

Accelerated Weathering of Natural Fiber-Filled Polyethylene Composites

Thomas Lundin¹; Steven M. Cramer²; Robert H. Falk³; and Colin Felton⁴

Abstract: The resistance of natural fiber-filled high-density polyethylene composite specimens to ultraviolet- (UV) and moisture-induced degradation was evaluated by measuring changes to flexural properties. High-density polyethylene (HDPE) served as the polymer matrix for four formulations: two formulations without fiber filler and two formulations one containing wood flour and the other containing kenaf fiber, each added at 50% by weight. Specimens were exposed for 4,000 h to UV radiation and moisture cycling in a laboratory weathering device to simulate the effects of exposure to sunlight and rain. Modulus of elasticity and modulus of rupture were measured prior to and after specific exposure periods. The flexural modulus and strength decreased significantly with increasing exposure for both natural fiber-filled HDPEs, but these composites still outperformed composites without filler. To explore a practical means to predict the changes in flexural properties to observed physical changes, a two-layer elementary mechanics model consisting of a degraded surface layer and an unaffected core layer was considered. The model was only partially successful—it provided reasonable predictions of flexural strength but did not accurately predict modulus of elasticity.

DOI: 10.1061/(ASCE)0899-1561(2004)16:6(547)

CE Database subject headings: Weathering; Polyethylene; Fiber reinforced plastics; Composite materials; Wood products; Durability.

Introduction

Natural fiber-filled thermoplastic composite products are rapidly penetrating the building products market. Wood/thermoplastic composite lumber, products with 50% or less plastic by weight, have been accepted by the construction industry and homeowners, primarily for decking applications. These composites are estimated to constitute 3–10% of the decking market (Eckert 2000). Currently, some 70 North American firms annually produce approximately 16–24 million board feet of plastic lumber (at least 50% plastic by weight), which was worth roughly \$40–\$60 million in 1996 (Powell 1996). The primary advantages claimed for these composite products are low maintenance, uniformity in properties and performance, and longevity. The disadvantages are their relatively low structural properties compared to those of structural lumber, which have limited their use to nonstructural or semi-structural applications such as landscaping, piers and docks, wall panels, and outdoor furniture. In addition, market development has been hampered by a lack of durability performance data

and standards, and by the reluctance of homebuilders and owners to utilize undemonstrated products.

Both thermoplastics and natural fibers are susceptible to environmental stresses, including temperature, moisture, light [ultraviolet (UV) radiation], and chemical agents such as organic solvents, ozone, acids, and bases, though most polymers are primarily degraded by oxidative reactions (Gillen et al. 1986). Outdoor weathering is a common case of oxidation enhanced by photochemical reaction, which is referred to as photodegradation.

Photodegradation of some thermoplastics can result in changes to polymer morphology because of chemical cross linking or chain scission (Davis and Sims 1983). Photodegradation is generally confined to the surface of thick (>3 mm, > 0.12 in.) polymer samples, either as a result of limited oxygen diffusion (Furneaux et al. 1981) or limited UV penetration (Ashbee 1993). For lignocellulosics, photodegradation can result in the breakdown of lignin, hemicelluloses, and cellulose, and the loss of fibrous material (Hon 2001). The mechanical properties of fiber-based composites are dependent upon the interaction and adhesion between the polymer matrix and the reinforcing fiber. Degradation of fiber, matrix, or the interface caused by weathering can reduce the ability of the composite to effectively transfer stress between these components, resulting in lowered mechanical properties. Artificial weathering tests have been developed for materials intended for use in outdoor environments. These tests have been shown to have an adverse effect on both physical and mechanical properties of polymers and lignocellulosics (Rowell et al. 1998; Dash et al. 2000; Falk et al. 2000a,b; Kiguchi et al. 2000; Matuana et al. 2001). The degree of degradation in these tests and its impact on composite mechanical properties have not been well documented.

The objective of the research was to evaluate the relative durability of natural fiber-filled high-density polyethylene composites (NF-HDPE) intended for exterior roofing applications based on a single accelerated weathering protocol. The NF-HDPE specimens were evaluated by measuring changes to flexural properties due to accelerated weathering (Lundin 2002). The effects of ac-

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Note. Associate Editor: Roberto Lopez-Anitlo. Discussion open until May 1, 2005. Separate discussions must be submitted for individual papers. To extend the closing date by one month, a written request must be filed with the ASCE Managing Editor. The manuscript for this paper was submitted for review and possible publication on March 14, 2003; approved on February 23, 2004. This paper is part of the *Journal of Materials in Civil Engineering*, Vol. 16, No. 6, December 1, 2004. ©ASCE, ISSN 0899-1561/2004/6-547-555/\$18.00.

Table 1. Constituent Materials (% Mass) and Composite Density (g/mm³)

Ingredient	Formulation			
	High-density polyethylene (HDPE)	Additives	Wood	Kenaf
HDPE, HMA018	100.0%	97.3%	48.6%	48.6%
Tinuvin 783 FDL	–	0.4%	0.4%	0.4%
Irganox B225	–	0.2%	0.2%	0.2%
Wood flour No. 40 mesh	–	0%	48.6%	0%
Kenaf fiber	–	0%	0%	48.6%
MAPE	–	2.1%	2.1%	2.1%
Density of composite formulation (g/mm ³)	9.1×10^{-4}	9.25×10^{-4}	1.108×10^{-3}	1.128×10^{-3}

celerated weathering on the mechanical properties of NF-HDPEs and the influence of a degraded layer on the effective cross section were studied.

Experimental Methods

Materials

The materials used in this study were virgin HDPE and two natural fiber fillers, wood flour and kenaf fiber (Kengro Inc., Charleston, Miss.). The HDPE was Mobil 30-melt (melt flow index = 30 g/10 min and density = 9.54×10^{-4} g/mm³) injection mold grade HMA018 (Muehlstein, Inc., Norwalk, Conn.) and the wood flour was 40-mesh (425 μ m) maple flour (AWF-4010, American Wood Fibers, Sheboygan, Wis.). The kenaf fiber was purchased from the recycled fiber market. To provide some control on kenaf fiber length, we utilized only fibers that passed through a 6.35 mm (1/4 in.) screen and retained on 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 Uniroyal Chemicals, MB226D maleic anhydride grafted polyethylene (MAPE). The compatibilizer reduced interfacial surface tension of otherwise immiscible polymers (Lu et al. 2000).

Four formulations were compounded for study by Teel-Global Resource Technologies, a natural fiber thermoplastic compounder located in Baraboo, Wis. (Table 1): (1) pure HDPE (no additives); (2) HDPE with antioxidant, HALS, and MAPE additives; (3) wood flour, 50% by weight; and (4) kenaf fiber, 50% by weight.

Specimen Preparation

After compounding, the formulations were granulated, passed through a 6.35 mm (1/4 in.) screen, and dried at 82°C (180°F) for 24 h prior to injection molding. Injection molding was performed at the Forest Products Laboratory using a 33 t reciprocating-screw injection molder (Vista Sentry VSX-33, Cincinnati Milcron, Cincinnati, Ohio). Flexural specimens were molded in accordance with *ASTM D790* (ASTM 1996), which recommends specimen dimensions of 127 by 12.7 by 3.2 mm (5 by 1/2 by 1/8 in.). Injection speeds were identical for Formulations 2, 3, and 4, and 30% faster for Formulation 1. Injection pressures for Formulation 2 were 40% higher than Formulation 1, and Formulations 3 and 4 were twice that of Formulation 1. Further detail on this process is presented by Lundin (2002).

Accelerated Weathering

Composite specimens were weathered using an Atlas Electric Devices (Wauconda, Ill.) 65-WT xenon-arc weatherometer. Because roofing was the intended application for these composites, exposure followed the International Conference of Building Officials (ICBO) Acceptance Criteria for Special Roofing Systems, which outline durability test procedures for synthetic roofing systems (ICBO 1997). Specimens were rotated by the weatherometer at 1 rpm around a spray nozzle and UV source. The device was set to provide 102 min of light followed by 18 min of light and water spray, exposing one side of the specimens to direct light and water spray while subjecting both sides to high levels of humidity. The irradiance was maintained at 0.35 W/m² when measured at 340 nm, with a bandwidth of 1 nm. The 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 backing for a total of 4,000 h, which is 2,000 h more than required by the ICBO criteria. At the end of every 500 h, selected specimens were removed for testing and not returned to the test chamber.

Flexural Testing

Flexural tests were performed on both unweathered (control) and weathered specimens in the Engineering Mechanics Laboratory of Forest Products Laboratory, using a 2 kN (450 lb) Instron 5540 testing machine, a three-point bending setup, and a linear variable differential transformer that recorded relative displacement between the loading head and the supports. Thirty specimens per formulation were held as controls, and two sets of 15 specimens were tested at the end of every 500 h of exposure. One set of 15 specimens was tested with the exposed surface oriented on the tension side; the other set was tested with the exposed surface on the compression side. Specimens of each formulation were tested in accordance with *ASTM D790* test method I, procedure B (ASTM 1996). The support span was 51.2 mm (2.0 in.), corresponding to a span to depth ratio of 16, and the rate of strain was 0.10 mm/mm min (in./in. min). Specimens were conditioned for at least 48 h at 20°C (68°F) and 50% relative humidity prior to testing. Continuous load and deflection data were recorded. Flexural properties of modulus and strength were calculated using the gross cross section. Flexural modulus was calculated using a four-parameter hyperbolic tangent function fit to the load displacement data. This method was chosen to coordinate with previous research (Falk et al. 2000a). The initial slope of this fitted function provides one estimate of the initial tangent modulus (Murphy 1992). Flexural strength was determined for failed specimens

Table 2. Effect of Accelerated Weathering on Flexural Modulus of Composites^a

Formulation	Flexural modulus (MPa) at various exposure times				
	0h	1,000 h	2,000 h	3,000 h	4,000 h
High-density polyethylene					
Exposed surface on compression side	668 (4.2)	944 (4.4)	1,140 (5.9)	522 (55.9)	567 (7.8)
Exposed surface on tension side	668 (4.2)	989 (3.8)	1,060 (6.3)	735 (8.8)	497 (8.6)
Additives					
Exposed surface on compression side	817 (2.7)	751 (2.2)	790 (11.9)	703 (3.5)	702 (3.5)
Exposed surface on tension side	817 (2.7)	751 (2.2)	762 (2.5)	703 (3.5)	686 (1.6)
Wood (50%)	3,950 (1.7)	2,800 (2.5)	2,630 (3.0)	2,040 (2.9)	1,770 (4.9)
Kenaf (50%)					
Exposed surface on compression side	5,950 (6.7)	3,600 (5.8)	3,390 (3.3)	2,550 (4.7)	2,280 (5.3)
Exposed surface on tension side	5,950 (6.7)	3,600 (5.8)	3,462 (5.4)	2,550 (4.7)	2,280 (5.3)

^aValues are averages. Coefficient of variation values (%) are given in parentheses.

Fifteen replications for each condition.

using linear elastic bending theory. For specimens that did not fracture before deflections became excessive (typical of ductile plastics), the stress corresponding to 3% strain was used as a measure of ultimate bending strength.

Results and Discussion of Effects of Accelerated Weathering

Density

Density was measured after production of the composite. The resulting mean values are shown in Table 1 for each formulation and the variations in measured values were insignificant. After an exposure period and before flexural testing, density was again measured with the general observation that accelerated weathering exposures prompted little or no change in density. At 4,000 h of exposure, density deviated from the initial value by no more than 1.5%.

Flexural Modulus

Flexural modulus (modulus of elasticity) values were calculated from the bending test data. Table 2 summarizes the effects of accelerated weathering on mean flexural modulus and coefficient of variation for all formulations and orientations of the degraded layer. To determine if the orientation of the degraded layer (surface nearest to light source) affected the flexural properties of the composites, a one-way analysis of variance was performed for all formulations for both flexural modulus and strength. If there were no statistically significant difference between the means at 95% confidence of those specimens tested with the degraded layer on the compression side and those tested with the degraded layer on the tension side in flexure, then only one value is shown at that exposure (Table 2).

For composites made with pure HDPE, the orientation of the exposed surface had a significant effect on flexural modulus for all exposure periods (Table 2). Loss in modulus was more severe for specimens tested with the exposed surface on the tension side. The high coefficient of variation at 3,000 h of exposure was a result of nonuniform crazing. For HDPE with additives, the orientation of the exposed surface was significant at 2,000 and 4,000 h of exposure. As for pure HDPE composites, loss in modulus was greater when the exposed surface was oriented on the tension side. For the NF-HDPEs, the orientation of the exposed surface in

general had no significant influence on flexural modulus. The addition of lignocellulosic fiber to unfilled HDPE appeared to reduce the influence of the orientation of the degraded layer on flexural modulus because there was no statistically significant difference in modulus between when the degraded layer was oriented on the compression and tension sides.

Fig. 1 shows least-squares regression fits of flexural modulus for all four formulations; error bars are equal to ± 1 s.d. based on both orientations per formulation. The flexural modulus for NF-HDPEs (composites filled with wood or kenaf fiber) was significantly greater than that for the unreinforced HDPE thermoplastics, regardless of length of exposure. The flexural modulus for all four formulations decreased over the 4,000 h of exposure. The flexural modulus of stabilized HDPE was reduced roughly 15% indicating that the stabilizer was effective in minimizing the changes in modulus caused by weathering. The most significant loss in modulus (62%) was for kenaf composites. Nonetheless, at the end of the exposure period, the flexural modulus of kenaf composites was still 3.3 times as great as that of HDPE composites with additives.

The flexural modulus for pure HDPE increased 70% during the first 2,000 h of exposure. This could indicate that this formulation was undergoing photoinduced molecular cross linking, resulting in longer molecular chains and greater modulus (Davis and Sims 1983). The flexural modulus of pure HDPE was greater than that of HDPE with additives after 1,000 and 2,000 h of exposure but was less than that of HDPE with additives at exposures of 3,000 h and greater. After 2,000 h, pure HDPE potentially underwent chain scission (division of tie molecules connecting the crystalline polymer regions), resulting in lowered modulus (Hoekstra 1997). The flexural modulus for HDPE with additives decreased between 0 and 1,000 h and between 2,000 and 4,000 h, but it increased between 1,000 and 2,000 h of exposure. After 4,000 h, the modulus was 85% of the unexposed modulus, which could indicate that some UV stabilizer was consumed.

For pure HDPE, the variability in modulus was quite high at 2,500 and 3,000 h of exposure (Fig. 1). This could be explained by the occurrence of small parallel surface cracks (crazes), the formation of which has been attributed to chain scission (Schoolenberg and Vink 1991). Crazes began to appear after 1,000 h of exposure and their concentration increased with increasing exposure. Crazing induced a slight camber in the HDPE composites (without fibers). Because crazing was unevenly distributed, the curvature of the specimen was not constant, which

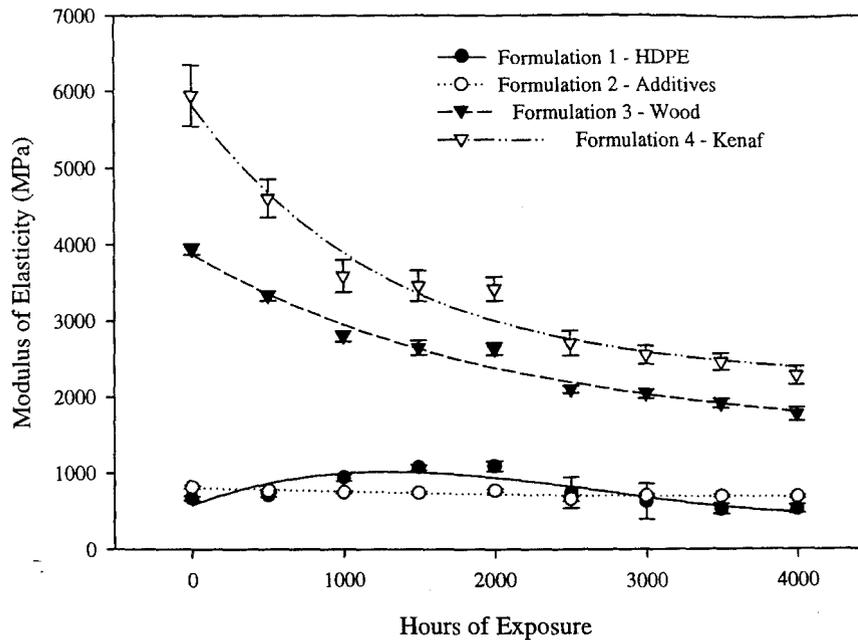


Fig. 1. Effect of accelerated weathering on flexural modulus of natural fiber-filled high-density polyethylene

resulted in a variety of deflected shapes and locations of failure. At 3,000 h of exposure, crazing on the specimen surface began to propagate through the thickness of the polymer and appeared sporadically along the length of the backside of the composite. After 3,500 h of exposure, crazing became regular along the backside, resulting in more consistent failures and lower variability.

The flexural modulus for wood and kenaf formulation specimens decreased with increasing exposure. For both NF-HDPEs, the greatest loss in modulus occurred during the first 1,000 h. For wood-filled composites, the modulus at 4,000 h of exposure was only 45% of that before exposure, though it was still twice as great as that of the neat and stabilized HDPE. The variability in properties was generally low for the NF-HDPEs. At the high filling volumes used in these composites, many fibers lie at or near the surface of the composite. Slight degradation to these fibers would likely result in significant reduction in interfacial bond, stress transfer, or reduction in effective cross section. Degradation or radical induced depolymerization of both lignin and hemicellulose, a result of UV oxidation (Evans et al. 1996), could induce a reduction in interfacial bond, stress transfer, or reduction in effective cross section.

Flexural Strength

Modulus of rupture was calculated for each fractured specimen. Pure HDPE specimens exposed for 500 h or less and specimens made with HDPE with additives, regardless of length of exposure, did not fracture until deflection became excessive. In these cases, the flexural yield stress was calculated as the stress corresponding to 3% strain. At 1,000 h of exposure, pure HDPE specimens tested with the exposed surface on the compression side did not fracture before 3% strain (full plastic action) was reached; however, pure HDPE specimens tested with the exposed surface on the tension side did fracture. All pure HDPE specimens fractured at exposures greater than 1,000 h.

Table 3 summarizes the effects of accelerated weathering on the mean flexural strength and its variation for all four formulations and both orientations in flexure. Variability in properties was

generally low and could be attributed to the use of a virgin polymer and dimensionally similar fillers. For pure HDPE, the orientation of the degraded layer had a significant effect at all levels of exposure; the highest strength values occurred with the degraded layer on the tension side when specimens were exposed for 2,000 h and greater. No consistent pattern of higher strength with any particular orientation occurred for the other formulations, although statistically significant differences in strength occurred in some situations.

Fig. 2 shows least squares regression fits of flexural strength, with error bars equal to ± 1 s.d. The flexural strength of NF-HDPEs was significantly greater than that of both unfilled thermoplastics (HDPE with and without additives), regardless of the length of exposure. Kenaf had the greatest flexural strength and pure HDPE the lowest flexural strength of all formulations. HDPE with additives was largely unaffected by the exposure conditions and even experienced a 5% increase in flexural strength at 4,000 h.

For pure HDPE, the variability in strength was greatest between 1,000 and 1,500 h and between 3,000 and 3,500 h of exposure; again, this is likely attributed to crazing. During these exposure periods, the cracks were located sporadically, which generally increased variability in the strength results. For the NF-HDPEs, flexural strength decreased with increasing exposure, with the most significant changes occurring in the first 1,000 h of exposure. The observed loss in flexural properties can likely be explained by the loss in the ability of polymer to transfer stresses to the reinforcing fiber.

Surface Degradation

To explore the source of the property reduction, several specimens were examined microscopically. Electron micrographs of the surface of NF-HDPEs were taken at various intervals of exposure (Fig. 3). The fiber is visible on the exposed surface [Fig. 3(b)], and there is little bond between the fiber and matrix. Without sufficient bond, stress cannot be transferred effectively and could be the cause of the reduction in flexural properties. To

Table 3. Effect of Accelerated Weathering on Flexural Strength of Composite^a

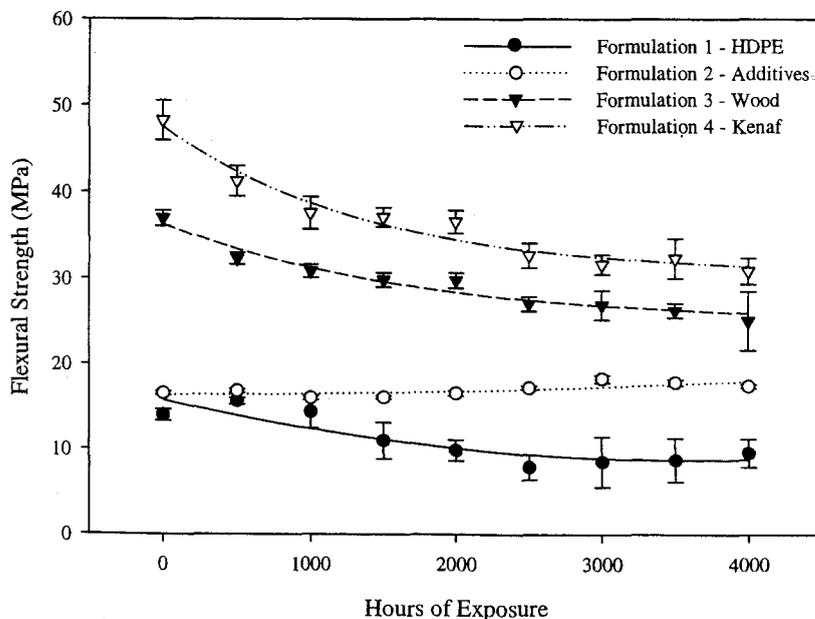
Formulation	Flexural strength (MPa) at various exposure times				
	0 h	1,000 h	2,000 h	3,000 h	4,000 h
High-density polyethylene					
Exposed surface on compression side	14.0 ^b (4.7)	15.8(6.8)	9.4(16.5)	5.6(6.5)	8.1(6.1)
Exposed surface on tension side	14.0(4.7)	13.0(11.0)	10.2(6.1)	11.4(4.0)	10.9(8.9)
Additives					
Exposed surface on compression side	16.5(1.3)	16.0(1.4)	16.6(1.2)	17.9(1.9)	17.3(1.4)
Exposed surface on tension	16.5(1.3)	16.0(1.4)	16.4(1.8)	18.5(1.2)	17.3(1.4)
Wood (50%)					
Exposed surface on compression side	36.9(2.4)	30.8(2.4)	29.7(3.0)	25.3(3.6)	26.2(4.0)
Exposed surface on tension side	36.9(2.4)	30.8(2.4)	29.7(3.0)	28.4(2.6)	24.8(7.4)
Kenaf (50%)					
Exposed surface on compression side	48.2(4.8)	37.6(5.0)	36.5(3.6)	31.6(3.6)	31.5(4.6)
Exposed surface on tension side	48.2(4.8)	37.6(5.0)	36.5(3.6)	31.6(3.6)	30.1(4.4)

^aValues are averages. Coefficient of variation values (%) are given in parentheses.

^bFifteen replications for each condition.

evaluate if weathering caused the formation of surface cracks and resultant reduction in interfacial bond, three specimens per formulation were monitored using a stereo microscope at two locations of interest for 2,000 h of exposure. The end of the specimen furthest from the injection mold gate was monitored because it had been observed to experience greater surface degradation than the other end. Fig. 4 (200 × magnification) shows the surface of a kenaf-filled composite before exposure and after 1,000 h. The fiber matrix interface along the top and bottom edges of the large piece of kenaf pith deteriorated during exposure. The observed changes can be attributed to the combination of the breakdown of lignocellulosic material by UV light and the removal of this material by the water spray, exposing previously protected material. It has been shown that the loss in weight of wood due to photodegradation is much greater in the presence of water and that the principle role of water is to facilitate light penetration into previously inaccessible regions (Hon 2001).

To quantify the degree of degradation of the weathered surface, we visually measured the depth of degradation d_i using the stereo microscope. We assumed that the depth of degradation corresponded to the whitened layer observed only in the lignocellulosic-filled composites. The whitened layer was observed on both sides of the exposed specimens; however, the depth of whitening on the side not directly exposed to the lamp was not measurable at the magnification employed (40 ×). This indicates that degradation occurred on both sides of the specimens, though the bleaching of fibers was amplified by the combination of direct UV light and water spray. Fig. 5 (40 × magnification) shows the edge of the kenaf-filled composite before exposure and after 4,000 h. An identifiable whitened layer appeared after weathering [Fig. 5(b)], which increased in depth with increasing exposure. The visible depth of degradation was greatest at the edges of the specimens and least in the middle of the specimen. For this reason, for all the specimens where the

**Fig. 2.** Effect of accelerated weathering on flexural strength of natural fiber-filled high-density polyethylene

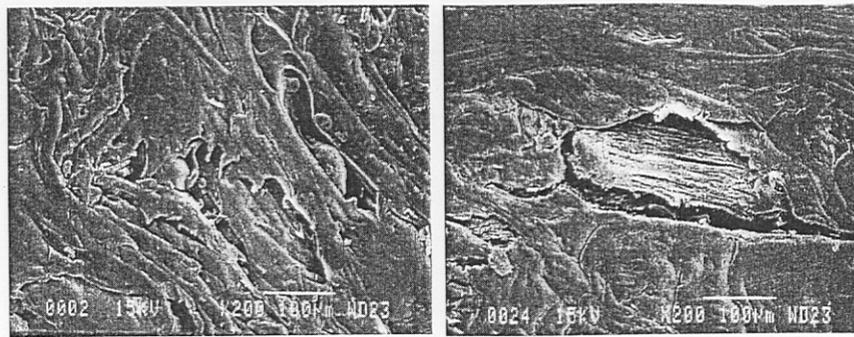


Fig. 3. Electron micrographs of surface of wood-filled natural fiber-filled high-density polyethylene before exposure (a) and after 2,000 h of exposure (b) to accelerated weathering

visible depth of degradation was measured, at least 2.0 mm of material was removed from the edge to eliminate any edge effects.

Fig. 6 shows the depth of degradation (d_d) for NF-HDPEs as a function of length of exposure. The mean values are plotted with error bars equal to one standard deviation and least squares regression lines using a two-parameter (a and b) exponential function of time (t) fit to the actual data as follows;

$$d_d = a(1 - e^{-bt}) \quad (1)$$

A rather large variation in measured depths of degradation was apparent. The coefficient of variation ranged from 18 to 30% for the wood-filled composite and 15 to 25% for the kenaf-filled composite. The large variation could be a result of the nonuniform orientation of fibers.

Analysis of Results

Prediction Model

The first and most direct approach to predicting flexural modulus and strength is to examine the viability of linear elastic bending theory as a prediction model. This method was investigated as a practical means to relate changes in mechanical properties to observed physical changes. In the model, gross specimen depth d , was reduced by assuming that the effective flexural rigidity was zero in the degraded whitened layer, d_w , and resulted in an effective depth, d_e (first equation). An effective moment of inertia, I_e (second equation), was calculated using d_e .

$$d_e = d - d_w \quad (2)$$

$$I_e = \frac{1}{12}b(d_e)^3 \quad (3)$$

General equations for the elastic modulus and strength (the following equations, respectively) were established for a simply supported beam with a center-point load using the effective moment of inertia [Eq. (3)]. These equations relate the material properties of modulus and strength to the load and deflection at length of accelerated weathering, x [Eq. (1)]

$$E = \frac{PL^3}{48I_e\delta} \quad (4)$$

$$\sigma = \frac{PLd_e}{8I_e} \quad (5)$$

where P = load required for deflection δ ; E = modulus of elasticity; and L = support span. Eq. (4) was evaluated by using the effective moment of inertia [Eq. (3)] based on the equation for effective depth [Eq. (2)] and then compared with results obtained from the test data. For both NF-HDPEs, the predicted modulus of elasticity at all periods of exposure was less than that calculated for the control specimens. The greatest difference for wood and kenaf formulations occurred at 1,000 h (27 and 42%, respectively). The smallest difference occurred at 500 h (9 and 20%, respectively).

Eq. (5) was evaluated in a similar manner. Again we found that at all periods of exposure the predicted strength was less than that calculated for the control specimens. The greatest difference for wood and kenaf occurred at 1,000 h (11 and 16%, respectively). For both formulations, the smallest difference (6%) occurred at 3,500 h.

It is likely that the visible depth of degradation was not entirely ineffective; this layer contributed in a limited way to the

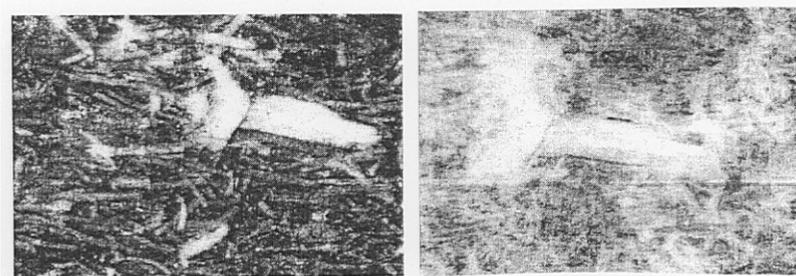


Fig. 4. Optical micrographs (40 \times) of surface of kenaf-filled natural fiber-filled high-density polyethylene before exposure (a) and after 1,000 h of exposure (b) to accelerated weathering

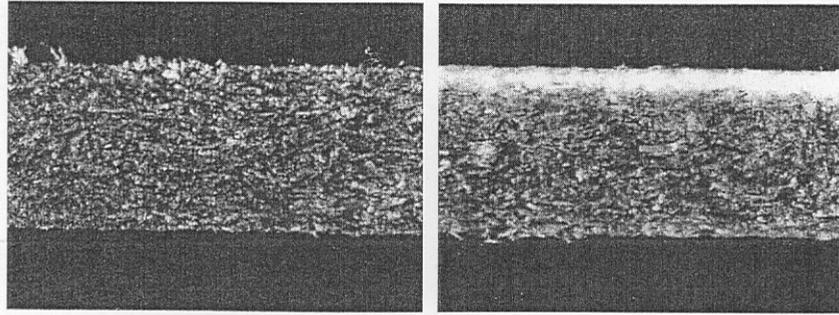


Fig. 5. Optical micrographs (40 ×) of edge of kenaf-filled natural fiber-filled high-density polyethylene before exposure (a) and after 4,000 h of exposure (b) to accelerated weathering

effective moment of inertia. In addition, on the side of the specimen not directly exposed to the lamp, moisture-induced degradation may have occurred, which was not measured and not considered in this simple model. A third possibility is that the modulus was not constant through the depth of the specimen, because the crystallinity and fiber orientation are generally anisotropic in injection-molded specimens (Osswald and Menges 1995). Overall, Eq. (4) provided an effective mechanistic prediction of flexural strength after exposure, especially for the wood formulation.

Flexural Modulus and Strength as Function of Specimen Depth

A set of experiments was conducted to determine the flexural modulus and strength of unexposed kenaf specimens as a function of depth. Kenaf was chosen due to the higher fiber aspect ratios, which would presumably result in greater effects on modulus and strength due to fiber orientation. A special set of specimens was produced by milling off the surface of extra unexposed injection-molded specimens. These specimens were milled on a vertical milling machine using a fly cutter with a carbide blade. The blade was oriented with a positive rake (shearing action) to produce the smoothest surface possible. A total of 44 specimens were milled at various depths. These specimens were tested in flexure with the milled surface on the tension side.

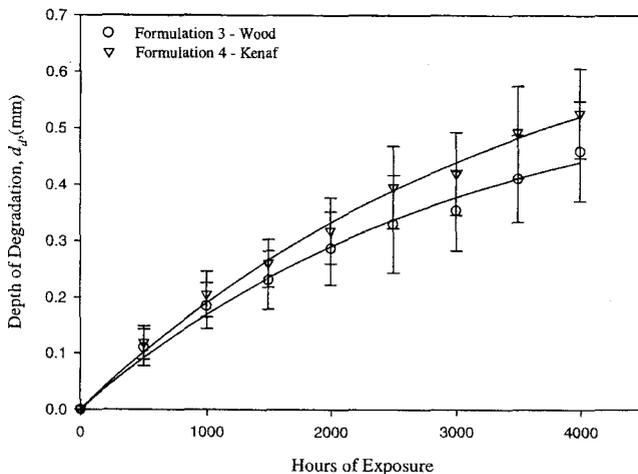


Fig. 6. Effect of accelerated weathering on visible depth of degradation for natural fiber-filled high-density polyethylene

Figs. 7 and 8 compare the predicted flexural modulus and strength of specimens with an effective cross section to the strength of milled specimens calculated using the prediction model. As these figures show, flexural modulus and strength are not constant and decreased with increasing depth removed. A 17% decrease in mean modulus was observed between specimens of full depth and those with 1.27 mm (0.05 in.) removed. Likewise, there was a 21% decrease in mean strength between specimens of full depth and those with 1.27 mm (0.05 in.) removed.

The modulus of the milled specimens was consistently greater than that of exposed specimens with an effective cross section (Fig. 7). This indicates that the effects of accelerated weathering on flexural stiffness were not confined to the visible degraded layer, as assumed in the two-layer model, suggesting the stiffness of the unwhitened material was also reduced as a result of exposure.

Both milled and weathered specimens had similar bending strengths at various specimen depths. Interestingly, the two-layer model appears to reasonably account for the loss in strength but to poorly predict the loss in modulus. The two-layer model is based on observable physical changes. These changes were more prominent on the side nearest the lamp but were not limited to this side, indicating that moisture-induced degradation occurred and that this degradation was amplified by direct water spray and UV light.

Conclusions

The degradation of NF-HDPE composites was investigated with a test protocol that combined cyclic UV exposure with moisture exposure. Using this weathering simulation, it was not possible to separate the individual effects of UV and moisture from the data. From test observations we can conclude that both aspects played an important role in the degradation of the NF-HDPEs. The flexural modulus of the NF-HDPEs decreased with increasing exposure, with the most dramatic changes occurring during the first 1,000h. Accelerated weathering reduced the modulus of the NF-HDPEs approximately 40%; however, the modulus was still 2.5 times greater than that of formulations without natural fibers. The flexural modulus was significantly affected by the orientation of the degraded layer for all formulations except the formulation containing wood flour.

The flexural strength of the NF-HDPEs decreased significantly during the first 1,000 h of exposure and retained approximately 65% of the original strength after 4,000 h of exposure. Despite the

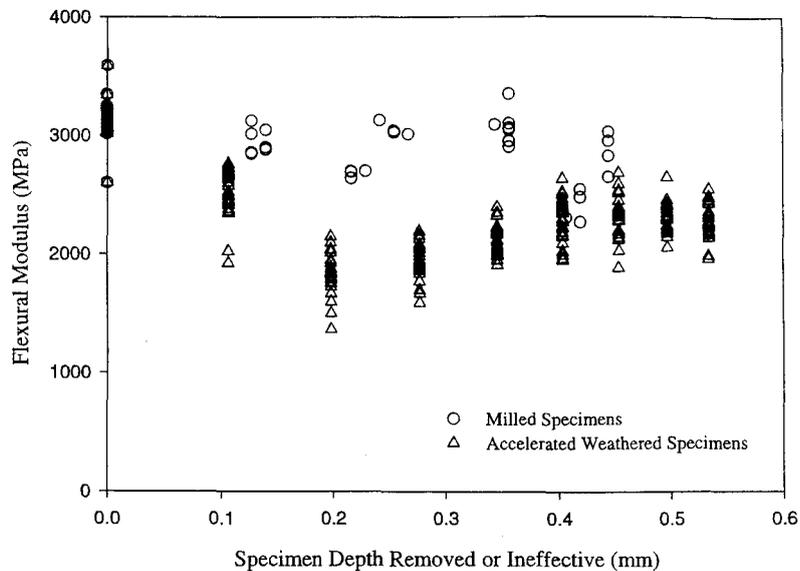


Fig. 7. Relationship between flexural modulus of milled specimens and weathered specimens using effective cross section

dramatic loss in strength with exposure, formulations containing wood flour and kenaf fiber outperformed unfilled polymer formulations. For all formulations, the orientation of the degraded layer significantly affected flexural strength. Both the flexural modulus and strength of weathered NF-HDPEs appear to have a consistent trend with exposure.

During accelerated weathering, NF-HDPEs developed a visually detectable degraded surface layer. The visible depth of degradation increased with greater exposure and could be characterized by loss in the interfacial bond between the fiber and matrix. The loss in interfacial bond was particularly noticeable around large pieces of lignocellulose fibers.

The two-layer linear elastic mechanics model did not predict flexural stiffness well but was effective in predicting the decrease in flexural strength with exposure. Although this theory is an obvious simplification of observed behavior, with further refinement it may offer a practical means to account for weathering degradation.

The flexural stiffnesses and strengths of injection-molded specimens containing 50% kenaf fiber were not constant through the depth of the specimens. Both bending stiffness and strength decreased approximately 20% through the depth of injection-molded specimens because of material flow characteristics.

Despite the significant changes to flexural properties caused by exposure, the NF-HDPEs were more predictable and possessed superior properties than similar unfilled HDPE composites.

Acknowledgments

The writers gratefully acknowledge the partial financial support of this research through the Partnership for Advancing Technologies in Housing (PATH). The Forest Products Laboratory is maintained in cooperation with the University of Wisconsin. This paper was written and prepared by U.S. Government employees on official time, and it is therefore in the public domain and not subject to copyright. The use of trade or firm names in this publication is for reader information and does not imply endorsement by the U.S. Department of Agriculture of any product or service.

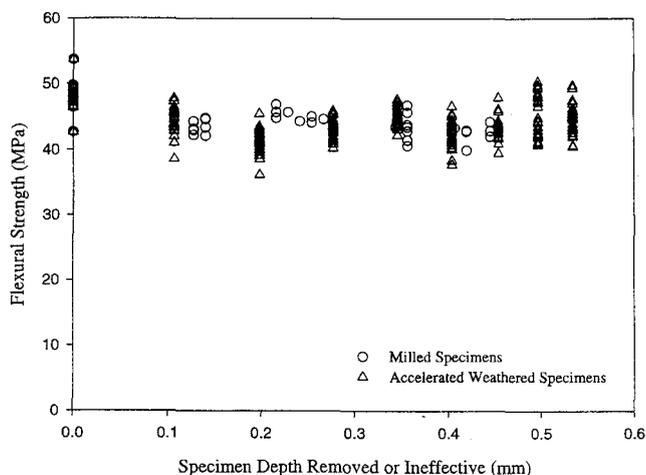


Fig. 8. Relationship between flexural strength of milled specimens and weathered specimens using effective cross section

Notation

The following symbols are used in this paper:

- a, b = parameters for least squares regression;
- d_d = depth of degradation;
- d_e = effective depth;
- d_i = gross specimen depth;
- E = modulus of elasticity;
- I_e = effective moment of inertia;
- L = support span;
- P = load required for deflection; and
- δ = deflection.

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