Influence of Coupling Agent in Polyolefinic Composites from Post-Consumer Waste with *Eucalyptus Grandis* Flour

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**Abstract:** This study investigates the feasibility of using recycled high density polyethylene (HDPE) and polypropylene (PP) with wood fiber from species *Eucalyptus Grandis* (EU) to manufacture experimental composite panels. The use of maleated polyethylene and polypropylene as coupling agent (CA) improved the compatibility between the fiber and plastic matrix and mechanical properties of the resultant composites compared well with only the polymer and composites without compatibilizer. It was evaluate the influence of coupling agent (CA) in polyolefin matrix at different concentrations of HDPE and PP in the composites, checking the physical, mechanical and morphological properties of wood plastic composites (WPC). HDPE and PP post-consumer, wood flour from *Eucalyptus grandis* (EU) species and graftized polyethylene and polypropylene with maleic anhydride (MA) as CA were used on formulations, being extruded and after injection molding. Results showed that composites with CA had a higher mechanical performance compared to no-compatibilized, it was confirmed also by morphological analysis. The results of contact angle showed that the incorporation of CA led an increase in surface energy of polymeric matrix promoting a better interaction with vegetable filler. Based on the findings in this work, it appears that recycled materials can be used to manufacture value-added panels without having any significant adverse influence on material properties.

**Key words:** Polyolefinic composite, HDPE, PP, plastic waste, coupling agent, wood flour.

**Nomenclature**

OE  
*Eucalyptus grandis*

PP  
Polypropylene

HDPE  
Polyethylene

EVA  
Ethylene vinyl acetate copolymer

CA  
Coupling agent

MA  
Maleic anhydride

**Greek letters**

$\theta$  
Contact angle

1. Introduction

The increasing amount of solid waste produced in urban centers, particularly those derived from plastic industries, has attracted the attention and motivated researchers to search solutions, aiming the use for the generation of new products [1]. In the context of concern about sustainability, there are two problems, the waste generation and depletion of natural resources. As the planet cannot renew what is consumed in modern times, it must then reconsider the standards of production, aiming at sustainable consumption. This can be achieved through less polluting energy sources, with emphasis on the life cycle of products, reducing waste production and recycling as much as possible [2]. So, much emphasis has been given to the preservation and conservation of the environment in order to ensure sustainable...
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devlopment. Among the various damages to the environment, one is related to the plastic waste and lignocellulosic. The plastic waste, in general, takes too long to spontaneous degradation and, when burned, they may produce different degrees of toxicity [3]. Therefore, there is a general tendency to use these lignocellulosic and polymeric wastes, considering the immense potential value of the materials processed and the implications and pollution arising from non-use of waste [4].

The development of composites involving the use of lignocellulosic materials as reinforcement in recycled polymer matrices has grown in response to environmental conservation, becoming more common in the polymer industry, because to the lignocellulosic be renewable, inexpensive, biodegradable and non-toxic. Among the lignocellulosic materials most used for this purpose are sawdust, a timber industry waste, vegetable fibers and lignocellulosic agricultural and agroindustrial waste [5]. But the preparation of polymer composites with wood flour requires good compatibility in the interface between the polymer matrix and the wood flour [6]. The main concern is the inherent incompatibility between the hydrophilic fibers and hydrophobic polymers surfaces, which results in a poor adhesion [7-18]. Thus, some coupling agents have been used in the modification of fibers, to increase the interfacial adhesion between reinforcement and polymeric matrix polyolefin cellulosic and thereby improve the mechanical properties of the polymeric composite [8]. The use of coupling agents is an efficient approach to improve the inter-phase compatibility of the composites [9-11]. A coupling agent is a chemical that functions at the interface to create a chemical bridge between the reinforcement and the matrix [12]. The agent improves the interfacial adhesion when one end of the molecule is tethered to the reinforcement surface and the functionality at the other end reacts with the polymer phase. Copolymers containing maleic anhydride, such as maleated polypropylene (MAPE), are the most commonly reported coupling agents used in wood plastic composites [13, 14]. The anhydride groups of the copolymers can react with the surface hydroxyl groups of wood polymers, forming ester bonds. The other end of the copolymer entangles with the polymer matrix because of their similar polarities [15, 16]. Isocyanates have also been used as coupling agents in natural fiber reinforced polymer composites. Urethane links can be formed between the isocyanate functionality and the hydroxyl group of natural fibers [17], consequently blocking the hygroscopic hydroxyl sites [18]. Silanes are recognized as efficient coupling agents as they can be hydrolyzed to expose the active silanol groups toward the hydroxyl groups of the wood, thereby reacting to form chemical bonding [19].

In this sense, this study aims to develop thermoplastic composites with polymeric matrix compatibilized and filled with wood flour, which have specific properties, from these post-consumer materials in order to reduce the environmental impact and add value to these materials.

2. Experiments

The materials used in this study were post-consumer HDPE (5 kg) from packaging of cleaning products collected at UFRGS; flakes of post-consumer PP (5 kg), from bottle caps from soda and mineral water supplied from the company Prisma Montelur Thermoplastics; EVA (ethylene vinyl acetate copolymer) from the “liners” inside the bottle caps; wood flour from Eucalyptus grandis (EU) species (3 kg) from the southern state of Rio Grande do Sul, Brazil; graftized polyethylene copolymer with maleic anhydride (PEGMA) Fusabond E-528 and graftized polypropylene copolymer with maleic anhydride (PPgMA) Fusabond MZ-109D from DuPont.

HDPE plastic waste were pre-washing (previously removed the covers and labels) with water, following
dried at room temperature for 24 hours. After, they were ground to size reduction in the form of flakes in a knife mill Retsch SM 200. The PP sample was already in form of flakes. The wood flour (EU) passed by size separation in a system of sieves brand Bertel series of 32 and 16 Tyler mesh, and the selected particle size between > 250 and < 500 μm.

In the preparation of thermoplastic composites, were made HDPE/PP blends in different ratios and mixed with 30% w/w eucalyptus powder, (PP-HDPE)/EU (70/30) and 3% w/w coupling agent (CA). The formulation of the substrates was performed as shown in Table 1.

The mixtures were processed in a single screw extruder (L/D: 22), with temperature profile from 170 °C to 190 °C, residence time of 80 s and screw speed of 65 rpm. PP, HDPE and blends of these polyolefins with the coupling agent (CA) were processed in the extruder in order to functionalize the polyolefins, and added after the wood flour (EU) sieved. The specimens for mechanical tests were prepared by injection process (injection Mini Thermo Scientific Haake MiniJet II) at temperature of 185 °C and pressure of 600 bar.

2.1 Characterization

2.1.1 Contact Angle and Surface Energy

The contact angle measurements were made with the equipment Ramé-Hart goniometer model 100-00, according to ASTM D 724-89. The liquids used for drop sessile test were deionized water and methylene iodide (CH₂I₂). For all samples were made 10 drops of each liquid, each drop image was measurement of angles using the software Surftens 3.0. The values obtained in the contact angle tests were used to calculate the surface energy of the samples. The calculation of the surface tension was carried out with the software Surftens 3.0.

2.1.2 Mechanical Testing

The tensile tests were performed on a universal machine, model DL 2000 from EMIC, based on ISO 527-1 standard, using a load cell of 1,000 N and the speed of separation of 2 mm/min, where were used eight specimens test. The flexure tests were performed on a universal machine, model 4200 from Instron, based on the ASTM D790, using a load cell of 5,000 N.

The impact resistance of a polymeric material is a parameter of high important because that the polymers, in most applications are subject to requests impact, ie extreme requests in a short time [20]. The method used is based on the principle of absorption of energy from the energy potential of a pendulum, and is called Izod impact test. The Izod impact was performed on CEAST equipment, model Impactor II, in accordance with ASTM D256. The specimens were prepared without notch and pendulum of 2.75 J for the sample of HDPE and other composites and pendulum of 11 J to PP sample, at room temperature.

2.1.3 Scanning Electron Microscopy (SEM)

This test was performed for analysis by scanning electron microscopy (SEM), using the electron microscope JEOL JSM 6060, with a voltage of 10 kV

| Sample | Matrix (70%) | Filler (30%) | PP (% w/w) | HDPE (% w/w) | % CA |
|--------|--------------|--------------|------------|--------------|------|
| 1      | PP           | -            | 100        | -            | -    |
| 2      | HDPE         | -            | -          | 100          | -    |
| 3      | HDPE         | EU           | -          | 100          | -    |
| 4      | PP           | EU           | 100        | -            | -    |
| 5      | PP-HDPE      | EU           | 80         | 20           | -    |
| 6      | PP-HDPE      | EU           | 50         | 50           | -    |
| 7      | HDPE         | EU           | -          | 100          | 3    |
| 8      | PP           | EU           | 100        | -            | 3    |
| 9      | PP-HDPE      | EU           | 80         | 20           | 3    |
| 10     | PP-HDPE      | EU           | 50         | 50           | 3    |
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and magnification 80 times. The samples were manually cut, fixed on stubs with double sided tape and coated with gold. The images of the samples were obtained from the fracture surfaces after tensile test.

3. Results and Discussion

3.1 Contact Angle and Surface Energy

Fig. 1 shows the results of contact angle of the water drop with the surface of polymers and their composite with and without CA. It is observed that the contact angles on the PP and HDPE surfaces are higher than the other materials, with values near to 90°, it is due to the high hydrophobicity of the polyolefinic materials [12, 16, 21]. The inclusion of polar groups for the surface oxidation process of apolar polymers makes the contact angle decreases [21]. Therefore, there is a change in the characteristics of hydrophobicity [22]. This is what can be seen from the results of the contact angle of the composite without CA, where there is a decreasing in contact angles because to the incorporation of vegetable filler. Among the composites the presence of CA also influence in the reduction of the contact angle of the samples, being more pronounced in the composites HDPE/EU and HDPE/EU/CA. In general, the composites with higher content of HDPE in their formulations showed smaller contact angles, both treated and those not treated with CA, indicating a higher wettability and consequently greater adhesion characteristics than other composites [23]. These results are unexpected, compared to PP. As the HDPE is more stable than the PP, it is believed to be because of origin of the HDPE (packaging cleaning products), wherein the internal surface has suffered contamination and/or surface modification by these products (hydrolytic degradation). According to research by Santana & Gondim [23] (evaluated the degradation of HDPE post consumer after various cleaning conditions with chemicals products - NaOH and detergent-DL) samples treated with NaOH solution showed an increase in OIT (oxidation induction time) in the carbonyl index and a decreased onset temperature of thermal decomposition of 10 °C compared to the treated with mixture solution of NaOH/DL.

For a better preview, in Fig. 2, it is shown the images of two drops on the surface of the composite PP80-HDPE20-EU without and with CA respectively representing the spreading on the surface thus indicating its wettability.

Formally, the contact angle between a drop of liquid with known surface tension and a solid surface depends on the relation between the adhesive forces (which would drop to spread over the surface) and the cohesive forces of the liquid (which want to contract the drop by a sphere with a minimum area).

Adhesion is the attractive force between two materials together in a strong interfacial contact where all work and mechanical power can be transferred along the interface of union between these bodies. The forces that contribute to this intimate contact between materials are Van der Waals forces (secondary bonds), and primary chemical bonds, in the polymer case is covalent bond [24]. Thus, through the contact angles calculated previously, is calculated by the surface tension of the polymers and composites.

In Fig. 3, it can be visualized the low surface energy on the HDPE and PP surface, when compared to the
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Fig. 2  Image of the water drop profile on the composites surface of: (a) PP80-HDPE20-EU composite, and (b) PP80-HDPE20-EU-CA.

Composites, fact characteristic of olefinic polymer to be non polar. It is observed that the composites without CA, presented lower surface energy than those with CA, fact due to that the intermolecular forces of cohesion acting on the polymer were mainly dispersion forces, which led to low surface energy and therefore poor adhesion with the polar filler [25].

By these results of surface energy, can be seen that the incorporation of the coupling agent (CA) was efficient, leading to an increase of surface energy of the polymeric matrix of PP and HDPE and therefore of the composites. This fact is due to presence of functional group C=O and C-O of the maleic anhydride from CA that promoted so a better interaction between non-polar polymeric matrix and polar lignocellulosic filler (EU), as mechanism of adhesion showed in Fig. 4 [26].

Redighieri [6] studied use of wood waste in the production of composites with recycled polyethylene. In this work, the polyethylene was functionalized with maleic anhydride (PE-g-MA) as coupling agent to provide better adhesion between the matrix and the dispersed phase. The results showed that treatment of the wood waste flour with PE-g-MA increased mechanical properties of the composite compared to composites containing untreated waste. The HDPE/EU/CA and PP/EU/CA composites were those that showed the best result, thus indicating a better adhesion between the phases.

3.2 Mechanical Properties

3.2.1 Tensile and Flexural Test

Table 2 shows the results of tensile strength of the PP, HDPE and of their composites reinforced with wood flour (EU), without CA and with coupling agent (PPgMA and PEgMA). It can be seen that all composites with CA had a higher tensile strength. Among the compatibilized composite, which showed the best performance was HDPE/EU/CA (0/100), with...
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Table 2  Results of tensile and flexural test of polymers and their composites without and with 3% CA.

| Samples              | Mechanical properties | Tensile test | Flexural test |
|----------------------|-----------------------|--------------|---------------|
|                      |                       | Tensile strength at rupture (MPa) | Modulus of young (MPa) | Flexural strength (MPa) | Flexural modulus (MPa) |
| PP                   | 37 ± 1.7              | 909 ± 23     | 14 ± 2.0      | 290 ± 52                |
| HDPE                 | 32 ± 1.5              | 520 ± 62     | 10.5 ± 1.5    | 247 ± 19                |
| HDPE/EU              | 35 ± 1.1              | 428 ± 69     | 17 ± 0.9      | 421 ± 36                |
| PP/EU                | 27.5 ± 1.4            | 289 ± 60     | 21.5 ± 0.7    | 476 ± 13                |
| PP80/HDPE20/EU       | 30 ± 1.3              | 865 ± 38     | 25 ± 3.0      | 559 ± 65                |
| PP50/HDPE50/EU       | 26 ± 1.1              | 564 ± 37     | 17.5 ± 1.8    | 417 ± 69                |
| HDPE/EU/CA           | 36 ± 0.5              | 519 ± 58     | 14.5 ± 0.5    | 303 ± 21                |
| PP/EU/CA             | 30 ± 1.4              | 356 ± 11     | 15.5 ± 1.6    | 282 ± 68                |
| PP80/HDPE20/EU/CA    | 30 ± 1.2              | 874 ± 30     | 20 ± 0.7      | 344 ± 42                |
| PP50/HDPE50/EU/CA    | 29.5 ± 1.1            | 717 ± 48     | 17 ± 2.5      | 379 ± 72                |

36 MPa. Among the blends without CA, the composition of PP/HDPE (80/20) showed the best mechanical performance.

Relative to the elasticity modulus or Young modulus also observed the same behavior, where the composite with CA presented module above those without AC, with best results (Table 2). It is also noted that between the blends, the composition PP/HDPE (80/20) showed the largest modulus.

The mechanical properties of polymer composites reinforced with short vegetable fibers depend on many factors that determine the surface area able transfer tensile of matrix to the fibers [27]. These factors include geometric parameters (length, diameter and aspect ratio) orientation and distribution of fibers in the polymer matrix, and these parameters are dependent on process conditions used for the manufacture of composites [28]. Also the adhesion between fiber and matrix is responsible for the efficient transfer tensile of the matrix to reinforcement [29].

The increase in tensile strength and modulus, observed in samples prepared with CA indicate a better transfer of stress from matrix to reinforcement in lignocellulosic wood-polymer interface due to a probable occurrence of anchoring mechanical and chemical interactions between anhydride groups and hydroxyl groups of cellulose. A similar result was obtained by Nachtigall et al. [30] when was verified that addition of the polymeric coupling agents in formulation produced composites with better performance, being this behavior can be attributed to the enhanced chemical compatibility between the components. This result may be related to the fact that the PPgMA and PEgMA are acting as compatibilizing agent between the wood particles and polymer matrix, making more dispersed *eucalyptus* particles and attached to recycled polymer matrix.

Evaluating only the post-consumer polymers used as matrix of composites, it is noted that the PP had a higher tensile strength and modulus as HDPE, unexpected result, probably because presence of EVA is mixed with the PP polymeric matrix. A study by Espert et al. [31] reported the use of a post-consumer polypropylene with a low percentage of EVA in its composition and in some cases leads to improved resistance to water absorption and better mechanical properties, when EVA can further to improve the compatibility between the fibres and the matrix. The lower mechanical performance presented by HDPE can also be related the possible degradation of this material due to merits of this residue (packaging that came from cleaning products) [23].

Evaluating the results of the flexural test of the PP, HDPE and of their composites shown in Table 2, indicate that composites with coupling agent showed
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inferior results compared to those not compatibilized. The influence of the proportion of wood on the flexural strength in thermoplastic composites has been observed by some authors [29, 32] where found that the flexural strength increases with increasing wood flour content in composite. In general, there is a good adhesion between the phases of fiber and matrix for this to occur, but even a system of low adhesion occurs this behavior.

Comparing the composites without coupling agents, those with higher levels of matrix PP presented a better maximum tensile of flexion, influenced by the origin of post-consume PP, because it contained EVA in its composition. The EVA has properties intermediate between its two constituent homopolymers, polyethylene and poly (vinyl acetate) [33]. Generally, the EVA is used to improve mechanical strength, processability, impact resistance and insulating properties. Furthermore, the ester groups present in the structure of the EVA, when introduced into the mixture, provide a better miscibility with inorganic fillers [34]. Relative the flexural modulus it can be seen that composites with polymeric matrix of PP or from the PP-HDPE blends with higher content of PP in its composition showed higher modulus results, probably because of characteristics given on the PP by the addition of EVA in the blend [35].

Comparing the flexural modulus of the composite with and without coupling agents, it is observed that those treated with CA presented modules lower than those untreated, indicating that the CA does not positively influence in the improvement. In relation to the maximum flexural strain and the module of the polymers (PP and HDPE), PP showed to be better than HDPE, probably influenced by the amount of EVA present in its composition.

3.2.2 Impact Energy Absorption

Fig. 5 shows the results of the impact resistance testing of the polymers and their composites. It is observed that the composites treated with CA showed higher values impact energy absorption than non-treated, a fact that is explained by the better interaction of coupling agents (PPgMA and PEgMA) in the composites, promoting a greater adhesion between polymer matrix and vegetable filler, where this filler would be acting as a reinforcement agent [36]. The composite HDPE/EU/CA showed a higher impact energy absorption that the PP/EU/CA. Among the composites obtained from the polymer blends that with a ratio of PP-HDPE (50/50) showed better results.

Results of the impact absorption of the polymers used in the composites are also shown in Fig. 4, where the PP had impact absorption energy higher than HDPE because to the presence of EVA in its composition, as already mentioned in this work, which improved this mechanical property [36]. Accordind Gupta et al. [37] and Martins et al. [38] the impact strength of the polypropylene increases rapidly with increasing EVA content up to 5 wt% of EVA and then this increase is moderated until the content of 30%, again increasing rapidly to higher concentrations of EVA. Furthermore, the higher the content of vinyl acetate in EVA, the greater the increase in impact resistance. The pendulum used for the sample of the PP was 11 J, while for the other samples was used 2.75 J.

3.2.3 Morphological Properties

The eucalyptus fiber, as all natural fiber, composed a bundle of micro-fibrils promoted by mechanical
shear in extrusion, a single fiber is “stripped” generating micro-fibrils [39]. The dispersion of the micro-fibrils in the polymer matrix can be evidenced by the micrographs of the composites analyzed by SEM. Analyzing these micrographs, it is clear that the micro-fibrils do not have a single orientation in the composites. This is expected in composites reinforced with short fibers, but the fibrillation compensates orientation lack in respect of mechanical properties [39]. Besides the orientation of micro-fibrils is also possible through the micrographs, check whether or not adhesion between matrix and reinforcement.

SEM was possible to evaluate the efficiency of the dispersion of the cellulosic filler in the thermoplastic matrix and the capacity of wetting the surface fiber for thermoplastic matrix as a way to evaluate the efficiency of coupling agent [40]. Aspects related with wettability and level of fiber-matrix interfacial adhesion can be observed in the micrographs presented.

Fig. 6a shows the micrograph of the surface of fracture after the tensile test of the composite PP/EU, where there is the presence of various structures like fibrils, characteristics for polymer matrix, and some holes promoted by removal of the filler, which indicates that there is a poor interaction between fiber and polymeric matrix [41]. Fig. 6b shows a micrograph of the fracture surface of the composite PP/EU/CA where can be seen that there are several particles of vegetable fiber presents, some that are not fully adhered to the polymer matrix, and smaller ones are fully adhered, demonstrating a good compatibility between the two phases.

Fig. 7 shows the micrographs of the composites HDPE/EU and HDPE/EU/CA where it is observed in the HDPE/EU composite many particles distributed in the matrix and some small voids, but many particles seem not be well adhered to the matrix.

The composite with CA also presents some gaps in its structure, however the composite is more homogeneous, where the filler particles are better attached to the HDPE matrix, indicating that the CA acts positively on the interfacial adhesion of the polymeric matrix with reinforcement [41], which can be also indicated by the results of tensile strength of the compatibilized composites that presented best results.

Fig. 8 shows micrographs of the surface after fracture of the composite tensile test PP80/HDPE20/EU (a) and PP80/HDPE20/EU/CA (b). In these composites, because of the matrix is composed of a mixture of two polymers, where 80% PP and 20% HDPE, which have a degree of incompatibility with each other, can be seen the composite without CA presented some voids and many fibrillated pieces characteristic of the PP, resulting from stretching of the polymer during the
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Fig. 7  SEM micrographs of fracture surfaces of composite (a) HDPE/EU and (b) HDPE/EU/CA.

Fig. 8  SEM micrographs of fracture surfaces of (a) PP80/HDPE20/EU and (b) PP80/HDPE20/EU/CA composites.

tensile test, and has not a uniform matrix, indicating that there was not good compatibility between the PP and HDPE. There is also no filler particles adhered to the matrix, which shows poor adherence between the phases of the composite. The composite made with PPgMA and PEGgMA no presented significant changes in the micrograph, but the tensile test proved that there is an improvement in mechanical properties because of the insertion of the CA [30, 42].

Fig. 9 shows the SEM micrographs of fracture surface after the composite tensile test (a) PP50/HDPE50/EU and (b) PP50/HDPE50/EU/CA. These composites, even though from blends of PP and HDPE composite as above, showed a better result than the composite PP80/PEAD20/EU, visible in composites treated with coupling agents indicated by arrows in the micrographs of Fig. 8b adhesion of particles of vegetable fiber in polymer matrix, showing the strong compatibility between matrix and reinforcement. Even in the composite without CA (PP50/HDPE50/EU), that had some gaps in its surface, it was find a better interaction between polymer matrix and the filler in relation to PP80/HDPE20/EU.

Thus, the morphology of fractured surfaces confirmed the correlation made about the mechanical properties and the fact of having made the choice of PPgMA and PEGgMA as coupling agent for PP and HDPE composites with eucalyptus fiber. Overall, analyzing the micrographs of the composites without
CA, it is observed that there are gaps between the fibers and matrix, and “holes” from the deployment of fiber because to poor interfacial adhesion and poor wettability of fiber within the matrix, which occurs probably due to the large difference in surface energy between the two [43]. However, the samples in which PPGMA and PEgMA was used as coupling agents showed an increase in interfacial adhesion, observed for the considerable decrease of gaps between the matrix and the dispersed phase.

Images shown clearly that the coupling agent provides an improved interfacial adhesion between the cellulosic fiber and thermoplastic matrix independent of wood size. The images confirm the results obtained in mechanical tests where the compatibilized samples showed an increase of strength in relation to the non-compatibilized compositions. It was also noted that composites containing HDPE presented more fragile fracture surface confirming results of mechanical properties and surface energy, demonstrating the thermoplastic matrix degradation.

4. Conclusions

The results obtained in this work, where was studied the compatibilization of thermoplastic matrix composites reinforced with vegetable filler using coupling agents were the following:

Results of the contact angle and SEM indicate an improvement of interfacial adhesion between the components of the composite (HDPE-PP-EU) when is used coupling agent, corroborating the results of performance in mechanical tests. Between PP-HDPE blends used as matrix, the composition PP80-HDPE20 showed the highest mechanical performance, especially those modified with CA.

The result of mechanical properties generally indicate that the use of recycled polyolefin is a viable alternative for the production of new materials and can therefore be used for many new applications, contributing to reduction of the impact environment and enabling new sustainable alternatives.

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