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Natural Fibers in Resin Transfer Molded Composites

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Abstract

Jute fiber nonwoven mats were used to reinforce resin transfer molded unsaturated polyester-styrene panels. Resin flow through the mat was similar to flow through a random glass mat. Tensile strength, and tensile and flexural modulus (ASTM 638, 790) for unmodified jute samples were half that of samples made with a commercial glass mat. Izod impact (ASTM D 256) and flexural strength were an order of magnitude lower than glass-reinforced specimens. Samples exposed in a Weatherometer for 1,200 hours showed minimal surface erosion or color change. Specimens showed negligible weight loss when exposed to wood degrading fungi (ASTM D 1413). Jute fiber pull-out from the matrix was seen in scanning electron micrographs, which indicates that improving adhesion at the fiber-polymer interface may increase mechanical properties.

Introduction

Renewable fibers are often considered only for markets that require low costs and high production rates and can accept low performance. These fibers have many properties that would be an advantage in other markets, such as light weight, high specific strength compared to glass and carbon, and low energy requirements for processing (Table 1). Natural fibers can be substituted for glass and carbon fiber in polymer composites. Their potential for use in molded articles not needing high strength for acceptable performance has been tried in equipment housings, roofing for low-cost housing, and in large diameter piping (9,10,13). These authors were primarily exploring production feasibility.

As in synthetic fiber composites, the mechanical properties of the final product depend on the individual properties of the matrix, fiber, and the nature of the interface between the two. Typically, glass fiber is treated with a silane or other coupling agent to improve the properties at the interface with the resin. Where the fiber is a renewable fiber, opportunities exist to tailor the end properties of the composite by selecting fibers with a given chemical or morphological composition and by modification of the fibers and fiber surfaces, either chemically or physically. Several studies of fiber composition and morphology found that cellulose content and microfibril angle tend to control the mechanical properties of cellulosic fibers

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TABLE 1.—Comparison of properties of different fibers.

Fiber	Specific gravity	Specific tensile strength	Cost	Energy to produce
		(GPa)	(US$/ton)	(GJ/ton)
Lignocellulosic	0.6 to 1.2	1.6 to 2.95	200 to 1,000	4
Glass	2.6	1.35	1,200 to 1,800	30
Carbon	1.8	1.71	12,500	130

TABLE 2.—Mechanical properties of natural fiber-thermoset resin composites.

Fiber and resin	Volume fraction	Tensile strength	Reference
	(%)	(MPa)	
Jute-polyester	30	125	(1)
Sun hemp-polyester	30	100	(6)
Sun hemp-polyester	30	125	(7)
Jute-polyester	22 (by weight)	84	(11)
Jute-epoxy	33 (by weight)	104	(11)

(2,3,8). Other authors have studied the mechanical properties of lignocellulosic/polyester composites (Table 2) with well-defined fiber orientation (unidirectional or bidirectional) (1,6,7,11,12). The samples were produced either by wet layup or by press molding of MgO thickened prepregs (similar to sheet molding compound). No studies were found that had been done using resin transfer molding to form specimens for testing.

In this study, resin transfer molded composites were made in commercial resin transfer molding equipment by direct substitution of jute bast fiber for glass random fiber mats. The goal was to determine their suitability for use in resin transfer molding equipment with regard to resin flow, air entrainment, and surface properties, Mechanical properties were determined as well as resistance of the composites to wood-degrading fungi and simulated weathering.

Materials and methods

Sample preparation

Panels used for testing were produced by a commercial manufacturer of resin transfer molded fiberglass products. They were reinforced with glass mat, jute felt, or modified jute felt using a commercial resin system including fillers, colorants, and cure promoters. The mats were precut to fit flat in the mold. Specimens for testing were cut from the flat section of the panel in such a way to ensure uniform fiber loading. The resin base was an orthophthalic-type unsaturated polyester-styrene (hereafter called polyester) resin solution (styrene <45%; Owens Corning, Inc.). It was chosen for its flexibility under dynamic loading conditions and low viscosity (150 cm/sec. at ambient temperature) in resin transfer molding. Owens-Corning reports typical mechanical properties for the resin (catalyzed w/ 1.0% BPO) of 120.7 MPa for flexural strength and 68.95 MPa for tensile strength (Table 3). $CaCO^3$ filler (particle diameter 3.0 mm) was used at a rate of 5 percent by weight. The free-radical catalyst was a commercial blend of methyl ethyl ketone peroxide (145%) in dimethyl phthalate (Elf-Atochem), at the rate of 1.5 percent of resin weight. Several promoters were also used, namely cobalt naphthenate, cobalt neodecanoate, and an aromatic amine. A commercial black pigment paste was added equaling < 5 percent of a proprietary composition.

TABLE 3.—Neat resin properties.

Flexural strength	17,500 psi (120.7 MPa)	ASTM D 790
Flexural modulus	645,000 psi (4.45 GPa)	ASTM D 790
Tensile strength	10,000 psi (68.95 MPa)	ASTM D 638
Tensile modulus	570,000 psi (3.93 GPa)	ASTM D 638
Tensile elongation	2.0%	ASTM D 638

Fiber volume fraction was 10 to 15 percent (vol./vol.). The mold temperature was 140°F (55°C) at the beginning of resin injection. Injection pressure of the resin was arbitrary, based on the operators judgment, but probably did not exceed 35 psi (240

kPa). In-mold pressure was not determined, but due to the design of the mold it was probably a time-dependent pressure distribution from 35 psi (240 kPa) at the inlet to atmospheric pressure at the edge vents throughout the resin injection. It is assumed that the pressure distribution would tend to equalize after resin stopped flowing, at least until gelation occurred. It should be recognized that mold filling is a complex phenomenon and quantifying flow parameters was not the subject of this study

Jute mats were 35 g/m^2 random fiber needle punched mat. A small amount of light machine oil was applied to the fibers for ease in processing. Some of the mats were treated with an aqueous glycol solution and air-dried before molding. The glass fiber mat was a commercial grade produced by Owens-Corning for use in general purpose fiberglass laminations. This random continuous mat has a thermoplastic binder to help fiber preforms hold their shape and to improve adhesion to the matrix. Three plies of mat were used in each panel used for property testing. In addition, four- and five-ply jute panels were made to determine what effect higher fiber loadings might have on resin flow, air entrapment, and void formation.

Mechanical testing

Tensile tests on specimens measuring 25 by 250 mm were conducted according to ASTM 638-90 and flexural tests on specimens measuring 11 by 140 mm by ASTM 790-90 at the Forest Products Laboratory Engineering Mechanics Laboratory with load applied perpendicular to the surface of the fiber mat. A crosshead speed of 5 mm/min. was used. The stress-strain curves were analyzed using a three-parameter hyperbolic tangent model that has been shown to accurately represent the stress-strain behavior. The composite specimens were all tested to failure. Information collected in the tests included tensile strength at failure, tensile modulus, flexural strength, and flexural modulus. Notched and unnotched high-speed impact performance of the composites (13 by 65 mm) was evaluated by Izod impact according to ASTM D 256.

Scanning electron microscopy of fracture surfaces

Jute-reinforced specimens fractured in the Izod impact test were used for scanning electron micrographs of the fracture surface. Since the glass-reinforced specimens did not fracture completely, the cracked samples were broken by hand to reveal the surface. Samples were attached to the holder with conductive adhesive and sputter-coated with gold. Micrographs were taken with a JEOL JSM-840 at 15.0 kV

Ultraviolet degradation

Samples were exposed to cycles of ultraviolet light and water spray in a weatherometer to simulate outdoor exposure. Test conditions were continuous UV exposure for 102 minutes dry, followed by 18 minutes of water mist. Samples were exposed for a total of 1,200 hours, then dried and weighed on completion of the test.

Fungal degradation

Specimens made with each fiber type were tested using a soil block fungal test. Specimens measuring 25 by 25 mm were conditioned at 80°F and 30 percent relative humidity to constant weight, sterilized by steam at 212°F for 20 minutes, then aseptically placed in previously prepared soil culture bottles with a pure culture of either *Coriolus versicolor* or *Gloeophyllum trabeum.* The bottles were incubated for 12 weeks. Upon removal, the samples were conditioned to 80°F, 30 percent relative humidity, and weighed to determine the weight loss due to fungal attack, reported as percent of initial sample weight. Details of the procedure can be found in the fungal test portion of ASTM D 1413-76.

Results and discussion

Appearance and processing

No problems were encountered during processing. The resin was observed to flow as readily through three plies of jute fabric as through three plies of glass. Increasing jute plies from three to four had little effect on resin flow or back pressure, while some increase in back pressure occurred when resin was injected through five plies of jute. All composites produced had the same minimal amount of air bubbles entrapped in the mat. Fiber wet-out was the same for jute and glass. The finished jute composites were lighter in weight than the glass panels and less abrasive to the cutting equipment.

Mechanical properties

Tensile testing was performed according to ASTM 638-90. Tensile strength and modulus of the tested composites are shown in Figures 1 and 2 and in Table 4. Flexural testing was according to ASTM 790-90 and the flexural strength and modulus are presented in Figures 3 and 4 and in Table 5.

The treated and untreated jute specimens have similar values of tensile strength, tensile modulus,

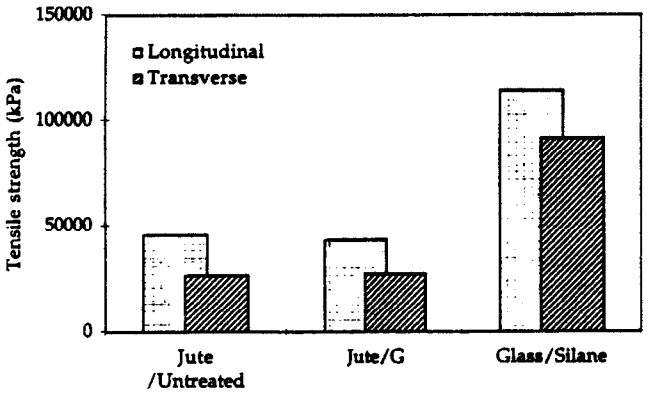

FIGURE 1.—Tensile strength of polyester composites.

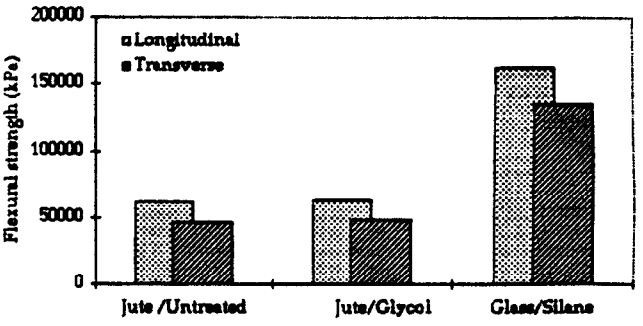

FIGURE 3.—Flexural strength of polyester composites.

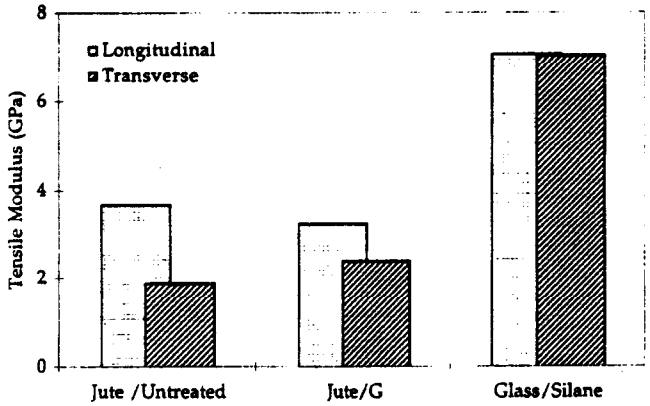

FIGURE 2.—Tensile modulus of polyester composites.

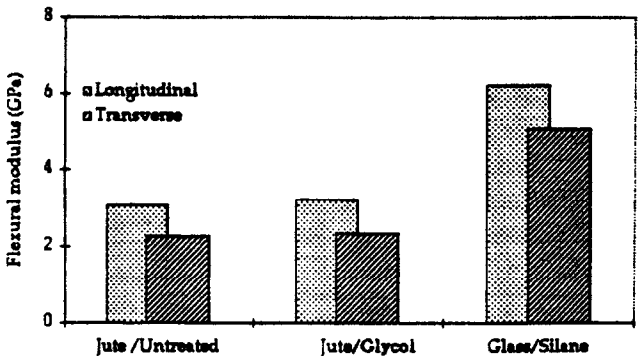

FIGURE 4.—Flexural modulus of polyester composites.

TABLE 4.—Comparison of tensile strength and modulus of polyester composites.[a]

Fiber/treatment	Strength (average kPa)	Modulus (average GPa)
Jute/untreated		
Longitudinal	45,818 (1,711.2)	3.6950 (0.3150)
Transverse	26,845 (707.1)	1.8840 (0.1192)
Jute/glycol		
Longitudinal	43,488 (2,930.6)	3.2250 (0.2033)
Transverse	28,076 (479.9)	2.3920 (0.0688)
Glass/silane		
Longitudinal	114,234 (1,669.5)	7.0867 (0.2922)
Transverse	91,358 (10,852.5)	7.0633 (0.7800)

[a] Standard deviation in parentheses.

TABLE 5.—Comparison of flexural strength and modulus of polyester composites.[a]

Fiber/treatment	Strength (average kPa)	Modulus (average GPa)
Jute/untreated		
Longitudinal	61,646 (1,931.0)	3.0500 (0.2033)
Transverse	46,759 (1,634.3)	2.2360 (0.0912)
Jute/glycol		
Longitudinal	63,153 (1,605.8)	3.2125 (0.1175)
Transverse	48,292 (895.8)	2.3383 (0.0756)
Glass/silane		
Longitudinal	161,916 (11,570.4)	6.2057 (0.3351)
Transverse	135,459 (4,799.6)	5.0700 (0.1800)

[a] Standard deviation in parentheses.

flexural strength, and flexural modulus, indicating that the treatment did not improve the properties of the composite. Values in the transverse direction are lower than in the longitudinal direction, because more fibers were oriented longitudinally than transverse. These samples have lower values than those reported for jute or sun hemp in the literature, primarily because in those studies the fibers were aligned along the axis of the test specimen and represent the maximum attainable values. Since our composites were made with a random mat, these values were expected to be lower. Jute compared with glass and with the neat resin indicates that jute composites are nearly the same as the neat resin, while the glass samples have better properties than unreinforced resin. Considering that the glass mats have a surface treatment to improve adhesion that has been highly optimized for composite applications, it is encouraging that the jute samples are within the same order of magnitude. The majority of the test specimens broke outside the gauge length. This indicates that a different specimen geometry should probably be used in future testing, and that the true values might prove to be higher than those reported here.

Results of Izod impact testing (ASTM D 256-90) are presented in Table 6. Both notched and unnotched specimens have a failure energy an order of magnitude lower than glass-reinforced specimens. The glass specimens in fact did not fracture during the test, and the actual energy required to fracture specimens is higher than the results indicate. In cross-linked thermoset resin systems, failure occurs by brittle fracture and depends on both crack initiation and crack propagation. Crack initiation begins at a flaw where stresses are concentrated. The crack propagates until the stresses are relieved or until it meets a higher strength material. Crack propagation in the impact test is very sensitive to adhesion between the matrix and the reinforcing fiber. Since the glass fiber-matrix interface has been highly optimized by the use of surface sizing, it can be assumed that stress transfer between matrix and glass fiber is good, leading to cracks stopping at the fiber. Figure 5 is a scanning electron micrograph of the fracture surface of the jute polyester Izod impact sample, showing fiber pull-out from the matrix. There is little fiber-matrix adhesion in the jute composites, therefore unmodified jute fibers cannot stop crack propagation. In fact, the fibers may serve as sites for crack initiation, thus reducing the energy required to break the specimens. In a study of jute fibers in polypropylene, the addition of a coupling agent increased failure energy of those composites by 80 to 100 percent (5). Modifying the jute fiber surface should lead to improved Izod impact strength.

TABLE 6.—Notched and unnotched Izod impact of polyester composites.

Fiber/treatment	Notched failure energy	Unnotched failure energy
	(average J/m)	
Jute/untreated		
Longitudinal	32.50 (3.45)	38.73 (4.26)
Transverse	39.75 (10.39)	58.85 (8.24)
Jute/glycol		
Longitudinal	28.64 (1.15)	32.40 (3.23)
Transverse	37.62 (1.41)	48.96 (4.25)
Glass/silane		
Longitudinal	376.81 (5.52)	376.63 (18.19)
Transverse	343.46 (15.31)	349.71 (13.80)

[a] Standard deviation in parentheses.

Physical testing

Jute and glass composites were exposed to white and brown rot wood-degrading fungi. Weight loss as a percent of initial weight is reported in Table 7. Though weight loss of jute specimens is higher than that of glass, the difference is within the accepted limits of error of the test. The glass and jute specimens had similar surface color change and unchanged structural integrity Specimens exposed to UV/water mist accelerated exposure showed no visual difference in color change or surface erosion between glass and jute specimens.

Conclusions

Jute fibers could be processed just as well as glass fibers in resin transfer molded composites. Tensile and flexural strength and tensile and flexural modulus of random jute fiber composites, though lower than glass fiber-reinforced composites, is in the same order of magnitude. Izod impact is an order of magnitude lower. Scanning electron micrographs of the Izod fracture surfaces show fiber pull-out, indicating poor fiber matrix interaction. Surface erosion when exposed to accelerated weathering is the same for jute and glass. Weight loss on exposure to wood-degrading fungi is slightly higher for jute composites than for glass, though quite small for both. Jute fibers can

FIGURE 5.—Scanning electron micrograph of jute fiber-reinforced polyester composite. Note the fibers pulled out of the matrix. which indicates poor adhesion of the matrix to the fiber.

be used unmodified in many composites where cost or lower weight is of greater importance than high strength. Future work should be to improve adhesion (using coating or fiber treatment) between the fiber and the matrix to improve strength and impact resistance. Higher fiber volume fractions can be used to reduce resin usage further, and properties of these higher fiber composites should be determined.

TABLE 7.—Percent weight loss for polyester composites exposed to white and brown rot fungi.

Fiber type	*Gloeophyllum trabeum*	*Coriolus versicolor*
	(%)	
Untreated jute	1.70	1.60
Glycol-treated jute	0.93	1.42
Glass	0.26	0

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