Tensile Properties of Short Sisal Fiber-Reinforced Polyethylene Composites

KURUVILLA JOSEPH,[1] SABU THOMAS,[1] C. PAVITHRAN,[2,*] and M. BRAHMAKUMAR[2]

[1]School of Chemical Sciences, Mahatma Gandhi University, Kottayam 686 631, India;
and [2]Regional Research Laboratory (CSIR), Trivandrum 695 019, India

SYNOPSIS

Sisal fibers (Agave-Veracruz) have been used as reinforcements in low-density polyethylene (LDPE). The influence of the processing method and the effect of fiber content, fiber length, and orientation on tensile properties of the composites have been evaluated. The fiber damage that normally occurs during blending of fiber and polyethylene by the melt-mixing method is avoided by adopting a solution-mixing procedure. The tensile properties of the composites thus prepared show a gradual increase with fiber content. The properties also increased with fiber length, to a maximum at a fiber length of about 6 mm. Unidirectional alignment of the short fibers achieved by an extrusion process enhanced the tensile strength and modulus of the composites along the axis of fiber alignment by more than twofold compared to randomly oriented fiber composites. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

Several cellulosic products and wastes such as shell flour, wood flour, and pulp have been used as fillers in thermoplastics primarily to achieve cost savings.[1] Over the past decade, cellulosic fillers of fibrous nature have been of greater interest as they would give composites with improved mechanical properties compared to those containing nonfibrous fillers.[2-5]

Lignocellulosic fibers like jute, sisal, coir, and pineapple have been used as reinforcements in thermoset matrices.[6-9] Among these fibers, sisal is of particular interest in that its composites have high-impact strength besides having moderate tensile and flexural properties compared to other lignocellulosic fibers.[10,11] A preliminary investigation has shown that sisal fiber can also be used as a reinforcement in a thermoplastic matrix and that it performs better than do wood fibers.[12]

The mechanical properties of a short-fiber-reinforced polymer composite depend on many factors like fiber–matrix adhesion, volume fraction of fiber, fiber aspect ratio (l/d), and orientation of the fiber. Fiber aspect ratio should be above a critical value for maximum effect of reinforcement by attaining maximum stress in the fiber before failure of the composite. Fiber orientation has a significant influence on the mechanical properties of the composite in that the stress value is maximum along the axis of the orientation of the fiber. In addition to the above, factors like processing conditions/techniques also have significant influence on the mechanical properties of fiber-reinforced composites, particularly thermoplastic composites wherein fiber breakage during processing is a commonly observed phenomena.[13] Therefore, a detailed investigation has been carried out on sisal–polyethylene composites, especially on the effect of fiber length, fiber orientation, and processing conditions on the tensile properties of the composites. The results are reported here.

EXPERIMENTAL

Materials

Polyethylene (LDPE-Indothene 16MA400) was supplied by Indian Petrochemicals Corp. Sisal fiber

---

[*] To whom correspondence should be addressed.

Journal of Applied Polymer Science, Vol. 47, 1731–1739 (1993)

CCC 0021-8995/93/101731-09

Table I Properties of Sisal Fiber

Fiber Diameter (μm)	Lignin Content (%)	Cellulose Content (%)	Tensile Strength (MPa)	Tensile Modulus (GPa)	Elongation at Break (%)
100–300	4–5	85–88	400–700	9–20	5–14

Table II Variation of Tensile Properties of MMC and SMC with Fiber Content (Average Fiber Length 5.8 mm)

MMC				SMC			
Fiber Weight (%)	Tensile Strength (MPa)	Modulus (MPa)	Percentage Elongation at Break (mm)	Fiber Weight (%)	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)
0	9.2	140	200	0	9.2	140	200.0
10	9.65	276	42	10	10.8	324	27.0
20	11.25	408	22	20	12.5	453	10.2
30	10.2	346	8	30	14.7	781	7.1

(Agave-Veracruz) was obtained from local sources. The fiber was washed with water and dried in an air oven at 80°C for 4–6 h before being chopped into the desired length ranging from 2 to 10 mm for preparation of composites.

Preparation of LDPE–Sisal Blends

The LDPE–sisal blends for molding composites were prepared by two different methods, viz., (1) melt mixing and (2) solution mixing and extrusion. In the melt-mixing method, the fiber was added to a melt of polyethylene and mixing was performed using a Brabender plasticorder model PLE 651. A mixing temperature of 120°C and a rotor speed of 30 rpm were maintained during mixing for 10 min. The mix was taken out from the mixer while hot and then subjected to extrusion using a hand-operated ram-type injection-molding machine. The extrudate was removed as about 10 mm-long and 4 mm-thick rods. In the solution-mixing method, fiber was added to a viscous solution of polyethylene in toluene that was prepared by adding toluene to a melt of the polymer. The mixing was carried out manually in a stainless-steel beaker using a stainless-steel stirrer. The temperature was maintained at 110°C during mixing for about 10 min. The mix was then transferred into a flat tray as lumps and kept in a vacuum oven at 70°C for 2 h to remove the solvent. The lumps were further subjected to extrusion as for melt-mixed blends. Blends containing 10, 20, and 30% by weight of fiber were prepared using fibers of length in the range 2–10 mm.

Preparation of Composite Sheets

a. *Randomly Oriented Fiber Composites*

Sisal–polyethylene blends prepared by both melt mixing and solution mixing were used for making

Table III Variation of Tensile Properties of SMC with Repeated Extrusion of the Blends (Average Fiber Length 5.8 mm, Fiber Content 20 wt %)

No. of Extrusions	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)	Density (gm/cm^3)
1	12.5	453	10.2	0.892
3	11.24	388	13.4	0.905
5	10.8	316	20.2	0.918
6	10.4	285	23.0	0.926

Table IV Variation of Tensile Properties of SMC with Fiber Length (Fiber Content 20 wt %)

Fiber Length (mm)	Tensile Strength (MPa)	Modulus (MPa)	Elongation at Break (%)
0	9.20	140	200.0
2.1	11.38	283	12.6
5.8	12.50	453	10.2
9.2	10.24	273	10.6

Table V Variation of Tensile Properties of SMC Unidirectionally Aligned Fiber Composites with Fiber Length (Fiber Content 30 wt %; Values Given in Parentheses Are the Properties in the Transverse Direction)

Average Fiber Length (mm)	Tensile Strength (MPa)	Tensile Modulus (MPa)	Elongation at Break (%)
0	9.2	140	200.0
2.1	20.5 (7.8)	1687 (261)	4.2 (3.4)
5.8	31.12 (6.1)	3086 (590)	1.8 (< 1)
9.2	25.9 (6.7)	1716 (334)	4.3 (3.4)

Table VI Variation of Tensile Properties of SMC Unidirectionally Aligned Fiber Composites with Fiber Content (Fiber Length 5.8 mm; Values Given in Parentheses Are the Properties in the Transverse Direction)

Fiber Weight (%)	Tensile Strength (MPa)	Tensile Modulus (MPa)	Elongation at Break (%)
0	9.20	140	200
12	15.61 (5.33)	1429 (295)	4 (2)
20	21.66 (5.91)	2088 (388)	3 (≈ 1)
30	31.12 (6.1)	3086 (590)	1.8 (< 1)

the randomly oriented fiber composites. Composite specimens of dimensions 120 × 12.5 × 3 mm were prepared by injection molding of the blends at 115 ± 5°C using a hand-operated ram-type injection-molding machine.

b. Oriented Fiber Composite

The specimens of oriented fiber composites were prepared from blends obtained by the solution-mixing method. A combination of injection- and compression-molding techniques was used for making the composite sheets. The blend was first extruded into 4 mm-thick cylindrical rods using the injection-molding machine. Rectangular specimens of a size measuring 120 × 12.5 × 3 mm were prepared by closely aligning the cylindrical extrudates (120 mm long and 4 mm thick) in a leaky mold[10,14] and then compression molding, employing a pressure of about 4 MPa and a temperature of 115 ± 5°C. The mold was cooled below 50°C before removing the composite specimens from the mold.

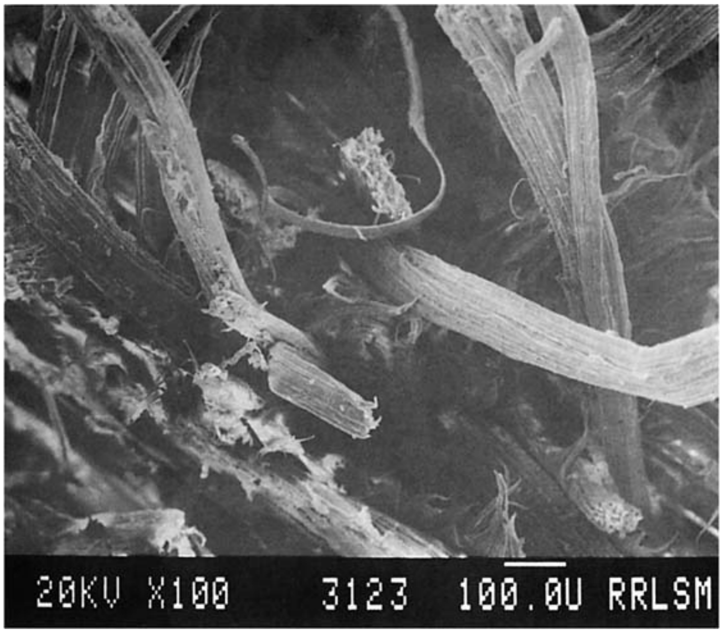

Figure 1 Fracture surfaces of sisal–polyethylene composite (SMC).

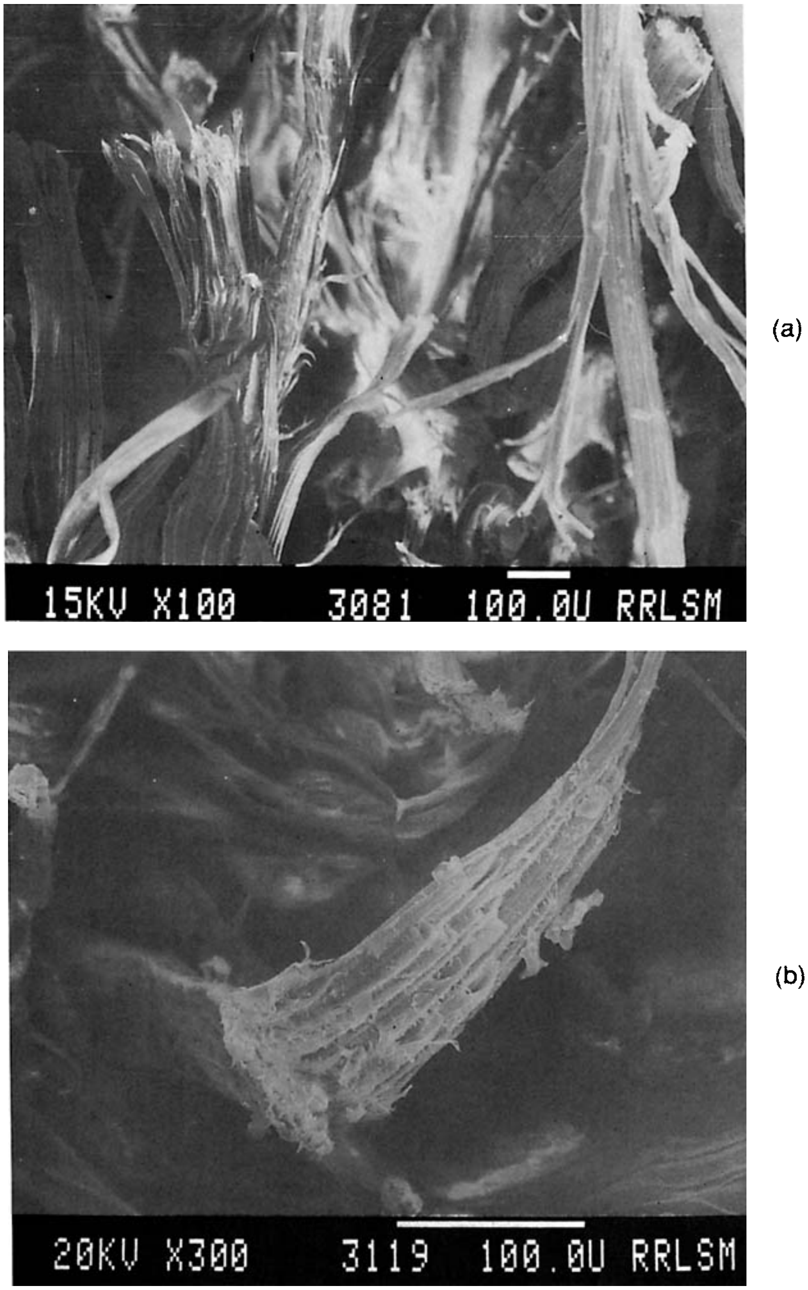

Figure 2 Fracture surfaces of MMC showing fiber damage by (a) splitting and (b) peeling.

Testing of Composites

Tensile testing of rectangular specimens of a size measuring 120 × 12.5 × 3 mm was carried out using an Instron testing machine at a crosshead speed of 200 mm/min^{-1} and a gauge length of 50 mm. The tensile modulus and elongation at break of the composite were calculated from the load-displacement curve. At least five specimens were tested for each set of samples and the mean values are reported.

Surfaces of the fractured specimens were examined using a JEOL scanning electron microscope. An optical stereomicroscope was used for observing the fiber orientation and also to study the fiber damage during preparation of the blends.

RESULTS AND DISCUSSION

The properties of the fiber used in the present study are presented in Table I. The diameter of the fiber

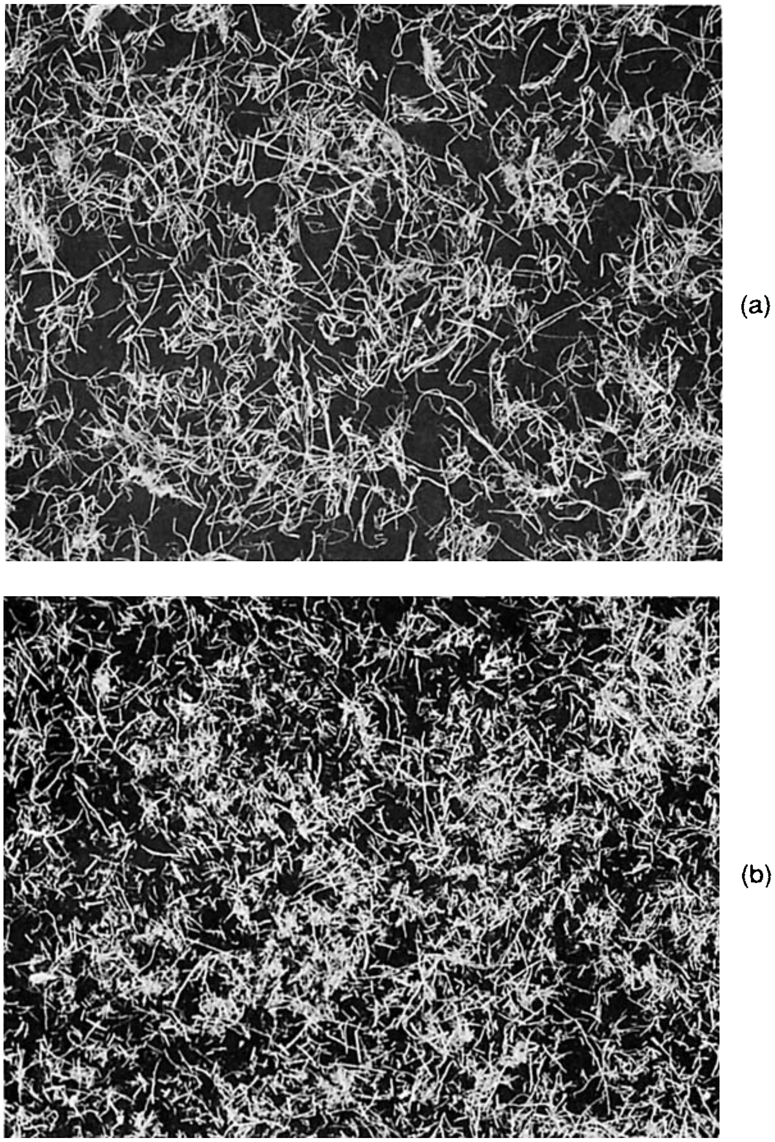

Figure 3 Optical micrograph of fibers extracted from (a) SMC and (b) MMC showing the extent of fiber breakage in MMC.

varied from 100 to 300 μm. The average diameter of the fiber calculated from the fraction of fibers of different diameter in a sample was about 190 μm.

Experimental results of processing conditions, fiber content, fiber length, and orientation on tensile properties of sisal–polyethylene composites are summarized in Tables II–VI. The composites made from the blends prepared by melt mixing and solution mixing are designated as MMC and SMC, respectively. Fiber content and length were limited to 30 wt % and 10 mm, respectively, since injection of the blend was found difficult at higher fiber loading and length.

Effect of Processing

Tensile properties of injection-molded MMC and SMC (randomly oriented fiber composites) are given in Table II. It can be seen that tensile properties of the composites are significantly affected by the processing method. In the case of SMC, the tensile strength and modulus steadily increase with fiber content, whereas tensile properties of MMC show a decrease in their values at 30 wt % fiber loading after showing an initial increase in the properties with fiber content. Further, the enhancement in the properties of MMC due to reinforcement is 30% less than that of SMC.

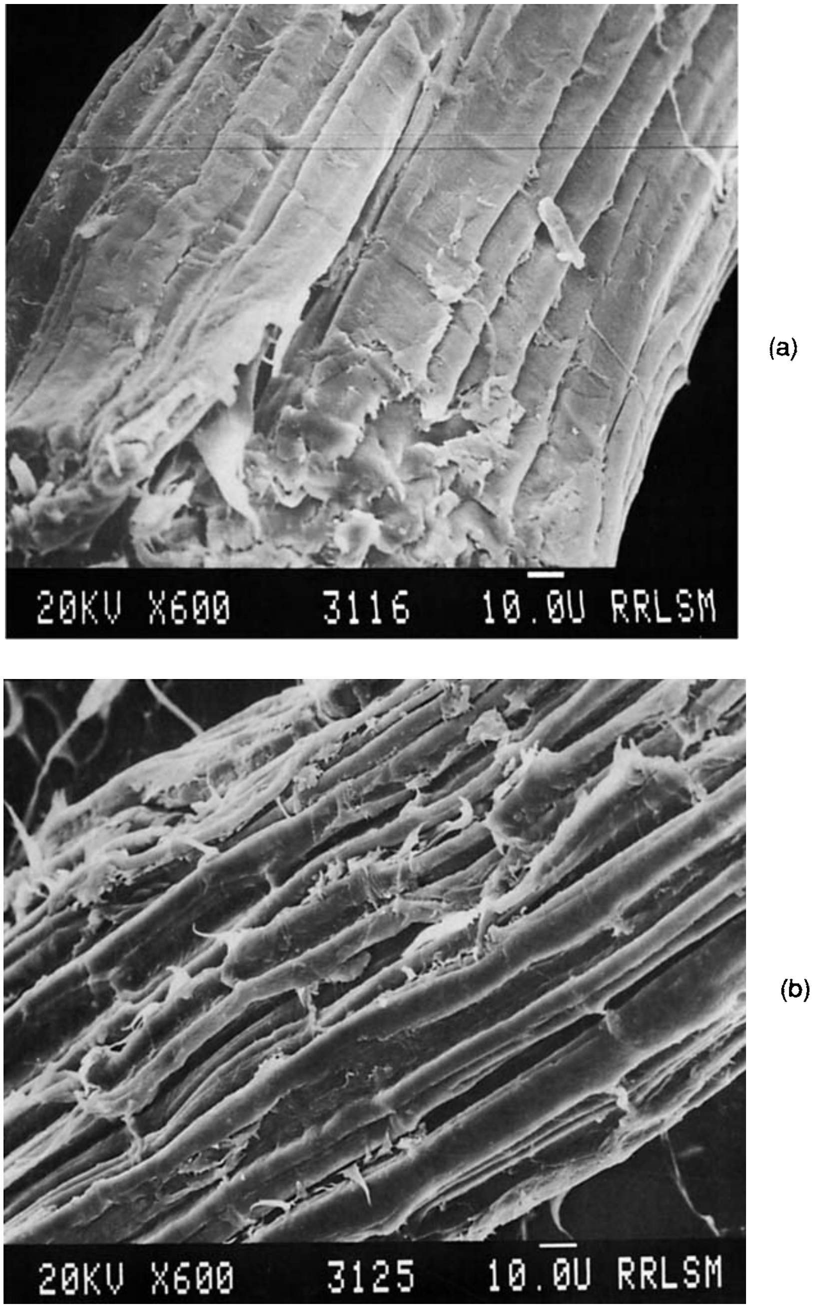

Figure 4 Surfaces of fibers extracted from the polyethylene blends after (a) first and (b) sixth extrusion showing degradation due to repeated extrusion of fiber.

Observations of the fractured tips of the composites indicated that the structure of the fiber is intact in SMC (Fig. 1), whereas the fibers in MMC undergo considerable damage, splitting and peeling as shown in Figure 2(a) and (b). The extent of fiber damage in MMC is clearer from the optical micrographs [Fig. 3(a) and (b)] of the fibers extracted from the composites by dissolving the matrix in toluene. Whereas the length of the fibers extracted from SMC is more or less uniform, that of the fibers extracted from MMC is distributed over a wide range. The degradation of fiber in MMC becomes more severe at high fiber content, which could be the reason for the decrease observed in its properties on increasing the fiber content from 20 to 30 wt %. Thus, better performance of SMC compared to

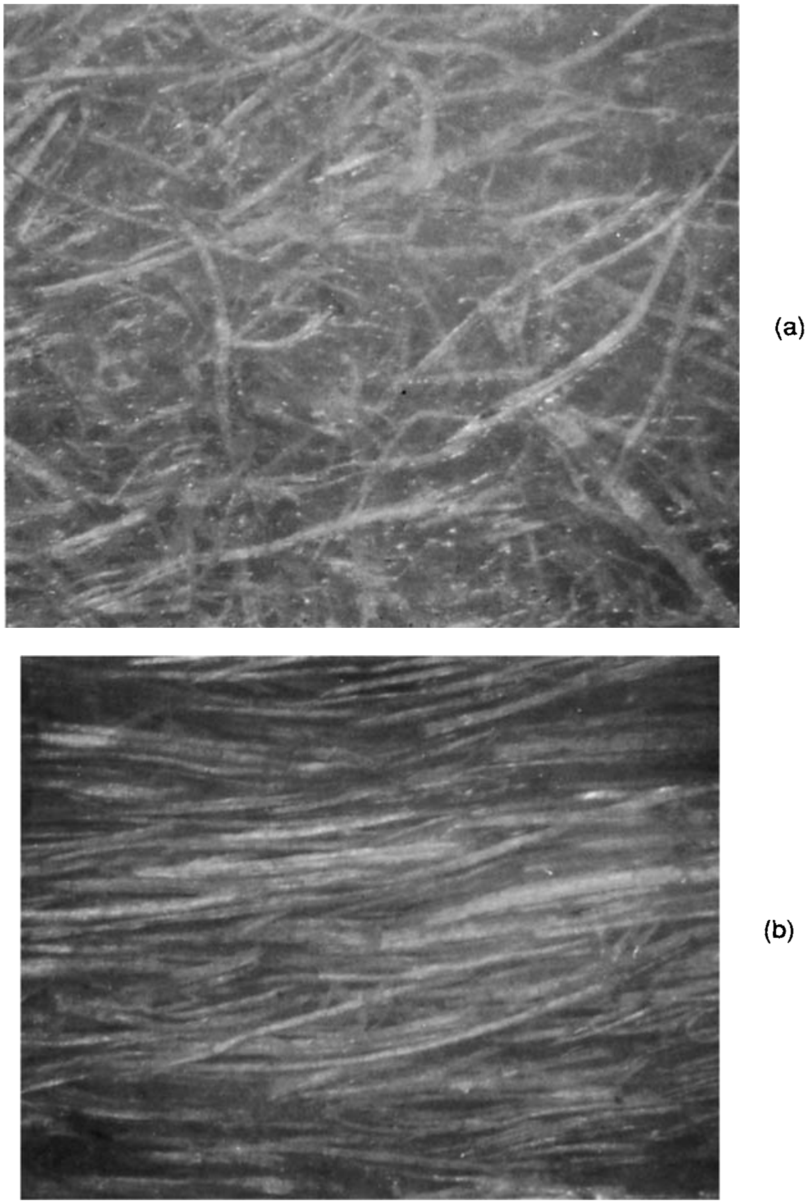

Figure 5 Optical micrographs of the surfaces of (a) randomly (b) unidirectionally oriented fiber composites.

MMC can be attributed to the preservation of fiber length in the former. However, MMC has shown a higher failure strain than its SMC counterpart, although both the composites exhibited reduction in failure strain with fiber content as generally observed with short-fiber-filled thermoplastics.

Density measurements of the composites have shown that higher failure strain associated with MMC is due to better compaction than that of SMC, possibly at the fiber–matrix interfacial region. Enhancement in both density and failure strain of SMC, as shown in Table III, can be achieved by repeated extrusion of the blends. But, contrary to expectations, the process resulted in a reduction of the strength and modulus of the composites. Observation of the fibers extracted from the blends after repeated extrusion [Fig. 4(a) and (b)] has shown that the process leads to interfibrillar debonding, possibly due to thermal effects, which should result in reduction of strength and modulus of the fiber and the composites. Therefore, further enhancement in tensile properties of SMC from the values shown in Table II may be possible if thermal degradation of fiber is prevented or minimized, the processing conditions for which are to be optimized.

Effect of Fiber Length

The effect of fiber length on tensile properties of sisal-polyethylene composites can be readily assessed from the data given in Table IV. The strength and modulus of the composites show an enhancement in their values by increasing the average fiber length from 2.1 to 5.8 mm followed by a decrease in the properties when a fiber length of 9.2 mm is employed. Observation of the composite specimens has shown that long fibers tend to bend or curl during molding. This causes reduction in the effective length of the fiber below the optimum length in a particular direction, which results in a decrease of the properties. The results indicate that there exists an optimum fiber length between 6 and 9 mm at which a maximum improvement in the properties of the composites can be achieved.

Effect of Fiber Orientation

In the injection-molded short-fiber-filled plastics, the fibers are oriented in a complex manner.[13] However, in cylindrical extrudates of SMC, where complex flow patterns evident in molded samples are absent, the fibers are aligned along the axis of the extrudate. As can be seen from Figure 5(b), orientation of fibers is not significantly affected, when the cylindrical extrudates are aligned and compression-molded into flat sheets in a leaky mold mentioned earlier.

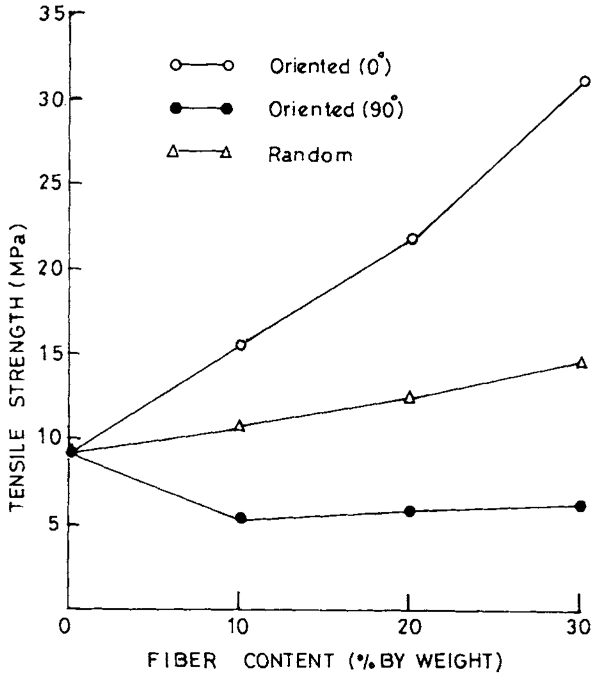

Figure 6 Effect of fiber orientation on tensile strength of sisal–LDPE composites (average fiber length 5.8 mm).

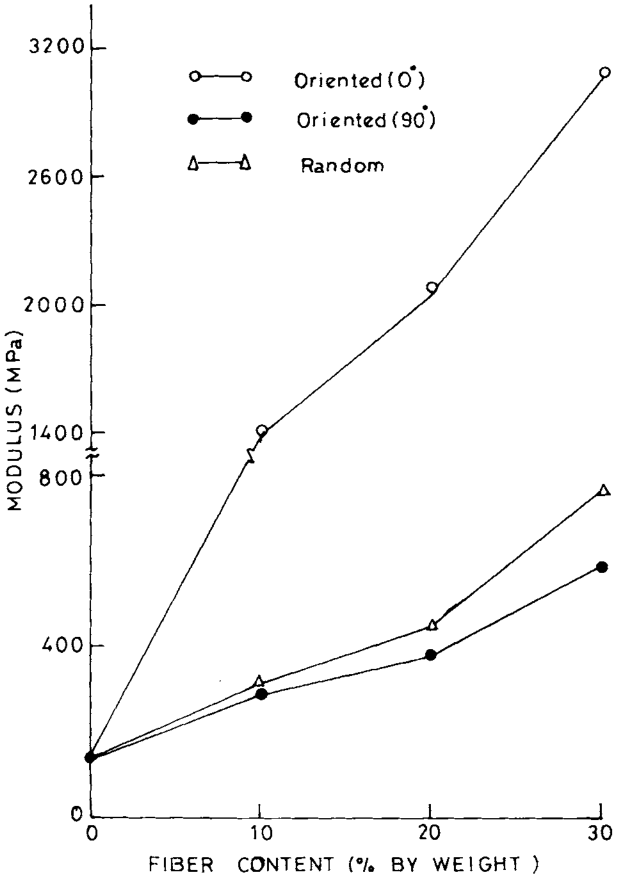

Figure 7 Effect of fiber orientation on tensile modulus of sisal–LDPE composites (average fiber length 5.8 mm).

The variation in tensile properties of oriented fiber composites with fiber length and fiber content given in Tables V and VI, respectively, follows a similar pattern to that observed with randomly oriented fiber composites. The increase in the strength values with fiber length is at a maximum at a length of about 6 mm due to the tendency of the fiber to bend or misalign when longer fibers were used.

The effect of fiber orientation can be readily observed from a plot of tensile strength and modulus of unidirectionally and randomly oriented fiber composites against fiber content (Figs. 6 and 7). The strength of the unidirectionally aligned fiber composite normal to the fiber alignment is less than that of the randomly oriented fiber composite, as expected. However, the tensile strength along the axis of the fiber alignment is 30% more than that of the randomly oriented composite at 10 wt % fiber content and the enhancement is twofold at a fiber content of 30 wt %. Similarly, the modulus of the composites is enhanced fourfold as a result of fiber alignment.

CONCLUSIONS

The tensile properties of sisal fiber–polyethylene (LDPE) composites are sensitive to fiber length, fiber content, and fiber orientation. The composites prepared from blends processed by a solution-mixing process, wherein fiber breakage is avoided, show a uniform increase in their tensile strength and modulus with fiber content. The composites also exhibited maximum properties at a fiber length of about 6 mm. Unidirectional alignment of the fiber enhanced the strength and modulus of the composites along the axis of fiber alignment by more than twofold compared to randomly oriented fiber composites.

REFERENCES

1. G. R. Lightsey, in *Polymer Application of Renewable Resource Materials*, C. E. Carraher, Jr. and L. H. Sperling, Eds., Plenum Press, New York, 1983, p. 193.
2. H. Dalvag, C. Klason, and H. E. Stromvall, *Int. J. Polym. Mater.*, **11,** 9 (1985).
3. D. Maldas, B. V. Kokta, and C. Daneault, *J. Appl. Polym. Sci.*, **37,** 751 (1989).
4. S. N. Maiti and M. R. Hassan, *J. Appl. Polym. Sci.*, **37,** 2019 (1989).
5. R. G. Raj, B. V. Kokta, and C. Daneault, *J. Appl. Polym. Sci.*, **40,** 645 (1990).
6. E. C. McLaughlin, *J. Mater. Sci.*, **15,** 886 (1980).
7. S. V. Prasad, C. Pavithran, and P. K. Rohatgi, *J. Mater. Sci.*, **12,** 1443 (1983).
8. R. A. Clark and M. P. Ansell, *J. Mater. Sci.*, **21,** 269 (1986).
9. C. Pavithran, P. S. Mukherjee, and M. Brahmakumar, *J. Reinf. Plast. Comp.*, **4,** 186 (1991).
10. C. Pavithran, P. S. Mukherjee, M. Brahmakumar, and A. D. Damodaran, *J. Mater. Sci. Lett.*, **7,** 825 (1988).
11. C. Pavithran, P. S. Mukherjee, M. Brahmakumar, and A. D. Damodaran, *J. Mater. Sci.*, **26,** 455 (1991).
12. K. Joseph, S. Thomas, and C. Pavithran, *J. Mater. Sci. Lett.*, to appear.
13. M. J. Folkes, *Short Fiber Reinforced Thermoplastics*, Research Studies Press, Wiley, New York, 1982.
14. N. H. Ladizesky and I. M. Ward, *Comp. Sci. Technol.*, **26,** 129 (1986).

Received January 30, 1992
Accepted March 12, 1992