The Sixth International Conference on Woodfiber-Plastic Composites
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Accelerated Weathering of Natural Fiber-Thermoplastic Composites: Effects of Ultraviolet Exposure on Bending Strength and Stiffness

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Abstract

Mechanical properties of bending stiffness and yield stress were used to evaluate the effects of ultraviolet exposure on natural fiber-thermoplastic composites. Four different specimen formulations were evaluated. Injection molded high density polyethylene (HDPE) served as the polymer base for all formulations. Two lignocellulosic fillers, wood flour and kenaf fiber, were added at 50 percent by weight. Additives consisted of an antioxidant, coupling agent, and ultraviolet (UV) stabilizer. Specimens were exposed in a laboratory weatherometer to high levels of UV radiation and moisture cycling to simulate the effects of sunlight and rain for a 2000-h exposure period. Bending stiffness (modulus of elasticity) and bending yield stress (modulus of rupture) were measured prior to and after specific exposure periods. For the 100 percent HDPE formulation, stiffness increased and yield stress decreased after exposure. For HDPE with additives, no significant changes in mechanical properties occurred after 2,000 hours of exposure. Significant loss in mechanical properties was observed for both fiber-filled formulations. A 42 percent drop in measured stiffness and 24 percent drop in measured bending strength were recorded for the kenaf fiber formulation due to the 2,000-hr. exposure. A less dramatic drop in stiffness (33%) and strength (20%) was recorded for the wood flour formulation. For all formulations except HDPE with no additives exposed for 1,000 hours, the orientation of the degraded surface in bending, whether on the tension or compression side, had no significant effect on stiffness or strength.

Introduction

The range of products manufactured from natural fiber-thermoplastic composites (NFTC) is broadening every year. NFTC can offer improved thermal dimensional stability, improved stiffness, and lower filler costs compared to many conventional plastic resin products. In addition, the use of these materials provides opportunities to utilize recycled plastics and renewable lignocellulosic re-

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Sources in building products. One potential market for NFTC is the roofing market, but there is a lack of durability performance data from which product lifetime estimates could be made,

Exterior building products can be exposed to harsh environments, such as extremes in temperature, moisture cycling, and biological hazards. Of particular importance for exposed NFTC is the effect of ultraviolet (UV) exposure and moisture cycling. UV exposure can result in the embrittlement of otherwise ductile plastics due to initial fracture of outer surface layers (3). UV exposure also leads to rapid degradation of lignin, resulting in loss of fibrous material (4). Degradation of either fiber or matrix could reduce the ability of the composite to effectively transfer load between these components, resulting in lowered mechanical properties.

The service life of a building product can be assessed by performance factors. In the case of roofing, two of the most important factors are durability (waterproofness, fastener performance) and aesthetics (color retention and uniformity). Earlier work by Falk et al. (7) investigated the fastener performance of wood-flour-filled thermoplastics. In a preliminary phase of this study, we evaluated the color fade of several pigmented and unpigmented NFTCs (5, 6). Our intention was to maximize service life based on color retention. The objective of this study was to evaluate the effect of weathering on engineering properties and to investigate if the degradation of these properties was measurable.

Methods

Materials

The materials used in this study were high-density polyethylene (HDPE) and two natural fiber fillers, wood flour and kenaf fiber. The HDPE was Mobil 30-melt injection mold grade HMA018, and the wood flour was 40-mesh maple flour (American Wood Fibers, Sheboygan, WI). The kenaf fiber was purchased from the recycled fiber market. In an attempt to control fiber length, we utilized only the fibers passing through a 6.35-mm (1/4-in.) screen and stopped by a 1.59-mm (1/16-in.) screen.

Additives used in some formulations included a hindered amine UV stabilizer (TINUVIN 783 FDL; HALS), an antioxidant process stabilizer (B225 IRGANOX), and a compatibilizer (maleic anhydride grafted polyethylene, MAPE).

Table 1. ~ *Matrix of formulations tested.*

		Additives			Filler	
Formulation	Polymer HDPE	HALS	Anti-oxidant	MAPE	Wood flour	Kenaf
1	×					
2	×	×	×	×		
3	×	×	×	×	×	
4	×	×	×	×		×

Four formulations were compounded by Teel-Global Resource Technologies (TGRT), a natural fiber thermoplastic compounder located in Baraboo, Wisconsin (Table 1). Formulation 1 consisted of pure HDPE (no additives). Formulation 2 consisted of HDPE with antioxidant and HALS and MAPE additives. Formulation 3 contained 50 percent kenaf fiber by weight and Formulation 4, 50 percent wood flour by weight. Proprietary compounding equipment at T-GRT was used to compound these formulations.

Specimen Preparation

After compounding, the formulations were granulated and passed through a 6.35-mm (1/4-in.) screen and dried at 82°C (180°F) for 24 hours prior to injection molding. Injection molding was performed at the Forest Products Laboratory (FPL) using a 30-metric ton reciprocating-screw injection molder. Bending specimens were molded in accordance with ASTM D790 (2), which recommends specimen dimensions of 127 by 12.7 by 3.2 mm (5 by 1/2 by 1/8 in.). Injection speeds and pressures were varied for the different formulations.

Accelerated Weathering

Weathering of the composite specimens was accomplished using an Atlas Electric Devices xenon-arc weatherometer (Fig. 1). Exposure followed the International Conference of Building Officials (ICBO) Acceptance Criteria for Special Roofing Systems, a test method that outlines durability test procedures for synthetic roofing systems (9). The weatherometer was operated at 1 rpm and was set to provide 102 minute of light followed by 18 minutes of light and water spray. The irradiance was maintained at 0.35 W/m^2 at 340 nm, with a bandwidth of 1 nm. Black panel temperature was maintained at 62°C (144°F). Two water-cooled borosilicate filters provided direct daylight simulation. Specimens were placed on racks without

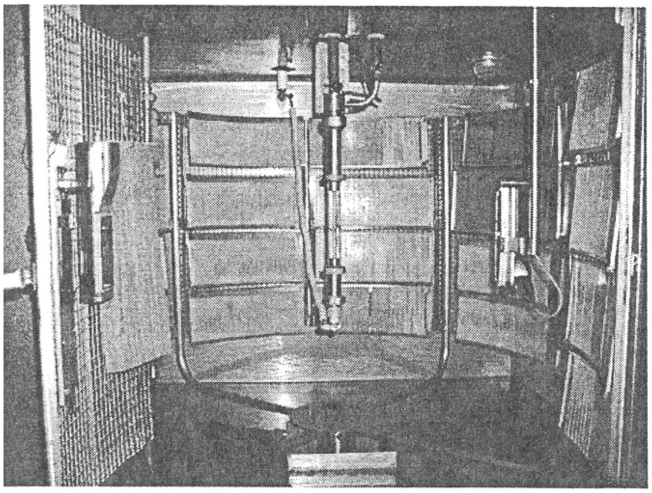

Figure 1. ~ *Interior of accelerated weathering chamber (Weatherometer).*

backing and exposed on one side for a total of 2,000 hours. At the end of every 500 hours, selected specimens were removed and tested.

Testing

Bending tests were performed on both unweathered (control) and weathered specimens at the FPL Engineering Mechanics Laboratory, using a 2-KN (450-lb.) Instron 5540 testing machine and a three-point bending setup (Fig. 2). Thirty specimens of each formulation were tested in accordance with ASTM D790 test method I, procedure B (2). The support span was 51.2 mm (2.0 in.); corresponding to an L/d of 16, and the rate of strain was 0.10 mm/mm minute (in/in. min.). Specimens were conditioned for at least 48 hours at 20°C (68°F) and 50 percent relative humidity (RH) prior to testing. Continuous load and deflection data were recorded. Because the specimens were weathered on only one side, flexural tests were performed on both the compression and tension side of the exposed surface. To calculate bending stiffness, a four-parameter hyperbolic tangent function was fit to the load displacement curve. This method was chosen to coordinate with previous research. The initial slope of this fitted function provides one estimate of stiffness (10). Bending strength was determined for failed specimens using linear elastic bending theory. For specimens that did not fracture (typical of ductile plastics), the stress corresponding to 3 percent strain was used as a measure of strength.

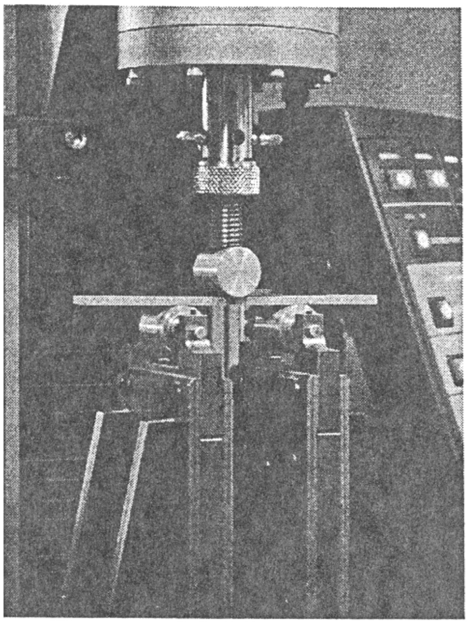

Figure 2. ~ *Three-point bending setup.*

Measurement of Degradation

The surface of a composite degrades throughout the weathering process. We attempted to measure this depth of degradation using an optical microscope. The depth of degradation was measured along the edge of the specimen corresponding to the whitened fiber layer.

Results and Discussion

Bending Stiffness

To provide an indication of changes induced by accelerated weathering, bending stiffness was calculated. Table 2 shows the effect of weathering on composite bending stiffness. The data are the mean values of 30 specimens per each formulation. Actual data are shown in Figure 3. For Formulation 1 (HDPE-no additives), a 64 percent increase in bending stiffness was found between 0 and 2,000 hours of exposure. This increase in modulus can likely be explained by an increase in crystallinity due to photo-oxidation and cross-linking (8). A 6 percent decrease in bending stiffness was found for Formulation 2 (HDPE-with additives).

More dramatic changes in composite stiffness were apparent for both Formulation 3 (kenaf fiber) and Formulation 4 (wood flour). As shown in Table 2, Formulations 3 and 4 showed a respective 42 percent and 33 percent reduction in stiffness. This reduction might be explained by the breakdown of

Table 2. ~ *Effect ofweathering on composite bending stiffness.*[a]

Formulation	Mean modulus of elasticity (MPa) at various exposure times				
	0 hr.	500 hr.	1,000 hr.	1,500 hr.	2,000 hr.
1. HDPE (no additives)	668.3 (4.2)	714.1 (3.1)	968.1 (4.3)	1,079.6 (3.2)	1,098.0 (6.1)
2. HDPE (w/additives)	816.8 (2.7)	773.1 (1.7)	751.3 (2.2)	744.4 (2.2)	768.9 (2.8)
3. 50% kenaf	5,947.5 (6.7)	4,577.4 (6.9)	3,595.9 (5.8)	3,470.6 (5.8)	3,426.6 (4.5)
4. 50% wood	3,950.8 (1.6)	3,329.3 (1.7)	2,798.2 (2.6)	2,650.0 (3.6)	2,631.3 (3.0)

[a] Average of 30 specimens. Coefficients of variation (%) are shown in parentheses.

the fiber or particle (flour) and/or the fiber-plastic bond due to the moisture cycling. Additionally, a lignin breakdown and a reduced bond between the cellulose and hemicelluloses likely occurred.

Table 2 and Figure 3 also show the dramatic increase in stiffness achieved by adding wood flour and kenaf fiber to HDPE. Before exposure, Formulation 4 specimens were over four times stiffer than Formulation 2 specimens. After 2,000 hours of exposure, Formulation 4 specimens retained more than three times the stiffness of Formulation 2 specimens. Similarly, Formulation 3 specimens were more than seven times stiffer than Formulation 2 specimens before exposure. After 2,000 hours of exposure, Formulation 4 specimens were more than four times stiffer than Formulation 2 specimens.

Bending Strength

Modulus of rupture (MOR) was calculated for each specimen that fractured. Formulation 1 specimens exposed for 500 hours or less and all Formulation 2 specimens regardless of length of exposure did not fracture until the deflection became excessive. In these cases, we calculated flexural yield stress using an assumed failure stress corresponding to 3 percent strain. After 1,000 hours of exposure, Formulation 1 specimens tested with the exposed surface on the compression side did not fracture before 3 percent strain was reached; however, Formulation 1 specimens tested with the exposed surface on the tension side did fracture. All Formulation 1 specimens fractured at exposures greater than 1,000 hours.

Weathering had quite a different effect on strength compared with stiffness. Table 3 shows the results of the effect ofweathering on composite yield stress; the actual data are shown in Figure 4. For Formulation 1 (HDPE-no additives), a 29 percent decrease in yield stress was recorded after 2,000 hours of exposure. No change in yield stress was found for Formulation 2 (HDPE-with additives). For Formulations 3 and 4, weathering caused a 24 percent and 20 percent reduction in MOR, respectively.

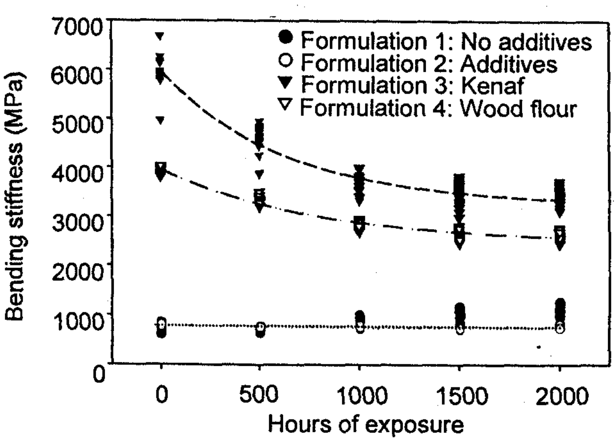

Figure 3. ~ *Efflect ofaccelerated weathering on bending stiffness.*

Table 3 and Figure 4 also show the increase in strength resulting from adding. wood flour and kenaf fiber to HDPE. Before exposure, Formulation 4 specimens were more than twice as strong as Formulation 2 specimens. After 2,000 hours of exposure, Formulation 4 specimens retained most of their strength (1.85 times) compared to Formulation 2 specimens. Similarly, Formulation 3 specimens were over 2.5 times stronger than Formulation 2 specimens. After 2,000 hours of exposure, Formulation 4 specimens were still 1.5 times as strong as Formulation 2 specimens.

Variability in Properties

Both Tables 2 and 3 indicate that the variability in the tested composite properties is generally low. The coefficient in variation (COV) for stiffness properties for all formulations ranged from 1 percent

Table 3. ~ *Effect of weathering on composite yield stress.*[a]

Formulation	Mean modulus of rupture (MPa) at various exposure times				
	0 hr.	500 hr.	1,000 hr.	1,500 hr.	2,000 hr.
1. HDPE (no additives)	14.0 (4.7)	15.6 (2.6)	14.3 (12.7)	11.0 (19.5)	9.9 (12.5)
2. HDPE (w/additives)	16.5 (1.3)	16.8 (1.3)	16.0 (1.4)	16.0 (1.2)	16.5 (1.6)
3. 50% kenaf	48.2 (4.8)	41.3 (4.3)	37.6 (5.0)	37.0 (3.0)	36.5 (3.6)
4. 50% wood	36.9 (2.4)	32.3 (2.1)	30.8 (2.4)	29.8 (2.6)	29.7 (3.0)

[a] Average of 30 specimens. Coefficients of variation (%) are shown in parentheses.

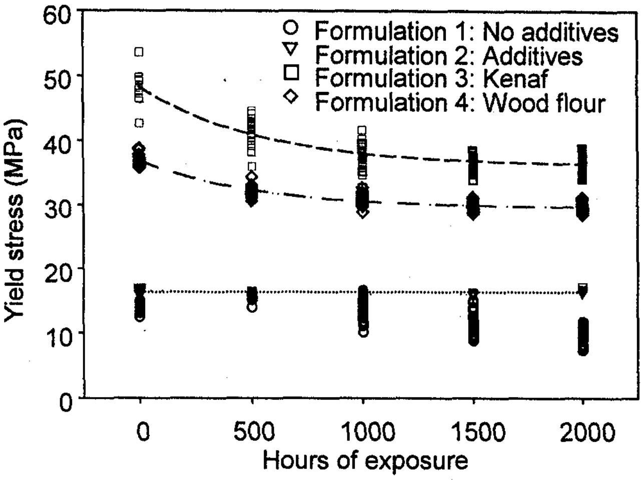

Figure 4. ~ *Effect of accelerated weathering on bending yield stress.*

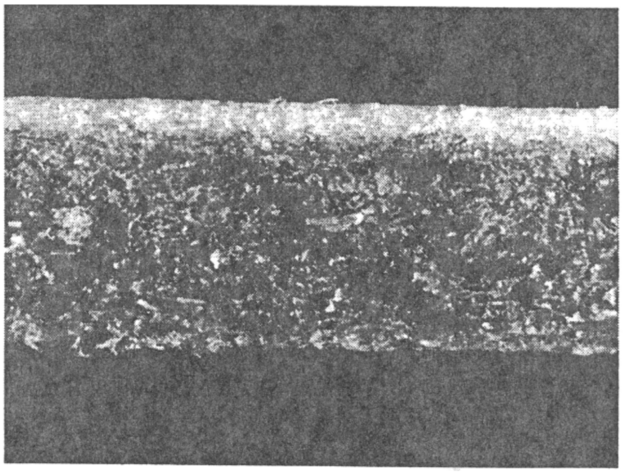

Figure 5. ~ *Edgewise depth of degradation for Formulation 3 (kenaf) at 2,000 hours of exposure. Whitened layer corresponds to degraded depth.*

to 8 percent; the higher end of this range was exhibited by Formulation 3 (kenaf).

For strength, both Formulations 3 (kenaf) and 4 (wood) exhibited lower variability (from about 2% to 5%) than that of the other formulations. Significantly higher variability was observed in MOR for Formulation 1 for exposures greater than 1,000 hours. Observations of the weathered specimens and the corresponding failure types offer an explanation for this variability. Nearly all the weathered Formulation 1 specimens began to exhibit surface crazing at irregular intervals across the surface at 1,000 hours of exposure. This crazing propagated surface cracks perpendicular to the mold filling flow (perpendicular to the specimen length). At 1,000 hours of exposure, these cracks were located sporadically and lowered the strength of only some specimens. Other specimens were not affected. This had the overall effect of increasing variability in the strength results. As exposure time increased, more cracks formed in more specimens. This lowered not only strength but also variability at the longer exposure levels (1,500 and 2,000 hr.).

Measurement of Surface Degradation

In an attempt to quantify the degradation of the weathered surface, we visually measured the depth of degradation using an optical microscope. We assumed that the depth of degradation corresponded to the whitened layer. Figure 5 shows edgewise depth of degradation of an exposed specimen (2,000 hr.) for Formulation 3 (kenaf); the visible depth of degradation for Formulation 4 (wood flour) is plotted in Figure 6. A rather large variation in measured depths of degradation is apparent, a result of the non-uniform orientation of wood particles. Similar variation in measured depths of degradation was observed for Formulation 3 (kenaf), but greater depth of degradation was observed for the same exposure period.

We used the measurement of degradation depth to adjust the effective depth of the specimen cross section for stress calculations. Our hope was that this effective depth modification serves as a pre-

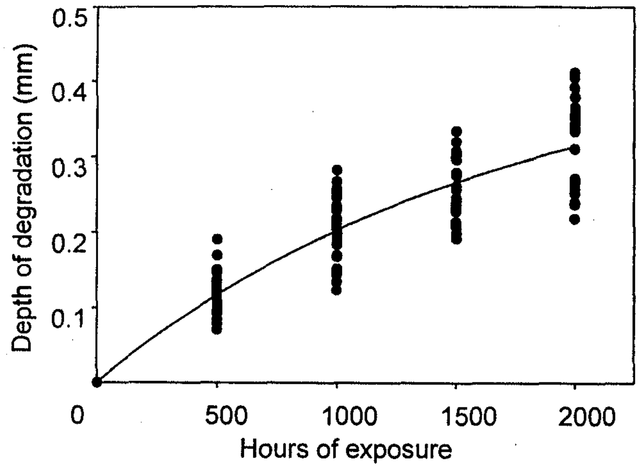

Figure 6. ~ *Effect of accelerated weathering on the visible depth of degradation for Formulation 4 (wood flour).*

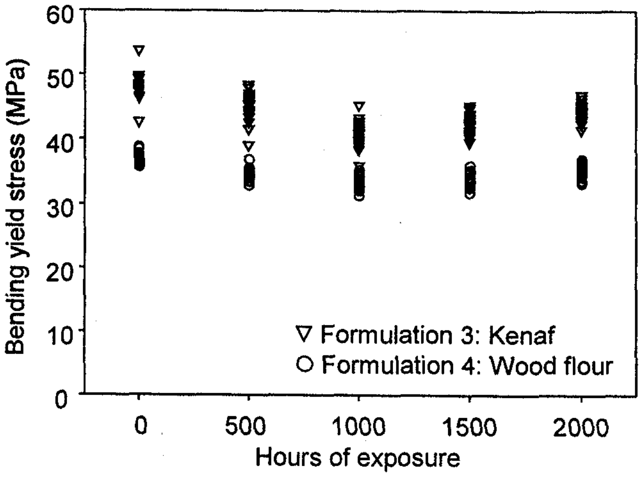

Figure 7. ~ *Bending yield stress calculated from visible depth of degradation measurements,*

dictor of the loss of bending strength with exposure. First, the assumption was made that the whitened layer does not contribute to the flexural properties of the weathered composite. Using this assumption, predicted failure stresses were calculated. Figure 7 shows the predicted bending yield stress calculated using the measured depth of degradation. If the measured depth accurately predicts a reduction in cross section, the calculated bending stresses shown in Figure 7 would be constant with exposure. It is apparent from Figure 7 that for both fiber-filled composites, the visible depth of degradation underpredicts the actual depth of degradation for short exposures and more accurately predicts the depth of degradation for longer exposures.

Conclusions

The results of bending tests of natural fiber-thermoplastic composites exposed to 2000 hours of accelerated weathering (UV light and water spray) led to the following conclusions:

1. Unfilled HDPE (no additives) increases in stiffness and decreases in strength with exposure.
2. The strength and stiffness of unfilled HDPE (with additives) is relatively unaffected by exposure.
3. The stiffness and strength of kenaf-filled HDPE decreases significantly with exposure (42% and 24%, respectively).
4. The stiffness and strength of wood-filled HDPE decreases significantly with exposure (33% and 20%, respectively).
5. The addition of natural fillers to HDPE results in significantly higher stiffness and strength than that of unfilled HDPE even after 2,000 hours of accelerated weathering.
6. The orientation of the degraded surface on the tension or compression side of a weathered bending specimen does not significantly affect flexural strength or stiffness.
7. Using a visual depth of degradation to adjust specimen effective depth for bending strength calculations underpredicts the depth of degradation, especially for short exposures.

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Sixth International Conference on Woodfiber-Plastic Composites

May 15–16,2001
The Madison Concourse Hotel
Madison, Wisconsin

Sponsored by the USDA Forest Service in cooperation with the
ACS Cellulose, Paper and Textile Division, the American Plastics Council,
the Market Development Alliance of the FRP Composites Industry,
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