Preparation of Pinewood Residues/Recycled HDPE Composites with Potential to Substitute Medium-density Fiberboards

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This work reports on the preparation of pinewood residues/recycled high-density polyethylene (HDPE) composites to evaluate their performance under flexion, extraction of nails and screws, and moisture absorption (MA) to assess their potential to replace medium-density fiberboards (MDFs). The effect of filler particle size (PS) was evaluated, and scanning electron microscopy (SEM) was conducted to elucidate the state of the interphase. The effect of UV-light accelerated weathering (AW) on samples with and without a UV stabilizer (UVS) was assessed. A dynamic mechanical analysis (DMA) was also conducted. The composites had better flexural performance, MA, and screw extraction resistance than the MDFs. However, AW affected the composites, mostly affecting those without UVS. Scanning electron microscopy showed the appearance of cracks on the surfaces with less UVS. The DMA results suggested that the composites with the largest PS showed a better resistance to creep.

Keywords: Pinewood-residues; Medium-density fiberboards; Mechanical performance; Recycling; Wood-plastic composites; UV degradation; Moisture absorption; Creep

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INTRODUCTION

Research interest in wood-plastic composites (WPCs) is increasing due to their promising potential to replace a variety of wood-made products in the construction industry (Clemons 2002; Schut 2005; Cruz-Estrada *et al.* 2006; Garnica 2010; Thompson *et al.* 2010; Saloni *et al.* 2011). Some of their potential applications include use as handrails, tiles, coatings, and profiles for windows and doors. One of the most notable features of WPCs is that they absorb much less moisture than wood (Klyosov 2007a; Kord 2011). Reinforcing recycled thermoplastic matrices with lignocellulosic particles brings many other advantages. Such advantages include the low cost and density of the reinforcing particles, their environmental benefits, and their ability to provide good mechanical properties to the composite (Zhang *et al.* 2007; López *et al.* 2013). Recycling is a way to mitigate deterioration of the environment that encourages the development of technology aimed at obtaining alternative materials and products. Such development provides value to solid residues that would otherwise be wasted.

One of WPCs' many possible applications is replacing medium-density fiberboards (MDFs) (Chaharmahali *et al.* 2008), which are commonly used for manufacturing home and office furniture. In contrast with WPCs, MDFs have serious problems with moisture absorption, causing them to swell and crumble. The MDFs are also used in outdoor applications, such as components for windows and doors including gabled windows, thresholds, frames, and pilasters. They are also used as stringers, lower guides, pediments, and for garage doors, among other applications. These materials are described as a type of hardboard, which is made from wood fibers glued under heat and pressure. Most of the time urea formaldehyde (UF) is used to bind the wood particles together. One of the problems faced by standard MDFs (*i.e.* in raw form) is that UF slowly releases from the surface, and possibly from the material through cutting and sanding. So, painting the whole product is recommended to seal it. The MDFs, can also be fixed together with screws and nails, but the material may split if care is not taken. Regarding the WPCs, they are produced by thoroughly mixing ground wood particles and heated thermoplastic resins. The most common method of production is to extrude the material into the desired shape, though injection molding is also used. WPCs may be produced from either virgin or recycled thermoplastics. Importantly, in comparison to conventional MDF boards, WPC boards do not need a compulsory lamination for end user applications. These boards can be directly applicable with aesthetically and technical harden surface properties in compare to even high-pressure laminate applied surfaces.

Although the use of WPC-made products has advantages, it is necessary to continue to improve their properties. For example, they are prone to creep and deform permanently under the influence of stationary mechanical stress when they are in service for long periods (Tajvidi *et al.* 2005; Xu *et al.* 2011; Kazemi-Najafi *et al.* 2012; Hidalgo-Salazar *et al.* 2013). This can happen as a result of long-term exposure to even low levels of stress. Deformation and creep occur at high temperatures but can also happen at room temperature. Depending on the magnitude and duration of the applied stress, the deformation may become so large that it can lead to failure and catastrophic fracture, preventing the object from functioning as intended. Another challenge is the optimization of the outdoor performance of WPC-made products. Although WPCs are promoted as low-maintenance, high-durability products, evidence of degradation of the thermoplastic matrix, wood particle decay, and susceptibility to mold during exterior use for larger periods have been documented (Schnabel 1981; Morris and Cooper 1998; Klyosov 2007b). Environmental modes of degradation, either abiotic or biotic, acting separately or synergistically, negatively affect a products' performance. For instance, the action of UV rays during outdoor exposure affects both the mechanical performance of the products and their aesthetic appeal. During outdoor applications, the surface of wood-polymer composites is affected. Although the damage is superficial, it promotes the occurrence of the other types of degradation already mentioned. Photostabilizers are often used to protect WPCs against weather effects, which increases their potential to replace MDFs for outdoor use. Hindered amine light stabilizers (HALS) have been extensively examined to protect polyolefins and combat UV degradation (Gijsman *et al.* 1993; Gugumus 1993).

Although the creep deformation and photodegradation of WPCs have been extensively studied, research on these issues should continue to explore their potential to replace conventional materials. Accordingly, this work focuses on studying the mechanical

performance of WPCs prepared with pinewood residues and recycled HPDE to compare their performance with that of commercial MDFs. Evaluation of the composites' performance under bending, removal of nails and screws, and moisture absorption was conducted. The effect of filler particle size, the use of a UV stabilizer, and the characteristics of the composites' creep were also evaluated.

EXPERIMENTAL

Materials

Wood residues

Pinewood residues (PR) supplied by Maderas Bajce (Merida, Mexico) were used as the dispersed phase. The material was a mixture of chips and sawdust (Fig. 1) produced by cutting wood boards without any pretreatment (*i.e.* without preheating or painting).

Fig. 1. Pinewood chips and sawdust

The wood residues were initially dried at 80 °C for 24 h in a convection oven (Fisher Scientific, Pittsburgh, PA) to eliminate excess moisture. Afterwards, the material was milled with a Pagani granulating machine (model 1520, Molino Pagani SpA, Borghetto, Italy) fitted with a screen plate drilled with holes 4 mm in diameter and then screened in a Tyler nest of sieves (meshes #20, #30, and #50) for 5 min using a sieve shaker (model RX-29, W.S. Tyler® Industrial Group, Mentor, OH, USA). The PR used consisted of particles with different sizes. Three different ranges of particle size were used in the experiment (all dimensions are approximate): (1) the milled PR which was not sieved (*i.e.* sizes less than approximately 4 mm); (2) the milled wood that passed through mesh #20, but was retained on mesh #30 (*i.e.* 0.84 mm > particles size > 0.60 mm); and (3) all the milled wood that passed through mesh #50 (*i.e.* particles size < 0.30 mm).

The different ranges of particle sizes described above will be referred to throughout the text as M4mm, R30, and TF, respectively.

Thermoplastic matrix

Recycled HDPE (from Recuperadora de Plásticos Hernández, Merida, Mexico) with a melt flow index (MFI) of 4.56 g/10 min at 190 °C was used as a polymer matrix. The as-received flake-shaped material was ground with a granulating machine (model TI 880804, C.W. Brabender® Instruments, Inc., South Hackensack, NJ, USA) fitted with a screen plate drilled with 1-mm holes in diameter.

Coupling agent and processing aid

The HDPE grafted with maleic anhydride (Polybond 3009 from Brenntag México S.A. de C.V., Cuautitlán Izcalli, Mexico) was used as coupling agent (CA). Its physical properties were as follows: MFI = 5 g/10 min at 190 °C, density = 950 kg/m^3 at 23 °C, and melting point = 127 °C. The maleic anhydride level was 1 wt.%. A blend of modified fatty acid esters (Struktol TPW113 from Struktol Company of America, Cuyahoga Falls, OH, USA) with a dropping point of 67 °C to 77 °C and a specific gravity of 1.005 was used as a processing aid (PA). Both the CA and PA were ground with the C.W. Brabender® instrument previously described.

UV stabilizer

A UV stabilizer (UVS) was purchased from GRUPO ALBE, S.A. DE C.V. (El Salto, Jalisco, Mexico). Its effect on the performance of the WPC prepared with PR TF was evaluated. This additive (off-white, slightly yellow solid microgranules) is a light stabilizer belonging to the hindered amine light stabilizer family of stabilizers (LOWILITE® 62). Its typical properties were as follows: softening range (°C) = 55 to 77, molecular weight (g/mol) = 3100 to 4000, and bulk density (kg/m^3) = 570.

Control materials

The performances of the WPCs obtained with solid residues were compared with that of medium-density fiberboard panels (Fig. 2) purchased from Placacentro MASISA (Merida, Mexico).

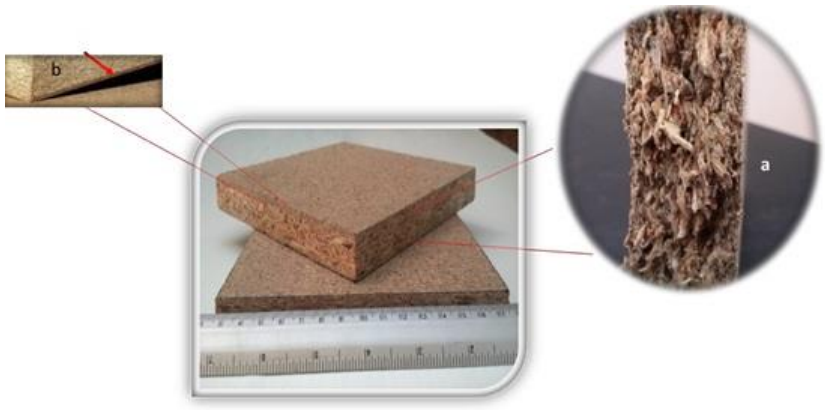

Fig. 2. MDF panel: (a) core composed of wood particles of different sizes and (b) layer formed by flour-like wood particles (the red arrow shows the lower layer)

The wood particles were bonded together by an adhesive composed of urea resin. A 15-mm-thick board and a 25-mm-thick board were acquired. These materials will be referred to throughout the text as MDF15 and MDF25, respectively. These materials consist of a core composed of wood particles of different sizes (Fig. 2a) and two layers (upper and lower) formed by flour-like wood particles that give a smooth finish to the board (Fig. 2b). The MDF15 was used to obtain test pieces for flexural and water absorption tests. The MDF25 was used to obtain specimens for the screw and nail extraction tests.

Composites preparation

Pinewood, HDPE, and additives were pre-mixed in a horizontal mixer with a helical agitator (model ML-5; Intertécnica Co., Mexico City, Mexico) and dried in the convection oven at 85 °C for 24 h before compounding. Some formulations were prepared with the three different PR particle size ranges and others were prepared with all of the additives, including different UVS content but using only PR TF. Details are shown in Tables 1 and 2, respectively. The reader should note that the composite PR TF presented in Table 1 is the same as that presented in Table 2 with the nomenclature PR TF 0.0 UVS.

Table 1. Formulations of WPCs Based on Pinewood Residue and HDPE

Composite	Wood (wt.%)	HDPE (wt.%)	CA (wt.%)	PA (wt.%)
PR M4mm	38.760	58.140	1.938	1.163
PR R30	38.760	58.140	1.938	1.163
PR TF	38.760	58.140	1.938	1.163

Notes: The wood/HDPE ratio is 40/60; the wt.% of CA with respect to wood is 5; the wt.% of PA with respect to wood is 3

Table 2. Formulations of WPCs Based on PR TF, HDPE, and UVS

Composite	Wood (wt.%)	HDPE (wt.%)	CA (wt.%)	PA (wt.%)	UVS (wt.%)
PR TF 0.0 UVS	38.760	58.140	1.938	1.163	0.000
PR TF 0.5 UVS	38.647	57.971	1.932	1.159	0.290
PR TF 1.0 UVS	38.536	57.804	1.927	1.156	0.578
PR TF 1.5 UVS	38.425	57.637	1.921	1.153	0.865

Notes: The wood/HDPE ratio is 40/60; the wt.% of CA with respect to wood is 5; the wt.% of PA with respect to wood is 3; the wt.% of UVS with respect to HDPE are 0.0, 0.5, 1.0, and 1.5, respectively

Compounding was conducted in a laboratory-scale conical twin-screw extruder (EP1-V5501, C.W. Brabender® Instruments, Inc., South Hackensack, NJ, USA) using a 4-cm long extrusion cylindrical die with a 5-mm internal diameter fitted to the extruder. During extrusion, the screw speed was 50 rpm and the barrel temperature and die temperature were set to 180 °C. The obtained extrudates were pelletized using a laboratory pelletizer machine (type 12-72-000, C.W. Brabender® Instruments, Inc., South Hackensack, NJ, USA).

Preparation of WPC test samples- samples without UVS

An automatic hydraulic press (model 5403CEB.4NE1001, Carver, Inc., Wabash, IN, USA) was used to prepare WPC boards approximately 280 mm wide × 470 mm long.

Approximately 15-mm-thick and 25-mm-thick boards were prepared using the pellets obtained *via* the twin-screw-extrusion procedure detailed above. Pellets with PR M4mm, R30, and TF without UVS were used. The 15-mm-thick boards were prepared by hot-pressing the pellets at 190 °C for 20 min with a pressure of approximately 14 MPa. The hot-pressing conditions for the 25-mm-thick boards were 200 °C, 30 min, and approximately 7 MPa. The resultant boards (Fig. 3) were processed to obtain specimens with the dimensions and geometry specified in the ASTM D1037 (2012) standard test method. Accordingly, the dimensions of the test specimens were as follows: 410 mm × 76 mm × 15 mm (flexural), 152 mm × 152 mm × 15 mm (moisture absorption), 152 mm × 76 mm × 25 mm (nail extraction), and 102 mm × 76 mm × 25 mm (screw extraction).

Fig. 3. A WPC board with approximately 25-mm-thick

The samples for creep characterization had the maximum dimensions allowed, indicated in the instructions brochure of the TA Q800 Dynamic Mechanical Analyzer (DMA) that was used for the tests (TA Instrument, New Castle, DE, USA) (2010). Accordingly, they were 50 mm long × 15 mm wide × 7 mm thick. The processing conditions to obtain them were the same as those used for obtaining the flexural test specimens; the only difference was that 7-mm-thick laminates were obtained. These specimens were machined to obtain samples with the required dimensions. Pellets with PR M4mm, R30, and TF without UVS were also used, respectively. The authors chose to use samples with the maximum dimensions because PR particles with dimensions up to 4 mm may exist in some of the studied WPCs.

Samples with UVS

Flexural tests samples with and without UVS and PR TF (Table 2) were prepared using the pellets obtained *via* the twin-screw-extrusion procedure detailed above. Pellets were hot-pressed using the same hydraulic press and processing conditions previously mentioned to obtain the creep characterization test specimens. In this case, 3-mm-thick flat plaques were obtained and the samples were cut from them. The test specimens' dimensions were those specified in the ASTM D790 (2015) standard test method (127 mm × 12.7 mm × 3.2 mm).

Density determination

The density of the 15-mm-thick and 25-mm-thick boards without UVS was determined following method A of ASTM D2395 (2017) standard. The boards were processed to obtain specimens with the following dimensions: 30 mm × 30 mm × 15 mm, and 30 mm × 30 mm × 25 mm. Ten samples of each board were evaluated.

Moisture absorption test

Composites PR M4mm, PR R30, PR TF without UVS, and material MDF15 were tested. The samples (10 replicates per material) were conditioned to a constant weight and were then placed in a container with distilled water. The samples were weighed at 2 h and 24 h after immersion to determine the percentage of absorbed water. The test was performed following the ASTM D1037 (2012) standard test method.

Accelerated weathering tests

An Uvcon tester (ATLAS MTT, Moussy Le Neuf, France) was used to expose test samples to 24 h cycles of continuous UV light irradiation at 60 °C with UVB-313 type fluorescent lamps (Atlas Electric Devices, Chicago, IL, USA). The ASTM G154 (2012) and ASTM D4329 (2013) standards were considered as references. Prior to their exposure, samples (10 replicates per material) were conditioned according to the ASTM D618 (2013) standard (105 °C for 24 h). The samples were subjected to weathering cycles for 0 h, 384 h, 576 h, and 1000 h and will be referred to throughout the text as 0AW, 384AW, 576AW, and 1000AW, respectively. The experiments were performed on the flexural test samples with and without UVS and PR TF.

Methods

Flexural characterization

Composites PR M4mm, PR R30, and PR TF, and material MDF15 were subjected to three-point bending tests (Fig. 4) using a universal testing machine (model AGS-X, Shimadzu Scientific Instruments, Columbia, MD, USA). Ten samples of each material were evaluated according to the ASTM D1037 (2012) standard. The tests were conducted at a crosshead speed of 7.2 mm/min with a 5 kN load cell.

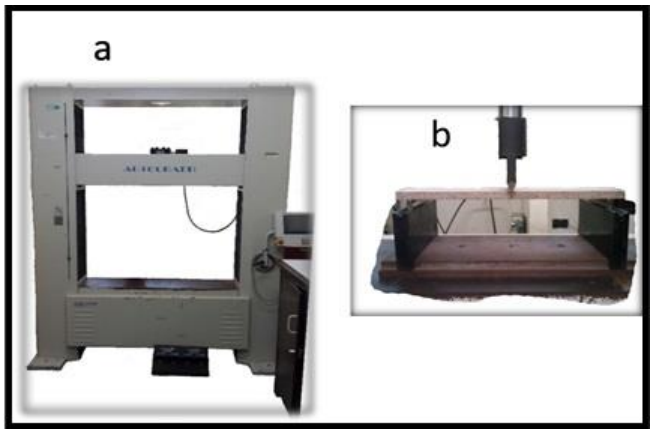

Fig. 4. Bending test: (a) universal testing machine and (b) test configuration

Composites PR TF with and without UVS (Table 2) and subjected and non-subjected to AW were three-point-bending tested using an Instron® 5500R universal tester machine (model 1125, Norwood, MA, USA) according to ASTM D790 (2015). The tests were conducted at a crosshead speed of 10 mm/min, using a 500 kg load cell. Ten specimens of each kind were tested.

All of the test specimens were conditioned at 23 ºC ± 2 ºC and 50% ± 5% relative humidity for at least 40 h before testing according to ASTM D618 (2013).

Nail and screw extraction resistance

The tests were performed with the Instron® universal tester machine previously mentioned using a 500 kg load cell according to ASTM D1037 (2012). The tests were performed at 6 mm/min for nail extraction and 15 mm/min for screw extraction. Ten specimens of each kind were tested.

Creep characterization

The tests were performed in a TA Q800 DMA instrument (TA Instrument, New Castle, DE, USA) using a three-point bending system and applying a constant stress of 2 MPa for 30 min, which was subsequently released according to the methodology reported by Xu *et al.* (2011). The tests were performed at 25 °C. Five specimens of each kind were tested.

Scanning electron microscopy

Morphological analysis was performed *via* scanning electron microscopy (SEM) on the transverse fracture surfaces obtained by cryogenically fracturing flexural samples of composites PR M4mm, PR R30, and PR TF (Table 1), and material MDF15. The samples for examination were gold-coated using a sputter coater (Denton Vacuum Desk II, Moorestown, NJ, USA). The samples were examined with an electron microscope (JSM-6360 LV, JEOL USA, Inc., Peabody, MA, USA). The samples' surfaces of composites PR TF with and without UVS (Table 2) and subjected and non-subjected to AW were also analyzed.

RESULTS AND DISCUSSION

Density Determination

The average density of the 15-mm-thick and 25-mm-thick WPC boards without UVS is presented in Table 3.

Table 3. Density of the 15-mm-thick and 25-mm-thick WPC Boards without UVS

Composite	Density (kg/m^3)	
	15-mm-thick board	25-mm-thick board
PR M4mm	1,017 (± 3.20)	1,014 (± 0.30)
PR R30	1,054 (± 4.90)	1,052 (± 2.70)
PR TF	1,069 (± 16.90)	1,069 (± 0.10)
Standard deviation is indicated in parenthesis.		

As can be seen, the variation of the density of the boards with the same range of wood particle sizes was practically negligible. On the other hand, it was observed that the density tended to increase slightly when the amount of smaller wood particles in the WPC was increased. With respect to the MDF, the technical data sheet reports densities of 620 (± 25) kg/m^3 for both the MDF15 and the MDF25.

Moisture Absorption Test

Figure 5 shows the results for composites PR M4mm, PR R30, PR TF without UVS, and material MDF15 after being submerged in water. The MDF was highly hydrophilic with average absorption rates of approximately 72% and 87% after 2 h and 24 h, respectively, which contrasted with WPC's absorption percentages of lower than 0.2% and 0.5%, respectively. The reader should note that the absorption percentages corresponding to the WPCs were practically imperceptible in Fig. 5, due precisely to the great difference with the absorption percentages corresponding to the MDFs. The WPC´s absorption percentages are shown more clearly in Fig. 6. The marked difference between the absorption percentages of the MDF and the WPCs is due to the fact that the thermoplastic matrix in the WPCs is hydrophobic, which protected the wood particles by encapsulating them and preventing them from absorbing water. This function is particularly important, as wood particles are hydrophilic. However, water absorption can still occur mainly due to the presence of fine pores, micro-cracks, and defects in the interface, which are believed to occur to a greater extent in standard MDFs.

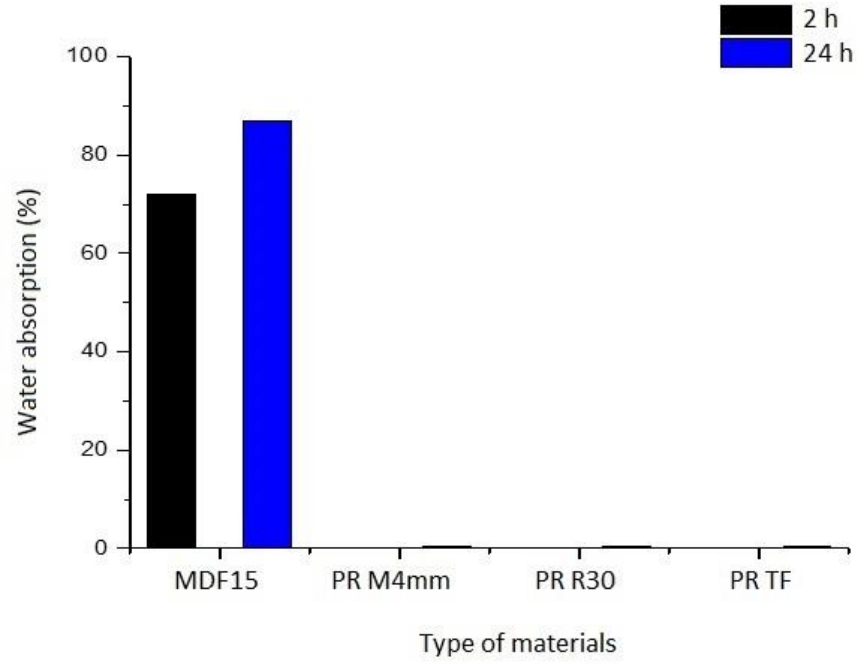

Fig. 5. Water absorption after 2 h and 24 h. The WPC´s absorption percentages are shown more clearly in Fig. 6

Unlike standard MDFs, WPCs exhibit less surface defects where water can contact the wood particles, which contributes to their very low water absorption rates. It should also be considered that, in the case of standard MDFs, only the use of a synthetic resin to

glue the wood particles during the manufacturing process does not guarantee an effective encapsulation of wood particles so as to avoid they absorb water. Differing from the MDFs, the extrusion process through which the WPCs were obtained favors a more effective mixing of the components of the material, which in turn propitiates that the wood particles are better encapsulated by the thermoplastic resin, which causes them to absorb less water. That is why it is recommended to apply a surface coating to standard MDFs to reduce water absorption, which, in most cases, is not necessary for WPCs. These results agree with the density difference between the WPCs and the MDFs, since the lower the number of surface defects or inside the materials, the higher their density, as it was the case for the WPCs. As such, it can be assumed that WPCs have much better fiber-matrix interphases than those of MDFs.

Figure 6 presents a comparison of water absorption in the different types of WPCs. As the wood particle size decreased, the percentage of water absorption also decreased. The composite PR TF had a better resistance to water absorption because it had smaller wood particles. Consequently, the surface area in contact with the polymer matrix was larger, which caused better adhesion between the wood particles and the matrix. As the wood particles were better encapsulated within the matrix, they were therefore more protected against water absorption. Again, these results are in agreement with the density of the WPCs (Table 3).

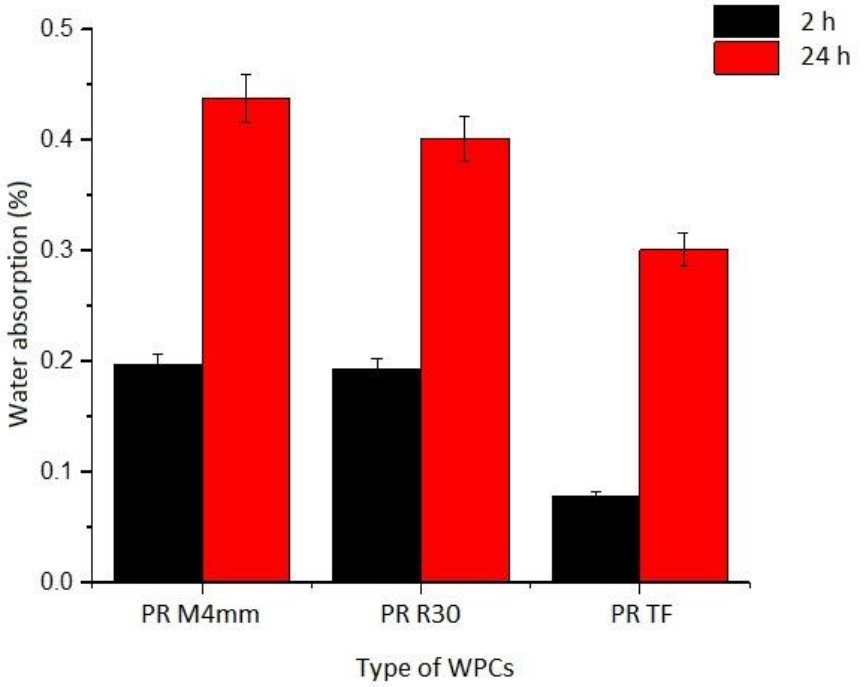

Fig. 6. Influence of wood particle size on water absorption of WPCs

Flexural Characterization

The average flexural strengths of composites PR M4mm, PR R30, PR TF, and material MDF15, are shown in Fig. 7. The WPCs had flexural strengths that ranged from approximately 18 MPa to 19 MPa.

The flexural strength of the MDF was approximately 12 MPa. In addition, the flexural strength of the WPCs increased slightly as the wood particle size decreased. As the wood particle size decreased, the average surface area that was in contact with the polymer matrix increased, which increased the bonding points, with a consequent increase in the interfacial resistance. This resulted in increased flexural strength as well. The low flexural strength presented by the MDF relative to those of the WPCs was because the former did not contain a coupling agent. In this regard, Chaharmahali *et al.* (2008) and Cui *et al.* (2010) commented on the positive effect of CAs on the mechanical performance of materials based on polymer resins and wood particles. These results agree with the density of the WPCs and the MDFs.

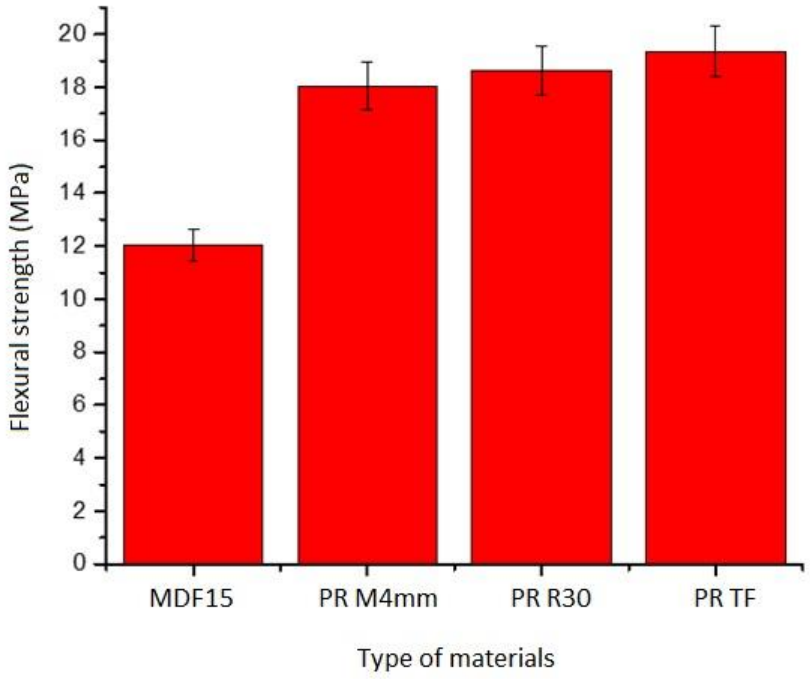

Fig. 7. Average flexural strength of composites PR M4mm, PR R30, and PR TF, respectively, and material MDF15

The flexural moduli of the different materials evaluated are presented in Fig. 8. The MDF modulus was higher than those of the WPCs, which suggested that it was more rigid and more susceptible to fracture under the same levels of applied flexural stress. These results suggested that the WPCs studied in this work have potential to replace MDFs.

Regarding the flexural properties of the WPCs, again, as the wood particle size decreased, there were larger surface areas, thus providing better load transfer between the wood particles and the polymer matrix. Also, it is very likely that wood particles with smaller sizes will disperse more homogeneously in the matrix, which caused the interaction at the wood particle-polymer matrix interface to be possibly stronger. Other authors have reported similar findings (Stark and Rowlands 2003; Khalil *et al.* 2006; Cui *et al.* 2008).

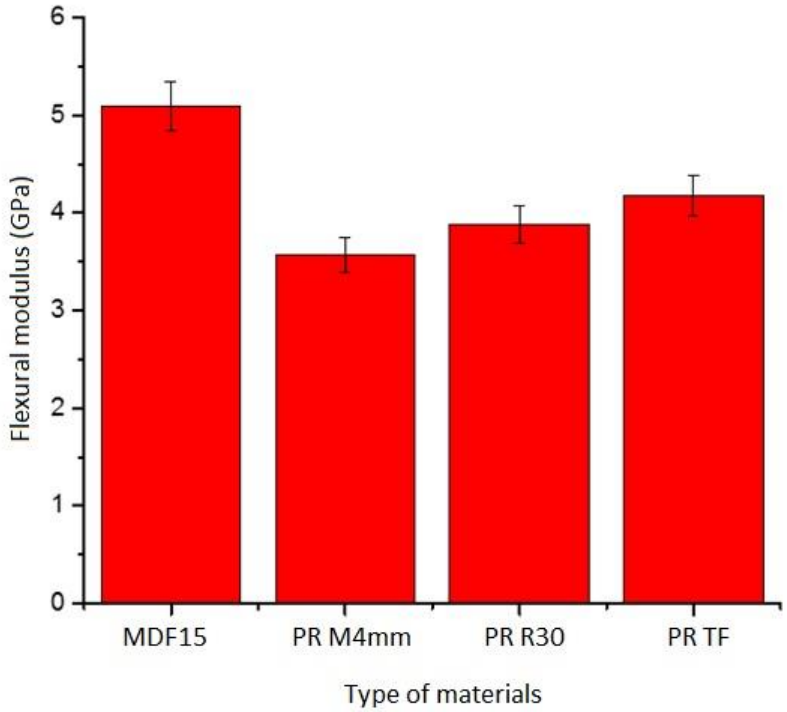

Fig. 8. Average flexural modulus of MDF and the WPCs

The average flexural strength of composites PR TF with and without UVS and subjected and non-subjected to AW is presented in Table 4. The reader should note that the average flexural strength of the composite PR TF presented in Fig. 7 is the same as that presented in Table 4 for the composite PR TF 0.0 UVS subjected to 0AW. As already mentioned before, both composites have the same composition. As can be seen in Fig. 7, the composite PR TF had the highest average flexural strength. For this reason, it was selected to analyze the effect of AW and UVS on the flexural strength of the material.

Table 4. Average Flexural Strength of Composites PR TF with and without UVS and Subjected and Non-subjected to AW

Composite	Flexural Strength (MPa)			
	Weathering Cycles			
	0AW	384AW	576AW	1000AW
PR TF 0.0 UVS	19.02 (± 0.39)	18.17 (± 0.58)	16.35 (± 0.78)	15.55 (± 0.24)
PR TF 0.5 UVS	18.96 (± 0.83)	18.40 (± 0.46)	17.71 (± 0.49)	16.91 (± 0.37)
PR TF 1.0 UVS	18.65 (± 0.78)	18.88 (± 0.65)	17.67 (± 0.42)	17.29 (± 0.61)
PR TF 1.5 UVS	18.96 (± 0.44)	19.07 (± 0.21)	17.66 (± 0.56)	17.46 (± 0.49)
Standard deviation is indicated in parenthesis				

The results presented in Table 4 show the effect of the weathering cycles and the content of UVS on flexural strength. The flexural strength decreased as AW increased. The WPC without UVS exposed to the largest period of AW (1000 h) was most notably affected. However, its flexural strength was higher (*ca.* 16 MPa) than that of the MDF (Fig. 7), which reinforced the hypothesis that the mechanical performances of the WPCs studied

in this work (as far as flexural behavior was concerned) were better. The variation in the flexural strength due to the exposure of the composites with and without UVS to AW was negligible. Although UV radiation damaged the surface of a WPC (it is a surface phenomenon), it alone was not entirely responsible for the decay of the mechanical performance of the material. Other factors must come into play to act synergistically with UV radiation to affect the material. Other authors have found similar findings. For example, studies have been conducted in which wood-HDPE composites were subjected to accelerated weathering, exposing them to either continuous UV light irradiation or cycles combining UV radiation and condensation (Stark and Matuana 2004, 2006; Stark 2006; Pech-Cohuo *et al.* 2016). These authors report that the observed loss of mechanical properties was due to moisture or moisture absorption-desorption cycles because of the damage caused on the WPCs' surface (appearance of cracks) by the UV radiation, which promoted moisture intake into the composite. López-Naranjo *et al.* (2013) examined the effects of termite attacks on AW and non-AW pinewood residue/HDPE composites and found that AW by itself did not produce a significant variation in the composites' flexural strength, but the combined effect of AW and exposure to termites diminished the composites' mechanical performance. Accelerated weathering produced cracks over the surfaces of all of the composites, leaving wood exposed to the environment and creating access routes to termites' mandibles, which caused significant changes in the mechanical properties. Ramírez-Chan *et al.* (2014) studied the effect of AW and *Phanerochaete chrysosporium* (Pc) on the mechanical properties of a composite prepared with discarded coir and recycled HDPE. Their results suggested that oxidative species were generated during exposure to AW and Pc. More damage was observed in the AW samples because Pc had better access to coir fibers, which was attributed to the damage on the samples' surfaces after exposure to AW. As presented in Table 4, the presence of UVS in the composites' formulations seemed to prevent the loss of their flexural strength. Similar results were obtained by other authors (Abu-Sharkh and Hamid 2004; Muasher and Sain 2006). As mentioned before, the effect of AW was negligible so as to contribute to the loss of flexural strength. However, the authors believe that major damage can occur long term if no UVS is used because it is likely that the appearance of cracks on the material's surface will increase, originated from chain scissions in the thermoplastic matrix. This can occur to different degrees depending on the formulation of the material as observed in Table 4.

Nail and Screw Extraction Resistance

Figure 9 shows the ultimate load (P_{max}) required to remove a screw from the specimens studied. As shown, the P_{max} was higher for the WPCs than for the MDF. For WPCs, the ultimate load tended to increase as the particle size of the pinewood residues decreased. It was noteworthy that the P_{max} for the composite made with PR TF was much larger than that of the other two WPCs. The variation in the P_{max} depending on the type of material and the size of the pinewood particles was similar to that observed in flexural strength. That is, the flexural strength was higher in the WPCs and it increased as the size of the pinewood particles decreased (Fig. 7). For the WPCs, higher P_{max} values were required to extract the screw than for the MDF. This was most likely because the threaded part of the screw adhered strongly to the polymer matrix in the WPCs (the WPCs were denser than the MDF) and when attempting to remove the screw, a matrix flow was

observed. The WPCs with the smallest pinewood particle sizes were most likely to generate more anchorage points with the screw thread. Consequently, a higher P_{max} was required for extraction.

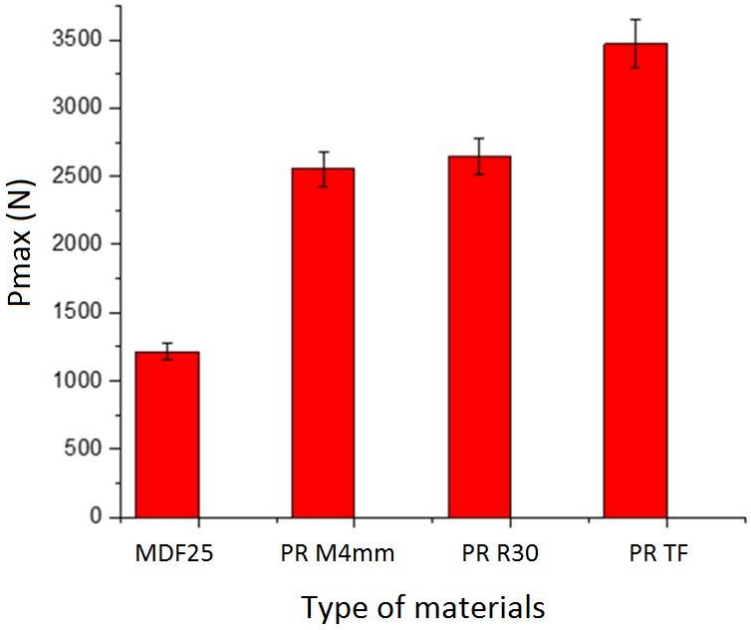

Fig. 9. Ultimate load (P_{max}) required to pull out a screw from the MDF and the WPCs

Figure 10 shows the P_{max} required to extract a nail from the specimens. Unlike the extraction of screws, the P_{max} was greater for the MDF than for all of the WPCs.

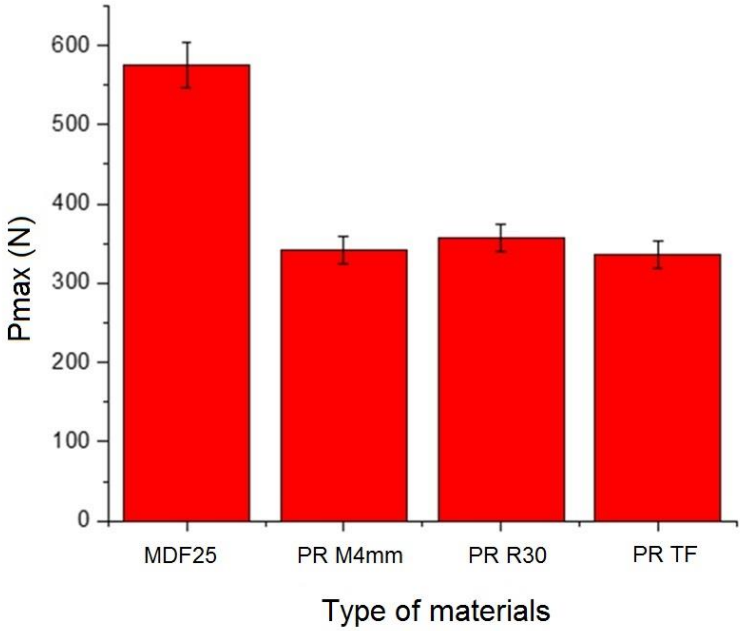

Fig. 10. Ultimate load (P_{max}) required to pull out a nail from the MDF and the WPCs

For WPCs, the variation of P_{max} was negligible. The P_{max} for the MDF was higher than that of the WPCs, which was likely due to the more heterogeneous particle composition of the MDF. Therefore, when the nail, which was not completely smooth, with a few millimetric reliefs, was introduced, the material around its surface had greater mobility. In addition, the way the particles were agglutinated gave them some freedom, which allowed them together with the binder resin, to be introduced into the reliefs of the nail, and provided a greater mechanical grip. In the WPCs, because the pinewood particles in the material were smaller, more homogeneous in size, and probably more compacted (in addition, the composite contained a coupling agent that strengthened the bonding with the polymer matrix), the particles did not have sufficient mobility to fill the reliefs of the nail. Therefore, the mechanical grip was smaller. Hence, the P_{max} was smaller than that required for the MDF.

In general, the results obtained were similar to others reported in previous literature. For example, Haftkhani *et al.* (2011) report that the withdrawal resistance of screws in WPC panels with 60% (W/W) wood flour, 30% (W/W) polymer (12% polypropylene, 18% polyethylene), and 10% (W/W) additives (4% talc, 4% calcium carbonate, 1% coupling agent, and 1% zinc stearate) was higher than that of MDF. They attribute this to the effect of the thermoplastic matrix through screw thread encapsulation. In addition, Chang *et al.* (2010) fabricated HDPE-based WPCs and evaluated the effect of formulation on nail withdrawal. They found that nail withdrawal was relatively unaffected by the different formulations and that the critical factor that affected the extraction resistance was the coupling agent, which can significantly improve the WPC properties. Studies done by Madhoushi *et al.* (2014) on polypropylene-based WPCs composed of MDF sawdust with maleated anhydride grafted polypropylene as a coupling agent showed that the withdrawal strengths of screws are much higher than those of nails.

Creep Characterization

The results presented corresponded to the composites PR M4mm, PR R30, and PR TF without UVS. It was not possible to test the MDF due to difficulty in obtaining good test specimens, because the wood particles began to separate when panels were cut to obtain specimens. This test was performed to elucidate the mechanical behavior under constant loads because WPC-based products are likely to experience it during their lifespan. Figure 11 shows the variation of strain over time. As shown, the composite with the largest amount of the largest wood particles (PR M4mm) deformed less. This may have been due to the fact that the presence of larger wood particles caused the material to creep less, making it behave more like wood, which is more resistant to deformation than the polymer matrix. Other authors have observed similar behavior (Xu *et al.* 2010). Xu *et al.* (2010) report that the creep resistance of a crushed sugarcane/recycled HDPE composite was the highest likely due to the larger particles of the lignocellulosic residues.

With respect to the MDFs, although it was not possible to test them, it has been reported that, unless a surface coating is applied to them, or they are subjected to some other treatment (for example, exposing them to a post-manufacture heat-treatment), they will experience higher levels of creep (Fernández-Golfín Seco and Díez Barra 1998; Ayrilmis *et al.* 2009). This happens because they absorb too much moisture. So, when they are in contact with water they swell, and a higher proportion of that swelling may not be

recoverable after drying. Basically, this happens due to the inherent hygroscopicity of the wood, and the residual stresses created during the product's manufacturing process. For these reasons, when a MDF panel has contact with water, the wood swells and some of that residual stress is released, causing an increase in the thickness of the panel.

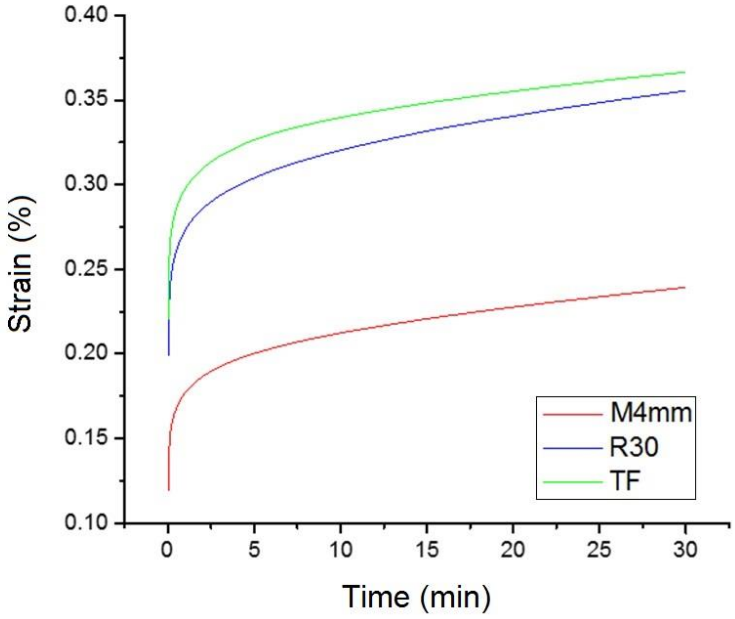

Fig. 11. Variation of strain with respect to time for composites PR M4mm, PR R30, and PR TF without UVS

Excessive thickness swelling not only causes a poor appearance, but also markedly weakens the material, which most of the time is reflected as a poor resistance to creep and deform under longer-term loadings. As noted already in this work, the MDF absorbed much more moisture than the WPCs under the same experimental test conditions. Thus, it is to be expected that they creep and deform more than the WPCs.

Scanning Electron Microscopy Analysis

Figure 12 shows a comparison of the cryogenic fracture surface of the material MDF15 with that of the composite PR TF without UVS. In the micrograph of the MDF, it was observed that cavities (indicated by red arrows) arose due to the extraction of wood particles during the fracture. This was attributed to poor adhesion between the wood particles and the polymer resin. In contrast, fewer voids were observed in composite PR TF, which suggested better adhesion between the wood particles and the polymer matrix due to the action of the coupling agent.

Figure 13 shows micrographs of the cryogenic fracture surface of composites PR M4mm, PR R30, and PR TF without UVS. The micrographs indicated that the wood particles had a strong bond with the polymer matrix due to the use of the coupling agent. Other authors, such as Zhang *et al.* (2008), suggest that this is due to the effectiveness of the maleic anhydride at the wood-HDPE interface reacting with the polar hydroxyl groups of wood to form strong covalent ester bonds.

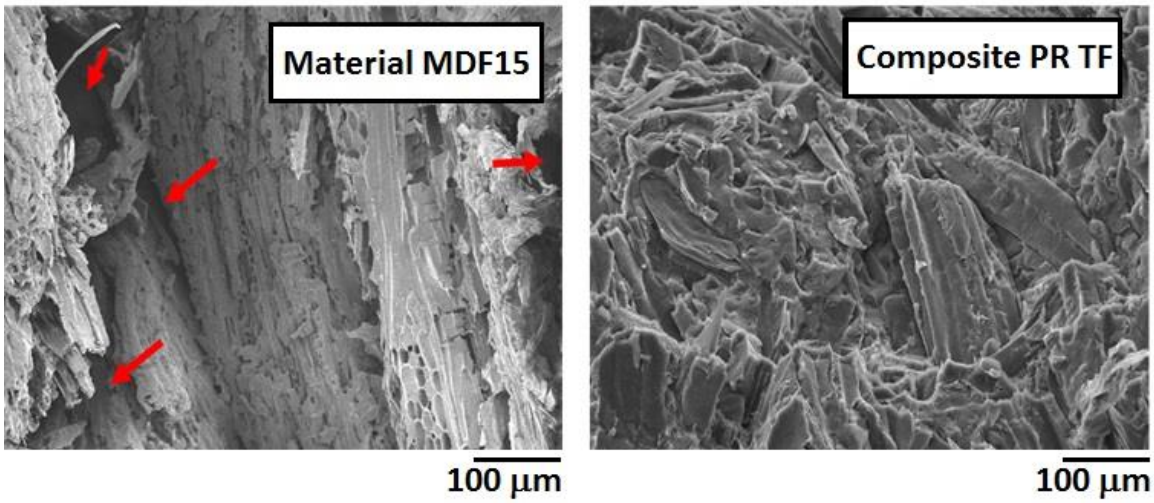

Fig. 12. SEM micrographs of cryogenic fracture surfaces of material MDF15 and composite PR TF without UVS; red arrows indicate cavities

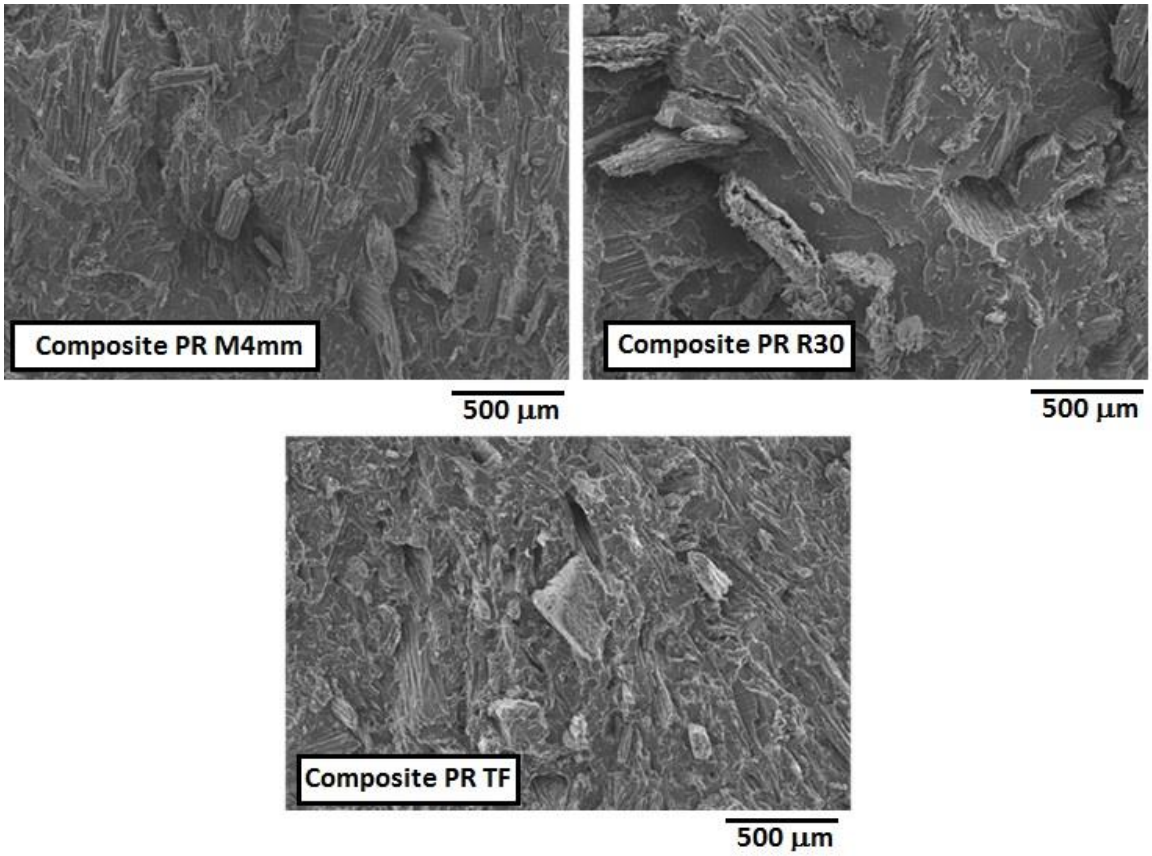

Fig. 13. SEM micrographs of cryogenic fracture surfaces of composites PR M4mm, PR R30, and PR TF without UVS

Zhang *et al.* (2008) also suggest that in WPCs that contain a coupling agent, the polymer matrix covers the surfaces of the wood particles and the fracture surface is rough, suggesting an increase in compatibility between cellulose and the polymer matrix. Other authors (Colom *et al.* 2003; Lai *et al.* 2003) also observed that after the fracture of composites when a coupling agent was used, their fracture surface was rough, and crusts were present on the surface of the wood. This suggested that there was good interfacial adhesion between the pinewood particles and the polymer matrix. Consequently, this improved the mechanical properties of the WPCs.

Figures 14 and 15 show the SEM micrographs of the surfaces of composites PR TF 0.0, PR TF 0.5 (Fig. 14), PR TF 1.0, and 1.5 UVS (Fig. 15) subjected and non-subjected to AW. It was observed that, for the non-weathered materials, the samples' surfaces were relatively smooth and the wood particles were well encapsulated by the polymer matrix. When the materials were exposed to 384 h of AW, cracks and holes appeared on those with 0% UVS, whereas the materials with 0.5% of UVS were less affected. In contrast, the materials with 1.0% and 1.5% UVS showed almost no damage on their surfaces. Upon exposure to 576 h of AW, the specimens without UVS showed larger cracks, which exposed wood particles to the environment. The contrary happened in the specimens with higher contents of UVS (*i.e.* the cracks on their surfaces were smaller). Exposure to 1000 h of AW caused laminar separation of the HDPE, which exposed and detached particles of the composites' disperse phase. Composites with 1.0% and 1.5% UVS content presented surface damage to a lesser extent. Similar findings are reported in previous literature. For example, Fabiyi *et al.* (2008) observed cracks in the surface of WPCs after exposure to weathering. They attributed it to chain breaks in the polymer matrix.

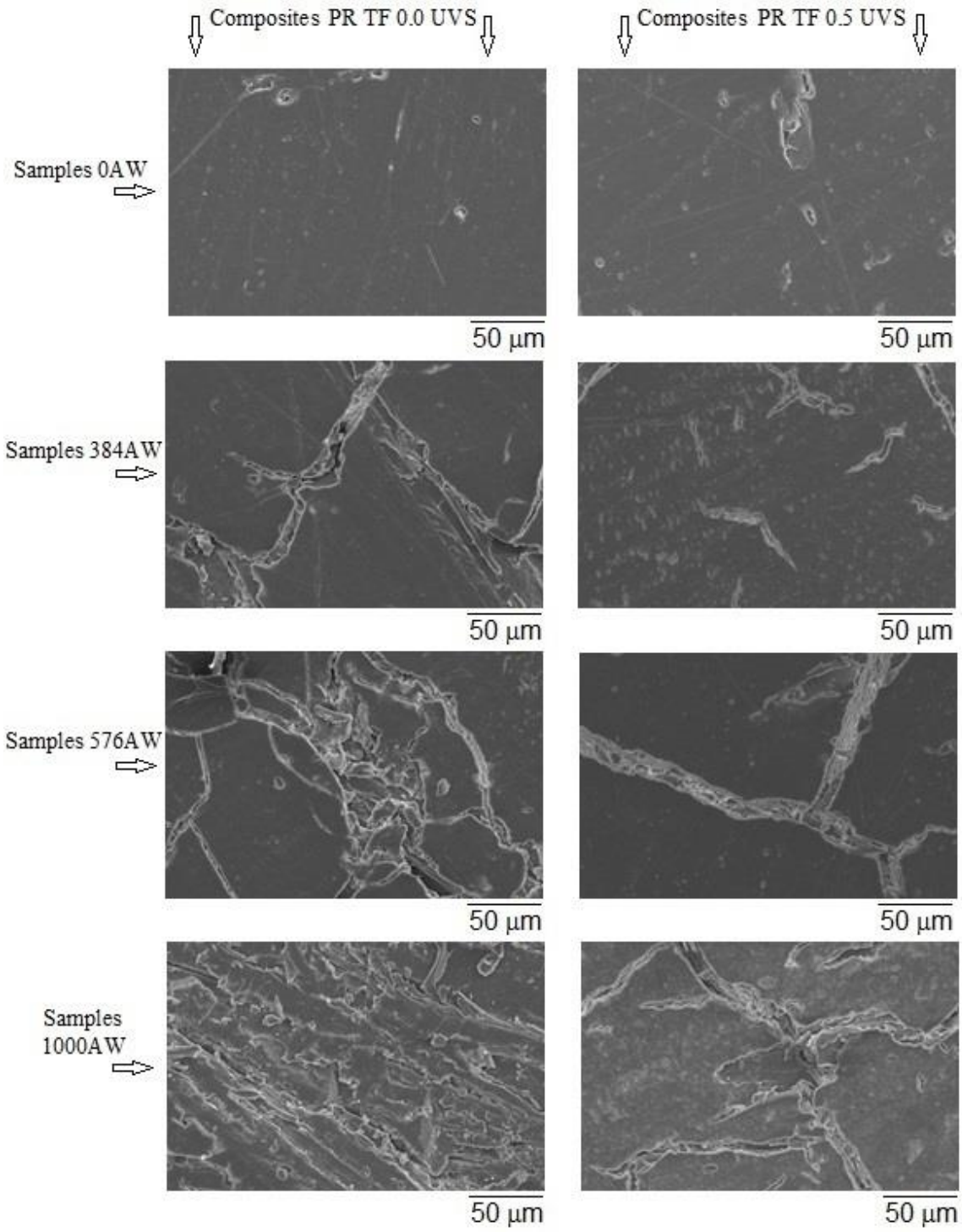

Fig. 14. SEM micrographs of surfaces of composites PR TF 0.0 and 0.5 UVS, subjected and non-subjected to AW

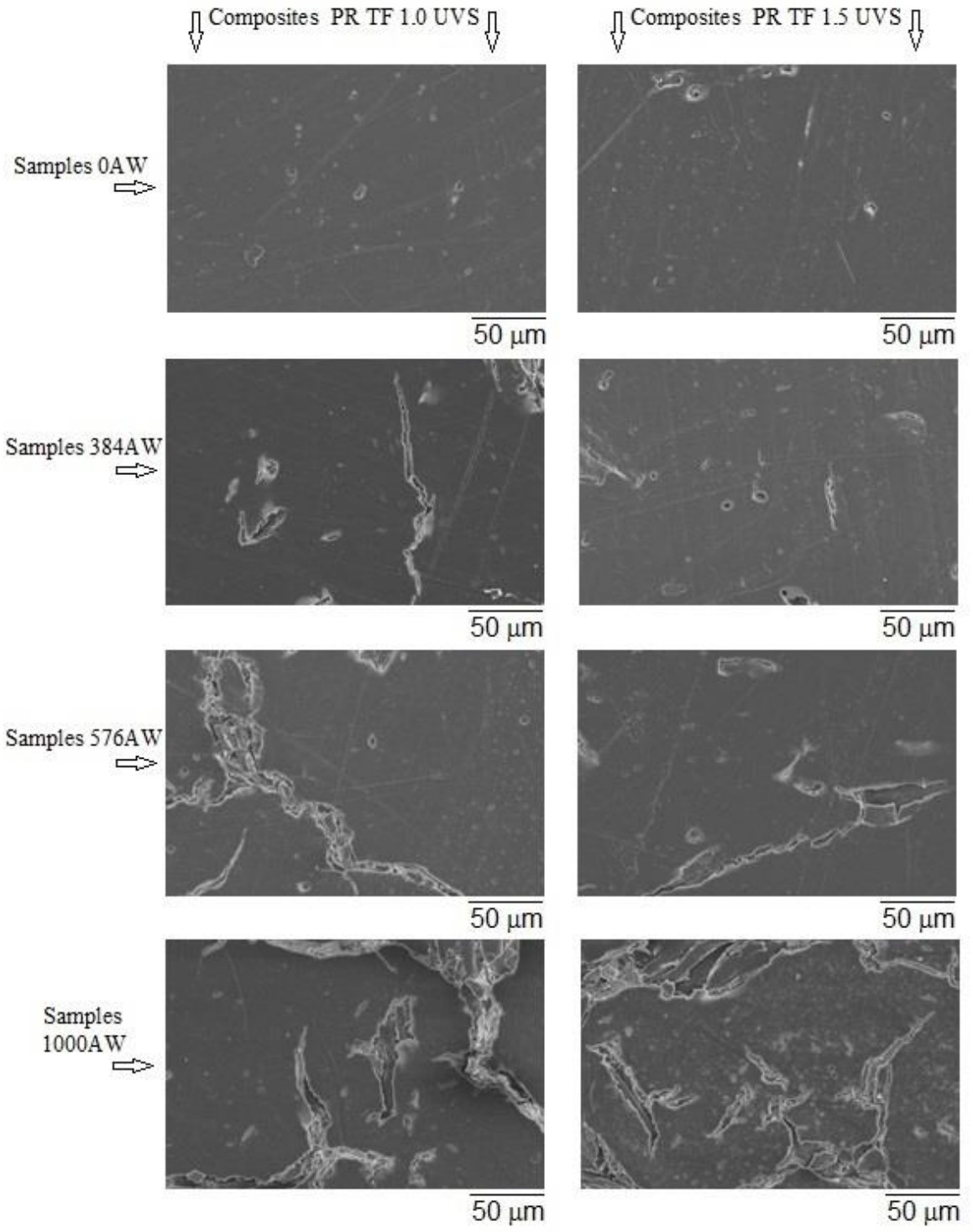

Fig. 15. SEM micrographs of surfaces of composites PR TF 1.0 and 1.5 UVS, subjected and non-subjected to AW

In general, the authors of this work consider that this is a valuable reference on some of the aspects to consider when proposing wood-based materials for a specific application. In the case of the WPCs, it was demonstrated that in general their performance as a construction material could be better than that of the MDFs. Regarding the effect of the AW and the UVS on the WPCs, is was found that their effect was practically negligible so as to contribute to the loss or not of the flexural strength. However, major damage appears to occur long term if UVS are not used because it will result in the appearance of more and larger cracks on the materials' surface due to chain scissions in the thermoplastic matrix. This can occur to different degrees depending on the formulation of the material. The authors emphasize that the effect of other elements of the environment, or the climate (whether biotic or abiotic, or their combined effect) on other characteristics of the materials (for example, their aesthetics, their performance under tension, compression, *etc*.) must be evaluated depending on the application intended to be given to them. On the other hand, we also reported that the MDFs studied in this work absorbed much more humidity than the WPCs under the same experimental test conditions. In this regard, we commented that the very high hygroscopicity of MDFs makes them more prone to experience higher levels of creep. In the literature there are reports that propose solutions for this problem. Especially worth noting among them are those that propose to apply a surface coating, or expose the product to a post-manufacture heat-treatment, among others. The authors of this work invite the readers to carry out an exhaustive search for scientific and technological literature related to the topic to better understand the behavior of these materials during a specific application. This will make it possible to establish their advantages and disadvantages, and mainly, to determine which are the most viable options to solve the problems they will face.

Future Work

Additional research can be carried out to a later stage to investigate the following:

- The effect of other elements of the environment or the climate on other characteristics of the WPCs, depending on the application that is intended for the material. It is suggested to subject the materials to accelerated and natural weathering, respectively. This will allow for estimating the durability of the materials by establishing a correlation between the damage occurring due to accelerated weathering and that due to natural weathering.
- In the same way, it is suggested to evaluate the combined effect of biotic and abiotic factors on the durability.
- Apply to the MDFs the most appropriate type of post-manufacture treatment, according to the intended application. Additionally, subject them to accelerated and natural weathering, and other biotic and abiotic degradation processes. This will be useful to estimate the durability of the materials.
- The findings from this research will be very useful for researchers and technologists to decide what type of material is best suited for a specific application. In the end, the authors of this paper consider that the decision will depend mainly on two factors: the functionality of the material, and the economic aspect.

CONCLUSIONS

1. The flexural strengths of the WPCs without UVS, which were not subjected to AW, were higher than those of MDF. The flexural strengths of the WPCs increased slightly as the particle size of the wood decreased. The MDF flexural modulus was higher than those of the WPCs, which suggested that it was more rigid and susceptible to fracture under the same levels of applied flexural stress. The flexural strength of the WPC without UVS exposed to the largest period of AW was the most affected. However, its flexural strength compared to that of the MDF was higher. This suggested that the WPCs studied in this work have potential to replace MDFs.
2. The MDF was much more hydrophilic than the WPCs. In the WPCs, it was observed that the percentage of water absorption decreased at small wood particle sizes.
3. The P_{max} values required to remove a screw from the WPCs were greater than those required for the MDF. In the WPCs, the P_{max} tended to increase as the particle size of the pinewood residues decreased. The P_{max} needed to extract a nail was greater for the MDF than for the WPCs.
4. The WPC with the largest amount of the largest wood particles deformed less under creep.
5. The SEM micrographs of the cryogenic fracture surface of the MDF showed much more cavities, which contrasted with the fracture surface observed in the micrograph of the WPCs without UVS. Scanning electron microscopy revealed that AW affected the WPCs with the lesser amount of UVS the most.

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