COMPARISON OF RESISTANCE AND BIODEGRADABILITY PROPERTIES OF WOOD-PLASTIC COMPOSITES FROM WOOD FLOUR/PHB/HDPE/STARCH

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Abstract. In this study, mechanical and biodegradability properties of wood-plastic composite were investigated. Beech wood flour (WF) 40 wt% was used as a reinforcing base material, maleic anhydride (MA) as a coupling agent, and nano clay to improve the properties were added. The polymer studied was polyhydroxybutyrate (PHB) and the other polymer was high-density polyethylene (HDPE). Three groups of composites were produced, in two groups each of the polymers alone and in the third group a combination of two types of polymers was used as a matrix. Starch 8 wt% and 12 wt% was used instead copolymer. The specimens were mixed using a twin-screw extruder, made with an injection-molding machine, and subjected to mechanical tests: tensile strength and modulus, bending strength and modulus, impact resistance tests and
biodegradability tests. In mechanical properties, it was observed that wood-plastic composite with PHB base material has lower resistance properties than composites containing HDPE. With the addition of starch, all resistance properties were significantly reduced compared with the control sample without starch in all three groups of composite samples. Starch could not play the role of copolymer well in any of the groups. The third group of samples showed better results in terms of mechanical strength resistance properties than the first group. Tensile strength and Modulus, bending strength and Modulus, impact resistance increased compared with the first group. In the biodegradability test, the samples were buried at a height of 25 cm for three months. Weight loss was due to the destruction of WF, starch, and natural polymers by soil microorganisms. The weight loss trend of the samples was increasing until the end of the second month and then decreasing.

**Keywords:** Wood-plastic composite, biodegradability, PHB, HDPE, starch.

### INTRODUCTION

Reinforced plastic composites are a class of composites in which high-modulus fibers are added to a polymer matrix to overcome the low modulus and thermal instability of the plastic. However, today they use organic fillers such as lignocellulosic fibers. These low-cost materials are low weight, renewable, available, and biodegradable. On the other hand, the use of these composites is a good alternative to wood and polymeric materials in different applications. With the increase of environmental awareness and interest of the international community and new environmental and oil consumption regulations, the use of environment-friendly materials has increased, and this is also true for wood-plastic composites (Casarin et al 2017).

To prevent environmental damage, high cost savings, and replacement of synthetic polymers, natural polymers were used in composite fabrication. In this regard, we examined the production of wood-plastic composites based on natural polymer. The largest group of biodegradable polymers are polyhydroxy alkanoates (PHAs). Despite the higher cost of production, these materials are probably much cheaper than traditional plastics because the cost of environmental degradation and postproduction recycling costs have never been calculated (Ienz and Marchessault 2005; Chen et al 2015). PHAs are generally made up of a subunit called β-hydroxy alkanoate by a simple pathway with three enzymes of acetyl coenzyme A, the most famous of which is polyhydroxybutyrate (PHB). PHB is a short-chain biological of PHA. Its advantages include availability, biocompatibility, and physical properties comparable to plastic thermoplastics (Isola et al 2017). This polymer is a good option for making the target composite (Chan et al 2020). Investigated the mechanical stability of PHA-based wood-plastic composite and used PLA/wood flour (WF) and polyethylene (PE)/WF composites as a comparison reference. PHA-containing biocomposites were degraded when continuously submerged, unlike PE- and PLA-containing composites. The specimens were mechanically stable indoors but a decrease in stability in mechanical properties was observed outdoors. Nikushi et al (2018) investigated the improvement of compatibility of physical-mechanical properties of polymer composites based on natural materials and biodegradability. They used polylactic acid (PLA), PHB, PHB with polyhydroxyvalerate copolymer (PHBV), bioflex (PLA blend) and solanyl (Starch-based). Biocomposites show poor mechanical properties due to the weak bond between the fibers and the matrix.

Vandi et al (2019) produced PHBV-based plastic wood composite by extrusion method and investigated its mechanical properties. In the mechanical properties of the composite containing 40% of wood, it showed higher strength, which showed a 73% increase in modulus and 80% in tensile strength compared with pure PHBV. Panaitescu et al (2020) investigated the thermal and mechanical properties of PHB composite and cellulose from wood waste. Composites showed better thermal stability than PHB. In composites with 5% fiber content, 13% and 10% fiber content increased by 25%. The highest rate of increase in the composite was 10% fiber, ranging from 25% at room temperature to 90% at 125°C. The best mechanical and thermal properties of the composite compared with PHB were when lignin was
present on the fiber surface. Gunning et al (2019) produced PHB-based biocomposites and three types of natural fibers by extrusion and compression injection. PHB was evaluated with hemp, jute, and lyocell with 10 wt% to 30 wt%. This study was performed to investigate the effect of fibers on the matrix. Tensile properties and impact resistance showed a significant decrease. Adding 30 wt% of hemp and jute fibers changed the modules. This value was very high in the flexural modulus. A value of about 591% was observed in hemp and 246% in jute compared with pure PHB. On the other hand, PHB/Lyocell composites showed the lowest flexural modulus. In total, the modulus values increased with fiber loading for all types of fibers.

Casarin et al (2017) to investigate the biodegradability, they made two types of composites: a composite with PHB/WF (80/20) composition and a composite with combination of PHB/fibers of sisal plants (80/20). They produced extruded plastic pipes by injection. Samples were placed in three degradation test devices for 30, 60, and 90 da. At the end of each experiment, the samples were weighed to measure the degree of degradation. Based on the results, it was observed that all samples lost their mass during the degradation test. Both types of composites lost more weight than pure PHB because of the adsorption of water due to the presence of natural fibers, which in turn creates a faster rate for PHB degradation. In flexibility tests, the modulus of elasticity decreased after composite degradation. After 30 and 60 da, the modulus of elasticity did not change much, but after 90 da, the amount of modules decreased by half. This indicates that the destruction is slow and accelerates over a period. Nicoleta Frone et al (2020), investigated the morphological structure, thermal and mechanical properties of biodegradable PLA/PHB/nanocellulose (NC) nanocomposites. They used three methods of injection, extrusion, and three-dimensional (3D) printing. The composites were fabricated by a combined single-step reaction process using dicumyl peroxide (DCP) as a bonding agent. They used plum seed shells to produce cellulose nanocrystals. Nanocrystals with a diameter between 30 and 80 nm were used. The results showed that with the addition of DCP the surface adhesion improved. The carbonyl index was calculated by infrared (IR) spectroscopy, which showed an increase in crystallinity after DCP was added to the composite mixture. This was confirmed by differential calorimetric scanning. The presence of NC and DCP caused nuclear activity and increased PLA crystallinity. PLA/PHB composite crystallinity increased from 16% to 38% and NC/PLA/PHB composite crystallinity up to 34% because of the addition of DCP, which increased crosslinking. In addition, DCP, due to low molecular weight product production increased with mobility, also affects melting processes and recrystallization. In the study of production method, the values of higher storage modulus were for extrusion films, 3D scan, and then injection mold, respectively. The best thermal stability and high degradation temperature belonged to biocomposites containing DCP. Torres-Tello et al (2017), used agave plant to produce green biocomposite with two types of polymers PHB and PHBV. Biocomposites were produced by compression molding with two purposes, first to reduce the amount of biopolymer used (because of high cost) in the final product and second to improve the mechanical properties of the material. To evaluate the effect of fiber content, 10 wt%, 20 wt%, and 30 wt% were used to prepare the composite. The results showed that the addition of fiber at 30 wt% in both matrices increased the tensile modulus by 80% compared with PHB and by 30% compared with PHBV and also the bending modulus by about 36% and 41% for both composites, respectively, shows. In addition, tensile strength and flexural strength did not have a negative effect and the impact strength increased significantly, which was 44% and 66% for the two matrix, respectively. Koller and Owen (1996) investigated the structure and mechanical properties of PHB/corn starch and PHB/HV copolymer. To make the samples, they used the compression molding method at 190 degrees. In the PHB/starch, composite, brittle cracks were observed below 1% of the pressure, indicating that the starch fails in the PHB, reduces the strength and increases the modulus. In another
part of composite fabrication, starch was treated with water, heat, and cut. This starch found better properties than the previous starch grains. However, there was not much change in the composite. Bledzki and Jaszkiewicz (2010) investigated the mechanical properties of natural fiber-reinforced PLA and PHBV composites in comparison with polypropylene (PP).

They used cellulose, jute, and abaca fibers plant as reinforcing fibers. Samples were made using injection molding. Resistance tests were performed to evaluate the mechanical performance of the composites. Electron microscopy was also used to examine the adhesion between the fibers and the matrix. In other samples, PP-containing composite was made with the same fibers studied for comparison. Fiber reinforcement significantly increased stiffness and tensile strength. However, depending on the type of fiber, there were differences in mechanical properties. Impact strength and tensile strength for composites containing cellulose fibers; the highest value was seen. In scanning electron microscope (SEM) images, matrix fiber bonding in PLA and PHBV composites was very different from PP composites. These bonds were much weaker in PHBV because of the very crystalline structure of PHBV. As a result, the bonding phase between the fiber and the matrix is reduced. As a result, the fibers are not well embedded in the matrix and a weak bond is created. The best fiber dispersion in the field belonged to Abaka fibers.

The purpose of this study is to use PHB to create the biodegradability of the composite after the service period for recycling in the natural cycle and to investigate the effect of the starch and combination of PHB and HDPE to increase the strength properties and degrade the composite after waste.

### MATERIALS AND METHODS

PHB made by the British company Good fellow and HDPE purchased from Shazand Petrochemical Company of Iran was used as the base material. PHB was in the form of granules with a size of 5 mm. Persian beech flour was used as a matrix reinforcing fibers. The starch was purchased from the German Merck brand under the brand name starch 10125 soluble GR, Maleic Anhydrid as a coupling agent graft with polyethylene (Pe-g-MA) from Aria polymer pioneer under the brand name Aria couple 1141 as compatible and nanoclay with grade 20A of the company American Saturn was produced under the brand name Cloisite 20A. In this research, extrusion and injection molding methods were used to make samples. The WF was placed in an oven at 90°C for 24 h to dry. The studied samples were made in three different categories in terms of composition and each group had 12 samples with different percentages of materials (Table 1).

To make each sample, a specified combination of raw materials and additives was alternately poured into an extruder with twin-screw nonaligning. The extruder has six areas for heating from 145 to 165 degrees with a distance of one temperature every 5 degrees. The rotation speed of the device was 60 RMP. Ingredients of the samples were hot extruded out of the extruder and after cooling were milled to become suitable granules for the injection machine. The raw materials were turned into test samples by injection machine. The samples were made according to the standard and tested. Beech flour was used as a matrix reinforcer in 80-mesh particle size with 40 wt%. Starch with an 8 wt% and 12 wt% was used as a copolymer. To improve the strength properties of 1 wt% nano and to improve the bonds of polymer and WF, 5 wt% maleic anhydride (MA) was used.

### Mechanical Tests

Tensile strength and modulus test, bending strength and modulus test, and impact resistance test were performed. The mechanical performance of the composites was examined. Table 1. Percent weight of sample compositions. Beech WF/ MA/ PHB/HDPE/nano clay/starch.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>40%WF/5%MA/8%starch/1%nano/46%PHB</td>
</tr>
<tr>
<td>2</td>
<td>40%WF/5%MA/12%starch/1%nano/42%PHB</td>
</tr>
<tr>
<td>3</td>
<td>40%WF/5%MA/8%starch/1%nano/46%HDPE</td>
</tr>
<tr>
<td>4</td>
<td>40%WF/5%MA/12%starch/1%nano/42%HDPE</td>
</tr>
<tr>
<td>5</td>
<td>40%WF/5%MA/8%starch/1%nano/46%PHB+HDPE</td>
</tr>
<tr>
<td>6</td>
<td>40%WF/5%MA/12%starch/1%nano/42%PHB+HDPE</td>
</tr>
</tbody>
</table>

WF, wood flour; MA, maleic anhydride; PHB, polyhydroxybutyrate; HDPE, high-density polyethylene.
performed according to ASTM D638, ASTM D790, and ASTM D250 standards, respectively. The results are shown in Table 2. The test of each sample was repeated three times.

According to the results (Table 2), a decrease in strength, especially in tensile strength, was observed in samples containing PHB alone. With the addition of HDPE to the samples, all resistances increased. This is because of the mixing of two polymers in combination with WF. HDPE by adding to the composite composition increases the strength and reduces the weakness and brittleness of PHB. However, the presence of WF is effective in improving the strength. Starch and its percentage increase showed a noticeable decrease in strengths, which produced weaker composites than nonstarch samples. Compared with mechanical properties, composites containing 1% nanoclay showed better mechanical properties than pure PHB. Addition of nanoclay to the polymer matrix increased the degree of crystallinity and increased the amorphous degree of the semicrystalline PHB.

**Biodegradability Test**

This test is one of the main objectives of this study. Soil burial test was performed according to ASTM G160 standard. The samples were buried at a height of 25 cm for 3 mo. The soil used is the soil of Aradkooh waste processing and disposal complex located at 23 km of the old Tehran-Qom road. This soil was collected in the required amount and transferred to the test environment under natural environmental conditions. The required amount of soil was poured into 12 plastic boxes with dimensions of $18 \times 35 \times 50$ cm. The samples were placed in each box according to the group and the time period of getting out of the soil. Soil characteristics of this center: 3% granules, 30% sand, 45% silica, and 22% clay.

The samples were placed horizontally in the soil and after certain periods, the first 2 wk, the first month, the second month, and the third month were taken out of the soil and cleaned with a soft brush to completely remove dust from their surface and then weighed. The results of each weighing step and the amount of weight loss were recorded. This weight loss is because of the destruction of WF, starch, and natural polymers by microorganisms that use these substances as food. In the samples of the second group, which contained HDPE as a synthetic polymer, the degradation was very small, which was done in WF and starch, and the polymer was not degraded, but in the first and third groups, because of the presence of PHB, in addition to WF and starch, weight loss and destruction also occurred in PHB. The amount of destruction in the first group was more than all groups. In the first 2 wk, the destruction took place slowly, but after a month, the percentage of destruction accelerated and after 2 mo, it decreased. The results are given in Tables 3 and 4.

**Weathering Test**

Wood-plastic composite samples were transferred to the weathering tester for weathering test. In this device, according to the standard, the samples were exposed to moisture and light rays and were tested during different hours as shown in the results diagram (2000, 1000, 500, 250 h). In the early stages, small but large cracks were seen on the surface of the samples, which due to ultraviolet (UV) radiation and exposure of the samples to

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**Table 2. Samples mechanical resistances (MPa).**

<table>
<thead>
<tr>
<th>N</th>
<th>Tensile strength</th>
<th>Tensile modulus</th>
<th>Bending strength</th>
<th>Bending modulus</th>
<th>Impact resistance</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>14.8</td>
<td>3321.5</td>
<td>21.7</td>
<td>1451.6</td>
<td>45.3</td>
</tr>
<tr>
<td>2</td>
<td>12.6</td>
<td>3301.09</td>
<td>18.87</td>
<td>1364.7</td>
<td>41.2</td>
</tr>
<tr>
<td>3</td>
<td>29</td>
<td>3633.6</td>
<td>36.1</td>
<td>1945.4</td>
<td>70.1</td>
</tr>
<tr>
<td>4</td>
<td>26.3</td>
<td>3477.3</td>
<td>27.86</td>
<td>1773.4</td>
<td>64.12</td>
</tr>
<tr>
<td>5</td>
<td>21.6</td>
<td>3385.4</td>
<td>27.3</td>
<td>1582.3</td>
<td>65.23</td>
</tr>
<tr>
<td>6</td>
<td>20.13</td>
<td>3314.1</td>
<td>26.1</td>
<td>1502.1</td>
<td>58.4</td>
</tr>
</tbody>
</table>
moisture caused the wood fibers to swell and create surface cracks. In the later stages, the cracks increased and were larger because of radiation. Light degradation occurred in cellulose, hemicellulose, and lignin. Because the samples also contained starch, degradation also occurred in starch. Starch swells and bursts because of the presence of glucose units due to water and heat and their semicrystalline structure is destroyed so that under weathering conditions the starch in the samples is destroyed, Griffin (1994). However, its sensitivity to moisture and poor mechanical stability are the main disadvantages when compared with other commercial bioplastics, which increased with increasing weathering time. As a result, the polymer chains in the inner layers are also broken and shortened and degradation occurs in the crystalline region. Therefore, with the increase of weathering time, the amount of degradation increased. This degradation was more in the first group. In this group, PHB was degraded earlier because of its fragile structure. The results can be seen in Figs 1-5.

RESULTS AND DISCUSSION

In this study, natural PHB polymer was used to investigate the strengths and biodegradability of composites. Starch was used instead copolymer to compare and create a new combination of HDPE, and MA was added as a coupling agent and nano was added to improve the resistance properties and beech WF 40 wt% was added to reinforce the matrix. One of the main bottlenecks of PHB/lignocellulosic fibers biocomposites is the poor affinity between lignocellulosic fibers and PHB resulting in limited tensile properties. Natural fibers have polar and hygroscopic properties, whereas plastics are mostly nonpolar and hydrophobic Therefore, because of the molecular structure, there is no bond between them. A coupling agent is used to make the connection. MA has a polar end and a nonpolar end that forms an ester bond from the hydrocarbon head by nonpolar bonding to the matrix and from the other end by carboxylic groups with hydroxyl groups maleic anhydrid as a coupling agent graft with polyethylene (MAPE) connects natural fibers with polymer. In previous research, various researchers have reported the properties of nanoclay on the increase of mechanical strengths in wood-plastic composites except for impact resistance (Wang et al 2006; Chowdhury et al 2006; Wu 2007; Han et al 2008; Abdous et al 2010). Nanoclays are layered. These layers can be quadrilateral or octagonal, which are attached to
the raw materials in three ways and to the other layers of nanoclay in one way. Because of the plate structure, they are placed between the raw materials and bond with van der Waals and hydrogen bonds, increasing the mechanical properties.

In mechanical properties, it was observed that plastic wood composite with PHB base material, which is a family of natural polymers, has lower strength properties than synthetic polymer base composites containing HDPE. With the addition of starch, all resistance properties showed a significant decrease compared with the control sample without starch in all three groups of composite samples. Starch could not play the role of copolymer well in any of the groups. The internal bonds between the polymer and the WF were weakened and in some cases prevented from forming a proper bond. Therefore, in terms of tensile strength, the composite showed low mechanical strength. The results showed poor mechanical properties because of insufficient adhesion and nonbinding effect and heterogeneous morphology of PHB/starch composition. This has been stated in other studies (Innocentini-mei et al 2003; Thire et al 2006; Jorres-Giner et al 2018; Gunning et al 2019).

This weakness was also seen in the second group, which did not contain PHB. PHB has a high crystallinity structure. When a composite is made due to melting, its structure changes to amorphous, which is why the bonds it make with other raw materials, are weak. These bonds are nonchemical. After cooling the resulting composite, the PHB structure returns to the crystalline state and as a result, the composite shows poor mechanical properties. As a result, the bonding phase between the fiber and the matrix is reduced. We divided the polymer 50 wt%-50 wt% between PHB and HDPE. In this way, we can form a stronger internal structure and stronger bonds. PHB breaks down quickly because of its brittle structure and has less tensile strength. Combining HDPE with a composite containing PHB and spreading it during the initial melting in different parts of the composite creates stronger bonds and increases the strengths.

Accelerated weathering test was performed on the samples by Atlas Xenon. The accelerated weathering process was performed for 2000 h. In the early stages, fine cracks were seen on the surface of the samples; in the later stages, the cracks were larger and larger in size. In general, the rate of water absorption and the rate of biodegradation of polymers are the main factors controlling the mechanical stability over time, so the composite is constantly moving toward the loss of mechanical properties. These factors cause cracks and crevices, which in turn weaken the performance of the material under mechanical load. The
composite showed a reduction in mass by weathering exposure. The molecular weight of PHB polymer and WF decreased steadily with increasing weathering.

During the biodegradability test, the samples were removed from the soil for 2 wk, 1 mo, 2 mo, and 3 mo after burial in the soil. Weight loss is because of the degradation of WF, starch, and natural polymers by microorganisms that use these substances as food. The weight loss trend of the samples was increasing until the end of the second month and then decreasing. With the addition of nano to the samples, the interlayer distribution of nanojoints improved. In composite specimens, the polymeric materials and additives are more compact, so that PHB and HDPE combine to form better fibers.

CONCLUSIONS

In mechanical properties, it was observed that wood-plastic composite with PHB base material has lower resistance properties than composites containing HDPE. With the addition of starch, all resistance properties were significantly reduced compared with the control sample without starch in all three groups of composite samples. Starch could not play the role of copolymer well in any of the groups. The third group of samples presented better results in terms of resistance properties than the first group. All mechanical properties increased compared with the first group. In the biodegradability test, the samples were buried at a height of 25 cm for 3 mo. Weight loss was because of the destruction of WF, starch, and natural polymers by soil microorganisms. The weight loss trend of the samples was increasing until the end of the second month and then decreasing.

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