

## Maleated Polypropylene as Coupling Agent for Polypropylene Composites Reinforced with *Eucalyptus* and *Pinus* Particles

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Waste from the processing of hardwood and coniferous wood generated in the timber industries is difficult to dispose of and can cause considerable environmental impacts, such as soil and groundwater contamination. In this context, composites with varying concentrations of polypropylene, maleated polypropylene, and particulate *Eucalyptus* and *Pinus* waste were produced in a twin screw extruder and injection molded as test bodies for tensile and flexural tests. The morphology of the composites was investigated via scanning electron microscopy. The thermal properties were identified through differential scanning calorimetry. The tensile and flexural results for the two waste formulations indicated that the addition of vegetable fillers increased the modulus of elasticity and bending, and the compatibilizer provided increased resistance to stress and maximum deflection. The scanning electron micrographs illustrated the wetting of the cellulosic charge by the thermoplastic polymer with the compatibilizer, which corroborated the possible occurrence of an esterification reaction and hydrogen bonding interactions in the matrix-particle interface. The incorporation of waste in the composite resulted in the reduction of the degree of crystallinity of polypropylene, regardless of the use of the compatibilizer. This was explained by the barrier capacity of the charge, which prevented the growth of the crystals.

**Keywords:** Composites; Polypropylene; Maleated polypropylene; Solid waste; Wood

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## INTRODUCTION

Timber industry solid waste accumulation is a continuous process that requires storage spaces that do not pose a risk of fire hazard. However, part of this volume of waste is used by small farmers in breeding and soil fodder crops, as well as by briquette industries, fertilizer companies, and potteries in the generation of energy. Wood waste can also be used as reinforcement in the manufacturing of wood-plastic composites, commercially known as WPCs.

Wood-plastic composites are widely marketed in the United States and Europe in construction, automotive, and structural applications. In Brazil, the use of this product is still low, although Brazil is one of the largest producers of planted forests in the world, generating large volumes of wood waste that can be used in the preparation of WPCs.

Approximately 47 million tons of wood waste from the wood processing and forest harvesting industries were generated in 2015, of which 33 million tons (70.5%) were generated by forestry activities and 13.8 million tons (29.5%) by industrialists (IBÁ 2017). However, the distance between the sawmills and the processing industries, as well as the lack of knowledge of the characteristics of the waste generated in the mechanical processing of wood, prevents the increase of the production of WPCs (Clemons 2002). Therefore, knowing the properties of the waste and using them properly is important to produce ecologically responsible products with reduced cost. These materials can have advantages in the production of components with greater dimensional stability, lower weight, and easy processing (Malkapuram *et al.* 2009; Leong *et al.* 2014).

Waste from species such as *Eucalyptus* spp. and *Pinus* spp. are interesting alternatives for reinforcements in polymer matrix composites. The genus *Eucalyptus* presents a great diversity of qualities, including high forest productivity, adaptability to diverse environments, and fast growth. The genus *Pinus* has low density, fast and easy growing cycles, and a shorter processing time compared to other species. The automotive sector was first to use vegetal wastes of the genera *Eucalyptus* and *Pinus* in composites for the manufacture of composite pieces with an approximate weight reduction of 40% when compared with fiberglass (Leão *et al.* 2009). The use of wood waste as reinforcement in thermoplastic resins has been studied by many authors (Yamaji and Bonduelle 2004; Bledzki *et al.* 2005; Paes *et al.* 2011; Dai and Fan 2014). The inclusion of particulate wood waste in a thermoplastic matrix provides consistent results for commercial use (Hillig *et al.* 2008) and low cost in relation to inorganic loads, such as fiberglass, talc, CaCO<sub>3</sub>, and mica, among others. According to López-Manchado *et al.* (2002) and Parente and Pinheiro (2008), the addition of lignocellulosic charges in a thermoplastic matrix increases the modulus of elasticity and attenuates time-dependent phenomena such as creep and relaxation.

Today, it is common to manufacture WPCs with polypropylene (PP) resin thermoplastic and wood, using a compatibilizer to improve adhesion between the matrix and the load. Polypropylene is widely exploited in a variety of applications that when combined with favorable economic conditions and excellent thermal and mechanical properties has encouraged a rapid expansion in the use of this material (Rosário *et al.* 2011). The maleic anhydride-modified polypropylene additive (PPMA) is one of the most widely used in WPCs because it presents an excellent adhesion between the matrix-particle interface.

Results from the literature show that the use of this compatibilizer improves the mechanical properties of the composites produced (Stark 1999; Albano *et al.* 2001; Keener *et al.* 2004; Kim *et al.* 2008; Yeh *et al.* 2013; El-Sabbagh 2014; Haq and Srivastava 2017). In this context, the main objective of this work is to compare the mechanical, morphological, and thermal properties of reinforced composites with planted forest waste (*Eucalyptus* spp. and *Pinus* spp.) in a PP matrix with and without PPMA coupling agent. In the literature no work was found comparisons between these species no treatment using PPMA in the same matrix PP.

This research contributes to the development of new wood-plastic composites and the reduction of environmental impact caused by the disposal of *Eucalyptus* and *Pinus* waste.

## EXPERIMENTAL

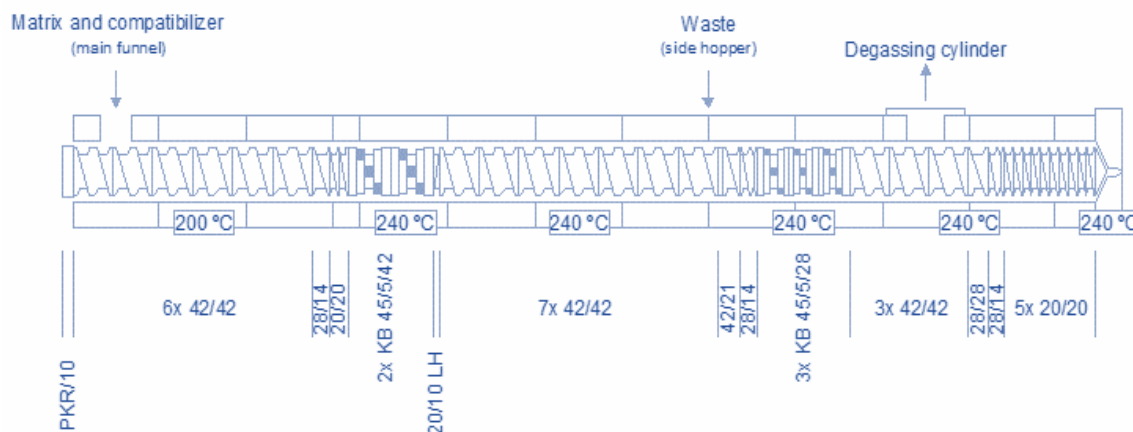
### Materials

The matrix used was the virgin isotactic homopolymer polypropylene (PP-Ho) in granules, grade H 301, flow index IF = 10 g/10 min (230 °C/2.16 kg), and a density of 0.905 g cm<sup>-3</sup> (Braskem, Sao Paulo, Brazil). The compatibilizer used was PPMA in granules, IF = 112 g/10 min (190 °C/2.16 kg), a density of 0.910 g cm<sup>-3</sup>, and 1% (w/w) maleic anhydride (MA) (Addivant, Danbury, USA). In its dispersed phase (ASTM D1921-18 and ISO 13322-14) as unmixed waste, granulometries were used in the range of 0.15 mm to 1.20 mm of *Eucalyptus* spp. (aspect ratio 0.4) and *Pinus* spp. (aspect ratio 0.5) without chemical treatment, a density of 0.11 ± 0.01 g cm<sup>-3</sup> and 0.20 ± 0.01 g cm<sup>-3</sup> with moisture contents of 9.9 ± 0.9% and 7.0 ± 0.2% for *Eucalyptus* and *Pinus*, respectively.

### Methods

#### Preparation of WPCs

The composites were prepared in a Werner-Pfleiderer ZSK-30 L/D 35 corrotative interpenetrating modular twin screw extruder (Coperion GmbH, Stuttgart, Germany) with a 100 rpm screw speed. Gravimetric feeders (Coperion K-Tron, Salina, USA) were used at a feed rate of 1.5 kg h<sup>-1</sup> (side hopper) feed and the feed rate of the polymer (main funnel) was varied to maintain the wood particles at concentrations of 10%, 20%, 30%, and 40% w/w. The use of PPMA at 1%, 5%, and 10% was determined according to the substitution of the equivalent quantity of polypropylene. For some formulations, only the concentrations of 20% and 40% w/w wood for 1% and 10% w/w of PPMA were tested, in order to evaluate the behavior of the largest intervals of each fraction. Figure 1 shows the mounting of the thread profile and the temperatures used in the six heating zones.



**Fig. 1.** Schematic of the thread profile and extrusion temperatures

Subsequently, specimens were molded from the extruded granules and oven-dried under vacuum at 80 °C for 12 h in an injector Arburg Allrounder (270V/300-120 L/D 30; Arburg GmbH, Loßburg, Germany) with the temperature profile of the feed to the injection nozzle at 210 °C, 215 °C, 225 °C, 230 °C, and 235 °C with an injection pressure of 450 bar.

### *Mechanical tests*

The tensile and flexure tests were performed in an EMIC universal test machine (DL 10000/700; Instron, Sao Jose dos Pinhais, Brazil). The tensile test followed the ASTM D638-14 (2014) standard using specimens with 'Type I geometry', a 1000 kgf load cell, 50-mm extensometer, and constant speed for 5 mm min<sup>-1</sup> assay. The flexural test followed the ASTM D790-10 (2010) standard using specimens with 'Procedure A' geometry, a 200 kgf load cell, and a deformation rate of 1.36 mm min<sup>-1</sup>.

### *Scanning electron microscopy (SEM)*

The adhesion between the matrix-particle interfaces was verified by means of the fracture surface of samples after tensile test. The observations were confirmed using a ZEISS® scanning electron microscope (EVO MA10; Carl Zeiss AG, Feldbach, Switzerland). The samples of the composites were fixed to a support with the help of double-sided self-adhesive carbon tape, metallized with a gold film, and analyzed at an acceleration voltage of 25 kV.

### *Differential scanning calorimetry (DSC)*

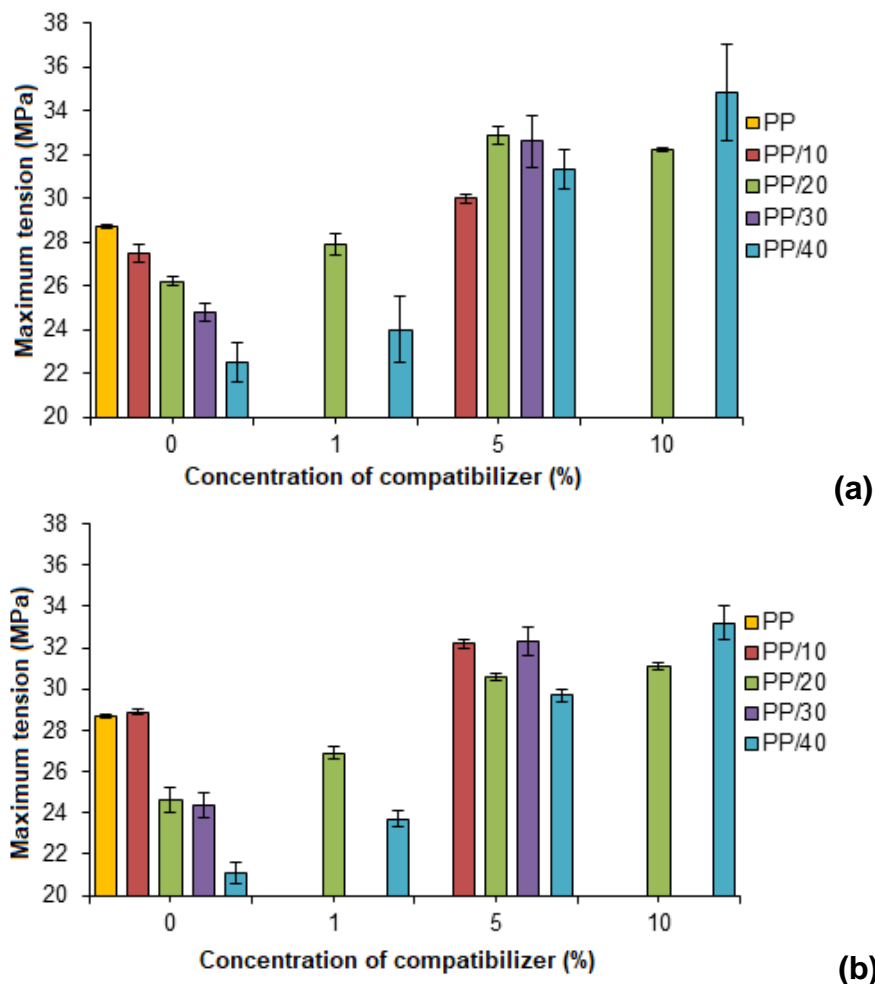
The samples were extracted from fractured traction bodies for thermal analysis. The tests followed ASTM D3418-12 (2012) standard and were conducted on DSC equipment, specifically a NETZSCH STA 449 F3 Jupiter® (NETZSCH-Gerätebau GmbH, Selb, Germany), at a heating rate of 10 °C min<sup>-1</sup> in the temperature range of 25 °C to 230 °C in an alumina crucible, inert argon atmosphere as the purge gas (60 mL min<sup>-1</sup>), and protection (20 mL min<sup>-1</sup>) at 0.5 bar. Subsequently, the samples were cooled to 25 °C and heated again under the same conditions. The samples were submitted to two heating cycles. The first cycle was to eliminate the thermal history of the material from extrusion and injection and the second was to analyze the thermal characteristics without influence of the processing conditions. The percentage of crystallinity of the polypropylene (Eq. 1) in the composites was calculated using  $\Delta H_m^o 100\% = 209 \text{ J g}^{-1}$  as reference (Hirayamaa *et al.* 2017),

$$X_c \% = \frac{\Delta H_m}{\Delta H_m^o \cdot (1-x)} \cdot 100 \quad (1)$$

where  $\Delta H_m^o 100\%$  is the melt enthalpy (J g<sup>-1</sup>) for crystalline PP and  $x$  represents the weight fraction of the particle in the composite. The values of melt temperature ( $T_m$ ), melt enthalpy of samples ( $\Delta H_m$ ), and percent crystallinity ( $X_c$ ) were taken from the curve of the second heating cycle.

## RESULTS AND DISCUSSION

The mean values obtained in the tensile tests for the PP/PPMA/*Eucalyptus* and PP/PPMA/*Pinus* composites are shown in Figs. 2, 3, and 4.



**Fig. 2.** Maximum tension variation for polypropylene and compatibilized and not compatibilized composites of PP/*Eucalyptus* (a) and PP/*Pinus* (b)

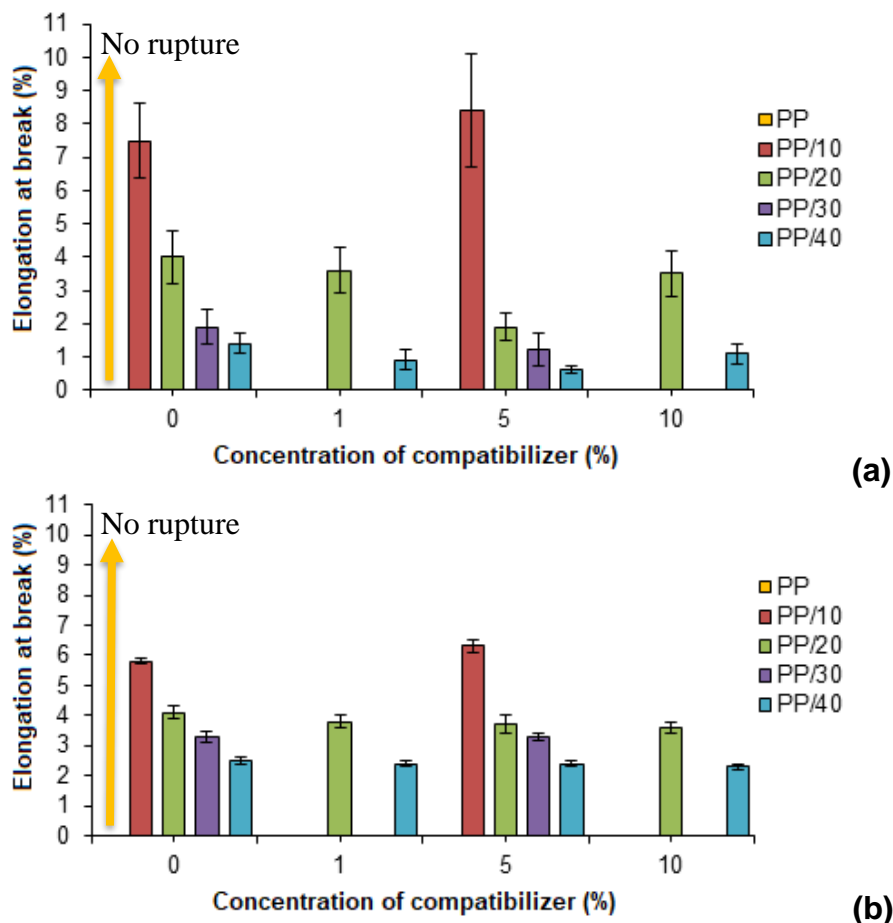
Particle-particle interaction, which is a combination of particle agglomeration and matrix-particle interaction, determines macroscopic behavior and material performance of a composite (Pukánszky 2005). Thus, it is observed in Fig. 2 that the maximum stress was reduced with the addition of wood particles in the polymer matrix, due to the possible propagation of cracks inside the composite caused by clusters of particles that acted as stress concentrators and, in this way, resulted in low mechanical strength of the material. Another indication was related to the weak matrix-reinforcement interaction caused by the lack of wettability between the reinforcement, which has a polar nature, and the polymer matrix, which has an apolar nature. It was noted that the PP/10%*Pinus* formulation in comparison with the thermoplastic matrix showed no decrease with the incorporation of the waste. This may have been related to the distribution of the charge in the matrix.

It was also observed that the same behavior of decline in the maximum tension with the addition of waste with 1% and 5% compatibilizer was observed for the PP/*Eucalyptus* composite. Unlike the PP/*Eucalyptus* and PP/*Pinus* composites with 10% compatibilizer, no decrease in tensile strength was observed with the waste concentration, in addition to a marked increase in the maximum stress supported by the material, in relation to the polypropylene matrix.

The addition of the PPMA compatibilizer led to a remarkable increase in the maximum stress when compared to the PP/*Eucalyptus* and PP/*Pinus* composites for the same mass waste concentration. This was associated with the possible improvement in the adsorption and interfacial adhesion as a function of the decrease in the surface tension between the phases that allowed a better transfer of tension between the matrix and the reinforcement. Thus, if the reinforcement percentage in the matrix is higher, the stress level supported by the composite will be higher as a consequence of the larger compatibilized matrix-particle interfacial area. Another possible factor for the improvement in wettability of solid surfaces and the consequent increase in maximum composite stress was due to the probable reaction of esterification and hydrogen bonding of cellulose with maleic anhydride modified polypropylene. According to Correa *et al.* (2003), this is due to a possible occurrence of mechanical anchoring or chemical interactions between the anhydride groups and the hydroxyl groups of the cellulose at the polymer-wood interface, promoting the effective transfer of forces from the matrix to the dispersed phase.

The PP/10%PPMA/20%*Eucalyptus* and PP/10%PPMA/20%*Pinus* showed an approximate 25% increase in the maximum stress, and PP/10%PPMA/40%*Eucalyptus* and PP/10%PPMA/40%*Pinus* exhibited an approximate 55% increase in the maximum stress, compared to the same not compatibilized formulations. The results for the PP/*Eucalyptus* and PP/*Pinus* composites with 1% and 5% of PPMA in relation to the not compatibilized composites and those with 10% PPMA presented interposed values. It was observed that the use of 10% compatibilizer was enough to avoid losses in the tensile strength of the polymer, even when 40% of wood particles were added in the PP/PPMA/*Eucalyptus* and PP/PPMA/*Pinus* formulations.

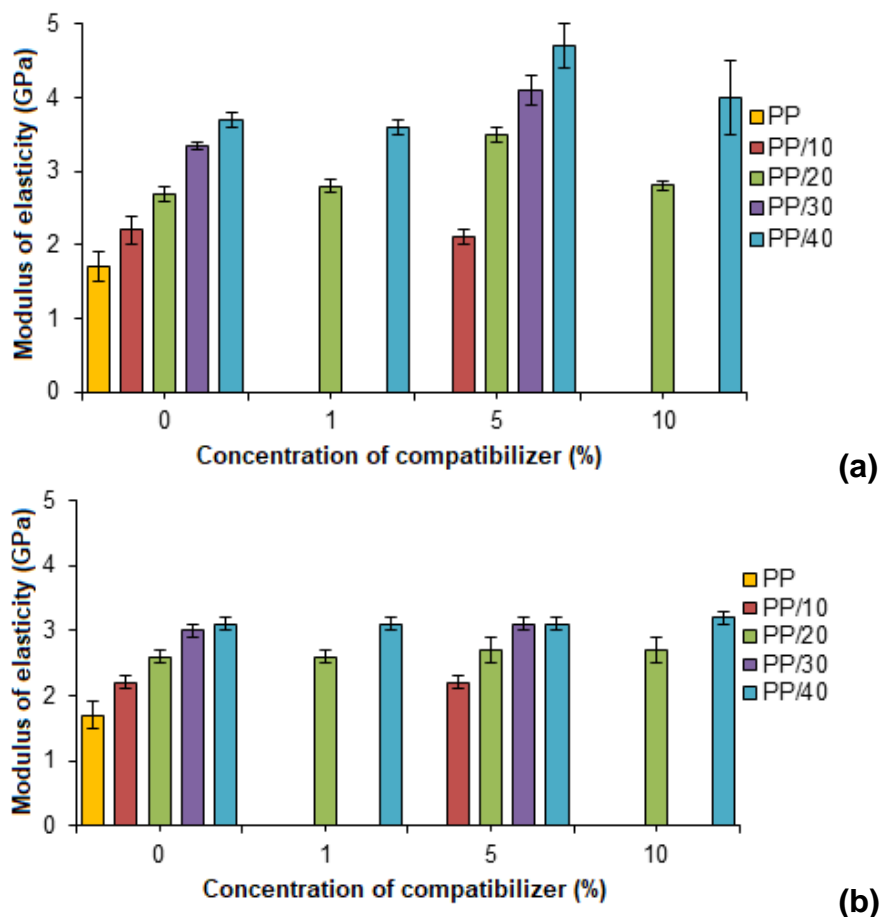
The elongation of the polymer is not shown in Fig. 3 because no rupture of the test specimens occurred at the extension limit of the claws of the test equipment. For all formulations, the ductility of the matrix decreased with the addition of waste, as the cellulosic load had high stiffness and low deformation. Lopez-Manchado *et al.* (2002) found that the addition of organic charges in a composite reduces the elongation at rupture because the fillers act as a reinforcing agent in the polypropylene, reducing the mobility of the polymer chains of the matrix, making the composite hard.



**Fig. 3.** Variation of elongation at break for polypropylene and compatibilized and not compatibilized composites of PP/*Eucalyptus* (a) and PP/*Pinus* (b)

In the composite materials, voids were formed during phase processing, which, in addition to acting as voltage concentrators, narrowed the cross-section in which the force was applied. The concentration of particles in the composites also acted as stress concentrators, causing material rupture with a lower deformation.

The addition of compatibilizer had no remarkable influence on the elongation and also did not produce noticeable effects on the modulus of elasticity of some formulated composites, but the increase in the modulus was evident with the increased concentration of the load, as shown in Fig. 4.



**Fig. 4.** Variation of modulus of elasticity for polypropylene and compatibilized and not compatibilized composites of PP/*Eucalyptus* (a) and PP/*Pinus* (b)

Polymer matrix composite materials usually have a higher modulus of elasticity than the matrix polymer. Thus, Fig. 4a shows that the addition of *Eucalyptus* particles in the thermoplastic matrix increased the elastic modulus approximately 30%, 60%, 95%, and 115% with the addition values of 10%, 20%, 30% and 40% w/w of waste, respectively, in the compositions without compatibilizer.

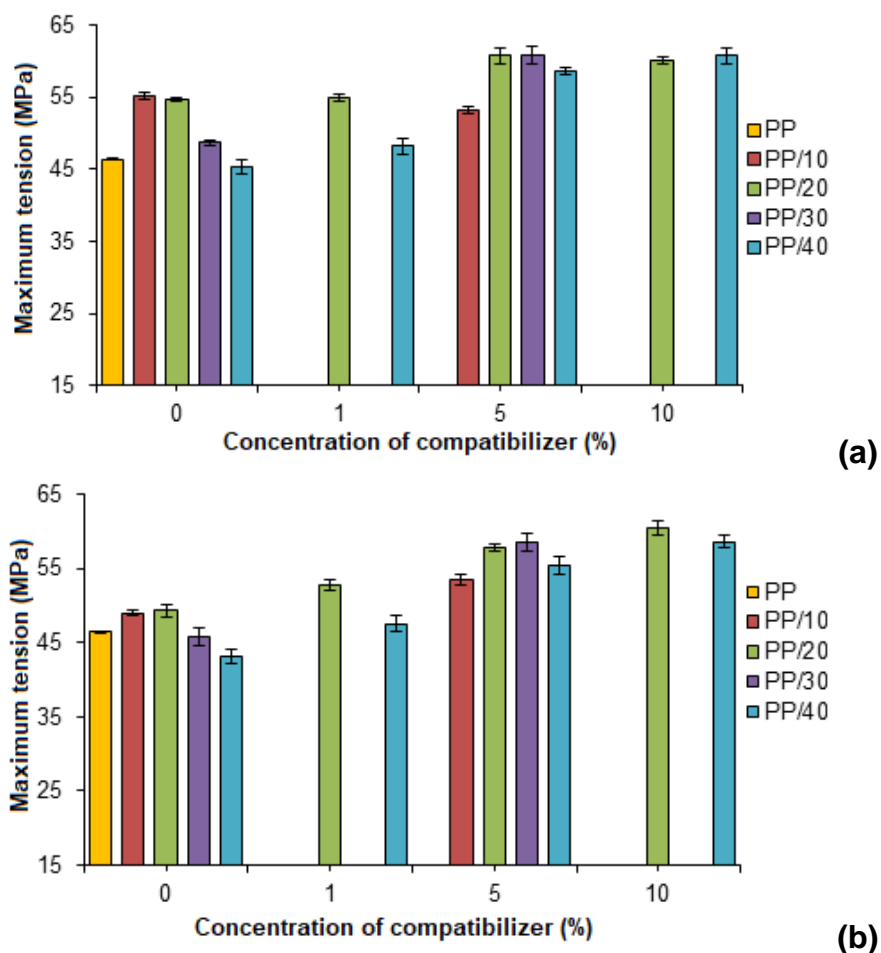
Figure 4b illustrates that an incorporation of *Pinus* particles in the matrix led to an increased modulus; this was expected because the tendency of composite materials is to increase stiffness as a higher percentage of residue in the matrix is added as reinforcement. The gains in the modulus were approximately 30%, 55%, 75%, and 80% for values of 10%, 20%, 30%, and 40% w/w of particulates, respectively, in the not compatibilized formulations.

According to Caraschi and Leão (2002), the composites have a higher modulus of elasticity with respect to the polymers because the incorporated dispersed phase has higher stiffness than the matrix and acts as a barrier to the mobility of the polymer chains of the polymer, thus avoiding deformation. Therefore, the increase in the modulus of elasticity *via* the incorporation of added waste was already expected due to the greater limitation to the deformation of the polymer. Studies have shown an increase in the modulus of elasticity with the addition of wood waste in a polypropylene matrix when compared to the polymer matrix (López-Manchado *et al.* 2002).

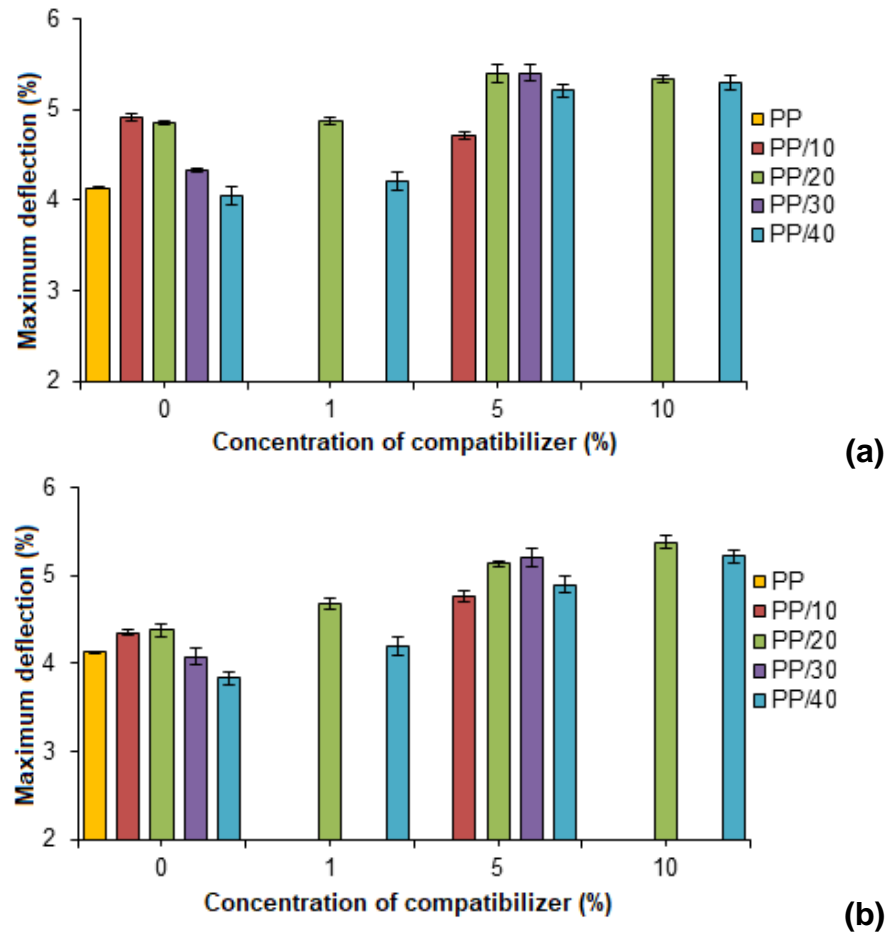


The results showed that there was influence in the modulus of tensile elasticity of polypropylene composites with 20%, 30%, and 40% w/w of *Eucalyptus* waste with 5% compatibilizer, due to a better transfer of the loading between the polymer matrix and dispersed phase. This result was in agreement with those obtained by Nuñez *et al.* (2003) and Qiu *et al.* (2003) in studies on compatibilization systems of PP/wood particles. An indication of matrix-particle interfacial interaction in mixtures with 20% and 40% of *Eucalyptus* waste that contained 1% and 10% of PPMA was observed, but without modulus increase. The presence of the compatibilizer did not have noticeable effects on the elastic modulus under traction of the PP/*Pinus* composites. Similar results obtained by Yeh *et al.* (2013) showed that the modulus of elasticity did not vary with the addition of compatibilizer PPMA in the PP composite/wood particles. Probably due to poor wettability of substrate with matrix-PPMA.

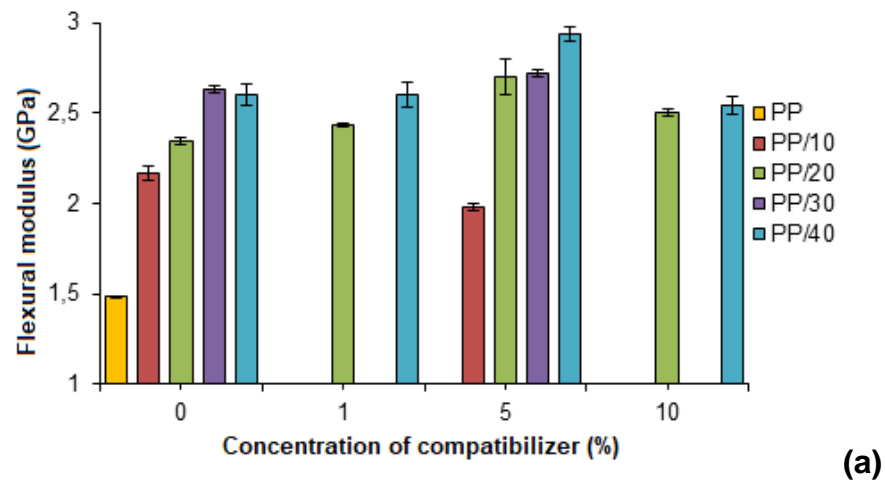
Figures 5, 6, and 7 show the average values obtained in the PP/PPMA/*Eucalyptus* and PP/PPMA/*Pinus* composite bending tests.

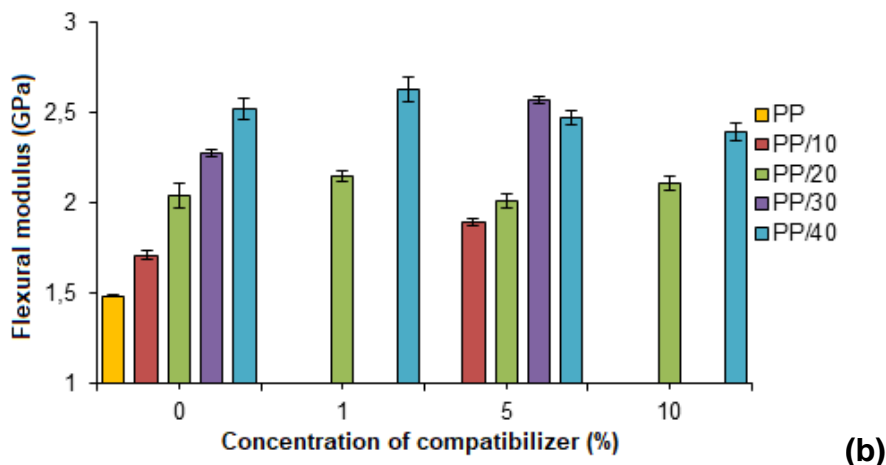


**Fig. 5.** Maximum tension variation for polypropylene and compatibilized and not compatibilized composites of PP/*Eucalyptus* (a) and PP/*Pinus* (b)



**Fig. 6.** Variation of maximum deflection for polypropylene and compatibilized and not compatibilized composites of PP/*Eucalyptus* (a) and PP/*Pinus* (b)



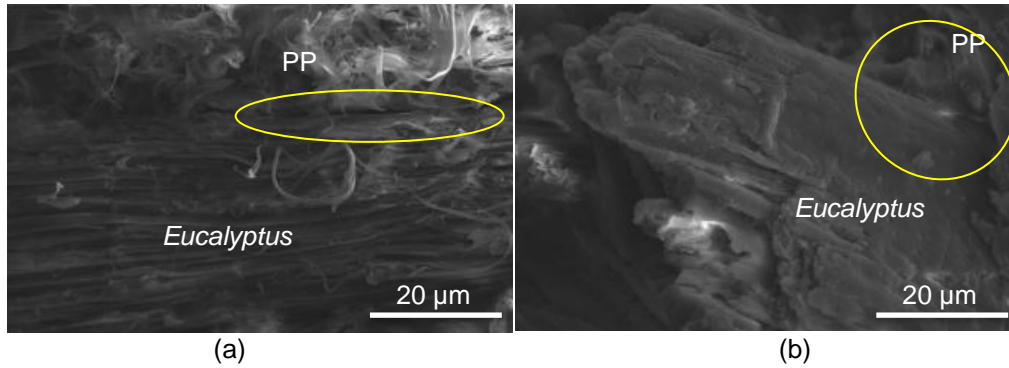


**Fig. 7.** Variation of flexural modulus for polypropylene and compatibilized and not compatibilized composites of PP/*Eucalyptus* (a) and PP/*Pinus* (b)

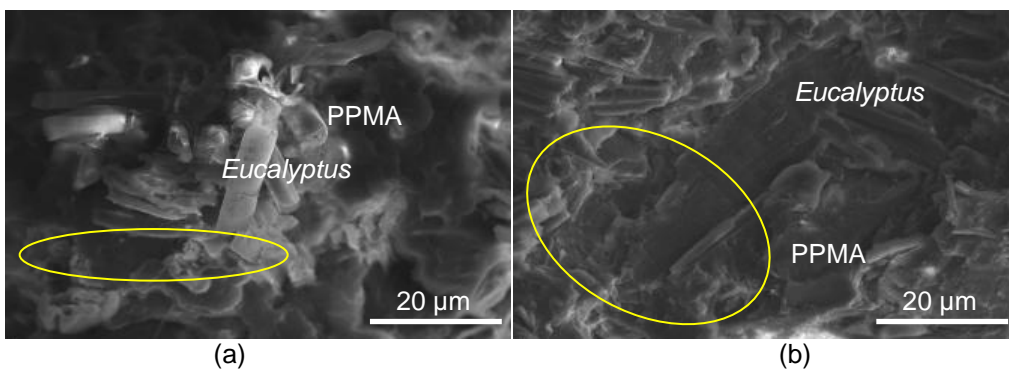
The results showed that as the waste levels increased, there was a decrease in the value of tension and maximum deflection. This was attributed to a low dispersion and high stiffness of the particles. The remarkable increase in stiffness was verified with the addition of *Eucalyptus* and *Pinus* waste, as indicated by the increase in modulus of elasticity in traction and bending. This result was in agreement with Dai and Fan (2014). For the PP/*Eucalyptus* composites, the approximate gains with the addition of wood particles were in the following order: 20% at the maximum stress for the PP/10% waste, 20% at the maximum deflection for the PP/10% waste, and 75% in the modulus of flexure for PP/40% waste, without compatibilizer. The PP/*Pinus* composites obtained smaller gains than the PP/*Eucalyptus* composites, with values of 5% in the maximum tension for the PP/10% waste, 5% in the maximum deflection for the PP/10% waste, and 70% in the flexural modulus PP/40% waste, without compatibilizer.

With the addition of up to 5% compatibilizer, an increase in tension and maximum deflection was observed. This result indicated that there was probable wetting and mechanical adhesion between the matrix-particle, as well as chemical interactions through polar groups in the structure of the polypropylene grafted with maleic anhydride with the hydroxyls of the cellulosic surface, which demonstrated improvements in the mechanical properties. About formulations with 10% of compatibilizer there was no significant difference for *Eucalyptus* but the flexural modulus increased with 40% *Pinus*, which may be related to the aspect ratio of the material.

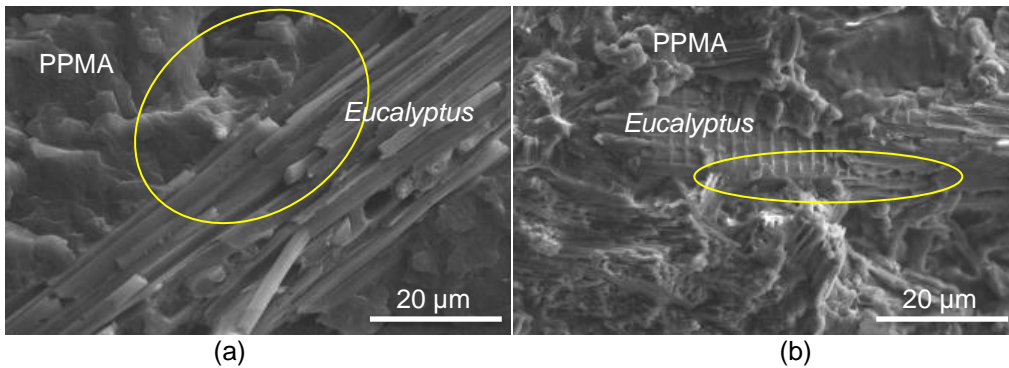
The photomicrographs show that the not compatibilized composites had lower matrix-particle transition than the compatibilized composites. These photomicrographs corroborated with the results obtained previously, in which the compatibilized formulations presented better performance compared to the not compatibilized samples. Figures 8 through 11 show fracture specimens morphologies from the PP/PPMA/*Eucalyptus* composites with different formulations. A predominance of ductile fracture surfaces for the not compatibilized composites and fracture of the fragile type for the compatibilized ones was observed, according to results in traction. The ellipses indicated the detachment of the dispersed phase of the unmodified thermoplastic matrix in Fig. 8 and the evident matrix-particle adhesion with the use of PPMA, according to Figs. 9 through 11.



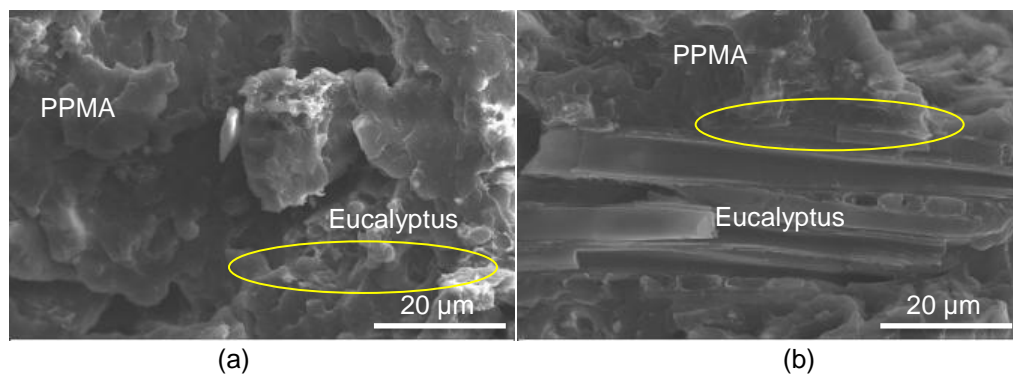
**Fig. 8.** Morphology of fracture surfaces of injected specimens of composites PP/20%*Eucalyptus* (a) and PP/40%*Eucalyptus* (b)



**Fig. 9.** Morphology of fracture surfaces of injected specimens of composites PP/1%PPMA/20%*Eucalyptus* (a) and PP/1%PPMA/40%*Eucalyptus* (b)

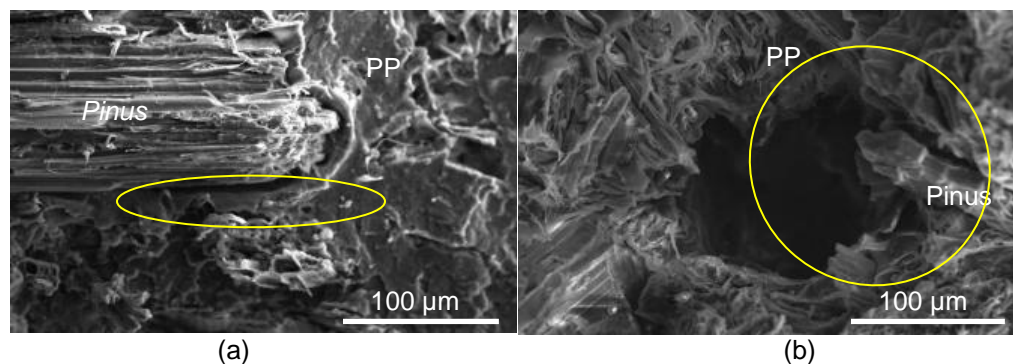


**Fig. 10.** Morphology of fracture surfaces of injected specimens of composites PP/5%PPMA/20%*Eucalyptus* (a) and PP/5%PPMA/40%*Eucalyptus* (b)

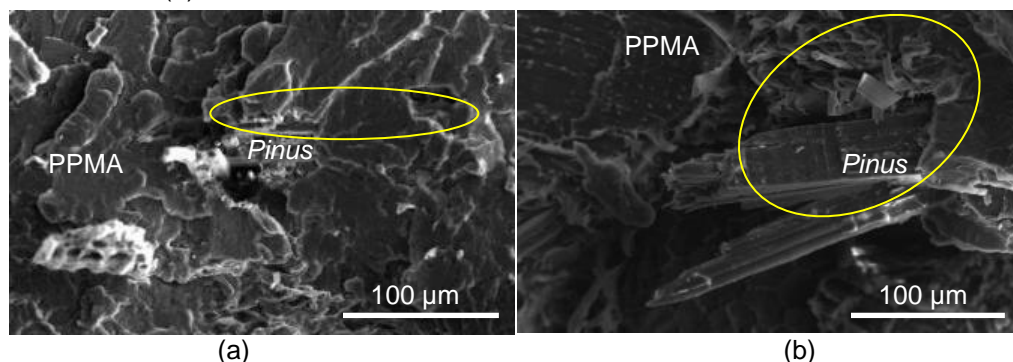


**Fig. 11.** Morphology of fracture surfaces of injected specimens of composites PP/10%PPMA/20%Eucalyptus (a) and PP/10%PPMA/40%Eucalyptus (b)

The scanning electron micrographs of the tensile fracture specimens of the PP/PPMA/*Pinus* composites with different formulations are shown in Figs. 12 through 15. Looking at Fig. 12, it was possible to identify that voids and particles detached from the polymer matrix without the added compatibilizer. This showed little affinity between the phases, resulting in poor matrix-particle anchoring. Figures 13 through 15 illustrate a great adhesion between the polymer matrix and the reinforcement with compatibilizing. According to Bledzki and Gassan (1999) and Hristov *et al.* (2004), polypropylene composites with wood particles, in the presence of compatibilizer, allow the production of composites with better dispersion of the waste to the polymer matrix, resulting in good wettability between phases.

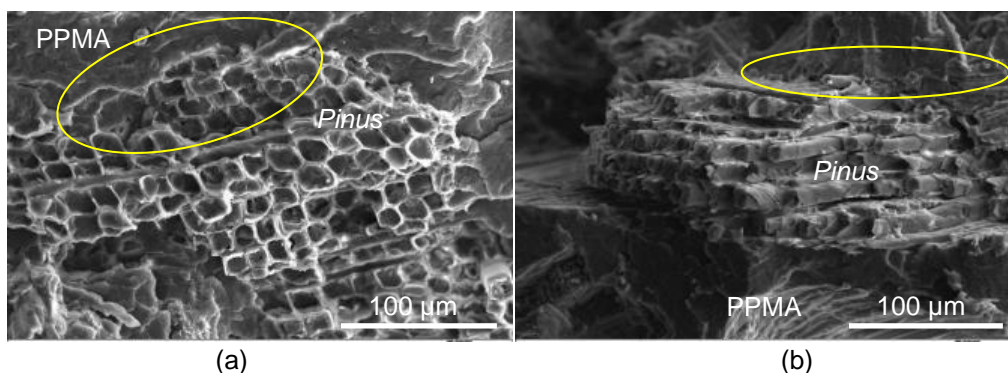


**Fig. 12.** Morphology of fracture surfaces of injected specimens of composites PP/20%*Pinus* (a) and PP/40%*Pinus* (b)

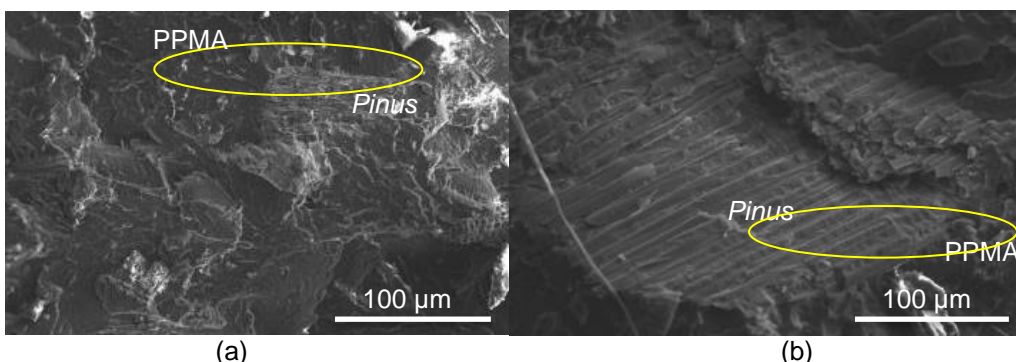


**Fig. 13.** Morphology of fracture surfaces of injected specimens of composites PP/1%PPMA/20%*Pinus* (a) and PP/1%PPMA/40%*Pinus* (b)





**Fig. 14.** Morphology of fracture surfaces of injected specimens of composites PP/5%PPMA/20%*Pinus* (a) and PP/5%PPMA/40%*Pinus* (b)



**Fig. 15.** Morphology of fracture surfaces of injected specimens of composites PP/10%PPMA/20%*Pinus* (a) and PP/10%PPMA/40%*Pinus* (b)

Table 1 presents the thermal properties of the PP/PPMA/*Eucalyptus* and PP/PPMA/*Pinus* composites, compatibilized and not compatibilized.

**Table 1.** Melt Temperature ( $T_m$ ), Melt Enthalpy ( $\Delta H_m$ ), and Percent Crystallinity ( $X_c$ ) of Virgin Polymer and PP/*Eucalyptus* and PP/*Pinus* Composites

Samples	<i>Eucalyptus</i>			<i>Pinus</i>		
	$T_m$ (°C)	$\Delta H_m$ (J g <sup>-1</sup> )	$X_c$ (%)	$T_m$ (°C)	$\Delta H_m$ (J g <sup>-1</sup> )	$X_c$ (%)
Virgin PP	167.1	85.22	40.8	167.1	85.22	40.8
<b>PP/waste</b>						
10%	167.1	67.57	35.9	168.7	65.46	34.8
20%	167.5	64.40	38.5	167.8	63.98	38.2
30%	167.2	54.35	37.1	168.0	56.91	38.9
40%	167.3	49.44	39.4	167.5	45.70	36.4
<b>PP/1%PPMA/waste</b>						
20%	167.7	64.46	38.6	167.9	66.18	39.6
40%	166.7	50.85	40.5	167.4	49.08	39.1
<b>PP/5%PPMA/waste</b>						
10%	167.2	66.47	35.3	168.2	74.10	39.4
20%	166.3	56.19	33.6	166.9	62.61	37.4
30%	166.0	46.52	31.8	167.4	56.50	38.6
40%	166.2	39.72	31.7	167.0	49.64	39.6
<b>PP/10%PPMA/waste</b>						
20%	164.3	57.17	34.2	167.8	63.79	38.1
40%	166.3	49.78	39.7	166.4	46.34	37.0

Table 1 shows that for the second heating cycle there were no noticeable variations in the melt temperature of the polypropylene in the presence of waste and compatibilizer for the varying concentrations. Studies conducted by Luyi and Kecheng (2001) using polypropylene rice husks, Kuruvilla and Mattoso (2002) using cellulose microfiber and sisal in a PP matrix, Correa *et al.* (2003) using three types of particulate waste of polypropylene *Pinus* wood for compatibilized and not compatibilized mixtures with PPMA, and Joseph *et al.* (2003) using short sisal fibers in PP, also showed that the melt temperature of the polypropylene ( $T_m$ ) remained practically unchanged with an increased percentage of cellulosic filler in the absence and presence of PPMA.

The melting enthalpy of the virgin PP ( $\Delta H_m$ ) in the composites showed a decrease with the addition of cellulosic filler, indicating an inhibitory effect on the crystallization of polypropylene. This showed that the addition of vegetable charges altered the crystallinity of the matrix, interfering in the mechanical properties of the composite materials.

It was observed that the addition of the particulate filler to the PP decreased the crystallinity of the polymer matrix for any series of the composites studied; that is, the presence of PPMA also showed no remarkable influence in the sense of increasing the crystallinity of the polypropylene. Correa *et al.* (2003) also found that although thermoplastic matrix reinforcement occurs, the crystallinity of the resin tends to decrease in the presence of cellulosic filler, which probably decreases the growth of the crystals. These results indicated that the barrier capacity of the load influenced the effects of the mechanical behavior through the change in crystallinity.

## CONCLUSIONS

1. The addition of loads of *Eucalyptus* and *Pinus* particulates to the polypropylene matrix conferred a reinforcing character to the conjugated materials, and the use of *Eucalyptus* waste in the formulations produced better effects on the results of mechanical resistance of the composites than the *Pinus* waste for the same formulations.
2. There was a high gain in the modulus of elasticity with the addition of *Eucalyptus* and *Pinus* waste in the polymer matrix and a positive influence on the maximum stress *via* the use of compatibilizer, which was shown to increase the reinforcing action of the composite materials, promoted by the best transfer of charges between matrix-particle. The improvement of the interfacial adhesion did not interfere in the elongation of the composites. An increase in the flexural modulus occurred with the addition of *Eucalyptus* and *Pinus* waste in the polypropylene (PP) matrix and a positive influence on the tension and maximum deflection *via* the use of maleic anhydride-modified polypropylene (PPMA).
3. In compositions with 5% PPMA, it was possible to add 30% to 40% of *Eucalyptus* and *Pinus* waste to obtain a small increase in the resistance of the composite materials compared to the thermoplastic matrix. In general, the use of PPMA was efficient in increasing the maximum stress of the developed composites, which proved a better matrix-reinforcement interfacial interaction. Photomicrographs confirmed the improvement of adhesion when the compatibilizer was used, due to the probable esterification and/or hydrogen bonding reactions at the material's surface. Finally, the degree of crystallinity of the thermoplastic matrix presented a decrease with the

addition of organic load in the composites compatibilized and not compatibilized, due to the limitation in the movement of the crystals.

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