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Recycled Polypropylene-Coffee Husk and Coir Coconut Biocomposites: Morphological, Mechanical, Thermal and Environmental Studies

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Abstract

In this work, biocomposites based on recycled polypropylene (r-PP) and two different natural fibers (coffee husk-CHF and coconut coir-CCF fibers) were prepared using extrusion and injection molding processes. Also, the addition of maleated polypropylene (MAPP) as a coupling agent on the biocomposites was explored. Recycled polypropylene and its biocomposites were tested following ASTM standards in order to evaluate tensile and flexural mechanical properties. Also, thermal behavior and the morphology of these materials have been studied by differential scanning calorimetry (DSC), thermogravimetric analysis (TGA), and scanning electronic microscopy (SEM). The experimental results showed that the addition of CHF and CCF to the r-PP resulted in an increase in the flexural modulus and thermal properties of the composites but resulted in poor impact properties. Thermal characterization showed that CHF possesses a better thermal stability compared to CCF. However, both fibers act as nucleating agents and generate an increase in the thermal stability of the r-PP phase. Finally, it was observed that addition of 4% of MAPP significantly improved the mechanical strength and impact behavior of the biocomposites. Regarding environmental issues, a cradle to gate life cycle assessment was made in order to define the carbon footprint of the materials.

Keywords: coconut coir, coffee husk, recycled polypropylene, biocomposites, MAPP

1. Introduction

Residual biomass is defined as a compound that contains mainly nonedible vegetal material called lignocellulose. Lignocellulose is the most important component found in plant tissues and is composed of three different polymers: cellulose,

hemicellulose, and lignin. Each of these components can be found on different parts of the biological structure of the plant, being the hemicellulose is the matrix that covers the cellulose skeleton and the lignin is the encrusting material or protective layer [1].

On the other hand, a plastic waste is defined as the material recovered by the final users after having complied with the use for which it was produced [2, 3]. This type of waste is classified in two categories: postconsumer plastic and postindustrial. The first one refers to residual plastics that have been previously used by people. In contrast, postindustrial or preconsumer plastics are defined as the industrial reject material (cuts of materials and damaged pieces, among others) that is not returned to the production line. These are recycled to a great extent, due to the high availability that exists and its relative degree of purity.

Around 140 billion tons/year of biomass wastes are generated in the world as a result of agricultural activities [4] and 230 million tons/year of plastic wastes [5] related to the production of these materials. In the case of Colombia, an estimated production of 72 million tons/year of residual biomass is reported [6]. Crops such as coffee, bananas, coconut, corn, and sugar cane contribute a large proportion to this production. Waste generated by the coconut processing industry includes its shell, water, and coir. Shell and coir represent 35% in weight of the entire fruit. In Colombia, about 4100 tons/year of this type of waste are produced that is the reason why some studies are being carried out in the biotechnology and construction fields to give them an adequate use [7]. There are two types of coir, the brown coir which is obtained from mature coconuts and the white coir which is extracted from green coconuts. Generally, this type of fiber has a length of 350 mm, a diameter between 0.12 and 0.25 mm, and a density of 1250 kg/m^3 . It is a material resistant to microbial degradation and salt water. It has a high content of lignin and is defined as a strong material with a high tensile strength [8]. On the other hand, one of the wastes generated in large quantities during the process of the coffee bean transformation is the coffee husk. This material represents 4.5% of the grain composition, and about 33.000 tons/year is produced in Colombia [9]. The proposed uses for this waste are fermentation in order to obtain enzymes, organic acids, or bioethanol. Also, it is used as a substrate for the growth of fungi and other microorganisms [10]. This type of vegetable fiber has an average diameter of 1.2 mm, a high content of holocellulose, as well as a significant proportion of lignin [11].

On the other hand, the national demand for plastic resins is close to 1.2 million ton/year [12], of which about 27.5% are recovered [13]. The rest of the material is disposed in landfills or inadequately in open dumps. **Tables 1** and **2** show the waste generation of the main agricultural crops and plastic resins in Colombia, respectively.

There are different studies from different areas related to the use of biomass waste. A great number of treatments have been proposed to add value to this type of material or simply to change its characteristics and make its final disposition simpler [15]. The main areas for the use of biomass waste are animal and human nutrition, energy generation, biotechnology industry, and the production of biocomposites (natural fiber reinforced polymers or NFRP).

Since several decades, biocomposites have emerged as an option aimed to solve several issues within the composite materials science. In most of published cases in literature, the use of natural fibers combined with polymers is carried out to achieve some degree of reinforcement from the fibers to the polymer. Many studies report the use of natural fibers such as flax, hemp, jute, sisal, coconut fiber, banana, and fique, among many others [16], using an extensive variety of polymer matrices like polyethylene [17, 18], polypropylene (PP) [19] polystyrene (PS) [20], epoxy resin (EP) [21], natural rubber [22], and recycled polypropylene (r-PP) [2]. Clear effects have been seen in the improvement of mechanical and thermal performance.

Crop	Production (Ton/year) [6]	Waste	Waste factor (Ton _{waste} /Ton _{product}) [6]	Waste mass (Ton/year)
Oil Palm	3.039.637	Coarse Leaves	0.22	401.232
		Fiber	0.63	1.148.982
		Palm Coir	1.06	1.933.209
Sugarcane	24.811.681	Leaves	3.26	10.110.760
		Bagasse	2.68	8.311.913
Coffee	850.500	Pulp	2.13	1.811.565
		Husk	0.21	178.605
		Stem	3.02	2.568.510
Corn	1.293.975	Leaves and stems	0.93	1.203.396
		Cob	0.27	349.373
		Fibers	0.21	271.734
Rice	2.243.981	Straw	2.35	5.273.355
		Husk	0.2	448.796
Banana	2.026.828	Coir	1	2.026.828
		Stem	5	10.134.140
		Discard fruit	0.15	304.024
		Skin	0.3	608.048
Coconut	129.956	Coir	0.35 [14]	45484.6

Table 1.
Generation of lignocellulosic waste in Colombia.

Polymer	National demand (Ton/year) [12]	Waste mass (Ton/year)
PVC	220.000	159.500
PS	78.000	56.550
LDPE	119.000	86.275
HDPE	160.000	116.000
PP	240.000	174.000
PET	163.000	118.175

Table 2.
Generation of plastic waste in Colombia.

These materials have the potential to be used in different industrial areas, mostly on automotive, industrial, construction, and decoration applications [23]. Due to its renewable nature, research and development of biocomposites have been constantly increasing, and its applications are spreading to multiple areas, being its main attractive is the combination between low price, biodegradability, availability, and their capability to substitute other compounds that use regular reinforcements as glass or carbon fibers [24, 25]. Characteristics as the ones mentioned before contribute to lower the environmental burden of these kinds of materials throughout their life cycle. When compared to traditional plastic products, the substitution of the polymeric material with a natural fiber fraction on the composite can reduce

environmental impacts derived from raw material acquisition, operation life of the product, and end of life processes. Since natural fibers are in most cases residues from agricultural practices, their incorporation on an industrial processes serves as a waste management alternative where the fibers are recycled as reinforcement for plastic materials, helping to minimize environmental burdens from their primary process in agriculture where they are treated as conventional waste. Also, the reincorporation of these residues contributes to assess environmental impacts of raw material transportation at a local scale. Furthermore, this material fraction substitution reduces the amount of plastic material needed to fabricate one product, and as a consequence, less quantity of polymers are demanded for production, and less extraction of fossil resources has to be made in order to supply this productive sector. Regarding processing, biocomposites offer a wide amount of advantages related to processing techniques. These materials not only can reduce the melting temperatures on the process, contributing to lower the embodied energy of the product and consequently the carbon emissions of the product, but also can be processed with existing tools and procedures, which means that the producer does not have to make major adjustments on his production line to work with them.

Nevertheless, not every composite is easy to process, and so traditional material may have a favored position related to biocomposites due to its advanced and well-studied processing techniques, and as direct result, fewer residues can be achieved during the fabrication process. Still some experiences with biocomposites have led to significant reduction of Greenhouse Gasses during processing and transformation stages [26]. In the case of thermoformed trays, it was found that by replacing talc fillers with starch fibers, the carbon footprint for this product was reduced around a 20% regarding gas emissions from processing [27].

Other outstanding characteristic of biocomposites compared to their traditional counterparts is the reduction of weight for the final product. For automotive applications, this characteristic could allow savings of carbon emissions by reducing the total weight of the vehicle and thus consuming less fuel without compromising the integrity of the material properties and the security guidelines of the automotive industry. Materials that possess high specific stiffness and specific strength are often very valuable in applications in which weight will be a critical factor [25], which makes biocomposites ideal candidates for automobile design and spare parts production. Different automakers believe that all advanced composite car body-work could be around 50–67% lighter than current similarly sized steel auto-body, 40–55% lighter than an aluminum auto-body, and 25–30% lighter than a steel auto-body. Nevertheless, there are not bio-based materials on commercial use or development that can be fully considered sustainable [28]. It is a fact that products derived from renewable resources tend to be competitive in the market if they prove to be similar or better than other products regarding performance and price. In fact, Reinders et al. [29] said that full bio-based brands usually have stronger purchase intentions than other brands, including those that are partially composed by bio-based products. However, the fact that a product has a renewable origin does not mean that its environmental performance is better when comparing it to traditional products in the market. A case-based evaluation is necessary to define the environmental aspects of a product and thus the sustainable nature of the product [30]. Virtually, biocomposites can be considered sustainable materials compared to traditional composites or fossil-based polymeric materials. The renewable provenance of these materials and the availability of the resource can suggest better environmental performance among its life cycle, easing pressures over the natural systems. However, critical aspects of the elaboration process during the materials life cycle can lead to different types of environmental impacts, which in turn, may be worse than the ones derived from traditional composite elaboration.

Manufacturing techniques should be strongly studied and refined in order to make them mainstream and reduce concerns and impacts regarding their development degree. Therefore, as mentioned before, this is the main reason why every case of composite material has to be reported on a case-based scenario in order to objectively define the sustainable nature of products developed with these kinds of materials. In this book chapter, biocomposites based on recycled polypropylene (r-PP) and two different natural fibers (coffee husk and coconut coir fibers) with maleated polypropylene (MAPP) as a coupling agent were prepared through extrusion and injection molding processes. Morphological, mechanical, and thermal properties of the biocomposites were investigated with the aim to understand the effect of fiber type and MAPP addition on the r-PP matrix properties. Also, the environmental performance of the materials was studied through a carbon footprint evaluation on a cradle to gate life cycle assessment.

2. Materials and methods

2.1 Materials

Coconut coir (CCF) was obtained from “Kiero Coco” S.A (Manizales-Colombia), and coffee husk fiber (CHF) was obtained from a local coffee mill located in Tuluá-Colombia. Recycled polypropylene (rPP) was a postindustrial waste collected from extrusion and injection processes carried out in the materials laboratory of the Autónoma de Occidente University (Cali, Colombia). Maleic anhydride grafted polypropylene (Licocene MAPP 6452 by Clariant) was used as coupling agent.

2.2 Natural fibers characterization

The time between the generation of the different fiber waste and its storage (at -20°C) was less than 8 hours, in order to minimize biochemical changes in the fibers. After separation, the fibers were dried in an oven at 45°C until reaching constant weight. Drying process was carried out at this temperature in order to avoid the elimination of volatile compounds and degradation of the lignocellulosic composition. After drying, the samples were milled (particle size $<1\text{ mm}$) in an impact mill (Retsch SR200). The milling time was 15 minutes for CHF and 30 minutes for CCF. Finally, fibers samples were stored in polyethylene bags with a hermetic seal at room temperature. After the characterization, the fibers were sieved in ASTM sieves, with the purpose of reaching a 60 mesh particle size, established by the ASTM standards for the analysis of solid samples.

The fibers (CHF and CCF) were characterized by proximate and elemental analysis, calorific power, and structural composition. These analyzes were performed in triplicate. Through the proximate analysis, the percentage of moisture content (M), volatile matter (VM), ash (A), and fixed carbon (FC) was determined according to ASTM D7582-12 [31]. These analyzes were performed using approximately 1.0 g of sample in a Leco brand thermogravimetric analyzer, TGA-601. **Table 3** shows the equations used in the determination of the proximate analysis of CHF and CCF. The calorific value was calculated using 1.0 g of sample in a Leco AC-350 calorimeter pump, following the ASTM 5865-13 standard. The calorific value establishes the amount of energy per unit mass that the waste can deliver when it is completely oxidized. This property was calculated using the equation also presented in **Table 3**.

Parameter	Equation
Moisture content at 105°C	$\%M_{105} = \frac{SW-DSW}{SW} \times 100 \text{ (1)}$
Ash	$\%A = \frac{ACW-CW}{DSW} \times 100 \text{ (2)}$
Volatile matter	$\%VM = \frac{DSW-VSW}{DSW} \times 100 \text{ (3)}$
Fixed carbon	$\%FC = 100 - \%M_{105} - \%A - \%VM \text{ (4)}$
Superior calorific power	$SCP = 20.7999 - 0.3214 \frac{VM}{FC} + 0.0051 \left(\frac{VM}{FC} \right)^2 - 11.2277 \frac{A}{VM} + 4.4953 \left(\frac{A}{VM} \right)^2 - 0.7223 \left(\frac{A}{VM} \right)^3 + 0.0383 \left(\frac{A}{VM} \right)^4 + 0.0076 \frac{FC}{A} \text{ (5)}$

SW: sample weight (gr), DSW: dry sample weight (gr), ACW: crucible weight + ashes (gr), CW: crucible weight, VSW: devolatilized sample weight (gr).

Table 3.
Equations used in the proximate analysis of lignocellulosic residues.

The elemental analysis was carried out in a Leco CHN-628 analyzer to determine the content of carbon (C), hydrogen (H), and nitrogen (N) according to ASTM D 5373-14 and in a Leco S-632 to quantify the sulfur content (S) according to ASTM D 4239-14. A weight sample of 0.1 g was used on both equipments.

The structural composition of the fibers was determined by the quantification of extractive compounds (EXT), lignin (LGN), cellulose (CEL), hemicellulose (HMC), and inorganic compounds (ashes). The preparation of the fibers was carried out following the standard NREL/TP-510-42620. In order to obtain the percentages of CEL, HMC, and LGN, it is necessary to perform two Soxhlet extractions to the fibers using water and ethanol as solvent, as indicated in the NREL/TP-510-42619 standard. The insoluble acid lignin percentage (LGN) or Klason lignin was calculated according to the standard NREL/TP-510-42618. The holocellulose percentage (HCL) was determinate by following the ASTM D1104 standard, while the cellulose percentage was determinate following the Han and Rowell methods [32]. **Table 4** shows the equations used for structural composition calculation of CHF and CCF.

Equation	Reference
$\%EXT_{water} = \frac{PEXT_{water}}{PR \times \%MS} \times 100 \text{ (6)}$	[33]
$\%EXT_{ethanol} = \frac{PEXT_{ethanol}}{PR \times \%MS} \times 100 \text{ (7)}$	
$\%EXT = \%EXT_{water} + \%EXT_{ethanol} \text{ (8)}$	
$\%LGN = \frac{PLGN}{PRIEXT \times \%MS} \times (100 - \%EXT) \text{ (9)}$	
$\%HCL = \frac{PHCL}{PRIEXT \times \%MS} \times (100 - \%EXT) \text{ (10)}$	[32, 33]
$\%CEL = \frac{PCEL}{PHCL} \times \%HCL \text{ (11)}$	[32]
$\%HMC = \%HCL - \%CEL \text{ (12)}$	

Where, % EXT: proportion of total extractives, % EXT_{water}: proportion of extractives in water, % EXT_{ethanol}: proportion of extractives in ethanol, PEXT_{water}: weight of extractives in water (gr), PEXT_{ethanol}: weight of extractives in ethanol (gr), PR: dry sample weight (gr), % MS: percentage of dry matter, % LGN: proportion of lignin, PLGN: weight of lignin (gr), PRIEXT: weight of the sample free of extractives (gr), % HCL: proportion of holocellulose, PHCL: weight of holocellulose (gr), % CEL: percentage of cellulose, PCEL: weight of cellulose (gr), % HMC: proportion of hemicellulose.

Table 4.
Equations used in the determination of the structural composition of lignocellulosic residues.

2.3 Preparation of the biocomposites

r-PP and its biocomposites were compounded in a co-rotating twin screw extruder (Harden Industries Ltd., China). For each case, MAPP, CCF, and CHF fibers were physically premixed with r-PP pellets in a plastic bag using 30% of fibers and 4% of MAPP in weight. A temperature gradient from 140 to 170°C from the feeder zone to the die was used. The rotation speed of the twin-screw was 50 rpm. The outcoming cord of r-PP and its biocomposites from the extruder were pelletized using a mill which produced pellets of about 5 mm long. After the pelletization process, the r-PP and its biocomposites samples were dried in an oven at 85°C followed by an injection molding process. A BOY XS (BOY Machines, Inc., USA) microinjection molding machine was used to prepare samples for flexural and impact tests. **Table 5** summarizes the injection-molding processing parameters used. **Figure 1** shows the injected specimens of neat PP, r-PP, r-PP-CHF, and r-PP-CCF biocomposites.

2.4 Characterization of the biocomposites

2.4.1 Flexural properties

Three point bending flexural tests were performed with an INSTRON universal testing machine model 3366 according to the ASTM D 790-17 as shown in **Figure 2**.

Parameter	Value
Barrel temperature(°C)	185
Nozzle temperature (°C)	180
Injection time (s)	3.8
Cycle time (s)	37.5
Screw travel (mm)	18.7
Back pressure (bar)	60
Injection pressure (bar)	80

Table 5.
Injection molding parameters used.

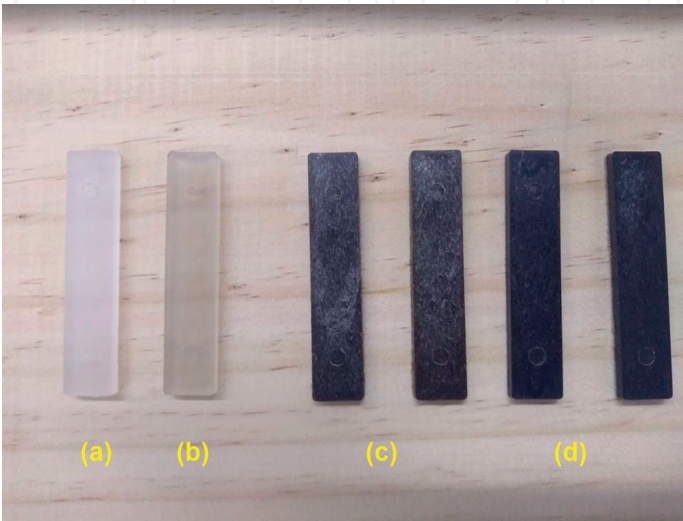


Figure 1.
Injected specimens of: (a) neat PP, (b) r-PP, (c)-PP-CHF and (d) r-PP-CCF biocomposites.



Figure 2.
Assembly used for flexural test (according to ASTM D790).

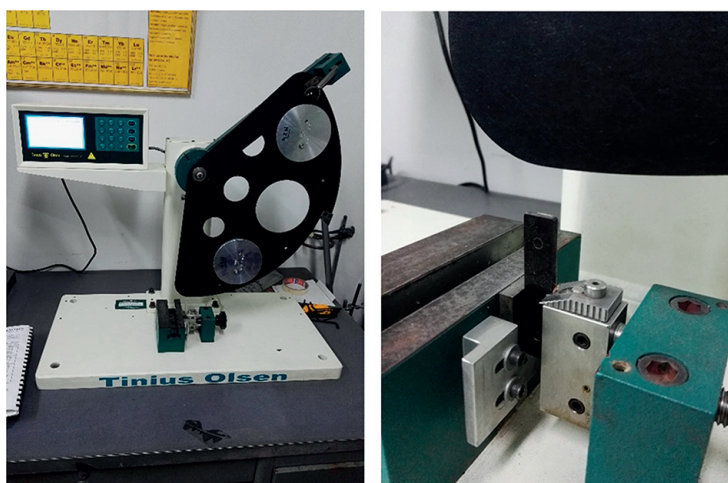


Figure 3.
Pendulum type impact test machine used for the notched IZOD impact measurements.

The tests were carried out on bars of rectangular cross section at 23°C and at a rate of crosshead motion between 1.34 and 1.44 mm/min. This rate was determined based on the dimensions of the specimens. Also, the distance between the supports was 50 mm and the tests were conducted up to 5% strain. All the results were taken as the average value of five samples.

2.4.2 Impact properties

The impact strength of PP and biocomposites was determined with an Izod Tinius Olsen impact pendulum equipped with a 4.53 N pendulum. Prior to the test, the materials were subjected to conditioning for 48 hours at 50% relative humidity and a temperature of 25°C. The specimens were made following the standard ASTM D256, and the starting angle of the test was 150° as shown in **Figure 3**. All the results were taken as the average value of five samples.

2.4.3 Thermal characterization

DSC test was carried out using a TA Q2000 differential scanning calorimeter under nitrogen atmosphere at a scanning rate of 10°C/min, with a sample of 10 mg in aluminum pans. The thermal history of the samples was erased by a preliminary heating cycle at 10°C/min from 20 to 200°C and maintaining it at that temperature for 10 min to melting residual crystals, cooling at 10°C/min to 0°C, and finally, they

were heated at °C/min from 0 to 200°C. The crystallization temperatures (Tc) and melting temperatures (Tm) were determined from cooling and second heating scans. The melting enthalpies values were normalized according to the proportion of the components in the samples. The crystallinity (χ_c) was determined from the Eq. (13):

$$\chi_c = \left(\frac{\Delta H_m}{[\Delta H_m^0 * (1-w_{Fiber})]} \right) * 100 \tag{13}$$

where w_{Fiber} is the CHF or CCF fiber mass fraction, ΔH is the melting enthalpy of the sample, and ΔH_m^0 is the specific enthalpy of melting for 100% crystalline PP. This value was reported in literature as 293 J/g [34].

2.4.4 Morphology

Scanning electronic microscopy (SEM) of the biocomposites was carried out on the cryogenic fracture surfaces of the specimens using a Quanta FEG 250 microscope operating at a voltage of 10 kV. The samples were previously sputter coated with gold to increase their electric conductivity. Determinations were performed in different areas of the SEM micrograph.

2.4.5 Statistical analysis

Flexural and impact properties of the materials were subjected to analysis of variance (ANOVA), and the Tukey’s test was applied at the 0.05 level of significance. All statistical analyses were performed using Minitab Statistical Software Release 12 (Pennsylvania, United States).

2.5 Environmental characterization of the materials

The environmental characterization of the materials was made through a cradle to gate Life Cycle Assessment based on the ISO 14041 parameters. The only indicator used for this evaluation was the carbon footprint, and the information for the emission factors was taken from secondary information found in literature.

3. Results and discussion

3.1 Natural fibers characterization

The proximate analysis of CCF and CHF is presented in **Table 6**. Comparing the obtained values, it is observed that the fiber with the highest moisture content is the

Natural fiber	%M ₁₀₅	%A	%VM	%FC	SCP (MJ/Kg)	References
CHF	11.61 ± 0.08	1.65 ± 0.05	83.08 ± 0.08	3.66	15.93	Obtained values
	9.22 – 25.3	1.71 – 2.5	68.2 – 81.87	16.42 – 18.5	19.8 – 21.41	[35–37]
CCF	12.15 ± 0.11	3.57 ± 0.08	73.07 ± 0.04	11.21	18.41	Obtained values
	9.65 – 10.1	3.2 – 5.56	69.35 – 75.5	11.2 – 15.44	14.67 -18.74	[38]

Table 6.
Proximate analysis of the natural fibers.

CCF. This parameter is directly related to the dispersion of the fibers in a polymer matrix during the melting processing [39]. Higher moisture content causes lower dispersion of a lignocellulosic material in a polymer matrix. This effect can be related with the final properties of the obtained biocomposites. Also, CHF presented the highest volatile matter value. Volatile matter is related to the cellulose and hemicellulose percentage in the fibers. It is reported that hemicellulose influences the distance of interfibrillary cellulose, impacting fiber stiffness [40]. For that reason, a higher content of volatile matter represents a significant proportion of holocellulose in the fiber and represents higher toughness and a better fatigue behavior in the fiber [40]. Therefore, better results can be expected in the mechanical properties of a biocomposite obtained from CHF compared to a CCF-based biocomposite. The results show that the moisture content value in CCF is higher than the data presented by several authors [38, 41–43]. Also, CHF volatile matter content is higher in comparison with the values reported in literature [35–37]. On the other hand, ash, fixed carbon percentages, and calorific value lower of CHF are lower than the values reported in literature [35–37, 41, 43–45].

The elemental analysis values of the fibers are presented in **Table 7**. The results show that the carbon content in the CCF (53.88%) was higher in comparison with the reported values in literature, while the oxygen content was lower. On the other hand, CHF elemental analysis values are within the ranges established by other authors. Atomic ratios O/C and H/C obtained for CHF and CCF were 0.61, 0.54, 1.63, and 1.45, respectively. These results are in accordance with the values of biomass established in the Van Krevelen diagram [31]. The values of the O/C ratio obtained can be attributed to a high content of cellulose and hemicellulose in the biomass [3, 47]. This O/C relationship can be used as a parameter to evaluate the polarity of fibers in the production of biocomposites materials, being related to the content of hydroxyl groups. These groups are reactive centers of high polarity, which influence the formation of hydrogen bonds and the compatibility between fibers and a polymer [48]. For this reason, the O/C ratio allows to estimate the interaction degree between the lignocellulosic reinforcement and the polar polymeric matrix used [3], and its value is higher for CHF compared to CCF.

The structural compositions of the fibers are presented in **Table 8**. CHF presented a higher cellulose content (33.38%) compared to CCF (22.24%). Cellulose is considered as a semicrystalline biopolymer with a fibrous and rigid structure, which positively affects the stiffness in biocomposites materials [52]. Also, a greater amount of cellulose is related to better compatibility between the fibers and the polymeric matrix and a better mechanical performance of the biocomposite [53]. Regarding the lignin content, CCF shows a greater quantity (25.42%) compared to CHF (17.31%). This type of compound is considered as an amorphous polymer with chemical heterogeneity and a low physical consistency. Bajwa et al. [48] mentioned that increase in the lignin content decreases the mechanical resistance of thermoset biocomposites.

Natural Fiber	C (%)	H (%)	N (%)	O (%)	S (%)	Reference
CHF	50.72	6.88	1.07	41.27	0.06	Obtained values
	40.1–52.56	4.9–7.08	0–5.2	39.54–49.1	0–0.35	[35–37, 46]
CCF	53.88	6.51	0.68	38.84	0.08	Obtained values
	47.25–48.58	5.7–6.74	0–3.04	43.74–45.6	0	[38, 46]

Table 7.
Elemental analysis of the natural fibers.

Natural Fiber	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Ashes (%)	Extractives (%)	References
CHF	33.38 ± 0.89	13.06 ± 0.60	17.31 ± 0.68	1.84 ± 0.06	34.42 ± 0.94	Obtained values
	29.17 – 35.4	18.2 – 28.96	22.35 – 23.2	1.4 – 4.6	17.67 – 21.8	[49, 50]
CCF	22.24 ± 1.46	15.62 ± 1.25	25.42 ± 0.81	3.71 ± 0.08	33.00 ± 0.28	Obtained values
	30.22 – 47.7	21.9 – 25.9	17.8 – 39.66	0.8 – 5.56	6.8 – 18.66	[8, 38, 51]

Table 8.
Structural analysis of the natural fibers.

3.2 Characterization of the biocomposites

3.2.1 Mechanical properties

The influence of CHF, CCF fibers, and MAPP addition on the r-PP flexural and impact properties was evaluated. The tensile behavior of the materials is shown in **Figure 4**. **Table 9** presents flexural modulus, flexural strength, and impact strength values of the materials.

The results show that CHF and CCF fibers incorporation induce a significant improvement of flexural properties of r-PP. r-PP-CHF and r-PP-CCF biocomposites flexural modulus (FM) increased 97 and 13%, respectively, in comparison with r-PP. Although the FM values were improved in both biocomposites, the effect was sharper for r-PP-CHF. This can be explained with the structural analysis of the fibers (Section 3.1). The cellulose content is related to the oxygen proportion and is associated with the resistance degree of the fiber. In this sense, lignocellulosic materials with a higher oxygen content or a higher value in the O/C ratio will have a better mechanical performance [54–57]. CHF presented a higher cellulose content

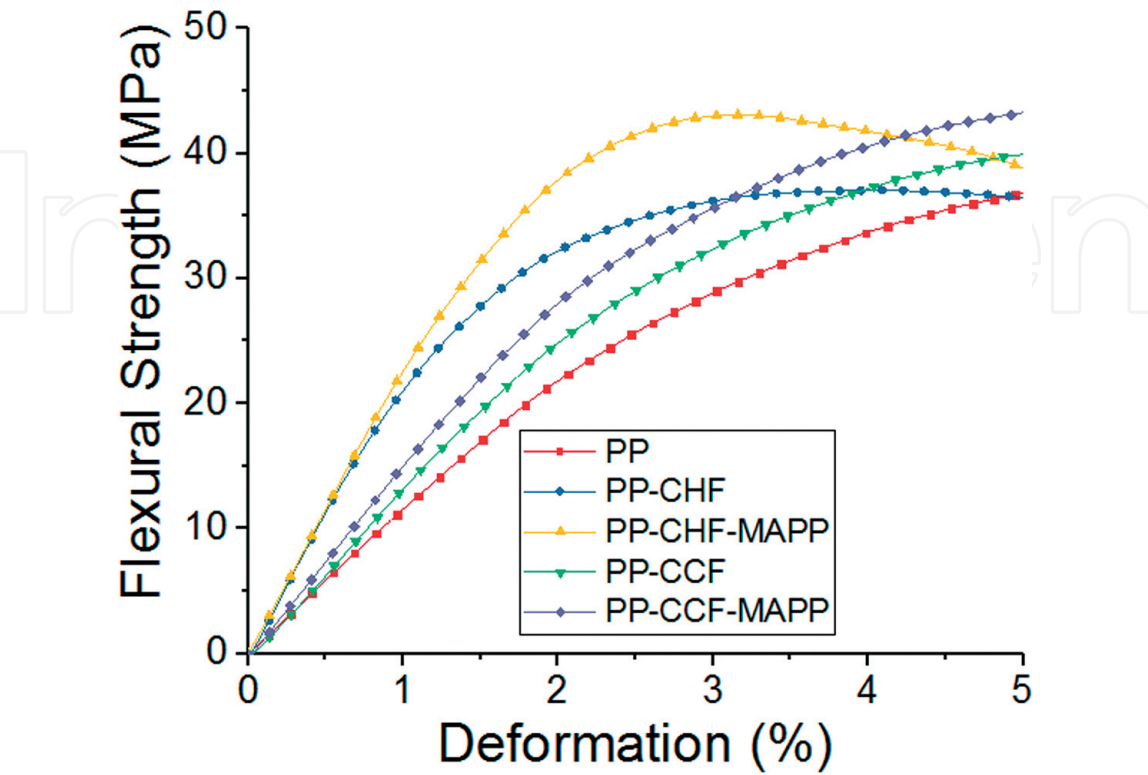


Figure 4.
Average flexural stress vs. deformation of r-PP and r-PP biocomposites.

Sample	Flexural and impact properties [*]		
	Flexural properties		Impact properties
	Modulus (MPa)	Strength (MPa)	Impact strength (kJ/m ²)
r-PP	1193 ± 50 ^a	37.1 ± 1.7 ^a	11.5 ± 1.8 ^a
r-PP-CHF	2350 ± 71 ^b	37.2 ± 0.3 ^a	7.3 ± 0.4 ^b
r-PP-CHF-MAPP	2309 ± 114 ^b	43.1 ± 0.9 ^b	15.5 ± 1.3 ^c
r-PP-CCF	1349 ± 10 ^c	40.1 ± 0.7 ^c	10.7 ± 1.2 ^a
r-PP-CCF-MAPP	1351 ± 12 ^c	43.3 ± 0.4 ^b	16.5 ± 0.7 ^c
<i>a–c Different letters in the same column indicate significant differences (p < 0.05).</i>			
<i>[*]Mean of five replications ± standard deviation.</i>			

Table 9.
Flexural and impact properties of r-PP and r-PP-natural fiber biocomposites.

in comparison with CCF (33.38 and 22.24%, respectively), which could explain the greater improvement in FM values with this fiber. It was also observed that MAPP addition did not generate significant differences ($p \geq 0.05$) on the FM values compared to r-PP-Fiber biocomposites.

On the other hand, CHF and CCF fibers addition generate slight improvements (1 and 8%) on the flexural strength (FS) compared to neat r-PP. These results agree with previous studies found in literature [58–60]. However, for both r-PP-fiber biocomposites, MAPP addition causes an increase in FS of 16% in comparison with r-PP.

Impact test results shows that CHF and CCF addition cause a decrease on the impact strength of 37 and 6%, respectively, in comparison with the r-PP. Similar results were reported by several studies about the morphology and mechanical properties of PP-natural fiber biocomposites [60–63]. However, for r-PP-CHF and r-PP-CCF, an increase on the impact strength of 35 and 44% was observed. This result shows that MAPP addition increases the capacity of r-PP to absorb energy. This phenomenon can be explained by a possible energy absorption promoted by fracture mechanisms, which involve detachment, slippage, and fragmentation of the fiber. Mechanisms are not present on the r-PP and r-PP biocomposites without MAPP.

The improvements in FS and impact strength with MAPP can be explained with the improved interfacial adhesion that MAPP caused. MAPP addition influences the chemical interaction between the hydrophobic matrix and the hydrophilic fiber through the formation of covalent bonds between the maleic anhydride groups and the hydroxyl groups present on the surface of the cellulosic fiber [57, 60]. In addition, Migneault et al. [55] mentioned that esterification reactions produced by the interaction between the natural fiber and the compatibility agent increase with the oxygen content of the fiber. Oxygen is directly related to the proportion of carbohydrates present in the surface of the fiber, creating a greater number of polar sites (hydroxyl groups) to react.

3.2.2 Thermal characterization

3.2.2.1 Differential scanning calorimetry (DSC)

DSC curves for r-PP and their biocomposites with CHF and CCF are shown in **Figure 5**. Numerical values of the thermal events are shown in **Table 10**.

The DSC cooling curve of r-PP (**Figure 5a**) shows a main exothermic peak located around 116°C corresponding to the crystallization of PP chains. When CHF, CCF, and MAPP were added into r-PP, a 3–6°C shift in the crystallization

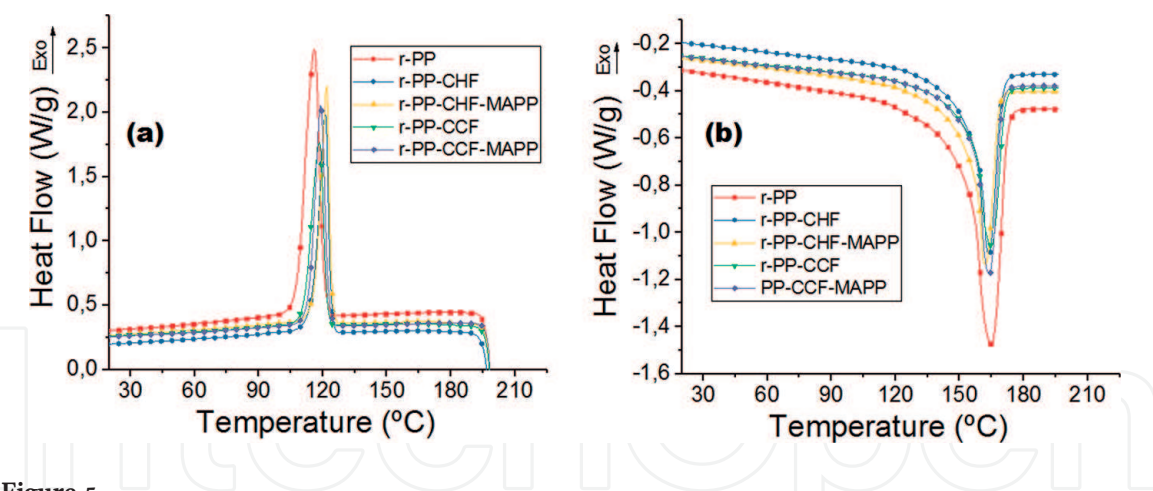


Figure 5.
(a) Cooling and (b) second heating DSC curves for r-PP and r-PP biocomposites.

Sample	Cooling		Second heating	
	Tc* (°C)	Tm* (°C)	ΔHm (J/g)	χ (%)
r-PP	116	165	82	40
r-PP-CHF	121	164	64	44
r-PP-CHF-MAPP	122	162	63	44
r-PP-CCF	118	165	61	42
r-PP-CCF-MAPP	119	164	63	44

*Tc and Tm were taken at the maximum peak of crystallization and melting peaks.

Table 10.
Thermal properties on cooling and second heating DSC scans of the samples.

temperature of r-PP was observed. This decrease indicates that natural fibers in biocomposites can act as a nucleation agent. The second heating runs of r-PP and r-PP biocomposites were shown in **Figure 5b**. All samples exhibit an endothermic peak between 162 and 165°C corresponding to the melting of the PP matrix. These results indicate that the addition of the CHF and CCF fibers does not disturb the melting processes of the PP matrix. Also, it is observed that PP crystallinity fraction melted during heating was 40%. For r-PP biocomposites, the crystalline phase content increases slightly up to 44%. These results show that CHF and CCF fibers promote the formation of crystalline phases in the r-PP present in the biocomposites. Some reports that were related to fiber-reinforced composites have found that the fibers act as nucleation points that increase the crystallinity of the polymer phase [64].

3.2.2.2 Thermogravimetric analysis (TGA)

TG and DTG curves were used to determine the thermal stability of coffee husk (CHF) and coir coconut fibers (CCF). The results are shown in **Figure 6**. Also, main thermal parameters obtained from these curves are summarized in **Table 11**.

As shown in TG curve (**Figure 6a**), fibers present three weight loss regions which are located around 60–100°C, 240–350°C, and 350–600°C. The first weight loss region below 100°C can be attributed to the evaporation of superficial water present in the sample, while the other regions might be associated with the decomposition of the fiber constituents. DTG curves (**Figure 6b**) show a first decomposition peaks at 299 and 284°C for CHF and CCF, respectively. These peaks correspond

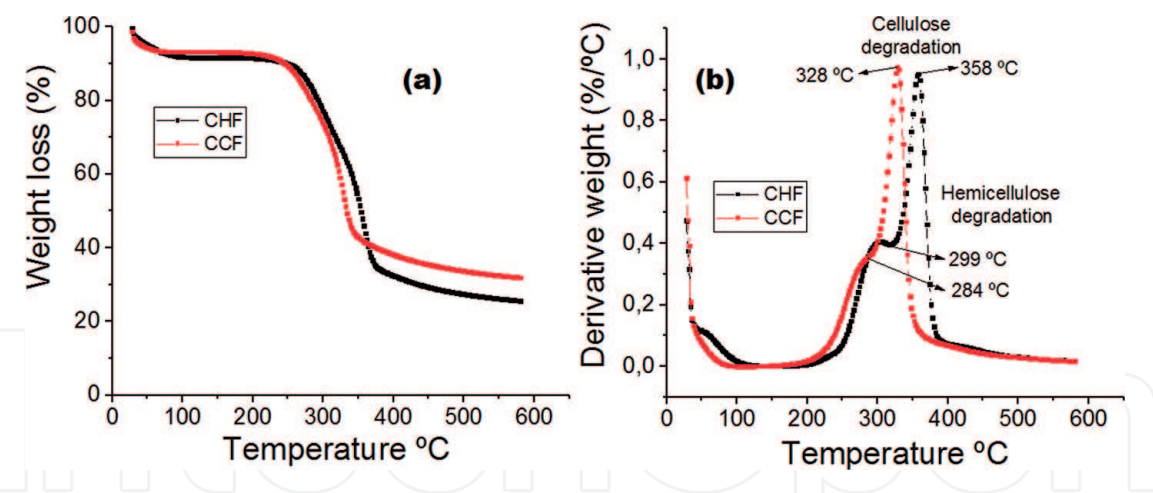


Figure 6.
(a) TG and (b) DTG curves of CHF and CCF fibers at heating rates of 10°C/min.

Sample	Degradation stage	To (°C)	Tmax (°C)	Residual Char (%)
Coffee husk fiber (CHF)	1	266	299	25
	2	336	358	
Coir coconut fiber (CCF)	1	245	284	31
	2	314	328	

Table 11.
Thermal degradation data of the fibers at 10°C/min in nitrogen atmosphere.

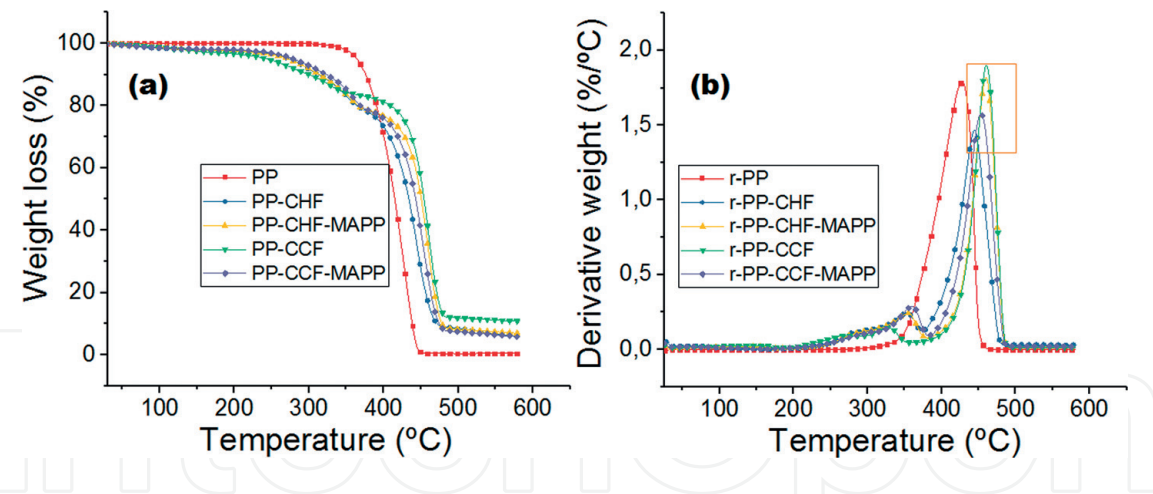


Figure 7.
(a) TG and (b) DTG curves of r-PP and r-PP biocomposites at heating rates of 10°C/min.

to the temperature of maximum weight loss rate (Tmax) of hemicellulose, while the second peaks located at 358°C and 328°C (for CHF and CCF) are related to the Tmax of α -cellulose. The residual weights of CHF and CCF have also been measured and are equal to 25 and 31% for CHF and CCF at 600°C. This results show that CHF possesses a better thermal stability compared to CCF.

TG and DTG curves for r-PP and r-PP biocomposites are shown in **Figure 7**. Also, main thermal parameters obtained from these curves are summarized in **Table 12**.

Recycled PP degradation occurs in a single step process with an onset temperature (To) located at 368°C and a Tmax of 428°C. The residue after final degradation was 0.4%. Regarding biocomposites, TG and DTG show that the addition of coffee husk and coir coconut fibers produces an increase in the thermal stability of the

Sample	Degradation stage	Tonset (°C)	Tmax (°C)	Residual Char (%)
r-PP	1	368	428	0.4
r-PP-CHF	1	260	354	8.4
	2	402	445	
r-PP-CHF-MAPP	1	264	355	8.2
	2	428	460	
r-PP-CCF	1	243	327	11.9
	2	427	460	
r-PP-CCF-MAPP	1	259	360	7.4
	2	411	453	

Table 12.
Thermal degradation data of r-PP and r-PP biocomposites.

r-PP phase. As shown in **Table 12**, T_o increases between 34 and 60°C. Also, T_{max} increases between 33 and 28°C in comparison to r-PP (as indicates in the orange area). This increment in the thermal stability of the biocomposites has been previously observed in different studies, indicating that the incorporation of fibers in the material induces spherulite nucleation points, increasing the crystallinity of the polymer and improving its thermal properties [64].

3.2.3 Morphology

Figure 8a and **b** shows SEM images of the fractured surfaces of r-PP-CHF and r-PP-CCF, respectively. In these images, gaps between the fibers and the surrounding r-PP matrix can be clearly observed, which indicate a poor interfacial adhesion between the r-PP matrix and the natural fibers [65]. For **Figure 9a** and **b**, with the MAPP addition, gaps between natural fibers and r-PP were significantly reduced, and as a consequence, an improvement over the interface for the composite can be appreciated. This result confirms that MAPP addition improved the interfacial property of the hydrophobic PP matrix and the hydrophilic natural fibers. Also, this can be related with mechanical properties enhancement observed in the biocomposites after the MAPP addition.

3.2.4 Environmental characterization of the materials

When assessing the carbon footprint of the material, the stages of the life cycle were limited to raw material acquisition, transport, and processing. Since the

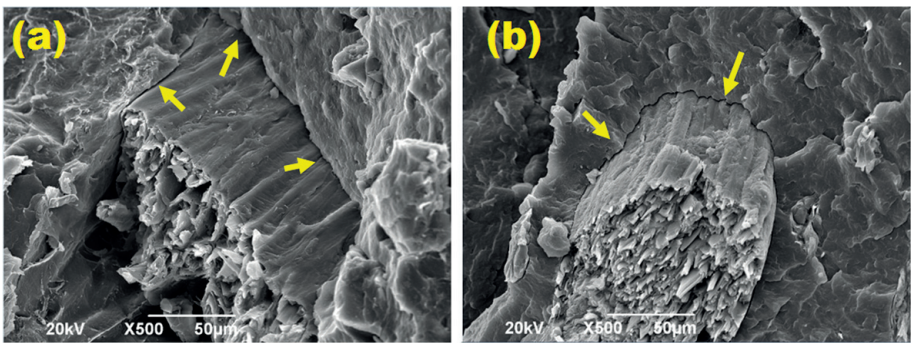


Figure 8.
SEM pictures for rPP-CHF and rPP-CCF biocomposites.

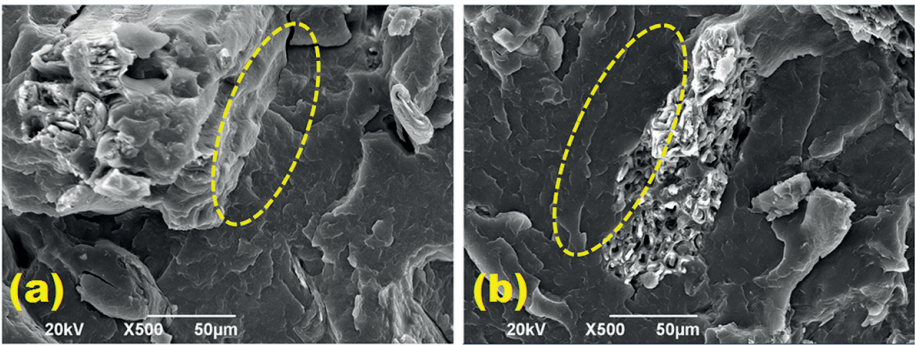


Figure 9.
SEM pictures for rPP-CHF-MAPP and rPP-CCF-MAPP biocomposites.

material was not transformed into a product, the functional unit was determined as the 1 kg of manufactured material and the carbon footprint was determined on a cradle to gate life cycle. The comparison was made between the biocomposite materials, recycled polypropylene, and neat polypropylene. Neat polypropylene was brought from Medellín 412 km away from the final user’s location. Coffee husk was delivered from Tuluá (104 km) and coconut coir from Manizales (270 km). All the materials were transported on a diesel-powered truck to the final user location. Fibers were blended with the rPP matrix through an extrusion process and subsequently pelletized with a 1.5 kW mill.

Emissions were determined through emission factors for every activity involved on the elaboration of the material using the Eq. (14). For each emission factor, there is one activity related in order to calculate the emissions for the product elaboration. The results of those emissions are listed for each biocomposite on **Table 13**.

$$Emission = Emission\ factor * activity \tag{14}$$

From **Figure 10** emission accounting for each category, it could be noted that transport is the largest contributor to the overall emissions. Emissions on this stage are directly proportional to the amount of fuel used to transport the materials, and so, the further away the place, the greater the associated emissions. In order to achieve a more sustainable product, materials should be taken from regional suppliers to lower the footprint from transporting activities. Since rPP is produced inside the processing site, there is no emission associated to the transport of this material as stated on the life cycle boundaries for this particular case. Regarding raw material acquisition, the incorporation of natural fiber seems to improve the impact of the material compared to neat PP and recycled PP. For the coffee husk composite case, the carbon footprint on this stage for the 10 kg of material corresponds to 4.84 kg CO₂ eq, and for coconut coir biocomposite, it corresponds to 4.29 kg CO₂ eq. When compared to neat PP, a reduction in terms of carbon emissions of 76.93 and 73.78%, respectively, for rPP-CFH and rPP-CCH biocomposites. Also when compared to recycled polypropylene, it could be noted that emissions of rPP-CHF composite were slightly lower, reducing emissions on a 1.98%. Nevertheless, for rPP-CCF composite, the emissions raised on this stage on an 11.26% compared to rPP. In order to elaborate the composite, the fibers had to be blended with the recycled material, and so an extra process is needed as mentioned before. This extrusion and grinding process generates emissions of 4.54 kg CO₂ eq for both biocomposites, adding emissions to the overall score. For this scenario, the carbon footprint for the finished materials is shown on **Table 14**. This table shows the carbon footprint in terms of a functional unit defined as 1 kg of processed material.

rPP-CHF Biocomposite					
Stage	Material	Sub-stage	Emission factor	Activity	Emission
Raw material	Coffee husk	Production	0.52 kg CO ₂ /kg [66]	3 kg	1,56 kg CO ₂
		Grinding	0.374 kgCO ₂ /kWh [67]	0.55 kWh	0.21 kg CO ₂
		Transport	2.61 kgCO ₂ /L [68]	36.4 L	95.01 kg CO ₂
	Recycled PP	Process	0.38 kg CO ₂ /kg [69]	7 kg	2.66 kg CO ₂
		Grinding	0.374 KgCO ₂ /kWh [67]	1125 kWh	0.42 kg CO ₂
		Processing	Biocomposite	Extrusion	0.374 KgCO ₂ /kWh [67]
Grinding	0.374 KgCO ₂ /kWh [67]			1.5 kWh	0.56 kg CO ₂
				Total	104.4 kg CO ₂
rPP-CCF Biocomposite					
Stage	Material	Sub-stage	Emission factor	Activity	Emission
Raw material	Coconut coir	Production	0.27 Kg CO ₂ /kg [70]	3 kg	0.81 kg CO ₂
		Grinding	0.374 kg CO ₂ /kWh [67]	1.1 kWh	0.41 kg CO ₂
		Transport	2.61 kg CO ₂ /L [68]	94.7 L	246.65 kg CO ₂
	Recycled PP	Process	0.38 kg CO ₂ /kg [69]	7 kg	2.66 kg CO ₂
		Grinding	0.374 kg CO ₂ /kWh [67]	1125 kWh	0.42 kg CO ₂
		Processing	Biocomposite	Extrusion	0.374 kg CO ₂ /kWh [67]
Grinding	0.374 kg CO ₂ /kWh [67]			1.5 kWh	0.56 kg CO ₂
				Total	255.49 kg CO ₂
Recycled PP					
Stage	Material	Sub-stage	Emission factor	Activity	Emission
Raw material	Recycled PP	Process	0.38 kg CO ₂ /kg [68]	10 kg	3.80 kg CO ₂
		Grinding	0.374 kg CO ₂ /kWh [67]	1.5 kWh	0.56 kg CO ₂
				Total	4.36 kg CO ₂
Neat PP					
Stage	Material	Sub-stage	Emission factor	Activity	Emission
Raw material	Neat PP	Process	1.86 kg CO ₂ /kg [71]	10 kg	18.60 kg CO ₂
		Transport	2.61 kg CO ₂ /L [68]	146.95 L	382.76 kg CO ₂
				Total	401.36 kg CO ₂

Table 13.
Results of the emissions for the compared materials.

3.3 Conclusions

Biocomposites based on recycled PP (r-PP) and two different natural fibers (coffee husk-CHF and coconut coir-CCF fibers) were prepared by a melt extrusion and injection processes. Proximate, elemental, and structural analysis performed to the natural fibers show that CHF contains a higher cellulose percentage and a higher ratio O/C in comparison to CCF. This condition makes CHF more attractive for biocomposites production. The effects of natural fibers and MAPP addition on the properties of the biocomposites were explored. Flexural characterization showed that MAPP incorporation induces a significant improvement of flexural properties

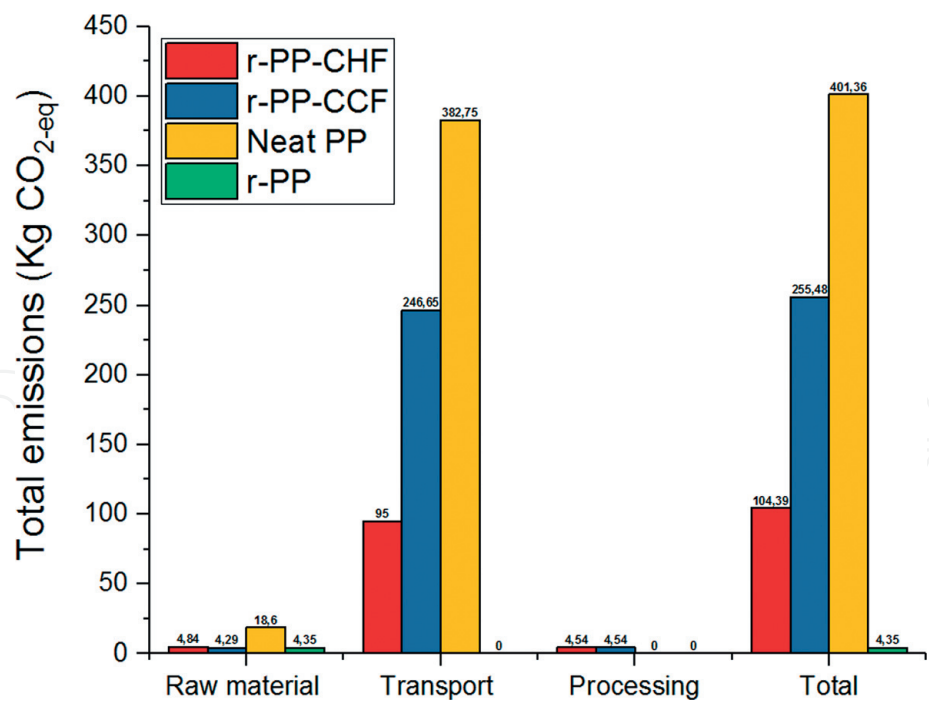


Figure 10.
Carbon emissions comparison for neat PP, r-PP, and r-PP biocomposites.

Material	Carbon footprint (kg CO ₂ eq/kg)
Neat Polypropylene	40.14
rPP-CHF	10.44
rPP-CCF	25.55
Recycled polypropylene	0.44

Table 14.
Carbon footprint of the compared materials.

of r-PP biocomposites. Also, the impact tests showed that the addition of MAPP increases the capacity of r-PP biocomposites to absorb energy. Thermal studies show that CHF and CCF fibers addition did not disturb the melting process and improves the thermal stability of the PP matrix. Despite that for this case scenario, the values of the carbon footprint for both biocomposites is considerably high compared to the recycled polypropylene, it is important to keep in mind that the evaluation was made on a cradle to gate analysis, this means that the benefits of the mechanical and thermal enhancements are not taken into account on this evaluation among the use and operational phases. Depending on the application, the use of these biocomposites has the potential to reduce the carbon footprint over the lifespan of a product made with it, saving emissions derived from usage and disposal. Regarding the performance of both biocomposites, it can be noted that rPP-CHF has a better operation regarding environmental issues due to the saved emissions from raw material transportation. This means that in order to elaborate more sustainable biocomposites, raw material should be delivered on a local extent. Also in order to lower the environmental impacts of the material, the fiber fraction is an important issue due to the replacement of polymer fraction over the composite and thus saving emissions from polymer primary elaboration process.

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Conflict of interest

The authors of this manuscript declare that do not hold any conflicts of interest that might have any bearing on research reported in their submitted manuscript.

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