Optothermal Properties of Fibers. V. Optical Anisotropy in Silk Fibers as a Function of Annealing Process

A. A. HAMZA, I. M. FOUDA,* T. Z. SOKKAR, and M. A. EL-BAKARY

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

SYNOPSIS

Silk fibers (Best Yellow Italian Silk, Ford & Co. Ltd) were annealed at constant temperatures of 100 and 150°C for different times ranging from 1 to 10 h and for different temperatures ranging from 60 to 160 ± 1°C for a constant time of 2 h. Refractive indices and birefringence were measured interferometrically. Two independent techniques were used to study the optical anisotropy in these fibers. The first technique was to study the effect of annealing on the swelling properties of fibers from its diffraction pattern using a He — Ne laser beam. The second is the application of a double-refracting interference microscope. The double-beam technique is considered the most favorable technique in determining the mean refractive indices and the double refraction of the annealed samples. The behavior of optical properties at different annealing temperatures and times are discussed. The results obtained clarify that the reorientations occurred due to annealing at different conditions. Microinterferograms and curves are given for illustration.

INTRODUCTION

Interferometry is a very useful tool in fiber science. In recent years, interferometric methods were used to determine the refractive indices and birefringence of both natural and synthetic fibers.[1-3] More recently, the application of double-beam and multiple-beam Fizeau fringes interferometry has stimulated interest in studying the effect of different types of thermal and mechanical properties of natural and man-made fibers.[4-6] Natural silk fibers have a double refraction of a positive value,[6] because their refractive index is greater for light vibrating parallel to the fiber axis.

The good thermal stability is another attractive characteristic that makes silk fibers suitable for several applications. Therefore, the purpose of improving the thermal resistance of silk by suitable treatments should be considered as a new approach for research in the field of structural proteins in order to make new materials and expand their utilization.[7]

Swelling of the fibers is another very important tool for the investigation of the structure of the fiber material. When a fiber is immersed in water, the water molecules infiltrate into the fiber and find their way in between the long-chain molecules, thereby pushing them apart. As the fiber-chain molecules are oriented in line with the axis of the fiber and as they are pushed apart, there will be a considerable increase in the diameter of the fiber.[8] Water molecules or liquid molecules can enter the amorphous regions, but not the crystalline region.[9] The value of the swelling factor is an indication of the fiber material: The higher the swelling factor, the greater the amorphous regions.[8]

In the present work, the refractive indices and birefringence and swelling factor and their effect on the variation of the annealing conditions of silk fibers was studied.

THEORETICAL CONSIDERATIONS

Double-beam Interferometry

For the determination of the mean refractive indices and birefringence, a Pluta polarizing interference

* To whom correspondence should be addressed.

Journal of Applied Polymer Science, Vol. 60, 1289–1299 (1996)
 CCC 0021-8995/96/091289-11

microscope was used[10,11] to produce a duplicated image for the samples using the following equation[3]:

$$n_a^{\parallel} = n_L + \frac{F^{\parallel}}{h}\frac{\lambda}{A} \quad (1)$$

where $n_a^{\parallel}$ is the mean refractive index of the fiber material for plane-polarized light vibrating parallel to the fiber axis; n_L, the refractive index of the immersion liquid; $F^{\parallel}$, the total area enclosed under the fringe shift; and h, the interfering spacing in the liquid region. A is the mean cross-sectional area of the fiber, and λ, the wavelength of monochromatic light used. Equation (1) has an analogous form for light vibrating perpendicular to the fiber axis for the determination of $n^{\perp}$.

The Pluta microscope can be used to determine, directly, the mean birefringence, Δn_a, by producing a nonduplicated image for the samples using the following equation[3]:

$$\Delta n_a = \frac{F}{h}\frac{\lambda}{A} \quad (2)$$

where F is the total area enclosed under the fringe shift in the nonduplicated image.

Polarizability per Unit Volume and Isotropic Refractive Index

For the determination of the polarizability per unit volume and the isotropic refractive index, the results of the refractive indices were employed in calculating these optical parameters according to the following relations[12]:

$$(n^2 - 1)/(n^2 + 2) = \tfrac{4}{3}\pi P \quad (3)$$

where P is the polarizability per unit volume. Also, the isotropic refractive index n_{iso} is given by the following equation[13]:

$$n_{iso} = (n_a^{\parallel} + 2n_a^{\perp})/3 \quad (4)$$

Swelling of the Fibers

For the determination of the swelling factor of the fibers, Jinricii et al.[14] used the diffraction of monochromatic light by a laser beam to a single fiber to estimate the average diameter of fibers before and after wetting in solvents using the following equation:

$$d = \pm\frac{\lambda L}{x} \quad (5)$$

where d is the diameter of the fiber; λ, the wavelength of monochromatic light used; x, the distance from the center of the pattern to the first minimum; and L, the distance between the fiber and screen on which the pattern was produced. The swelling factor can be determined using the following equation[14]:

$$q = 100(d_s - d_d)/d_d \quad (6)$$

where q is the swelling factor; d_s, the diameter of swollen fiber; and d_d, the diameter of the dry fiber.

EXPERIMENTAL AND RESULTS

Sample Preparation (Annealing)

The scoured silk fibers were distributed in three groups in small glass dishes, then left in an electric oven whose temperature was adjusted to be constant to within ±1°C. The first group was heated at a constant temperature of 100°C, the second was heated at a constant temperature of 150°C for different annealing times ranging from 1 to 10 h, and the third was heated at a constant time of 2 h for different annealing temperatures ranging from 60 to 160°C. The samples after heating were then left to cool at a room temperature of 31 ± 3°C.

Measurement of Cross-sectional Area

Plate 1 shows the cross section of untreated silk fibers seen by high-power optical microscopy. It is clear that the majority of the individual fibers have a triangular cross section with rounded corners and also a wide range of cross-sectional areas. A statis-

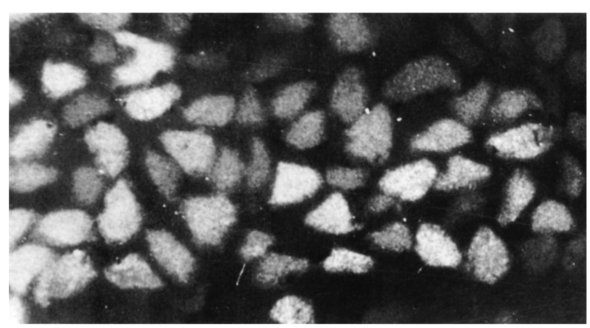


Plate 1 Optical micrograph of a cross section of natural silk fibers.

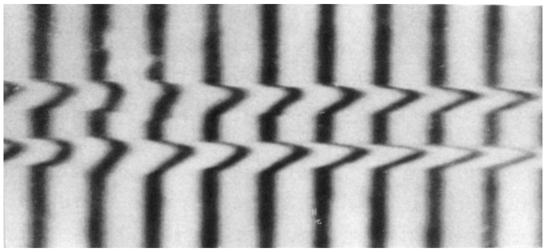


Plate 2 Totally duplicated image of natural silk fiber; using a Pluta microscope, monochromatic light of wavelength 546 nm was used at 21°C.

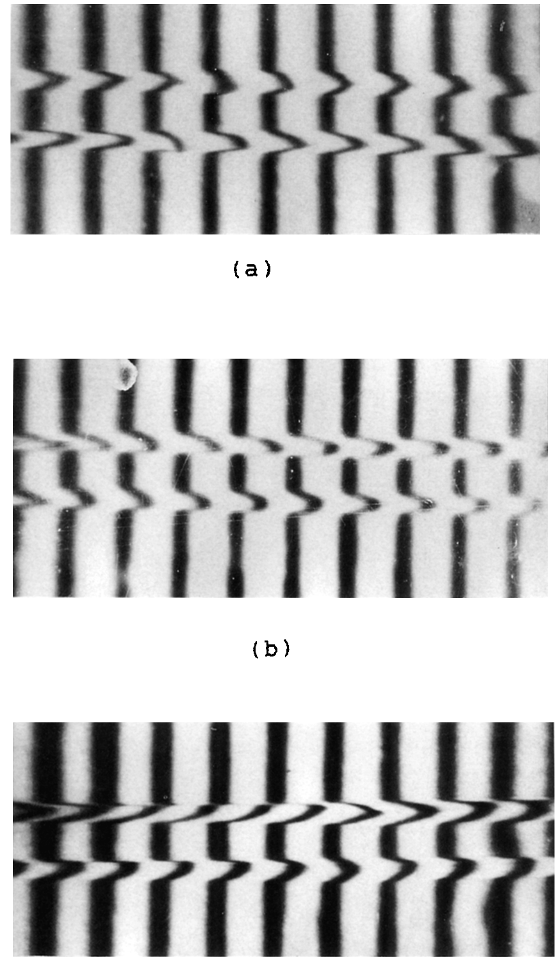
tical determination for the average cross section (A) of silk fibers was determined and found to be $A = 64.3 \pm 8.5\ \mu m^2$.

Double-beam Interferometry

Plates 2 and 3 show microinterferograms of duplicated and nonduplicated images of natural silk fiber without annealing, respectively. Monochromatic light of wavelength 546 nm was used. The refractive index of the immersion liquid was 1.5715 at 21°C. By using these interferograms and eqs. (1) and (2) the mean refractive indices and birefringence were calculated.

Plate 4(a–c) shows microinterferograms of totally duplicated images for annealed silk fibers at a constant temperature of 100°C for different times of 1, 4, and 10 h using monochromatic light of wavelength 546 nm. The refractive index of the immersion liquid was 1.5715 at 22°C.

Plate 5(a–c) shows microinterferograms of totally duplicated images of annealed silk fibers at a constant time of 2 h for different annealing temperatures of 80, 100, and 160°C. Using monochromatic light of wavelength 546 nm, the immersion liquid refractive index was 1.5715 at 31°C.



(a)

(b)

(c)

Plate 4 Totally duplicated image of annealed silk fibers at a constant temperature of 100°C for different times: (a) 1, (b) 4, and (c) 10 h. Using a Pluta microscope, the wavelength of monochromatic light used was 546 nm at 22°C.

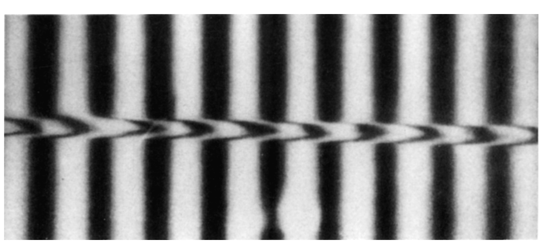
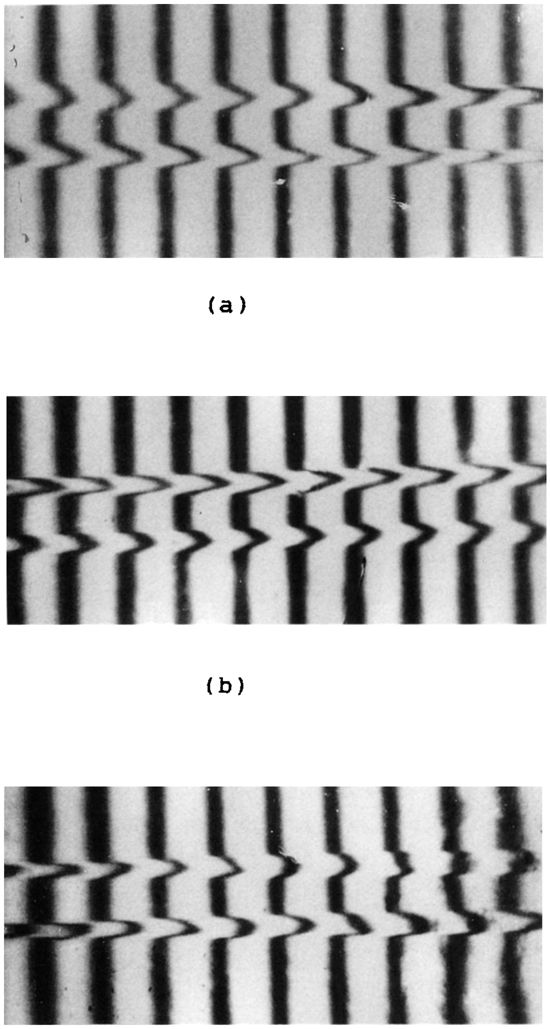


Plate 3 Nonduplicated image of natural silk fiber. Using a Pluta microscope, the wavelength of monochromatic light used was 546 nm at 21°C.

Plate 6(a–c) shows microinterferograms of nonduplicated images of annealed silk fibers at a constant temperature of 150°C for different times of 1, 4, and 10 h. Using monochromatic light of wavelength 546 nm, the refractive index of the immersion liquid was 1.5715 at 22°C.

Plate 7(a–c) shows microinterferograms of nonduplicated images of annealed silk fibers at a constant time of 2 h for different annealing temperatures of 80, 100, and 160°C. Using monochromatic



(a)

(b)

(c)

Plate 5 Totally duplicated images of annealed silk fibers at a constant time of 2 h for different temperatures: (a) 60, (b) 80, and (c) 160°C. Using the Pluta microscope, the wavelength of light used was 546 nm at 31°C.

light of wavelength 546 nm, the refractive index of the immersion liquid was 1.5715 at 31°C.

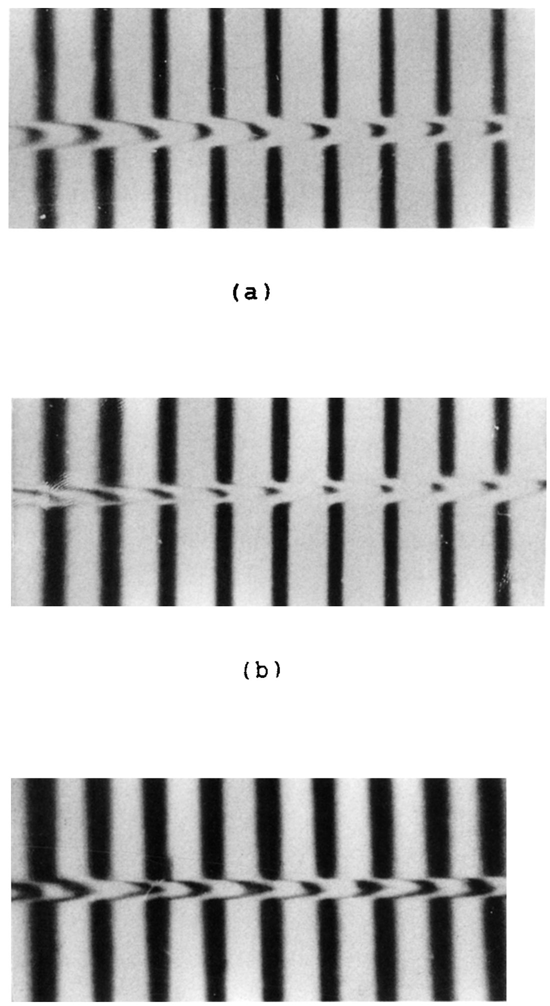
Table I shows the values of birefringence obtained for annealed silk fibers at a constant temperature of 100°C for different times by duplicated and nonduplicated images and gives the results obtained for natural silk fiber annealed at a constant temperature of 100°C for different annealing times using a Pluta microscope considering the average values.

Figure 1 shows the variation of both $n_a^{\parallel}$ and $n_a^{\perp}$ of silk fibers by increasing the annealing time for a constant temperature of 100°C as obtained using a Pluta interference microscope. It is clear that the parallel refractive index $n_a^{\parallel}$ slightly decreased with increasing annealing time, while the perpendicular refractive index $n_a^{\perp}$ decreases, which gives the result that the birefringence Δn decreases with increasing annealing time, as shown in Figure 2.

Empirical formulas of the refractive indices $n_a^{\parallel}$ and $n_a^{\perp}$ as a function of annealing time for annealed silk fibers at a constant temperature of 100°C are assessed as follows:

$$n_a^{\parallel} = n_{a_0}^{\parallel} + Ct$$

$$n_a^{\perp} = n_{a_0}^{\parallel} + Dt$$

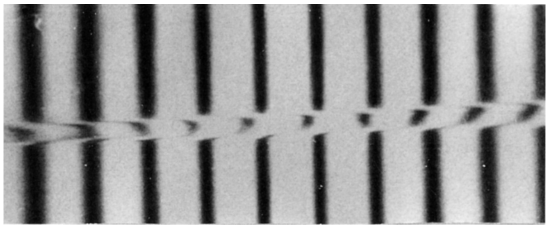


(a)

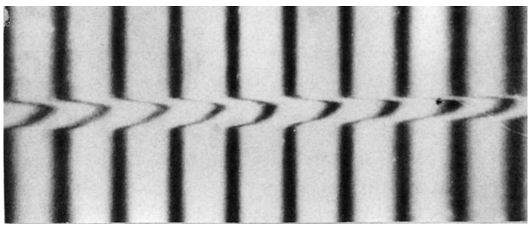
(b)

(c)

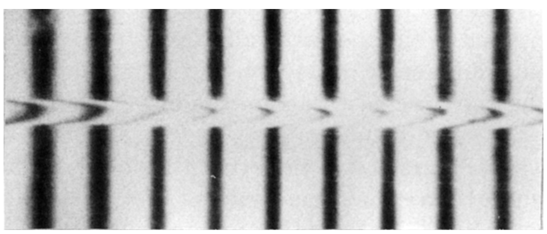
Plate 6 Nonduplicated image of annealed silk fibers at a constant temperature of 150°C for different times: (a) 1, (b) 4, and (c) 10 h. Using a Pluta microscope, the wavelength of monochromatic light used was 546 nm at 22°C.



(a)



(b)



(c)

Plate 7 Nonduplicated images of annealed silk fibers at a constant time of 2 h for different temperatures: (a) 60, (b) 80, and (c) 160°C. Using a Pluta microscope, the wavelength of light used was 546 nm at 31°C.

where $n_{a_0}^{\parallel}$ and $n_{a_0}^{\perp}$ are the refractive indices of unannealed fiber for the two directions of the vibration, and C and D are constants that characterize the orientation process due to the annealing. The refractive indices $n_{a_0}^{\parallel}$ and $n_{a_0}^{\perp}$ and the constants C and D were calculated for the annealed silk fibers and were found to be

$$n_{a_0}^{\parallel} = 1.593, \quad n_{a_0}^{\perp} = 1.551 \quad \text{and} \quad C = -0.00061, \quad D = 0.000651$$

Figure 1 shows this linear relation.

Figures 3–5 show the relation between the polarizabilities $p^{\parallel}$ and $P^{\perp}$ and the isotropic refractive index n_{iso} with the annealing time. A dashed curve is the theoretical curve obtained from the calculations from the constants of the above empirical formulas and the solid curve is obtained from the experimen-

Table I Values of (a) Birefringence and (b) Some Optical Parameters for Annealed Silk Fibers

(a)

Annealing Time (h)	$\Delta n_a = n_a^{\parallel} - n_a^{\perp}$		Δn_a (Directly)		Δn_a (Average)	
	Max	Min	Max	Min	Max	Min
0	0.042	0.041	0.043	0.035	0.042	0.038
1	0.053	0.040	0.034	0.026	0.043	0.028
2	0.036	0.027	0.034	0.026	0.035	0.026
4	0.049	0.037	0.030	0.023	0.039	0.029
6	0.039	0.030	0.032	0.025	0.035	0.027
8	0.035	0.025	0.038	0.028	0.036	0.026
10	0.036	0.027	0.043	0.033	0.039	0.030

(b)

Annealing Time (h)	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a	n_{iso}	$p^{\parallel}$	$p^{\perp}$
0	1.595	1.553	0.042	1.567	0.081	0.076
1	1.594	1.547	0.047	1.563	0.081	0.076
2	1.590	1.559	0.032	1.569	0.081	0.077
4	1.592	1.549	0.043	1.563	0.081	0.076
6	1.589	1.555	0.035	1.567	0.081	0.077
8	1.589	1.559	0.030	1.565	0.080	0.077
10	1.589	1.557	0.032	1.568	0.080	0.077

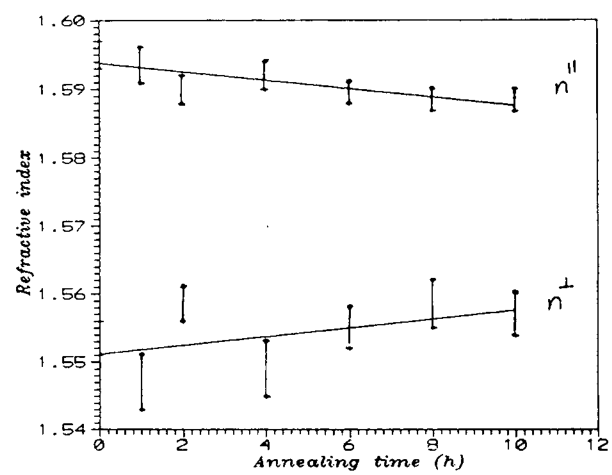


Figure 1 Relation between the refractive indices $n_a^{\parallel}$ and $n_a^{\perp}$ of annealed silk fibers and the annealing time at a constant temperature of 100°C.

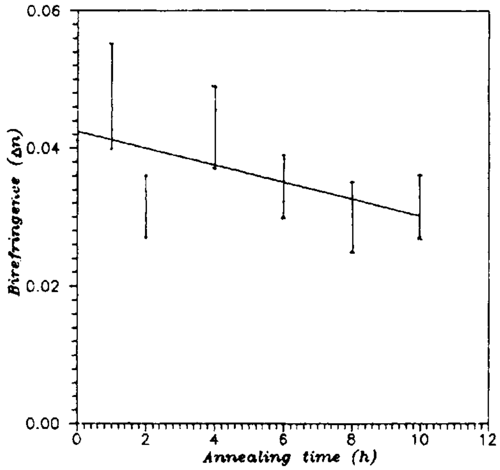


Figure 2 Relation between the birefringence Δn_a of annealed silk fibers and the annealing time at a constant temperature of 100°C.

tal data of the refractive indices. It is clear that the polarizabilities have the same behavior of $n_a^{\|}$ and $n_a^{\perp}$, while the isotropic refractive index n_{iso} increased with increasing time.

Table II shows the values of birefringence obtained for annealed silk fibers at a constant temperature of 150°C for different times (1–10 h) by duplicated and nonduplicated images and gives the results obtained for natural silk fibers annealed at a constant temperature of 150°C for different annealing times using a Pluta inference microscope.

Figure 6 shows the variation of both $n_a^{\|}$ and $n_a^{\perp}$ of annealed silk fibers by increasing the annealing

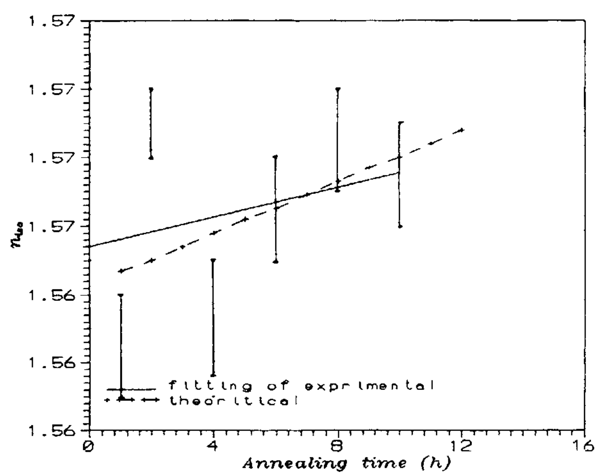


Figure 3 Relation between the polarizability $p^{\|}$ of annealed silk fibers and the annealing time at a constant temperature of 100°C.

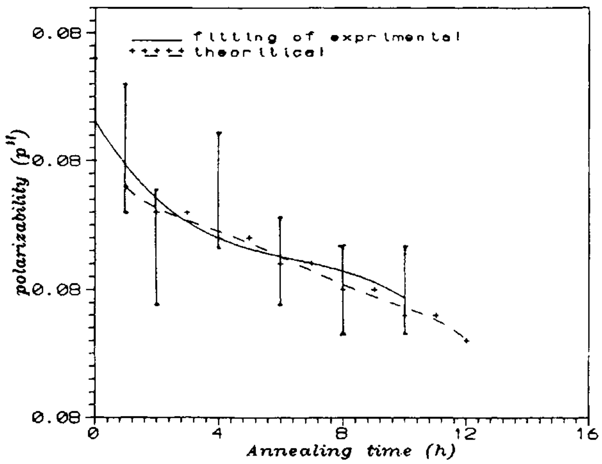


Figure 4 Relation between the polarizability $p^{\perp}$ of annealed silk fibers and the annealing time at a constant temperature of 100°C.

time for a constant temperature of 150°C, as obtained using a Pluta interference microscope. It is clear that the parallel refractive index $n_a^{\|}$ is decreased at nearly 2 h and increased at 7 h and then again decreased, while the perpendicular refractive index $n_a^{\perp}$ is increased at nearly 2 h and decreased until 7 h and then increased again; the birefringence Δn_a has the same behavior of $n_a^{\|}$ at the same times as given in Figure 7. It is clear that the polarizabilities $p^{\|}$ and $p^{\perp}$ have the same behavior of $n_a^{\|}$ and $n_a^{\perp}$, respectively, at the same times of annealing, which is given in Figure 8.

Figure 9 shows the variation of the isotropic re-

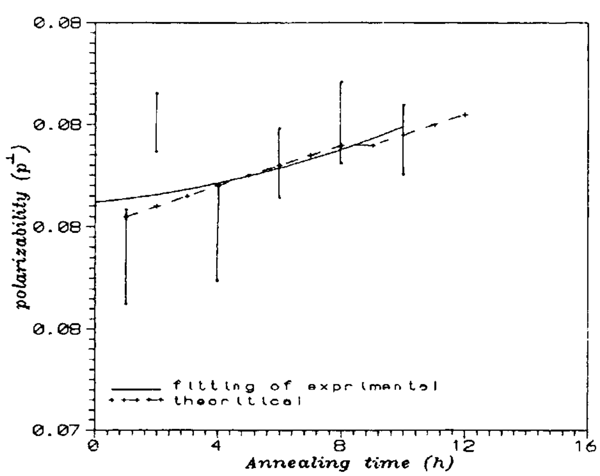


Figure 5 Relation between the isotropic refractive index n_{iso} of annealed silk fibers and the annealing time at a constant temperature of 100°C.

Table II (a) Values of Birefringence Obtained for Annealed Silk Fibers at a Constant Temperature of 150°C for Different Times by Duplicated and Nonduplicated Images; (b) Results Obtained for Natural Silk Fibers Annealed at a Constant Temperature of 150°C for Different Annealing Times Using a Pluta Interference Microscope

(a)

Annealing Time (h)	$\Delta n_a = n_a^{\parallel} - n_a^{\perp}$		Δn_a (Directely)		Δn_a (Average)	
	Max	Min	Max	Min	Max	Min
0	0.042	0.041	0.043	0.035	0.042	0.038
1	0.036	0.027	0.035	0.026	0.035	0.026
2	0.033	0.026	0.036	0.025	0.034	0.025
4	0.034	0.025	0.031	0.022	0.032	0.023
6	0.041	0.031	0.040	0.029	0.040	0.030
8	0.037	0028	0.039	0.031	0.038	0.029
10	0.025	0.019	0.031	0.022	0.028	0.020

(b)

Annealing Time (h)	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a	n_{iso}	$p^{\parallel}$	$p^{\perp}$
0	1.595	1.553	0.042	1.567	0.081	0.076
1	1.591	1.560	0 031	1.570	0.081	0.077
2	1.591	1.561	0.030	1.571	0.081	0.077
4	1.589	1.559	0.030	1.569	0.080	0.077
6	1.593	1.557	0.036	1.569	0.081	0.077
8	1.595	1.562	0.033	1.573	0.081	0.077
10	1.587	1.565	0.022	1.572	0.080	0.078

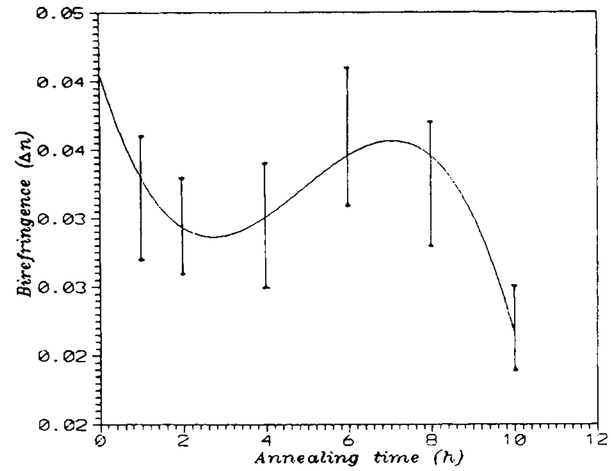


Figure 7 Relation between the birefringence Δn_a of annealed silk fibers and the annealing time at a constant temperature of 150°C.

fractive index n_{iso} with annealing time, which has nearly the same behavior of $n_a^{\perp}$.

Table III shows the values of the birefringence obtained for annealed silk fiber at a constant time of 2 h for different annealing temperature by Pluta polarizing microscope.

Figure 10 shows the variation of both $n_a^{\parallel}$ and $n_a^{\perp}$ of natural silk fibers by increasing the annealing temperature, as obtained using a Pluta microscope. It is clear that both $n_a^{\parallel}$ and $n_a^{\perp}$ were increased with increasing the annealing temperature by approximately the same ratio and, hence, the birefringence shows nearly constant values, as given in Figure 11. The polarizabilities $p^{\parallel}$ and $p^{\perp}$ have the same behavior

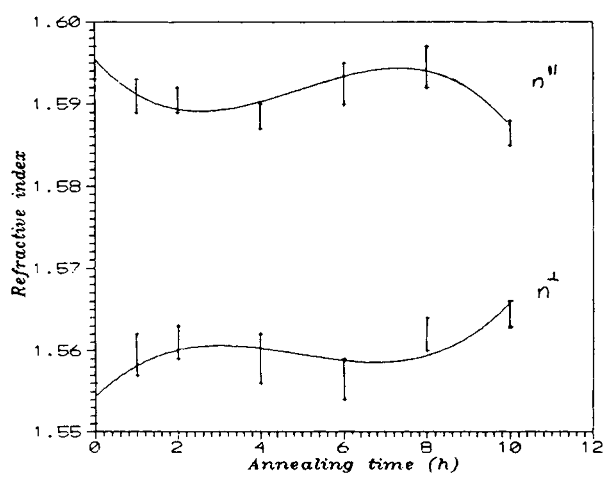


Figure 6 Relation between the refractive indices $n_a^{\parallel}$ and $n_a^{\perp}$ of annealed silk fibers and the annealing time at a constant temperature of 150°C.

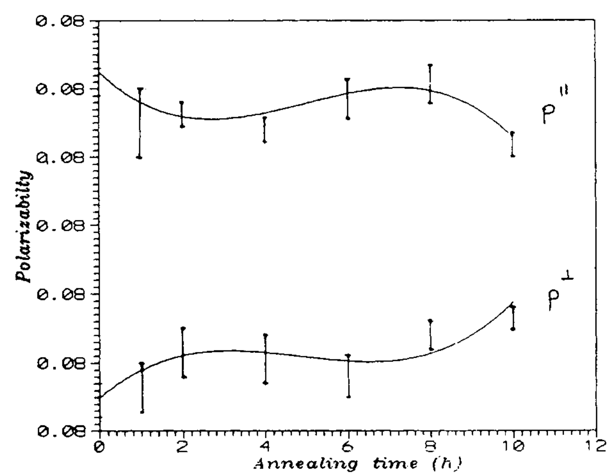


Figure 8 Relation between the polarizabilities $p^{\parallel}$ and $p^{\perp}$ of annealed silk fibers and the annealing time at a constant temperature of 150°C.

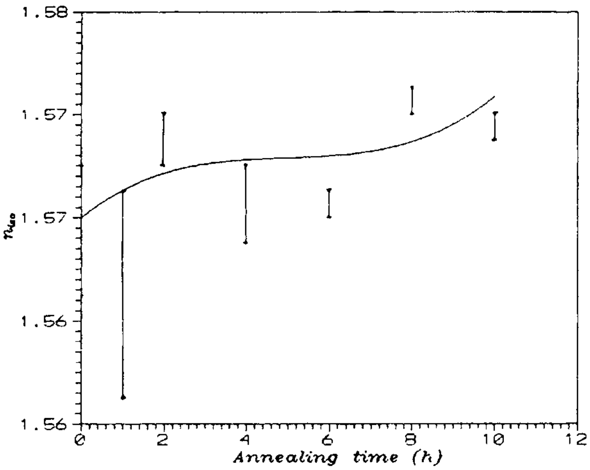


Figure 9 Relation between the isotropic refractive index n_{iso} of annealed silk fibers and the annealing time at a constant temperature of 150°C.

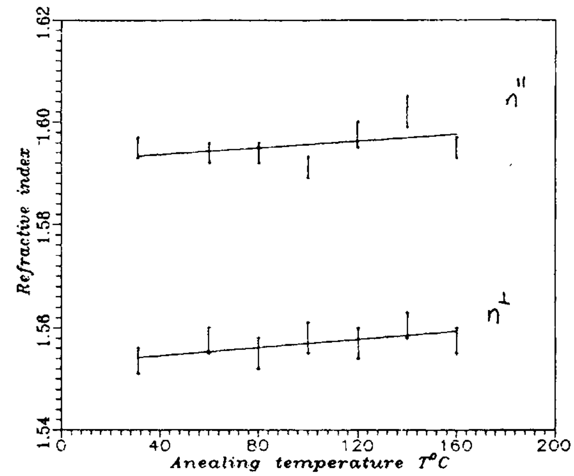


Figure 10 Relation between the refractive indices $n_a^{\parallel}$ and $n_a^{\perp}$ of annealed silk fibers and the annealing temperature at a constant time of 2 h.

Table III (a) Values of Birefringence Obtained for Annealed Silk Fibers at a Constant Time of 2 h for Different Annealing Temperatures by Duplicated and Nonduplicaed Images; (b) Results Obtained for Natural Silk Fibers Annealed at a Constant Time of 2 h for Different Temperatures, Using a Pluta Microscope

(a)

Annealing Temp (°C)	$\Delta n_a = n_a^{\parallel} - n_a^{\perp}$		Δn_a (Directly)		Δn_a (Average)	
	Max	Min	Max	Min	Max	Min
31	0.042	0.041	0.043	0.035	0.042	0.038
60	0.041	0.032	0.042	0.032	0.041	0.032
80	0.044	0.034	0.041	0.032	0.042	0.033
100	0.038	0.028	0.037	0.029	0.037	0.028
120	0.046	0.035	0.046	0.035	0.046	0.035
140	0.047	0.036	0.044	0.033	0.045	0.034
160	0.042	0.033	0.045	0.034	0.043	0.033

(b)

Annealing Temp (°C)	$n_a^{\parallel}$	$n_a^{\perp}$	Δn_a	n_{iso}	$p^{\parallel}$	$p^{\perp}$
31	1.595	1.553	0.042	1.567	0.081	0.076
60	1.594	1.557	0.037	1.569	0.081	0.077
80	1.594	1.555	0.039	1.568	0.081	0.077
100	1.591	1.558	0.033	1.569	0.081	0.077
120	1.598	1.557	0.041	1.571	0.082	0.077
140	1.602	1.560	0.042	1.574	0.082	0.077
160	1.595	1.558	0.037	1.570	0.081	0.077

of $n_a^{\parallel}$ and $n_a^{\perp}$, increased by the same ratio as given in Figure 12. The isotropic refractive index is increased by increasing the annealing temperature, which is given in Figure 13.

Measurement of the Swelling Factors for Different Liquids

The diffraction of monochromatic light by a laser beam is used for fibers to estimate the average thickness of natural silk fibers annealed at a constant temperature of 150°C for different times. The measurements were made before and after wetting the samples in different solvents.

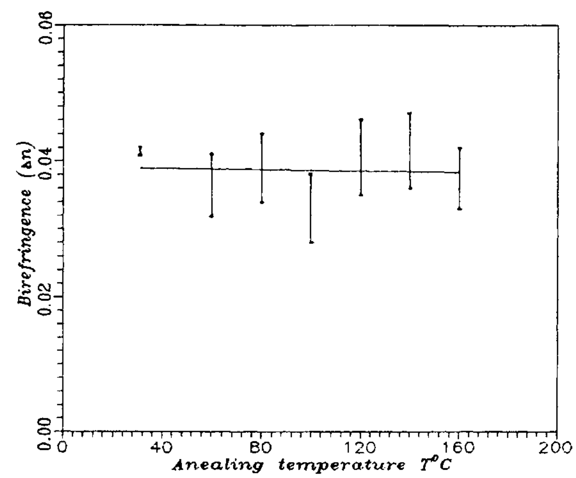


Figure 11 Relation between the birefringence Δn_a of annealed silk fibers and the annealing temperature at a constant time of 2 h.

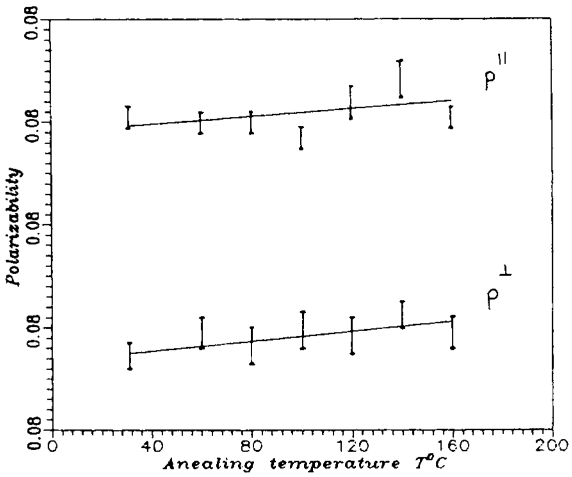


Figure 12 Relation between the polarizabilities $p^{\parallel}$ and $p^{\perp}$ of annealed silk fibers and the annealing temperature at a constant time of 2 h.

Table IV clarifies the results obtained using this method for annealed silk fibers at constant temperature 150°C for different time taking the average value of examined samples. All the samples were wetted in liquids for 1 h at room temperature; then, the average values for the examined fibers were given.

Figure 14 shows the variation of the swelling factor of samples of annealed silk fibers when wetted in water for 1 h. It is clear that the maximum swelling for water is at 5 h, while for wetting these samples in paraffin liquid for 1 h, the curve shows the same behavior of the water curve, but the maximum swelling is at 7 h, as shown in Figure 15.

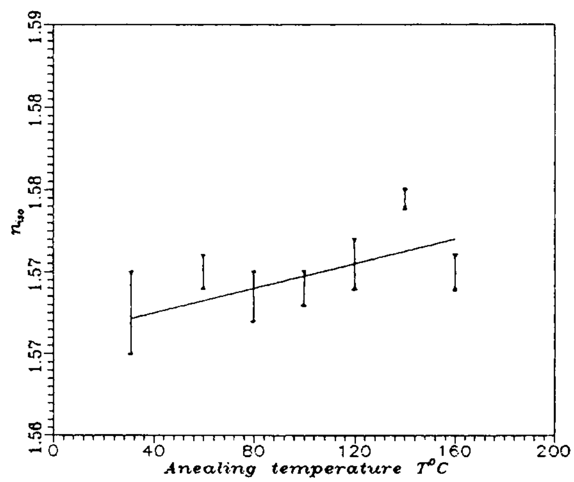


Figure 13 Relation between the isotropic refractive index n_{iso} of annealed silk fibers and the annealing temperature at a constant time of 2 h.

Table IV Swelling Factors of Silk Fibers Annealed at 150°C

Annealing Time (h)	Solvents			
	q_{water}	$q_{paraffin}$	$q_{methanol}$	$q_{acetic\ acid}$
1	7.56	8.19	6.83	4.99
2	10.11	6.11	11.49	6.9
4	12.15	14.08	8.91	12.69
8	8.35	16.67	12	13.79
10	4.08	13.67	9.81	5.82

Figure 16 shows the variation of the swelling factor of the same samples when wetted in acetic acid (pure) for 1 h. The maximum swelling was at 5 h, while when wetting the same samples in methanol liquid, the maximum swelling was at 8 h, as given in Figure 17. From the above results of the swelling factor measurements, it is clear that the range in which the maximum swelling occurred was between 5 and 8 h.

CONCLUSION

From the measurements carried out in the present work to investigate the change in optical properties due to the annealing process for natural silk, the following conclusions may be drawn:

1. The direction dependence of the refractive index was cited by early workers as a proof of the presence of crystalline elements within a fiber. As $n_a^{\parallel}$ decreases with different annealing times (at a constant temperature of

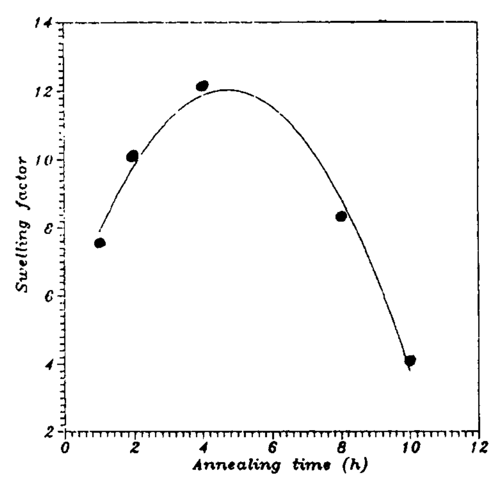


Figure 14 Relation between the swelling factor of annealed silk fibers and the annealing time at a constant temperature of 150°C for water liquid.

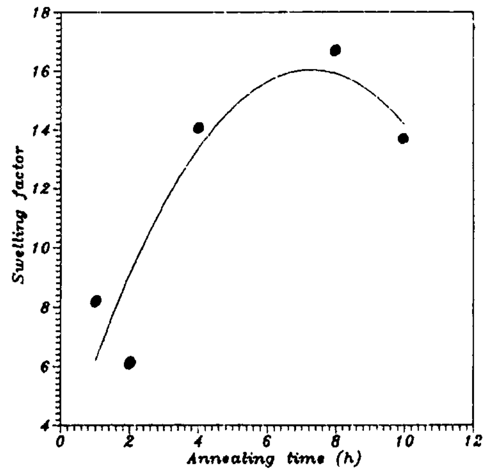


Figure 15 Relation between the swelling factor of annealed silk fibers and the annealing time at a constant temperature of 150°C for paraffin liquid.

100°C), the crystallinity of the fiber must also decrease.

2. Decreasing birefringence at (100°C) means decreasing the crystallized volume in the silk medium.
3. Birefringence decreased at $\simeq$ 2 h and then its value increased at 7 h, thus decreasing and increasing the crystallized volume of the silk medium. The crystallized volume of the material is varied due to the annealing conditions, possibly due to the number of nuclei formed per unit time according to the nucleation process.

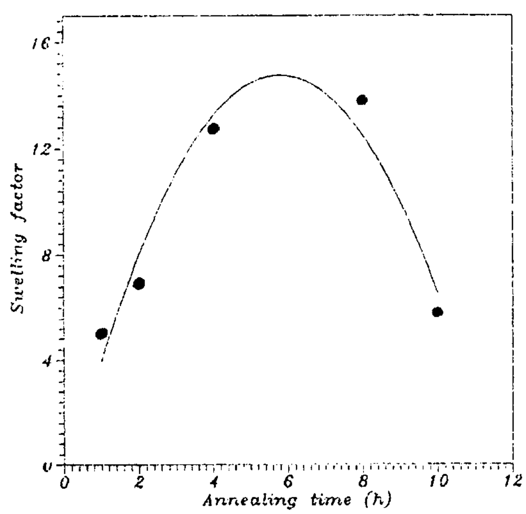


Figure 16 Relation between the swelling factor of annealed silk fibers and the annealing time at a constant temperature of 150°C for acetic acid liquid.

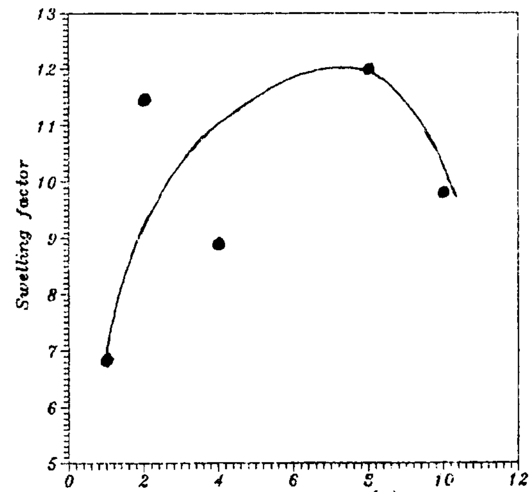


Figure 17 Relation between the swelling factor of annealed silk fibers and the annealing time at a constant temperature of 150°C for methanol liquid.

4. It may be inferred that due to that the annealing process on natural silk fibers depends on the time and temperature of annealing that new crystallized regions are formed.
5. Changes in n_{iso} with annealing time indicate a change in the specific volume of silk fibers on annealing.
6. The value of the swelling factor differs for different liquids and depends on the time and the temperature of annealing and also on the nature of the absorbant liquids and the chemical structure of the fiber.
7. Measuring the absorption factor throws light on the degree of the crystalline and amorphous areas, and, in fact, all degrees of order and disorders area exist.
8. The change of isotropic refractive indices is related to the degree of order and crystallinity of the fiber as well as to the density of the sample[7] $[(n_{iso} - 1)/\rho = k]$.
9. Empirical formulas are suggested for the relation between the mean refractive indices and the annealing time as $n_a^{\parallel} = n_{a_0}^{\parallel} + Ct$ and $n_a^{\perp} = n_{a_0}^{\perp} + Dt$ for the annealed silk fibers at a constant temperature of 100°C; the values of $n_{a_0}^{\parallel}$ and $n_{a_0}^{\perp}$ agree with the refractive indices without annealing. The constants of these formulas characterize the orientation process due to the annealing process.

The present results show good agreement with the previous studies of the effect of annealing on silk fibers.[6]

REFERENCES

1. A. A. Hamza, T. Z. N. Sokkar, and M. A. Kabeel, *J. Phys. D Appl. Phys.*, **18,** 1773 (1985).
2. I. M. Fouda, M. M. El-Tonsy, and K. A. El-Farahaty, *Arab. Gulf J. Sci. Res.*, **8**(2), 61–78 (1990).
3. I. M. Fouda, M. M. El-Nicklawy, K. A. El-Farahaty, and T. El-Dessouki, *Arab. Gulf J. Sci. Res.*, **5,** 378 (1987).
4. A. A. Hamza, I. M. Fouda, K. A. El-Farahaty, and K. A. El-Sayed, *Acta Physiol. Pol. A*, **73**(5) 767 (1988).
5. A. A. Hamza, *J. Microsc. I*, **142,** 35 (1986).
6. I. M. Fouda and M. M. El-Tonsy, *J. Mater. Sci.*, **25,** 21 (1990).
7. M. Tsukada, Y. Goto, G. Freddi, H. Shiozaki, and H. Ishikawa, *J. Appl. Polym. Sci.*, **45,** 1719 (1992).
8. R. W. Moncrieff, *Man-made Fibres*, New Nes–Butterworths, London, 1975, pp. 77–79.
9. S. R. Cockett, *An Introduction to Man-made Fibres*, Sir Isaac Pitman, London, 1966, p. 21.
10. M. Pluta, *Opt. Acta*, **18,** 661 (1971).
11. M. Pluta, *J. Microsc.*, **96,** 309 (1972).
12. J. R. Samuels, *Structured Polymer Properties*, Wiley, New York, 1974, pp. 50–60.
13. W. I. Word, *Structure and Properties of Oriented Polymers*, Applied Science, London, 1975, p. 57.
14. V. Jindricii, B. Brancik, and A. Datyner, *Text. Res. J.*, **47,** 662–665 (1977).

Received April 19, 1995
Accepted October 6, 1995