

Article

# Durability of High-Density Polyethylene (HDPE)- and Polypropylene (PP)-Based Wood-Plastic Composites—Part 1: Mechanical Properties of the Composite Materials

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**Abstract:** Wood-plastic composites (WPCs) have shown exceptional promise as a building material, especially for outdoor uses. Using renewable wood fiber as the reinforcing filler in WPCs increases the material's environmental sustainability. While virgin commodity thermoplastics are primarily used in these composites, using post-consumer plastic further contributes to their sustainability. While they are beginning to be used in the Gulf countries, information on their performance, especially durability under harsh desert climates, is sparse. The present investigation on WPCs is based on the two most popularly used thermoplastics in WPCs, virgin high-density polyethylene (HDPE) and polypropylene (PP), with the wood content varying between 0 and 36 wt. %. These were prepared with melt processing from a masterbatch and characterized primarily using thermal methods and tensile properties of their injection molded test pieces. Variations in tensile properties, especially the tensile modulus (MPa), the tensile strength (MPa), and the ultimate extensibility (%) of the composite samples were investigated to determine an optimal wood-fiber loading. For either polymer type, exceeding 27 weight percent of wood fiber resulted in unacceptably low ultimate extensibility of the material.

**Keywords:** wood-plastic composites; polymer degradation; polymer lifetime; HDPE; PP



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## 1. Introduction

In 2020, the energy demand by the building sector was about 127 EJ or 36% of the overall energy demand globally, with residential buildings accounting for the largest fraction of 22% of the expenditure. The associated carbon emissions from the sector were about 9 gigatons, with 37 percent of overall carbon emissions ascribed to the building sector [1]. Furthermore, the number of buildings worldwide is expected to double by 2060 [2], with the addition of 230 billion m<sup>2</sup> of new floor area (relative to 2017), suggesting an increased use of building materials, including wood and plastics. Global volumes of plastic resin and industrial roundwood production are already at the highest levels reported over the last half a century! The anticipated additional production over the next several decades will translate into a higher energy demand and carbon emissions, thus reducing the sector's environmental sustainability. The trend in the building industry towards environmental sustainability encourages builders to qualify for LEED (Leadership in Energy and Environmental Design) [3] certification in the US or the Building Research Establishment

Environmental Assessment Method (BREEM) in Europe. The present emphasis on environmental sustainability demands lowering energy costs, reducing carbon emissions, conserving non-renewable materials such as plastics, and reducing waste generation in the use of building materials.

Wood has always been regarded as a low-cost, sustainable building material with a relatively low carbon footprint compared to materials such as concrete, metals, glass, or plastics. It is a natural composite made up of lignin, cellulose, and hemicellulose, and the ratio of components varies with the wood species. Using more wood, a renewable material, in buildings increases the environmental sustainability of the structure. However, there are several drawbacks to wood as a building material including the energy costs of harvesting or processing and the material's susceptibility to solar UV radiation and biological degradation. Some of these limitations are avoided when using wood-plastic composites (WPCs) instead of wood in buildings, especially in members exposed routinely to outdoor environments. Therefore, all factors that impact the sustainability of a building product need to be considered when determining the merits of WPCs over wood or plastics. This generally requires a lifecycle analysis (LCA) and valid durability assessments that qualify WPCs as the most environmentally sustainable material adequate for a given application.

The design freedom afforded by WPCs that can be formed into single-piece building products can often result in considerable energy and emission savings. For instance, a recent Finnish study [4] found wood-plastic composites (WPCs) to have a lower carbon footprint relative to wood. Using recycled post-consumer plastics in the WPC further decreased its carbon footprint. Embedding the biodegradable, environmentally susceptible wood fibers in a thermoplastic matrix affords the material some degree of protection from the elements. Not only is the thermoplastic matrix hydrophobic but, depending on the type of polymer, it can serve as a UV screener protecting the wood component.

Wood-plastic composites (WPCs) are a recent development that dates back to the 1970s and have been popularly used as a building material in North America since the 1990s [5]. Given the present emphasis on environmental sustainability in the building sector, WPCs with renewable wood fillers are an attractive building material [6]. Natural fibers are also significantly lower in density than glass fibers (1.15–1.50 g/cm<sup>3</sup> in contrast to 2.4 g/cm<sup>3</sup> for glass) commonly used in fiber-reinforced composites and can result in a lower density of composites. However, WPCs have a lower elastic modulus than glass or carbon fiber [7,8], and they obtain significant weight reduction in the final products, thus reducing transportation costs when distributing products. Over 70 percent of WPC production is used in buildings, with polyethylene (PE) and polypropylene (PP) being the most-used plastic in their manufacture [9]. Other plastics such as poly(vinyl chloride) (PVC) and poly(styrene) (PS) are also used as the plastic fraction [10]. PE-based WPCs are typically used in exterior building components such as decking, fencing, and siding, while those with PP are used more in automotive applications. Wood/PVC composites are primarily used in window frame manufacturing and in decking applications [11]. WPCs building products typically have a 15-years warranty and are competitive in cost with treated wood. These composites have desirable sustainability characteristics because they are based on a renewable material [12] and because post-consumer plastic waste is used in the composites [10,13].

WPC materials are composites that contain plant fiber as a filler, typically in the thermoplastic matrices, and are generally produced by intimately mixing the wood component with the polymer. The mix is molded under pressure at high temperatures [14]. Most wood fibers tend to degrade above 210 °C (410 °F), and thus the processing temperatures must be controlled below this limit [15]. The most widely used resins in WPCs, polyethylene (PE), poly(vinyl chloride) (PVC), and polypropylene (PP) [16], can be processed below these temperatures. Even during outdoor use, the WPC building products enjoy decades of service life [17] and may even be recyclable for reuse in composites in the future.

A basic difficulty when using wood as a filler is that it is hydrophilic, while the polymer matrix is hydrophobic. Even when the wood fiber is exhaustively dried before

compounding, the interface between the phases will be weak. The surface functionalities on the wood fiber, especially the oxidized groups, determine how strong an interaction might be expected [18–20]. A weak interface obtains poor stress transfer through the composite, negatively impacting its mechanical properties [21,22]. To overcome these shortcomings, the wood fibers are chemically surface treated to render them hydrophobic [23] or a compatibilizer such as maleated polyolefin is used in the compound [24,25].

A related limitation is the propensity of WPCs to undergo surface biodegradation, the primary failure mechanism for untreated wood [26]. Exposed wood fibers at the surface can serve as entry points for moisture. Therefore, moisture absorption by WPCs can be much higher than that of virgin polymers. Embedding the wood fibers in the polymer matrix is expected to restrict moisture absorption and avoid the fungal growth responsible for decaying the wood fraction. The poor weatherability of the polymer fraction in the composites leads to UV-induced discoloration and exacerbates moisture absorption, leading to weakening, cracking, and bio-deterioration in the WPC [27].

WPCs have been recently introduced into the Middle East building industry. However, the material remains largely untested for its performance and durability when exposed to desert conditions. The current study will contribute to our understanding of the mechanical properties and weathering to improve their durability, cost, and efficiency in local applications. WPCs have already proven to be successful under moderate outdoor exposure conditions [28]. However, the materials and specifications developed for other regions cannot be used in the extreme weathering conditions encountered in Saudi Arabia without relevant studies conducted under realistic use conditions.

In this study, two types of WPCs were formulated with three different weight percentages of the wood component. The formulation contains HDPE or PP, wood, and a compatibilizer (maleated polyolefin) with the mixes closely resembling commercially available WPC materials. Individual ASTM tensile test pieces (dogbones) were injection-molded with care taken to ensure good dispersion of the wood in the plastic phases. In addition, the process temperature was kept as low as possible to avoid any degradation in the wood component.

Part 1 of this paper addresses the mechanical characteristics of the wood-plastic composites, and Part 2 will be on the weatherability of the WPCs under outdoor and accelerated weathering exposure.

## 2. Experimental Methods

The wood fiber was obtained from Jelu-Werk (Rosenberg, Germany) as masterbatches with 50 wt. % wood fiber (Spruce and Fir) content in PP (Grade PP-H50-500-14) and in HDPE (Grade HDPE-H50-500-09). The bulk density of the masterbatches were 550 g/L, and their melt flow index (190 °C/21.6 kg) was 33 and 60 g/10 min according to DIN ES ISO 1133, respectively. The masterbatches were dried in the hopper for 5 h at 80 °C before injection molding. Moldings were carried out at a screw temperature of 190 °C and mold temperature of 80 °C with the screw speed maintained as low as possible to avoid overheating the wood fiber. Standard ASTM Type I dumbbell test pieces were molded and stored refrigerated in the dark.

Tensile testing was carried out according to ASTM D 638 using an Instron Tensile Testing machine, Model 3367, at room temperature (20 to 22 °C) under a 10 mm/min strain rate. Load displacement data were recorded digitally using a computer attached to the machine.

Microstructural characterization was carried out using a Scanning Electron Microscope, Model JEOL JSM-6064LV. The micrographs were taken at 15 kV accelerating voltage and at various magnifications of  $\times 2000$ ,  $\times 4000$ ,  $\times 8000$ , and  $\times 30,000$ .

A Mettler Toledo Model DSC 822 Model Differential Scanning Calorimeter: DSC 822 was used to determine the thermal transition temperatures. Typically, the samples were heated from room temperature to 200 °C at a heating rate of 10 °C/min under argon flow.

Melting temperature was detected as the peak temperature, and the melting heat was obtained from the area of the peak after the baseline correction.

Fourier transform infrared (FTIR) spectroscopy is commonly used to obtain information on the molecular structure of wood as well as plastics. The infrared (IR) spectrum is related to vibrations in the molecules, is unique to each compound, and yields a 'fingerprint' of the polymer composition. Reflectance FTIR was used with the present thicker samples using an IR microscope. Infrared spectra were recorded using a Perkin Elmer Model 16F PC FT-IR spectrophotometer loaded with Spectrum V 2.00 software (Waltham, MA, USA).

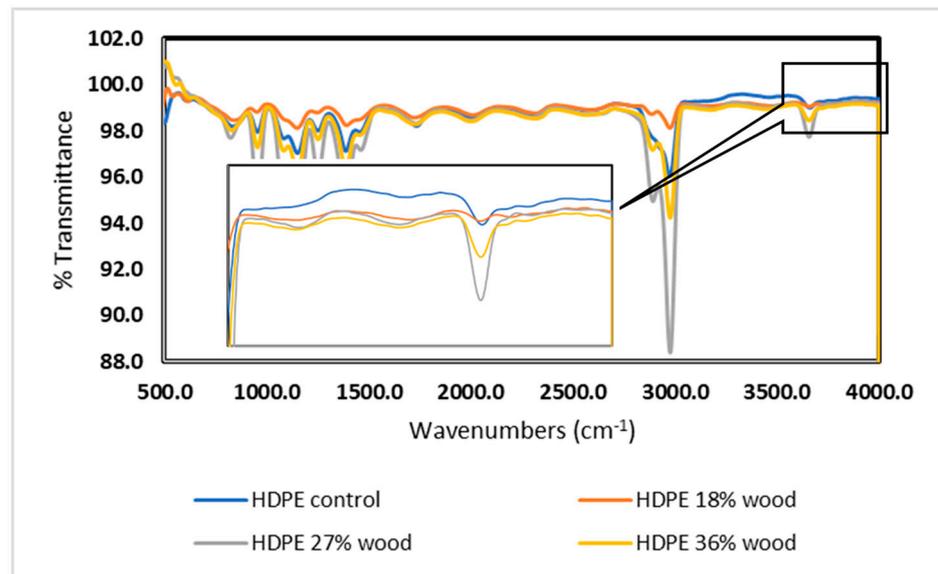
The average molecular weight of HDPE and PP were determined using Gel Permeation Chromatography (GPC). GPC is a size exclusion chromatography (SEC) separation technique that separates analytes on the basis of molecular size and is applied to polymers. Chain scission during degradation causes a general decrease in molecular weights—the number average molecular weight ( $M_n$ ), the weight average molecular weight ( $M_w$ ), the size average molecular weight ( $M_z$ ), and the polydispersity index (PDI) can be assessed conveniently using GPC. Polystyrene standards with a PDI of less than 1.2 were used to calibrate the GPC procedure. Gel Permeation Chromatography was carried out at column/detector temperature of 160 °C in 1,2,4-trichlorobenzene (TCB) at a flow rate of 1.0 mL/min and injection volume of 200  $\mu$ L in the Viscotek HT-GPC 350A model.

### 3. Results and Discussion

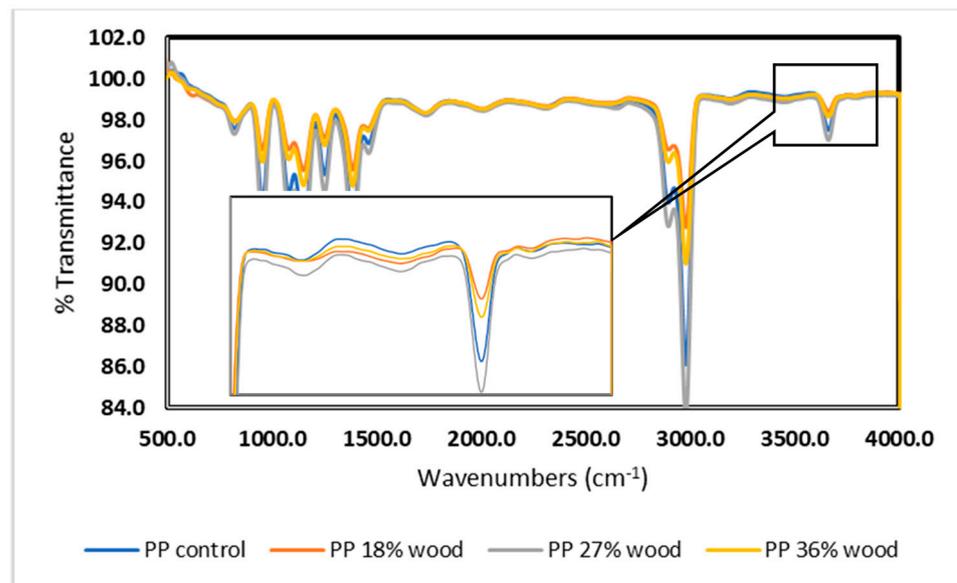
Incorporating the wood fiber into a polymer matrix can be monitored with FTIR used in the Attenuated Total Reflection (FTIR-ATR) mode. The spectra of dry composites display the characteristic -OH absorption peak associated with cellulose and hemicellulose fractions, which appears around 3500  $\text{cm}^{-1}$ . As expected, both HDPE and PP show this absorption band in that wavelength region. In addition, the intensity of the absorption band in the present samples increased with the fraction of wood fiber in them, as shown in the insert in Figure 1. The blending of wood does not result in a significant change in the melting characteristics of the polymers as the fibers do not undergo any phase transition upon heating. The melting transition in the composites, as measured using Differential Scanning Calorimetry (DSC), shows that the heat flow decreases with increasing wood content (see Figure 2), as expected, but the transition temperature of the polymer fraction is not significantly affected by the presence of wood fibers. The changes in both FTIR and thermal measurements qualitatively relate to the wood content, which suggests that the wood fibers and the polymer were intimately mixed in the composite. The spectra do not show a predominance of either wood fiber or polymer spectral bands associated with polymer- or wood fiber-rich domains.

The mechanical stresses in the melt experiences during compounding and in the injection molding of the test pieces are expected to degrade the polymers compounds to some extent. This is especially important as the molding has to be carried out at low temperatures to ensure that the wood fibers do not degrade. Any such degradation during processing would occur in both the virgin polymer and polymer/wood mixes, but the degradation in the latter case is expected to be more severe because of the additional friction afforded by the wood filler. This was investigated for the virgin polymers and their composites with low (18 wt. %) wood fiber content using GPC of the polymer fraction before and after processing. The data in Table 1 show a reduction in the number average molecular weight  $M_n$  (g/mol) of virgin HDPE and PP as well as their composites. As might be expected from structural considerations of the two polymers, the degradation rate in PP on processing will be relatively larger than for HDPE. The tertiary carbon radical likely generated during the mechanical and mechano-oxidative degradation of PP is more reactive relative to the largely secondary radical species generated by HDPE, leading to higher rates of degradation in the former. The observed percentage decrease in  $M_n$  (g/mol) for HDPE and PP in the composites are 15% and 12.6%, respectively, and are not significantly different, at least at 18% weight fraction of wood fiber. The polydispersity of the HDPE samples (PDI) decreased slightly upon being mixed with wood fiber. In the more reactive PP, the

PDI suggests that the molecular weight distribution broadened upon mixing with wood fiber. Molecular weights were not determined in composites with higher wood content.



(a)

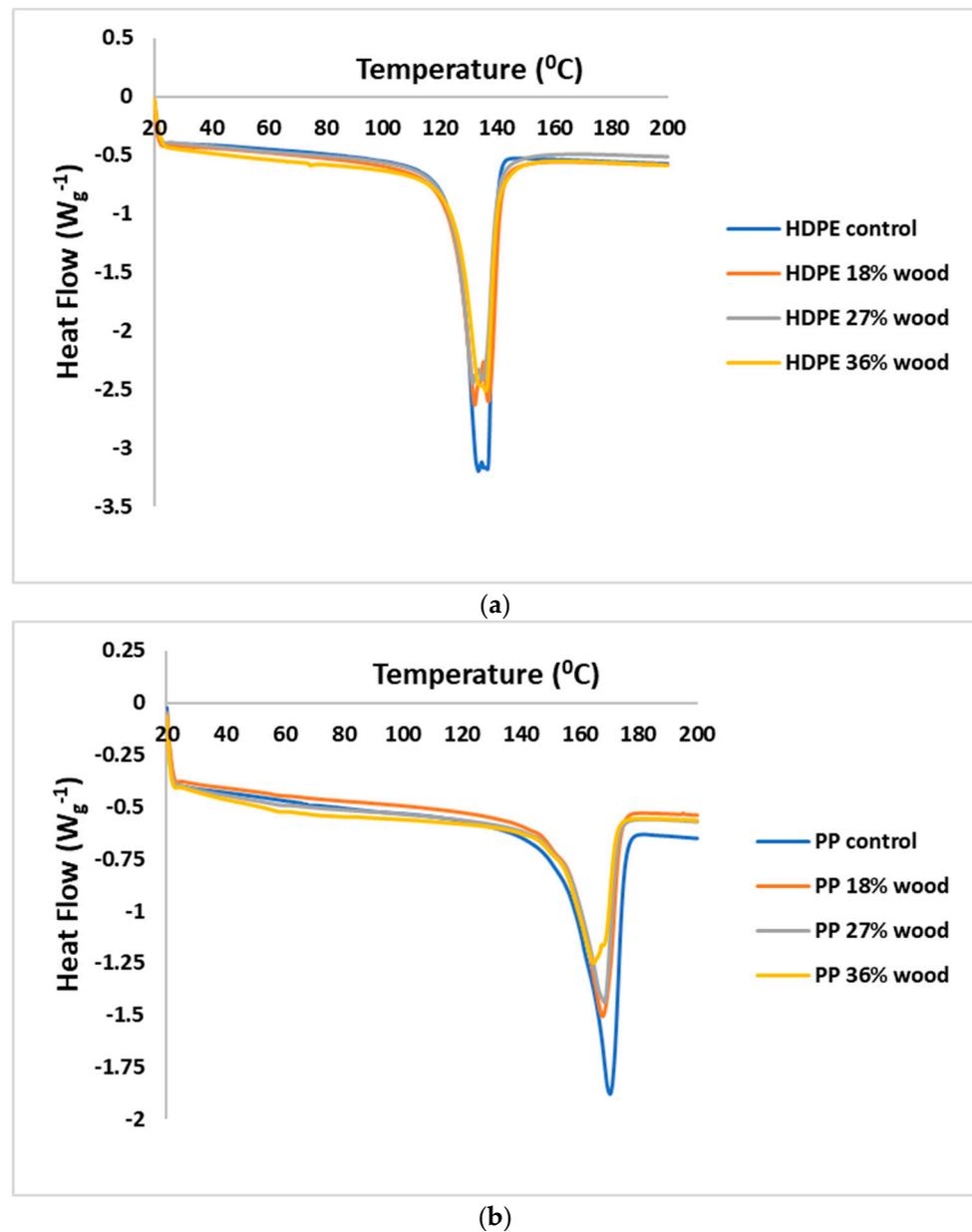


(b)

**Figure 1.** Overlay of FTIR spectra for the (a) HDPE wood composite and (b) PP wood composite samples with different percentage weight of filler.

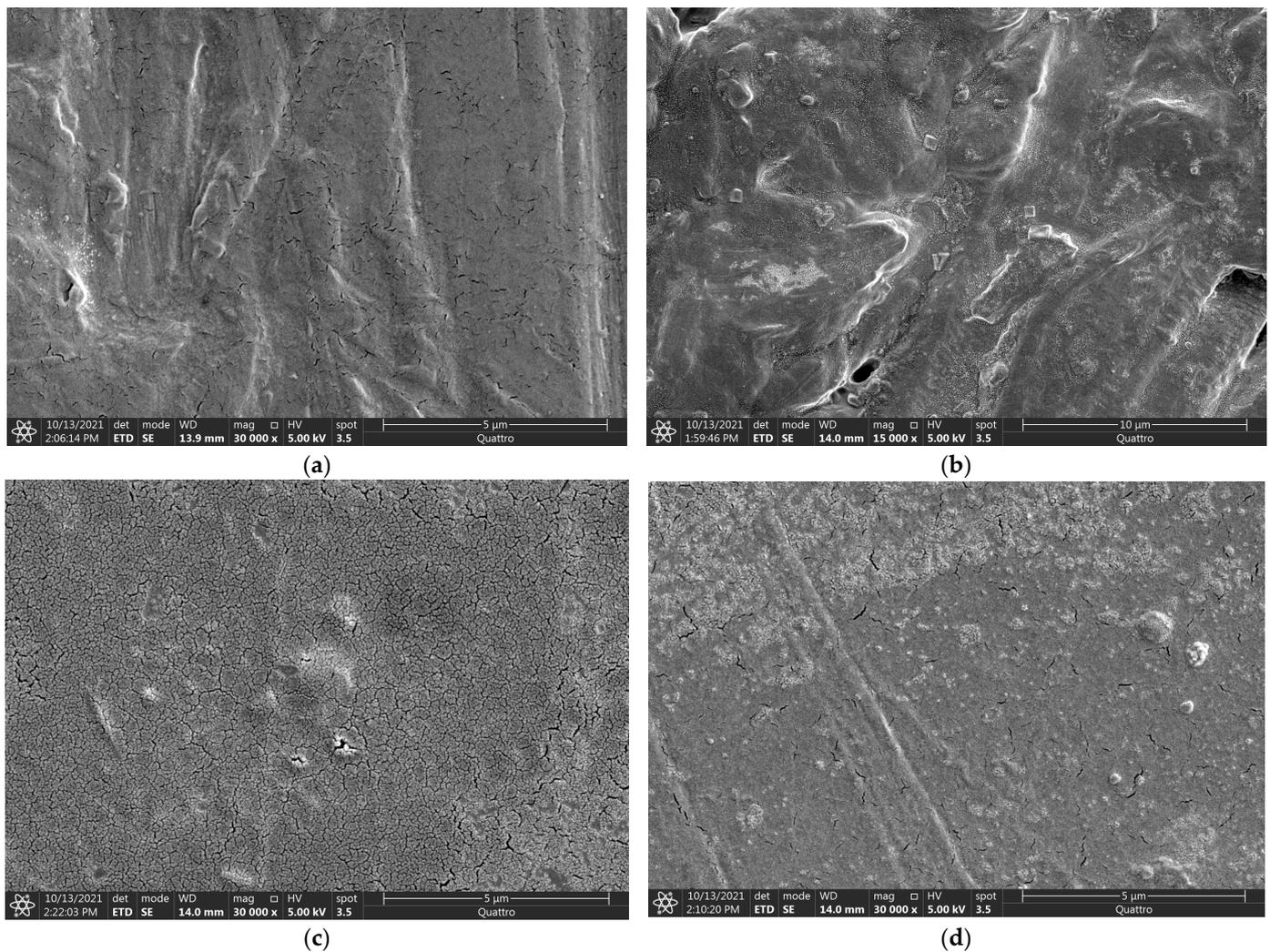
**Table 1.** Changes in the average molecular weights (g/mol) of the HDPE and PP composites with 18% wood filler.

Material	M <sub>w</sub>	M <sub>n</sub>	PDI
HDPE Control	78,428	15,208	5.16
HDPE-18% wood	61,839	12,871	4.80
PP Control	184,387	37,189	4.96
PP-18% wood	190,151	32,495	5.85



**Figure 2.** Differential scanning calorimetry of the (a) HDPE wood composite and (b) PP wood composite samples with different percentage weight of filler, shown as an overlay to illustrate the lack of any significant difference in the crystalline melt temperatures.

The primary objective of incorporating wood fiber into a thermoplastic was to obtain reinforcement that improves the mechanical integrity of the material. Any reinforcing effect of the wood filler is best assessed using tensile property measurements. Effective reinforcing generally depends on intimate mixing of the filler and polymer that in turn requires a low enough melt viscosity at the processing temperature to allow the polymer to flow into the porous wood morphology and obtain good bonding at the interface. A robust interface between the wood fiber and plastic is critical to obtaining good mechanical characteristics in composites. A coupling agent, such as maleated polyethylene used in these samples, often contributes towards a good interface between the hydrophilic and hydrophobic phases in the composite. The scanning electron micrographs (SEMs) of the composites illustrate the very good dispersion achieved in the present samples during processing. No large aggregates of wood fiber were noticed during the SEM study on the different mixes. Selected micrographs are shown in Figure 3.



**Figure 3.** Scanning electron microscopy of wood composite samples at  $\times 30,000$  magnification for the (a) HDPE-control sample, (b) HDPE-18 wt. % wood sample, (c) PP-control sample, and (d) PP-18 wt. % wood sample.

The tensile strength of composite materials is determined by the fiber's strength and its volume fraction in the composite. Generally, the ultimate strength of the fiber is much higher than that of the polymer matrix. Unlike thermoplastics that are produced using closely managed, highly controlled processes, bio-based materials such as wood fibers show a high degree of variability in their tensile properties depending on the species and growth conditions of the materials. Consequently, the coefficient of variation in tensile strength of high-grade wood is 20% to 40%, whereas that of thermoplastics such as PP is only about 5% [7,29]. The wood fiber strengths are at least an order of magnitude higher (typically 15–40 GPa) [30] than that of the polymer matrix and, consequently, the strength of the matrix has limited influence on the in-plane tensile strength of composite materials. Selecting the appropriate type of wood fiber is important to ensure high tensile strength of the composite. Variables such as differences in morphology, density, and aspect ratios across wood species used in composites account for varying reinforcement properties in thermoplastic composites [31]. With synthetic fibers, such as carbon, it is possible to design unidirectionally aligned composites with exceptionally high strengths and moduli.

The presence of a reinforcing wood filler in the polymer matrix typically increases the mechanical integrity of the composite [32,33], and this is usually reflected in their tensile strength (MPa) as well as their tensile modulus (MPa). Table 2 summarizes the

tensile data obtained for the present composites with 0%, 18%, 27%, and 36% of the filler. Attempts to mold test pieces with 50 wt. % of wood fiber were unsuccessful as the injection molded samples showed charring and insufficient polymer to hold the wood fraction together. With wood/HDPE composites, the strength and moduli increase linearly with the weight percent of filler up to 36 wt. % of wood fiber content. However, with the PP composites, the tensile strength increased only marginally with the weight fraction of wood fiber. The gradient in the tensile strength plot for HDPE and PP is 32.5 MPa/wt. % and 2.5 MPa/wt. %, respectively (Figure 4). Low extension modulus (MPa) calculated from the stress–strain curves, as shown in Table 1, shows significant increases for both the HDPE and PP composites (23 MPa/wt. % for HDPE and 23 MPa/wt. % for PP).

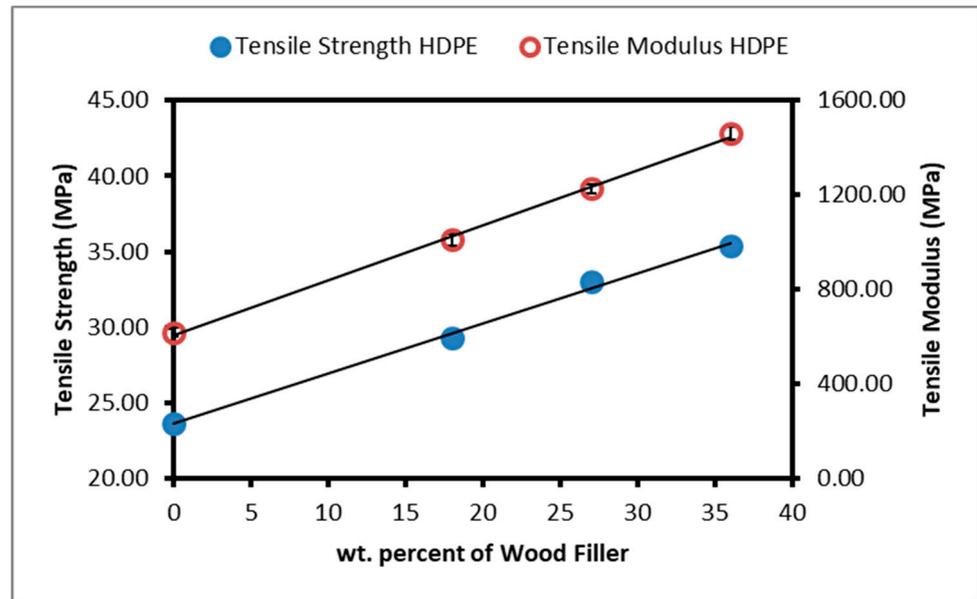
**Table 2.** Summary of tensile test data for the control samples and three formulations of HDPE and PP.

Material	Tensile Strength (MPa)		Elongation at Break (%)		Modulus of Elasticity (MPa)	
	Average	Std. Error	Average	Std. Error	Average	Std. Error
HDPE Control	23.67	0.11	1011.87	1.84	619.01	17.94
HDPE-18% wood	29.35	0.36	24.53	1.58	1010.31	22.84
HDPE-27% wood	33.05	0.38	13.75	0.68	1226.15	17.43
HDPE-36% wood	35.36	0.35	9.45	0.40	1461.73	25.46
PP Control	37.84	0.11	43.15	4.28	991.73	1.77
PP-18% wood	38.42	0.17	9.52	0.25	1370.90	5.68
PP-27% wood	38.73	0.13	6.93	0.16	1553.33	19.15
PP-36% wood	38.77	0.28	5.87	0.09	1712.30	6.49

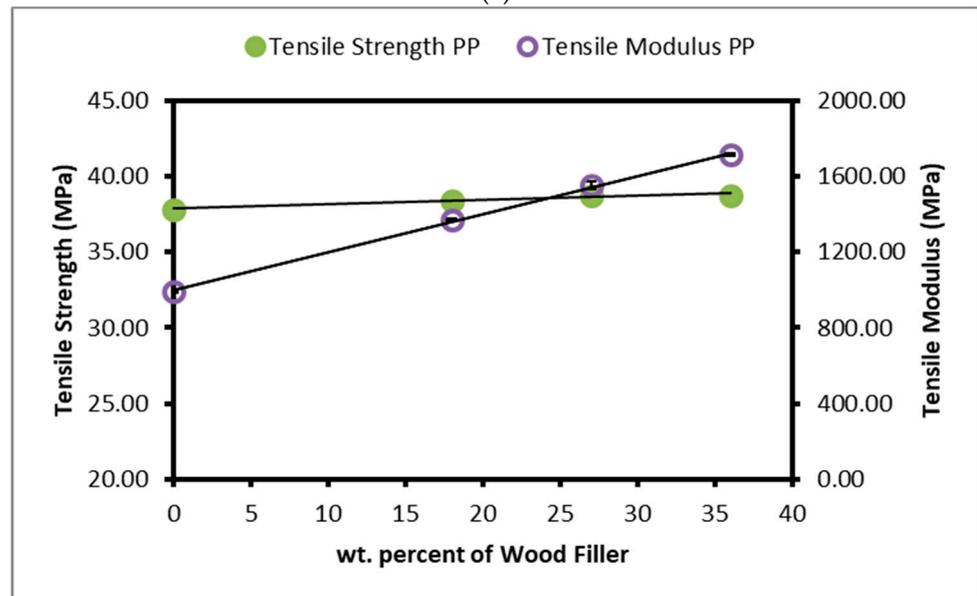
This discrepancy between HDPE and PP, as seen in Figure 4, is likely a result of the low screw temperature (190 °C) used when molding the composite samples to avoid damage to wood fibers. With the less-than-ideal melt viscosities used in PP processing, the melt did not flow well enough to allow the formation of a good interface at the wood fiber. The effects of the poor interface are more apparent at higher strains in tensile extensibility but not in low-extension moduli. The present findings are in agreement with data reported by Lu et al. on the mechanical integrity of WPCs [34]. They found the mechanical properties of the WPCs to increase with wood fiber content only at low fractions of the wood, with a maximum tensile strength obtained at 15% of wood in the WPC. At higher fractions of wood filler, the strength decreased. At higher filler content, the wood fibers aggregate, resulting in reduced interface volume that is responsible for transferring stress between the polymer and wood phases [35]. Bouafif et al. [36], working with WPCs of Cedar wood fiber in HDPE, also reported an increase in tensile strength and the low-extension tensile modulus with a wood content from 0% to 45% wood fiber. The data, however, show considerable scatter compared to the present data. Consistent with the present observations, the tensile extensibility drops drastically with wood content over the same range (from 0% to 45% wood). Therefore, the energy to break also decreases similarly. This result agrees with the present work that extends the observation to PP matrices as well.

The present result agrees with that reported earlier [37] for PP-based WPCs, where both tensile strength and the low-extension modulus increased with wood content for up to 40 wt. %. The polymer and wood fibers were mixed in a Branbury kneader prior to being melt-pressed into laminates. Others [38,39] have reported a similar dependence of tensile properties with wood content for PP-based WPCs. Earlier reports [40] studied melt-pressed laminates of WPCs with wood fiber (*Acacia* sp.) in both PP and HDPE, but without pre-mixing wood and plastic powder intimately under high temperatures (except for manual mixing), their WPCs showed opposite behavior: the tensile strength and moduli decreased with wood content for both polymers. These results, taken together with the

present data, underline the importance of mixing and dispersion to obtain WPCs with good mechanical properties.



(a)



(b)

**Figure 4.** A comparison of the change in the tensile modulus and tensile strength with different percentage weight of filler for (a) HDPE wood composite and (b) PP wood composite samples.

However, in contrast, the tensile extensibility or the elongation at break of the test pieces decreased sharply with increasing wood content in both the HDPE and PP composites. The reduction in extensibility below 10% severely limits the use of composites in some building applications. However, HDPE with 18% or 27% and PP with 18% wood fiber have sufficient extensibility to warrant further study, especially for their weatherability and moisture absorption.

Interestingly, only the high-deformation metrics measurements, the stress, and extension at break, are significantly affected by increasing the wood fiber content in both polymer types. The low extension is not so affected, suggesting that failure at the interface between the two phases at high deformations is likely responsible for the observed results.

#### 4. Conclusions

The average tensile moduli of the wood-plastic composites investigated changed with wood content, increasing linearly with wood weight fraction in both the HDPE and PP composites studied. However, the average tensile strength similarly increased only for the HDPE composites; for the PP composites, no dependence of the tensile strength on wood content was observed. This was attributed to potential inhomogeneity in the mixes resulting from the use of low processing temperatures and is apparent only for high-deformation measurements. Ultimate extensibility of both sets of composites was reduced with the wood content. Up to 36 wt. % of wood fiber in either polymer yielded a well-reinforced material. However, where flexibility is also a requirement, the upper limit of wood content would be 27 wt. % for HDPE and 18 wt. % for PP. Continued weathering studies will establish the durability of these composites under desert exposure conditions.

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