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Section 1 **Biology**

Termite resistance of wood-plastic composites made with acetylated wood flour, coupling agent or zinc borate

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Termite resistance of wood-plastic composites made with acetylated wood flour, coupling agent or zinc borate

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ABSTRACT

There is little published scientific literature on the laboratory or field testing of wood-plastic composites (WPC) against termite attack. Therefore, the objective of this research was to investigate termite resistance (both laboratory and field) of 5 different extruded WPC blends of 50% western pine wood flour (WF) and high-density polyethylene (HDPE), and untreated pine solid wood. The study included two unmodified control WPC blends (one with cut surfaces and one with plastic rich surfaces); one blend with 3% coupling agent (maleated polyethylene, MAPE); one blend with acetylated WF; and one blend with 1% ZB. These 3 modifications/treatments were chosen because (1) coupling agents are known to promote bonding between the plastic and unmodified wood fibers when added to WPCs; (2) acetylating the wood esterifies the hydroxyl groups, making the wood more hydrophobic, dimensionally stable, and biologically durable; and (3) ZB is a known fungicide and insecticide used in some commercial WPC formulations.

All WPC blends tested performed well (0.1% - 0.7% weight loss; 66% – 71% termite mortality) in the 10 week dry-wood laboratory termite test compared to the untreated solid wood (9.1% weight loss; 33.8% mortality.) Field exposure ratings are on a scale from 0 (complete failure) to 10 (sound, one to 2 small nibbles should be permitted.) After 30 months field exposure to *Macrotermes gilvus* Hagen in Bogor, Indonesia the acetylated WPCs performed the best with ratings of all 10s and only 0.8% weight loss, followed by the 1% ZB blend with ratings of all 9s and 3.9% weight loss. The coupling agent blend had ratings of all 8s and 13.6% weight loss, while the unmodified WPC controls had ratings of 7 and from 11.3-22.6% weight loss. The solid wood was completely destroyed with 100% weight loss and 0 rating.

Keywords: termite resistance, laboratory, field, wood-plastic composite, acetylation, zinc borate, coupling agent.

1. INTRODUCTION

There is increasing demand for wood-plastic composites (WPC) in the construction industry and the key driver for the market growth is in the applications of decking, fencing, molding, and siding. The mechanical strength, low maintenance and weight reduction offered by WPCs are expected to increase the demand in the construction industry over the next eight years. 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).

With time and weathering in outdoor exposures, moisture can penetrate into the WPC and if given the appropriate conditions can ultimately result in decay. The mechanisms and impact of moisture, UV, and fungal deterioration and the mechanical loss on WPCs has been the focus of our research (Chow et al. 2002, Clemons and Ibach 2004, Ibach et al. 2007, Ibach and Clemons 2007, Segerholm et al. 2012, Ibach et al. 2013, Ibach et al. 2016, Ibach et al. 2017, Sun et al. 2017).

Termites damage untreated wood materials in many parts of the world, however little is known of the termite resistance of WPC materials. There are few publications on the durability of WPC materials against termites and most of these are limited to laboratory studies (Chow et al. 2002, Klyosov 2007, Shirp et al. 2008, López-Naranjo et al. 2012, Kartal et al. 2013, Xu et al. 2015, López-Naranjo et al. 2016). Overall, when compared to untreated solid wood, the WPC materials have greater termite resistance, but attack can occur over time. The incorporation of ZB increases the termite resistance (Kartal et al. 2013, López-Naranjo et al. 2016). Higher wood contents (70%) increased the mass losses from termite attack and particle size is an important factor in the termite degradation of WPC. As particle size decreased from 30 to 100 mesh, mass losses decreased (Kartal et al. 2013). The particle size used in this research was nominally -40, +80 mesh. Recently, longer term field studies were published (Gardner and Bozo 2018, Ibach and Clemons 2017, Tascioglu et al. 2018). After 10 years in-ground exposure in Santiago, Chile, all the WPC blends performed well (Gardner and Bozo 2018). After 7 years above ground exposure of WPC in southern Japan, it was concluded that ZB levels needed to be higher than 2% for full protection (Tascioglu et al. 2018). After 7 years in-ground field exposure in Saucier, Mississippi, WPC made with acetylated wood flour showed no termite attack (Ibach et al. 2017).

Here we summarize our initial investigations on the termite resistance of WPC materials including a comparison of different protection approaches in both laboratory and field tests. Specifically, our objective was to investigate the termite resistance of 5 wood-plastic composite (WPC) blends and 1 untreated solid wood: 1.) HDPE with 50% wood flour, 2.) HDPE with 50% acetylated wood flour, 3.) HDPE with 50% wood flour and coupling agent, 4.) untreated Southern pine sapwood, 5.) HDPE with 50% wood flour and 1% zinc borate (cut from larger deck board), 6.) HDPE with 50% wood flour (cut from larger size deck board, i.e. no plastic rich surface). Both the addition of coupling agents and acetylation of the wood component offer ways for reducing moisture sorption, while the ZB is an effective biocide. The goal of this study is to understand the extent of termite damage to WPC materials and to identify methods for improving the long term durability of these materials.

2. EXPERIMENTAL MATERIALS AND METHODS

2.1 Materials:

- Plastic (HDPE): High density polyethylene (BP Solvay Polyethylene, grade A60-70-162; MFI approximately 0.72).
- Wood flour (WF): grade 4020 pine, (nominally -40, +80 mesh pine flour, mostly ponderosa pine). American Wood Fibers (Schofield, WI).
- Acetylated wood flour (AWF): Above wood flour that was acetylated to approximately 20% weight gain.
- Maleated Polyethylene Coupling Agent (MAPE): Polybond 3009, a maleated high-density polyethylene (MA-HDPE) supplied by Crompton Corporation (Middlebury, CT).
- Lubricant (Lub): A blend of calcium stearate and a proprietary amide, (Struktol TR 016, Struktol Company of America). Added at a level of 6-8%.
- Biocide (ZB): 1% Zinc Borate (Boragard ZB, U.S. Borax)
- Solid wood: untreated Southern pine sapwood

2.2 Processing

Experimental design and formulations for each blend are shown in Table 1. For blends 1-3, field stakes (19 by 19 by 457 mm) and laboratory specimens (3 by 13 by 89 mm) were profile extruded by a direct extrusion process using a 32 mm corotating twin-screw extruder. Wood and HDPE were added into the main feed throat. The molten material is then shaped to size by forcing it through a die at the end of the extruder. A sizing die, water spray tank, and puller were used to solidify the plastic in the desired shape and continuously remove it from the extruder. Addition of lubricant (Lub) was used to prevent tearing of the melt as it exited the die. Blends 5 and 6 were processed by a commercial-scale extruder at the University of Maine's Advanced Engineered Wood Composite Center. Radius edge deck boards were produced (30 mm by 140 mm) using an in-line twin-screw extruder profiling system on a 94 mm profiling extruder (Davis Standard, Woodtruder). Laboratory and field stakes were then cut from these deck boards. A surface of extruded WPC boards was covered with a plastic rich skin. To eliminate the impact of the skin on test results, this plastic rich layer was removed from one group of control samples (control cut) by cutting.

Table 1: Experimental design and formulations of each blend.

Materials ID Number and Description	Plastic	Wood Type	Wood Content (%)	Coupling Agent (%)	Biocide (%)
1 - Control	HDPE	WF	50	0	0
2 - Acetylated	HDPE	AWF	50	0	0
3 - MAPE	HDPE+ MAPE	WF	50	3	0
4 – Solid wood	0	Solid Wood	100	-	-
5 - ZB	HDPE	WF cut surfaces	50	0	1% ZB
6 – Control-cut	HDPE	WF cut surfaces	50	0	0

The formulations to be studied were the following: 1) unmodified wood flour, 2) acetylated wood flour 3) coupling agent (MAPE) 4) unmodified solid wood, 5) Zinc borate (ZB) biocide (cut surface, no plastic rich surface), 6) unmodified wood flour (cut surface, no plastic rich surface). For each termite exposure test, 10 replications per blend was used.

2.3 Laboratory termite tests

2.3.1 Dry wood termite test

In laboratory no-choice tests, specimens were evaluated against the feeding by the dry wood termite, *Cryptotermes cynocephalus* Light (Hadi and Febrianto 1991, Hadi et al. 1995). Fifty healthy and active workers were put into each box (10 by 5 by 5 cm in L, W, T) containing specimens of one type, which were maintained in a dark room at an average temperature of 20 to 32 °C and 81 to 89% relative humidity (RH) for 10 weeks. At the end of the test, termite mortality and percent wood weight loss were determined.

2.3.2 Subterranean termite test

Specimens were evaluated in a no-choice test against feeding by the subterranean termite, *Coptotermes curvignathus* Holmgren, using the Japanese method (Association 2004). Each specimen was placed in an acrylic cylindrical tube (60 mm height; 80 mm diameter), with 150 workers and 15 soldiers. A wet tissue was placed in each tube to maintain humidity. The tubes were maintained in a dark room at an average temperature of 28 to 30 °C and 81 to 89% relative humidity (RH) for 3 weeks. At the end of the test, the percent weight loss of each specimen was determined as well as termite mortality (Standard 2006, Arinana et al. 2012).

2.5 Field tests

Field stakes were exposed in-ground to termite feeding by two different subterranean termite species: *C. curvignathus* or *Macrotermes gilvus* (Hagen). *C. curvignathus* was already identified in one plot, but in another field, with special arrangement, we cultured for *M. gilvus*. The procedure followed was a consensus of the Indonesian Wood Preservation Association which was adapted from some standards (Standard 2006). Before the Indonesian National Standard was published, the consensus was followed, so there was no standard test method number. Specimens were placed horizontally on the surface of field soil and covered by a plastic box to eliminate exposure to sunlight. The test was carried out in Serpong, Indonesia, which has an average of 2562 mm/year rainfall, 87.6% RH and 27.5 °C temperature. After 3, 6, 12, and 30 months specimens were inspected (pictures taken) and percentage weight loss was determined. The specimens were cleaned, oven dried at 60 °C for 2 days and then weighed. This lower drying temperature (60 °C) was used to minimize the effect of heating on the WPC. Visual ratings for each wood specimen were determined using the rating system shown in Table 2 for termites following AWPA E7 (AWPA 2007). Only data from the 30 months inspection is presented in this paper.

Table 2: Rating system for field test termite resistance.

Rating	Condition of Specimen
10	Sound. 1 to 2 small nibbles permitted
9	Slight evidence of feeding to 3% of cross section
8	Attack from 3 to 10% of cross section
7	Attack from 10 to 30% of cross section
6	Attack from 30 to 50% of cross section
4	Attack from 50 to 75% of cross section
0	Failure

3. RESULTS AND DISCUSSION

3.1 Laboratory termite tests

The results of specimens exposed to *C. cynocephalus* showed high termite mortality and low weight loss (less than 1%) in all of the WPC blends compared to the untreated solid wood (9.1±2.2% weight loss) (Fig. 1 and Fig. 2). This high mortality could be from lack of appropriate moisture/water in the WPC material since moisture sorption is much slower than in solid wood, or from access to appropriate food in the WPCs, as opposed to a toxicity mechanism. ZB was the only bioactive compound used in any of the WPC blends and it had a weight loss of 0.47±0.02%, as seen in Fig. 2. Interestingly, the 2 blends based on lowering the equilibrium moisture content (EMC) had the lowest weight losses of 0.23±0.10% for the MAPE coupling agent and 0.18±0.05% for the acetylated WPC blend.

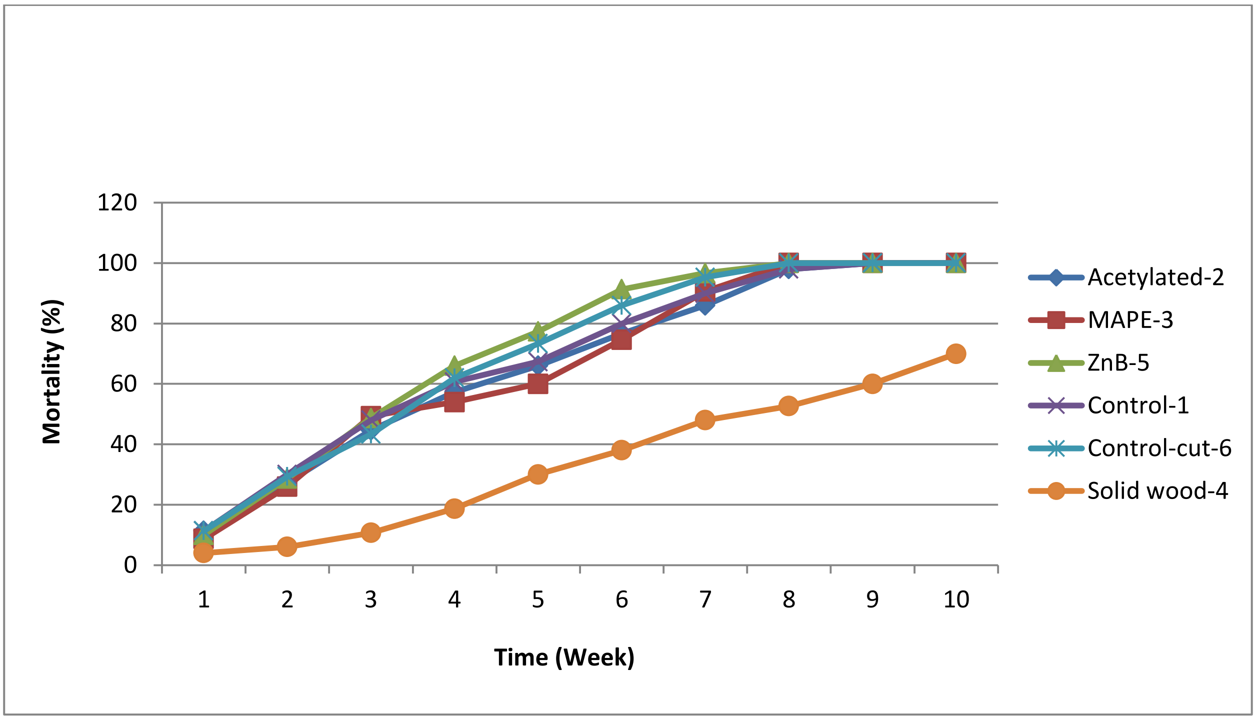

Figure 1: Laboratory dry wood termite test results after 10 weeks exposure to *C. cynocephalus*.

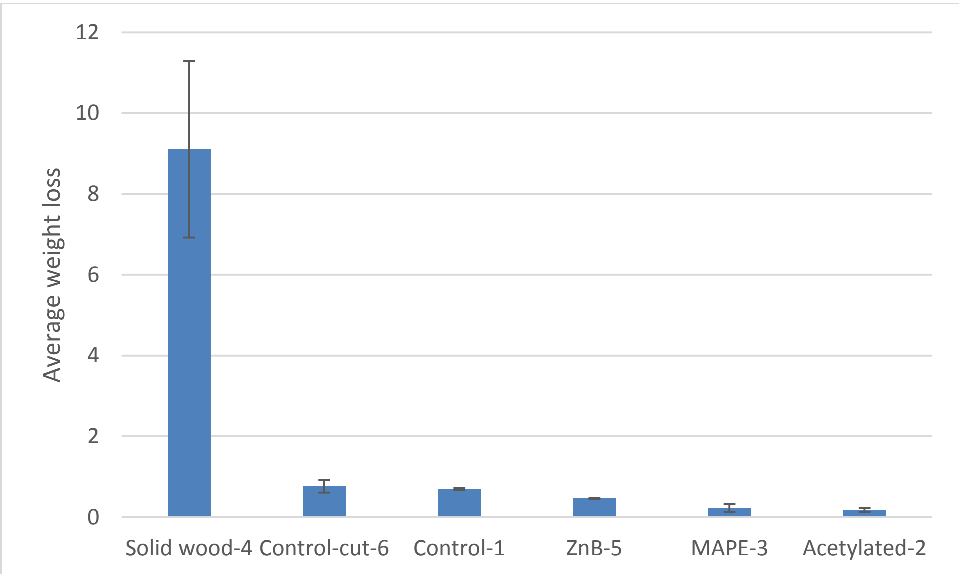

Figure 2: Average weight loss after the 10 week dry wood laboratory termite test.

Similarly, laboratory tests with subterranean termites, *C. curvignathus* also showed high termite mortality and low weight loss in the WPC specimens (not shown) (specimens after 3 weeks exposure - Fig. 3). Only blend 4, (untreated solid wood) showed deterioration. Both laboratory standards followed were for solid wood, not WPC. Both sets of laboratory results indicate that the test procedure could be modified for WPC by perhaps water soaking the specimens before placing in test to make sure it is not just a lack of moisture in the test specimen that is leading to low weight losses and high mortality. This was found necessary in previous work investigating decay using soil block evaluations (Clemons and Ibach 2004). Additionally, another option would be to perform a laboratory choice test.

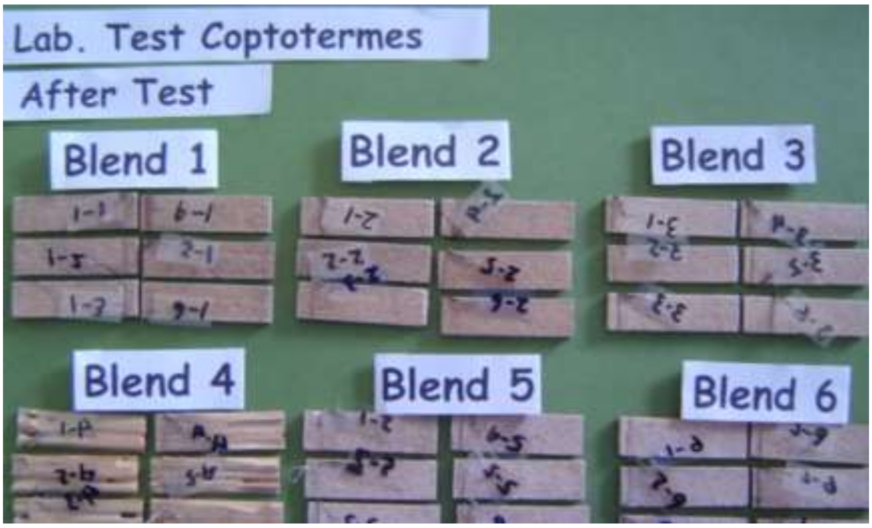

Figure 3: Specimens after 3 weeks exposure to *C. curvignathus*.

3.2 Field tests

Overall, the WPC specimens were in good condition after 30 months outdoor field exposure to *C. curvignathus* (Fig. 4). The control (uncut surfaces, control-1) had an average rating of 9 and all of the rest of the blends had average ratings of 10. All specimens tested showed little weight loss (less than 4%). The ZB and the acetylated WPC blend had the lowest weight losses at 0.47±0.71% and 0.73±0.09% for ZB and acetylated WPC, respectively. These differences could be due to the longer duration of the outdoor field test (30 months) compared to the laboratory testing (3 weeks). From the weight loss perspective, the cut specimens (1.9±0.3% weight loss) performed better than the uncut control specimens (3.9±0.9% weight loss). This could be due to the method of processing (deck board size vs extruded field stake), the density/hardness of the WPC materials, the accessible fiber material on the surface, or the amount of lubricant (needed for the extrusion process) on the surface.

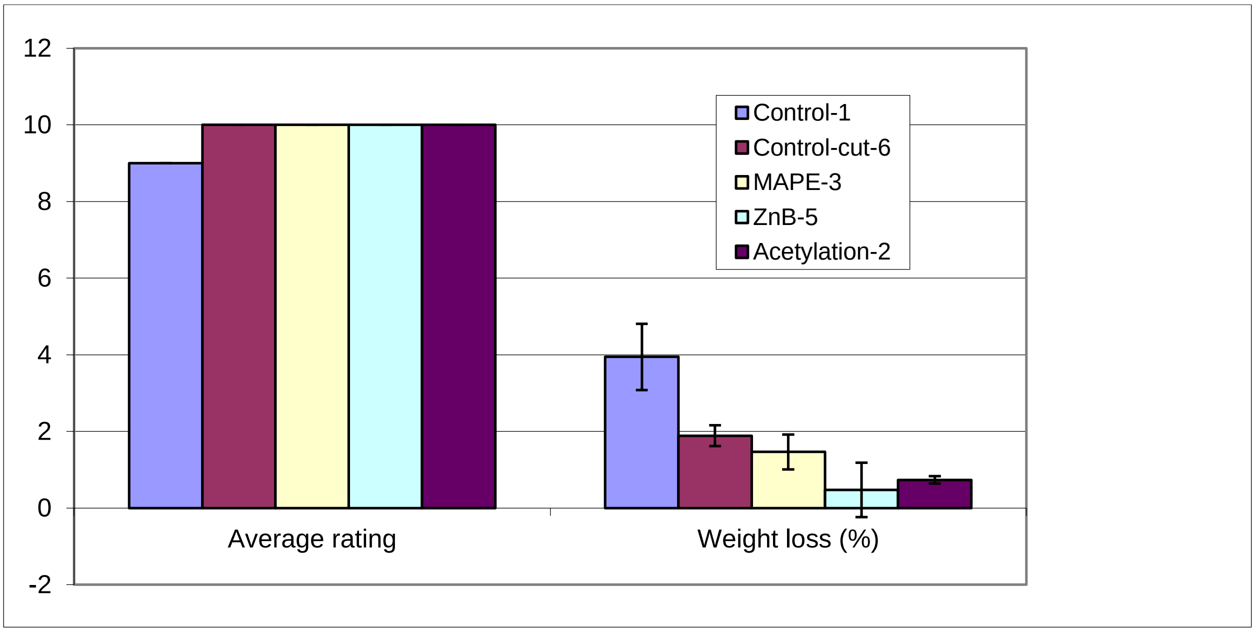

Figure 4: Field test results after 30 months exposure to *C. curvignathus*. (Not shown: Solid wood pine specimens had an average rating of 0.9±1.91, and 92.2±16.8% weight loss.)

Visual ratings and weight loss of the WPC blends after 30 month field exposure to *M. gilvus* are shown in Fig. 5. In contrast to the *Coptotermes* field test, there was significant termite damage in all the WPC blends, except for the ZB (3.9±1.4% weight loss) and acetylated (0.8±0.5% weight loss) WPC blends. All the WPC specimens were blended from HDPE plastic and wood flour so that the materials were mostly homogeneous. The WPC specimens were attacked randomly from the surface to the inner part but not too deeply. Termites do not feed on the plastic HDPE, they are just looking for the cellulose in the wood flour, and therefore, 100% solid plastic stakes are not attacked.

The WPC blend made with 1% ZB performed well against *M. gilvus* (visual rating of 9 and 3.9±1.4% weight loss). The mechanism of effectiveness for ZB is toxicity. Depending upon the specific termite and field exposure conditions, it was found that higher levels of ZB (up to 3%) may be necessary for long term protection (López-Naranjo et al. 2016, Tascioglu et al. 2018). The 1% ZB level used in this study was effective for the 30 month testing period, but may need to be increased for longer exposures.

The WPC blend made with acetylated wood flour performed very well against *M. gilvus* (visual rating of 10 and 0.8±0.5% weight loss). These results are similar to our in-ground field studies located on the Harrison Experimental Forest in Saucier, MS, USA. In this prior study, the MAPE and acetylated WPCs showed no termite attack on any of the WPCs after 2 years 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 average rating for MAPE specimens was 7.6±1.8 and the control was 8.6±0.9. The acetylated WPC stakes showed no termite attack after 7 years with a rating of 10 (Ibach and Clemons 2017).

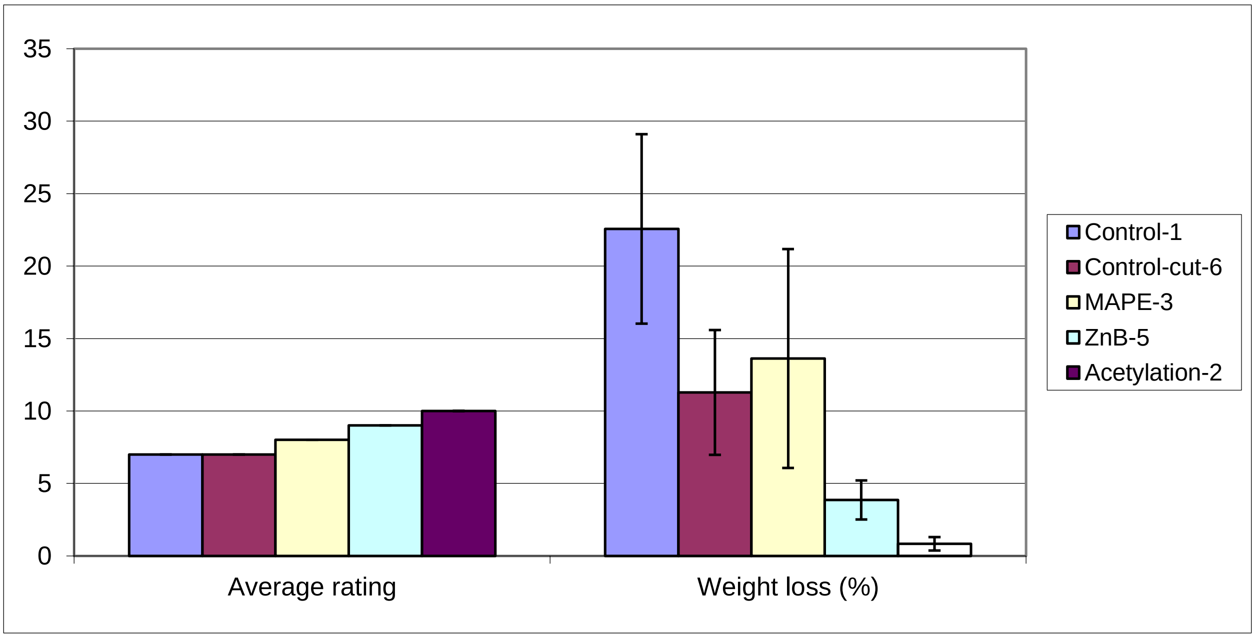

Figure 5. Field test results after 30 months exposure to *M. gilvus*. (Not shown: All solid wood pine specimens had average rating of 0 and 100% weight loss after 30 months.)

Acetylated solid wood has been shown to impart excellent protection against termite attack if there is a high degree (about 20% weight gain) of acetylation (Rowell 2006). The acetyl content used in this study was 22.0±0.01% and the unmodified flour was 2.3±0.09%. The mechanism of termite resistance of acetylated wood may be the result of several factors: 1) reducing the equilibrium moisture content (EMC) below that needed for attack; 2) increasing the hardness; 3) modifying the typical nutrients, such as hemicellulose acetate and lignin acetate resulting in no recognition message; and/or 4) the actual digestion of the wood is done by bacteria living inside the termite and since the acetylated wood is resistant to attack by microorganisms, the termites may graze but do not attack the acetylated wood (Gascón-Garrido et al. 2013, Rowell 2014). Further investigation is needed to determine if the mechanism of effectiveness of acetylated wood flour is due to lowering the moisture of the WPC specimen, or if the acetate group renders the wood flour component unrecognizable as a food source for the termites.

When performing termite testing of WPC in the laboratory, modification of the standard solid wood protocols may need to be considered when comparing various formulations due to the slow moisture sorption of WPCs. Weight losses in the laboratory were less than the field tests, but the field tests could also include weight loss from fungal decay and weathering.

4. CONCLUSIONS

Preliminary results from this study suggest that:

- Laboratory testing was less discriminating than field tests for WPCs. This was because lab tests were performed as no-choice tests while termites do have a choice in field tests. Future work should include running laboratory choice tests on WPCs and conditioning specimens in water.
- Control WPCs have a higher termite protection level than control solid southern pine.
- In field tests, WPC blend performance was as follows:
  Acetylated WPC > Zinc Borate >> Coupling agent, Untreated WPC Controls, both cut and uncut surface
- Future work should include more controlled investigations to verify these preliminary results and to help elucidate mechanisms of termite feeding resistance.

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