

Chapter 29

Biological Degradation of Wood–Plastic Composites (WPC) and Strategies for Improving the Resistance of WPC against Biological Decay

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Much of the research on wood-plastic composites (WPC) has focused on formulation development and processing while high biological durability of the material was assumed. The gap between assumption and knowledge in biodeterioration of WPC needs to be reduced. Although some information on the short-term resistance of WPC against biological degradation is available, long-term data are required in order to adequately evaluate the performance of these relatively new materials and to enable use of WPC for structural applications. The influence of abiotic factors on material performance also has to be considered. Thus, further market growth and differentiation, especially in Europe, may be achieved while simultaneously standardization issues are dealt with.

Introduction

Wood–plastic composites (WPC) or natural fiber reinforced thermoplastic composites represent a relatively new class of hybrid materials which have been gaining rapid market share in North America, primarily as a substitute for wood decking (1-3). Applications for WPC also include siding, fencing, windows, doors, car interior parts, furniture, sinks, concrete formwork, crates, boxes, flower pots, dowels and tool shafts. The use of WPC for load-bearing purposes is currently being investigated. In 2003, 400,000 t of WPC were produced in North America (4) and approximately 30,000 t in Europe (5). In contrast to North America, the European market is still undifferentiated but significant market growth is predicted.

As commercial WPC formulations contain a nutrient source in the form of wood or other natural fibers, it can be assumed that this material is susceptible to microbial decay. However, some WPC manufacturers initially believed that these materials were resistant to biodegradation due to encapsulation of the wood particles by the plastic. It is well known that polyolefins are highly resistant to biodegradation, especially without prior abiotic oxidation (6-8), due to their backbone being solely built of carbon atoms. Scheffer and Morrell (9) applied this knowledge in the wood preservation area by using polyethylene films for below-ground decay protection of small ponderosa pine sapwood stakes over a two-year period. It must be emphasized, however, that biodeterioration of synthetic polymers can be initiated due to decay of additives included in plastics.

Evidence for the presence of fungal decay and discoloration on WPC decking material in service was first presented by Morris and Cooper (10). Morris and Cooper (10) isolated and identified brown and white rot fungi as well as one blue stain fungus growing on a WPC boardwalk in Florida which had been in service for four years. This boardwalk consisted of 50% recycled wood fiber and 50% recycled plastic bags and film.

Biological degradation of WPC may include attack by decay fungi, moulds, algae, termites, and marine borers. In addition, abiotic influences such as moisture, UV-light and temperature affect WPC performance, but these are not considered in this review. This chapter will provide a state-of-the-art review on:

- Biological degradation of WPC due to wood-decaying, mould and staining fungi, insects, and marine borers;
- Current test methods for biodegradation of WPC;
- Antimicrobial treatments for WPC;
- Strategies for improvement of WPC durability;
- WPC standardization initiatives in North America and Europe.

Biological Deterioration of Wood-Plastic Composites: State of the Art

Influence of moisture on development of biological decay

The availability of moisture is a prerequisite for biological decay in a material. Although moisture uptake in WPC occurs relatively slowly, moisture levels in the outer 5 mm of commercial products were shown to be sufficient for fungal attack (11, 12). Once water has entered into the material, it will leave only very slowly since the plastic provides a barrier to gas evaporation and, as a result, biodegradation may occur.

Individual WPC processing methods have an influence on the moisture sorption. Clemons and Ibaach (13) used a two-week soaking or cyclic boiling-drying procedure to infuse moisture into composites made from high-density polyethylene (HDPE) filled with 50% wood flour and processed by extrusion, compression molding, and injection molding. It was determined that extruded composites absorbed the most moisture, compression-molded composites absorbed less, and injection-molded composites absorbed the least.

It is well known that moisture alone greatly reduces the strength and stiffness of wood (14), however, the reduction in strength properties due to fungal decay generally outweigh the effects of moisture in wood. It was recently shown that this does not apply to WPC (13, 14). See the section on “Mechanical Property Losses in WPC” for further information on this aspect.

Decay Fungi

Test methods

A multitude of methods are available to test for fungal degradation of wood and plastics, however, there is no laboratory standard currently available for testing the fungal durability of WPC. At present, in North America, the soil-block test for wood (16, 17) has been adopted for fungal durability tests of WPC in which weight loss serves as an indicator of decay. In Europe, the agar-block test, according to EN 113 (18), is commonly used in fungal decay testing. While ASTM D2017-05 (16) aims at determining the natural decay resistance of woods, EN 113 (18) is intended to determine the efficiency threshold of wood preservatives against wood-decay fungi. A new European standard, prCEN/TS 15083-1 (19), will be used to identify the natural durability of woods and is currently under review. In prCEN/TS 15083-1, the test fungus (*Poria placenta* for softwoods and *Coriolus versicolor* for hardwoods) is grown on sterile malt-agar in a Kolle flask until the medium is well covered with mycelium. then two sterile glass support rods are placed on the fungal mat, and two test or two

reference specimens are placed on the rods. Following 16 weeks of incubation, specimens are withdrawn from the vessels, any adhering mycelium is removed, and each specimen is weighed and dried to constant mass. Mass loss of each specimen is calculated in percent based on its initial dry mass. The soil block test is principally very similar to the agar-block test, however, soil and a so-called feeder strip, made from a non-durable wood species, are used as the substrate for growing the fungus. In ASTM D 2017-05 (16), *Gloeophyllum trabeum* and *Poria placenta* are obligatory test fungi for testing both softwoods and hardwoods, and *Coriolus versicolor* is a test fungus for testing hardwoods. When information on strength properties is sought as part of a soil or agar block test, the test method is usually changed in two ways: longer specimens are used and incubation vessels are placed horizontally instead of vertically (13, 15).

Schirp and Wolcott (15) compared the North American and European methods for WPC fungal durability testing. The agar-block test was modified such that no support rods were employed to accelerate moisture uptake by WPC specimens which had not been pretreated, only steam-sterilized in an autoclave. It was determined that modified agar- and soil-block tests are equally suited for determining weight loss in WPC, but that agar-block tests can be completed in a shorter time span.

Dynamic mechanical analysis (DMA) is a laboratory method which facilitates the investigation of the effects of wood decay fungi in WPC (20). Dynamic mechanical analysis can potentially provide valuable molecular and morphological information about a material in the solid state by subjecting it to dynamic loads over a broad range of temperature and frequency. A significant advantage of DMA as compared to commonly used static mechanical strength tests is that incubation times in fungal decay experiments can be drastically shortened due to the employment of very small specimens.

Field tests provide additional valuable information on the durability of a material. The objective is to determine the types and causes of failure not only from moisture, fungal and termite degradation but also from other environmental factors such as UV radiation, thermal cycles and freeze-thaw cycles. Verhey et al. (21) exposed field stakes at a site near Hilo, Hawaii, and samples were pulled for impact and flexural strength testing as well as fungal isolations at three-month intervals over the course of one year. Additional field tests with WPC, based on AWWA Standard E7 (22), were sponsored by the U.S. Navy and conducted by Michigan Technological University.

Scientists at the USDA Forest Products Laboratory are currently evaluating the field performance of extruded WPC (23). In-ground and above-ground specimens were installed in Madison, Wisconsin, and Saucier, Mississippi, in October 2000. Criteria for evaluating the in-ground stakes are decay, moisture, termites, cracking, checking and swelling. Decay and termite grades have been determined according to ASTM D 1758-02 (24). The above-ground stakes have been evaluated for color change, mold/mildew, warping/twisting, checking/cracking and flexural strength and stiffness.

Specific adjustments will have to be made to existing wood durability standards so that they can be appropriately used in WPC fungal decay testing. For example, weathering of WPC prior to fungal testing may be an effective way of accelerating the laboratory test by simulating outdoor conditions (25). In addition, suitable reference(s) for WPC in fungal decay testing will have to be determined, for example, a durable wood species, wood treated with a preservative, or a material based on 100% plastic. Finally, the fungal species that colonize wood and wood composites outdoors may not be the fungi that are predominant on WPC during long-term exposure. Hence, research is required to establish the predominant microorganisms occurring on WPC.

Weight losses in WPC

Mankowski and Morrell (26) conducted scanning electron microscopy to determine patterns of fungal attack in WPC following exposure in a soil block test. It was demonstrated that a formulation containing a 70/30 wood-HDPE mixture was most susceptible to fungal attack while two different 50/50 wood-HDPE composites displayed little or no degradation. Decay fungi were prevalent in the voids between wood and HDPE. Verhey et al. (27) and Pendleton et al. (28) also determined that an increase in plastic reduced weight losses during laboratory soil block tests. Maximum weight loss due to *Poria placenta* after 12 weeks was 8.5% when the formulation contained 70% wood filler (28).

Weight loss of an extruded WPC formulation with 70% wood filler and incubated with *T. versicolor* was twice as high as that of redwood in a modified agar-block test (6% versus 3%), however, only 1% weight loss was obtained when the formulation contained 49% wood filler (15). These results indicate that WPC can be designed to provide high fungal durability by controlling the material composition of the formulation. Generally, reported weight losses of WPC obtained in laboratory testing are below 10%.

Verhey and Laks (29) determined that weight loss in WPC due to fungal decay increased as the wood particle size increased, probably due to a more effective encapsulation of smaller wood particles by the polymer matrix. A similar protective effect due substrate encapsulation has been reported for wood-cement composites (30).

Weight losses in WPC are usually calculated based on the wood content of a formulation because of the aforementioned resistance of plastic matrices to biodeterioration. Only one publication regarding polyethylene degradation by white-rot fungi has been published (31), and there is no information available on PE-degradation by brown-rot fungi. The white-rot fungi *Phanerochaete cryosporium*, *T. versicolor* and one unidentified fungal isolate degraded high-molecular-weight PE-membranes under nitrogen- or carbon-limited culture conditions (31).

Mechanical Property Losses in WPC

It is well known that strength properties are the most sensitive indicators of fungal decay in solid wood (32-35) but this concept does not necessarily apply to a composite material consisting primarily of a thermoplastic polymer matrix and wood filler. At low filler concentrations, the wood flour used in WPC does not greatly contribute to the strength of the composite unless coupling agents such as MAPP and MAPE are included in the formulations. However, the wood filler tends to increase the stiffness of the composite (2, 36) and may therefore be useful as an indicator of fungal decay in WPC. It is desirable to compare potential losses in weight and stiffness in WPC formulations caused by decay fungi to determine which of the two methods is more sensitive as an indicator of fungal decay. Khavkine et al. (37) performed three-point bending tests with compression-molded samples which had been subjected to water boiling, boiling and freezing cycles, or fungal exposure. They found that the flexural properties of the composites were more affected by cyclic exposure than either by 2 hours water boiling or fungal exposure, however, no significant weight losses following 12 weeks incubation with decay fungi were observed.

Research conducted by Silva et al. (38), Verhey et al. (39, 21) and Ibach and Clemons (40) determined strength properties of WPC following exposure to decay fungi. The effects of moisture sorption and fungal decay on strength and stiffness were separated by Clemons and Ibach (13). Clemons and Ibach (13) determined that water soaking and cyclic boiling resulted in large losses in strength of the extruded composites, however, strength losses caused by fungal attack were found for boiled composites but not water-soaked ones. In contrast, weight loss results showed that both water-soaked and boiled composites sustained weight loss.

Schirp and Wolcott (25) separated the effects of fungal decay and moisture absorption on flexural strength and stiffness of extruded WPC specimens which had not undergone any preconditioning treatment, i.e., water-soaking or boiling. Stiffness of WPC was affected more severely by moisture absorption than by fungal colonization (Table 1). Strength of WPC was not affected following incubation with decay fungi but significantly reduced by moisture absorption for a formulation containing 70 % wood filler. However, no strength reduction due to moisture was observed when the formulation contained 49 % wood filler. Weight loss was determined to be a more sensitive indicator of fungal decay in WPC than flexural strength tests for the formulations investigated.

In a parallel study (20), it was shown that after short-term (24 days) exposure, wood decay fungi may improve interfacial adhesion and reinforcement of WPC. Using dynamic mechanical analysis, an increase in the storage modulus (E'), which is closely related to stiffness as determined in static tests, was observed for fungal treated WPC. This increase in E' was attributed to a reinforcing effect of the fungal hyphae present in the interfacial gaps between wood filler and polymer matrix (Figure 1). It was supported with the observation

Table 1. (previously published in Wood and Fiber Science 37(4), 2005 643-652; copyright: Wood and Fiber Science): Stiffness (MOE), flexural strength (MOR) and weight loss (based on wood fraction) of two WPC formulations, yellow-poplar and redwood, following three months of incubation with *T. versicolor* and *G. trabeum*. Formulation #3 consisted of 49% wood, 45% HDPE and 6% additives; formulation #7 contained 70% wood, 24% HDPE and 6% additives. Each value represents the average of 16 replicates, except for yellow-poplar in the soil block test (15 replicates). Figures in bold indicate significant ($p < 0.0001$) difference between samples and incubated controls.

Material,treatment	Type of test	Density* (g/cm ³)		MOE [†] (MPa)		MOR (kPa)		Weight loss (%)	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
#3, <i>T.v.</i>	agar	1.05	0.01	1497	212	26274	2344	1.17	1.25
#3, <i>G.t.</i>	agar	1.07	0.01	1345	94	28096	1490	-1.41	0.13
#3, incub. control	agar	1.07	0.01	1461	194	26920	2926	-1.26	0.09
#3, non-incub. control	n.a.	1.09	0.01	1979	130	26862	1101	n.a.	n.a.
#7, <i>T.v.</i>	agar	0.99	0.02	797	163	11975	2226	6.32	0.47
#7, <i>G.t.</i>	agar	0.98	0.01	839	109	10590	2166	0.38	0.74
#7, incub. control	agar	1.02	0.01	806	116	11171	1948	-0.46	0.21
#7, non-incub. control	n.a.	1.12	0.01	1004	216	14696	1139	n.a.	n.a.
YP, <i>T.v.</i>	agar	0.21	0.03	2525	479	16355	3500	55.94	5.59
YP, <i>G.t.</i>	agar	0.40	0.05	6258	2098	43098	21167	25.21	5.82
YP, incub. control	agar	0.48	0.02	9719	787	97598	7374	-1.19	0.70
YP, non-incub. control	n.a.	0.61	0.02	15366	623	145183	8190	n.a.	n.a.

RW, <i>T.v.</i>	agar	0.44	0.03	9380	1661	81643	14405	3.03	0.51
RW, <i>G.t.</i>	agar	0.45	0.01	8325	1733	68763	13596	2.76	1.56
RW, incub. control <i>T.v.</i>	agar	0.45	0.02	10096	1157	89621	10501	1.29	0.54
RW, incub. control <i>G.t.</i>	agar	0.47	0.03	9244	1327	91625	21824	-0.78	0.94
#7, <i>T.v.</i>	soil	0.91	0.04	963	164	7203	1915	7.90	2.97
#7, incub. control	soil	0.94	0.04	560	180	8092	2434	0.77	0.94
YP, <i>T.v.</i>	soil	0.22	0.03	2758	948	13525	4050	61.85	4.32
YP, incub. control	soil	0.54	0.04	13479	1518	126235	14101	2.03	0.19

T.v. = *Trametes versicolor*

G.t. = *Gloeophyllum trabeum*

YP = yellow-poplar

RW = redwood

n.a. = not applicable (equilibrated only).

* determined at time of flexural strength test.

† Calculation based on the values for 20-40 % of the maximum load in the stress-strain curves.

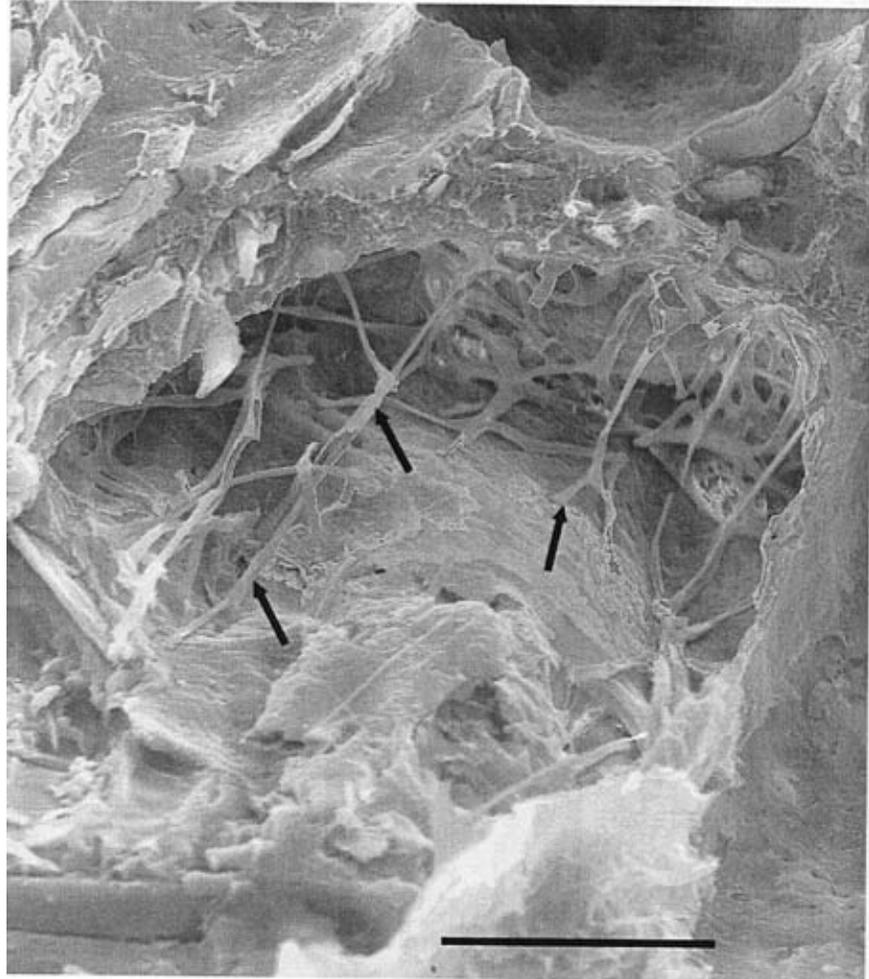


Figure 1. (previously published in *Journal of Applied Polymer Science* 99, 2006, 3138-3146: copyright: *Journal of Applied Polymer Science*): Reinforcement effect of fungal hyphae of *T. versicolor* (arrows) in WPC after 24 days of incubation (formulation #7, i.e., 70% wood, 24% HDPE and 6% additives). Bar represents 40 μm .

that higher activation energies were required for a-transition, i.e., chain rotation (41), in WPC samples incubated with the white-rot fungus *T. versicolor* as compared to untreated controls.

When using field tests, it is difficult if not impossible to separate the effects of moisture and fungal decay (21, 23). In any case, a significant reduction in the flexural modulus of WPC stakes following three years of exposure above-ground in Mississippi and Wisconsin was observed (Figure 2, 23). A significant loss in flexural strength was determined only for the above-ground WPC stakes placed in Mississippi (Figure 3, 23). Examples of extruded WPC following three years of outdoor exposure are shown in Figure 4. Decay fungi could be identified in the wood flour component of the stakes only upon examination in the laboratory. The failure break of a WPC field stake after exposure in-ground for three years in Saucier, Mississippi, is depicted in Figure 5.

Field tests on WPC were also sponsored by the U.S. Navy and conducted by Michigan Technological University over a 54-month period near Hilo, Hawaii, ending in May 2004. In these tests, AWWA Standard E7 (22) was used. Test stakes, composed of HDPE with 2% and 5% zinc borate as well as PVC with 5% zinc borate, were placed in ground contact along with creosote-treated and untreated Southern yellow pine stakes as controls. Randomly selected stakes were pulled every six months and tested for physical and mechanical properties. At the conclusion of the study, there was no visual indication of decay in any of the WPC stakes whereas the untreated Southern yellow pine stakes were all destroyed by decay fungi within 30 months. Mechanical testing of the WPC stakes showed that there was a significant loss of strength and stiffness after six months but no further significant reduction in these properties occurred over the next 48 months.

At present, no published information on the durability of WPC against soft-rot and soil fungi is available.

Mould and Staining fungi

Test methods

Resistance of WPC against mould and staining fungi is currently tested either according to plastic, wood or paper standards. Some of the laboratory test methods for mould growth on WPC which may presently be used are:

- ASTM G21-96 (42);
- ASTM D3273-94 (43);
- ASTM D 4445-91, reapproved 1996 (44);
- AWWA Draft: Standard Method of Evaluating the Resistance of Wood Product Surfaces to Mold Growth (45);
- BS 3900: Part G6: 1989 (46);
- ISO 16869 (47);

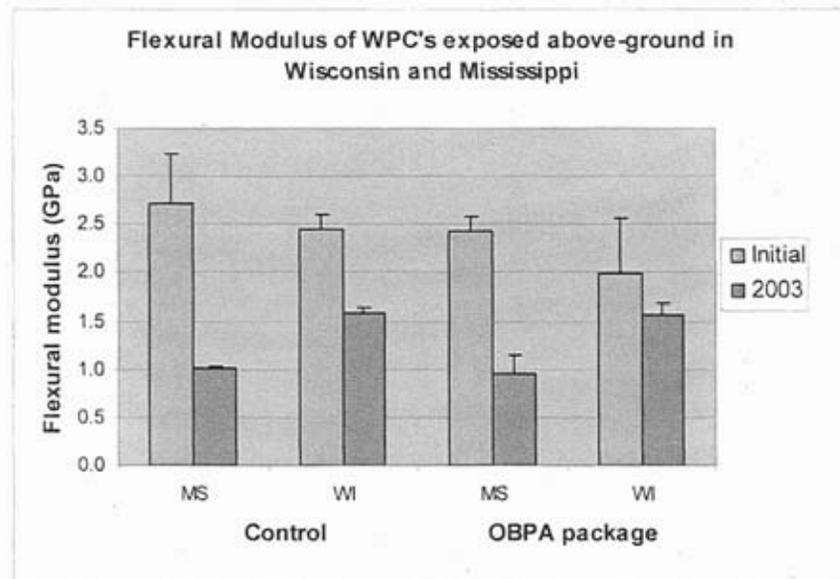


Figure 2. (previously published in *Conference Proceedings, Progress in Woodfiber-Plastic Composites, Toronto, Canada, 2004, 1-13*): “Control” consists of 48.5% HDPE (reprocessed milk bottles; H. Muehlstein and Co., Inc., Norwalk, CT). 50% Western pine wood flour (40 mesh, i.e., 420 μm , American Wood Fibers, Schofield, WI) and 1.5% light stabilizer package (Ciba Speciality Chemicals Co., Terrytown, NY). “OBPA package” contains 47.5% HDPE, 50% Western pine wood flour, 1.5% light stabilizer package plus a 1% OBPA package (Rohm and Haas, Philadelphia, PA).

(Drr page 9 of color inserts.)

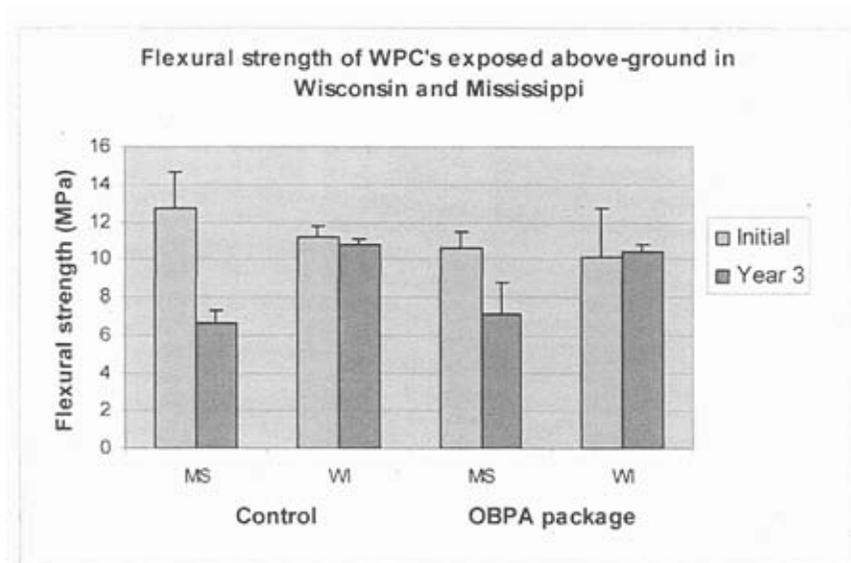


Figure 3. (previously published in *Conference Proceedings. Progress in Woodfiber-Plastic Composites*, Toronto, Canada, 2004, 1-13): “Control” consists of 48.5% HDPE (reprocessed milk bottles; H. Muehlstein and Co., Inc., Norwalk, CT), 50% Western pine woodflour (40 mesh, i.e., 420 μm , American Wood Fibers, Schofield, WI) and 1.5% light stabilizer package (Ciba Specialty Chemicals Co., Terrytown, NY). “OBPA package” contains 47.5% HDPE, 50% Western pine woodflour, 1.5% light stabilizer package plus a 1% OBPA package (Rohm and Haas, Philadelphia, PA). (See page 9 of color inserts.)

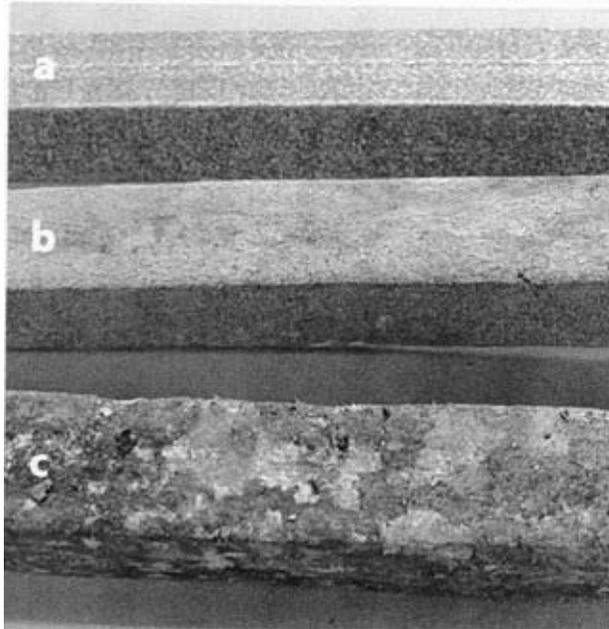


Figure 4. Extruded WPC: a) unexposed; b) above-ground exposure for 3 years in Saucier, Mississippi; c) in-ground exposure for 3 years in Saucier, Mississippi. (See page 10 of color inserts.)

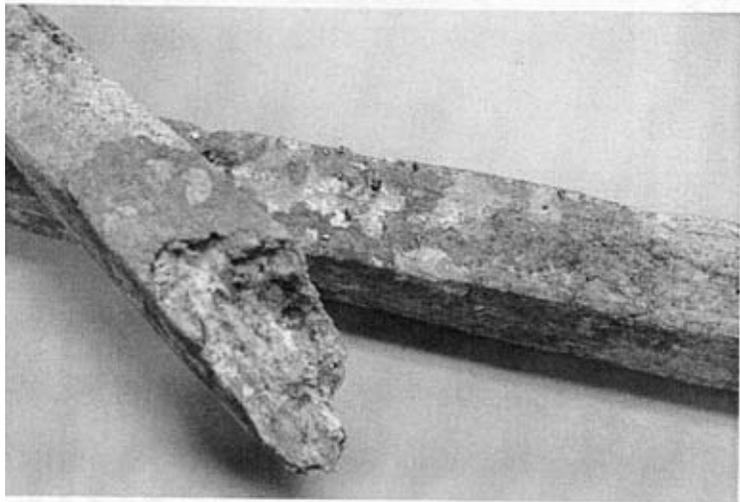


Figure 5. Failure break of a WPC field stake after exposure in-ground for 3 years in Saucier, Mississippi. (See page 10 of color inserts.)

TAPPI T 487 pm-99 (48).

Fungal species used in mould testing are *Aspergillus niger*, *Aureobasidium pullulans*, *Penicillium purpurogenum*, *Stachybotrys chartarum*, *Chaetomium globosum* and others. Generally, a mixed fungal spore suspension containing a defined number of spores per mL is prepared and sprayed on the test specimens which are placed on an agar-based medium in a Petri dish. Specimens are incubated at a defined temperature (around 30°C) and high (around 85 %) relative humidity for a set amount of time, for example, 28 days (42).

All mould test methods listed above are based on visual quantification of fungal growth on the material surface. For example, in ASTM G21-96 (42), observed growth on specimens is rated between 0 (no growth) and 4 (heavy growth (60% to complete coverage)).

Effect of mould and staining fungi on wood and the environment

Fungi which cause discoloration of wood in storage and service are generally described as staining fungi while those that grow superficially on wood are called moulds (49). Staining and mould fungi are members of the *Ascomycetes* and *Deuteromycetes (Fungi Imperfecti)* although a few of the moulds are *Zygomycetes* and are classified in the Mucorales. Both mould and staining fungi lower the aesthetic quality of wood-based products due to discoloration. It is well known that sapstaining fungi produce extracellular enzymes to utilize nonstructural components of sapwood, namely sugars, proteins, and extractives (50-53). Extracellular cellulase is not secreted by sapstaining fungi, however, selected species were shown to produce low amounts of xylanase and pectinase, and one species (*Ophiostoma piceae*) produces mannanase (54). The apparent inability of staining fungi to decay wood cell walls is not universal because some of these fungi are known to cause soft rot in hardwoods under optimal conditions (49).

Staining and mold fungi do not significantly affect the mechanical properties of wood (55-58), however, some species of sapstaining fungi may reduce impact bending strength or toughness of wood (59-63). Due to increased permeability in sapstained wood, the use of this material for appearance purposes is limited because it absorbs solutions excessively resulting in uneven finishes. Allegedly, some mould species such as *Stachybotrys chartarum* (syn. *Stachybotrys atra*) potentially produce health-related building issues. It was reported that 8,000 to 10,000 lawsuits were pending nationwide in the United States for mold litigation in the year 2002 alone (64). It must be stressed that mold and mold spores are always present in the air, indoors as well as outdoors, and that infections may occur only in immune-compromised human beings or following prolonged exposure to exceedingly high airborne mold concentrations, such as in an

agricultural setting (65). The amount of mold that must be inhaled to cause an allergic response is unknown (65). In the United States, there are currently no regulations or exposure limits for molds or mycotoxins in homes and commercial or public settings. However, in the occupational setting, a general duty clause may apply to mold exposures, i.e., the rule that requires employers to provide workers with a safe and healthy work environment (65).

Effect of mould and staining fungi on WPC

In the short term, mould and staining fungi may represent a more relevant issue with WPC than decay fungi since these first two types of fungi develop more quickly on the material. Many of the mould fungi known to grow on wood have also been isolated from plastics, for example, *Aureobasidium*, *Aspergillus* and *Penicillium*, and therefore are of importance as screening organisms for both plastic and wood (66). According to anecdotal evidence (10), WPC are susceptible to mould and staining fungi, however, very little detailed information on this issue is available in the scientific literature (66, 67).

Laks et al. (67) examined the effect of several manufacturing variables (i.e., wood loading, extrusion lubricants, fungicides and extruder temperature) on mould susceptibility of wood-plastic composites. It was determined that an increase in wood content as well as surface roughness increased mould growth and that lubricants generally increased mould growth rates. Both in-process fungicides tested, chlorothalonil and zinc borate, significantly reduced mould growth on WPC, however, both fungicides were used in different concentrations. Thus, the efficiency of both biocides cannot be directly compared. The effect of extruder temperature on mould development could not be conclusively determined and needs to be investigated further.

In conclusion, so far the effect of staining and mould fungi on WPC has been evaluated only by visual quantification of microbial growth on the material. A drawback of this method is that it is based on subjective evaluation, unless this quantification could be achieved using a photometric colour coordinate system. In addition, measuring mould growth on a material cannot provide information on which components included in the plastic are degraded and if mould growth leads to a reduction in WPC strength properties. It must be emphasized again that synthetic polymers themselves are resistant to fungal growth but that generally, other components such as plasticizers, lubricants, stabilizers and colorants are responsible for fungal colonization and decay on plastic materials (42). Although mould fungi do not significantly affect the mechanical properties of wood, as previously mentioned, some fungi may be able to degrade certain additives in plastic and WPC, especially following oxidation (8), and thus reduce the WPC strength.

Termites

As previously mentioned, scientists at the USDA Forest Products Laboratory have been evaluating the field performance of extruded WPC, including deterioration by termites according to ASTM D1758 (23). Termite nibbling was visible on in-ground stakes after the third year of exposure in Saucier, Mississippi, but not on test material in Wisconsin due to the lack of termites in this state.

Results from termite field testing of WPC over a 27-month period were also obtained through work sponsored by the U.S. Navy and conducted by Michigan Technological University. Three WPC formulations were tested, HDPE with zinc borate, HDPE without zinc borate, and PVC without zinc borate. For the test, ten replicates were used for each of the formulations. The test specimens (50 x 125 x 25 mm or 50 x 90 mm) were randomized and placed horizontally on top of hollow concrete blocks four inches above the ground at a field site near Hilo, Hawaii. The test site is characterized by a naturally occurring, large Formosan termite population. Untreated aspen (*Populus tremuloides*) bait stakes (200 mm length x 25 mm thickness x 25 mm width) were driven vertically into the ground through holes in the concrete blocks to attract termites. In addition, untreated aspen feeder stakes (450 mm length x 25 mm thickness x 25 mm width) were placed between the rows of test blocks. The bait stakes were in direct contact with the feeder stakes which in turn were touching the test blocks. The assembled units were enclosed with a boxed lid to maintain dark conditions and provide shelter from rainfall. Termite-damaged bait and feeder stakes were replaced every six months during inspection of the test blocks, resulting in a permanent high termite hazard for the blocks. The rating system was according to AWWA E7 (22) in which a block rating of 10 equals perfect condition while a rating of 0 means failure. No termite activity on any of the WPC test blocks was observed throughout the test period whereas seven out of ten aspen control specimens were attacked by termites resulting in four failures.

There is no published information available in the scientific literature on WPC laboratory tests using termites.

Algae

No information is available in the scientific literature on algal growth on WPC although tests involving algae have been performed (68). Test method VDL RL 07 (69) was used to measure algal growth on WPC (68). This method is intended to determine the resistance of coating materials against algal growth by adding the coated test specimen to a suitable agar medium and inoculating the specimen with a standard algal solution followed by incubation for a specific duration. An alternative test method is IBRG/P98/03, however, it is available for members of the International Biodeterioration Research Group (www.ibrg.org) only.

Bacteria

Bacteria compete poorly with fungi on wood, however, where favourable conditions exist, they may represent a major form of decay of man-made structures (70). True wood degrading bacteria exist in all parts of the world, and have a tendency to be aerobic and exist in a diverse range of aquatic and terrestrial environments. Anaerobic and aerobic bacteria are known to cause a pronounced increase in the permeability of round wood, including refractory species like spruce, during ponding or water sprinkling.

Fungi are the most important microorganisms which colonize and degrade synthetic polymers, however, bacteria such as *Pseudomonas aeruginosa*, *Serratia marcescens* as well as several species of the genus *Micrococcus*, *Bacillus* and *Streptomyces* are also involved in deterioration of synthetic polymers (71). Lee et al. (72) provided the first report demonstrating degradation of oxidized, starch-containing polyethylene by *Streptomyces* species in pure culture. *Streptomyces* and other filamentous bacteria (actinomycetes) have also been reported as important colonizers of wood in ground contact although few reports showing degradation of lignified wood fibres are available (70). The effect of bacteria on WPC merits further research due to a lack of investigations carried out in this area.

Marine Organisms and Seawater

Marine test procedures with WPC according to ASTM D2481 (73) were conducted in Pearl Harbor, Hawaii (74). The test method is generally used to determine the relative efficacy of preservatives in small wood specimens exposed to a natural marine environment and was adapted to evaluate small WPC-specimens. A qualitative index of physical condition, determined visually during periodic inspections, was used to measure resistance to marine borer attack. In addition, flexural strength tests according to ASTM D790 (75) were performed periodically. The selected test site is known to have marine borer populations, including several *Teredo* spp. (shipworm), *Limnoria tripunctata* (gribble) and *Martesia striata* (pholad) that commonly cause failure of timber piling.

Two commercial WPC formulations were included in the study by Pendleton and Hoffard (74) consisting of a 70/30 mixture (by weight) of wood flour and HDPE and a 50/50 mixture of wood and LDPE. The LDPE specimens were added after the study had been in progress for two years. Untreated Southern yellow pine specimens were included as controls and as bait. All specimens were cut from larger pieces and measured 15.24 cm x 3.81 cm x 1.27 cm. Wood-plastic and wood specimens were attached to a test rack which was suspended from a pier about five feet below mean low tide. The wood specimens

were replaced annually because of heavy borer attack by *Limnoria* and *Teredo*. Fouling organisms were removed annually to facilitate inspections.

The HDPE and LDPE specimens exhibited no marine borer attack after three years and one year of exposure, respectively. A loss in flexural strength and stiffness of the formulations was observed but attributed to wetting during exposure and not to marine borer degradation. Similar strength losses were found in the materials after simple wetting and drying.

Due to the high wood content and lack of any preservative in the HDPE formulation tested it may be concluded that current commercial WPC formulations with a low wood content are not at risk to *Limnoria* or *Teredo* attack. A remote risk exists that boring clams, such as *Martesia striata*, may present an occasional problem as these clams can bore into any material, including plastics and WPC, which are softer than the clam's shell. However, as no boring clams were observed in the wood specimens, no conclusions could be drawn in this regard.

Antimicrobial treatments for WPC

WPC manufacturers responded to the risk of fungal attack in their products by incorporating zinc borate into formulations prior to extrusion. Zinc borate has become the dominant preservative used for this purpose because of its relatively low water solubility, resistance to leaching, ability to withstand common extrusion temperature, low cost and very low environmental and work toxicity hazard (76, 77). Zinc borate is effective against wood decay fungi and insects but, as mentioned previously, not highly effective against mould and staining fungi at low concentrations (66, 77). It is suited for exterior applications with a low to moderate leaching hazard (American Wood Preservers Association H2 use level), e.g., siding, exterior trim and window components (76). Zinc borate at 2% concentration was shown to prevent any weight loss in extruded WPC and will take at least 20 years to completely dissolve and leach from the material (28). Simonsen et al. (78) determined that the addition of zinc borate or a blend of calcium and sodium borates before molding dramatically reduced weight losses of WPC due to decay fungi. Weight losses overall were below 2% when calculated on the basis of the wood content, however, the maximum weight loss of a WPC without any biocide was only 6%. For solid wood, according to ASTM D 2017-05 (16), a species displaying a mass loss due to Basidiomycete attack below 10 % is considered highly resistant. In European standard prCEN/TS 15083-1:2004 (19), a provisional durability rating scale is provided which classifies a timber as very durable at 5 % or less mass loss.

Dylingowski (66) compared the efficiency of an isothiazolone biocide, specifically, dichloro-octyl-isothiazolone (DCOIT), and of zinc borate against mould growth on WPC following testing according to a variety of methods and

fungal isolates. DCOIT was shown to be more effective against mould growth than zinc borate. DCOIT is incorporated into a plastic pellet, added to the extrusion process, dissolved into the WPC matrix and slowly migrates to the WPC surface. It has a high thermal stability, very low water solubility and broad-spectrum activity against fungi, actinomycetes and gram positive bacteria (66).

Once mold growth has appeared on a WPC surface, several commercial WPC producers recommend cleaning the material with water-diluted sodium hypochlorite (bleach). However, bleach treatments do not completely eliminate fungal growth from wooden surfaces since they lack broad spectrum antifungal activity (79), and this may also apply to WPC. It may be necessary to use biocides to prevent mold growth or delay re-growth of mold on WPC. Periodic cleaning of WPC decking is advised to prevent the build-up of debris and pollen that can cause mold growth.

Some commercial WPC products, especially for use as decking material, contain broad spectrum treatments against bacteria, mold and mildew such as Microban® or Sanitized®. A bacteriostat, disinfectant (bacteriocide / germicide) and fungicide, 10, 10'-Oxybisphenoxarsine (OBPA), is currently being tested in field trials with extruded WPC formulations by scientists at the USDA Forest Products Laboratory (23).

Strategies for Improvement of WPC Durability

Introduction

All biological agents of deterioration have four basic requirements (80):

- Nutrient source;
- Moisture (generally greater than 30%);
- Temperature (generally between 5°C and 40°C, but optimally between 24°C and 30°C);
- Oxygen.

Since it is not feasible for WPC applications such as decking to limit oxygen supply and/or to adjust temperature to an out-of-optimum level for microorganisms, most strategies to inhibit biodegradation are either based on moisture or nutrient exclusion. Wood is rendered an unfavorable microbial nutrient if it is of high natural durability or if it is of low natural durability and treated with preservative(s), and synthetic polymers are relatively inert to biodegradation.

Nutrient exclusion or modification

WPC durability may be improved by minimizing the amount of nutrients available for microorganisms, i.e., wood and specific additives. As previously

mentioned, laboratory tests have shown that no significant weight loss occurs in WPC containing 50% wood by weight. Similar baseline thresholds for additives (lubricants etc.) cannot be provided at present due to a lack of systematic investigations. If higher amounts of wood are incorporated into a formulation it may be advisable to also include zinc borate, but long-term field tests are required to clarify this aspect.

While it is recognized that fungal durability may be improved by reducing the amount and size of wood in a formulation, at the same time strength and especially stiffness of the WPC may be reduced and material costs will be increased because more plastic will be included. It was shown that the incorporation of particles with a high fiber aspect ratio may improve the mechanical properties of WPC (36).

Another strategy for nutrient exclusion would be to achieve inaccessibility of the degradable wood particles in WPC by encapsulating them with the plastic matrix.

The incorporation of wood originating from a naturally durable species may also improve biological durability of WPC. A promising alternative to the use of naturally durable wood is to chemically modify the wood filler prior to compounding and extrusion or molding. Chemical modification involves treating wood with chemicals that alter the water-sorption properties either by bulking the cell lumens to the point where water is excluded or by cross-linking the cellulose hydroxyls to reduce hygroscopicity (81). In order to improve compatibility of wood fibers with a thermoplastic matrix and improve material performance, chemical wood modification is aimed at changing fibers from a hydrophilic into a permanent hydrophobic state. The three best-investigated groups of modification agents for wood are anhydrides, isocyanates and epoxides (82). Acetylation has been the subject of most investigations due to the relatively low cost of the reagent and its relatively low toxicity (82). Liu et al. (83) investigated the potential of acetylation for modification of wood in WPC and determined that acetylation increased the interfacial shear strength between polystyrene and wood.

Compression-molded WPC based on wood flour modified with either acetic anhydride, butylene oxide or propylene oxide were prepared by Ibach and Clemons (40). A correlation between moisture and fungal decay was determined. Weight loss due to fungal decay increased with increasing specimen moisture content. Lowest weight losses were obtained with acetic anhydride modification, followed by butylene oxide modification, untreated control, and propylene oxide modification.

Despite the relatively low cost of acetic anhydride, the cost of modification includes the removal and recovery of acetic acid which may add complexity to the process. Thus, any composite based on an acetylated lignocellulosic substrate may have to be aimed at a value added market (82).

Moisture exclusion

Microorganisms depend on water in order to grow and cause deterioration. Thus, if moisture content of the wood filler can be kept out or at least below 20%, decay in WPC may be prevented. This could be achieved by complete encapsulation of wood particles by the plastic matrix, hydrophobation of the WPC surface, or by chemical modification of the wood substrate. It must be emphasized that no reasonable amount of zinc borate will prevent moisture entry into the composite. As a consequence, moisture exclusion is of paramount importance when designing and processing WPC.

Microbial pathway exclusion

Voids between wood and plastic represent entry points and proliferation pathways for microbes; thus, these should be eliminated or reduced. With regard to polymer-starch composites, biodegradation kinetics were examined based on scalar percolation theory (84). Scalar percolation theory deals with the connectivity of one component randomly dispersed in another (84). When the starch fraction exceeds a specific percolation threshold, significant pathways for microbial invasion are generated and degradation is accelerated.

It can be expected that the shape of the wood substrate in WPC, i.e., flour versus fibers, as well as wood density differences also have an influence on the degradation rate of WPC, however, this aspect has not been investigated.

WPC Standardization Initiatives in North America and Europe

With the improvement in WPC technology, it is likely that new product applications will result. Thus, standardization issues will become increasingly important on a world-wide level. The development of adequate WPC standardization is required to ensure market acceptance, growth and diversification, especially in Europe.

In North America, standards for plastic lumber as well as for WPC have existed for a number of years. Acceptance criteria based on ICC ES Acceptance Criteria AC174(3) are being used to address one of the significant WPC product applications in the United States, deckboards and guardrails (85). According to these criteria, a particular product is tested according to code-prescribed loads in the configuration for which the proponent has requested a code listing. Due to the unique nature of WPC in a specific application, acceptance criteria include test requirements such as temperature, moisture, UV and freeze-thaw effects. The important point is that in the United States, all materials for a specific

application, must perform according to the same performance limits (86). For example, WPC may be compared with preservative-treated lumber when used as decking material. Currently, there is an on-going effort to eliminate termite and fungal decay testing in the ICC ES Acceptance Criteria for products with less than 70% wood content (86). The motivation for this effort is that no commercial WPC product with less than 70% wood has been shown to behave worse than preservative-treated lumber in tests according to ASTM D2017 (16), ASTM D1413 (17), ASTM D3345 (87), AWPA E1 (88) and AWPA E10 (89). At the same time, biological durability issues have become a top concern of the North American WPC industry due to a few early product failures (86). Hence, it remains to be clarified if the standards used for fungal durability testing of wood adequately address the long-term durability performance of WPC.

In Europe, several initiatives are currently working on WPC standardization (4):

- CEN/TC 249 Plastics, WG 13: Plastics - Wood-plastic composites (convenor: Claudine Bloyaert, Technical Marketing and Development, SolVin, Brussels, Belgium);
- CEN/TC 112 Wood-based composites, WG 12: Wood Plastic Composites (convenor: Dr. Alfred Teischinger, Institute of Wood Science and Technology, Department of Material Sciences and Process Engineering, University of Natural Resources and Applied Life Sciences, Vienna, Austria);
- ON (Österreichisches Normungsinstitut, Austrian Standardization Institute), Fachnormenausschuss 087 Holz, AG Holz-Kunststoff-Verbundwerkstoffe (convenor: Dr. Alfred Teischinger).

Standardization to be developed by CEN/TC 249/WG 13 has the following objectives:

- To provide an exhaustive description of the test methods applicable to WPC;
- To define the common characteristics of the different applications;
- To provide characteristics and corresponding requirements for specified applications.

With regard to biological durability of WPC, publications and recommendations from experts in this field are currently compiled to serve as the basis for a durability standard to be issued by CEN/TC 249/WG13. CEN/TC 112 is currently working on the development of a scope for standardization, i.e., raw material characterisation and compounds, composites and products. The current main challenge in Europe will be to coordinate standardization initiatives such that the requirements of both the plastics and wood industries and research organizations will be met.

Conclusions

Despite a significant increase in WPC research in recent years, some fundamental questions on the biological durability of WPC still need to be addressed, for example:

- How durable is WPC under long-term outdoor exposure, including biological and abiotic factors?
- Can long-term outdoor exposure be adequately simulated by using an accelerated laboratory test?
- Which fungi colonize and degrade WPC under natural, i.e., outdoor conditions? Is there an ecological succession with regard to different types of fungi (moulds, decay fungi)?
- Which fungi and/or bacteria should be used as test organisms in WPC standards?
- How does the shape (flour versus fibers) and type (durable versus non-durable) of wood substrate influence degradation of WPC?
- Under which conditions can polymer matrices be biologically degraded?
- Are additives in WPC degradable and do they contribute to degradation?
- Which applications require fungicides in WPC formulations?
- Is there a possible interference between fungicides and coupling agents?

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