Long term durability of wood-plastic composites made with chemically modified wood

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Keywords: acetylation, coupling agent, moisture sorption, fungal decay, extrusion, field exposure

ABSTRACT

Wood-plastic composites (WPCs) have slower moisture sorption than solid wood, but over time moisture can impact the strength, stiffness, and decay of the composite. These changes will become increasingly important if WPCs are used in more challenging environments such as in ground-contact applications. There are several options for mitigating the moisture sorption of WPCs. A coupling agent can be added to potentially improve the wetting and dispersion of the wood flour and improve its adhesion with the plastic matrix. In previous investigations, the use of a coupling agent has been shown to further slow, though not stop, moisture intrusion in WPCs. Alternatively, the wood flour can be chemically modified to make the wood itself more hydrophobic and, therefore, reduce the driving force for moisture intrusion rather than just slow it.

While the effects of such approaches to moisture reduction have been investigated somewhat in the laboratory setting, field exposure is ultimately needed to verify the overall long-term durability of such WPCs, which includes the potentially synergistic effects of exposure to sunlight, moisture, wind, temperature fluctuations, freeze/thaw, wet/dry cycling, termites, mold and decay fungi. WPCs containing either a coupling agent or chemically modified wood flour were fabricated. These modified WPCs and a control were installed both above-ground and in-ground in Saucier, Mississippi and Madison, Wisconsin and observations (e.g., appearance, thickness swell, and fungal attack) were recorded over a 12 year time period. Results will be discussed, as well as correlation between laboratory and field measurements.

INTRODUCTION

The global wood plastic composite (WPC) market is expected to grow at about 13.2% over the next decade to reach approximately $9.7 billion by 2025 (Cole 2017). In this very dynamic market, there is an expectation by consumers for improved performance characteristics, such as high durability, low maintenance requirements, and enhanced appearance. However, some of these improvements may still not meet consumer expectations. Additionally, WPCs may be increasingly used in more challenging environments such as in ground contact applications.

It was first thought that mixing plastic and wood together would result in plastic encapsulation of wood, which would prevent moisture sorption and fungal decay. However, evidence of fungal decay and discoloration on WPC decking in service was found (Morris and Cooper 1998). Since then, considerable research has investigated the moisture and biological durability of WPCs. Although WPCs have slower moisture sorption than wood (Rowell et al. 2002, Wang and Morrell 2004), water affects the mechanical properties and ultimately promotes biological decay of the material (Clemons and Ibach 2002, 2004; Ibach and Clemons 2007a; Ibach et al. 2017; Shirp and Wolcott 2005). Shirp et al. (2007) reviewed the literature on biological degradation of WPCs and strategies for improving resistance – from the addition of toxic chemicals to chemically modifying the wood.

However, when used in outdoor conditions, WPCs are exposed to not only biological and water degradation but also UV, thermal, chemical, and mechanical degradations. To begin understanding the interaction of some of these combined exposures, new laboratory testing protocols have been investigated. For example, Ibach et al. 2004 investigated combined moisture and ultraviolet (UV) exposures in laboratory fungal decay tests.

Additionally, it is necessary to evaluate different strategies of mitigating the negative effects related to the different types of exposure found in outdoor use. One such candidate is the use of coupling agents, which are incorporated into WPCs to improve the compatibility and adhesion between the polar wood fibers and non-polar polymers (Bledzki et al. 1998, Lu et al. 2000, Bledzki and Omar 2003). The most common coupling agents are maleic anhydride grafted to polypropylene (MAPP) or polyethylene (MAPE.) Addition of such coupling agents have been shown to improve polymer–wood adhesion and dispersion of wood particles (Ichazo et al. 2001, Kazayawoko et al. 1999) as well as improve mechanical properties such as flexural moduli, yield stress, and some types of impact behavior (Hristov et al. 2004, Stark and Rowlands 2003). Maleated polyolefins have also shown an ability to reduce water absorption in WPCs (Ichazo et al. 2001), which can affect their durability.

Chemical modification of wood cell walls can also prevent or slow degradation reactions (Rowell 2013). Acetylation is one of the most studied wood modification chemistries. Reaction of acetic anhydride with wood results in esterification of accessible hydroxyl groups in the cell wall with acetyl groups and reduced moisture sorption. Because moisture plays a key role in fungal decay of WPCs, it is prudent to evaluate WPCs made with acetylated wood fiber and research has been conducted showing improvements in a variety of laboratory durability experiments (Glasser et al. 1999, Ibach and Clemons 2002, Khalil et al. 2002, Glasser 2004, Segerholm et al. 2005, 2012, 2013). For example, acetylated wood-fiber–plastic composites that were compression molded had better mechanical properties and water resistance than unmodified WPC controls (Glasser et al. 1999, Khalil et al. 2002). Acetylated wood-fiber–plastic composites that were injection molded and extruded had decreased moisture content and were highly resistant to brown-rot decay compared with unmodified WPC controls (Segerholm et al. 2005). The acetylated fiber in Segerholm's research was produced by grinding previously acetylated solid wood. Ibach and Clemons found that compression molded composites of acetylated ground wood fiber and high-density polyethylene (HDPE) decreased the equilibrium moisture content (EMC) and fungal decay of the WPC compared with composites made with unmodified wood or butylene- or propylene-oxide modified wood (Ibach and Clemons 2002).

Both the addition of coupling agents and acetylation of the wood component appear to offer potential for reducing moisture sorption and improving biological durability. In previous work directly comparing the two approaches, we found that acetylating the wood component in WPCs reduced moisture sorption and related performance losses such as decay and flexural property loss in laboratory tests (Ibach, Clemons et al. 2007b). Reduction in moisture sorption and decay with addition of coupling agent were significant but not as large as with acetylation. Reductions in mechanical properties during soil block tests appeared to be primarily due to moisture sorption and not fungal attack. The overall WPC moisture content exceeding about 15% led to significant weight losses. Also, the type of processing method used to prepare the composites influenced the moisture sorption and fungal resistance.

However, long-term field tests are also needed to evaluate such strategies for multiple types of exposures during outdoor use and would provide a useful comparison with the laboratory tests. Therefore, the objective of this portion of the investigation was to compare the strategies of: 1) adding a coupling agent or 2) chemically modifying the wood flour in improving the moisture and biological resistance of WPCs during outdoor field exposure. Early field evaluation of these composites were previously presented but results were quite preliminary (Ibach, Clemons et al. 2007b). Here we present the field evaluations over a much longer time frame.

EXPERIMENTAL

Materials

The wood filler was ponderosa pine wood flour (AWF-4020), nominal 40 mesh (420 µm), from American Wood Fiber (Schofield, Wisconsin). The plastic material was high-density polyethylene (HDPE) from reprocessed milk bottles (Muehlstein and Co., Inc., Roswell, Georgia), with a melt flow index of approximately 0.7 g per 10 min. The flour was modified with acetic anhydride from Aldrich Chemical Company (Milwaukee, Wisconsin). The coupling agent was maleated polyethylene (MAPE), Polybond 3009, from Chemtura Corporation (Middlebury, Connecticut), which contains 1% maleic anhydride. The lubricant was TPW-113 from Struktol Company of America (Stow, Ohio). It is a complex blend of modified fatty acid ester.

Acetylation of wood flour

The flour was oven dried and then boiled in acetic anhydride in a 1-L glass reactor for 4 h. The treated flour was washed and oven dried, and its weight gain percentage (WPG) was calculated. Percentage acetyl content was determined using anion exchange high-performance liquid chromatography (HPLC) with a suppressed conductivity detector, following a previously described methodology (Ibach et al. 2000).

Profile Extrusion

Profile extrusion was performed on a co-rotating 32-mm twin-screw extruder (Davis Standard, Connecticut). All components were fed into the main feed throat. HDPE was melted and then blended with the wood flour and additives. The material was then forced through a 19 by 19-mm square die, cooled and cut into 46-cm long specimens for in-ground field testing and 23-cm long stakes for above-ground field testing. Addition of 6% lubricant helped prevent tearing of the material as it exited the die. Blends of three different compositions were extruded (Table 1).

Field Testing

The stakes were installed in the ground in Saucier, Mississippi on the Harrison Experimental Forest and Madison, Wisconsin at the Valley View site following AWPA Standard E7-01. Stakes were evaluated periodically for decay/moisture and termites for 7 years in Mississippi and for decay/moisture for 12 years in Wisconsin. Observations such as cracking, checking, and swelling were also recorded. Thickness measurements of the stakes were taken with a Mitutoyo digimatic caliper in three locations on the stake: below the ground line, at the ground line and above the ground line (2 readings each.)

Stakes were also installed 1 m above-ground horizontally on a CCA-treated wooden rack and evaluated by the following criteria: color change, mold/mildew, checking/cracking, and warping/twisting. Thickness measurements of the stakes were also taken (2 readings each.)

The Mississippi site contains a sandy loam soil with an annual rainfall of 1575 mm (62 in.) per year. The Wisconsin site contains a clay loam soil with 787 mm (31in) of rainfall per year. Specimens at the Wisconsin site were also subjected to freeze/thaw cycles.

RESULTS AND DISCUSSION

Acetylation of wood flour

Acetylation of the wood flour was straightforward, except for the calculation of weight gain. There was loss of fine flour particles during refluxing and removal of the acetic acid by-product by water washing. Therefore, the percentage acetyl content was determined using HPLC (Ibach et al. 2000). The average acetyl content of the acetylated pine flour was 22.65% ± 0.01 (average of 24 batches) compared with that of the unmodified pine flour, which was 1.95% ± 0.00 (average of 4 samples.)

Field testing

Wood-plastic composite field stakes of the same formulations were evaluated in-ground and above-ground in both Mississippi and Wisconsin.

In-Ground Stakes

To verify the fungal viability of the field plots untreated southern pine solid wood stakes were included and evaluated at each test site. In Wisconsin the untreated solid wood controls had 4 out of 10 stakes remaining after 7 years and all were decayed by 10 years. Comparatively, the solid wood controls in Mississippi had only 1 left after 2 years and all were decayed after 3 years exposure. This shows that decay fungi were present in the soil at each site and that the decay hazard was more severe in Mississippi than Wisconsin which was expected.

Subterranean termites are present in Mississippi, but not at the Wisconsin site. Figure 1 shows the termite rating of the WPC stakes over 7 years. Stakes were visually rated from 0-10, with 10 being sound with 1 to 2 nibbles permitted, and 0 being a failure with attack on greater than 75% of the cross section. No termite attack was found on any of the WPCs after 2 years of exposure, but nibbles were found on the unmodified control and MAPE WPC blends after 3 years and this attack continued over the next 4 years. After 7 years exposure the MAPE rating was 7.6±1.8 (where 1.8 represents one standard deviation) and the control was 8.6±0.9. The acetylated WPC stakes showed no termite attack after 7 years with a rating of 10.

In-ground stakes were visually rated from 0-10, with 10 having no decay and 0 being failure with decay on greater than 75% of the cross section. Figure 2 shows the average in-ground decay rating after 12 years exposure in Wisconsin. None of the stakes showed signs of decay after 6 years in the ground. Signs of decay were present during the next 6 years, with ratings of 7.8±0.7 and 8.4±0.9 after 12 years exposure for the control and MAPE WPC blends, respectively. The acetylated WPCs had no decay after 12 years.

Figure 3 shows the average in-ground decay rating after 7 years exposure in Mississippi. The control and MAPE showed signs of decay and after 7 years had ratings of 8.2±0.4

and 8.6±0.7, respectively. The acetylated showed no decay over 6 years and then a rating of 9.1±0.3 at 7 years.

Not surprisingly, the largest thickness increases were found below the ground line. Figure 4 shows the thickness change below ground after 12 years exposure for the in-ground stakes in Wisconsin. The highest thickness changes came at 6 years, 5.5% for MAPE, 5.3% for the control, and 0.7% for the acetylated stakes. In comparison, Figure 5 shows the Mississippi stakes had the greatest thickness change after the first year for the control (6.5%) and MAPE (6.4%) and then subsequent decreases in the next 6 years. The highest thickness change for the acetylated stakes was 2.2% after 6 years.

Changes in thicknesses are most likely a combination of year-to-year variability in climate and time of rating as well as permanent damage. For example, major events such as Hurricane Katrina during year 1 are likely a significant factor in the large thickness increases in the first year of the stakes in Mississippi. Never-the-less, it is clear that acetylating the wood component led to significant reductions in thickness swell. This is consistent with laboratory findings where large reductions in moisture content were found when the wood component was acetylated (Ibach et al. 2007b).

Above-Ground Stakes

Above-ground stakes were mounted on racks and visually rated (0-10) for each of 5 criteria: 1) cracking and checking, 2) color change, 3) decay, 4) mold and stain, and 5) termite attack (Mississippi only). Figures 6 shows the above ground ratings after 12 years exposure in Wisconsin, and Figure 7 shows the 7 year ratings in Mississippi. The stakes performed similarly at both sites until year 5 when lichen started to develop in Wisconsin. This bioaccumulation made it especially difficult to rate the stakes visually. Although the stakes were only rated for 7 years in Mississippi, they looked better than the ones from Wisconsin exposed over a similar timeframe. The acetylated stakes had higher ratings for checking/cracking, decay, and mold/stain than the control and MAPE stakes after 12 years exposure in Wisconsin.

Color change was significant in both Mississippi and Wisconsin. This is not surprising since colorant or light stabilizers were not added to the composites. All composites faded similarly for 7 years. After 11 years in Wisconsin, all stakes had ratings of 0. This again was in large part due to the samples being heavily covered in lichen and moss.

After 2 years, mold and stain growth were found on the composites at the Mississippi site only, which typically gets twice the rainfall as the Wisconsin site. The Wisconsin site saw mold growth after 4 years. Despite the potential for moisture reduction, acetylating the wood component did not appear to inhibit the formation of mold on the surfaces of the composites. No termite activity was found on the above ground stakes after 7 years in Mississippi.

The above-ground average thickness change after 12 years exposure in Wisconsin is shown in Figure 8. Little swelling was found until year 4 for the controls. The greatest thickness change was after 12 years (3.1% MAPE, 2.8% control, 1.2% acetylated.) The above-ground thickness change after 7 years exposure in Mississippi as shown in Figure 9. The greatest thickness change was after 6 or 7 years (3.4% MAPE, 2.6% MAPE, 1% acetylated.) The significant swelling after one year can be explained by Hurricane Katrina, which brought significant rainfall.

Observations on Field Testing of WPCs

Several methods of moisture reduction were evaluated by long term field tests. There were challenges in evaluating and interpreting WPC field data (both above-ground and in-ground) when half the material is plastic compared to solid wood standards. When solid wood decays, it gets soft on the outside of the stake and moves inward. The WPC does not get soft like solid wood, making it difficult to examine visually. Therefore, we are exploring other methods of evaluation with magnetic resonance imaging (MRI) to track moisture and internal voids analysis with computed tomography (CT) (Ibach et al. 2016; Sun, et al. 2016).

CONCLUSIONS

The findings reinforced the need for exposure sites in multiple locations that have different climatic conditions. There is a higher decay hazard in Mississippi, but moisture was still an issue in Wisconsin along with freeze/thaw cycles and a greater bioaccumulation on above-ground stakes. There is less precipitation in Wisconsin, but the lower temperatures may provide more favorable conditions for fungal growth on the above-ground stakes. As a result of these climatic differences, there were different initiation periods before in-ground decay occurred at each location (6 years Wisconsin vs. 1 year Mississippi). Although it takes a longer time for in-ground decay in Wisconsin, the above ground stakes showed more bioaccumulation than in Mississippi.

The acetylated WPCs had the highest performance ratings in both Wisconsin and Mississippi, i.e. the highest decay and termite ratings and least thickness change for both above-ground and in-ground exposures compared to the control and those containing MAPE. The MAPE slowed the thickness change above-ground in Mississippi compared to the control, but did not stop the effects of exposure below-ground. Decay results presented here were similar to laboratory fungal decay tests (not shown), both of which demonstrated that moisture reduction can improve WPC performance. Field observations and measurements help to improve both accelerated laboratory test methodology as well as the understanding of the long term durability of WPCs.

ACKNOWLEDGMENTS

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TABLES

Table 1. Composition of three extruded WPC blends.*

Modification	Wood content (%)	Wood form	HDPE content (%)	Lubricant content (%)	Coupling agent content (%)
Control	50	Flour	44	6	-
Acetylation	50	Flour	44	6	-
Coupling Agent	50	Flour	41	6	3

*HDPE=high density polyethylene, MAPE= maleated polyethylene. All percentages are weight percent.

FIGURES

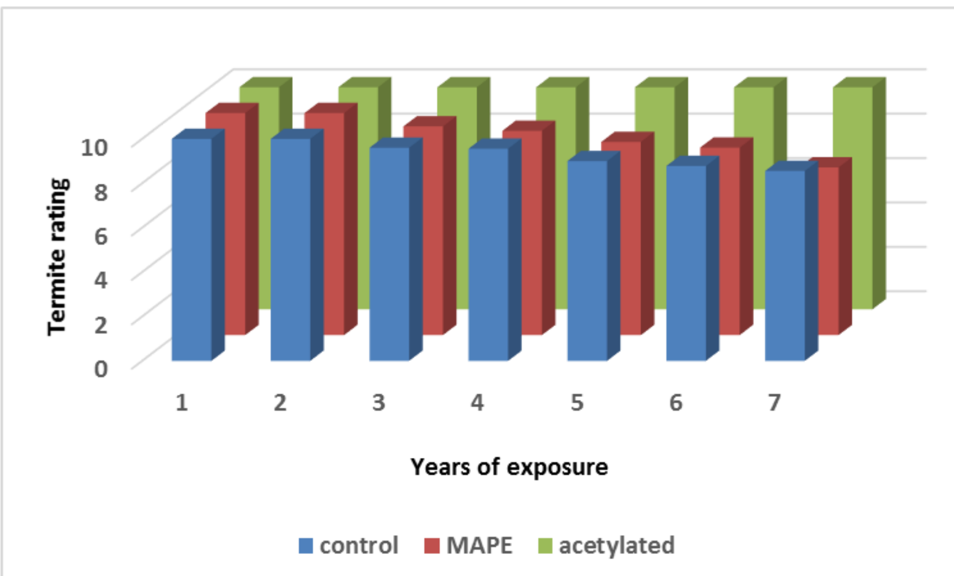

Figure 1. Termite rating after 7 years in-ground in Saucier, Mississippi. (Rating of 10 is sound with 1-2 nibbles permitted, and 0 is a failure with attack on greater than 75% of the cross section)

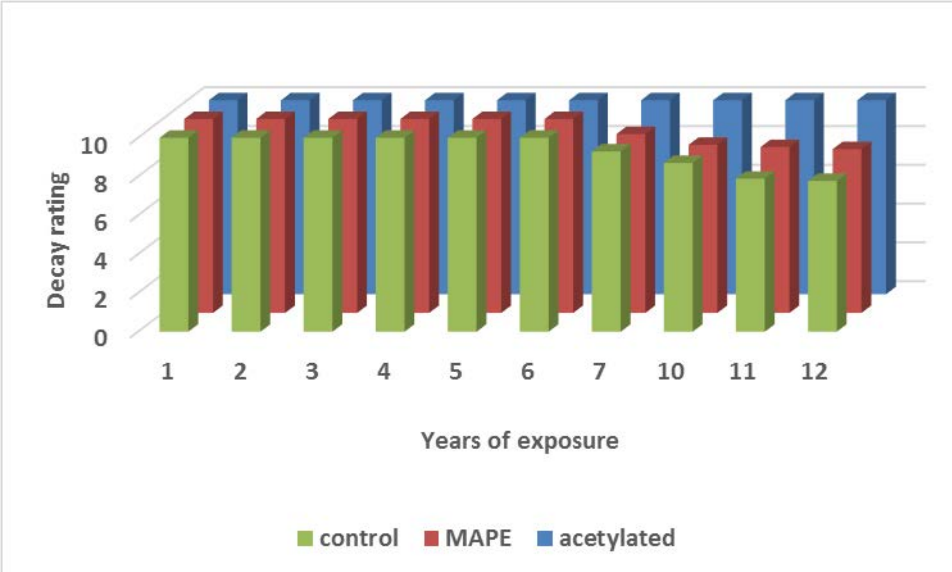

Figure 2. In-ground decay ratings after 12 years outdoor exposure at the Valley View site in Madison, Wisconsin. (Rating of 10 is sound, and 0 is a failure with decay on greater than 75% of the cross section)

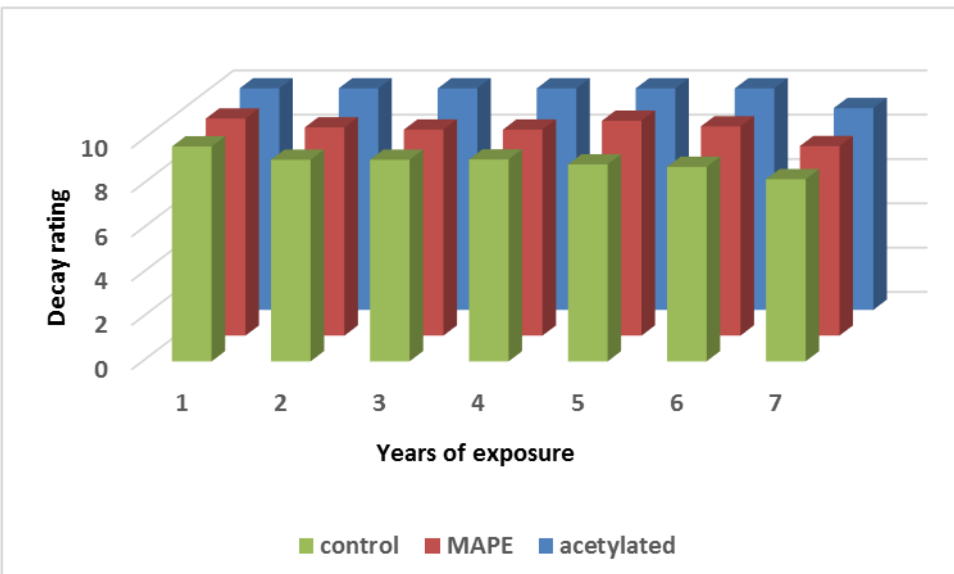

Figure 3. In-ground decay ratings after 7 years exposure at Saucier, Mississippi. (Rating of 10 is sound, and 0 is a failure with decay on greater than 75% of the cross section)

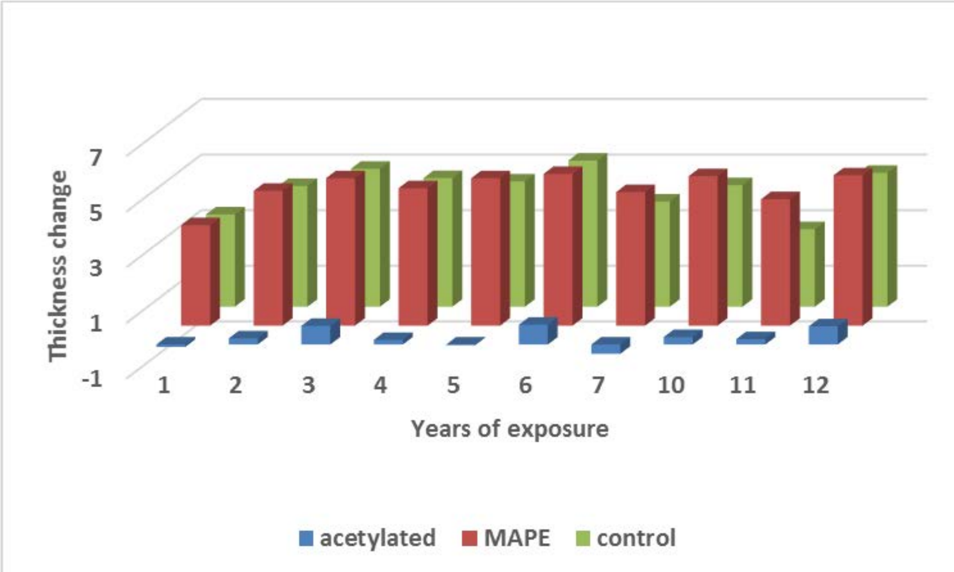

Figure 4. Average percent thickness change (below ground) after 12 years exposure for in-ground field stakes in Wisconsin

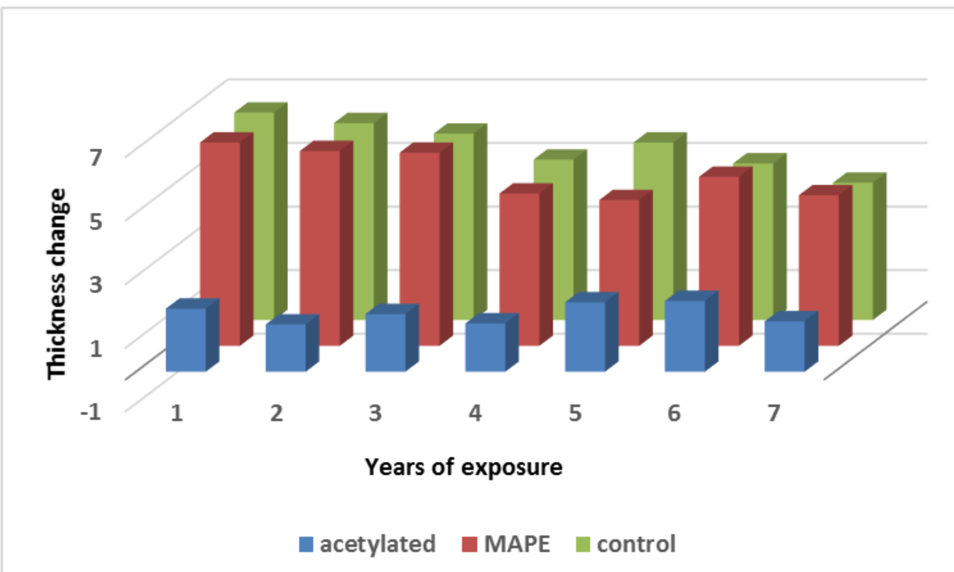

Figure 5. Average percent thickness change (below ground) after 7 years exposure for in-ground field stakes in Mississippi

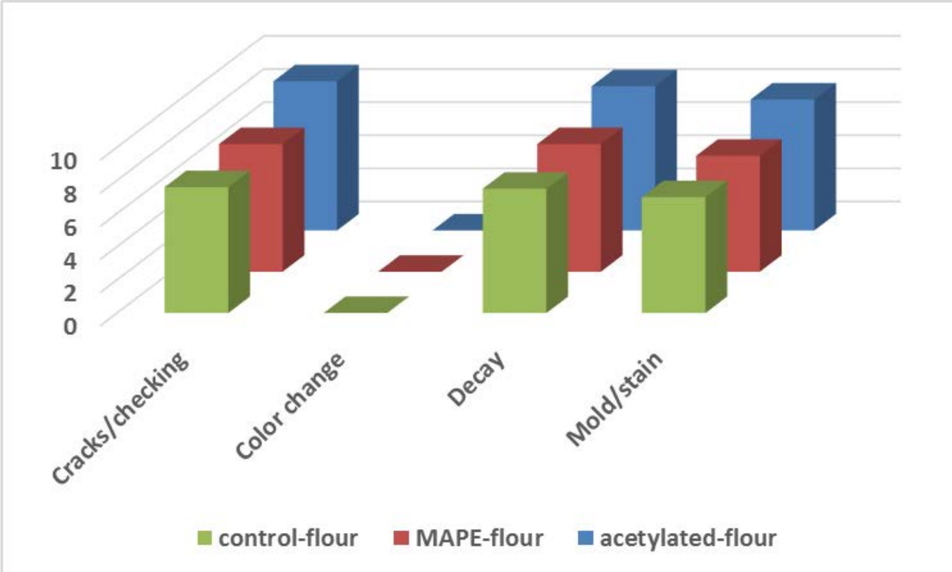

Figure 6. Above-ground ratings after 12 years outdoor exposure at the Valley View site in Madison, Wisconsin. (Rating of 10 is good, 0 is failure)

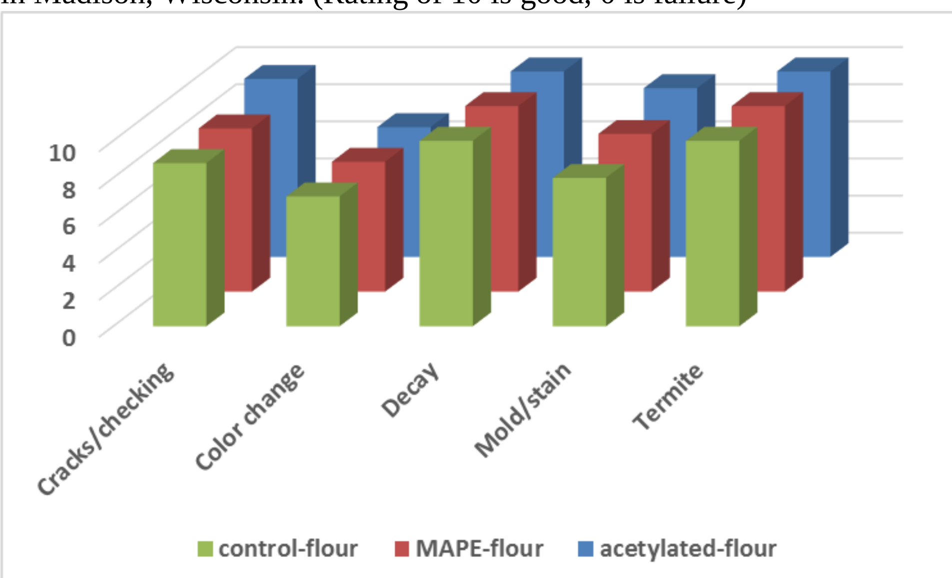

Figure 7. Above-ground ratings after 7 years outdoor exposure in Saucier, Mississippi. (Rating of 10 is good, 0 is failure)

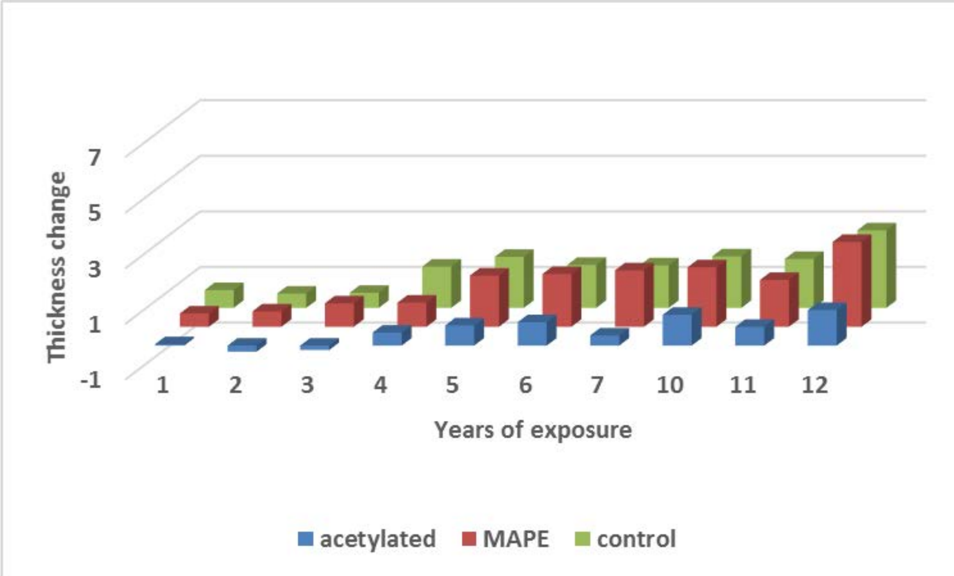

Figure 8. Average percent thickness change after 12 years exposure for above-ground stakes in Wisconsin

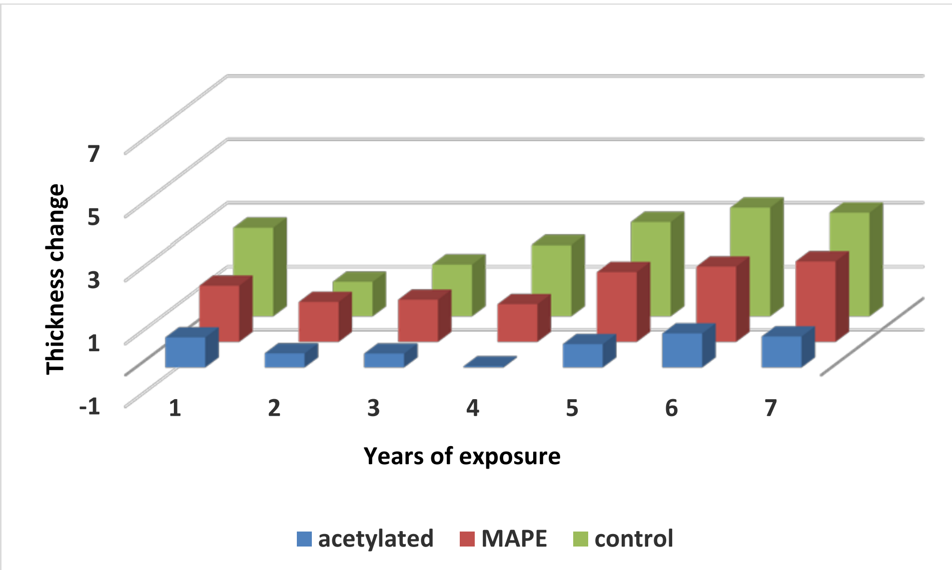

Figure 9. Average percent thickness change after 7 years exposure for above-ground stakes in Mississippi.

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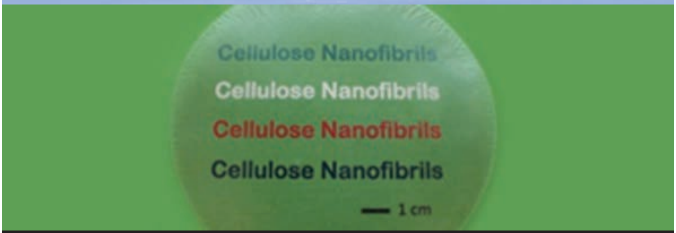

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