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# Degradation in the mechanical and thermo-mechanical properties of natural fiber filled polymer composites due to recycling

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## HIGHLIGHTS

- Wood flour and high density polyethylene based composites (WPC) can be efficiently recycled.
- Six reprocessing cycles showed relative decrease in strength and stiffness properties of WPC.
- The strain properties of WPC increased with recycling.
- The crystallinity of WPC decreased after recycling.
- The thermal stability of composites slightly increased after six reprocessing cycles.

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## ABSTRACT

To manage the plastic waste, recycling is recognized as the most environment-friendly and non-destructive method. The aim of this research is to investigate the recyclability of oak wood flour (WF) filled high density polyethylene (HDPE) composites. Two different composite formulations (30 and 50 wt% filler) were considered, each with 3 wt% coupling agent maleic anhydride (MA). Both composites were individually reprocessed six times by extrusion. Test samples were injection molded, to measure mechanical and thermo-mechanical properties. Fiber length measurement and gel permeation chromatography (GPC) were performed respectively to examine the change on fiber length and molecular weight of polymer. Differential Scanning Calorimetry (DSC), Thermogravimetric Analysis (TGA), Scanning Electron Microscopy (SEM), and Fourier Transform Infra-Red Spectroscopy (FTIR) were also carried out to better understand the impact of recycling on composite properties. After reprocessing six times, WF-HDPE composites showed relative decrease in strength and stiffness properties and slight increase in strain properties as compared to corresponding virgin composite. The strain properties saw an increase in their value with recycling. The crystallinity of HDPE decreased but thermal stability of the composite increased with reprocessing.

## 1. Introduction

Wood Plastic Composites (WPCs) are typically manufactured from two fundamental constituents a natural fiber or filler, and a thermoplastic resin [1,2]. Nowadays, WPCs are widely accepted as building materials especially because of their durability, higher specific strength and stiffness, and zero or very low health concern along with other advantages [3–5]. Among all WPCs, wood fiber or filler (WF) based high density polyethylene (HDPE) composites are extensively used in household apparatus (e.g., doors, decking, windows, railing, and furniture), and automotive industry (e.g., door panels and seat covers) [6]. HDPE is preferred because it has a lower melting point around 130 °C as compared to the degradation temperature of most natural fibers that varies between 200 and 220 °C, which helps in processing of its composites [6]. HDPE also shows higher toughness, stiffness, chemical resistance, thermal stability, and electrical insulation [7]. For improved properties incorporation of WF in HDPE becomes necessary since elastic modulus and strength of WF, in general is 40 and 20 times respectively higher than that of HDPE [8,9]. WF is also considerably cheap since a substantial amount of wood waste is generated in wood industry [10].

Along with a lot of advantages, WF-HDPE composites come with some imperfections. The major problem is the incompatibility between the fiber and matrix. Natural fibers are hydrophilic while thermoplastic polymers are hydrophobic. This incompatibility causes poor interfacial adhesion between fiber and matrix when

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mixed together. Poor adhesion leads to less stress transfer from matrix to fibers that finally results in lower mechanical and thermal properties of the WPC. However, this problem could be resolved by adding specialty additives such as coupling agent (CA) or compatibilizers in the composites [11,12]. The CA helps to improve the interfacial adhesion that leads to improved bonding in the interface region. For improved properties of WF/HDPE composites, maleated polyethylene (MAPE) has been reported the better CA compared to other CA such as maleated polypropylene (MAPP) and carboxylated polyethylene (CAPE). This is due to the better wetting and higher compatibility of MAPE in HDPE polymer. In general, the typical amount of MAPE in the WF/HDPE composite-ranges from 1.5 wt% to 4.5 wt%. However, 3 wt% of MAPE has been reported the optimum level of CA for improved properties of WPCs made of HDPE polymer [3,7]. That is why incorporation of 3 wt% of MAPE has been considered for WF/HDPE composites in this research.

Another way to solve this problem is to separately modify the fiber surface and polymers, especially with silane CA. Surface modification of natural fibers improves the fiber surface functionalization for better chemical bonding and increases the fiber surface roughness for better mechanical interlocking. Silane treatment of polymers improves the chemical resistance and physico-mechanical properties of the polymers [13–15]. Therefore, a composite formulated by silane treated natural fibers and polymers also have higher thermal, chemical, and mechanical properties.

However, one major issue with WPC's are that they are not fully biodegradable at the end of their useful life and can raise an environmental concern if discarded in the landfills [1,16]. Plastic waste can cause soil and air pollution. All petroleum based plastics take a very long time to degrade in soil and impact the soil fertility [17]. In addition, plastic waste-floating on sea surface-reduce the sea life in a greater amount. Because of these environmental concerns, plastic waste management is now a very important issue all over the world.

There are several traditional methods that are used for plastic waste management. But most of these methods are detrimental to environment and destructive to material. For example, when disposed in landfills, plastic waste creates soil and air pollution; and when incinerated, this waste creates noxious gases that helps global warming [18]. On the contrary, recycling is the most environment friendly and undamaging process that can help to reuse the material without significant degradation in properties. The other advantages of recycling is the fluctuating price of crude oil based plastics which can add risk to pricing strategy. In addition, the need for landfills for discarding plastic is an environmental concern [18].

Considering all these advantages, recycling is acknowledged as best alternative for WPC waste management. To date, a lot of research have been done on the recycling of WPCs but the information is somewhat conflicting. Lei et al. added pine or bagasse flour to recycled HDPE, and concluded that the mechanical properties of the recycled composites compared fairly well with virgin composites [3]. Bourmaud et al. recycled virgin WPCs of PP by injection molding and grinding up to 7 times, and found comparable mechanical properties of recycled WPCs with virgin WPCs [4]. Shahi et al. recycled virgin WPCs of HDPE once by grinding and extrusion, and reported decrease in strength properties but increase in water uptake of recycled WPCs [6]. Adhikary et al. made recycled HDPE/virgin fiber WPC, and reported that the recycled WPCs showed only a marginal change in mechanical and dimensional properties as compared to virgin ones [10]. Augier et al. recycled virgin WPCs of PVC by extrusion and milling up to 20 times, and found that mechanical properties of the composite increased or remained almost constant with ascending no. of reprocessing cycles [11]. Beg et al. reprocessed WF/PP composites by injection molding and grinding up to eight times, and reported that the mechanical properties of the composite decreased with increased number of reprocessing cycles [16]. Petchwattana et al. mixed virgin and post-consumer WPC at a weight ratio of 70:30, extruded that WPC eight times, and reported insignificant degradation in mechanical properties of recycled WPCs [18].

However, it should be noted that the all these studies are not exhaustive and comparable, due to multiple grades of the polymers, type of wood fibers/fillers, grade of coupling agents and their percentage content, presence of other specialized additives, and the manufacturing and recycling processes of the WPCs itself [3]. Since there is a lot of discrepancy in the literature, an in-depth study was conducted to understand the influence of reprocessing on the mechanical and thermo-mechanical properties of oak wood flour (WF) filled HDPE composites containing coupling agent (MAPE) with two different filler loadings. The material was individually recycled up to six times by extrusion, and test samples were injection molded after each reprocessing cycle.

## 2. Experimental

### 2.1. Materials

Oak wood-flour was provided by Southern Wood Services (Macon, GA). It is widely produced by the furniture industry as an operational waste. Wood particle size was in the range of 250 μm–400 μm. The thermoplastic polymer was high density polyethylene (Marlex 9012), manufactured by Chevron Phillips Chemical Company, TX. The polymer has a MFI of 11.5 g/10 min, density of 0.952 g/cm$^3$, and vicat softening temperature of 124 °C. The coupling agent-maleic anhydride grafted polyethylene (MAPE), with density 0.92 g/mL, $T_m$ of 107 °C was purchased by Sigma-Aldrich, St. Louis, MO.

### 2.2. Methods

#### 2.2.1. Composite manufacturing

Fig. 1 shows the manufacturing process of WF-HDPE composites. Composite pellets were manufactured from 30% nd 50 wt% wood flour, HDPE resin and 3 wt% MAPE, labelled at WF 30 HDPE and WF 50 HDPE respectively. The material was processing by using a twin-screw co-rotating extruder (Leistriz Micro 18 GL 40 D, NJ, USA). Test samples were prepared from pellets by using a single screw injection molder (Model SIM-5080, Technoplas Inc., OH). The extruder had seven different temperature zones. The temperature of these seven zones, from feed section to melting section, was 160 °C, 193 °C, 199 °C, 204 °C, 207 °C, 210 °C, and 213 °C respectively. The temperature of the die and gate adapter was controlled at 213 °C. The screw rpm of the extruder was set at 150. Prior to extrusion, wood flour, MAPE, and HDPE were dried in an oven at 80 °C for a minimum period of 24 h to remove moisture. The dried wood flour had a moisture content of less than 0.5%. The extruded material in the form of strands was cooled bypassing it through a water bath followed by pelletizing. A portion of these pellets were dried in an oven at 80 °C for at least 24 h and then molded to make tensile and flexural testing samples. This material was named as 'cycle 0' or 'virgin' material. The remaining pellets of 'cycle 0' material were dried in an oven at 80°C for 24 h and again extruded at the same processing conditions as mentioned above and was named as 'cycle 1' or 'first time recycled' material. The remaining pellets from 'cycle 1' material were again consecutively dried, extruded, cooled, and pelletized. A portion of this pelletized composite was dried, and injection molded to get 'cycle 2' or 'second time recycled' material. This process was repeated up to six times, in total, to get 'cycle 6' or 'sixth time recycled' composites.