Performance of Natural-Fiber–Plastic Composites under Stress for Outdoor Applications: Effect of Moisture, Temperature, and Ultraviolet Light Exposure

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ABSTRACT: The effects of moisture, temperature, and ultraviolet (UV) light on performance of natural-fiber–plastic composites (NFPC) were assessed. We conducted short-term tests in the laboratory and long-term tests under natural exposure and measured changes in mechanical properties and color in samples of the composite. Chemical changes of the composite’s materials were measured by X-ray photoelectron spectroscopy to elucidate the mechanisms of chemical transformations on the material surface. Relative humidity highly affected the modulus of rupture (MOR) and the modulus of elasticity (MOE), and had a greater effect than temperature and UV exposure on performance of the composite. The lightness of the composite was increased by the UV effect in the short- and the long-term tests. The X-ray photoelectron spectroscopy (XPS) analysis suggested that the composite was protected by the UV absorber.© 2005 Wiley Periodicals, Inc. J Appl Polym Sci 99: 2570 –2577, 2006

Key words: mechanical properties; natural-fiber–plastic composite; XPS; UV irradiation; photo- and thermo-oxidation

INTRODUCTION

Durability of natural fiber composites is uncertain and performance is still under investigation. Outdoor durability of wood and nonwood–plastic composites might be influenced by thermal stability, moisture uptake, fungal resistance, and ultraviolet (UV) stability. To overcome the detrimental effects of these agents, the industry enhances properties to prolong the lifespan of the natural fiber composites, using coupling agents, fungicides, and UV absorbers during the manufacturing process.

Durability is measured through performance and appearance. When physical and mechanical properties, as well as esthetic properties, deteriorate, the service life of the product shortens. Some of the most common damaging effects in the composites are the following: color change and fading, surface erosion, loss of mechanical properties, and weight loss. Results of recent studies show that solar irradiance (UV component of the sunlight), relative humidity, temperature, and/or other environmental variables are the causal agents of this deterioration in weathered natural–fiber–plastic composites. Each component of a natural-fiber–plastic composite absorbs UV energy.

Free radical photo-oxidation and thermo-oxidation reactions occur as a result of sunlight and high temperatures, respectively. This is relevant because the polymer chain scissions originating from these reactions lead to deterioration of the mechanical properties in the composites, reducing their durability.

Degradation of polymers through photo-oxidation has been studied in different materials. In the pulp and paper industry, yellowing and bleaching of newsprint and mechanical pulps has been extensively studied using light at different wavelengths. Lignocellulosic materials such as wood and paper readily undergo light-induced yellowing. Lignin contains numerous chromophores that efficiently absorb UV radiation. As much as 80–95% of the absorption coefficient of wood can be ascribed to the lignin fraction. Forsskahl and Tylli summarize the results of several studies, showing that the maximum yellowing effect on wood fibers was found at 310–328 nm and at 310–320 nm. Results of their study on chemimechanical pulps confirmed these values.

In the case of polyolefins, Xingzhou and Zhenfeng et al. showed that the most effective spectral regions for causing PE and PP photo-oxidation are in the ranges 330–360 nm and 335–360 nm, respectively.

Stark and Matuana determined the effectiveness of various photo-stabilizers and their interactions on the weathering of wood flour filled HDPE composites. Under the conditions of their experiment they found...
that the UV absorbers are more effective photo-stabilizers on the composites than hindered amine light stabilizers.

Li Studies the environmental degradation of a wood–high density polyethylene composite in Australia. He stated that daily temperatures and sunshine times are believed to be less crucial than rainfall and UV intensity. Apparently the reductions in the modulus of rupture (MOR) and the energy absorption were about 50%. The stiffness (MOE) declined by about 70% after outdoor weathering for 205 days. The weathering also degraded about 50% of the tensile properties of the composite, including strength, elastic modulus, and energy absorption.

Falk et al. found a 24% reduction in the modulus of elasticity (MOE) and 19% reduction in the MOR for a wood–plastic composite (HDPE/wood) protected with a red pigment and containing 50% wood flour after 1500 h of exposure to UV light. Results of research conducted by Stark and Matuana on specimens sized to fit in the drum of a xenon arc-type light-exposure apparatus operating according to ASTM D2565 illustrate significant changes in flexural strength (MOR) and stiffness (MOE) of HDPE/wood composites after 2000 h of exposure.

Stark and Matuana have also used X-ray photoelectron spectroscopy (XPS) to elucidate oxidation of wood–plastic composites after accelerated weathering. They found that, even though both neat HDPE and the wood–fiber composite experienced surface oxidation after exposure, the surface oxidation of the latter occurred to a greater extent than that of the former. They state that these results suggest a more weather-related damage to the HDPE matrix with the addition of wood fiber.

Most of these studies are related to performance and durability of wood–plastic composites based on laboratory studies only. In this study, we assess the effect of moisture, temperature, and UV light in mechanical properties and color of the composite in short-term accelerated tests. We also look at performance of the composites in weathering long-term tests using commercial dimension samples. They have not used commercial dimension material in their studies.

The elucidation of mechanisms governing changes in composite materials during weathering is critical to assess durability and also to improve the current methods of protection and preservation of the composites.

**MATERIALS AND METHODS**

The tested materials in the short-term as well as in the long-term tests were commercial extruded profiles of NFPC protected with UV absorbers used in outdoor deck railings. The composition of materials was 40% HDPE and ~60% natural flour. The specimens were hollow pieces with a 41 × 41 mm² cross section, 5 mm thickness, and 75 cm length. The rail components were obtained directly from the manufacturing company.

Previous research in our laboratory found that the stress–strain curve obtained for the extruded profiles in the bending test was elastic up to 22% of the flexural strength or 0.4% strain. The maximum stress calculated at this point was close to 4 MPa. To calculate the uniformly distributed load at this maximum stress, we used the following formula:

\[ W = \frac{\sigma_{\text{max}} I}{L^2 c} \]

where \( W \) is the uniform distributed load, \( \sigma_{\text{max}} \) the stress at maximum distance \( (c_{\text{max}}) \) from the neutral axis (4 MPa), \( I \) the moment of inertia of the beam cross section, \( L \) the span (16" = 406.4 mm), and \( c \) is the distance from the neutral axis. From the above, it was determined that \( W = 764 \text{ lb/ft}^2 (4106 \text{ kg/m}^2) \).

A uniform distributed load of 4300 kg/m² (800 lb/ft²) was placed on the samples to accelerate the short-term tests. This load was placed on the central 12 in. (30.5 cm) of the specimens with a total span of 16 in. (40.6 cm) (Fig. 1).

To compare effects of exposure to two different UV wavelengths, coupons of 20 × 160 × 3 mm³ were obtained from the extruded profiles. Specimens of 1 × 1 cm² were cut from the exposed samples to be used for the XPS surface analysis.

**Short-term accelerated tests—commercial dimension samples**

Four tests were carried out under controlled conditions of temperature and relative humidity inside the environmental chamber. The profiles of the composite (composite 1) were exposed to several treatments,
combining different levels of relative humidity, temperature, and UV light as follows: relative humidity 34 and 93%, temperature 23 and 40°C, and UV 0.85 mW/cm² and no UV exposure.

The specimens were exposed to UV radiation using a fluorescent lamp with a nominal wavelength of 365 nm (peak emission). This wavelength provides a similar spectrum to that of sunlight in the short and middle UV wavelength regions simulating daylight very closely. The UV wavelengths to which the composite’s polymers are most susceptible to degradation are well covered by this lamp. The UV intensity plays a significant role as well, since it reflects sunlight conditions in different places at different hours. A typical sunny summer day in Toronto at noon has peak UV intensity close to 1.9 mW/cm² at a 365 nm wavelength. However, this intensity is limited to a few hours during the day and decreases significantly with the presence of clouds and humidity. The UV intensity applied to the specimens was appropriate to obtain realistic correlations with outdoors exposures.

Moisture, in combination with radiation, is often a key contributor to material weathering. Moisture contributes physically to degradation through mechanical forces imposed when moisture is absorbed or desorbed. The frequency and duration of the exposure to moisture is often a critical parameter.16

**Long-term tests—commercial dimension samples**

Samples of the same composite (composite 1) were exposed to the effects of variable weather factors such as precipitation, snow, sunlight, temperature, moisture, wind, etc. on the roof of the Forestry Building at the University of Toronto as depicted in Figure 2. Mechanical properties of unloaded specimens of composite 1, which had been exposed since October 2002, were measured in this test after 18 months of exposure.

Similar extruded profiles of a natural-fiber–plastic composite made with the same materials and the same fiber content (composite 2) had been formerly exposed to the weathering conditions of Toronto since March 2002. Changes in mechanical properties and color were also measured after 12, 18, 24, and 30 months and compared with the changes of the composites tested in the short-term tests for 2000 h.

**Mechanisms of degradation of the composite’s materials**

Susceptibility to different wavelengths and XPS surface analysis were used to determine the mechanisms of degradation of the composite’s components. Coup[ons](20 \times 160 \times 3 \text{ mm}^3) of the composite, the wood (aspen) and the plastic, were used for the materials degradation tests. The coupons were exposed to two different wavelengths under ambient conditions (34% RH and 23°C). The purpose was to detect the major degradation effect of the two wavelengths on each of the materials, as well as the effect on the composite, and correlate their behavior. Two UV fluorescent lamps were set up to illuminate the samples for two periods: 2000 and 4000 h. The peak emissions of the UV light were 365 (UVA) and 313 nm of wavelength (UVB) (Fig. 3).

Tensile strength tests and XPS analysis were carried out after 2000 h of exposure. The tensile strength was also measured for the specimens exposed for 4000 h. XPS surface analysis technique was used to detect the chemical reactions taking place at the surface of the composite and the composite’s materials. From the initial carbon surface spectra, the deconvolution process shows the proportion of different types of carbon bondings at distinct binding energies (Table I).17–19 The oxygen/carbon ratio was calculated for the three materials under both wavelengths after 2000 h of UV exposure.

**Mechanical properties**

Flexural strength (MOR) and flexural stiffness (MOE) are critical for the composite’s performance when the product is in service. Bending tests were carried out...
after 14 and 18 months of outdoors exposure for composite 1, and after 12, 18, 24, and 30 months for composite 2 to measure the MOR and the MOE. The specimens in all cases were conditioned to 50% relative humidity and 23°C for 72 h before testing. The same properties were measured for the indoors samples of composite 1 after 2000 h of UV exposure for all treatments. The four-point loading method was applied to the specimens according to ASTM D 6109. The specimens were deflected until rupture, using a Universal Testing Machine.

The tensile test was conducted according to standard ASTM D-638 – 02a. In the case of wood, the tensile strength parallel to grain was measured in clear specimens (20 × 160 × 3 mm³) of Aspen.

Color

Measurements of color of exposed and unexposed specimens of composites 1 and 2 were done with a spectrophotometer MINOLTA, using the CIELAB color space method described in the MINOLTA booklet, Precise Color Communication.

RESULTS AND DISCUSSION

Effects of moisture, temperature, and UV light in the short-term accelerated tests

Figure 4 illustrates the MOR of the composites under the different combinations of temperature and relative humidity. The higher the relative humidity, the lower the MOR for both unexposed and UV-exposed specimens. Temperature had the second greatest effect on MOR after moisture. The MOR was not highly affected by the UV irradiation.

Figure 5 shows the change of MOE under different conditions of temperature and relative humidity for unexposed and UV-exposed samples of the composite. The effect of the relative humidity on the MOE was similar to that on the MOR. At lower levels of relative humidity, the increase in temperature reduced the MOE; whereas at higher RH levels, the higher temperature exposure increased the MOE. It is possible that the high temperature at high levels of moisture promotes embrittlement of the plastic component at the surface of the composite, making it less elastic.

As seen in Table II, the MOR and the MOE values decrease when the relative humidity is increased from 34 to 93% under the same temperature and UV exposure conditions. The increase of relative humidity had a major effect on the degradation of the composite under the conditions of this experiment.

At low relative humidity, the increase of temperature reduces the MOE of the UV-exposed and unexposed samples.

<table>
<thead>
<tr>
<th>RH (%)</th>
<th>MOR (MPa)</th>
<th>MOE (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>No UV exposure (T = 23°C)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>34</td>
<td>17.4 (0.15)</td>
<td>2906 (89.2)</td>
</tr>
<tr>
<td>93</td>
<td>13.6 (0.45)</td>
<td>1913 (130.7)</td>
</tr>
<tr>
<td>UV exposure (T = 23°C)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>34</td>
<td>17.0 (0.27)</td>
<td>2783 (68.69)</td>
</tr>
<tr>
<td>93</td>
<td>14.5 (0.66)</td>
<td>2227 (24.8)</td>
</tr>
<tr>
<td>No UV exposure (T = 40°C)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>34</td>
<td>14.0 (0.24)</td>
<td>2539 (59.2)</td>
</tr>
<tr>
<td>93</td>
<td>13.0 (0.98)</td>
<td>2459 (22.1)</td>
</tr>
<tr>
<td>UV exposure (T = 40°C)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>34</td>
<td>16.8 (0.50)</td>
<td>2655 (69.2)</td>
</tr>
<tr>
<td>93</td>
<td>11.8 (1.56)</td>
<td>2312 (40.3)</td>
</tr>
</tbody>
</table>

Values in parentheses indicate standard deviations.
posed specimens of the composite, whereas at high relative humidity, the MOE is increased as explained above.

Finally, the MOE and the MOR were susceptible to changes in UV exposure under the same conditions of temperature and relative humidity. The effect of UV light on the MOE was significant for those cases in which the relative humidity was 93%.

Performance of the composites in the long-term tests

Specimens of the same composite used in the laboratory and exposed to the climatic conditions of Toronto experienced decrease of 8.7% in flexural strength and 17.6% in stiffness after 18 months, showing statistically significant differences in both cases (Fig. 6). As illustrated in Figure 7, the MOR and MOE of extruded profiles of fiber–plastic composite 2, with similar composition to composite 1 also decreased after 2.5 years of weathering. These two properties are consistently affected by moisture, temperature, UV light, snow, ice, and other weathering factors, and show a continuous linear trend to decrease after this period of time. This trend is not necessarily linear in nature and might be verified after longer periods of outdoors exposure.

In some cases, changes of these properties in the lab are similar to changes of these properties outdoors and might reflect performance in the long term. The MOE decreased by 17.6% for composite 1 after 18 months of exposure. From Figure 8, the MOE of the same composite in the lab declined by 23.0% after 2000 h of exposure when the relative humidity increased from 34 to 93% and the UV irradiation went up from zero to 0.85 mW/cm2 under the same temperature. The change of the MOE in the long-term is similar to the change of the MOE in the short-term test. Longer periods of outdoors exposure are required for the composite to show major changes in stiffness that could be reflected by the results of the short-term tests.

The Acceptance Criteria AC174 from the International Conference of Building Officials (ICBO) Evaluation Service, Inc.23 were used as a reference to assess durability of the composites.

Flexural strength (MOR) and stiffness (MOE) of the composites show similar trends in performance when

![Figure 6](https://example.com/fig6.png)  
**Figure 6** Change of the MOR and the MOE of a fiber–plastic composite (composite 1) exposed to the weathering conditions of Toronto for 18 months. (a) MOR (change after 18 months, −8.7%); (b) MOE (change after 2000 h, −17.6%).

![Figure 7](https://example.com/fig7.png)  
**Figure 7** Change of the MOR and the MOE of a fiber–plastic composite (composite 2) exposed to the weathering conditions of Toronto for 2.5 years. (a) MOR (change after 30 months, −10.1%); (b) MOE (change after 2000 h, −9.0%).
these are exposed to short- and long-term tests under the conditions of this experiment.

Change of color and fading is illustrated in the pictures of Figure 9. The color of the composite 1 was measured after exposure in the short- and the long-term tests using the CIELAB color space method and the following values were obtained: The L value, which stands for lightness, had a similar increase under the two conditions. The outdoors weathered samples turned bluish, whereas the short-term exposed specimens had a slight change of color. Longer times of exposure under the short-term conditions might better reflect the observed changes in color of the outdoors weathered samples.

Mechanisms of degradation of the composite’s materials

Mechanical properties

The values of tensile strength of the specimens of different materials measured after 2000 and 4000 h of UV exposure to two different wavelengths are depicted in Table III.

<table>
<thead>
<tr>
<th></th>
<th>Short-term (2000 hr)</th>
<th>Long-term (one year)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>L  a  b</td>
<td>L  a  b</td>
</tr>
<tr>
<td>Unexposed samples</td>
<td>60.8  8.0  16.7</td>
<td>61.7  5.8  16.9</td>
</tr>
<tr>
<td>UV exposed samples</td>
<td>66.1  5.2  16.7</td>
<td>69.6  0.4  2.4</td>
</tr>
</tbody>
</table>

The tensile strength of the natural fiber composite did not experience a large change with either of the wavelengths after 4000 h of UV exposure.

The high density polyethylene experienced a decrease of almost 23% of the tensile strength after 4000 h of UVB exposure, turning more brittle and less elastic under both wavelengths.

The tensile strength of wood highly increased with both wavelengths after 4000 h (Table III). Possible crosslinking reactions promoted by the UV radiation on the lignin component might be a factor playing a role in this change. Considering the high proportion of area exposed to the UV effect (42%) and the high area/volume ratio of the specimens, this factor might affect this property. Additional experimentation would be required for the verification of this effect.

XPS surface analysis

Figure 10 shows the oxygen/carbon ratio of the plastic, wood, and natural fiber composite before and after 2000 h of UV radiation under both wavelengths.

The carbon’s deconvolution values for the three materials before and after UV exposure are illustrated in Table IV.

Although the two component materials of the natural fiber composite have potential for oxidation, its oxygen/carbon ratio did not experience any increase with the UV light exposure (Fig. 10).

According to the results depicted in Table IV, the increase in carbon–carbon and carbon–hydrogen bonds, and the decrease in signal intensities of carbon–oxygen bonds and oxygen–carbon–oxygen bonds suggest that the surface of the natural fiber composite was not oxidized.

Because the dominant material at the surface of the composite is the HDPE, the high resolution carbon spectrum of the composite is similar to that of this material. Since both the components appeared to be oxidized by UV exposure, while the combination did not, the UV absorbers appear to have been effective in
preventing significant oxidation of both components. It is also possible that the effect of UV on HDPE was negligible and that the surface layer of polymer protected the wood. The small increase in the oxygen/carbon ratio of the HDPE after UVA and UVB exposure suggests a slight oxidation of this material.

Similar results to Hon’s⁶ were obtained for the surface analysis of wood. The oxygen/carbon ratio increased after the specimens were exposed to UVA and UVB. Obvious oxidation of this lignocellulosic material is confirmed by the decrease in carbon–carbon and/or carbon–hydrogen bonds, as well as the increase of oxygen–carbon bonds and carbonyl and carboxyl groups observed in Table IV.

With longer times of UV exposure, a more advanced state of degradation of the composite would be expected. The interaction between the UV light and the moisture would facilitate water uptake, internal stresses, and erosion. The oxidation of the wood component would play a major role in changes to color and mechanical properties of the natural fiber composite.

The natural fiber composite had an increase in lightness and no major change of color. The exposed specimens turned whiter than the unexposed specimens. This behavior is similar to that of the high density polyethylene, which became clearer after exposure to both UV wavelengths.

The UVA- and UVB-exposed samples of wood were oxidized, turning darker than the unexposed ones.

Even though the chemical changes experienced by the plastic after UV exposure were not as high as those by the wood, its pattern of color change was more similar to that of the composite. It is possible that the composite’s lightness is very susceptible to small changes in the HDPE chemical composition. Because the natural fibers of the composite are encapsulated by the plastic, the latter is the dominant material at the surface. Chemical changes should happen at the surface of the composite when it is exposed to UV light. At the beginning of the degradation process, the plastic should be the most affected material.

### CONCLUSIONS AND RECOMMENDATIONS

Under the short-term test conditions of this study, exposure to high relative humidity highly reduced the MOR and the MOE, and had a greater effect than temperature and UV exposure on performance of the composite.

### TABLE III

<table>
<thead>
<tr>
<th>Material</th>
<th>Wavelength</th>
<th>0 h Tensile strength (MPa)</th>
<th>2000 h Tensile strength (MPa)</th>
<th>4000 h Tensile strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE</td>
<td>UVA</td>
<td>22.57 (0.72)</td>
<td>23.31 (0.25)</td>
<td>15.63 (0.74)</td>
</tr>
<tr>
<td></td>
<td>UVB</td>
<td>22.57 (0.72)</td>
<td>17.49 (0.58)</td>
<td>15.80 (0.32)</td>
</tr>
<tr>
<td>Wood</td>
<td>UVA</td>
<td>42.30 (12.64)</td>
<td>61.77 (14.76)</td>
<td>76.73 (0.57)</td>
</tr>
<tr>
<td></td>
<td>UVB</td>
<td>42.30 (12.64)</td>
<td>49.47 (5.27)</td>
<td>84.30 (16.96)</td>
</tr>
<tr>
<td>NFC</td>
<td>UVA</td>
<td>13.42 (0.37)</td>
<td>13.45 (0.40)</td>
<td>14.29 (0.14)</td>
</tr>
<tr>
<td></td>
<td>UVB</td>
<td>13.42 (0.37)</td>
<td>13.52 (0.36)</td>
<td>13.67 (0.13)</td>
</tr>
</tbody>
</table>

Values in parentheses indicate standard deviations.

### TABLE IV

<table>
<thead>
<tr>
<th>Material</th>
<th>Conditions</th>
<th>C1 (%)</th>
<th>C2 (%)</th>
<th>C3 (%)</th>
<th>C4 (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NFC</td>
<td>No UV</td>
<td>86.0</td>
<td>8.6</td>
<td>3.8</td>
<td>1.6</td>
</tr>
<tr>
<td></td>
<td>UVA</td>
<td>87.6</td>
<td>8.0</td>
<td>2.8</td>
<td>1.6</td>
</tr>
<tr>
<td></td>
<td>UVB</td>
<td>87.8</td>
<td>8.0</td>
<td>2.8</td>
<td>1.4</td>
</tr>
<tr>
<td>HDPE</td>
<td>No UV</td>
<td>87.8</td>
<td>8.9</td>
<td>2.7</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>UVA</td>
<td>87.8</td>
<td>7.7</td>
<td>3.6</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>UVB</td>
<td>88.7</td>
<td>7.7</td>
<td>2.4</td>
<td>1.2</td>
</tr>
<tr>
<td>Wood</td>
<td>No UV</td>
<td>52.7</td>
<td>30.3</td>
<td>11.4</td>
<td>5.5</td>
</tr>
<tr>
<td></td>
<td>UVA</td>
<td>38.9</td>
<td>35.8</td>
<td>14.3</td>
<td>11.0</td>
</tr>
<tr>
<td></td>
<td>UVB</td>
<td>32.2</td>
<td>40.6</td>
<td>15.5</td>
<td>11.7</td>
</tr>
</tbody>
</table>

Figure 10  Oxygen/carbon ratio at the surface of HDPE, wood, and composite samples before and after 2000 h of UV exposure to short and long wavelengths.
The change of lightness of the composite in the long term is well reproduced by the change of lightness of the UV-exposed composites in the short term.

Flexural strength (MOR) and stiffness (MOE) of the composites show similar trends in performance when these are exposed to short- and long-term tests under the conditions of this experiment.

From the XPS surface analysis, the UV-exposed natural-fiber–plastic composite was apparently protected by the antioxidant, even though the plastic (HDPE) and the lignocellulosic material (wood) showed potential for oxidation.

Because the change of lightness in the composite was more similar to that in the plastic than in the wood, it is possible that the composite’s lightness is more susceptible to small changes in the HDPE’s chemical composition.

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