Morphology and Mechanical Properties of Unidirectional Sisal–Epoxy Composites

KRISTIINA OKSMAN,[1] LENNART WALLSTRÖM,[2] LARS. A. BERGLUND,[2] ROMILDO DIAS TOLEDO FILHO[3]

[1] SICOMP AB (Swedish Institute of Composites), Box 271, SE-941 26, Piteå, Sweden

[2] Luleå University of Technology, Division of Polymer Engineering, Sweden

[3] Federal University of Rio de Janeiro, Brazil

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ABSTRACT: Plant fibers are of increasing interest for use in composite materials. They are renewable resources and waste management is easier than with glass fibers. In the present study, longitudinal stiffness and strength as well as morphology of unidirectional sisal–epoxy composites manufactured by resin transfer molding (RTM) were studied. Horseshoe-shaped sisal fiber bundles (technical fibers) were nonuniformly distributed in the matrix. In contrast to many wood composites, lumen was not filled by polymer matrix. Technical sisal fibers showed higher effective modulus when included in the composite material than in the technical fiber test (40 GPa as compared with 24 GPa). In contrast, the effective technical fiber strength in the composites was estimated to be around 400 MPa in comparison with a measured technical fiber tensile strength of 550 MPa. Reasons for these phenomena are discussed. © 2002 Wiley Periodicals, Inc. J Appl Polym Sci 84: 2358–2365, 2002

Key words: natural fiber; sisal; composites; resin transfer molding (RTM); mechanical properties; morphology

INTRODUCTION

Northern Brazil is the largest producer of sisal in the world, and more than a million people are depending on its crops. The sisal plants can be cultivated in an extremely dry climate where other plants cannot survive. Typically, small farmers are working with the sisal crop. The harvesting of the plants and production of sisal fibers is done in an old-fashioned and risky way. More than two thousand people have lost their hands or arms during sisal processing, mainly during the decorticating step. The sisal fiber extraction from the plant is done by cutting and decorticating the leaves (mechanical scraping), and then drying, brushing, and cleaning the fibers. The mechanical process yields about 2–4% fiber, one person can produce approximately 15 kg fibers in 8 h.[11] After processing, the fibers are transported to different industries for further processing to ropes, textiles, carpets, rugs, and sacks. Sisal is also used as paper fiber. Rope used to be a major application for sisal but plastic ropes are nowadays replacing sisal. New applications for sisal fibers are therefore of interest. Sisal fiber reinforcement in composite materials is one such possibility.

The interest in using different plant and wood fibers as reinforcement in plastics has increased dramatically during the last few years. Environmental concern is one driving force. We are par-

Correspondence to: K. Oksman, Norwegian University of Science and Technology, Department of Machine Design and Materials Technology, Richard Birkelandsvei 2B, 7491 Trondheim, Norway (kristiina.oksman@immtek.ntnu.no).

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Table I Fiber Type, Fiber Content, and Densities of Studied Materials

Sample	Fiber Type	Fiber Extraction Method	Weight fraction (w/o)	Volume Fraction (v/o)	Density (g/cm^3)
Epoxy	—	—	—	—	1.15
S28	Sisal	Decortication	30	28	1.16
S35	Sisal	Decortication	37	35	1.17
S46	Sisal	Decortication	48	46	1.17
UD32	Flax	Dew retting	37	32	1.23
G48	Glass	—	66	48	1.71

ticularly interested in replacing glass fibers with cellulose fibers in composite materials for structural applications. Natural fibers have some advantages compared to man-made fibers. They have low density, are biodegradable, and incineration is straightforward. They are renewable raw materials, and have relatively high strength and stiffness and cause no skin irritations.[1–10] On the other hand, there are also some disadvantages such as moisture sensitivity, quality variations, and low thermal stability. Many investigations have been performed on the potential of natural fibers. In several cases, the results have shown that although the composite modulus is high, the strength is much lower than for glass fiber composites.[7–10]

Manufacturing methods for natural fiber thermoset composites have been modified lay-up/press molding, pultrusion, resin transfer molding (RTM), and vacuum infusion molding.[1,3–14,19] The review article of sisal fiber and its composites written by Yan Li et. al. showed that manufacturing methods such as RTM and resin film injection (similar to vacuum infusion) have not been used to economically make composites with good mechanical properties.[1] Other areas needing more research are composite microstructure, fiber–matrix adhesion, toughness and failure mechanisms, new processing methods, and recycling.[1]

The objective of the present study is to characterize composite morphology and study longitudinal stiffness and strength in unidirectional sisal–epoxy composites manufactured by RTM.

EXPERIMENTS

Materials

The sisal fibers were supplied from northern Brazil; the extracted and cut fibers were washed in warm water in order to remove water soluble extractives and waxes. After washing the fibers were dried, heckled and sewed by hand to unidirectional fiber mats. The fiber mats were dried at 103°C during 4 h before RTM processing. Reference materials were unidirectional glass fiber mat, L 500-E11-1, from Devold AMT AS with 90 w/o fibers in the main direction and retted flax fibers. An epoxy resin LY 5082 with an amine type hardener HY 5083 from Ciba Geigy was used as matrix. This epoxy is especially designed for RTM. The sisal–epoxy composites were molded with 3 different fiber contents and the mechanical properties were compared to those of glass fiber and flax fiber composites. Table I shows the compositions of the laminates, weight fraction of the fibers, and the estimated volume fractions. The densities of the composites were measured using an Accupyc 1330 according to ASTM standard D-2856. The volume fraction of the fiber V_f was obtained knowing the mass of constituents M and density of the resin ρ_r from the relationship

$$V_f = (V_c - (M_c - M_f)/\rho_r)/V_c \tag{1}$$

where V_c is composite volume fraction, M_c is the mass of composite, and M_f is the mass of the fibers.[4] The value obtained for volume fraction sisal fiber includes the empty lumen.

Manufacturing Process

Composites were manufactured using the RTM processing technique. RTM is a closed mold process where laminates are formed between two stiff mold halves. The dry fiber mats were placed into the mold cavity, the mold was closed, and then the degassed epoxy resin was injected. The laminate size was 300 × 200 × 4 mm. The pressure during injection was varied depending on the fiber content, a pressure of 2 bar was used for higher fiber content and 1 bar for lower fiber content. The tool temperature was held at 50°C during the injection and afterward the laminates were cured in the mould having a temperature of 80°C during 8 h. The flow behavior of the resin in the sisal weaves was good compared to the flax mats (higher permeability), this perhaps because of the larger diameter of the sisal technical fibers and the longer fiber length.

Mechanical Testing

The mechanical properties of the sisal fiber bundles and composites were measured using an Instron universal testing machine (model 4411) with a crosshead speed of 2 mm/min. The measured sisal fiber bundles were dried at 103°C during 24 h. Four 80 mm long bundle samples were tested and the strain was estimated from crosshead movement. Composites were tested according to ASTM 3029 standard. Composite samples for tensile testing were cut to the width of 25 mm and a length of 250 mm. The laminate thickness was 4 mm. At least five samples of each composition were tested and the results are presented as an average for the tested samples.

Microscopy

The morphology of the fibers and the composites was studied using scanning electron microscopy (SEM) and optical microscope (OM). Fractured cross sections were sputter-coated with gold and studied in a SEM at an acceleration voltage of 30 kV. The fibre and void contents were studied on polished cross sections using an OM and image analysis software.

RESULTS AND DISCUSSION

Fiber Morphology

The sisal fibers (*Agave sisalana*) are extracted from the sisal plant leaves in the form of long fiber bundles called technical fibers.[15] The sisal leaf is a sandwich structure. Each leaf contains approximately 700–1400 technical fibers with a length of 0.5–1 m. Every technical fiber contains numerous individual fibers (here called "fibers") about 3 mm in length (1–8 mm) and 10 to 30 μm in diameter.[1,15–17] The total sisal leaf mass contains 2–4% technical fibers.

The morphology of the sisal fibers is apparent in Figure 1: (a) shows the longitudinal section of the sisal technical fiber bundle at two different magnifications and (b) the cross section. In (a) the remaining parts of parenchyma cells surrounding the technical fibers are observable. The individual fiber diameter varies along the fiber length, being smallest toward the ends. In the higher magnification micrograph (b) we therefore see large variations in fiber diameter. The single fiber cross section has an irregular shape, often polygonal with five sides.

Fiber Properties

For comparative purposes, mechanical properties, density, and geometry of sisal (current data and literature data), flax and glass fibers are presented in Table II. Flax demonstrates higher modulus and strength than sisal due to smaller lumen and smaller microfibril angle. The present sisal data are rough estimates from only four tests giving a Young's modulus of 24 GPa and a tensile strength of 550 MPa. Literature data for natural fibers show large variations in both Young's modulus and tensile strengths.[1,9,10,14,16,17] This depends on quality variations between fibers (microstructural variations, damage state) but also on the fact that fibers are tested in different ways. Mukherejee and Satyanarayana[16] showed that different fiber test lengths and strain rates result in different tensile properties for sisal fibers. Increased test length decreases the strength of the fibers because the number of defects (i.e., fiber ends) is increased.[16] It has also been shown that the strength of technical sisal fibers is not uniform along the length of the fiber bundle. The fibers taken from the root and lower part of the plant have lower tensile strength and Young's modulus and a higher strain to failure.[14] Also, in order to simplify experimental procedures, technical fibers are usually tested rather than individual fibers. The present data are also technical fiber data. Single fibers will show significantly higher strength than technical fibers. The reason is that the probability of finding significant defects is much larger in a technical fiber than in a single fiber. In addition, the load distribution is much more inhomogeneous in the technical fiber.

Morphology of the Composites

Figure 2 shows the fractured cross sections of the sisal–epoxy composites. There was no epoxy resin located in the fiber lumen. The total lumen area in the technical fiber cross-section was found to be 15% and the total void content (including lumen) about 17%. The void content of the composite

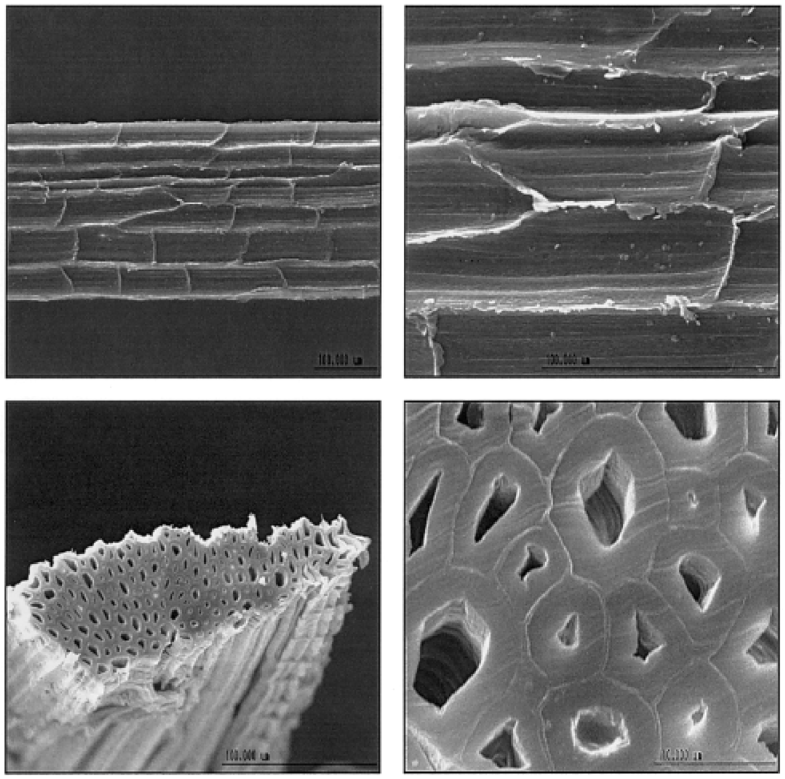

Figure 1 Technical fiber of sisal (fiber bundle): (a) longitudinal section and (b) cross section.

cross section was measured using image analysis of OM micrographs.

Oksman and Lindberg (1995) studied thermoplastic wood fiber composites and showed that lumen was filled with the polymer matrix.[18] An important function of wood fibers in the tree is water transport from the root to the top. Wood fibers therefore have distribution channels between the single wood cells. It is possible that the absence of such channels in sisal, due to the different function of sisal fibers, is the reason for lack of polymer in the lumen. The total leaf mass contains only 2–4% technical sisal fibers, other cells are taking care of the water distribution.

In Figures 2 and 3, the protruding fibers demonstrate the mechanism of fiber pull-out during the fracture process. In Figure 3, technical fiber imprints in the matrix are also apparent. The imprint in Figure 3(b) even shows the pattern from residual parenchyma cell walls. In several locations, for instance in Figure 2(b), a gap between technical fiber and matrix can be observed. The clear imprints and the gaps indicate weak fiber-matrix adhesion.

Figure 4 presents optical micrographs of polished composite cross sections. The horse-shoe shape of technical sisal fibers is apparent. We may also note the distribution in technical fiber

Table II Physical Properties of Sisal and Flax Fibers Compared to Glass Fiber[1,9,10,13–17]

Fiber Type	E-modulus (GPa)	Strength (MPa)	Strain to Failure (%)	Fiber Length (mm)	Single Fiber Length (mm)	Single Fiber Diameter (μm)	Cell Wall Density (g/cm^3)
Glass	72	2000–3400	1.8–3.2	—	Continuous	10	2.56
Flax	45–100	600–1100	1.5–2.4	300–900	13–70	10–30	1.4–1.5
Sisal (lit.)	7–20	400–700	2–14	500–1000	1–8	10–40	1–1.45
Sisal (meas.)	24 ± 6	550 ± 100	2.4 ± 0.4	—	—	—	1.37

cross-sectional area and shape. During processing, many technical fibers are pulled apart parallel to the fiber length direction resulting in a different cross-sectional shape. The technical fibers are distributed nonuniformly in the matrix with resin- and fiber-rich areas. It is helpful to consider cross-sectional micrographs when material properties are considered. For instance, each technical fiber contains 15% of empty lumen. Therefore, we should view the technical fiber as a separate phase consisting of a cellular solid.

Mechanical Properties of Sisal Composites

The longitudinal tensile property data of the present sisal composites are presented in Table III. As expected, the sisal fibers significantly increase the tensile strength and Young's modulus of the epoxy resin. The composite with 46% by volume of sisal fibers has a Young's modulus of about 20 GPa. The tensile strength of the same material is 210 MPa. These results are difficult to compare with earlier results in the literature on sisal–epoxy since the literature data are in compression and bending. Bisanda and Ansell[11] reported a flexural modulus for unidirectional sisal–epoxy composites of 16 GPa at 40% fibers by volume. The flexural strength was 266 MPa. Since flexural strength is usually significantly higher than tensile strength, our data are in good agreement with this previous study. For nonwoven sisal mats, Singh et al.[12] reported that sisal–polyester composites (fiber content 50% by volume) had a tensile strength of 30 MPa and a tensile modulus of 1.15 GPa. The composites were manufactured by impregnation of the nonwoven sisal mats and then compression moulded during 2 h.

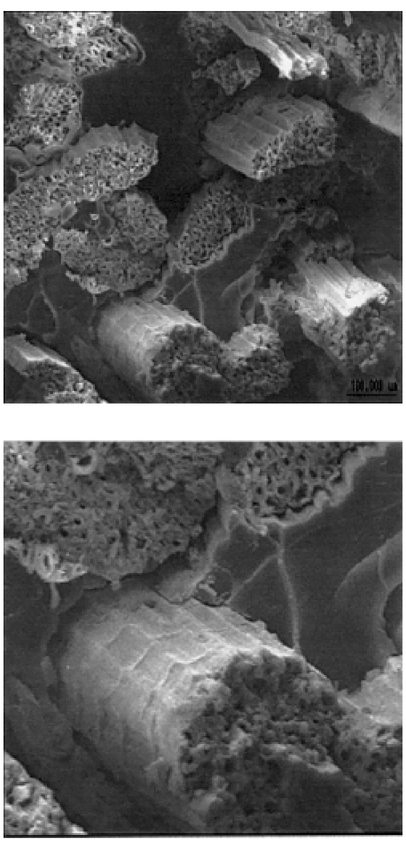

Figure 2 Cross section of fracture surface of the sisal–epoxy composite.

a)

b)

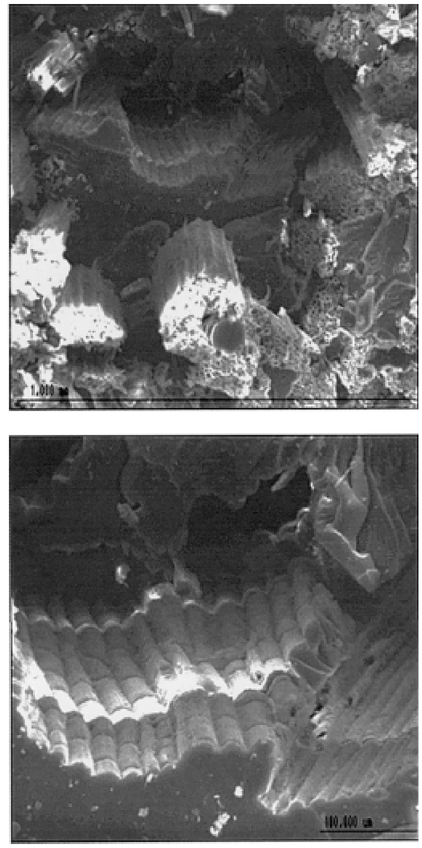

Figure 3 Cross section of fracture surface of the sisal–epoxy composite, showing the (a) fiber pull-outs and (b) print of the fiber surface in the epoxy.

Figure 5 shows measured values and theoretical estimates of Young's modulus vs fiber volume fraction. The solid line is based on eq. (2) and a technical fiber modulus E_f of 40 GPa. The dotted line assumes $E_f = 20$ GPa.[16,17] The predictions are based on the rule of mixture:

$$E_C = E_f V_f + (1 - V_f) E_m \tag{2}$$

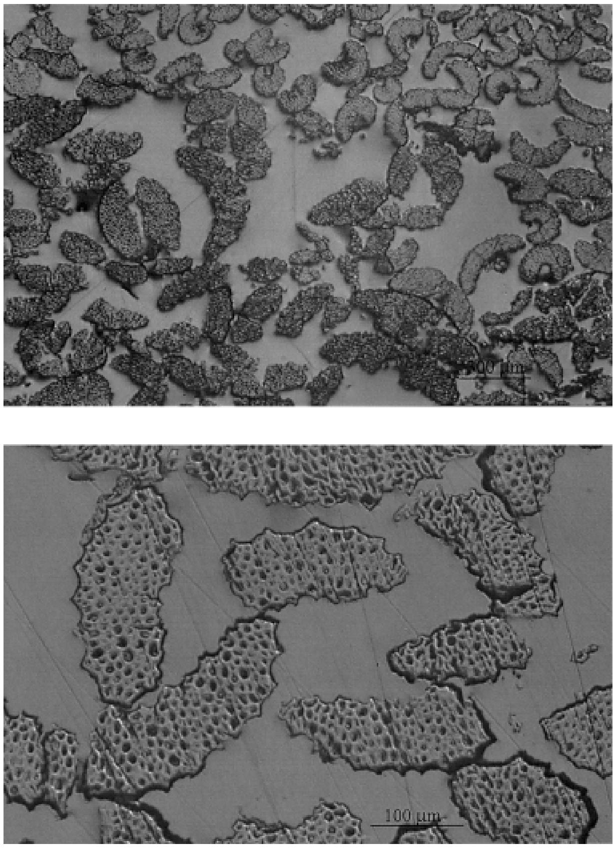

Figure 4 OM pictures on the cross sections of the sisal–epoxy composite. Note the nonuniform fiber distribution and horseshoe-shaped technical fibers.

where E_C, E_f, and E_m are the moduli of the composite, fiber and matrix, respectively, and V_f is fiber volume fraction. Figure 5 demonstrates that 40 GPa is a better estimate of the effective fiber modulus than the 20 GPa reported in refs.16 and 17. Note also that our measured technical fiber modulus is 24 GPa. Fiber modulus data, from tests of bundles of technical fibers, are lower than the real fiber modulus since the individual fibers are not loaded uniformly (some single fibers are not loaded at all). In addition, the cross-sectional area of the technical fiber is nonuniform. However, in the composite the load-bearing situation of the technical fiber is very different as it is embedded in polymer matrix. That the effective technical fiber modulus in the composite is 40 GPa is very interesting. This points to the limitations of current technical fiber testing procedures and also demonstrates a more favorable stress distribution in the technical fibers in the composite material. The effective fiber strength in the composite can be estimated using the following relationship:

$$\sigma_f^* = (\sigma_c^* - \sigma_m(1 - v_f))/v_f \qquad (3)$$

where the σ_f^* is effective fiber strength, σ_m stress in the epoxy at fracture, σ_c^* strength of the composite and v_f fiber volume fraction. This somewhat simplistic relationship is based on the assumption of constant strain in matrix and fiber. We assume that all technical fibers fracture at the same critical stress and this corresponds to final fracture. For $v_f = 0.28$, 0.35, and 0.46, this results in effective technical fiber strengths of 420, 400, and 390 MPa. Considering the real nature of the fracture process (progressive fracture of individual fibers and technical fibers followed by crack growth and final fracture), the consistent nature of our data for different v_f is encouraging. An effective technical fiber strength around 400 MPa is also reasonable compared with our measured average technical fiber strength of 550 MPa. In reality, inhomogeneous stress distribution and a wide distribution in individual fiber and technical fiber strength will cause many technical fibers to fail at stresses much lower than 400 MPa. Al-

Table III Absolute and Specific Properties of the Sisal Composites and Pure Epoxy Resin

Sample	Tensile Strength (MPa)	Specific Strength (MPa/g cm^{-2})	Tensile Modulus (GPa)	Specific Modulus (GPa/g cm^{-2})	Elongation at Break (%)
Epoxy	76[a]	66	3.1–3.2[a]	2.7	7.3[a]
S28	169 (±23)	146	14.2 (±1.6)	12.2	2.3
S35	183 (±16)	157	14.5 (±1.6)	12.4	2.2
S46	211 (±12)	180	19.7 (±1.5)	16.8	1.9

Standard deviation in parentheses.
[a]Manufacturer supplied data

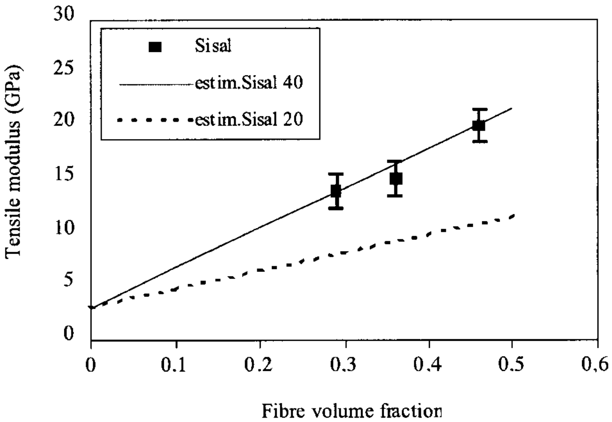

Figure 5 Modulus vs sisal fiber content, both measured values and theoretical estimations.

though we have no observations of failure mechanisms, comparison between 400 MPa and the 550 MPa average technical fiber strength indicates that failure of the composite material occurs at a low concentration of failed technical fibers. We may further speculate that poor interfacial adhesion leads to fiber–matrix debonding close to fracture sites and corresponding load redistribution from fractured technical fibers to neighboring fibers. The inhomogeneous fiber distribution is no advantage in this context. As fibers experience local load concentrations, the failure process continues, leading to ultimate failure.

The results from mechanical testing demonstrate that the technical fiber shows higher modulus when embedded in a matrix material as compared with technical fiber test. On the other hand, the effective technical fiber strength in the composite is lower than the measured technical fiber strength. This indicates that the low strength fraction of the technical fiber population controls composite strength. More uniform fiber distribution and improved fiber–matrix adhesion may improve the strength of sisal–epoxy composites.

Comparison with Flax and Glass Fiber Composites

Table IV shows the absolute and specific mechanical properties of the compared composites. Generally, the tensile strength of natural fiber composites does not reach the level of the glass fiber composites.[7–10] Note that sisal–epoxy composites have better strength than retted flax–epoxy composites. Sisal is also a cheaper fiber than flax. It is interesting to note that the specific modulus of sisal composites is similar to glass fiber composites. The specific modulus was 17 GPa/g cm^{-2} for sisal compared to glass fiber composites 18 GPa/g cm^{-2}. The specific tensile strength of sisal–epoxy composites was 186 MPa/g cm^{-2} compared to 470 MPa/g cm^{-2} for the glass fiber composites while the absolute values were 219 and 817 MPa, respectively.

CONCLUSIONS

The morphology study of sisal–epoxy composites showed horseshoe-shaped technical fibers non-uniformly distributed in the matrix. In contrast with many wood composites, lumen was not filled by polymer matrix. About 15% of the technical fiber cross-section consisted of lumen. Micrographs of fractured composites indicated weak fiber–matrix adhesion.

Technical sisal fibers showed higher effective modulus when included in the composite material as compared with technical fiber test (40 GPa as compared with 24 GPa). In technical fiber tests, the individual fibers are not loaded uniformly

Table IV Absolute and Specific Properties of the Composite and Pure Epoxy Resin

Sample	Tensile Strength (MPa)	Specific Strength (MPa/g cm^{-2})	Tensile Modulus (GPa)	Specific Modulus (GPa/g cm^{-2})	Elongation at Break (%)
Epoxy	76[a]	66	3.1–3.2[a]	2.7	7.3[a]
S35	183 (±16)	157	15 (±1.6)	12	2.2
S46	211 (±12)	180	20 (±1.5)	17	1.9
UD32	132 (±4.5)[b]	107[b]	15 (±0.6)[b]	12[b]	1.2[b]
G48	817 (±35)[b]	478[b]	31 (±1)[b]	18[b]	2.8[b]

Standard deviation in parentheses.
[a]Manufacturer supplied data.
[b]Oksman.[13]

(some single fibers are not loaded at all). In addition, the cross-sectional area of the technical fiber is nonuniform. This points to limitations with technical fiber test procedures and also demonstrates a more favorable stress distribution in the technical fibers embedded in composite materials.

The effective technical fiber strength in the composites was estimated to be around 400 MPa in comparison with a measured technical fiber tensile strength of 550 MPa. We expect the real fracture mechanism to depend on the inhomogeneous fiber stress distribution and the wide distribution in individual fiber and technical fiber strength. Therefore, many technical fibers fail at stresses much lower than 400 MPa and the low strength fraction of the technical fiber population is of importance. Poor interfacial adhesion and inhomogeneous fiber distribution is no advantage in this context. Compared with previous tensile strength data for dew-retted flax composites, sisal composites offer higher strength at lower fiber cost.

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