

PAPER

Evaluating the influence of coupling agents in the structural properties of polypropylene coconut fiber composites

To cite this article: Luciano Pisanu *et al* 2019 *Mater. Res. Express* **6** 115320

View the [article online](#) for updates and enhancements.



IOP | ebooks™

Bringing you innovative digital publishing with leading voices to create your essential collection of books in STEM research.

Start exploring the collection - download the first chapter of every title for free.

Materials Research Express



PAPER

Evaluating the influence of coupling agents in the structural properties of polypropylene coconut fiber composites

RECEIVED
3 July 2019

REVISED
29 August 2019

ACCEPTED FOR PUBLICATION
19 September 2019

PUBLISHED
4 October 2019

Luciano Pisanu^{1,4} , Josiane Barbosa¹, Ricardo Souza² and Marcio Nascimento³

¹ SENAI-CIMATEC, Institute of Innovation for Forming and Joining of Materials, Av. Orlando Gomes 1845, Piatã. 41650-010 Salvador, Brazil

² University of Aveiro, 3810-193 Campus Santiago, Aveiro, Portugal

³ UFBA—Federal University of Bahia, Escola Politécnica, R. Aristides Novis 2, Federação. 40170-115 Salvador, Brazil

⁴ Author to whom any correspondence should be addressed.

E-mail: luciano.pisanu@fieb.org.br, josianedantas@fieb.org.br, rsousa@ua.pt and mlfn@ufba.br

Keywords: natural fibers, polymer-matrix composites, thermal properties, mechanical properties, coconut

Abstract

Coupling agents have been widely used to improve the adhesion of composites reinforced with vegetable fibers. In this study, the influence of three different coupling agents was analyzed: polyethylene and polypropylene both grafted with maleic anhydride and titanate grafted in polyethylene. The composites with 30 wt% percentage of coconut fibers and 6 wt% of compatibilizers were processed in a co-rotating twin-screw extruder. Specimens were submitted to mechanical and thermal treatments to evaluate these properties. Data showed that coupling agents significantly influence composite properties. Polypropylene-based coupling agent shown good results to the composites increased the yield strength by 23% and the tensile strength by 32% with respect to the polymer matrix. These results represented thus an adequate and positive effect of the coupling agents related to the more relevant mechanical properties to manufacture structural composites using vegetable fibers. Based on results achieved in this investigation, the composite containing coconut fiber presented the optimum set of mechanical properties and improved thermal stability, which make it suitable for applications such as structural components.

1. Introduction

Thermoplastic polymers have an undisputed presence in the routine of human beings and have conquered applications that had once belonged to metallic and/or ceramic materials. However, thermoplastic polymers require a long time to degrade spontaneously [1]. Composite materials have received much attention in industrial applications in the plastics, automobile, furniture, and packaging industries to cut down on material cost. The use of natural reinforcement fibers in polymer matrices has been widely applied, resulting in materials with excellent mechanical and thermal properties [2]. Due to the growing demand for the use of renewable resources, the number of industrial applications of composites that contain vegetable fibers has increased, including the 3D printed parts improving significantly their mechanical properties [3]. Coconut fibers have a low cellulose content and a high microfibril angle and thus provide large elongations and lower values of tensile strength when compared with other types of vegetable fibers [4].

As a disadvantage, in general, vegetable fibers are extremely hydrophilic, which results in a weak interaction with the thermoplastic matrix, which is normally hydrophobic. This low interfacial adhesion is associated with the chemical affinity between the matrix and the vegetable fiber, which can form voids at the interface and result in failures that would compromise the mechanical performance of the composites. To minimize these problems, chemical and physical processes have been used to modify both the fiber surface and surface tension of the polymer to improve the matrix-fiber interfacial adhesion [4, 5].

The coupling agents are deliberately designed to contain chemical functional groups that can react with hydroxylated inorganic surfaces, producing covalent bond linkages. The coupling agent then acts as a bridge to

bond the fiber to the polymer matrix with a chain of primary bonds that, in principle, could be expected to lead to the strongest interfacial bonds [6].

Several studies showed that fiber–polymer bonding can be improved by the use of coupling agents [6–9]. In some cases it was verified that the use of coupling agents also served to moderate and somewhat mitigate moisture movement through the composite, thus improving the mechanical properties of the materials [10].

The interface region where contact occurs between the polymeric matrix and the vegetable fiber is responsible for the mechanical load transfer from the matrix to the reinforcement. The application of such compatibilizers should improve the dispersion of fillers in a polymeric matrix, as noted by many authors [12–32].

In this work we considered commercial and customized coupling agents, used to improve some polypropylene mechanical and thermal properties. As far as authors know, there are dozens of coupling agents and natural fibers mixed in polymers used since the pioneering work of Meyer in 1968 [13]. The reader is invited to follow the interesting work of Lu *et al* [14] about this subject.

The addition of polyethylene to polypropylene should improve composite characteristics, promoting a low temperature processing impact and diminishing environmental stress cracking. Due to immiscibility, in order to enhance the ultimate properties, frequently a compatibilizer like ethylene-propylene rubber (EPR) has to be used to ascertain a good relation between the tensile impact strength and HDPE content [15, 18]. In this context, the primary objective of this work was to evaluate the influence of different coupling agents on the mechanical, morphological and thermal properties of a polypropylene composite with 30 wt% percentage of coconut fibers. For this purpose, a 6 wt% content of coupling agents relative to the fiber weight was applied in all of the studied cases and compared to a formulation without the addition of coupling agents. Such percentage choices for vegetable fibers and coupling agents correspond to usual values found in literature [16]. For example, Saheb and Jog [11] verified that the fiber loading can be as high as 80%, considering their analysis from different fiber reinforced polymers. Lu *et al* [14] verified that different coupling agents were usually applied between 2 up to 8% by weight of natural fibers, similarly to the work of Bettini *et al* [16] that studied polypropylene and coconut fibers.

2. Materials and methods

The natural fibers came from the region of Conde—Bahia, Brazil. The average length of coconut fibers after grinding in a knife mill was (10.9 ± 5.2) mm and the average diameter was $(212,2 \pm 10,3)$ μm . The density of coconut fiber depends on the region and drying time, may range from 0.67 to 1.5 g cm^{-3} as quoted by Siakeng *et al* [17] in your research. The polypropylene EP 440L was acquired from Braskem with a melt flow index of 6.0 g/10 min according to ASTM D 1238. This copolymer contains as dispersed phase (grafted) an Ethylene Propylene Rubber (EPR).

The following commercial coupling agents with the percentage of 6% wt% related to coconut fiber percentage were used: (i) Orevac CA 100, a polypropylene (PP) grafted with 1 wt% maleic anhydride content; (ii) Orevac 18507, a high density polyethylene (HDPE) also grafted with 1 wt% maleic anhydride (both supplied by Arkema company); and (iii) Cesa[®] Mix PEA0601031 a combination of lubricant based in stearamine and titanate as a coupling agent, grafted with high density polyethylene, an additive from Clariant Company. This additive customized in our laboratory at SENAI CIMATEC acted as both a coupling agent by modifying the surface energy of the dispersed phase of the composites and worked as lubricant to help in phase dispersion.

Therefore, in this study, coupling additives with PE and PP bases were tested to verify which has the best interaction between the polymer matrix of the copolymer and coconut fiber. The polypropylene and polyethylene are immiscible and partially compatible [18] and this blends have been attract a lot of attention due to their potential industrial applications for impact improvement and environmental stress cracking proprieties [15, 16].

All such coupling agents showed compatibility with materials used with the specific proportions of this work. In the present investigation another main objective was to develop compatibilization of fiber composites to attain maximum improvement in ultimate mechanical properties.

2.1. Preparation of the composite

To assess which formulation would provide better adhesion and wetting between polymer and vegetable fiber, four composite formulations were developed with coconut fiber and polypropylene.

The compositions were weighed on a semi-analytical balance (Toledo 9094) with polypropylene and the respective coupling agent, as shown in table 1. The contents of coconut fiber and the coupling agent are relative to the total mass of the composite sample (30%), with the percentage of 6% additive associated to the fiber content, resulting 1.8% total mass.

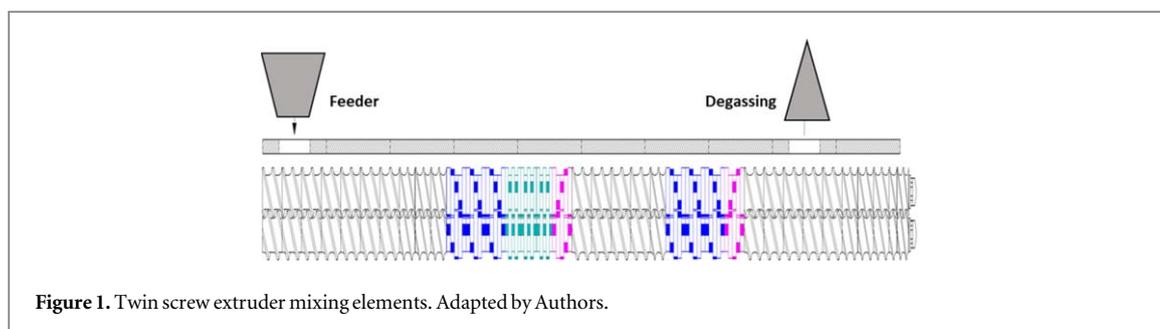


Figure 1. Twin screw extruder mixing elements. Adapted by Authors.

Table 1. Formulations used in the sample preparations (wt%).

Formulations	PP EP 440L	Coconut fiber	Orevac CA 100	Orevac 18507	Cesa® Mix
PPFC	70	30	—	—	—
PPOCA100	68.2	30	1.8	—	—
PPO18507	68.2	30	—	1.8	—
PPCM	68.2	30	—	—	1.8

Legend: PP EP 440L—Polypropylene; PPCF—Composite of polypropylene and coconut fiber without coupling agent; PPOCA100—Composite of polypropylene and coconut fiber with Orevac CA 100 as coupling agent; PPO18507—Composite of polypropylene and coconut fiber with Orevac 18507 as coupling agent; PPCM—Composite of polypropylene and coconut fiber with Cesa® Mix as coupling agent.

After weighing the formulations, the premix was dosed at the main feed point of a corotating screw extruder manufactured by Imacom, model DRC 30:40 IF with thread diameter of 30 mm and L/D ratio = 40 between length and diameter. The processing conditions for these composites were: (i) screw speed: 140 rpm; (ii) feeding speed: 8 rpm; (iii) mass temperature: 193 °C. The thread profile used can be considered typical for the production of composites with vegetable fibers and is composed of two mixing zones formed with kneading blocks of 45° and 90° and the other elements designed to the transport of material as showed in figure 1.

2.2. Preparing injected samples

After the composite formulation step, the materials were dried at 100 °C for a period of 8 hours and used to prepare the specimens by an injection process in an ROMI machine (Model Primax 100 R). The injection of the specimen was given at a pressure of 950 bar, $120 \text{ cm}^3 \text{ s}^{-1}$ and a temperature of 200 °C of the molten material.

2.3. Characterization of the composites

The mechanical characterization of the composites under tensile strength followed ISO 527 standard. Ten samples were used for mechanical tests. The experiments were conducted in a universal testing machine (Emic Model DL 2000) at a strain rate of 1 mm min^{-1} . The IZOD impact strength test was conducted in a machine (model Emic Pendulum Machine) with a 2.7 J hammer according to ISO 180 standard. The composites fracture surface morphologies were evaluated using a scanning electron microscope (Jeol, model JSM—6510LV) with a voltage ranging from 15 to 20 kV. Differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) of the samples of pure polypropylene and the composites were performed in a differential scanning calorimeter (Netzsch, model DSC 200) using a heating rate of 10 °C min^{-1} and a cooling rate of 20 °C min^{-1} under inert argon atmosphere with a flow rate of 100 ml min^{-1} in a sealed aluminum sample holder. After milling, samples weighing about 10 mg were used in measurements. For the TGA analysis, a Shimadzu equipment (model TGA 50H) was used with the same heating DSC conditions and with an open platinum crucible under nitrogen atmosphere with a 50 ml min^{-1} flow rate.

3. Results and discussion

The analysis of mechanical properties of the composites associated with microscopy and thermal analysis will be discussed in the next topics.

3.1. Mechanical characterization of the composites

The mechanical properties of the yield stress, ultimate tensile strength and Young modulus under tension are shown in table 2.

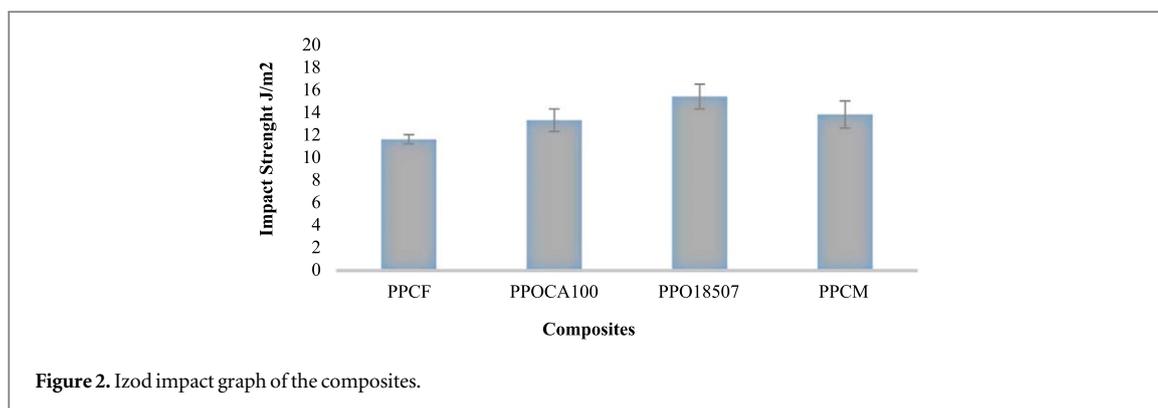


Figure 2. Izod impact graph of the composites.

Table 2. Mean and deviation values of mechanical properties under uniaxial tension.

Formulation of the composite	Yield strength (MPa)	Tensile strength (MPa)	Young modulus (MPa)
PP—EP440 L ^a	17.18 ± 0.12	14.6 ± 1.2	903 ± 35
Coconut Fiber (CF)	100.9 ± 25.7	100.1 ± 26	2657 ± 514
PPCF	16.95 ± 0.29	14.32 ± 0.57	1608 ± 63
PPOCA100	22.35 ± 0.32	21.51 ± 0.30	1530 ± 24
PPO18507	17.41 ± 0.22	13.60 ± 0.77	1398 ± 79
PPCM	16.17 ± 0.29	13.45 ± 0.44	1302 ± 55

^a PP- EP 440 L was submitted to an extrusion in the same conditions of composites to injection of samples.

The values of the mechanical properties for the coconut fiber (CF) exhibited approximately 588% higher than that of the polymer matrix used for the composites. After analyzing such results, it was found that the best value of tensile stress was obtained for the composites with Orevac CA 100. The addition of polypropylene functionalized with maleic anhydride in this composite provided a higher interaction between fiber and the polymer matrix and could transfer the tensile strength observed in CF to this particular composite. In fact, tests using composites with Orevac 18507 and Cesa[®] Mix, which have a polyethylene base, the interaction of the additive with the polypropylene matrix was quite satisfactory considering tensile test results, in spite of the heterophasic character of the polymer matrix. HDPE has lower Young's modulus than PP [19], so probably the composites with HDPE content and the coupling agent contributed to decrease this particular result. Tensile measurements indicated that the tensile strength as well as the Young modulus decreased with PE addition [15, 17]. Figure 5 shows the stress-strain curve of composites where is possible to observe insignificant variations in the values of the yield strength in PPO18507 and PPCM composites in relation to the composite without a coupling agent (PPFC). The increased stiffness in the composites with coconut fibers can be attributed to the hindrance of movement of the polymer matrix chains due to the strong interaction of the additive reacting with the free OH⁻ on the fiber surface. This behavior has been observed particularly in studies using vegetable fibers with polypropylene composites [11]. Commonly, the effect of a coupling agent can be better observed at higher stresses, when measuring the tensile strength, because a strong interface is crucial. In fact, this behavior was more strong in PPOCA100 and has been observed particularly in studies using vegetable fibers with polypropylene composites [5].

Based on the IZOD impact strength test results illustrated in figure 2, it was possible to observe that in general the composites exhibited better results than their counterparts without additives.

This result could indicate the participation of the coupling agent interacting more strongly with the fibers through possible strong covalent bonds or via secondary interactions as tie and molecular entanglements [20]; these interactions would result in a better interfacial adhesion between the coconut fiber reinforcement and the polymer matrix [4, 5]. Furthermore, the addition of a small HDPE amount, that presented more ductile behavior [15], as noted in Orevac 18507 and Cesa[®] Mix samples, improved the impact result as observed in figure 3. However, the main result was that the introduction of fibers reduced dramatically the impact strength of the polymer matrix where the results of impact resistance obtained was around 40 J m⁻². In a similar study with vegetable fibers and thermoplastic polypropylene matrix [7], it was found that the impact strength in thermoplastic composites depends on the ability of the matrix to dissipate deformation energy at high shear rates, where the characteristics of the interface are crucial for this process.

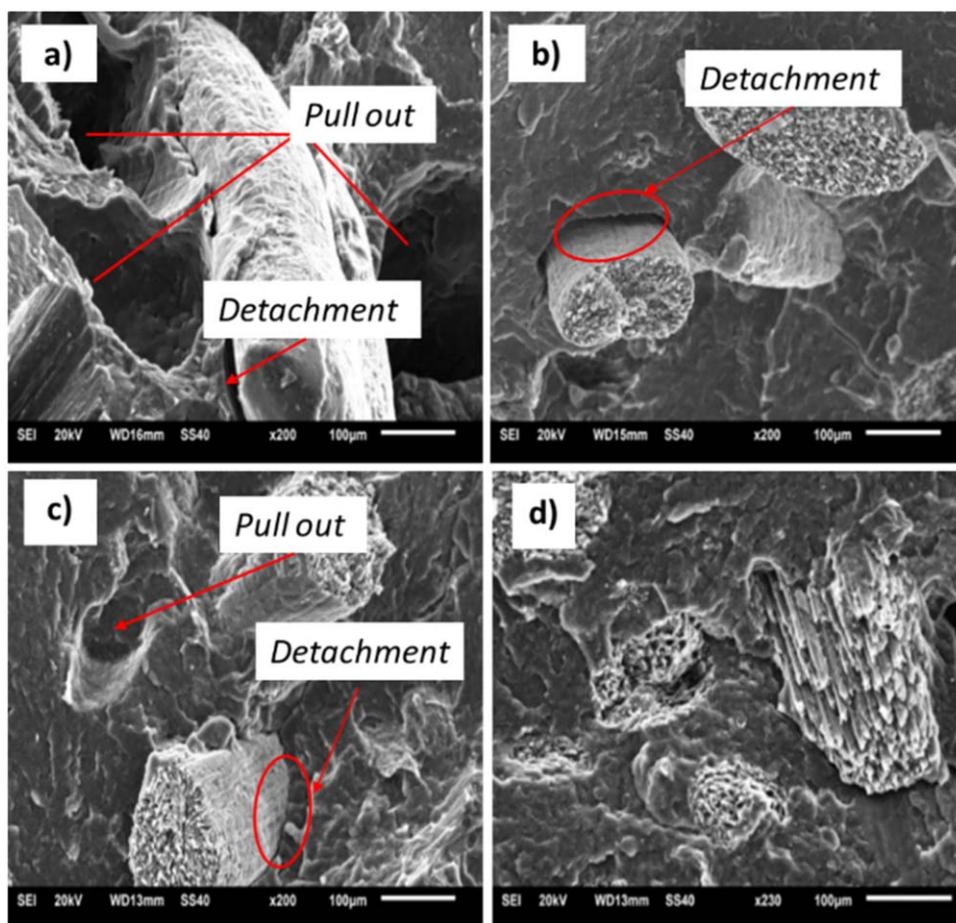


Figure 3. SEM images of the composite surfaces. (a) CF sample without additives; (b) with Cesa[®] Mix; (c) with Orevac 18507; and (d) with Orevac CA 100.

In general, the filler causes a decrease of impact resistance and the crack travel more easily through the weaker interfacial regions hence decreases this property the composite [21].

3.2. Morphological analysis of the composites

For a better understanding of the results of the mechanical properties associated with reinforcement theory, the cryogenically fractured samples were subjected to scanning electron microscopy, as shown in figure 4. When the material is stretched, a portion of the load is absorbed by the fiber, restricting the polymer mobility, which therefore increases the tensile strength composite [6]. If the bond between the fiber and the matrix is strong, the fibers break, which means that the interface is stronger than the fiber itself. When the interaction is weak, it cannot support all stress, and many fibers no longer adhere to the matrix. Palaniyandi and Simonsen [12] observed something similar to this result considering some compatibilizers added with wood flour in HDPE matrix. Figure 3(a) shows the presence of voids and a fiber detachment in the composite without additive, which may indicate the existence of a low affinity of the polymer matrix with coconut fibers. These voids may occur due to incomplete wetting of the resin on fibers, forming for example air bubbles (or other gases) that may get stuck due to resin high viscosity or by virtue of volatile compounds that are released during the composite preparation process [11]. The gap between filler-matrix also can be related due to poor adhesion. Besides, some voids might found in composites, and it also probably due to fiber pull out [22]. The image in figure 3(b) indicates a partial adhesion of the composite using Cesa[®] Mix because there were voids in the interfacial region between fiber and matrix; the same figure also exhibited a porous fracture surface and disrupted/broken fibers. Similar behavior can be observed for the compound with Orevac 18507 in figure 3(c), where it was observed some regions with poor adhesion and fiber pull-out, but is also possible to note evidences of partial adhesion. It was not possible to note phase segregation between the HDPE contained in these coupling agents and the polymer PP matrix, indicating an apparent compatibility of the materials used in this work. Figure 3(d) demonstrates the behavior in the composite with Orevac CA 100 added, in which, due to the influence of the coupling agent, it was impossible to observe voids and signs of fiber pull-out.

The images show that there was effective shear on fibers, which confirms the results of mechanical tests, via a better interfacial adhesion between phases as compared to the non-coupled composite. The results provided by

SEM complement the characterization study, where the samples with additives had an increase in the ultimate tensile stress up to 23% compared with a composition without additives; these results are also in agreement with the studies performed by other researches [7, 8].

3.3. Analysis by differential scanning calorimetry of the composites

Table 3 shows the results of the DSC tests regarding the melting enthalpy (ΔH), melting temperature (T_m), the degree of crystallinity peak and the characteristics melting temperatures (T_{onset} , T_{endset}) of the composite materials obtained during the second heating run.

According to Ehrenstein *et al* [23], the degree of crystallinity is defined from the area bounded by the enthalpy of melting change curve and the baseline of a thermal apparatus (as DSC or DTA). This energy is necessary to melt the crystalline fraction, named heat of fusion, and can be considered as a way to calculate the crystalline fraction of a polymer.

In DSC, the crystallinity fraction is conventionally measured by dividing the enthalpy change associated with T_m ($^{\circ}\text{C}$), ΔH_m (J/g), by the enthalpy of fusion of a 100% crystalline polymer sample, ΔH_m° . The heat of fusion for pure polypropylene crystals is 209 J g^{-1} [23, 34]. Equation (1) represents the calculation of the degree of crystallinity of polymers:

$$X_c(\%) = 100 \frac{H_m}{H_m^{\circ}} \quad (1)$$

Where X_c is the degree of crystallization or crystallinity.

From the values of the melting enthalpy (ΔH) and considering a proportion of 30% of coconut fibers in the composite, it can be concluded that the mean value of ΔH melting is lower than 80% of the melting ΔH value of the PP EP 440L, leading to the conclusion that the fiber caused significant decrease in the crystallinity matrix degree. This behavior was also observed by Techawinyutham *et al* [24] in studies with polypropylene/coconut fiber composites and similar to that reported by Karsli *et al* [25].

The crystalline melting point (T_m) during second heating revealed that the coupling agent and coconut fiber had no effect on the melting points of composites. It can be noted that in the composites with Orevac 18507 and Cesa[®] Mix, a broadening of the peak area occurred. This temperature difference (ΔT) between T_{onset} and T_{endset} larger indicate that happen a wider distribution of the sizes of the crystals in composites with coupling agents PEAD base. Jose *et al* [26], in studies using PP and HDPE blends, reported that the addition of HDPE decreased PP spherulite size by occupying the interspherulite region and thereby increased the impact resistance.

By analyzing the curves of the second heating shown in the thermograms on figure 4, peaks located by red circles were observed at lower temperatures, between 120 and 130 $^{\circ}\text{C}$. These peaks correspond to a smaller or less crystallizable component of the sample, possibly polyethylene (PE) or a PP-PE copolymer [27]. This event was more accentuated with the coupling agents as Orevac 18507 and Cesa[®] Mix that contain PE in their formulations. These values are in agreement with the thermograms presented by Jose *et al* [26] in their study. We only could observe this particular peak in PPCF and PPCA100 in the first heating, probably the low content of PE in these composites became the peak more dispersed.

In the thermograms of figure 5 it is also possible to note that for the composites with PE matrix additives, the crystallization temperature shifts occur delayed. Blom *et al* [28], observed the same behavior in their studies, and argued that there is a certain degree of interaction between PP and HDPE at low concentrations, that basically reducing the number and size of the regions of the high density segments, thus delaying nucleation and the crystallization of PP phase.

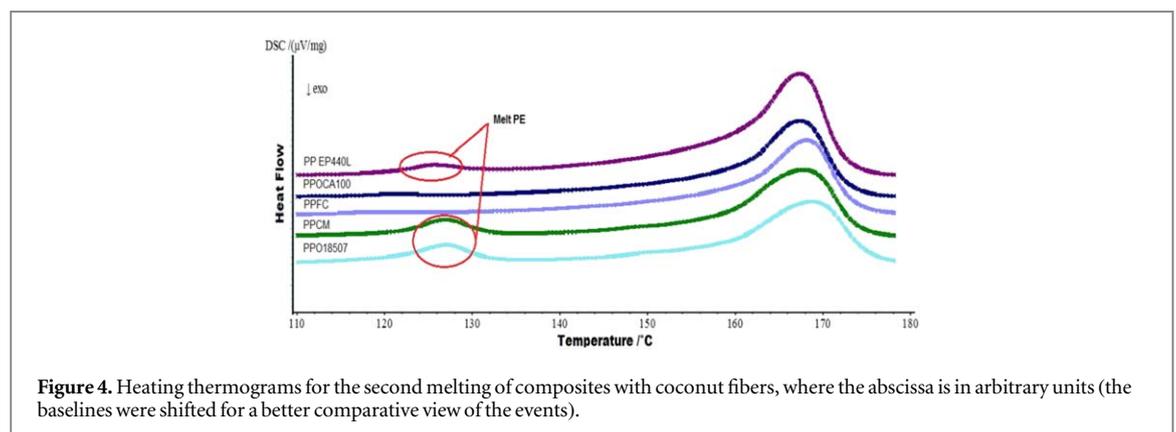


Figure 4. Heating thermograms for the second melting of composites with coconut fibers, where the abscissa is in arbitrary units (the baselines were shifted for a better comparative view of the events).

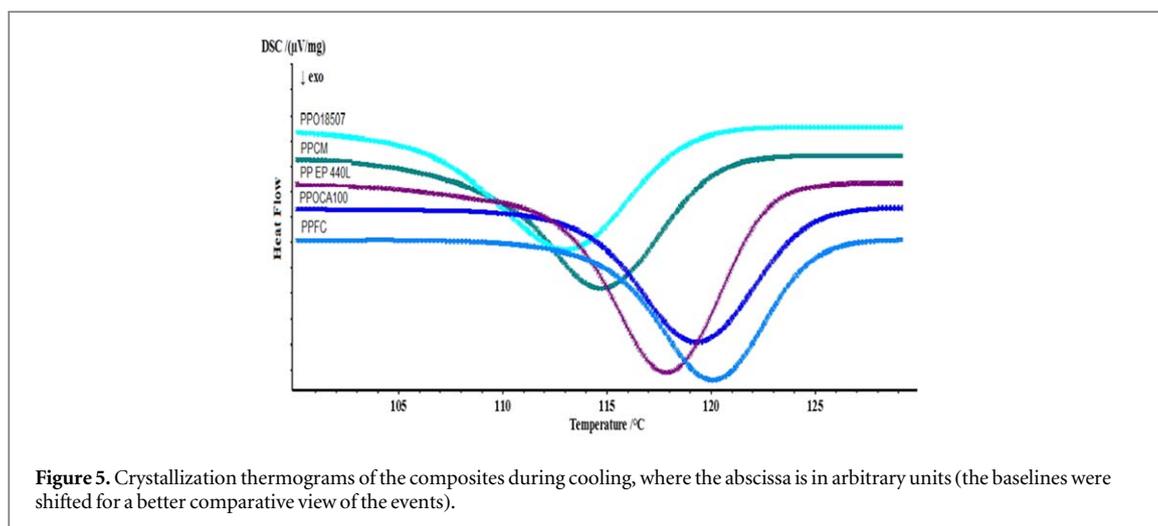


Figure 5. Crystallization thermograms of the composites during cooling, where the abscissa is in arbitrary units (the baselines were shifted for a better comparative view of the events).

Table 3. Values of the characteristic DSC thermal variables in the second heating.

Samples	$\Delta H_m(\text{J g}^{-1})$	$X_c\%$	$T_m(\text{peak})^\circ\text{C}$	$T_{onset}^\circ\text{C}$	$T_{endset}^\circ\text{C}$	$\Delta T^\circ\text{C}$
PP EP 440 L	73,60	36	167,3	160,2	172,4	12,2
PPCF	54,26	26	168,1	160,6	173,3	12,7
PPOCA100	55,03	27	167,2	160,2	172,8	12,6
PPO18507	49,40	24	168,7	158,6	174,9	16,3
PPCM	52,37	25	167,8	158,2	173,8	15,6

Table 4. Values of the characteristic DSC thermal variables in second cooling treatment.

Samples	$\Delta H_c(\text{J/g})$	T_c (peak) $^\circ\text{C}$	$T_c^{onset}^\circ\text{C}$	$T_c^{endset}^\circ\text{C}$
PP EP 440L	92.57	117.90	122.80	112.40
PPCF	65.83	120.00	124.90	114.70
PPOCA100	67.14	119.30	124.70	113.80
PPO18507	71.46	113.00	118.90	114.90
PPCM	71.34	114.70	120.30	109.80

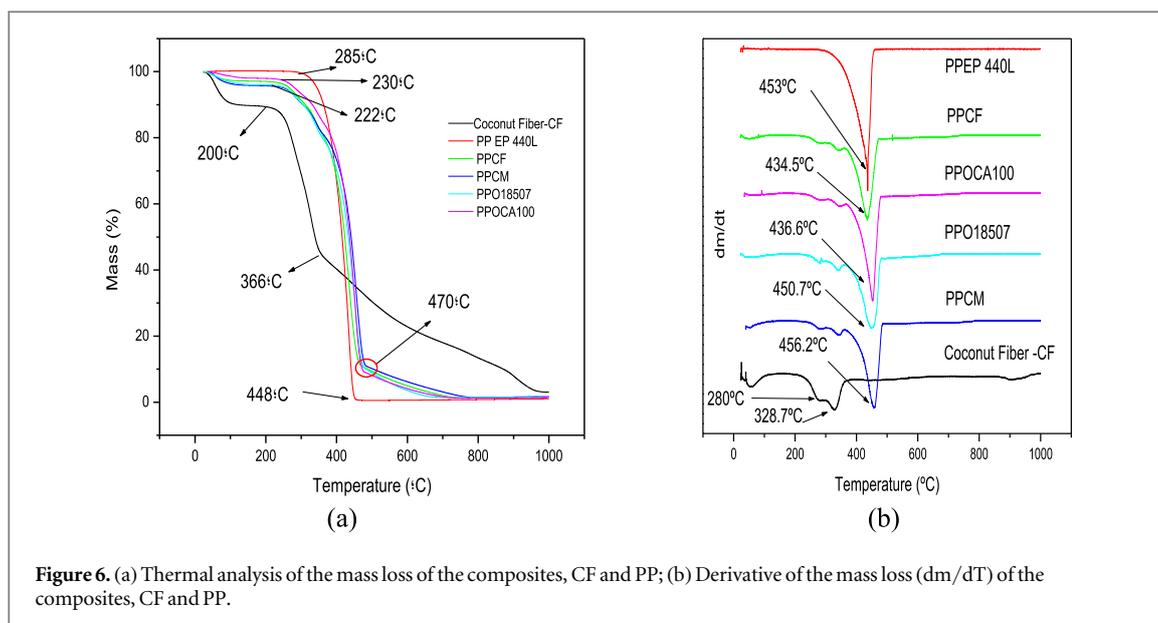
It was observed that the crystallization temperature (T_{onset}) moved to higher temperatures for the composite without additives PPFC. This effect was also observed with PPOCA100 composite as showed in table 4 during the second cooling treatment. This result may be explained by considering the ability of the fibers for nucleating the crystallization of polypropylene [2].

Other hypothesis for this behavior is related to the formation of a layer with different crystallinity, known as the transcrystalline layer, that may have occurred along the fiber in contact with the polymer matrix [29]. Pracella *et al* [30] pointed that the highest nucleation density of PP spherulites occurred preferentially on the wall surface of fibers. This effect was also evident for compounds with maleic anhydride grafted with PP as a consequence of the stronger interactions of coconut fibers with polymeric matrix.

The increase in the impact properties of PPO18507 composite could be related to the addition of HDPE with the coupling agent, thus changing the crystallinity temperature lead to a decrease of PP spherulite sizes [26]. During this process, molten HDPE flowed into the region between PP spherulites growing near the interface. This process promoted a deformation of the interface between PP and PE, which was only interrupted after the complete crystallization of polypropylene. The result increased the interfacial area represented by ΔH_c , as well as reduced the smaller T_c (*onset*) values [26]. The PPCM presented the same temperature displacement but the interaction of the fibers using titanate as a coupling agent were not so effective to mechanical properties analyzed.

3.4. Thermogravimetric analysis of the composites

By analyzing the thermograms of the coconut fibers TGA curve, PP EP 440L and the composites obtained (with and without coupling agents), it was observed that all composites had increased its thermal stability in the region



between 448 °C–470 °C. This is probably due to the lignin content present in the coconut fiber that has a higher temperature for its degradation onset. In fact, the polymer matrix thermally ‘protects’ fibers before thermal degradation and this would cause a small shift in mass loss at the beginning of the process, as expected. The composite curves, regardless the presence of additives, resulted between the curves of coconut fibers and pure polypropylene. It can be concluded that the composites with coupling agents exhibited a higher thermal stability than untreated composites probably due to the same mass loss behavior at higher temperatures, i.e., samples can withstand higher temperatures until thermal degradation.

According Chun *et al* [22] the natural fiber reinforced composite usually exhibited an earlier thermal degradation due to removing of hemicellulose and moisture. Then, the thermal degradation of lignin may yield high amount of char, which causes char formation effect/thermal shielding effect on composites, thus the composite shows better thermal stability at higher temperature.

This result is similar to that found by others researches [5, 29] about polypropylene and vegetable fibers. In the analysis of the derivative of the TGA curve (DTG), an event was found at approximately 280 °C, which can be attributed to the degradation of the cellulose and the peripheral regions of the lignin. Another event at 328 °C was attributed to be the highest lignin degradation. The thermal degradation of compounds reached peaks around 470 °C. This weight loss was attributed to thermal depolymerization of hemicellulose and cleavage of the glucosidic linkages of cellulose. These events observed in figures 6(a) and (b) were noted in all compositions at the same temperature ranges, which characterize the cumulative effect of the vegetable fibers in the composite.

4. Conclusions

In this work it was observed that the addition of coupling agents to polypropylene had a positive effect on the mechanical properties of the composite, particularly in conjunction with the Orevac CA 100, which had an increase in the ultimate tensile strength of 23% at break and of 32% in relation to the polymer matrix. The impact strength of the composites showed significant reductions regardless of the use of coupling agents. The composite PPO18507 with HDPE content in coupling agent decreases the PP spherulite size by occupying the interspherulite region and thereby increased impact resistance results. The PPCM had the same polymer in its formulation, but the interaction between fiber and the titanate as coupling agent was not so effective.

SEM micrographs of fractured surfaces also showed conclusive evidence regarding the increase in the wetting ability of the cellulose fiber by the polymeric resin in the presence of the coupling agent. The DSC and TGA thermal analyses of the composites with coconut fibers showed that the addition of coconut fibers promoted a decrease in the crystallinity degree of the polymer matrix. The coconut fiber content provided the highest nucleation density of PP spherulites on the wall surface of the fibers. This effect was also evidenced for compounds with polypropylene grafted with maleic anhydride. The composites PPCM and PPO18507 resulted in a more ductile behavior between the composites as result of PE contained in their coupling agents. Given the results described in this study, the use of coconut fibers with a coupling agent can be considered as sustainable and adequate for the manufacture of new composites. In addition, to reducing the amount of solid waste, the low cost of this natural fiber makes possible to obtain polymer composites with better mechanical properties under

tension than the polymer matrix. The best compatibility of the matrix with coupling agent was obtained by Orevac CA 100, but considering impact resistance as an important factor in any given application, polyethylene could be integrated as an impact modifier.

The mechanical, thermal and morphological properties enhanced by the addition of the coconut fiber and coupling agents in biocomposites. The reinforced and sustainable appearance of these materials may allow being suitable for structural engineering, furniture, decking, automotive and home appliance parts. The natural fiber composites reduce the environmental impact and help to generate incomes for local communities.

Acknowledgments

The present work was supported by SENAI CIMATEC, Federal University of Bahia, via the Graduate Program in Industrial Engineering. We are also thankful for the support of the Brazilian National Research Council, CNPq Grant Nos. 300938/2012-8, 471546/2012-7 and 304705/2015-2.

ORCID iDs

Luciano Pisanu  <https://orcid.org/0000-0001-7727-6978>

References

- [1] Baker A M and Mead J 2004 Handbook of plastics, elastomers, and composites *Digital En.* (Massachusetts: McGraw-Hill Companies-Digital Engineering Library)
- [2] Leão R M, Luz S M, Araujo J A and Novack K 2015 Surface treatment of coconut fiber and its application in composite materials for reinforcement of polypropylene *J. Nat. Fibers* **12** 574–86
- [3] Parandoush P and Lin D 2017 A review on additive manufacturing of polymer-fiber composites *Compos. Struct.* **182** 36–53
- [4] Morandim-Giannetti A A, Agnelli J A M, Lanças B Z, Magnabosco R, Casarin S A and Bettini S H P 2012 Lignin as additive in polypropylene/coir composites: thermal, mechanical and morphological properties *Carbohydr Polym* **87** 2563–8
- [5] Catto A L, Stefani B V, Ribeiro V F, Marlene R and Santana C 2014 Influence of coupling agent in compatibility of post-consumer HDPE in thermoplastic composites reinforced with eucalyptus fiber *Mater. Res.* **17** 203–9
- [6] Owen M J, Corning D and Midland C 2002 Coupling agents: chemical bonding at interfaces *Surfaces, Chem. Appl.* (Midland, USA: Elsevier B.V) p 403–31
- [7] John M J and Anandjiwala R D 2008 Recent developments in chemical modification and characterization of natural fiber-reinforced composites *Polym. Compos.* **16** 101–13
- [8] Paul S A, Joseph K, Mathew G D G, Pothen L A and Thomas S 2010 Influence of polarity parameters on the mechanical properties of composites from polypropylene fiber and short banana fiber *Compos Part A Appl Sci Manuf* **41** 1380–7
- [9] Roberts D H 1999 Chemical coupling of polypropylene systems containing nonglass fillers *J Vinyl Addit Technol* **5** 231–4
- [10] Nachtigall S M B, Cerveira G S and Rosa S M L 2007 New polymeric-coupling agent for polypropylene/wood-flour composites *Polym. Test.* **26** 619–28
- [11] Saheb D N and Jog J P 2016 Natural fiber polymer composites: a review *Adv Polym Technol* **2329** 351–63
- [12] Palaniyandi V and Simonsen J 2007 Effect of compatibilizers on the crystallization kinetics of cellulose-filled high density polyethylene *Compos. Interfaces* **14** 73–83
- [13] Meyer A J 1968 Crosslinking affects sanding properties of wood-plastic. In *Journal FP* editor. Wood fiber Sci., Forest Production Journal **1** 89–94
- [14] Lu J, Wu Q, McNabb H, Harold S and McNabb J J 2000 Chemical coupling in wood fiber and polymer composites: a review of coupling agents and treatments *Wood Fiber Sci.* **32** 88–104
- [15] Mourad A H I 2010 Thermo-mechanical characteristics of thermally aged polyethylene/polypropylene blends *Mater. Des.* **31** 918–29
- [16] Bettini S H P et al 2010 Investigation on the use of coir fiber as alternative reinforcement in polypropylene *J. Appl. Polym. Sci.* **118** 2841–8
- [17] Siakeng R, Jawaid M, Ariffin H and Sapuan S M 2019 Mechanical, dynamic, and thermomechanical properties of coir/pineapple leaf fiber reinforced polylactic acid hybrid biocomposites *Polym. Compos.* **40** 2000–11
- [18] Shanks R A, Li J and Yu L 2000 Polypropylene-polyethylene blend morphology controlled by time-temperature-miscibility *Polymer (Guildf)* **41** 2133–9
- [19] Tai C M, Li R K Y and Ng C N 2000 Impact behaviour of polypropylene/polyethylene blends *Polym. Test.* **19** 143–54
- [20] Rabello M S and White J R 1997 Polypropylene photodegradation: An inhomogeneous process. *Polimeros Ciência e Tecnol* (Brazil: Associação Brasileira de Polímeros - ABPol) 47–57 (http://www.scielo.br/scielo.php?script=sci_arttext&pid=S0104-14281997000200007)
- [21] Sarkar N, Sahoo G, Khuntia T, Priyadarsini P, Mohanty J R and Swain S K 2017 Fabrication of acrylic modified coconut fiber reinforced polypropylene biocomposites: study of mechanical, thermal, and erosion properties *Polym. Compos.* **38** 2852–62
- [22] Chun K S, Husseinayah S and Syazwani N F 2016 Properties of kapok husk-filled linear low-density polyethylene ecocomposites *J. Thermoplast. Compos. Mater.* **29** 1641–55
- [23] Ehrenstein G W, Riedel G and Trawiel P 2004 Thermal analysis of plastics *Therm Anal Plast* (Germany: Hanser) I–XXIX 978-3-446-43414-1
- [24] Techawinyutham L, Frick A and Siengchin S 2016 Polypropylene/maleic anhydride grafted polypropylene (MAGPP)/coconut fiber composites *Adv Mech Eng* **8** 1–9
- [25] Karsli N G and Aytac A 2011 Effects of maleated polypropylene on the morphology, thermal and mechanical properties of short carbon fiber reinforced polypropylene composites *Mater. Des.* **32** 4069–73

- [26] Jose S et al 2004 Phase morphology, crystallisation behaviour and mechanical properties of isotactic polypropylene/high density polyethylene blends *Eur. Polym. J.* **40** 2105–15
- [27] Pereira C et al 2019 Heterophasic polypropylene and wood flour composites: processing and properties *Mater. Res. Express* **6** 085321
- [28] Teh J W, Blom H P and Rudin A 1994 A study on the crystallization behaviour of polypropylene, polyethylene and their blends by dynamic mechanical and thermal methods *Polymer (Guildf)* **35** 1680–7
- [29] Luz S M, Del Tio J, Rocha G J M, Gonçalves A R and Del'Arco A P 2008 Cellulose and cellulignin from sugarcane bagasse reinforced polypropylene composites: effect of acetylation on mechanical and thermal properties *Compos Part A Appl Sci Manuf* **39** 1362–9
- [30] Pracella M, Chionna D, Anguillesi I, Kulinski Z and Piorkowska E 2006 Functionalization, compatibilization and properties of polypropylene composites with Hemp fibres *Compos. Sci. Technol.* **66** 2218–30
- [31] Cantero G, Arbelaz A, Mugika F, Valea A and Inaki M 2003 Mechanical behavior of wood/polypropylene composites: effects of fibre treatments and ageing processes *J. Reinf. Plast. Compos.* **22** 13
- [32] Dányádi L, Janecska T, Szabó Z, Nagy G, Móczó J and Pukánszky B 2007 Wood flour filled PP composites: compatibilization and adhesion *Compos. Sci. Technol.* **67** 2838–46
- [33] Li J, Shanks R A, Olley R H and Greenway G R 2001 Miscibility and isothermal crystallisation of polypropylene in polyethylene melts *Polymer (Guildf)* **42** 7685–94
- [34] Canevarolo S V 2010 *Ciência dos Polímeros* 1a edn (São Paulo: Artliber Editor) (<https://doi.org/03-5795>)