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X-Ray Study of Crystalline Character and Mechanical Properties of Cotton Fibers Swollen in Zincoxen Solution

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Synopsis

Swelling of cotton in zincoxen solutions of various concentrations was performed for 1 hr at 0°C. The crystallite orientation and the crystallite dimensions of the swollen cotton fibers were determined by x-ray techniques. The swelling in zincoxen improved the orientation and Young's modulus. The relationships between the modulus and (a) the crystallite orientation and (b) the crystallite dimensions were measured, and the correlation was found to be good with the former and poor with the latter.

INTRODUCTION

Zincoxen [tri(ethylenediamine)zinc hydroxide solution] was first prepared by Jayme and Neuschäffer[1,2] as a cellulose solvent. Jayme et al.[3] have used zincoxen and other solvents to study microscopically the fine structure of cellulose pulp. Nevertheless, no work appears to have been reported in the literature on the swelling effect of zincoxen.

In the first paper of this series,[4] the degree of swelling of cotton fiber in zincoxen solutions of various concentrations was investigated and found to be complex. The absorption of the swelling agent and the x-ray diffraction pattern of the swollen fibers were also studied, and it was shown that the swelling in zincoxen was mostly interfibrillar. The present paper describes the swelling effect of zincoxen on the crystalline character of cotton fibers as measured by x-ray diffraction techniques. The interrelations between these and the mechanical properties are also included. Recently, Shenouda and Viswanathan[5,6] have used the term "crystalline character" to cover three aspects, namely, degree of crystallinity, crystallite orientation, and crystallite sizes. However, they concluded that the degree of crystallinity is not very meaningful and has little influence on the strength of the fiber, and thus, crystallinity measurements have not been undertaken in the present study.

EXPERIMENTAL

Samples and Treatments

Giza 66 cotton was used throughout this investigation. The cotton was purified according to a standard procedure.[7] Slack swelling of cotton in zincox-

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en solutions, of different ethylenediamine (EDA) and zinc concentrations, was performed for 1 hr at 0°C; the cotton was then washed in distilled water for 15 min and air dried at room temperature. The details of the preparation of zincoxen and the estimation of the EDA and zinc concentrations were given in a previous paper.[4]

X-Ray Measurements

Crystallite Orientation. X-Ray photographs of bundles of parallel fibers were taken using a flat-film camera. A specimen-to-film distance of 4 cm was maintained throughout the study. Filtered CuK_α radiation from a Philips sealed tube, working at 40 kV and 30 mA, was used. The exposure time was 2 hr. The diffraction patterns were analyzed using a Joyce Leobi Automatic Recording Microdensitometer. The optical density along the arc of the (002) reflection was measured using the rotating table. Before proceeding to measure the intensity of the reflection, the center of the x-ray film was located using a template with a series of concentric circles drawn on it. The film was mounted on the rotating table so that their centers coincided. The table was then moved until the measuring beam lay in the middle of the (002) arc. Both (002) arcs of each fiber bundle were recorded. Then the azimuthal intensities were measured in steps of 5° after subtracting a linear background. By means of a computer program,[5] Hermans' orientation factor[8] f_x

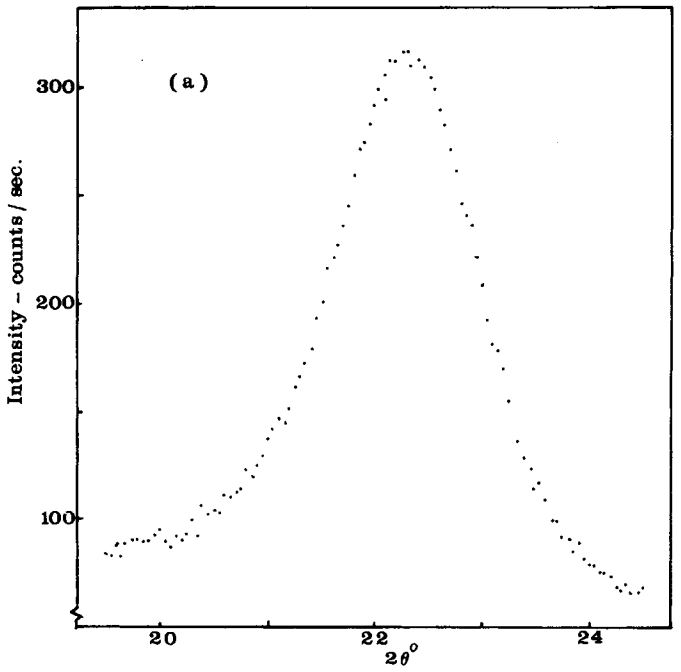

Fig. 1 (*continued*)

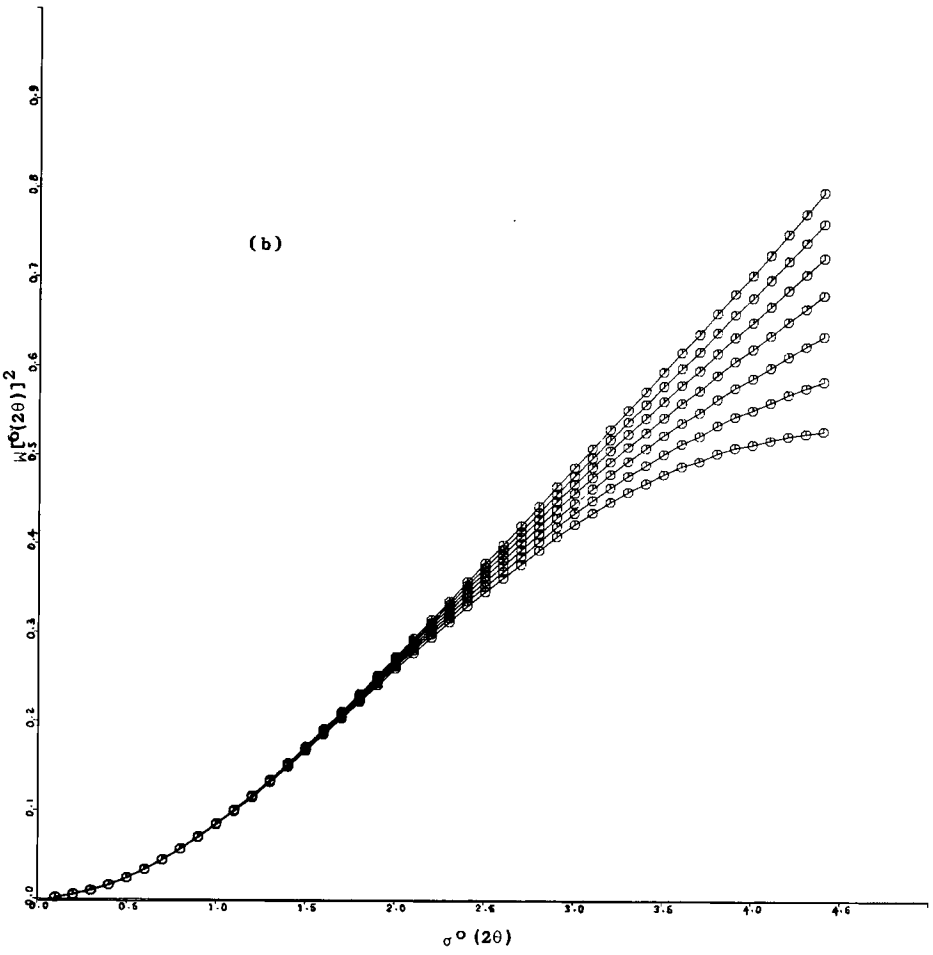

Fig. 1. Measured line profile of 002 reflection of a bundle of cotton fibers (a) and the computed variance range analysis (b).

and the mean inclination angle β to the fiber axis were evaluated according to the following relations:

$$f_x = 1 - \frac{3}{2}\sin^2\beta \tag{1}$$

$$\sin^2\beta = 2\sin^2\alpha \tag{2}$$

$$\sin^2\alpha = \frac{\int_0^{\pi/2} F(\alpha)\sin^2\alpha\cos\alpha\, d\alpha}{\int_0^{\pi/2} F(\alpha)\cos\alpha\, d\alpha} \tag{3}$$

where α and $F(\alpha)$ are the azimuthal angle and the intensity distribution along the (002) reflection, respectively.

Crystallite Dimensions. A Philips wide-angle goniometer PW1050 together with a diffractometer PW1051 and a pulse-height analyzer were used

for a transmission geometry of a fiber bundle. The line profiles of (002) and (040) of cotton were scanned in steps of 0.05° (2θ) using step scanning and fixed time (100 sec) techniques, Figure 1a. Hexamethylenetetramine was used as a standard for correcting the instrumental line broadening. The variance range analysis[9,10] of line profile was used to estimate the apparent crystallite size through a computer program. Other details are as described earlier.[6] Figure 1b shows the variance W versus range σ (where σ is half the angular range of the profile) of the (002) reflection.

Paracrystalline Lattice Distortions. Bonart and co-workers[11] have suggested a means of separating the line breadths due to particle size β_P and paracrystalline lattice distortions β_D from the observed line breadth β_{PD} according to the equation

$$\beta_{PD}^2 = \beta_P^2 + \beta_D^2 = \frac{1}{L_{hkl}^2} + \frac{(\pi g n)^4}{d_{hkl}^2} \tag{4}$$

where L_{hkl} and d_{hkl} are the ideal crystallite size and the repeat distance in the direction perpendicular to (hkl) planes, respectively, and g is the degree of paracrystalline lattice distortions.

If the experimental data are available for at least two orders of reflection from a given set of planes (hkl), eq. (4) could be used to estimate both L and g. This procedure has been used for ramie,[6,12] the highly crystalline naturally occurring cellulosic fiber, since (020) and (040) reflections are experimentally measurable. In extending this study to cotton, where only the (040) reflection is obtained in practice, it is assumed that the ideal crystallite length in cotton would have been equal to that in ramie but for the different degrees of lattice distortions.[6] Substituting in eq. (4) for the ideal length L_{010} obtained from ramie and for β_{PD} calculated in the present work for cotton from the slope of the variance range analysis after correction for instrumental line broadening, the paracrystalline lattice distortions along the fiber axis can be estimated.

Mechanical Properties Measurements

The mechanical properties of native and swollen cotton fibers were determined using an Instron tensile tester according to a standard procedure. A gauge length of 1 cm was used. A single fiber was first fixed on a small paper frame, of 1 cm-length, by rapidly acting adhesive and mounted between the jaws of the Instron. Then, the two sides of the paper frame were cut with a pair of scissors before starting the experiment. Forty fibers were tested from each specimen. The cross-head and chart speeds were 0.5 cm/min and 30 cm/min, respectively. The load cell A (0–50 g range) with 10 g full-scale load calibration was used. All experiments were performed at 65% R.H. and 20°C.

From the load–extension curves, values of initial modulus (static Young's modulus) were calculated. The slope of the initial straight portion of each load–extension curve was measured in terms of grams-per cent extension of the initial gauge length. The average value of this slope was divided by the average tex value of the fiber (obtained by cutting, counting, and weighing 1-cm lengths).

RESULTS AND DISCUSSION

Crystallite Orientation

The effects of the swelling of cotton fibers in zincoxen solutions on Hermans' x-ray orientation parameters (f_x and β) are summarized in Table I and Figure 2. The values obtained for the 50% x-ray angle ψ from the experimental azimuthal intensities are also included for comparison. It must be remembered that the limiting factor in each particular concentration of ethylenediamine (EDA) is the maximum amount of zinc that can be dissolved.

TABLE I
Effect of Swelling in Zincoxen Solutions on the X-Ray Orientation Factor f_x, Angle of Inclination of Crystallites β, and 50% X-Ray Angle ψ

EDA in zincoxen, %	Zn in zincoxen, %	f_x	β, degrees	ψ, degrees
22.3	0.00	0.618	30.94	31.7
22.3	1.10	0.617	30.44	31.8
22.3	1.50	0.610	30.66	31.6
22.3	2.10	0.627	28.91	31.2
22.3	2.40	0.633	29.63	30.3
22.3	2.79	0.625	30.00	31.2
22.3	4.04	0.631	29.76	30.8
22.3	4.91	0.626	29.96	30.6
22.3	6.45	0.646	29.07	30.8
22.3	7.18	0.655	28.64	30.7
27.2	0.00	0.614	30.34	31.4
27.2	1.21	0.647	29.00	29.5
27.2	1.94	0.676	27.67	26.4
27.2	2.16	0.691	27.01	26.4
27.2	2.76	0.717	25.76	26.4
27.2	3.32	0.720	25.60	24.6
27.2	3.63	0.704	28.38	23.4
27.2	4.00	0.699	26.60	26.1
27.2	5.45	0.677	27.66	27.5
27.2	6.62	0.680	27.50	28.0
27.2	7.85	0.682	27.41	28.2
30.4	0.00	0.611	30.61	31.5
30.4	1.09	0.620	30.22	31.2
30.4	2.04	0.632	29.69	31.1
30.4	3.15	0.646	29.07	30.2
30.4	4.05	0.669	28.46	28.9
30.4	4.77	0.699	26.57	27.3
30.4	5.96	—	—	—
30.4	6.91	0.695	26.80	24.9
30.4	7.85	0.685	27.27	25.2
41.4	0.00	0.634	29.62	31.4
41.4	1.07	0.642	29.24	28.2
41.4	1.64	0.665	28.66	28.1
41.4	2.07	—	—	—
41.4	3.30	—	—	—
50.3	0.00	0.623	30.09	30.9
50.3	0.75	0.636	29.52	29.8
50.3	1.41	0.693	26.88	24.6
Native cotton		0.616	30.38	30.5

The results show that, in dilute EDA zincoxen solution (22.3% by weight EDA), the crystallite orientation improves slowly with concentration of zinc in solution. In EDA solutions of intermediate concentrations (27.2% and 30.4%), the orientation first increases with zinc concentration, passes through a maximum (f_x = 0.720 and 0.700, respectively), then decreases again. In concentrated EDA zincoxen solutions (41.4% and 50.3%), the crystallite orientation increases rapdily with concentration of zinc in solution. In 41.4% EDA zincoxen solutions, the cotton fibers start to dissolve at a zinc concentration of about 2%, at first partially and then completely. The fibers also partially dissolve at 30.4% EDA and 6.0% zinc. The highest x-ray orientation factor of 0.720 for cotton was obtained with a zincoxen solution of 27.2% EDA and 3.32% zinc.

The swelling treatment of cotton in zincoxen solution has been found to increase the fiber orientation. This behavior may be attributed to the partial straightening of the convoluted fiber during swelling. Such a deconvolution would align the axes of the crystallites more nearly parallel to the fiber axis. Similar results were obtained for cotton by Radhakrishnan and co-workers[13] and Varma and Bhatia,[14] who worked on the swelling behavior of NaOH and cadoxen solutions, respectively. On the other hand, the interaction of cellulose with zincoxen may affect the chain length and chain length distribution, yet improvement in crystallite orientation due to accompanying swelling may give better and close packing.

It must be mentioned that, despite the high degree of swelling of cotton

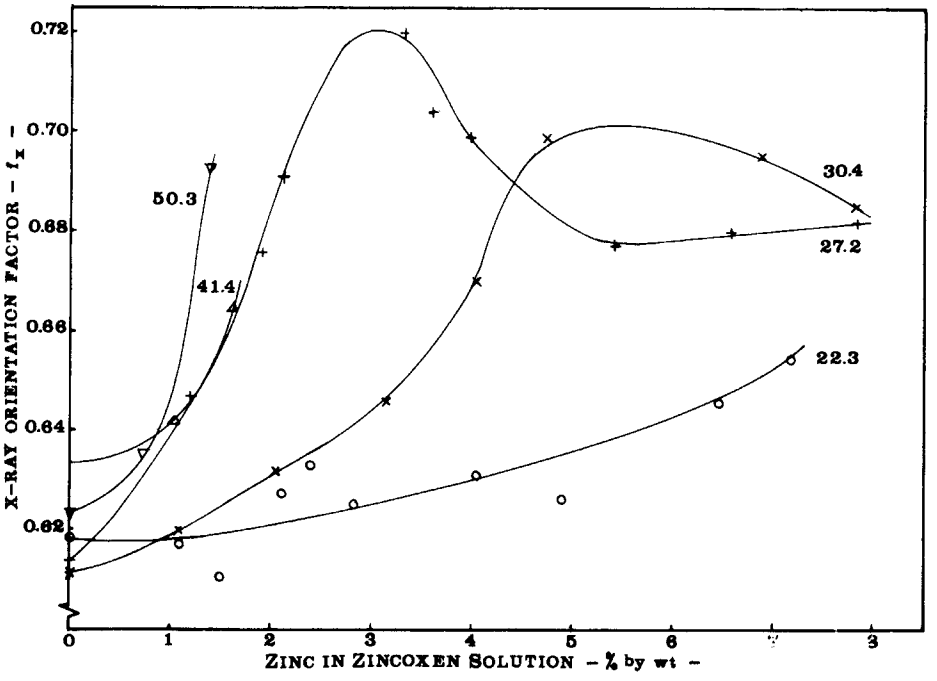

Fig. 2. Effect of swelling in zincoxen solutions on the x-ray orientation factor of cotton fibers. The concentration of EDA (wt-%) in each solution is indicated on the figure and on the consequent ones.

fiber achieved in zincoxen solutions, no evidence could be found by x-ray methods for any lattice change of the swollen fibers.

Crystallite Dimensions

The effects of zincoxen solutions of different concentrations on the lateral size and length of the cellulose crystallites of cotton are shown in Figures 3 and 4 and Table II. In this study, the apparent crystallite size estimated only from the slope of the variance curve, Figure 1b, has been reported. It is seen from the previous work[6] that the slope estimate is less susceptible to the residual strain in the reference material[10] and values obtained for the crystallite size are in fair agreement with the ones reported in the literature. These values are derived from the slope of the variance range function without taking the paracrystalline lattice distortions into account. It must, however, be emphasized that although the corrected slope is used to obtain an estimate of the crystallite size, such numerical results are highly affected by the prevalence of lattice distortions.

Table II shows that the lateral size of the crystallites for the raw and zincoxen-treated samples range from 48 Å to 70 Å, while the crystallite length ranges from 73 Å to 105 Å. It may be mentioned in passing that the values obtained for the crystallite lengths, using the variance-range analysis for cotton, are in accordance with the values obtained by Kulthreshtha and coworkers[15] for tufcel using the same method, but are smaller than the values reported before for cotton using other x-ray methods. On the other hand, Nieduszynski and Preston[16] have obtained a value of 61 Å for the lateral crystallite dimension from the line broadening of (002) reflection of cotton using the Fourier analysis method of Stokes.[17] The effect of zincoxen solution on the crystallite dimensions is complex and can be summarized as follows: In

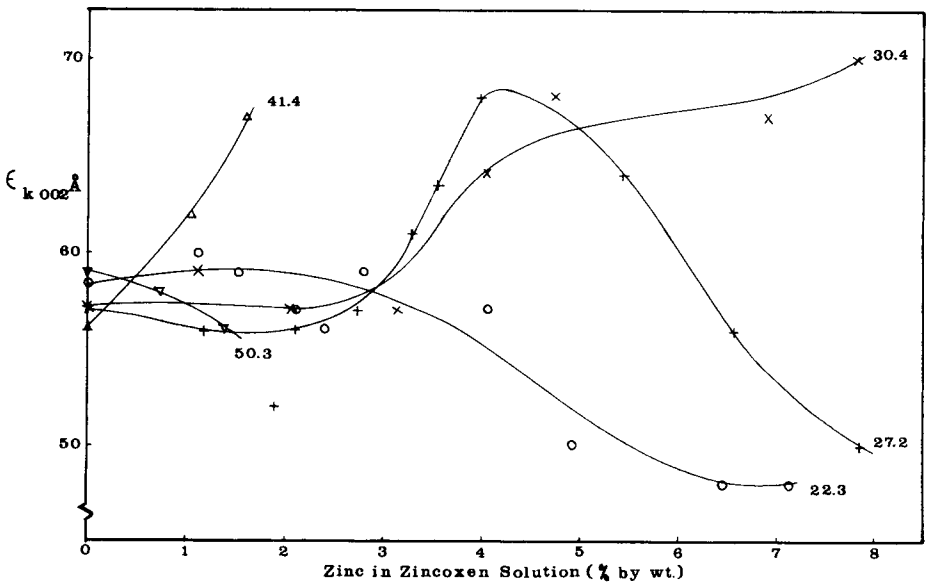

Fig. 3. Effect of swelling in zincoxen solutions on the lateral crystallite sizes of cotton.

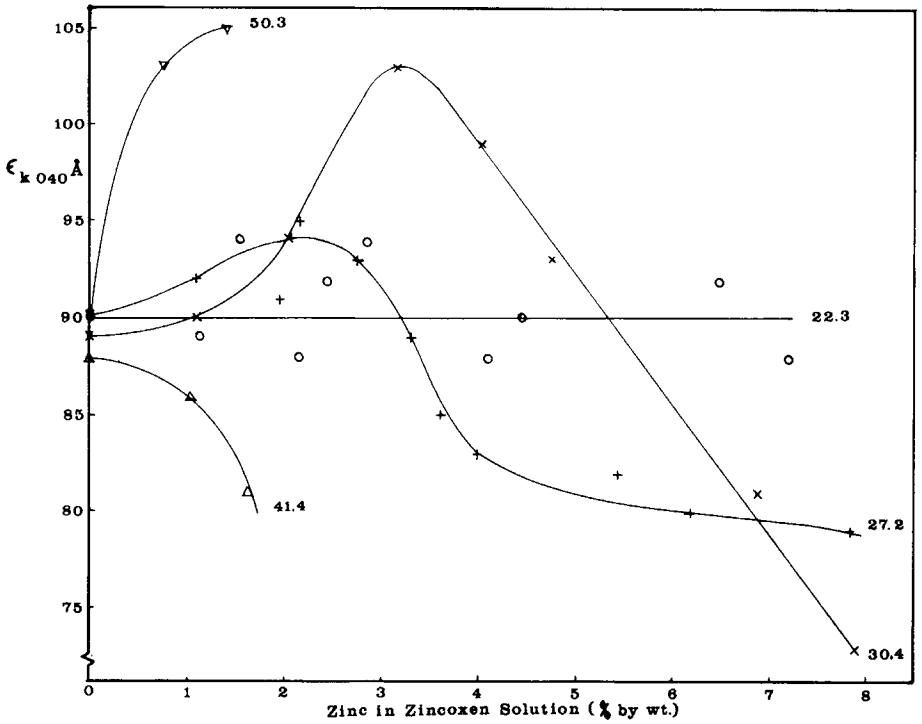

Fig. 4. Effect of swelling in zincoxen solutions on the crystallite length of cotton fibers.

dilute EDA zincoxen solutions (22.3% by weight EDA), the lateral sizes decrease at high zinc concentrations, while the crystallite lengths remain unchanged. In 27.2% EDA solutions, the crystallite dimensions first increase with zinc concentration, pass through maxima, and then decrease again. These maxima were at about 4% and 2.5% zinc for lateral and length of crystallites, respectively. In zincoxen solutions of intermediate concentrations of EDA (30.4%), the lateral sizes increase with zinc concentration, but the crystallite lengths first increase, pass through a maximum at about 2.5% zinc, and then decrease again. In 41.4% EDA solutions, the lateral sizes increase with zinc concentration, but the crystallite lengths decrease. In the more concentrated EDA solutions of 50.3%, the crystallite lengths increased with zinc concentration, while the lateral sizes remained virtually unchanged.

Paracrystalline Lattice Distortions

The values obtained for the degree of paracrystalline lattice distortions g in the axial direction of the zincoxen-treated cotton samples are given in Table II. The values of g range from 2.34% to 2.90%, while the mean value is found to be 2.59%. In dilute EDA solutions (22.3%), the values of g do not show any trend with zinc concentrations. In zincoxen solutions of intermediate concentrations of EDA (27.2% and 30.4%), g values first decrease with zinc concentration, pass through a minimum, and then increase again. In 41.4% EDA solutions, g values increase with zinc concentration, while in the case of 50.3%

TABLE II
Effect of Swelling in Zincoxen Solutions on the Dimensions of the Crystallites and the Paracrystalline Distortions of Cotton Fiber

EDA in zincoxen, %	Zn in zincoxen, %	(002) ϵ_k, Å	(040) ϵ_k, Å	g, %
22.3	0.00	58	90	2.57
22.3	1.10	60	89	2.59
22.3	1.50	59	94	2.51
22.3	2.10	57	88	2.61
22.3	2.40	56	92	2.54
22.3	2.79	59	94	2.51
22.3	4.04	57	88	2.61
22.3	4.91	50	90	2.57
22.3	6.45	48	92	2.54
22.3	7.18	48	88	2.61
27.2	0.00	58	90	2.57
27.2	1.21	56	92	2.54
27.2	1.94	52	90	2.57
27.2	2.16	56	95	2.49
27.2	2.76	57	93	2.52
27.2	3.32	61	89	2.59
27.2	3.63	64	85	2.66
27.2	4.00	68	83	2.70
27.2	5.45	64	82	2.72
27.2	6.62	56	80	2.76
27.2	7.87	50	79	2.78
30.4	0.00	57	89	2.59
30.4	1.09	59	90	2.57
30.4	2.04	57	94	2.51
30.4	3.15	57	103	2.37
30.4	4.05	64	99	2.43
30.4	4.77	68	93	2.52
30.4	5.96	—	—	—
30.4	6.91	67	81	2.74
30.4	7.85	70	73	2.90
41.4	0.00	56	88	2.61
41.4	1.07	62	86	2.64
41.4	1.64	67	81	2.74
41.4	2.07	—	—	—
41.4	3.30	—	—	—
50.3	0.00	59	90	2.57
50.3	0.75	58	103	2.37
50.3	1.41	56	105	2.34
Native cotton		57	89	2.59

EDA solutions, the reverse is true. It is seen that some of the treatments increased the lattice distortions while some others reduced them. Using the infrared-deuteration technique for cadoxen-treated cotton, Evans and Jeffries[18] have shown that the hydrogen bond disorder increases with the increase in cadmium content of the swelling solution.

Mechanical Properties

From the load–elongation curves of single fibers of native and swollen cotton, the initial modulus, strength, and elongation at break have been calculat-

TABLE III
Mechanical Properties of Swollen Cotton Fibers in Zincoxen Solutions[a]

Composition of zincoxen		Strength		Elongation		Initial modulus	
EDA, wt-%	Zn, wt-%	g/tex	C.V., %	%	C.V., %	g/tex	C.V., %
22.3	0.00	37.9	(31)	9.3	(30)	613	(29)
22.3	1.10	37.4	(41)	9.8	(36)	616	(39)
22.3	1.50	36.7	(32)	10.0	(28)	616	(31)
22.3	2.10	37.8	(35)	9.2	(34)	717	(32)
22.3	2.40	35.7	(32)	9.5	(23)	646	(23)
22.3	2.79	35.8	(32)	9.3	(22)	650	(37)
22.3	4.04	35.2	(38)	8.6	(30)	582	(29)
22.3	4.91	36.4	(32)	9.2	(29)	751	(42)
22.3	6.45	37.5	(34)	7.6	(28)	805	(29)
22.3	7.18	35.9	(34)	7.4	(29)	790	(30)
27.2	0.00	35.6	(30)	7.7	(36)	612	(34)
27.2	1.21	35.6	(31)	7.3	(32)	625	(31)
27.2	1.94	36.5	(32)	10.7	(35)	679	(40)
27.2	2.16	35.2	(31)	8.2	(26)	709	(37)
27.2	2.74	36.4	(28)	8.7	(26)	772	(35)
27.2	3.32	36.8	(32)	10.0	(29)	861	(29)
27.2	3.63	28.2	(37)	8.6	(43)	1010	(33)
27.2	4.00	38.5	(37)	6.7	(40)	1000	(34)
27.2	5.45	37.1	(30)	6.5	(31)	1017	(34)
27.2	6.62	35.8	(31)	6.9	(28)	1000	(28)
27.2	7.87	32.8	(44)	5.5	(35)	996	(25)
30.4	0.00	37.6	(41)	7.2	(42)	661	(33)
30.4	1.09	38.1	(38)	7.0	(37)	722	(29)
30.4	2.04	38.0	(34)	6.7	(32)	901	(31)
30.4	3.15	38.1	(41)	6.4	(37)	1029	(25)
30.4	4.05	36.6	(33)	6.4	(31)	945	(28)
30.4	4.77	30.2	(44)	7.3	(38)	844	(27)
30.4	5.96	—	—	—	—	—	—
30.4	6.91	—	—	—	—	—	—
30.4	7.85	29.3	(28)	5.9	(34)	800	(34)
41.4	0.00	33.1	(35)	7.5	(30)	734	(33)
41.4	1.07	36.6	(40)	7.6	(43)	890	(25)
41.4	1.64	35.2	(34)	7.1	(32)	766	(31)
41.4	2.07	—	—	—	—	—	—
41.4	3.30	—	—	—	—	—	—
50.3	0.00	35.8	(31)	7.0	(33)	781	(30)
50.3	0.75	34.9	(44)	5.6	(42)	924	(29)
50.3	1.41	35.0	(39)	6.6	(34)	979	(22)
Raw cotton		38.3	(33)	5.7	(20)	993	(30)

[a] C.V. = Coefficient of variation.

ed, and the results are given in Table III. It is seen from the results that most of the swollen cotton fibers show a loss of tensile strength compared to native cotton. This reduction in strength of the swollen fibers in zincoxen is quite similar to the ones obtained on slack mercerization. The results also indicate that there is no definite relationship between the strength and the zinc content of the swelling solution. However, it can be seen that maximum breaking strength is obtained from swelling cotton in zincoxen solution of

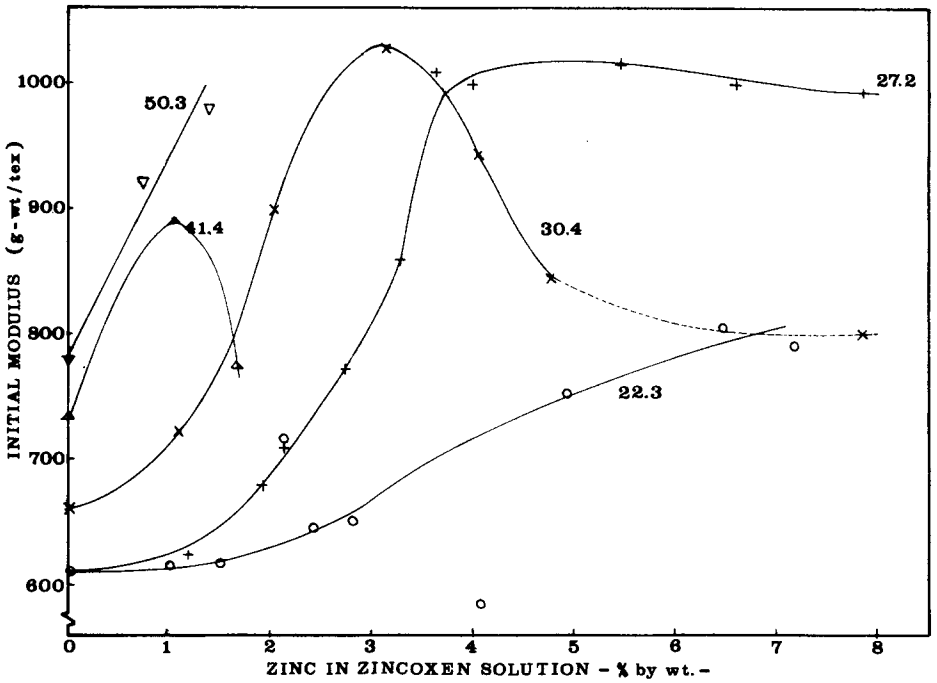

Fig. 5. Effect of swelling in zincoxen solutions on the modulus of cotton fibers.

27.2% EDA and 4% zinc. On the other hand, the swelling has considerably increased the elongation at break from 5.7% for native cotton fibers to 10.7% for swollen fibers. As with strength, so also the elongation at break of swollen fibers does not reflect any definite trend.

Figure 5 illustrates the effect of swelling in zincoxen solutions on the initial (Young's) modulus of cotton fibers. It is seen from the figure that in zincoxen solutions of the lowest and the highest EDA concentrations (22.3% and 50.3%), the modulus increases with zinc concentration. In EDA solutions of moderate concentrations (27.3%, 30.4%, and 41.4%), the modulus first increases rapidly with zinc concentration, passes through a maximum, and then decreases again. It is clear from the results that there are critical zinc concentrations in zincoxen solutions which are responsible for maximum modulus values of swollen cotton fibers, and any increase or decrease in zinc content sharply reduces the modulus.

Modulus–Crystalline Character Relationship

Table IV gives a complete picture of the correlations between the initial (Young's) modulus of the swollen fibers in zincoxen solutions and the various crystalline characteristics investigated in the present study. It is known that the changes in mechanical properties of the fibers can be attributed almost exclusively to the changes in the orientation. There is an obvious flaw in correlating mechanical properties like the initial modulus with crystallite orientation factor, rather than overall longitudinal orientation. However, it is assumed by analogy to earlier reported work[19] that trends in overall longitudi-

TABLE IV
Coefficients of Correlation r Between Initial Modulus and Crystalline Characteristics

Between initial modulus and	r	Significance level[a]
Hermans' orientation factor f_x	0.572	1%
50% x-ray angle ψ	−0.545	1%
Apparent crystallite length ϵ_{k040}	−0.025	N.S.
Apparent lateral size ϵ_{k002}	0.209	N.S.
Paracrystalline distortions g	0.068	N.S.
Amount of zincoxen absorbed	0.569	1%
Amount of zinc absorbed	0.700	1%

[a] N.S. = Not significant.

nal order are duplicated by trends in x-ray orientation factor. It is seen from Table IV that the orientation factor and the 50% x-ray angle are well correlated with Young's modulus of the fiber. It appears, therefore, that the swelling effect of zincoxen solutions is sufficient to separate the microfibrils. This displacement of the microfibrils and molecular chains can result in improved orientation along the fibre axis, which accounts for the change in the mechanical properties.

According to the results expressed in Table IV, the apparent crystallite dimensions have no correlation with the modulus of the fibers. This indicates that the effect of various concentrations of zincoxen solution altered the crystallite dimensions. Between critical concentrations of zincoxen (e.g., 41.4% EDA and 2.07% zinc), cellulose will completely dissolve[20] as has previously been reported.[4] At concentrations close to the critical compositions, the solvent action is reduced, but a powerful swelling effect is evident. In these concentrations, the fibers are disrupted.[4] The x-ray line profile depends on both the size of the crystallites and their perfection. In the present study, both parameters are influenced by the swelling effect of zincoxen. From the results shown in Table IV, it is confirmed that the modulus of the fibers is not correlated with the degree of paracrystalline lattice distortions.

Besides the correlations of the initial modulus with the crystalline characteristics, the correlations for the amounts of zincoxen and zinc absorbed by the fibers, as described before,[4] are also included in Table IV. A strong dependence was found between the initial modulus and the amounts of zincoxen or zinc absorbed during the swelling. As mentioned in the previous paper,[4] the swelling of cotton in zincoxen solutions is mostly interfibrillar. It appears, therefore, that the absorbed zinc must be associated with the cellulose on the surface of the microfibrils. So it is the amount of swelling agent absorbed that helps to separate and interact (through hydrogen bonding) between microfibrils, which produces further access to the microfibrils. This would also explain the marked improvement of crystallite orientation and initial modulus of the swollen cotton fibers.[21]

CONCLUSIONS

The swelling of cotton in zincoxen solutions at specific conditions for maximum swelling markedly improve the crystallite orientation and initial modu-

lus. The effect of swelling in zincoxen solutions of different concentrations on the crystallite dimensions and paracrystalline lattice distortions did not show any definite trend.

The crystalline character–modulus relationship of the zincoxen-treated cotton showed that the orientation parameters are well correlated with Young's modulus, while, on the other hand, the crystallite dimensions are not. Also, a strong dependence was found between the modulus and the amount of zincoxen or zinc absorbed by the fiber during the swelling.

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