffraction profiles of single fibres.

2. Theoretical considerations

2.1. Determination of the fibre diameter

To determine the fibre diameter [3] the following formula is used

$$d = \pm \lambda l / x \tag{1}$$

where d is the fibre thickness, $\lambda = 632.8\,\text{nm}$ the wavelength of the He–Ne laser used, x the distance from the centre of the pattern to the first minimum and l the distance between the fibre and the screen on which the diffraction pattern is produced.

2.2. Determination of the refractive index

For the determination of the refractive index using multiple-beam Fizeau fringes the following formulae are used.

(i) For a circular multiple skin [4]

$$\frac{\mathrm{d}z}{h} = \frac{4}{\lambda}\sum_{k=0}^{m} A_k \tag{2}$$

where dz is the value of the fringe shift, h the distance between any two consecutive straight-line fringes, λ the wavelength of monochromatic light used and

$$A_k = (n_k - n_{k+1})\,(r_k^2 - x^2)^{1/2}$$

where n_k is the refractive index of the layer k of the fibre, n_{k+1} is the refractive index of the medium in which the fibre is immersed, r_k is the radius of the skin of order k from, the core, and x is the distance measured from the centre of the fringe shift at which the fringe shift tends to zero.

(ii) The mean refractive index, n_a of the fibre, having a core of n_c surrounded by a skin with n_s was determined using the following formula [5]

$$n_a = n_c \frac{t_c}{t_f} + n_s \frac{t_s}{t_f} \tag{3}$$

where t_f, t_c and t_s are the thickness of the fibre, core and skin, respectively.

(iii) For multiple-beam Fizeau fringes considering the area enclosed under the fringe shift we use the following formula [6]

$$n_a = n_L + \frac{F}{h}\frac{\lambda}{2MA} \tag{4}$$

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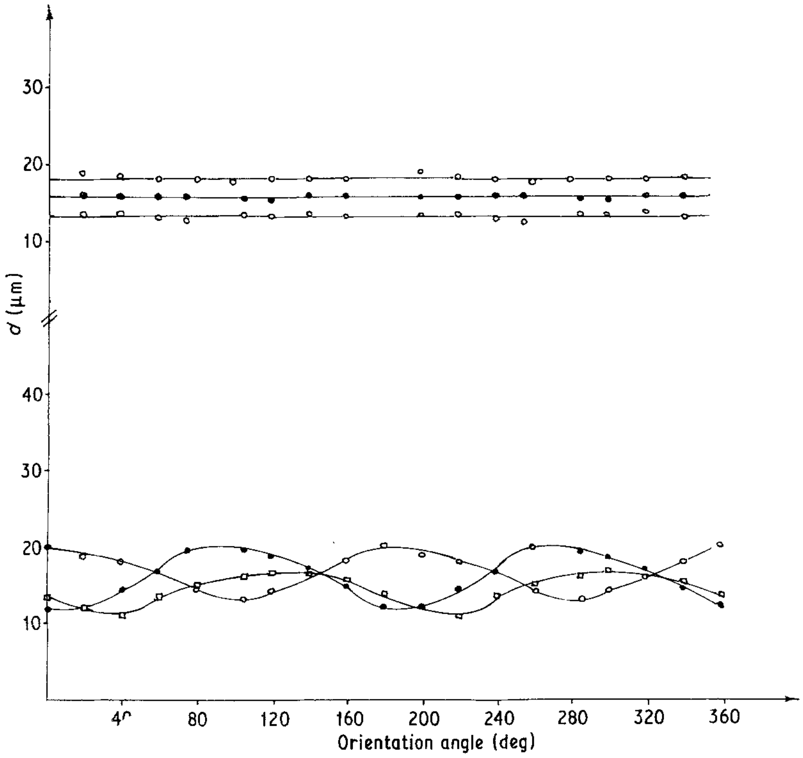

Figure 1 Radius of different angular orientations of vinyon fibres.

where F is the area enclosed under the fringe shift, A is the mean cross-sectional area M the magnification and n_a, n_L and λ are as previously defined.

2.3. Determination of the mean refractive indices and birefringence

For the determination of the mean refractive indices and birefringence the Pluta [7, 8] polarizing interference microscope was used for the double-beam technique and the following formulae [9] were applied.

$$n_a^{\parallel} = n_L + \frac{F^{\parallel}}{h}\frac{\lambda}{MA}$$

$$n_a^{\perp} = n_L + \frac{F^{\perp}}{h}\frac{\lambda}{MA} \tag{5}$$

$$\Delta n = \frac{F^{\parallel} - F^{\perp}}{h}\frac{\lambda}{MA} \tag{6}$$

where $n_a^{\parallel}$ and $n_a^{\perp}$ are the mean refractive indices of the fibre for light vibrating parallel and perpendicular respectively to the fibre axis, Δn_a is the mean birefringence and M the magnification.

3. Experimental results

We used the methods discussed in previous work to produce optical diffraction profiles [3], Beke-line method [11], Pluta microscope [8, 9], and multiple-beam Fizeau fringes in transmission and in reflection [5].

3.1. Cross-sectional parameters of vinyon fibres

About 25 fibres were used for diffraction measurements. The mean values of the experimental results for the diameter and the ellipticity of the transverse sections for vinyon fibres are given in Table I.

Fig. 1 shows the relation between the diameter and the orientation angle with respect to the fibre axis for each fibre. It is clear from these figures that some of vinyon fibres give straight lines and some show wave character curves. These data have been plotted to determine graphically the shape of the fibre cross-section by plotting the fibre-radius vector against the angle of rotation of the fibre around the centre. Fig. 2 shows the experimental results pertaining to the shape of the transverse sections of vinyon fibres. From Figs 1 and 2 it is clear that some fibres have nearly circular cross-sections and others have irregular shape cross-sections.

The mean value of the transverse-sectional areas measured from Fig. 2 are also given in Table I.

3.2. Becke-line method

We have used the Becke-line method to determine the refractive indices $n_s^{\parallel}$ and $n_s^{\perp}$ of the skin of vinyon fibres for light vibrating parallel and perpendicular to the fibre axis, respectively. Birefringence of the fibres Δn_s were calculated, and results are given in Table II. The wavelength of the light used was 546.1 nm.

TABLE I Geometrical parameters of vinyon fibres measured by the diffraction method

Average mean diameter (μm)	Maximum mean diameter (a) (μm)	Minimum mean diameter (b) (μm)	Average (K)* b/a	Average cross-sectional area (μm^2)
15.23	18.4	12.1	0.658	182

TABLE II Principal refractive indices ($n_s^{\parallel}$, $n_s^{\perp}$) and birefringence Δn_s for vinyon fibres

Temperature (°C)	$n_s^{\parallel}$ ($\pm 5 \times 10^{-4}$)	$n_s^{\perp}$ ($\pm 5 \times 10^{-4}$)	Δn_s
20	1.5402	1.5337	0.0065
29	1.5380	1.5317	0.0063
32	1.5373	1.5311	0.0062

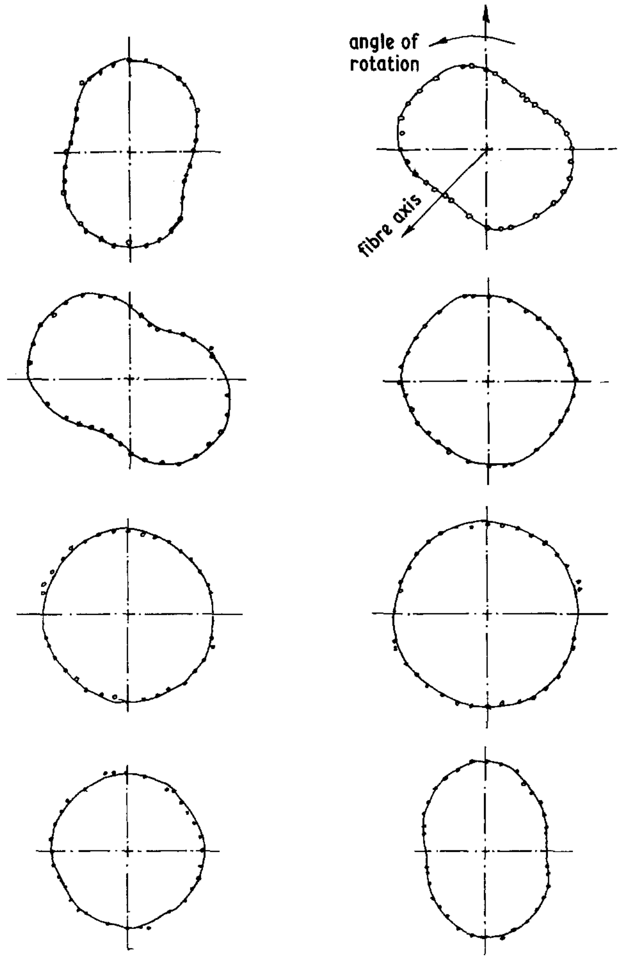

Figure 2 The graphical cross-sectional shape of vinyon fibres.

3.3. Application of double-beam technique

Fig. 3 shows microinterferograms for the duplicated image of the vinyon fibres using a Pluta microscope [8, 9]. White light vibrating parallel and perpendicular to the fibre axis, respectively, was used. The refractive index of the immersion liquid $n_L = 1.4841$ at 18°C.

To determine the mean birefringence of the fibre directly, the differentially sheared (non-duplicated) image of the fibre was used.

The results of the mean refractive indices $n_a^{\parallel}$ and $n_a^{\perp}$ for plane-polarized light vibrating parallel and perpendicular to the fibre, respectively, are given in Table III. The mean birefringence Δn_a of these is also given in this table.

3.4. Application of multiple-beam Fizeau fringes

In this work vinyon fibres are considered to have circular transverse sections at one time and irregular transverse sections at another time. The results are compared with each other in every case.

Fig. 4 shows transmission multiple-beam Fizeau fringes for light vibrating parallel and perpendicular to the fibre axis. Monochromatic light of wavelength $\lambda = 546.1$ nm has been used.

Fig. 5 was produced by applying multiple-beam Fizeau fringes at reflection to a vinyon fibre using plane-polarized light of wavelength $\lambda = 546.1$ nm vibrating parallel and perpendicular to the fibre axis, respectively.

Table IV gives the values of $n_a^{\parallel}$, $n_a^{\perp}$, Δn_a, $n_s^{\parallel}$, $n_s^{\perp}$, Δn_s, $n_c^{\parallel}$, $n_c^{\perp}$, Δn_c using Equations 2, 3 and 4.

3.5. Application of multiple-beam Fizeau fringes to the calculation of the constants of Cauchy's dispersion formula for vinyon fibres

The calculated values of the refractive indices and birefringence for different wavelengths of light are given in Table V.

Fig. 6 shows the relation between ($n_s^{\parallel}$, $n_c^{\parallel}$, $n_a^{\parallel}$ and $1/\lambda^2$); while Fig. 7 shows the relation between ($n_s^{\perp}$, $n_c^{\perp}$, $n_a^{\perp}$ and $1/\lambda^2$). These relations were constructed to evaluate the constants A and B of Cauchy's dispersion formula

$$n = A + B\lambda^{-2}$$

The values of A and B are shown in Table VI.

3.6. Variation of the principal refractive indices and birefringence along the axis of vinyon fibres

Fig. 8 shows transmission multiple-beam Fizeau fringes for light vibrating parallel and perpendicular to the vinyon fibre axis with $n_L = 1.5318$ at 31.5°C and $\lambda = 546.1$ nm. There were observable changes of the refractive indices along the axis of the fibre as indicated in Table VII.

3.7. Determination of the thermal coefficient of the refractive indices for vinyon fibres

The thermal coefficients of the mean refractive indices dn_a/dT of vinyon fibres were determined by forming

TABLE III The measured values of the mean refractive indices and birefringence of vinyon fibres $\lambda^* = 550$ nm, temperature = 18°C, $A = 182 \times 10^{-6}$ mm^2, $M = 510$

n_L	Area enclosed under fringe shift (mm^2)		h_{mm}	$n_a^{\parallel}$	$n_a^{\perp}$	$\Delta n_a = n_a^{\parallel} - n_a^{\perp}$	Δn_a from differentially sheared image
	$F^{\parallel}$	$F^{\perp}$					
1.4841	80	72	8	1.5434	1.5374	0.0060	
1.5339	21	–	9	1.5477	–	–	0.0067

*Taken as the average value for white light.

TABLE IV Refractive indices and birefringence of vinyon fibres. $A = 182 \times 10^{-6}$ mm^2, $\lambda = 546.1$ nm $M = 20.75$ (for Fig. 10) $M = 28$ (for all other figures).

Temperature (°C)	n_L	According to Equation 4									According to Equation 2					According to Equation 3							
		h (mm)	$F^{\parallel}$ (mm^2)	$F^{\perp}$ (mm^2)	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a	Radius (μm) r_s	r_c	$\frac{dz_s^{\parallel}}{h}$	$\frac{dz_s^{\perp}}{h}$	$n_s^{\parallel}$	$n_s^{\perp}$	Δn_s	$\frac{dz_c^{\parallel}}{h}$	$\frac{dz_c^{\perp}}{h}$	$n_c^{\parallel}$	$n_c^{\perp}$	Δn_c	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a
29	1.5322	1.8472	0.5556	0.3333	1.5483	1.5419	0.0064	7.10	3.30	0.5340	0.2860	1.5438	1.5384	0.0054	0.6840	0.3911	1.5472	1.5412	0.0060	1.5454	1.5397	0.0057
31	1.5318	1.4444	0.3333	1.6670	1.5442	1.5380	0.0062	5.96	2.97	0.4138	0.1926	1.5427	1.5369	0.0058	0.5677	0.3267	1.5469	1.5417	0.0052	1.5448	1.5393	0.0055
29	1.5322	1.2639	0.2917	0.1389	1.5446	1.5381	0.0065	6.30	3.20	0.4395	0.2197	1.5433	1.5377	0.0056	0.5823	0.3406	1.5464	1.5414	0.0050	1.5449	1.5396	0.0053
31.5	1.5315	1.8082	0.3214	0.1731	1.5444	1.5384	0.0060	6.80	3.50	0.4957	0.2550	1.5431	1.5375	0.0056	0.6782	0.4070	1.4570	1.5417	0.0053	1.5451	1.5347	0.0054
31	1.5318	1.8472	0.5000	0.2920	1.5463	1.5403	0.0060	7.50	3.70	0.5867	0.3010	1.5441	1.5381	0.0060	0.7522	1.4358	1.5469	1.5414	0.0055	1.5455	1.5397	0.0058
29	1.5322	1.5278	0.4583	0.2778	1.5483	1.5420	0.0063	6.85	3.28	0.5003	0.2637	1.5436	1.5382	0.0054	0.6636	0.3728	1.5474	1.5412	0.0062	1.5454	1.5396	0.0058
29	1.5322	1.5139	0.4722	0.2917	1.5489	1.5425	0.0064	7.50	3.45	0.5596	0.3120	1.5437	1.5386	0.0051	0.7339	0.4585	1.5477	1.5428	0.0049	1.5455	1.5405	0.0050
31.5	1.5318	1.5566	0.3451	0.1593	1.5437	1.5373	0.0064	5.90	2.41	0.4626	0.2339	1.5435	1.5377	0.0058	0.6108	0.3354	1.5495	1.5423	0.0072	1.5459	1.5396	0.0063

TABLE V Variation of the refractive indices with wavelengths for vinyon fibre $A = 182 \times 10^{-6}$ mm^2, $M = 24.42$; $r_s = 8.5$; $r_c = 2.55$; $T = 32.5$°C

According to Equation 4								According to Equation 2					According to Equation 3							
λ (nm)	n_L	h (mm)	$F^{\parallel}$ (mm^2)	$F^{\perp}$ (mm^2)	n_a			$\frac{dz_s^{\parallel}}{h}$	$\frac{dz_s^{\perp}}{h}$	n_s			$\frac{dz_c}{h}$	$\frac{dz_c}{h}$	n_c			n_a		
					$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a			$n_s^{\parallel}$	$n_s^{\perp}$	Δn_s			$n_c^{\parallel}$	$n_c^{\perp}$	Δn_c	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a
589.3	1.5292	1.104	0.2277	0.1282	1.5429	1.5369	0.0060	0.666	0.347	1.5413	1.5355	0.0058	0.740	0.396	1.5437	1.5374	0.0063	1.5420	1.5361	0.0059
578.0	1.5295	0.987	0.2108	0.1181	1.5434	1.5373	0.0061	0.685	0.354	1.5417	1.5358	0.0059	0.760	0.404	1.5441	1.5377	0.0064	1.5424	1.5364	0.006
546.1	1.5300	0.818	0.1940	0.1096	1.5446	1.5382	0.0064	0.754	0.392	1.5427	1.5366	0.0061	0.841	0.452	1.5452	1.5388	0.0066	1.5435	1.5373	0.0062
535.1	1.5310	0.779	0.1800	0.0953	1.5449	1.5384	0.0065	0.740	0.364	1.5432	1.5370	0.0062	0.827	0.420	1.5459	1.5390	0.0069	1.5440	1.5376	0.0064
436.0	1.5381	0.636	0.1600	0.0692	1.5504	1.5434	0.0070	0.781	0.290	1.5486	1.5420	0.0066	0.877	0.348	1.5511	1.5439	0.0072	1.5494	1.5426	0.0068

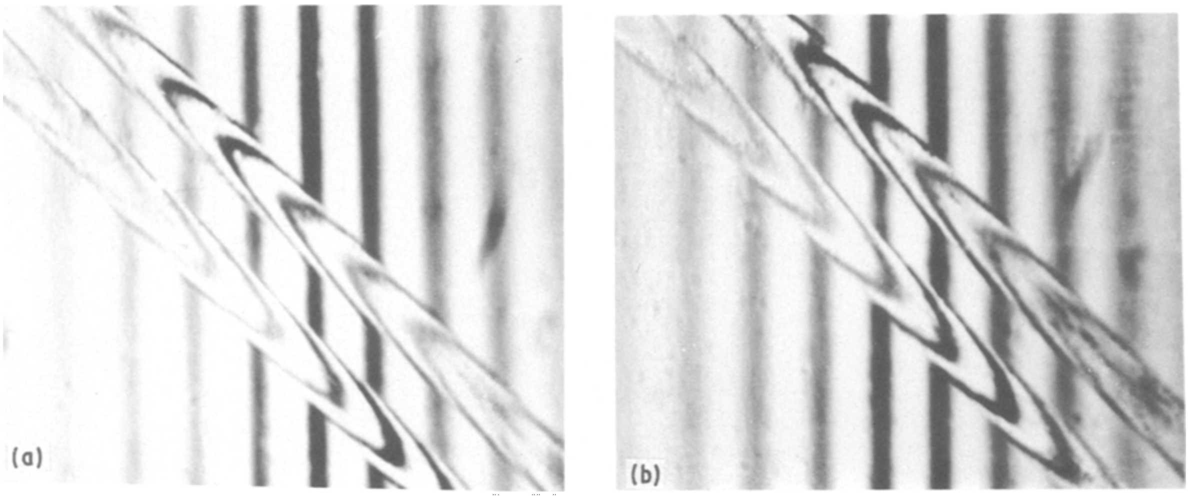

Figure 3 Photomicrograph of totally duplicated image of a vinyon fibre using white light vibrating (a) parallel and (b) perpendicular to the fibre axis.

Fizeau fringes for the same section of the fibre when immersed in a silvered liquid wedge at two temperatures T_1 and T_2. Table VIII gives the estimated values $dn_a^{\perp}/dT$ using light of wavelength 546.1 nm vibrating perpendicularly to the fibre axis. A knowledge of these thermal coefficients is very valuable in order to explore the transmissivity and reflectivity of the fibrous medium.

4. Discussion and conclusion

Part of the modern trend in fibre research is to alter fibre properties; one of the methods for property modification involves the formation of copolymerization at the resin preparation stage.

It is generally more difficult to chemically characterize copolymers which consist of more than one kind of repeated unit along the backbone and also to correlate with their physical properties [10].

Vinyon HH and CF (American viscose) is vinyl chloride-vinyl acetate copolymer (HH unstretched, CF stretched form). Vinyon has a different microscopic appearance depending on the manufacturing process and on the degree of stretching after spinning. The stretched fibre (usually a continuous filament, CF) has an irregular shape in cross-section, whereas the unstretched fibre (usually the staple fibre, HH) is circular in cross-section.

Recent mathematical formulae are applied to estimate the basic optical parameters for the skin and core of vinyon fibres with irregular and circular cross-sectional shape. The measured values for the refractive indices and birefringence are in good agreement with the values that have been previously published [11]. The slight variation in the obtained results with respect to the published data has been due to the drawing and spinning process and the technique used.

In conclusion the recent mathematical expressions provide a solution to, and accurate results for a fundamental problem for copolymer fibres encountered by the final product characteristics involved with

TABLE VI Values for Cauchy's dispersion formula constants for vinyon fibres

Layer	A		B	
	$A^{\parallel}$	$A^{\perp}$	$B^{\parallel}$ (nm^2)	$B^{\perp}$ (nm^2)
Mean value	1.5341	1.5291	3110.7	2719.1
Skin	1.5325	1.5275	3059.7	2748.4
Core	1.5350	1.5296	3076.6	2726.1

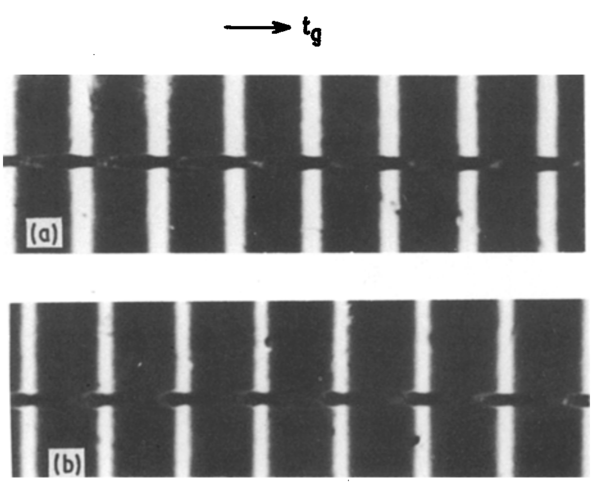

Figure 4 Transmission multiple-beam Fizeau fringes for light vibrating (a) parallel and (b) perpendicular to the fibre axis.

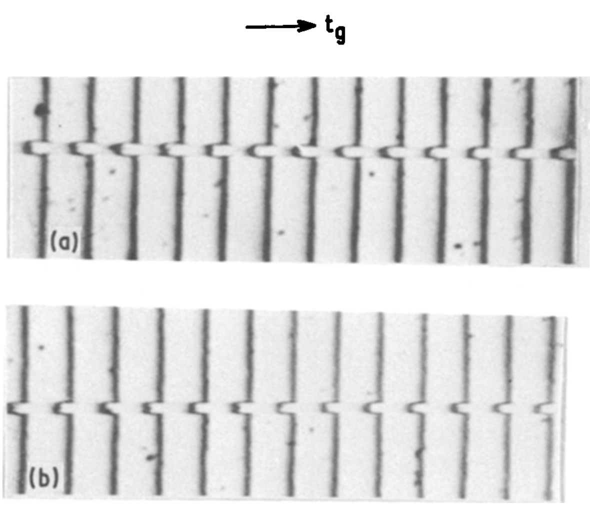

Figure 5 Reflection photomicrograph of multiple-beam Fizeau fringes for light vibrating (a) parallel and (b) perpendicular to the fibre axis.

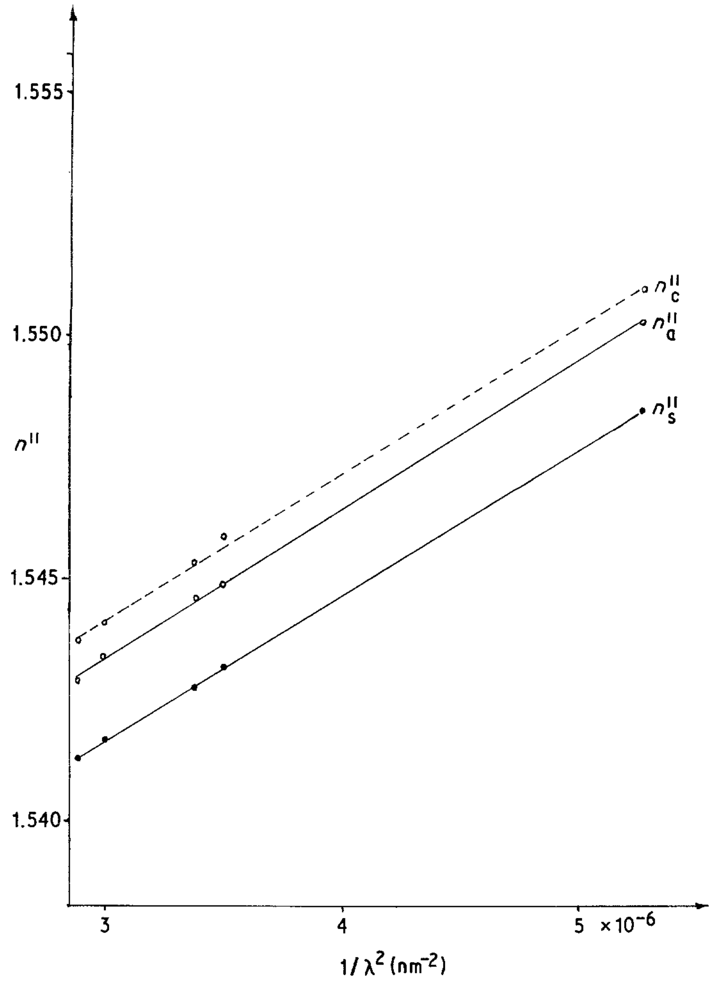

Figure 6 The variation of refractive indices $n_s^{\parallel}$, $n_c^{\parallel}$ and $n_a^{\parallel}$ with $1/\lambda^2$.

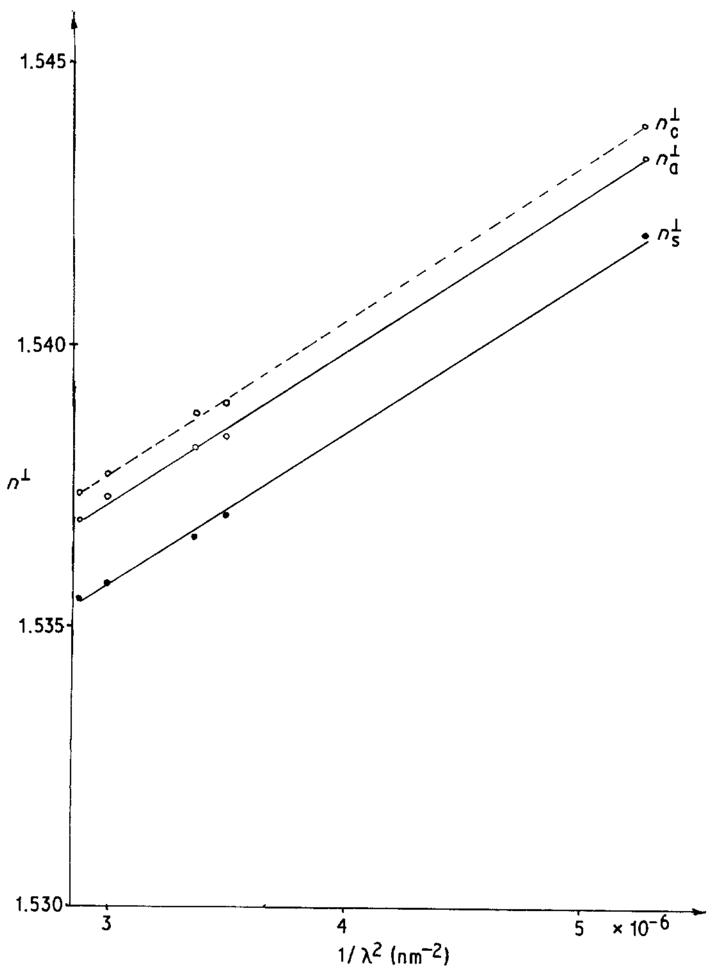

Figure 7 The variation of refractive indices $n_s^{\perp}$, $n_c^{\perp}$ and $n_a^{\perp}$ with $1/\lambda^2$.

TABLE VII Variation of the principal refractive indices ($n_a^{\parallel}$, $n_s^{\parallel}$, $n_c^{\parallel}$ and $n_a^{\perp}$, $n_s^{\perp}$, $n_c^{\perp}$) and the birefringence (Δn_a, Δn_s, Δn_c) along the axis of multiple-skin vinyon fibre. $A = 182 \times 10^{-6}\,\text{mm}^2$, $M = 25$, $h = 2.06\,\text{mm}$, $\lambda = 546.1\,\text{nm}$, $n_L = 1.5318$. $I = 31.5°\text{C}$.

No	Distance (μm)	According to Equation 4							According to Equation 2					According to Equation 3							
		$F^{\parallel}$ (mm^2)	$F^{\perp}$ (mm^2)	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a	Radius (μm) r_s	r_c	$\frac{dz_s^{\parallel}}{h}$	$\frac{dz_s^{\perp}}{h}$	$n_s^{\parallel}$	$n_s^{\perp}$	Δn_s	$\frac{dz_c^{\parallel}}{h}$	$\frac{dz_c^{\perp}}{h}$	$n_c^{\parallel}$	$n_c^{\perp}$	Δn_c	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a
1	0.000	0.2741	0.0712	1.5398	1.5339	0.0059	8.40	2.20	0.44	0.11	1.5392	1.5337	0.0055	0.47	0.13	1.5407	1.5345	0.0062	1.5396	1.5339	0.0057
2	82.4	0.2884	0.0819	1.5402	1.5342	0.0060	8.40	2.20	0.46	0.12	1.5395	1.5338	0.0057	0.50	0.14	1.5411	1.5348	0.0063	1.5399	1.5341	0.0058
3	164.8	0.3097	0.1000	1.5408	1.5347	0.0061	8.45	2.20	0.50	0.13	1.5402	1.5340	0.0062	0.54	0.15	1.5414	1.5349	0.0065	1.5405	1.5392	0.0063
4	247.2	0.3311	0.1175	1.5414	1.5352	0.0062	8.45	2.30	0.53	0.16	1.5407	1.5345	0.0062	0.57	0.18	1.5418	1.5353	0.0065	1.5410	1.5347	0.0063
5	329.6	0.3382	0.1246	1.5417	1.5354	0.0063	8.45	2.40	0.55	0.19	1.5411	1.5350	0.0061	0.59	0.21	1.5419	1.5357	0.0062	1.5413	1.5352	0.0061
6	412.0	0.3489	0.1353	1.5420	1.5357	0.0063	8.45	2.40	0.57	0.21	1.5414	1.5353	0.0061	0.61	0.23	1.5423	1.5361	0.0062	1.5417	1.5355	0.0062
7	494.4	0.3560	0.1424	1.5422	1.5359	0.0063	8.48	2.65	0.58	0.23	1.5416	1.5357	0.0059	0.63	0.25	1.5427	1.5361	0.0066	1.5419	1.5358	0.0061
8	576.8	0.363	0.1440	1.5424	1.5360	0.0064	8.48	2.65	0.59	0.24	1.5418	1.5359	0.0059	0.64	0.26	1.5428	1.5362	0.0066	1.5421	1.5360	0.0061
9	659.2	0.363	0.1440	1.5424	1.5360	0.0064	8.46	2.46	0.59	0.24	1.5418	1.5359	0.0059	0.64	0.26	1.5428	1.5362	0.0066	1.5421	1.5360	0.0061
Average value		0.3303	0.1179	1.5414	1.5352	0.0062	8.45	2.41	0.53	0.18	1.5408	1.5349	0.0059	0.58	0.20	1.5419	1.5355	0.0064	1.5411	1.5350	0.0061

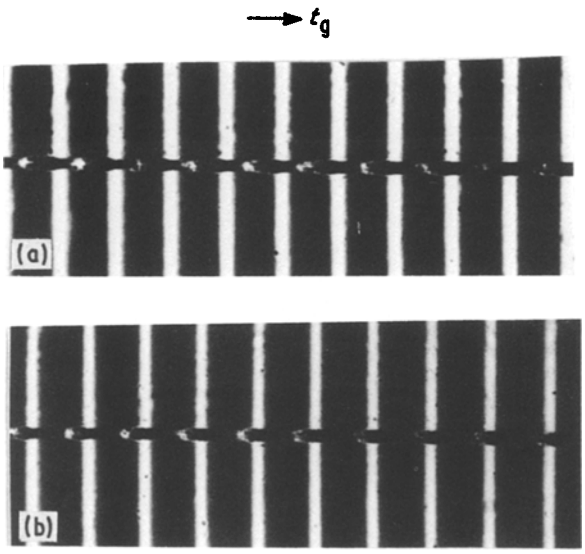

Figure 8 Multiple-beam Fizeau fringes in transmission for light vibrating (a) parallel and (b) perpendicular to a vinyon fibre axis to show the variation of ($n_s^{\parallel}$, $n_c^{\parallel}$, $n_a^{\parallel}$, $n_s^{\perp}$, $n_c^{\perp}$ and $n_a^{\perp}$) along this axis.

irregular and circular skin and core cross-sectional fibres.

The birefringence is not characteristic of the material itself but subject to a great variation within the same fibre due to skin (0.0055–0.0062) and core (0.0062–0.0066) orientation.

Finally, the practical importance for the present study lies in emphasizing the importance of measuring the optical concept of copolymer fibre orientation and its behaviour with temperature and different wavelengths. Hence, the radiative properties of the medium used must be properly defined with respect to the fibre orientation, size distribution and optical properties [12]. The structural properties must also be correlated with other physical properties.

TABLE VIII Determination of the thermal coefficient of the mean refractive index of vinyon fibre

Temperature (°C)	n_L	$n_a^{\perp}$	$\frac{dn_a^{\perp}}{dT}$ (°C^{-1})
30.5	1.5350	1.5386	
32.5	1.5340	1.5370	-8×10^{-4}

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