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# DEVELOPMENT OF WOOD PLASTIC COMPOSITE WITH REDUCED WATER ABSORPTION

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**ABSTRACT**

In this work, wood-plastic composites were developed using plastic lumber (recycled high-density polyethylene, or rHDPE) and wood residues (sawdust). First, raw materials were characterized by infrared spectroscopy (FTIR) and scanning electron microscopy (SEM). Second, the formulations of rHDPE/sawdust (100/0, 80/20, 60/40, 50/50, 40/60, weight percentage) were mixed manually and processed by extrusion in a twin-screw extruder (TeckTril, DCT model) equipped with ten temperatures zones. The pelletized samples obtained by extrusion were pressed to obtain the films. The WPC films were characterized by density (ASTM D792-13), melt flow index (MFI-ASTM D1238-13), hardness (ASTM D2240–10), SEM, FTIR and water contact angle measurements. The density results were 0.81, 0.87, 0.66, 1.01 and 0.98 g.cm$^{-3}$ using 0, 20, 40, 50 and 60% sawdust, indicating good fiber-matrix adhesion, confirmed by SEM images. The FTIR spectra showed peaks of rHDPE and sawdust. No chemical interaction occurred between the components after extrusion. The MFI for WPC decreased in relation to the matrix (100/0), but no significant change was observed regardless of the fiber content used. The wettability of the composites' surface, evaluated by water contact angle measurements, revealed angles close to 90$^{o}$, which can be considered an positive result, since no coupling agent was used. The absorption water value for composites was less than 3%, a reference value for absorbing MDF panels.



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# INTRODUCTION

Wood-plastic composites (WPCs) are environmentally friendly materials because the source of the raw material can be either virgin or recycled materials. The use of recycled thermoplastics like high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP) and polyvinylchloride (PVC) has increased in recent years, since recycling of plastics can reduce not only pollution but also manufacturing costs, energy consumption and depletion of virgin materials (Najafi 2019; Hyvärinena *et al.*, 2019). As reported by Chaudemanche *et al.* (2018) and Lopez *et al.* (2020), sawdust is one of the main residues resulting from the wood processing. When stored under uncontrolled conditions, it can be an important source of environmental pollution and damage to the human respiratory system due to $NO_2$ (nitrogen dioxide) emissions. Among the most used species, *Cedrelaodorata L.* from the Amazon rainforest is one of the main woods in terms of economic value. It can be used for making furniture, fine interior and exterior joinery, sculpture, coverings, moldings, skirting boards, friezes, doors, window frames, decorative veneers and plywood (Locatelli *et al.*, 2006; Lopez *et al.*, 2020). Thermoplastic materials have a very short useful life and are typically discarded in landfills or dumps, contaminating water bodies and soil, besides causing death of animals that ingest them, but they can be recycled (Spinacé and de Paoli, 2005). Many studies are reported in the literature about the development of WPCs for industrial uses (Spinacé and de Paoli, 2005; Lei and Wu, 2010; Zhang *et al.*, 2019; Hyvärinena *et al.*, 2019; Marins *et al.*, 2019; Liu *et al.*, 2019; Lazrak *et al.*, 2019). They are mainly used in long-life applications such as paving, plastic wood, civil construction and the automotive and electronics industries (Spinacé and de Paoli, 2005). Plastic lumber is produced from recycled plastic waste, mainly HDPE, with fillers and additives, aiming to improve the material's properties. Martins *et al.* (2019) tested the use of inorganic solid waste from the manufacture of fluidized-bed catalytic cracking catalyzers (FCC catalysts) as flame retardant in polymer–matrix composites of plastic lumber.

The results indicated that for applications where fire resistance is the preponderant factor, the 40/60 (rHDPE-FCC) formulation was the most appropriate, but the physical properties may limit its application. Liu *et al.* (2019) studied the use of two kinds of silane coupling agents to improve the bond between virgin HDPE and wood fiber. Treatment with these two agents significantly changed the contact angle of the wood veneer, which increased from 46º before treatment to greater than 120º afterward. Lazrak *et al.* (2019) studied the hydrophobicity of the surface of WPCs (rHDPE/pine wood) using water contact angle measurements in two ways: the direct method and theoretical method. They observed a contact angle value of 67.63º for the composite using 30% of wood. It decreased to 33.97º for composite containing 70% wood. The roughness of the composites' surfaces was improved by adding HDPE.

The advantages of WPCs include increased flexural strength and stiffness, reduced thermal expansion and cost compared to unfilled thermoplastics (Liu and Wu, 2010). Due to the hydrophilic nature of wood, outdoor applications can be limited, so research regarding engineering aspects of WPCs is important. The goal of the present work was to obtain WPCs using plastic lumber - recycled HDPE (rHDPE) - as the polymer matrix and sawdust (of *Cedrella Odoratta L.*) as filler without coupling agent. WPCs (rHDPE/sawdust) obtained by extrusion were compounded in proportions of 100/0, 80/20, 60/40, 50/50 and 40/60 (weight percentage). The raw materials were characterized by scanning electron microscopy (SEM) and Fourier-transform infrared spectroscopy(FTIR). WPCs were characterized as to density (ASTM D792-13), melt flow index - MFI (ASTM D1238-13), SEM, FTIR, contact angle measurements and hardness (ASTM D2240–10).

**Experimental Procedures**

**Raw Materials:** Plastic lumber waste (recycled high-density polyethylene, or rHDPE) was provided by Companhia Municipal de Limpeza Urbana (Comlurb, the municipal sanitation company in the city of Rio de Janeiro). The material was processed in the following steps: separation, cutting and magnetization, to remove possible contaminants. Wood residues of *Cedrelaodorata L.* (kindly donated by a local company in the city of Rio de Janeiro) were used in the form of sawdust. The sawdust was ground and dried at 100 ºC in an oven for 24 hours before processing. To reduce the costs of the process, sawdust was used without sieving or treatment, and no coupling agent was used.

**WPC processing:** Sawdust was previously dried in an oven with forced air circulation at 100 °C until constant weight (approximately 24 hours) and stored in a desiccator for another 24 hours before processing. The materials were mixed manually, and the rHDPE/sawdust composites were compounded in ratios of 100/0, 80/20, 60/40, 50/50 and 40/60 (weight percentage). Each formulation was fed into a twin-screw extruder (TeckTril, DCT model) equipped with ten temperatures zones, ranging from 115 to 175 ºC, from the feed to die, and rotating at 116 rpm. Then, a water jet at 25 ºC cooled the extruded material. The specimens for the WPC characterization were obtained by pressing the pelletized samples from the extrusion, at a temperature of 190 ° C for 300 s, with pressure of 7 tons for film formation, and then cooled in a cold press for 60 s.

**Characterization**

**Density, melt flow index (MFI) and hardness**

The WPCs were characterized according to density (ASTM D792-13), melt flow index, MFI (ASTM D1238-13) and hardness (ASTM D2240–13).

**FTIR:** The raw materials and the WPCs were characterized by Fourier-transform infrared spectra (FTIR), with a Nicolet 6700 FTIR spectrometer (Thermo Scientific). The samples were mounted on an attenuated total reflectance (ATR) accessory equipped with ZnSe crystal prior to scanning. The spectra were obtained with an accumulation of 120 scans and resolution of 4.182 $cm^{-1}$.

**SEM:** Scanning electron microscopy (SEM) analysis was performed using a Quanta FEG 250 microscope, to observe specimens coated with gold. Cryogenically fractured transversal sections of the samples were assessed, and the images were obtained at 200, 500 and 1000 x magnification, at 15 kV.

**Contact angle measurements:** The wettability of r-HDPE/sawdust surface was examined through water contact angle measurements using an NRL A-100-00 Ramé-Hart goniometer. The evolution of the droplet shape was recorded with a CCD camera every 15 s for a period of 225 s for each sample, at room temperature.

**Water absorption:** The water absorbing test was carried out according to ASTM D-570.Three conditionedspecimens (20 x 20 x 20 $mm^3$) wereplaced in a container of distilled water maintainedat a temperatureof 23±1°C and rested one dgeand been tirelyimmersed. At theendof 2 h and 24 h, thespecimenswereremovedfromthewateroneat a time, allsurfacewaterwipedoffwith a drycloth, andweighed in a analytical balance (Marte, UX4200H model) 0.001 g immediately.

# RESULTS AND DISCUSSION

Figure 1 shows the results of density analysis of WPCs. According to Rahman*et al.* (2013), the most important factor of WPC performance is density, which affects all other properties. The low density of wood waste is one of the factors that causes inconsistencies in the product, such as material separation, leading to production of low-quality WPC (Rahman*et al.*, 2013; Koenig and Sypens, 2002). The density results were 0.81, 0.87, 0.66, 1.01 and 0.98 $g.cm^{-3}$ using 0, 20, 40, 50 and 60% sawdust, indicating good fiber-matrix adhesion. The results showed a tendency for slightly increased density of the composite with sawdust content of 50% or more. Similar results were found by Rahman*et al.* (2013), who developed PET/sawdust composites and found density results ranging from 0.856 $g.cm^{-3}$ to 1.048 $g.cm^{-3}$, and by Souza *et al.* (2018), who found average densitiy values of particleboard (epoxy/wood waste) of 0.55 - 0.89 $g.cm^{-3}$.

According to the standards ABNT NBR 14810:2 (Brazilian Association of Technical Standards, 2013) and ANSI/A208.1 (American National Standards Institute, 2009), particleboards are classified as follows: up to 0.59 g/cm³ - low density; 0.60 - 0.79 g/cm³ - medium density; and above 0.80 g/cm³ - high density (Souza *et al.*, 2018). Thus, it can be inferred that all WPCs developed in this work met the normative requirements regarding density for application as panels with medium density (60/40) and high density (80/20, 50/50 and 40/60). The MFI results are shown in Figure 2. There was a decrease in the flow rate in the samples after the insertion of the sawdust. The composites in general did not undergo any significant change in this property, presenting flow rates below that of the pure sample (rHDPE). This effect can be attributed to mechanical obstruction of the polymer molecules due to the sawdust. The presence of fibers in the melt and their partial misalignment significantly affect the viscoelastic dynamics of the melt, hampering molecular chain mobility and therefore flow (Bastos*et al.*, 2018). According to the literature (Kajaksa*et al.*, 2018; Soccalingame*et al.*, 2015; Pereira *et al.*, 2015), for the extrusion process the lowest permissible MFI values are about 0.1–0.3 g/10 min, and for compression molding it can be lower than 0.1 g/10 min. All WPCs maintained sufficient fluidity, since the MFI value changed from 0.6 g/10 min for rHDPE to 0.26–0.14 g/10 min for filled systems (20–60 wt% sawdust). Therefore, the MFI values were high enough to process these materials with the traditional polymer processing methods. Table 1 and Figure 3 show the water contact angle measurements and images of the droplets on WPC surface, respectively. The water contact angle measurements revealed values around 84°, thus showing a hydrophobic tendency (angles close to 90°). We expected the contact angle for composites with more wood at the surface to be lower, since wood is a hydrophilic material.

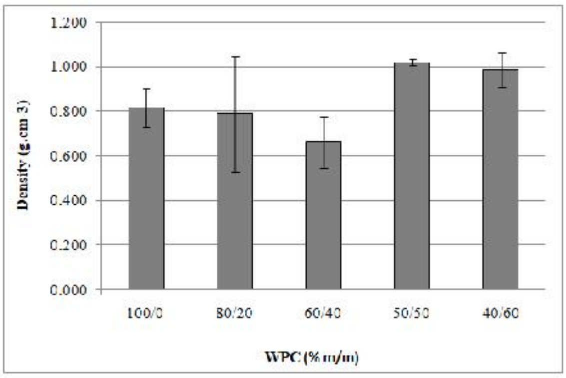


**Figure 1. Density results for WPCs (rHDPE/sawdust)**

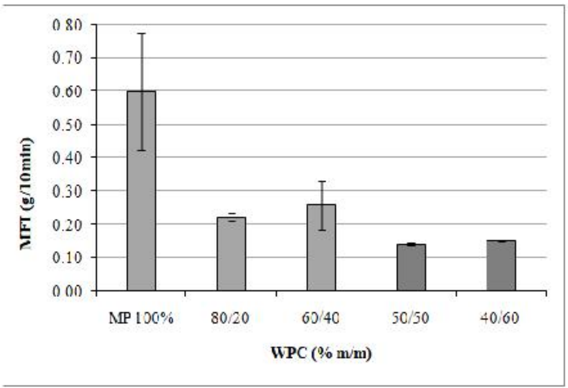


**Figure 2. Results of Melt Index Flow (MFI) for WPCs**


**Figure 3. Droplet photos: contact angle between water and WPCs (rHDPE/sawdust) surface**

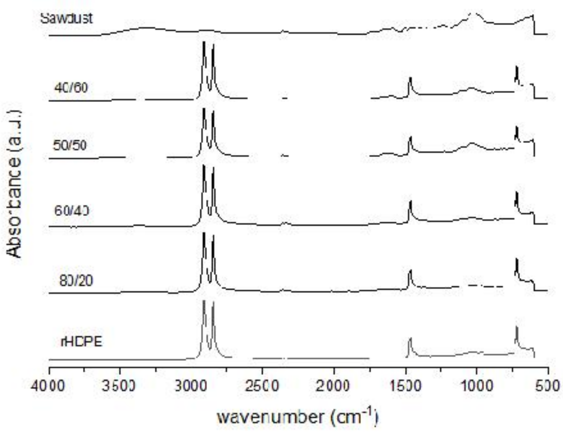


**Figure 4. ATR-FTIR spectra of sawdust and WPCs (rHDPE/sawdust)**

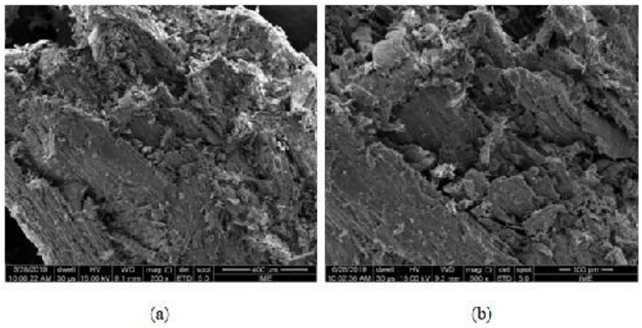


**Figure 5. SEM micrographs of sawdust: 200x magnification (a) and 500x magnification (b).**

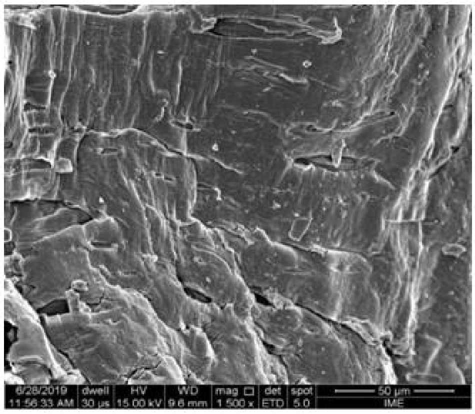

**Figure 6. SEM micrograph of rHDPE: 1500xmagnification**

**Table 1. Water contact angle measurements results**

| WPC (wt %) | Contact Angle (°) |
|---|---|
| 100/0 | 68.4±1.06 |
| 80/20 | 85.2± 0.27 |
| 60/40 | 84.1±1.01 |
| 50/50 | 79.5 ±2.19 |
| 40/60 | 75.7 ±0.66 |

**Table 2. Effect on the absorbed water of rHDPE/sawdust composites**

| WPC (wt %) | Water absorption (2 h) (wt %) | Water absorption (24 h) (wt %) |
|---|---|---|
| 100/0 | 0 | 0 |
| 80/20 | 0.021±0.000 | 0.261±0.005 |
| 60/40 | 0.096±0.000 | 1.154±0.004 |
| 50/50 | 0.155±0.000 | 1.864±0.002 |
| 40/60 | 0.171±0.000 | 2.048±0.000 |

**Table 3. WPCs hardness results.**

| WPC (wt %) | Hardness (Shore D) |
|---|---|
| 100/0 | 74,3±4,25 |
| 80/20 | 71,4±2,07 |
| 60/40 | 71,0±2,00 |
| 50/50 | 69,0±2,92 |
| 40/60 | 72,2±2,68 |

However, the sample-water interface had the same behavior regardless of the wood proportion (20, 40, 50 and 60 wt%). This behavior can be attributed to the additives used to improve the flow of post-consumer polymer (plastic lumber) during reprocessing of the material, as pointed out by Martins *et al.* (2019), and to improve fiber-matrix adhesion, in accordance with the density results obtained in this work. Similar behavior was reported by Stark and Mutuana (2007) for WPCs (virgin HDPE/wood flour).

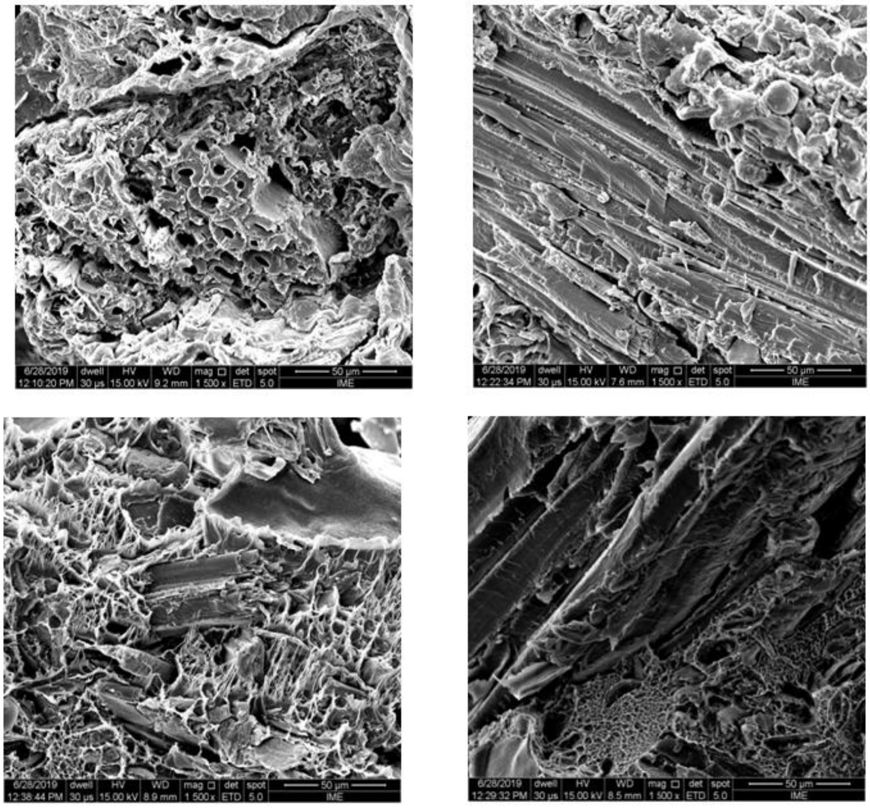

**Figure 7. SEM micrographs of WPCs (rHDPE/sawdust): (a) 80/2, (b) 60/40, (c) 50/50, (d) 40/60. 1500x magnification**

The effects of sawdust on absorbed water of rHDPE composites are shown in Table 2. The absorbed water increases with sawdust addition in relation to matrix (rHDPE). To determine the durability of WPC, water uptake is a significant parameter, because moisture causes interfacial cracks resulting from wood particles welling (Chaudemanche *et al.*, 2018). Water up take is the result of surface porosities and absorption by wood particles. In this work, at room temperature after 2 h and 24 h, no significant differences between the extruded composites were observed. The absorption of all composites was less than 3%, a reference value for absorbing MDF panels. Figure 4 shows the ATR-FTIR spectra of sawdust and WPCs. Four major bands can be detected from the sawdust spectrum: the band at 3300 $cm^{-1}$ (O-H stretching); 2950 $cm^{-1}$ (C-H stretching); 1720 $cm^{-1}$ (hemicellulose and lignin stretching) and 1270 $cm^{-1}$ (hemicellulose and lignin C-O stretching) (Lunz et al., 2012). Polyethylene peaks corresponding to CH, $CH_2$ and $CH_3$ appeared at 2923 $cm^{-1}$ and the peak at 1475 $cm^{-1}$ referred to angular deformations of $CH_3$ (Koenig and Sypens, 2002). HDPE also exhibited C-C bending, which is normally observed with absorption bands in the 1000-1250 $cm^{-1}$ region (Torres and Almeida, 2010; Gerado *et al.*, 2020).

In WPCs, the signal corresponding to the hydroxyl (3300 $cm^{-1}$) present in the sawdust was attenuated, showing there was a reduction in O-H bonds in the composite (Lunz *et al.*, 2012; Bastos *et al.*, 2018), which is in accordance with contact angle measurements. Some other bands could be attributed to the additives used to improve the flow of post-consumer polymer during reprocessing. The rHDPE/sawdust interface is physical, since there were no changes in the WPCs' infrared peak (Bastos *et al.*, 2018). SEM micrographs of sawdust are presented in Figure 5. According to Hill *et al.* (1998), fiber is considered a natural polymer consisting basically of cellulose, hemicellulose and lignin. These constituents contribute differently to mechanical strength. SEM images showed sawdust covered by substances such as pectin, lignin and amorphous wax present in fiber cuticles (Borsoi *et al.*, 2014). In the formulation with 100% plastic residue (Figure 6), it was possible to observe the presence of some dispersed solid particles. The fact that the material is recycled explains the presence of these particles, because of the use of additives to improve material reprocessing.

Figure 6 also shows the formation of typical "alveoli" of plastic fractures (Martins *et al.*, 2019). The WPC micrographs are shown in Figure 7.It is possible to observe the wetting of the sawdust by the polymeric matrix, indicating there was good fiber-matrix adhesion with all proportions used, which can be attributed to the presence of additives in the recycled matrix. The micrographs corroborated the other results. Table 3 shows the results of hardness. A slight decrease in the hardness of the WPCs in relation to the matrix (100/0) was observed. According to Borsi *et al.* (2014), this occurs due to the possible non-uniformity in the distribution of fibers in the matrix. No significant changes were observed for the WPCs with 20, 40, 50 or 60 wt% sawdust.

# CONCLUSION

In this work, wood-plastic composites from recycled HDPE and sawdust were obtained by extrusion and characterized. The results are summarized as:

- For the rHDPE/sawdust composites (80/20, 60/40, 50/50 and 40/60, weight percentage), the MFI results were sufficient for these materials to be submitted to traditional polymer processing methods.
- The density results showed that WPCs developed in this work should be used as panels of medium (60/40) or high density (80/20, 60/40, 50/50 and 40/60).
- FTIR peaks of WPCs showed there is only physical interaction between rHDPE-sawdust.
- The slight decrease in the hardness of WPCs in relation to the matrix is associated with uneven distribution of fibers in the matrix.
- The water contact angle measurements for WPCs were around 84°, a result reported in the literature only with the use of compatibilizers.
- The water absorption test showed low absorption and pointed out the possibility of using the WPCs obtained in this work in civil construction. The absorption of all composites was less than 3%, a reference value for absorbing MDF panels.

The results of this work are promising, since WPCs were obtained without the use of coupling agent, indicating the possibility of reducing the costs of final products and pollution, since they only use waste in the formulations. Considering the analyses realized, the formulation 80/20 (rHDPE/sawdust) is the most suitable for panel production (water contact angle value of 85.2$^o$).

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