

## Studies on Mechanical Performance of Wood-Plastic Composites: Polystyrene-*Eucalyptus globulus* Labill

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The effects of size and concentration of wood particles on the properties of composites, obtained by extrusion, were evaluated based on polystyrene and wood particles from *Eucalyptus globulus* Labill. Wood-plastic ratios were 10:90, 30:70, and 50:50 (weight / weight), and wood particles were retained in 40, 50, 65, and 100-mesh sieves. The density, flow index, water absorption, and the mechanical properties were evaluated. Scanning electron microscopy revealed poor adhesion between the wood particles and the polystyrene. The size and content of wood particles were found to have a strong influence on the mechanical properties of the composite. The introduction of the wood particles induced a reduction of the Young's modulus, ultimate strength and deflection, as well as an increment in the elongation at break. The impact resistance also increased with the size and concentration of the wood particles. Furthermore, with increasing content of wood particles, the value of the melt flow index decreased and the water absorption rose.

*Keywords:* Wood-plastic-composite; *Eucalyptus* flour; Mechanical properties; Extrusion

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### INTRODUCTION

The growing attention given to environmental issues has forced industry to develop new sustainable techniques and processes. For this reason, research concerning environmentally friendly materials has been flourishing over the last couple of years. Composite materials are defined as materials formed by two components (phases) that can mix on a macroscopic level but are not miscible with each other. The quest for more versatile and sustainable composite materials that can function in a wide range of fields has generated a constant growth in the market for thermoplastic materials (Sheikh-Ahmad 2009). Wood-plastic composites (WPCs) are materials that have remarkable properties, such as high strength and durability, low density, and high elastic moduli (Sanjuan and Jasso 2009). Other characteristics that support their application are their low cost as a result of the vast availability of natural fibers (Lightsey 1983; Zadorecki and Michell 1989), an excellent dimensional integrity under humidity exposure (Maldas and Kokta 1991; Flores *et al.* 2014), and a significant resistance against fungus and termites (Mankowski and Morrel 2000; Verhey *et al.* 2001; Lomelí *et al.* 2009).

The market for WPCs is enormous because of the large-scale production of plastics and wood, constituting a noteworthy amount of solid waste that, unfortunately, is

not being used (Adhikary *et al.* 2008). Furthermore, WPCs, because of the huge quantity of cheap waste generated every day, hold a great promise as advanced components that can be used for new value-added products. Commercial products based on WPCs are replacing more and more products used in a variety of different applications, such as structural and construction applications (Youngquist *et al.* 1992; Yeh *et al.* 2009).

For WPC preparation, several thermoplastics have been used as matrices, such as polyethylene, polypropylene, polystyrene, and polyvinyl chloride. These materials are processed as thermoplastics; however, they have better overall properties than woods and plastics. The WPC technology includes the concepts of compatibility and processability, involving important challenges in the optimization of formulations, as well as processing and stability of composites in natural weathering conditions (El-Haggag and Kamel 2011).

Maldas and Kokta (1991) reported the effects of pairs of contrasting reinforcing materials, *i.e.* hybridization, on the mechanical properties of composites (based on polystyrene) of a mica/wood floor, treated on its surface. They demonstrated that the mechanical properties depend on the type of wood and treatment used on the fibers. Additionally, Simonsen and Rials (1996) applied a pretreatment using phenol-formaldehyde on wood fibers before processing the composite, finding evidence of interactions between the fibers and polystyrene. In another study, Kosonen *et al.* (2000) used coupling agents of poly(styrene-acrylic acid) and poly(styrene-methacrylic acid) and observed an increase in the ultimate tensile strength. Manikandan and Thomas (2003) investigated the effects of aging on the mechanical properties of composites based on polystyrene and short sisal fibers. They reported that the dimensional stability and mechanical properties of composites containing treated fibers are superior to those of composites without treated fibers. Furthermore, Lisperguer *et al.* (2007) studied the effect of the acetylation of wood on the thermal behavior of composites based on wood and polystyrene. Flores *et al.* (2014) reported the physical and mechanical properties of composites formed by polystyrene/white oak, finding that the melt fluid index and water absorption strongly depended on the wood particle concentration. Moreover, the Young's modulus increases as the size of the wood particles decreases.

Here, composite materials were prepared based on eucalyptus wood fibers and polystyrene. The mechanical properties as a function of the wood fiber size and concentration were evaluated. These composites have competitive mechanical properties and have the potential to be used in structural outdoor applications.

## EXPERIMENTAL

### Materials

The natural fibers used in this study were from eucalyptus (*Eucalyptus globulus* Labill), obtained from the State of Jalisco in Mexico. First, the fibers were triturated using a blade mill. The resulting wood flour was then separated by particle size and dried at room temperature for two weeks. The wood flour fractions of 40- (590 to 421  $\mu\text{m}$ ), 50- (420 to 298  $\mu\text{m}$ ), 65- (297 to 211  $\mu\text{m}$ ), and 100- (210 to 150  $\mu\text{m}$ ) mesh were employed for composite preparation. The moisture content of wood flour was between 2% to 3%. Polystyrene (PS) pellets, Resirene HF 777, with a density of 1.05  $\text{g}/\text{cm}^3$  and melt index of 7.87  $\text{g}/10 \text{ min}$  (200°C, 5 Kg), were acquired from Resirene plastic company, Mexico.

### Analysis of the Dimensions of the Wood Particles

The dimensions, particle length, and diameter of the wood particles were measured by optical microscopy (stereomicroscope Leica, from the MZ 7.5 series). The average L/D ratio was calculated according to,

$$\bar{\lambda} = \frac{\sum_{i=1}^n \left( \frac{L_i}{D_i} \right)}{n} \quad (1)$$

where  $\lambda_i$  is the  $L_i/D_i$  ratio. A total of 300 wood particles for each mesh were analyzed.

### Preparation of Composites

The polystyrene-eucalyptus wood mixture was processed using a co-rotating twin-screw extruder (Leistritz Micro 27 GI/GG 32D, Germany), equipped with a round profile die. A temperature ramp from 170 to 190 °C was selected. The resulting filaments were cooled, collected, and cut into pellets. Subsequently, the pellets were subjected to thermo-compression in a press (Schwabentan model Polistat 200T, Germany). The obtained materials were used to prepare the samples for testing according to ASTM standards. The samples are identified as PS (polystyrene), Eu (eucalyptus), 40, 50, 65 or 100 (retained particle size at the mesh) and at the end 10, 30 or 50 (concentration of wood particles). For example, the sample PSEu4010 is the composite that was filled with wood particles retained in the mesh 40 and with a concentration of 10 wt%.

### Water Sorption

For water retention studies, the samples (0.5 x 1.0 x 10.0 cm) were submerged in stagnant double-distilled water (pH 7) at room temperature for four weeks. After the samples were removed from the water, their surfaces were dried using blotting paper. Changes in weight were studied using an electronic balance with an accuracy of 0.1 mg. We calculated the diffusion coefficient ( $D$ ) according to the method described by Crank (1975) which states that,

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left(-\frac{D(2n+1)^2\pi^2 t}{l^2}\right) \quad (2)$$

where  $l$  is the thickness of the sample,  $M_\infty$  is the mass of water absorbed at the equilibrium and  $M_t$  corresponds to the mass of water absorbed in the sheet at time  $t$ .

### Melt Flow Index (MFI)

Eucalyptus flour concentrations of 10%, 30%, and 50% retained in each mesh (40, 50, 65, and 100 mesh) were tested. A Tinus Olsen Plastometer (USA) was used to measure the MFI according to ASTM D1238-13 (2013). A temperature of 190 °C for the PS composites (PS-E) was applied. To avoid thermal degradation of the wood flour, the lowest temperature was chosen.

### Mechanical Testing

Mechanical traction and flexion tests were performed in a United universal testing machine model ASMF-100 (Canada), with a speed run of 5.0 mm/min and equipped with a load cell of 8896.443 N for traction tests. ASTM D638-14 (2014) and ASTM D790-

15e2 (2015) were followed for traction and flexion tests, respectively. The speed run in the crosshead was 4 mm/min, and a load cell of 889.644 N was used for the flexion tests. The impact tests were performed in a Custom Scientific Instrument (USA) with a free fall impact weight of 0.1724 kg.

### Scanning Electron Microscopy (SEM)

Cross sections of the tensile tested samples were observed using a model TM-1000 Hitachi Tabletop Microscope (Japan). It was not necessary to pre-treat the samples since this SEM equipment does not require special preparation such as metal coatings.

## RESULTS AND DISCUSSION

The length/diameter ( $L/D$ ) relationship determines the mechanical resistance that a wood particle provides to the matrix (Migneault *et al.* 2009). Small diameters are ideal to reduce the number of surface defects and help to reduce crack propagation. Table 1 displays the results of the granulometric analysis and the  $L/D$  values for the four groups of sizes. As expected,  $L/D$  decreases when the particle size decreases. Based on the analysis made by Callister (2007), the wood particles used in this work can be considered short fibers since the values of  $L/D$  were below 10. Figure 1 shows the particles retained in the 40-mesh sieve.

**Table 1.** Granulometric Analysis Results and  $L/D$  Ratio

Mesh	Retention (%)	$L / D$
40 (507 $\mu\text{m}$ )	40.53	4.56
50 (359 $\mu\text{m}$ )	21.63	4.29
65 (254 $\mu\text{m}$ )	17.04	4.17
100 (130 $\mu\text{m}$ )	17.28	4.03
Residues fines	3.52	---



**Fig. 1.** Retained particles in 40-mesh sieve. Micrograph (10X lens)

The densities are reported in Table 2. In Fig. 2, the densities of processed PS samples as a function of the fiber concentration are shown. There was a close, linear relationship, having correspondent R-squared values of 0.992 (Mesh 40), 0.908 (Mesh 50), 0.966 (Mesh 65), 0.955 (Mesh 100). Concentrations of 10% showed an increment of

up to 4.04 wt% compared with the processed PS concentrations. For concentrations of 30%, this increment rose from 4.04 wt% to 7.07 wt% and, in the case of the concentration of 50%, the increment went from 8.08 wt% to 11.1 wt%. Evidently, when the concentration of the wood particles rises, the densities of the composites also increase. This is because of the higher density of the wood particles with respect to the density of the PS. This behavior is in agreement with the density results reported by Sommerhuber *et al.* (2016) and Porebska *et al.* (2015).

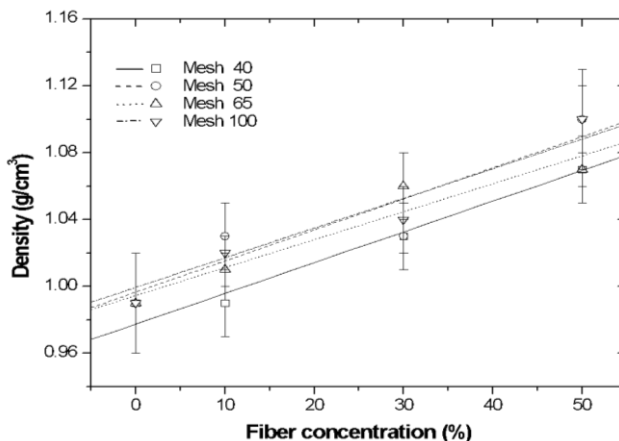


Fig. 2. Density as a function of fiber concentration

### Water Sorption

Table 2 shows the highest values of water absorption at equilibrium (168 h). The equilibrium was reached in approximately 96 h. In Fig. 3, the swelling kinetics in water are shown. The composites with higher concentrations of wood particles absorbed water at a faster rate over the first 24 h. The water absorption increased proportionally to the size and concentration of the wood particles. Changes in dimensions or deformations were not observed in the tested samples. Mishra and Verma (2006) and Tajvidi *et al.* (2006) suggested that this is due to the increased number of free OH groups present on the cellulose from the wood particles. When these OH groups are exposed to water, hydrogen bonds are formed, which results in weight gain in the composite. Water absorption by WPCs is an important indicator of quality because these materials absorb less humidity and at a slower rate compared with wood. Furthermore, WPCs are more resistant against the attack of fungus and have a good dimensional stability under humidity exposure (Clemons 2002; Yang *et al.* 2006). The water absorption capacity was affected by the nature of the wood particles and the thermoplastic matrix (Saeed *et al.* 2008). Ideally, the polymeric matrix embeds the wood particles completely, avoiding contact between the water and the particles. In reality, there is contact between the wood particles and the water on the surface of the composite. The capacity of water absorption is very slow because the underlying wood particles absorb water only through capillarity. The polymeric matrix affected this process. In this context, asymptotic behavior was reached after 72 h of immersion in distilled water.

The diffusion coefficient was calculated according to Crank (1975), in order to study the diffusive process of water inside the composite. The results of the water diffusion coefficient (Table 2) exhibit a linear relationship with respect to the quantity of wood particles. When the concentration increases, the water diffusion coefficient also increases, most likely because of more contact between the particles in the composite.

This is consistent with the results reported by Porebska *et al.* (2015) for PS/cellulosic fiber composites, Moscoso *et al.* (2013) for polystyrene/agave fiber composites, and Rao *et al.* (1984) for jute/epoxy composite systems.

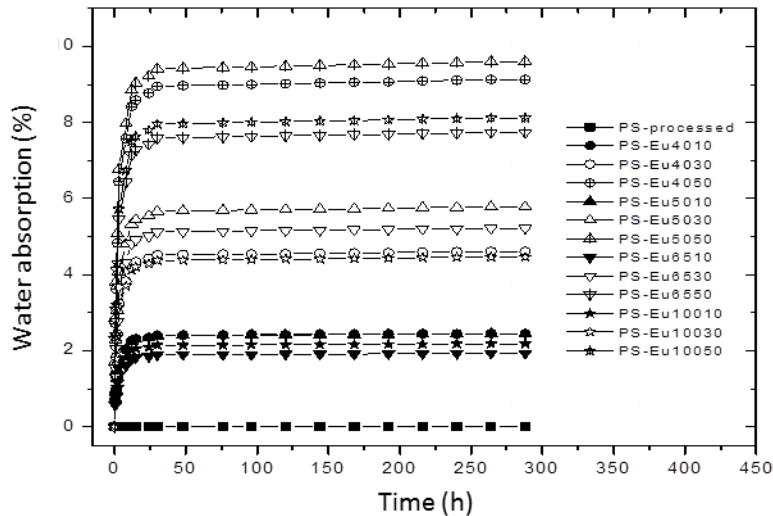


Fig. 3. Water absorption as a function of time

Table 2. Water Absorption, Density, and Melt Flow Index

Composites	Water absorption (%)	Water Diffusion Coefficient (10 <sup>-10</sup> m <sup>2</sup> /s)	Density (g cm <sup>-3</sup> )	Weight employed (Kg)					
				2.1	5.00	10.0	12.2	14.9	20.0
				Melt Flow Index (g/10 min)					
PS-Processed	0	0.32	0.99 ± 0.03	5.3	8.6	28.5	36.3	49.7	104
PS-Eu4010	2.45 ± 0.06	0.98	0.99 ± 0.02	3.9	11.4	24.5	---	---	---
PS-Eu4030	4.61 ± 0.09	1.15	1.03 ± 0.02	Nf	7.6	19.8	30.1	---	---
PS-Eu4050	9.13 ± 0.16	1.25	1.07 ± 0.01	Nf	Nf	Nf	6.5	14.6	17.2
PS-Eu5010	2.44 ± 0.08	1.07	1.03 ± 0.02	4.1	10.1	22.4	---	---	---
PS-Eu5030	5.78 ± 0.14	1.18	1.03 ± 0.02	Nf	7.7	20.7	31.8	---	---
PS-Eu5050	9.60 ± 0.19	1.28	1.10 ± 0.02	Nf	Nf	3.6	6.9	14.5	---
PS-Eu6510	1.93 ± 0.09	1.08	1.01 ± 0.01	4.2	10.8	26.4	---	---	---
PS-Eu6530	5.22 ± 0.15	1.22	1.06 ± 0.02	Nf	3.4	11.9	24.5	---	---
PS-Eu6550	7.74 ± 0.19	1.25	1.07 ± 0.02	Nf	Nf	4.2	9.7	14.6	---
PS-Eu10010	2.18 ± 0.07	1.07	1.02 ± 0.01	4.9	9.2	26.3	---	---	---
PS-Eu10030	4.46 ± 0.11	1.18	1.04 ± 0.02	Nf	8.3	21.3	28.5	---	---
PS-Eu10050	8.12 ± 0.18	1.23	1.10 ± 0.03	Nf	Nf	Nf	3.9	8.2	18.3

Nf: No flow; PS-Eu: polystyrene and Eucalyptus

**Melt Flow Index**

Table 2 displays the MFI values for each studied composition at various loads. Composites with 10% flow had the lowest weight (2.16 kg). On the other hand, composites with 30% and 50% did not flow at the temperature and weight of the test. In addition, an emanation of extractable gases (from the eucalyptus wood) was generated on those samples, preventing the completion of the test (Nf).

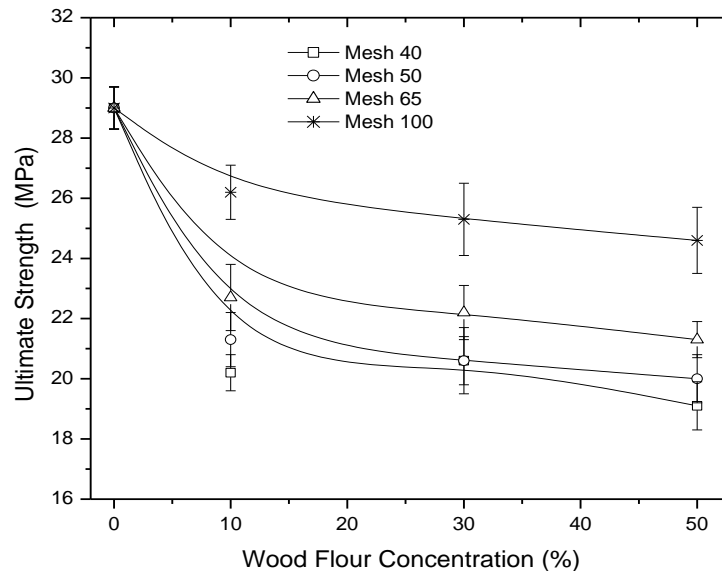
The capacity to flow of the thermoplastic matrix was affected considerable by the quantity of wood particles added. The test samples that showed the highest resistance to

flow were those that contained 50% wood particles, from the 40-mesh sieve. This result suggested that smaller sizes of particles could reduce the friction of the sample against the walls of the plastometer. The composites that contained wood particle concentrations of 10%, retained in 65- and 100-mesh sieves, presented lower resistance to flow against the walls of the capillary tube.

## Mechanical Properties

### Traction resistance

Table 3 shows the values of the tensile mechanical parameters. The values of the ultimate strength of the PS-Eu samples were reduced when the size of the wood particles increased and the concentration of the wood particles decreased. These are common behaviors for thermoplastic composites containing reinforcements (Simonsen and Rials 1996; Sanjuan and Jasso 2009). Their resistance to traction is strongly associated with the concentration of wood particles. The composites with concentrations of wood particles of 30% or more presented a reduction in their traction resistance values (Fig. 4). The elongation capacity of the composites increased as the size and concentration of the wood particles decreased. This behavior can be attributed to an inverse relationship between the size of the particles and the elongation. The composites made with wood particles from the 40- and 50-mesh sieves showed an elongation 50% higher than the elongation of the processed PS. For samples made from wood particles retained in the 65- and 100-mesh sieves, this increment in elongation was higher than 75%.



**Fig. 4.** Ultimate strength as a function of wood flour concentration

The Young's modulus showed a sharp reduction for all composites when the concentration of the wood particles augmented (Fig. 5). This behavior is attributed to a low compatibility between the PS and the wood particles. These values are comparable to those reported by Moscoso *et al.* (2013) for polystyrene/agave fibers composites.

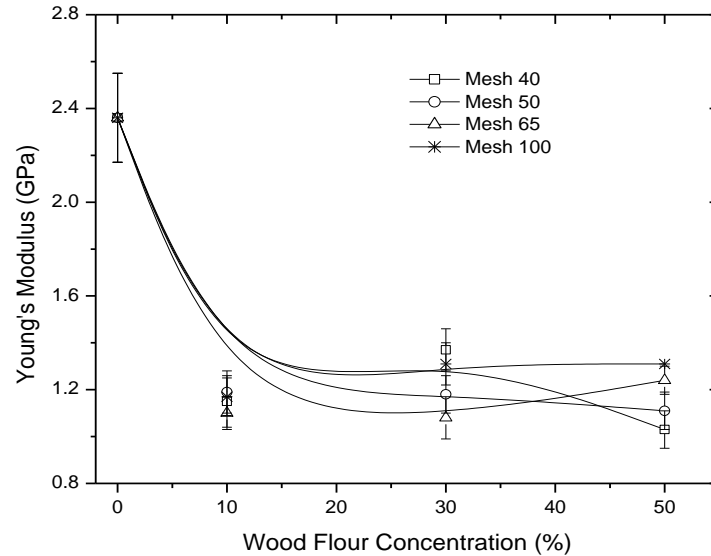


Fig. 5. Young's modulus as a function of wood flour concentration

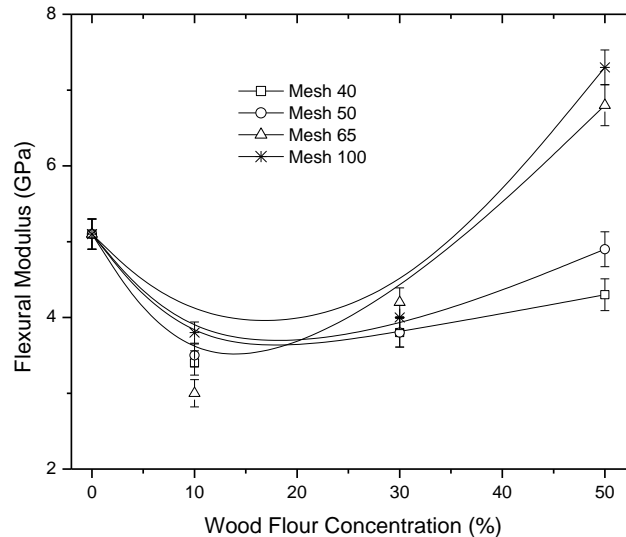
Table 3. Mechanical Properties of PS-Eu Composites

Formulations	Tensile			Flexural			Impact
	Ultimate Strength (MPa)	Elongation at break (%)	Young's modulus (GPa)	Strength maximum (MPa)	Deflection maximum (%)	Flexural modulus (GPa)	Bending Strength (KJ / m <sup>2</sup> )
PS-Virgen	31.9 ± 0.5	1.3 ± 0.01	2.53 ± 0.33	46.2 ± 5.0	2.2 ± 0.06	5.1 ± 0.18	0.77 ± 0.06
PS-Processed	29.0 ± 0.7	1.2 ± 0.02	2.36 ± 0.19	40.1 ± 4.7	1.8 ± 0.03	5.1 ± 0.20	0.74 ± 0.02
PS-Eu4010	20.2 ± 0.6	1.5 ± 0.08	1.15 ± 0.11	30.7 ± 1.7	1.2 ± 0.03	3.4 ± 0.16	0.64 ± 0.02
PS-Eu4030	20.6 ± 1.1	1.8 ± 0.09	1.37 ± 0.09	36.9 ± 1.7	1.2 ± 0.06	3.8 ± 0.19	0.80 ± 0.03
PS-Eu4050	19.1 ± 0.8	1.9 ± 0.11	1.03 ± 0.08	35.5 ± 1.6	1.1 ± 0.01	4.3 ± 0.21	1.05 ± 0.04
PS-Eu5010	21.3 ± 0.9	1.7 ± 0.12	1.19 ± 0.09	37.8 ± 1.7	1.4 ± 0.02	3.5 ± 0.15	0.62 ± 0.01
PS-Eu5030	20.6 ± 0.8	1.8 ± 0.09	1.18 ± 0.08	39.6 ± 1.5	1.4 ± 0.01	3.8 ± 0.19	0.83 ± 0.02
PS-Eu5050	20.0 ± 0.8	1.9 ± 0.13	1.11 ± 0.08	38.7 ± 1.4	1.2 ± 0.04	4.9 ± 0.23	1.14 ± 0.05
PS-Eu6510	22.7 ± 1.1	2.1 ± 0.09	1.10 ± 0.07	33.2 ± 1.1	1.9 ± 0.06	3.0 ± 0.18	0.75 ± 0.02
PS-Eu6530	22.2 ± 0.9	2.1 ± 0.08	1.08 ± 0.09	37.5 ± 1.4	1.4 ± 0.03	4.2 ± 0.19	0.86 ± 0.01
PS-Eu6550	21.3 ± 0.6	2.2 ± 0.11	1.24 ± 0.06	33.7 ± 1.3	1.2 ± 0.05	6.8 ± 0.27	1.24 ± 0.01
PS-Eu10010	26.2 ± 0.9	2.2 ± 0.17	1.17 ± 0.08	38.0 ± 1.8	1.9 ± 0.04	3.8 ± 0.14	0.86 ± 0.02
PS-Eu10030	25.3 ± 1.2	2.3 ± 0.14	1.31 ± 0.09	39.0 ± 1.5	1.6 ± 0.06	4.0 ± 0.16	0.96 ± 0.03
PS-Eu10050	24.6 ± 1.1	2.3 ± 0.09	1.32 ± 0.07	38.4 ± 1.7	1.1 ± 0.03	7.3 ± 0.23	1.44 ± 0.02

### Flexion Resistance

The results of flexion resistance tests are shown in Table 3. The size of the wood particle was found to affect the flexion resistance. The composites based on larger wood particles present a reduction in their flexion resistance. The maximum deflection of the composites with a concentration of 10% of wood particles, from the 65- and 100-mesh sieves, exhibited similar values to those of processed PS, while the rest of composites had lower values than the processed PS. The flexural modulus of the WPCs increased proportionally to the quantity of wood particles and with a decrease in the size of the wood particles (Fig. 6). This behavior may be related to the fact that the cellulose fibers have a greater flexion modulus than the polymer matrix (Bengtsson *et al.* 2007; Le Baillif and Oksman 2009; Poletto 2016), and also can be because of the orientation of the wood particles, which are perpendicular to the applied force.





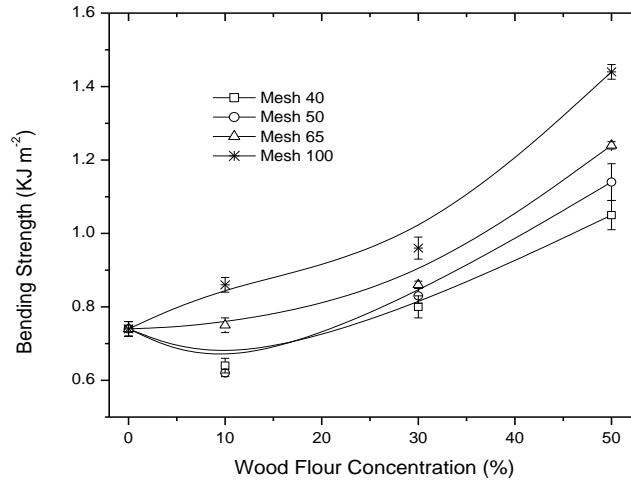
**Fig. 6.** Flexural modulus as a function of wood flour concentration

### Impact Bending Strength

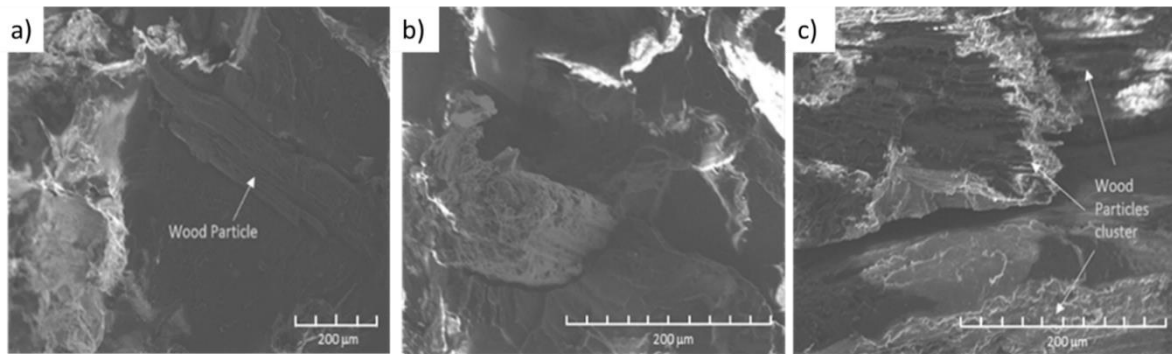
Table 3 shows the results of the impact tests. The energy needed to cause rupture in the material increased when the quantity of wood particles increased and the particle size decreased. The composites with a concentration of 10% of the wood particles from the 40- and 50-mesh sieves showed lower values than the processed PS (Fig. 7). According to Stark and Berger (1997a), an increase in the size of particles induces a reduction in the impact resistance because of the creation of a fracture. Larger sizes of wood particles provide higher stress concentrations, where a fracture can most likely form. In other words, less energy is required to create a fracture. This tendency was observed in this study with composites using larger wood particles (Flores *et al.* 2014). Some of the wood particles were totally covered by the polymer (Fig. 7), while others aggregated in groups, exhibiting a non-homogenous distribution in the composite (Fig. 7c). This has a negative effect on the interaction in the wood/polymer interphase. Regarding the composites with the smallest particles size, from the 65- to 100-mesh sieves, the impact resistance increased with increasing content of wood particles. The reinforcements provided concentrations of stresses, where less energy was needed to form a fracture. As the sizes of the particles increased, the energy required to create a fracture was reduced, which in consequence decreased the resistance to the impact. This tendency has been reported for many types of reinforcements (Stark and Berger 1997b).

### Microscopic Characterization

Figure 8a shows a single wood particle embedded in a homogeneous polymeric matrix with a low adhesion in the wood/polymer interphase. This was expected for a low concentration of wood particles. In Fig. 8b, the wood particles do not show good compatibility. This explains the reduction seen in the mechanical properties. The wood particles were aligned in the flow direction (Fig. 8c) in the WPCs that contain a concentration of 50% of wood particles from the 65-mesh sieve. The particles are well oriented in the direction of extrusion, showing a uniform distribution in the polymeric matrix and a good polymeric coating of the wood particles. The presence of agglomerates or clusters (Fig. 8c) promotes incompatibility with a hydrophobic polymer matrix reducing the composite mechanical properties (Le Baillif *et al.* 2009; Poletto 2016)



**Fig. 7.** Bending strength as a function of wood flour concentration



**Fig. 8.** SEM images of the cross sections of tested samples (traction)

## CONCLUSIONS

1. The tensile ultimate strength decreased when the concentration of wood particles was increased.
2. Composites with smaller sizes of particles combined with a higher content of reinforcements showed an increase in the flexural modulus. On the other hand, composites containing larger wood particles showed a reduction in the flexural modulus.
3. The increments seen in the bending strength were associated with higher contents and sizes of wood particles. Moreover, the MFI depends on the size and content of the wood particles, and lower MFI values are strongly associated with an increase in the concentration of the reinforcement.
4. The content of wood particles was associated with the water absorption behavior and the density. When the concentration of particles rose, both water absorption and density increased.

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