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Structure and Properties of Natural Cellulose Fibers Obtained from Sorghum Leaves and Stems

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For the first time, sorghum leaves and stems have been used to produce natural cellulose fibers with properties suitable for composite, textile, and other high-value fibrous applications. The leaf and stems fibers produced are multicellular and have similar cellulose contents. The breaking tenacity and elongation of the fibers are similar to that of natural cellulose fibers such as kenaf and cornstalk fibers. However, the sorghum fibers have a modulus of about 113 g/denier (15 GPa) similar to the modulus of cornstalk fibers but higher than that of cotton and cornhusk fibers. At least 7 million tons of natural cellulose fibers can be produced by using the sorghum stems and leaves available as byproducts every year. Using the sorghum byproducts as a source for cellulose fibers will help to add value to the sorghum crops and also make the fiber industry more sustainable.

KEYWORDS: Cellulose; biofibers; sorghum; value addition

INTRODUCTION

Sorghum is one of the major food crops of the world, and about 58 million tons of sorghum grains was produced in 2005 (*1*). The production of sorghum also leaves about 58 million tons of byproducts mainly composed of cellulose, hemicellulose, and lignin. These byproducts are of limited use at present and are a cheap and abundant source that could be used to produce natural cellulose fibers for textiles, composites, and other fibrous applications. Although it has been reported that sorghum stems could be potential feedstock for the paper and pulp industry, the characteristics of the pulp produced or the properties of the sorghum fibers (single or ultimate cells) used for paper have not been reported (*2*−*4*). In addition, the pulping process reduces the cellulose in sorghum to single cells that are about 1−2 mm in length and not suitable for textile and other high-value fiber applications. High-value fiber applications require fibers to have lengths of at least 25 mm with adequate strength, elongation, and other properties. Such long length (>25 mm) fibers are formed by a bundle of single cells held together by lignin and other binding materials. To the best of our knowledge, no previous reports are available on the production of such fiber bundles from sorghum leaves or stems.

Producing fibers from byproducts of major agricultural crops such as the sorghum will not only add value to the crops but also make the fiber industry more sustainable. There are several disadvantages of using both the natural and the synthetic fibers currently in use. For example, synthetic fibers are from the nonrenewable petroleum resource, and the natural fibers need scarce land, water, and other resources to grow. Therefore, attempts are being made to use alternative sources to produce natural cellulose, protein, and synthetic fibers from various annually renewable resources. Such attempts have been successful to produce high-quality natural cellulose fibers from cornhusks, cornstalks, rice straw, pineapple leaves, sugarcane rind and switchgrass, protein fibers from wheat gluten, soy proteins, and zein and synthetic fibers such as corn from PLA (*5*−*14*). However, some of these fibers have poor properties, and efforts are being made to understand and improve the properties of the fibers (*15*−*18*). Similar to these crops, sorghum stover could also be used to obtain natural cellulose fibers. Because most of the sorghum is grown in dry and arid climates, especially in developing countries, using the byproducts will help to add value to the crop and benefit the farmers economically.

In this paper, we have used both sorghum stems and leaves as sources for natural cellulose fibers. Cheap and environmentally safe chemicals were used to obtain the fibers, and the fibers obtained were characterized for their structures and properties. Our goal was to obtain fibers from sorghum stems and leaves with the length, strength, and other properties required for high-value fibrous applications.

EXPERIMENTAL PROCEDURES

Materials. Sorghum stover was collected from a research field at the University of Nebraska—Lincoln, 21 days after postharvest. The outer leaves and the stems in the stover were used separately for fiber extraction. Sodium hydroxide, acetic acid, chromic acid, and nitric acid were reagent grade chemicals purchased from VWR International (Bristol, CT). The structures and properties of sorghum fibers have been compared with fibers obtained from cornhusks, cornstalks, and also cotton and linen. The data for cornhusks, cornstalks, cotton, and linen are from the literature (*6*−*8*, *19*). We have included a range of data for cotton and linen to cover the values reported by several authors.

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Table 1. Composition (% on Dry Weight) of the Sorghum Leaf and Stem Fibers

material	cellulose	lignin	ash
sorghum leaves	64.8 ± 1.7	9.0 ± 0.7	2.9 ± 0.7
sorghum stems	65.1 ± 2.1	6.5 ± 1.1	1.2 ± 0.4

Fiber Extraction. A series of experiments were conducted to develop optimum conditions of fiber extraction. This optimization process included varying the extraction conditions such as the concentration of sodium hydroxide, time and temperature of treatment, and the liquor to material ratio used. The range of extraction variables studied was selected based on our previous experiences in extracting fibers from corn stover, rice straw, and switchgrass (*6−9, 11*). Our goal was to obtain the finest possible fibers with the highest yield and with good tensile properties. Under the optimized conditions developed, the sorghum leaves were treated using a 2% (w/w) sodium hydroxide solution at 95 °C for 30 min using a liquor to material ratio of 10 to 1. Similarly, the sorghum stems were treated with a 4% (w/w) sodium hydroxide solution at 95 °C for 45 min using a liquor to material ratio of 15 to 1. The treated fibers were washed in water to remove the dissolved substances and to obtain the fibers. The fibers obtained were neutralized using a 2% (w/w) acetic acid solution, washed again, and air-dried.

Fiber Composition. The amount of cellulose in the sorghum leaf and stem fibers was determined using the acid detergent fiber method according to AOAC method 973.18. Lignin in the fibers was determined as Klason lignin according to ASTM standard method D1106-96. Ash in the fibers was determined according to ASTM standard E1755-01. Two replications were done for each compositional analysis, and the average ± one standard deviation is reported.

Physical Structure. The physical structure of the fibers was studied in terms of the % crystallinity, crystallinity index (CI), multifibrillar angle (MFA), and the orientation of the cellulose crystals by observing the diffraction patterns of the fibers. Two types of X-ray diffraction machines were used to determine the physical structure of the fibers. A Rigaku D-Max/B Θ/2Θ X-ray diffractometer with Bragg−Brentano parafocusing geometry, a diffracted beam monochromator, and a copper target X-ray tube set to 40 kV and 30 mA were used to determine the % crystallinity and CI. These measurements were taken on fibers that were made into pellets of about 5 mm thick for a 2θ range from 5 to 45°. To make the pellets, fibers were powdered in Wiley Mill grinder using a 250 μm mesh, and the powdered fibers were pressed into a pellet using a hydraulic press operated at about 12000 PSI. CI was determined from the intensity differences in the peak positions at 18 and 22° (*20−22*).

A Bruker D8 Discover model diffractometer equipped with an area detector and GAADS software was used to obtain the diffraction patterns and to calculate the MFA. To obtain these measurements, a parallel bundle of fibers was mounted vertically in a specially designed sample holder with the axis of the fiber perpendicular to the X-ray beam. The 002 peak intensities in the diffraction patterns were fit into two Gaussian curves using a nonlinear least-square algorithm with the software program Microcal ORIGIN to obtain the MFA.

Maceration. Sorghum leaf and stem fibers were macerated to obtain the single cells in the fiber. Maceration was done using a 10% (w/w) nitric acid and 10% (w/w) chromic acid solution. Fibers were dipped in equal volumes of the two solutions for about 24 h after initiating the reaction by heating the solution at 60 °C for 5 min (*23*).

Morphological Structure. A Hitachi S3000N model variable pressure scanning electron microscope (SEM) was used to observe the morphological features of the untreated sorghum leaf and stem, and the fibers and single cells were obtained. The specimens to be observed were mounted on conductive adhesive tape, sputter coated with gold palladium, and observed in the SEM using a voltage of 15 kV. The lengths of the single cells were measured using a Motic image plus digital microscope. About 100 single cells were measured for their length, and the averages and standard deviations are reported. The widths of the single cells were measured from the SEM pictures.

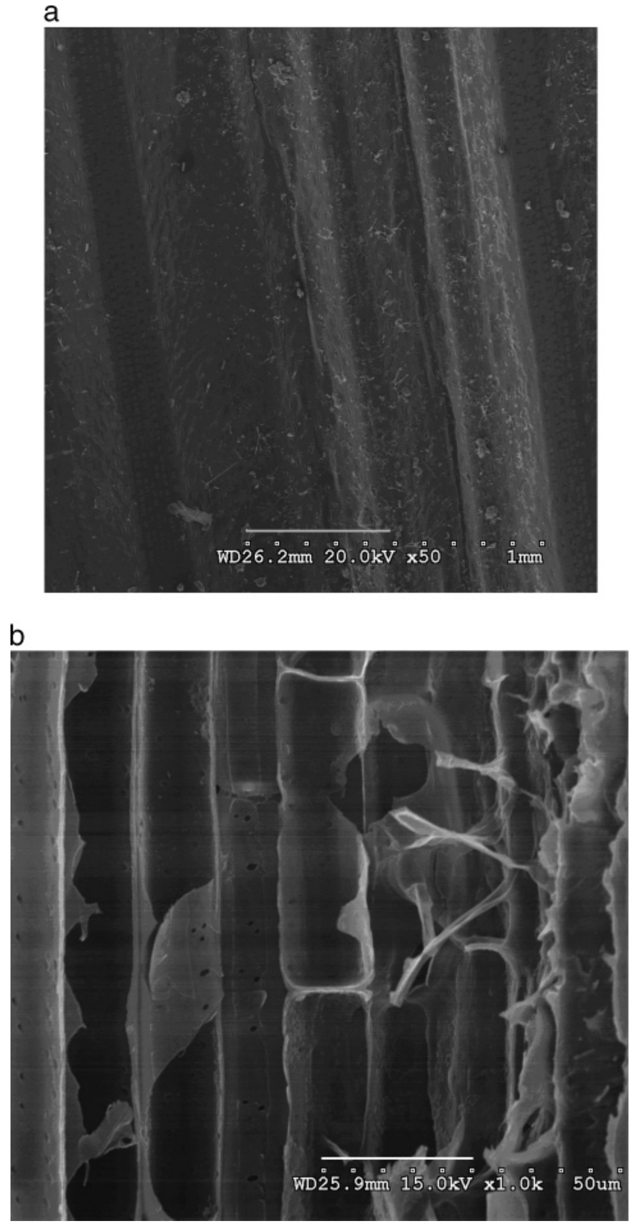

Figure 1. SEM picture of the surface of an untreated sorghum leaf (**a**) and stem (**b**) shows a rough outer surface with surface deposits that protect the cellulose inside. Bar = 0.5 mm.

Fiber Properties. The tensile properties of the fibers in terms of the strength, % elongation, and modulus were determined using an Instron (model 4000) tensile testing machine. A gauge length of 25 mm and a crosshead speed of 18 mm/min were used for the testing. About 100 fibers were tested for each of the leaf and stem fibers, and the averages and standard deviations are reported.

Moisture Regain. The moisture regain of the sorghum leaf and stem fibers was determined according to ASTM method 2654.

RESULTS AND DISCUSSION

Fiber Composition. The composition of the sorghum leaf and stem fibers is given in **Table 1**. As seen from the table, the leaf and stem fibers have very similar cellulose content, but the leaf fibers have higher lignin and ash contents than the fibers obtained from sorghum stems. The cellulose content in the sorghum fibers is lower than the cellulose in cornhusk and cornstalk fibers but similar to that in rice fibers (*6−9*). The lignin and ash contents in the sorghum fibers are similar to those in the fibers obtained from other agricultural byproducts (*5−9*,

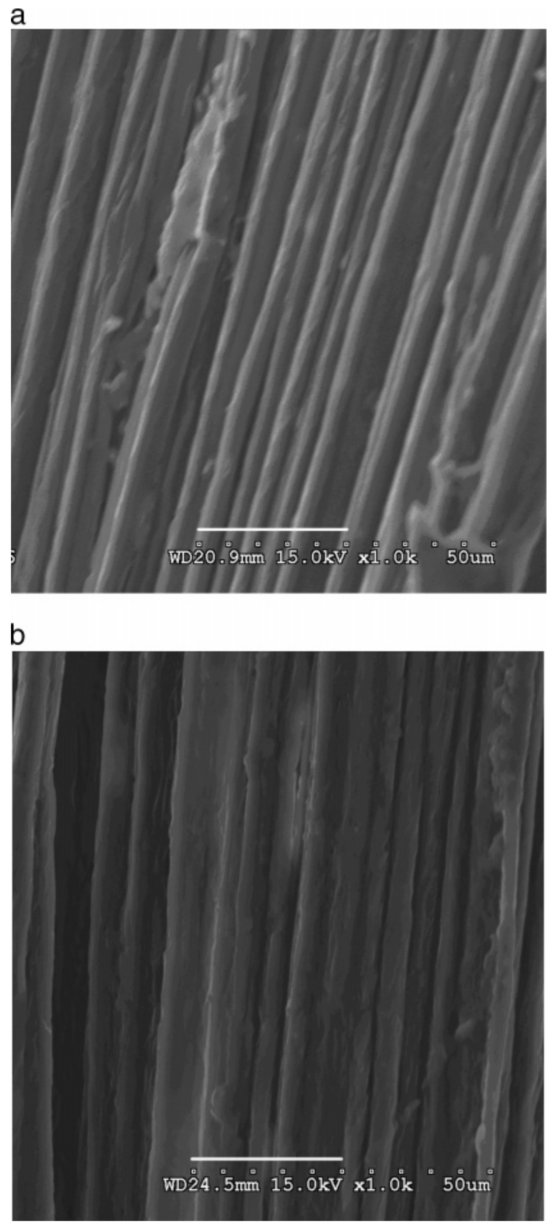

Figure 2. SEM picture of the extracted sorghum leaf (**a**) and stem fibers (**b**) showing a number of single cells held together to form a fiber bundle. The sorghum leaf fibers were obtained by treating with 2% sodium hydroxide at 95 °C for 30 min, and the sorghum stem fibers were obtained by treating with 4% sodium hydroxide at 95 °C for 45 min. Bar = 25 μm.

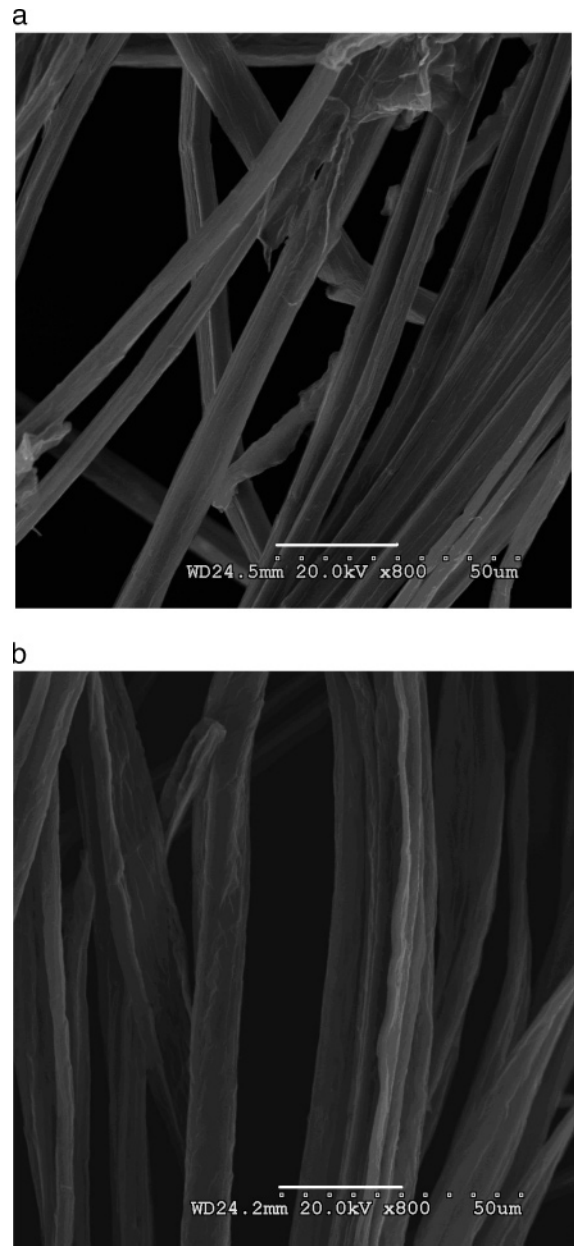

Figure 3. Single cells obtained by maceration from the sorghum leaf (**a**) and stem (**b**) are smooth and have a clean surface. Maceration was done using 10% chromic acid and 10% nitric acid solutions in equal proportions. Bar = 25 μm.

11). A few attempts have previously been made to determine the composition of the leaf and stem of various sorghum species. Sorghum stems of various species have been reported to have cellulose contents ranging from 41 to 65%, and sorghum leaves have been reported to have cellulose in the range of 59−73% (*24*−*27*). The cellulose content in the sorghum leaves and stems is higher than that reported for other agricultural byproducts such as cornstover, rice, and wheat straw, which have cellulose contents in the range of 30−40% (*5*−*9*). Although a report by Volenec et al. reports a lignin content of about 5% in various species of sorghum stems, other reports have determined the lignin content in sorghum stems in the range of 12−16% (*4*).

Fibers with higher cellulose contents have better properties and are also relatively easier to process. This is because noncellulosic contents such as lignin are responsible for the natural color, which may make it difficult to bleach the fibers, and lignin also makes the fibers stiff and harsh to handle (*26*). However, because the sorghum fibers are multicellular, lignin is necessary to hold the single cells together to form a fiber bundle required for textile and other high-quality applications. The composition of the sorghum fibers will therefore be determined by the inherent composition of the sorghum leaf and stems, the methods used to extract the fibers, and also the applications intended for the fibers. The method of fiber extraction also determines the yield of the fibers obtained. Stronger extraction conditions give lower yields, but the fibers will be relatively finer. The conditions used in this research produce about 25% fiber yield from both the sorghum stems and the leaves. With the 25% fiber yield and assuming 50% of the total sorghum stems and leaves could be made available for fiber production, about 7 million tons of sorghum fibers can be produced every year.

Morphological Structure. Figures 1−3 depict the morphological features of the sorghum leaf and stems and also the fibers

Table 2. Morphology of Single Cells and Physical Structure of Fibers from Both Sorghum Leaf and Stems as Compared with Cornhusk and Cornstalk Fibers[a]

	sorghum		cornstover			
parameter	leaf	stem	husks	stems	cotton	linen
			single cell dimensions			
length (mm)	1.7 ± 0.8	1.3 ± 0.7	0.5–1.5	0.8 ± 0.3	15–56	4–77
width (μm)	7.6 ± 2.4	15.8 ± 7.2	10–20	27 ± 8.9	12–25	5–76
			physical structure			
% crystallinity	32	39	48–50	52	65–70	65–70
MFA	15.5	16.5		10.9	20–30	8–12
CI	56	81		74	60	73–81

[a] Errors are ± one standard deviation. Data for cornhusk, corn stems, and cotton and linen are from refs *6*, *7*, and *19*, respectively.

and single cells obtained from the sorghum leaves and stems. The untreated leaves and stems shown in **Figure 1a**,**b**, respectively, have a layer of deposits on the surface. This surface layer is probably composed of waxes, hemicellulose, lignin, and other binding materials also seen in other lignocellulosic sources such as wheat straw (*28*, *29*). This surface layer protects the cellulose fibers inside. During fiber extraction, the surface layer is removed, resulting in fibers that have a relatively clean and smooth surface as shown in **Figure 2a**,**b**. **Figure 3a**,**b** shows several single cells in the leaf and stem fibers, respectively. As seen from the figures, the single cells are clean and have a smooth surface.

Single cells in the sorghum leaf fibers are longer and much smaller in width than the single cells in sorghum stems as seen from **Table 2**. The single cells in sorghum stems are similar to the single cells in cornhusk fibers, but the single cells in sorghum leaves are finer and longer than the single cells in both the cornhusk and the cornstalk fibers (*6*−*8*). Both the sorghum and the corn fibers have lower lengths and wider widths than cotton and linen except for the width of sorghum leaf fibers. No previous reports are available on the dimensions of single cells in sorghum stems or leaves. The leaf fibers having longer and smaller width single cells mean that sorghum leaves could produce finer fibers as compared to the fibers produced from sorghum stems.

Physical Structure. Sorghum leaf and stem fibers have relatively low % crystallinity as compared to the % crystallinity of the cornhusk, cornstalks, cotton, and linen fibers as seen from **Table 2**. In fact, the % crystallinity of the sorghum leaf and stem fibers is lower than most other lignocellulosic fibers (*6*−*9*). Low crystallinity means that the fibers will have relatively high amorphous regions. These amorphous regions facilitate moisture and other chemical absorptions for the sorghum fibers. One of the reasons for the low % crystallinity of sorghum fibers could be the inherently low crystallinity of cellulose in the sorghum plants, probably associated with the growth and nature of the sorghum plants. Sorghum is relatively easy to grow and supports grains that are lesser in weight than, for example, corn. Therefore, the plants may inherently have less crystallized cellulose. However, both the sorghum leaf and the stem fibers produce similar diffraction patterns as seen from **Figure 4**.

Although the sorghum fibers have low % crystallinity, the cellulose crystals in the both the leaf and the stem fibers are well-oriented as seen from the diffraction patterns in **Figure 5a**,**b**, respectively. The diffraction patterns have small and bright diffracting arcs, indicating good orientation of the cellulose crystals to the fiber axis. The sorghum fibers have an MFA lower than cotton but similar to linen. Fibers that have low MFAs will typically have lower elongations and vice versa. Because the sorghum fibers have an MFA closer to that of linen

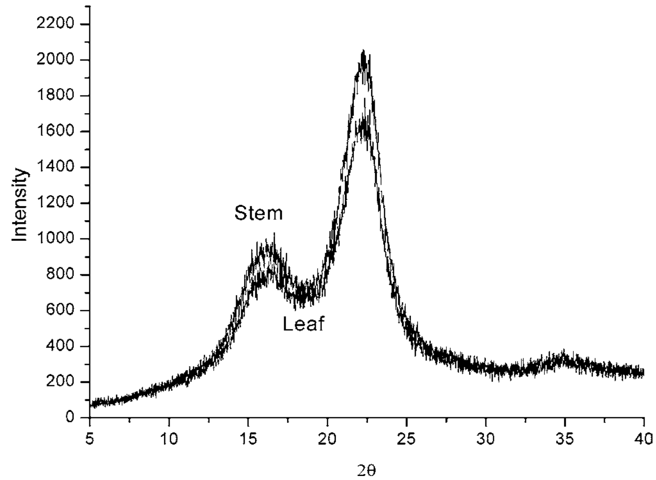

Figure 4. Diffraction pattern of sorghum stem and leaf fibers.

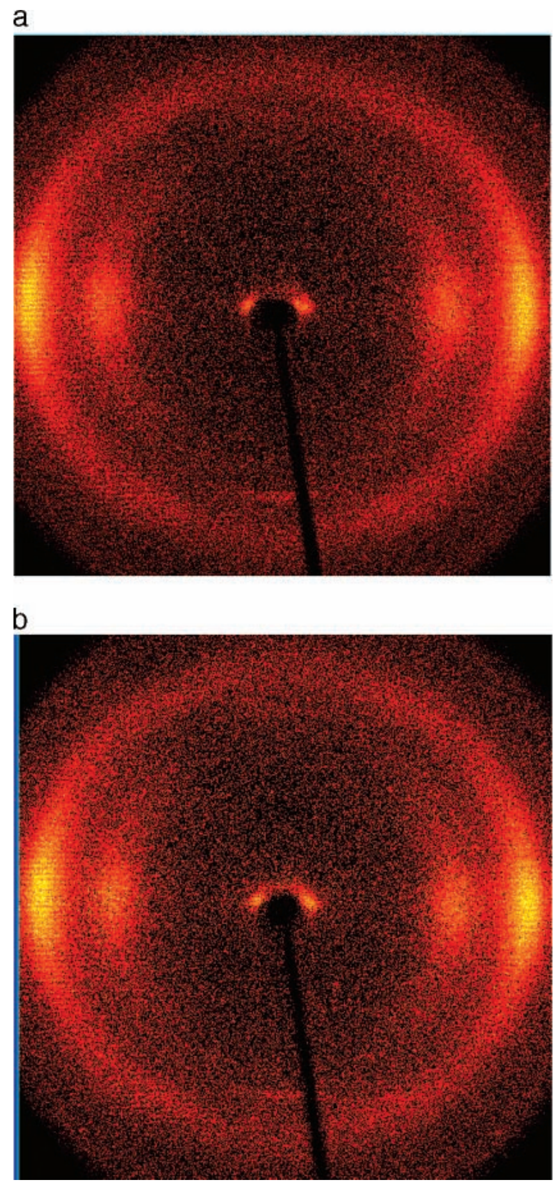

Figure 5. Diffraction pattern of sorghum leaf (**a**) and stem (**b**) fibers shows bright and sharp diffracting arcs.

Table 3. Dimensions, Tensile Properties, and Moisture Regain of Sorghum Leaf and Stem Fibers as Compared with Cornhusks and Cornstalk Fibers[a]

	sorghum		cornstover			
parameter	leaf	stem	husks	stems	cotton	linen
			fiber properties			
denier	65–105	59–95	18–120	35–120	3–8	1.7–17.8
length (cm)	4.5–9.5	4–11.5	2–20	1.5–8.5	1.5–5.6	20–140
strength (g/den)	2.4 ± 0.6	2.3 ± 0.5	2.7 ± 0.1	2.2 ± 0.9	2.7–3.5	4.6–6.1
elongation (%)	2.6 ± 0.7	2.6 ± 0.6	15.3 ± 2.1	2.2 ± 0.7	6.0–9.0	1.6–3.3
modulus (g/den)	114 ± 9.9	113 ± 10.3	70 ± 1.7	127 ± 56	55–90	203
moisture regain (%)	9.8	9.5	9.5	7.5–8.4	7.5	12.0

[a] Errors are ± one standard deviation. Data for cornhusk, corn stems, and cotton and linen are from refs *6*, *7*, and *19*, respectively.

than cotton, the sorghum fibers should have elongation similar to that of linen, as will be shown later (*19*, *30*).

Fiber Properties. The length, fineness, tensile properties, and moisture regain of sorghum stem and leaf fibers are compared with that of cornhusk, cornstalks, cotton, and linen fibers in **Table 3**. As seen from the table, sorghum fibers have lengths similar to that of cornstalks and cotton but lower than that of cornhusk fibers. However, commercially used natural cellulose fibers such as cotton, which is processed on the short staple system, typically have lengths of about 2.5−3.5 cm, and linen, which is processed as a long staple fiber, is used in lengths of about 8−12 cm (*19*, *30*). Therefore, both the sorghum leaf and the stem fibers can be processed on either the short or the long staple spinning system. The fineness of the fibers is similar to that of cornhusk and cornstalk fibers, but the fibers are coarser than cotton and linen.

The tensile properties of the sorghum leaf and stem fibers are very similar to each other, and the breaking tenacity of the fibers is similar to that of cotton, cornhusk, and cornstalk fibers but lower than the strength of linen as given in **Table 3**. The presence of high amounts of cellulose and the better orientation of the cellulose crystals in the fibers could be responsible for the good strength of the sorghum fibers despite the fibers having relatively low % crystallinity. Elongation of the sorghum fibers is similar to that of linen and cornstalks but lower than that of cornhusk and cotton fibers (*6*−*8*). Cornhusk fibers are unique in that they have high elongation, unlike the common bast fibers and the fibers obtained from other lignocellulosic agricultural byproducts. Sorghum fibers have good breaking tenacity and elongations, which are two basic properties of fibers. This along with the length and fineness indicate that sorghum fibers could be suitable for use as textile fibers.

Young's modulus of sorghum fibers is higher than that of cotton and cornhusk fibers, similar to that of cornstalk, but lower than linen fibers as seen from **Table 3** and **Figure 6**. A higher modulus indicates that the fibers will be stiffer than cornhusk fibers but similar or softer than cornstalk fibers when similar denier fibers are used. As seen from **Figure 6**, the sorghum fibers have similar stress−strain behavior to that of cornstalk fibers, but the cornhusk fibers are much softer and also have higher elongation whereas linen has high strength and modulus. The moisture regain of the sorghum and corn fibers is similar but higher than cotton and lower than linen as given in **Table 3**.

Fibers obtained from both sorghum stems and leaves have structures and properties similar to that of cotton and also to the natural cellulose fibers obtained from cornhusk and cornstalks. Although the sorghum fibers have relatively low % crystallinity, the fibers have good orientation of microfibrils and the cellulose crystals along the fiber axis. The sorghum stem and leaf fibers have strengths similar to cotton and corn fibers, but the elongation of sorghum fibers is lower than that of cotton. The moisture regain of the sorghum fibers is higher than cotton but similar to most other natural cellulose fibers; therefore, products made from sorghum fibers will be comfortable to wear. Overall, sorghum leaves and stems are cheap and abundantly

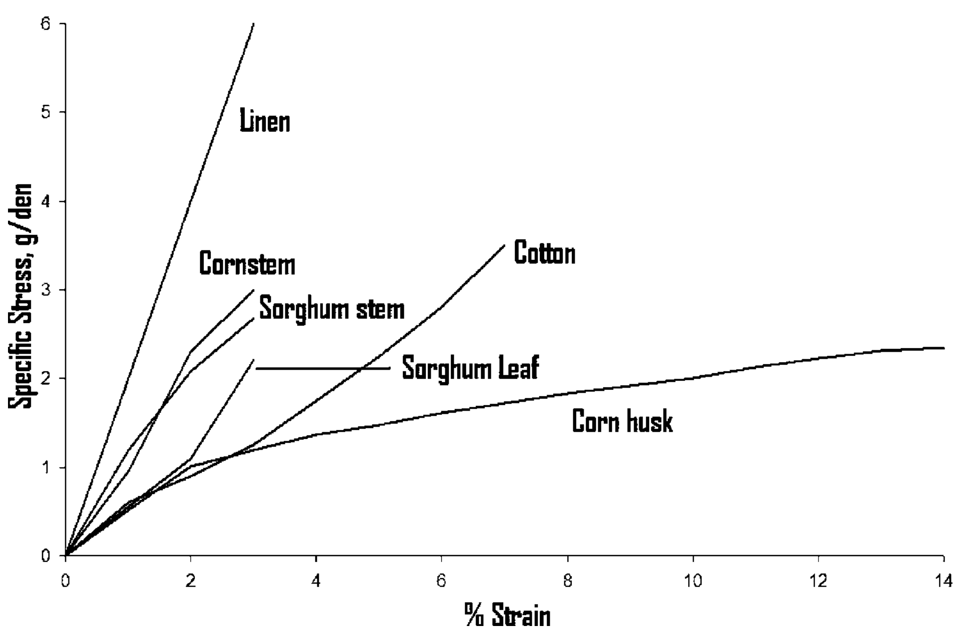

Figure 6. Stress–strain curves of fibers obtained from sorghum leaf and stem as compared with that of cornhusk, cornstalk, cotton, and linen fibers.

available byproducts suitable for fiber production. Using the sorghum stems and leaves as a source for fibers will add value to the crops, benefit the farmers economically, and also make the fiber industry more sustainable. However, methods to collect and preserve the stover for long periods of time and support to develop suitable fiber production and processing machinery are required for commercial production of fibers from sorghum and other lignocellulosic sources.

LITERATURE CITED

(1) http://faostat.fao.org/site/339/default.aspx.
(2) Clark, T. F.; Nelson, G. H.; Cunningham, R. L.; Kwalek, W. F.; Wolff, I. A. *Tappi J.* **1973**, *56* (3), 107−110.
(3) Billa, E.; Koullas, D. P.; Monties, B.; Koukios, E. G. *Ind. Crops Prod*. **1997**, *6*, 297−302.
(4) Wall, J. S.; Blessin, C. W. In *Sorghum Production and Utilization*; Wall, J., Ross, W. M., Eds.; The AVI Publishing Company, Inc.: Connecticut, 1970; pp 118−127.
(5) Reddy, N.; Yang, Y. *Trends Biotechnol*. **2005a**, *23* (1), 22−27.
(6) Reddy, N.; Yang, Y. *Green Chem*. **2005**, *7*, 190−195.
(7) Reddy, N.; Yang, Y. *Polymer* **2005b**, *46*, 5494−5500.
(8) Reddy, N.; Yang, Y. *AATCC Rev.* **2005**, *5* (7), 24−27.
(9) Reddy, N.; Yang, Y. *J. Agric. Food Chem.* **2006**, *54*, 8077−8081.
(10) Reddy, N.; Yang, Y.; McAlister, D. D., III. *Indian J. Fibre Text. Res.* **2006**, *31* (4), 537−542.
(11) Reddy, N.; Yang, Y. *Biotechnol. Bioeng.* In press, DOI 10.1002/bit.21330.
(12) Collier, B. J.; Collier, J. R.; Agarwal, P.; Lo, Y. *Text. Res. J.* **1992**, *62* (12), 741−748.
(13) Reddy, N.; Yang, Y. *Biomacromolecules* **2007**, *8* (2), 638−643.
(14) Doraiswamy, I.; Chellamani, P. *Text. Prog.* **1993**, *24* (1), 1−25.
(15) Karst, D.; Yang, Y.; Genzo, T. *Polymer* **2006**, *47*, 6464−6471.
(16) Karst, D.; Yang, Y. *Polymer* **2006**, *47*, 4845−4850.
(17) Yang, Y.; Huda, S. *J. Appl. Polym. Sci.* **2003**, *90*, 3285−3290.
(18) Yang, Y.; Huda, S. *AATCC Rev.* **2003**, *3* (8), 56−61.
(19) Batra, S. K. In *Handbook of Fiber Science and Technology*; Lewin, M., Pearce, E. M., Ed.; Marcel Dekker, Inc.: New York, 1998; Vol. 4, Fibre Chemistry, pp 727−803.
(20) Hindeleh, A. M. *Text. Res. J.* **1980**, *11*, 667.
(21) Hu, X.; Hsieh, Y. *J. Appl. Polym. Sci.* **1996**, *34*, 1451−1459.
(22) Mwaikambo, L. Y.; Ansell, M. P. *J. Appl. Polym. Sci.* **2002**, *84*, 2222−2234.
(23) Ruzin, S. E. In *Plant Microtechnique and Microscopy*; Oxford University Press: New York, 1999; pp 127−136.
(24) Reed, J. D.; Tedla, A.; Kebede, Y. *J. Sci. Food Agric.* **1987**, *39*, 113−121.
(25) Volenec, J. J.; Cherney, J. H.; Moore, K. J. *Crop Sci.* **1986**, *36*, 307−310.
(26) Cherney, D. J. R.; Patterson, J. A.; Cherney, J. H.; Axtell, J. D. *J. Sci. Food Agric.* **1991**, *54*, 645−649.
(27) Ali, M.; Islam, M. N.; Mian, J. A.; Chowdhury, S. A. W. *J. Text. Inst*. **2001**, *90* (1), 34−39.
(28) Liu, R.; Yu, H.; Huang, Y. *Cellulose* **2005**, *12*, 25−34.
(29) Yan, L.; Zhu, Q. *J. Appl. Polym. Sci.* **2003**, *88*, 2055−2059.
(30) Mauersberger, H. R. *Matthews' Textile Fibers*; John Wiley & Sons: New York, 1954; p 282.

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