NOTE

Questions About Constancy of True Spiral Angle in Cotton

INTRODUCTION

X-ray orientation studies in never-dried cotton reported in this journal indicated that much of the differences in orientation factor found among different cotton varieties in the air-dried state is attributable to the presence of convolutions.¹ These authors¹ used the 50% X-ray angle as a measure of the average spirality for computing the values of true spiral angles in cotton. But 40 and 50% Xray angles are purely arbitrary measures,²⁻⁵ although 75% X-ray angles have been shown to be a closer measure of the true spiral angles in native cotton.⁶ A more accurate measure of spirality for each of the cotton varieties studied by Iyer et al.¹ should have been the average angle of crystallite orientation $(\alpha_m)^{2-5}$ deduced from the $\overline{\sin^2 \alpha_m}$ values using the Hermans expression [Eq. (1)], instead of the 50% X-ray angles, when the Hermans crystallite orientation factors in respect of both the air-dried and solventdried fibers of varieties of the four species had actually been measured by them.¹

$$f = 1 - \frac{3}{2} \overline{\sin^2 \alpha_{\rm m}} \tag{1}$$

where α is the angle made by the <u>molecular</u> chain in the crystallite with the fiber axis and $\sin^2 \alpha_m$ is the average value of $\sin^2 \alpha$. If α_{hkl} is the angle between hkl and the equator, then the average values of the distribution factor on the right of the Eq. (1) are determined from the azimuthal intensity scans of (002) and (101, $10\overline{1}$) taken as combined reflection and using the following relationship and the graphical integration procedure of Hermans.²⁻⁵

and

$$\overline{\sin^2 \alpha_{hkl}} = \frac{\int_0^{\pi/2} I(\alpha_{hkl}) \sin^2 \alpha_{hkl} \cos \alpha_{hkl} \, d\alpha_{hkl}}{\int_0^{\pi/2} I(\alpha_{hkl}) \cos \alpha_{hkl} \, d\alpha_{hkl}}$$

 $\overline{\sin^2 \alpha_m} = (\overline{\sin^2 \alpha_{002}} + \overline{\sin^2 \alpha_{101}})$

Journal of Applied Polymer Science, Vol. 44, 1121–1124 (1992) © 1992 John Wiley & Sons, Inc. CCC 0021-8995/92/061121-04\$04.00 The reduction in the range of true spiral angle values (calculated by subtracting the convolution angle from the 50% X-ray angle) in the solvent-dried cotton as compared to the air-dried cotton of the same varieties is attributed to the elimination or a substantial reduction in the number of convolutions on cotton fibers.¹ Basically it is inappropriate to take the range within genetically diverse species of cotton for convolutions, since it is known that the fibers of *Gossypium hirsutum* varieties are the most convoluted as compared to the fibers of *Gossypium barbadense* and the number of convolutions on fibers of *Gossypium barbadense* and the number of convolutions on fibers of *Gossypium herbaceum* and *Gossypium arboreum* species are far less so.⁷⁻¹¹.

A reexamination of the data reported by P. Bhama Iyer et al.¹ and their hypothesis proposed in respect to the variation in orientation due to the presence of convolutions on cotton is reported here. The average angle of crystallite orientation (α_m) has been used as a measure of spirality instead of the 50% X-ray angle. The following discussion indicates that the conclusion of these authors¹ leads to erroneous information about the structure of cotton fiber hitherto known.

EXPERIMENTAL

The values of the average angles of crystallite orientation (α_m) were calculated using the Hermans expression given in Eq. (1) and the values of the Hermans crystallite orientation factor of air-dried and solvent-dried cotton varieties of the four species reported by Iyer et al.¹ The true spiral angles were then computed by subtracting the values of convolution angles $(\theta)^1$ from the (α_m) , applying the same logic as explained by these authors.¹ The recalculated data are presented in Table I.

RESULTS AND DISCUSSION

It is evident from Table I that the Hermans crystallite orientation factor¹ is higher for the solvent-dried cotton in all the four species than for the corresponding air-dried fibers, indicating increased orientation of cellulose crystallites to the fiber axis. Even the α_m values, calculated (Table I, columns 2 and 6) in respect of these cotton varieties, also indicate the same conclusion, i.e., lower spiral

Sr. No.	Species and	Air-dried Cotton (AD)				Solvent-dried Cotton (SD)			
		f(AD)	$lpha_{m(AD)}$ (°)	Convolution Angle (°)	True Spiral Angle (°)	$f_{(\mathrm{SD})}$	$\alpha_{m({ m SD})}$ (°)	Convolution Angle (°)	True Spiral Angle (°)
No.	Variety of Cotton	1	2	3	4	5	6	7	8
	G. barbedense								
1	Giza-7	0.63	29.76	10.65	1911	0.70	26 56	1 33	25.23
2	ERB-4600	0.66	28.42	6.57	21.85	0.71	26.07	1.38	20.20
3	SUVIN	0.69	27.03	7.63	19.40	0.71	26.07	1.63	24.00
4	IBSI-25	0.69	27.02	6.97	20.05	0.71	26.07	0.90	25.17
5	IBSI-53	0.72	25.58	7.78	17.80	0.75	24.08	1.25	22.83
6	Avg. within the species	0.678	27.56	7.92	19.64	0.716	25.77	1.298	24.47
7	Range within the species	0.09	4.18	4.08	4.05	0.05	2.48	0.73	2.40
	G. hirsutum								
8	IAN-579	0.60	31.08	12.35	18.73	0.73	25.10	1.41	23.69
9	MCU-5	0.63	29.76	12.10	17.66	0.72	25.58	1.17	24.41
10	HH-35	0.62	30.21	11.13	19.08	0.71	26.07	1.60	24.47
11	Hybrid-4	0.64	29.32	8.68	20.64	0.72	25.58	1.33	24.25
12	Hybrid-5	0.69	27.03	6.40	20.63	0.73	25.10	1.91	23.19
13	G.COT. 11	0.61	30.65	9.58	21.07	0.70	26.56	1.95	24.61
14	G.COT. 10	0.68	27.50	7.28	20.22	0.70	26.56	2.65	23.91
15	IAN-4975	0.67	27.96	8.23	19.73	0.75	24.08	2.63	21.45
16	Avg. within the species	0.642	29.18	9.46	19.72	0.720	25.58	1.83	23.74
17	Range within the species	0.09	4.05	5.95	3.41	0.05	2.48	1.48	3.16
	G. arboreum								
18	SANJAY	0.74	24.59	4.70	19.89	0.75	24.08	0.70	23.38
19	K-9	0.63	29.76	5.41	24.35	0.70	26.56	2.13	24.43
20	AKH-4	0.68	27.50	5.50	22.00	0.69	27.01	0.83	26.18
21	AKH-235	0.65	28.88	6.47	22.41	0.69	27.01	1.28	25.73
22	Avg. within the	0.675	27.68	5.52	22.16	0.707	26.16	1.23	24.93
	species								
23	Range within the species	0.11	5.17	1.77	4.46	0.06	2.93	1.43	2.35
	G. herbaceum								
24	SUYODHAR	0.63	29.76	6.47	23.29	0.72	25.58	1.60	23.98
25	JAYADHAR	0.64	29.32	7.27	22.05	0.72	25.58	1.38	24.20
26	SUJAY	0.71	26.07	4.40	21.67	0.73	25.10	1.05	24.05
27	DIGVIJAY	0.65	28.88	6.80	22.08	0.71	26.07	1.11	24.96
28	Avg. within the	0.657	28.50	6.23	22.27	0.720	25.58	1.285	24.27
29	species Range within the	0.08	3.69	2.87	1.62	0.02	0.97	0.55	0.98
	species	0.00	0.00						0.00
30	Combined avg. within all the species	0.663	28.23	7.28	20.94	0.715	25.77	1.41	24.35
31	Range within all the species together	0.14	6.49	7.95	6.55	0.06	2.93	1.85	4.73

Table IHermans Orientation Factor, Average Angle of Crystallite Orientation Convolution Angle, andTrue Spiral Angles in Air-Dried and Solvent-Dried Cotton Fiber

angles correspond to increased orientation to the fiber axis. However, the true spiral angles of the air-dried fibers (Table I, columns 4 and 8) in all the four individual species are smaller than the true spiral angles of the corresponding solvent-dried fibers. The average values of the true spiral angles for solvent-dried cotton of all four species reported here (Table I, column 8) is very close to the value 24.25 \pm 3.34° for air-dried Indian cottons reported by Betrabet et al.¹² The average value of true spiral angles for air-dried cotton of all the four species on the other hand are very close to the value (ca. 21°) reported by Meredith.^{13,14} Most appropriately the true-spiral angle values for the same cotton varieties, corrected for the contribution of convolutions, independently for air-dried and solvent-dried fibers, must be nearly identical within limits of experimental error. However, the original data of true spiral angle of Iyer et al.¹ and the recalculated data (Table I, columns 4 and 8) indicate that the true spiral angle values for the air-dried cotton are smaller than the corresponding values for the solvent-dried fibers in all the varities, irrespective of the species of cotton. This is contrary to the expectation since the Hermans orientation factors for solvent-dried cotton is higher than that for the air-dried cotton and correspondingly the α_m values for the solvent-dried cotton are smaller than the α_m values for the air-dried cotton. These discrepancies imply several possibilities:

- 1. The convolution angles for the solvent-dried cotton are considerably undervalued in measurement. If not,
- 2. The higher values of true spiral angle for the solvent-dried cotton (Table I, column 8) deny the assumption¹ that the solvent exchange dehydration procedure has not modified the spiral structure of the cotton, and the solvent-dried cotton conforms to the never-dried state.
- 3. Alternately, if the solvent exchange dehydration procedure retains the never-dried state, then the presence of convolutions on fibers in fact decreases the true spiral angle in cotton as evident from Table I, columns 4 and 8. And since lower spiral angle values correspond to increased strength of cotton fibers as pointed out by several workers, ^{2-5,11-16} the presence of convolutions on fibers must be taken to add to the strength of fibers. This interpretation is in direct conflict with the observations of Meredith and other workers.^{8,9,12,13,17} Moreover, it may be interesting to note from Table I, Sr. Nos. 30 and 31, columns 3 and 7, that the range of variation in the convolution angles in all the four species of cotton reported by Iyer et al.¹ is more than the average value for the species.

Therefore, the objection to the use of the reduced range in convolution angle in solvent-dried cotton of diverse genetic species as a measure of the contribution of convolutions to spiral angle is fully justified. It is therefore clear that unless the discrepancies as pointed out above in respect of the true spiral angle are satisfactorily explained, it would be premature to conclude that all observed differences in the orientation of air-dried cotton are attributable to the presence of convolutions. In reality, such differences must be seen to arise as a result of the complex genotype-metabolic-environmental interaction at the place of growth of cotton^{18,19} in view of the recently known facts that there are no basic differences between the mass density of cellulose in never-dried fibers,²⁰ orientation of crystallites,²¹ and the size of the crystallographic units of cellulose²² within different varieties and species of cotton and the argument that the reasons for apparent differences in cotton varieties must be sought in some higher order of structural organization, 20-23 because cellulose synthesis, packing, cell-wall thickness, and convolution number induced are very much interrelated with each other.

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A. V. MOHARIR

Laboratorium de Meulemeester voor Technologie der Textielstoffen State University of Ghent Grote Steenweg Noord-2, 9052, Zwijnaarde Gent, Belgium

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