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# Effects of chemical preservative treatments on durability of wood flour/HDPE composites

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ABSTRACT

The main objective of this study was to determine the durability of chemical preservatives treated wood flour/high density polyethylene (HDPE) composites to white-rot fungus (*Coriolus versicolor*). Specimens, containing treated and untreated poplar wood flour (PF, 60%), were mixed with HDPE (38%) as polymer matrix and maleic anhydride grafted polyethylene (MAPE, 2 wt.%) as coupling agent. Two fungicide materials, namely 3-iodo-2-propynyl butylcarbamate (IPBC, 0.3, 0.6 and 0.9 wt.%) and 2 thiazol-4-yl-1H-benzoimidazole (TBZ, 0.3, 0.6 and 0.9 wt.%) were used in preparation of wood plastic composites (WPCs). Then, treated and untreated composites were exposed to the fungal decay for 12 weeks according to the European Union (EN) 113 standard. Mechanical and physical properties of the composites were evaluated before and after fungal incubation. The experimental results indicated that treated composites were more resistant to decay, with strength losses significantly lower than the untreated (control) sample. Physical properties in terms of water absorption and thickness swelling were improved by the incorporation of fungicide agents, but no significant differences were observed between the treaded samples. Weight losses for the various treated composites ranged from 1.1% to 4.5%. In addition, IPBC treated samples showed slightly lower weight loss compared with the treated composites with TBZ. The highest weight loss corresponds to the control. Accordingly, IPBC and TBZ can be effectively used as preservatives for WPC. However, IPBC showed superior results compared to the TBZ and it is recommended for the WPCs preservation.



## 1. Introduction

The term “wood plastic composite” (WPC) refers to any composite that contains wood and thermoplastics or thermosets. WPC products are commonly substituted for solid wood in today’s building structures. The objective of WPC development is to produce a product with performance characteristics that combine the positive attributes of wood and plastics [1]. The combination turns out to be a whole new material with improved characteristics and properties compared to the characteristics of the individual components of the composite [2]. The wood fibers offer a combination of attractive properties such as low density, renewability, biodegradability, wide availability, and low cost, which make them alternatives to traditional synthetic fibers in many applications. However, the disadvantages of using wood fibers are their low bulk density, low thermal stability, high tendency to absorb moisture, and susceptibility to biological degradation [3]. Plastics are generally resistant to fungal attack; however a major concern with these materials is the wood in the WPCs. Initially, it was presumed that thermoplastic material completely encapsulated the wood component of the composite, protecting it from wetting and further decay, but a number of tests suggest that wood encapsulation by polymer matrix is incomplete [4,5]. As a result, the wood component in WPCs reaches moisture levels suitable for fungal attack [2,6,7]. Morris and Cooper [8] were the first to prove the presence of fungal decay and discoloration on WPC decking material in service. Laks et al. [9] reported that WPCs containing more than 50% weight ratio of wood particle were degraded by brown rot and white-rot fungi.

There is little data on decay patterns or effects of fungal degradation on physical and mechanical properties of WPCs, although new reports are emerging in this rapidly expanding area [10,11]. In general, fungal decay has a negative effect on the mechanical and physical properties of WPCs. A number of researches have focused on the properties of such composites as their applications are expanding [4,5,12–14]. For example, Nadali et al. [15] reported that the mechanical properties of WPCs including bending strength, elastic modulus, and hardness decreased due to exposure to rainbow fungus. Verhey et al. [16] conducted a research on the variations of mechanical properties of composites made of wood fibers – thermoplastic polymer as the result of exposure to white

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and brown-rot fungi. They concluded that because of contact with the studied fungi, the mechanical properties of the composites decreased. The protection of WPCs against fungal decay is reported to be possible through the addition of anti-fungal agents [4,9,17]. Therefore, most of WPCs are being treated with wood preservatives during their service life to protect them from biological deterioration [7,18]. The selection of anti-fungal agents to protect a particular product, such as WPCs, is based on their specific properties (i.e., the ability to control the growth of a particular fungi and biodegradability). To protect WPCs, commercially available solutions of 3-iodo-2-propynyl butylcarbamate (IPBC) and 2-thiazol-4-yl-1H-benzoimidazol (TBZ) are currently employed. Treatment efficiency may be further enhanced by the use of two or more wood preservatives, such as IPBC and TBZ [19].

To the best of our knowledge, there are a few reports about HDPE composite degradation by *Coriolus versicolor* (white-rot fungus) [7,12,15] and there is no published information on preservation of wood flour/HDPE composites. The main objective of current study is to evaluate the effects of pre-treatment WPCs with the two above-mentioned different fungicide agents against *C. versicolor* attack. It is to be noted that this is one of the most common and dangerous saprophytes that attacks hardwoods all around the world [7]. In addition, physical and mechanical properties of treated and untreated composites were investigated.

## 2. Materials and methods

### 2.1. Materials

*Lignocellulosic material*: waste poplar (*Populus deltoides*) flour (PF) was collected from a local sawmill in Karaj, Iran. The untreated sawdust was milled down to particle size of 60 mesh and then dried at 102 °C for 24 h to less than 3% moisture content. The PF was kept in sealed plastic bag for further use.

*Polymer matrix*: high density polyethylene (HDPE), with trade name of HD5620EA, an injection molding grade was supplied by Arak Petrochemical Co. (Iran), in the form of pellets. Its melting flow index (MFI) was 20 g/10 min (190 °C/2.16 kg), with a density of 0.95 g/cm$^3$.

*Coupling agent*: maleic anhydride grafted polyethylene (MAPE), in the form of powder (grade KJS 111) with a density of 0.92 g/cm$^3$ and a melting flow index of 0.4 g/10 min, was obtained from Kimia Javid Sepahan Co., Iran.

*Wood preservatives*: two fungicide agents, including 3-iodo-2-propynyl butylcarbamate (IPBC) and 2-thiazol-4-yl-1H-benzoimidazol (TBZ) were obtained from Sigma–Aldrich Co., South Korea. The solutions of IPBC and TBZ were prepared by dissolving in ethanol (96%) and dimethyl sulfoxide (DMSO) at room temperature by stirring for 1 h, respectively.

*Purified white-rot fungus*: a Petri dish containing the purified *C. versicolor* was obtained from Alborz Research Institute in Karaj, Iran. Malt extract agar (MEA) was used as the culture medium. The medium was supplied by Merck and was used at a concentration of 38 g/L in all laboratory tests. All other chemicals were of analytical grade and obtained from various sources.

### 2.2. Preparation of composites

In the first stage, the fungicide solutions were sprayed onto PF. The treated and untreated PF, HDPE and MAPE were then premixed based on each formulation (Table 1) before being fed into the extruder. Consequently, the mixtures were compounded in a co-rotating twin screw (Collin) extruder. The barrel temperatures of the extruder were controlled at 160, 165, 170, 175 and 180 °C for zones 1–5, respectively, while the temperature of the die was held at 185 °C. The melt temperature was kept at 185 °C to prevent wood degradation, and screw speed was set at 60 rpm. The extruded strand was passed through a water bath, granulated, and subsequently dried at 105 °C for 24 h to remove any moisture. The granules were then injection molded at 180 °C and a pressure of 6 MPa to produce standard ASTM specimens. After molding, the test specimens were conditioned at 23 ± 2 °C and 50 ± 5% relative humidity for at least 40 h according to ASTM D 618.

**Table 1**
Compositions of the evaluated treatments (wt.%).

| Codes | PF (wt.%) | HDPE (wt.%) | MAPE (wt.%) | IPBC (wt.%) | TBZ (wt.%) |
|---|---|---|---|---|---|
| Control | 60 | 38 | 2 | 0 | 0 |
| $A_1$ | 60 | 37.7 | 2 | 0.3 | 0 |
| $A_2$ | 60 | 37.4 | 2 | 0.6 | 0 |
| $A_3$ | 60 | 37.1 | 2 | 0.9 | 0 |
| $B_1$ | 60 | 37.7 | 2 | 0 | 0.3 |
| $B_2$ | 60 | 37.4 | 2 | 0 | 0.6 |
| $B_3$ | 60 | 37.1 | 2 | 0 | 0.9 |
| $A_1B_1$ | 60 | 37.4 | 2 | 0.3 | 0.3 |
| $A_3B_3$ | 60 | 36.2 | 2 | 0.9 | 0.9 |

### 2.3. Inoculation of composite specimens

The inoculation of composite specimens was carried out according to the procedure of European Standard EN 113 [20]. The purified fungus was transferred to Petri dishes containing MEA. The dishes were then kept in the laboratory at 25 °C for 1 week until the culture medium was fully covered by the fungus. The cultured fungus was then transferred into Kolle flasks containing the culture medium that was incubated for 1 week at 25 °C. To prevent direct contact of the specimens with the culture medium, the specimens were mounted over two 3-mm platforms and were placed in the Petri dishes. The dishes containing the fungus and the specimens were then stored in an incubator for 12 weeks at 25 °C and 75% relative humidity (Fig. 1). After the exposure period, the specimens were carefully removed from the dishes, the mycelium was brushed off the surfaces with a soft sponge, and they were oven-dried at 103 °C before testing.

### 2.4. Testing

#### 2.4.1. Mechanical properties

All specimens were tested following ASTM D 790 [21] for three-point static bending to obtain modulus of elasticity (MOE) and modulus of rupture (MOR), and D 256 [22] for notched Izod impact (NI) strength. Bending tests were conducted using an Instron Universal Testing Machine (model 4486) at the crosshead speed of 8 mm/min. The dimensions of specimens for MOE and MOR were 115 mm × 13 mm × 3.2 mm. An Instron impact tester (model Wolpert PW5) was used for the Izod impact tests. For each treatment, 4 replications were conducted.

#### 2.4.2. Physical properties

Physical properties, namely water absorption (WA) and thickness swelling (TS) were tested in accordance with ASTM D 570 [23]. Before the testing, the weight and thickness of each specimen (5 mm × 1 mm × 0.8 mm) were measured. Conditioned samples of each type of composite were soaked in distilled water at room temperature for 24 h. Samples were removed from the water, patted dry and then measured again. For each treatment, four samples were tested.

#### 2.4.3. Property losses

Changes after fungal attack for each physical and mechanical property of the composites served as good indicator of the effect