Polyethylene-coffee husk eco-composites for production of value-added consumer products

Leyla Y. Jaramillo[1], Mauricio Vásquez-Rendón[2], Sergio Upegui[1], Juan C. Posada[2] and Manuel Romero-Sáez[2]*

Abstract

The annual worldwide production of coffee exceeds 10 million tons, and more than 90% of this production is waste, including the husk. On the other hand, plastic consumption increases every year, and sustainable alternatives are necessary to decrease it. This work arises to solve these two problems, and seeks to produce products at an industrial level from polyethylene/coffee husk eco-composites. Both Low Density Polyethylene and High Density Polyethylene were used, and the amounts of coffee husk added as filler were 20 and 40 wt%. The composites were characterized by different morphological, thermal and mechanical techniques. Scanning Electron Microscopy images showed husk particles embedded in the polymer matrix, but with some gaps between the polymer and the filler, because no compatibilizer agent was used. The addition of large amounts of natural filler negatively affected the tensile strength and elongation at break, but increases eco-composites crystallinity, and hence, their Young modulus and hardness. The industrial applicability of the eco-composites was verified through the production of five different consumer products by extrusion and injection processes, using mixtures with 40 wt% coffee husks. All products were obtained without significant defects. If only 3.25 wt% of the polyethylene products produced each year in Colombia did so with the eco-composites developed in this work, all the coffee husk produced in the country would be used, and the emission of about 5.390 million m^3 of greenhouse gases would be avoided.

Keywords: Waste valorization, Eco-composite, Natural fibers, Value–added products, Coffee husk, Polyethylene

Introduction

Coffee is one of the most important agricultural commodities worldwide, and the most consumed beverage in the world after water. It is cultivated in around 80 countries, with a global production of ~ 10.2 million ton in 2019 [1]. It is estimated that around 25 million families worldwide produce coffee, and most of them are small producers whose incomes depend heavily on its harvests [2]. Approximately 70% of coffee supply comes from four main countries: Brazil, Vietnam, Colombia, and Indonesia. In the particular case of Colombia, in 2019, 820,600 tons of coffee were produced, cultivated on approximately 839,700 ha [3].

Coffee consumption increases every year, which means that the residues and by-products associated to its production also do so. From all coffee production, roasted bean, that is ground to prepare the drink, accounts only 9.5 wt% of the total fruit. The remaining 90.5 wt% are different wastes, such as parchment, pericarp, pulp, mucilage and husk. These can be used for different applications, such us ethanol production, animal feed, manure production, polyphenols extraction, adsorption of heavy metals and biochar production [4, 5]. However, nowadays most coffee wastes are not used, but dumped into land or water, causing contamination problems and degradation of the ecosystem.

* Correspondence: manuelromero@itm.edu.co
[2]Quality, Metrology and Production Research Group, Instituto Tecnológico Metropolitano, Medellin 050036, Colombia
Full list of author information is available at the end of the article

On the other hand, the need to produce new materials that are non-toxic and eco-friendly, with specific characteristics for specific purposes, is an increasing challenge for all industries, due to rise of environmental pollution and the lack of resources [6]. Polymer industry, for instance, faces a great challenge in this matter, as its production increases year after year. According to Plastics Europe, the world plastic production in 2018 was 359 million tons, and it grows by around 4% every year [7]. Along with this, come several environmental concerns related to the final disposal of millions of tons of post-consumer plastic product wastes every year [8].

One strategy to decrease the use of polymers, without compromising their properties, is the use of fillers. These are materials, commonly fibers that are incorporated into the formulation of polymers with the aim of improving some of their properties, or merely for saving raw materials. Traditionally, carbon and glass fibers have been the most widely used fillers. Over the past decade, some natural fibers are gaining interest as substitutes for synthetic ones. In polymers reinforcement technology, some of the most used natural fibers are wood, bamboo, oil palm, cotton, wheat straw, rice husk, date palm, peanut shell and bagasse, to name a few [9, 10]. In all these works the use of polymer-fiber compatibilizing agents to improve the interaction with the polymer matrix is reported. Regarding coffee husk (CH), there are few works that use it as a filler in polymeric matrix, and all of them do so with a previous husk surface treatment or with the use of a compatibilizing agent [11, 12].

The use of natural fibers in thermosets or thermoplastics composites has some advantages like ease of availability, reduction of materials density and lowering processing costs [13]. Natural fiber composites are used in different fields, including applications in automotive industry, building and construction industry, aerospace, sports and others, such as panels, frames, windows, decking and bicycle frames [14].

In the present work, polyethylene eco-composites filled with 20 and 40 wt% CH were produced, without any husk surface modification or the use of compatibilizing agent. CH, polymers and composites were characterized by different thermal, morphological and mechanical techniques. In order to verify the eco-composites processability and applicability, five commonly used products were obtained with the higher waste content at an industrial scale using extrusion and injection molding processes. Additionally, possible environmental impacts of eco-composites were analyzed considering the life cycle of similar products and the projection of greenhouse gases emissions.

Materials and methods
Materials
High-density polyethylene (HDPE, SGF4950 extrusion grade) and low-density polyethylene (LDPE, PB208 injection grade) were purchased from Braskem company (São Paulo, Brazil). CH were obtained from coffee plantations of Andes, Antioquia, Colombia. In Table 1 are listed the designations used in this work and some useful materials properties for composites processing and formulation, such as density and melt flow index (MFI).

Preparation and characterization of CH
The main metal elements content presented in the CH was determined by atomic absorption in a Perkin Elmer Aanalyst 800 equipment. Thermal stability was evaluated by thermogravimetric analysis (TGA), using an SDT-Q600, TA instruments equipment. TGA tests were carried out at a heating rate of $10\,°C\,min^{-1}$ within a temperature range between 25 and $900\,°C$, and under a synthetic air atmosphere ($100\,cm^3\,min^{-1}$). The size and shape of CH particles was evaluated by scanning electron microscopy (SEM), using a JEOL JSM-7100F electron microscope. Before the analyses, and in order to improve the electrical conductivity, the samples were coated with gold in a sputter coating system (Q300TD, Quorum). Bulk density of CH was determined according to the ASTM E873–82 standard.

Prior incorporation into the polymer matrices, CH was dried at $80\,°C$ for 4 h, blade-milled and sieved to obtain a particle size ≤4 mm. The particle size distribution was carried out by dry sieving through a series of sieves with decreasing mesh size (5, 8, 10, 14, 20, 25, 30, 40, 45, 50 and 60 mesh).

Composites processing
Composites between PE-E and CH, and PE-I and CH were prepared in a Haake Polylab QC internal mixer equipped with roller-type rotors. A processing temperature of $190\,°C$, a rotation speed of 60 rpm, and a total mixing time of 6 min were used. The mass of polymer and CH were obtained by using Eq. (1):

$$m = \rho_C V f \qquad (1)$$

where ρ_C is the density of the composite calculated from polymer and CH densities and their weight fraction for

Table 1 Raw materials characteristics and designation

Material	Designation	MFI (g/10 min)[a]	Density (g cm^{-3})
Coffee husk	CH	N/A	0.429
HDPE	PE-E	0.34[b]	0.956[a]
LDPE	PE-I	22[b]	0.923[a]

[a]Data obtained from supplier's data sheet
[b]Obtained at 190 °C and 2.16 kg

each composition, V is the free volume inside the mixing chamber ($V = 78$ cm^3 for roller type rotors), and f is the filling factor ($f = 0.7$).

The amount of CH used in the composites production, as well as the nomenclature of each one, is shown in Table 2.

Polymers and composites characterization
Differential scanning calorimetry
Thermal behavior of polymers and composites was analyzed using a differential scanning calorimeter DSC Q600TD from TA Instruments. All tests were made according to ASTM 3417–99 and ASTM 3418–99 standards, at a heating rate of 20 °C min^{-1} within a temperature range between 40 and 250 °C and under a nitrogen atmosphere. Composites crystallinity percentage (X_c) was calculated according to Eq. (2) [15].

$$X_c = \frac{\Delta H_m}{\Delta H_0 w_x} *100 \qquad (2)$$

where ΔH_m is the melting enthalpy of the composite calculated from the melting peak in the thermograms, in J g^{-1}, ΔH_0 is the enthalpy of fully crystalline polymer, in J g^{-1}, and w_x is the weight fraction of polymer in the composites, expressed without units.

Mechanical properties
Tensile tests were performed to dog bone shape specimens according to the ASTM D638 standard. Samples were mechanized from compressed molded sheets obtained at a constant temperature of 190 °C, a pressure of 689 kPa, and a holding time of 4 min. Tests were made in a universal Shimadzu AGX 1000 kN testing machine at a crosshead speed of 50 mm min^{-1} under controlled room temperature of 23 ± 2 °C. Tensile modulus, tensile strength, and elongation at break were obtained from the average values of five specimens. Additionally, hardness of samples was measured using a Shore D digital durometer from YPED.

Products processing conditions
Two sets of products were obtained: extruded profiles and mold-injected products. For extrusion process, virgin PE-E and CH were pre-mixed in a rotating container, dried and mixed in a co-rotating twin-screw extruder at 60 rpm using a temperature profile from 130 °C, at the feeding section, to 195 °C, at the extrusion die. The twin screw extruder had a diameter of 25 mm and L/D = 45, with a side-feeder. The head pressure was 25 MPa, and the mass flow rate was 40 kg h^{-1}. The screw profile, barrel sections temperatures and geometry are detailed in Fig. S1 and Table S1 of Supplementary Material.

For injection molding, PE-I/CH extruded composites were cut into small pieces and crushed into smaller granules using a blade milling machine. Injection molding conditions were defined after several experiments, and the best conditions (where samples did not exhibit apparent visual defects), were settled as follow: temperature at the barrel, nozzle, and mold was set at 200–180-40 °C, respectively, for a rotation rate of 80 rpm, injection pressure of 20 MPa, and holding pressure of 35 MPa. Injection time was 3 s and the mold clamp force 20 ton. The reciprocating screw extruder had a diameter of 35 mm with L/D = 45. The screw profile, sections temperatures and setting parameters are detailed in Fig. S2 and Tables S2 and S3 of supplementary material.

Environmental analysis
A general analysis considering positive and negative environmental impacts from a product life cycle perspective was made, using the model proposed by the U.S. Environmental Protection Agency (EPA) for landfill gas emission model [16]. The software LandGEM versión 3.02 was used to calculate greenhouse gases emissions.
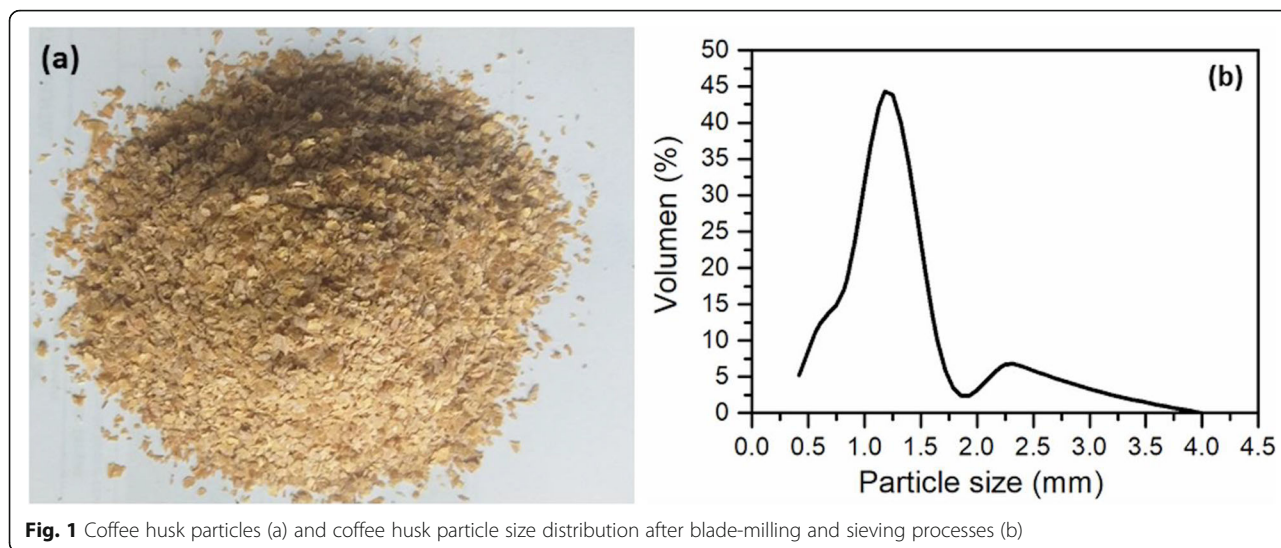
Results and discussion
CH characterization
Figure 1a shows the appearance of CH as received from coffee producers, prior to grinding process. It was seen that the husk was a dry material with an appearance similar to sawdust, with irregular particle size. After blade-milling and sieving processes, CH particle size distribution was much narrow (Fig. 1b). About 90% of the particle volume was less than 1.8 mm in size, while the remaining 10% presented a size between 1.8 and 4 mm.

SEM offers an excellent opportunity of studying the morphology on the surface of milled CH. In Fig. 2, it was noticed that CH exhibited variable rough surfaces that may enhance the mechanical adhesion to the

Table 2 Composites formulation and designation

Materials	Polyethylene (wt%)	Coffee husk (wt%)	Composite designation
80 HDPE + 20 CH	80	20	PE-E-20
60 HDPE + 40 CH	60	40	PE-E-40
80 LDPE + 20 CH	80	20	PE-I-20
60 LDPE + 40 CH	60	40	PE-I-40

Fig. 1 Coffee husk particles (a) and coffee husk particle size distribution after blade-milling and sieving processes (b)

matrix. Figure 2a for instance, shows two CH particles constituted from agglomerated fibers of irregular lengths, as well as some smaller particles left after blade-milling process. This result suggests that CH has a clear tendency to exfoliate when subjected to mechanical stimuli leading to formation of smaller fibers and particles. Figure 2b on the other hand, presents a closer look to the surface of a single CH fiber. The rough surface presented a pattern of valleys and peaks that were distributed along the entire surface. This surface irregularity is highly beneficial when it is pursued a good mechanical adhesion to the polymeric matrix, particularly, if not chemical pretreatment is made to the fiber or no compatibilizing agent is used [17].

Table 3 shows the chemical composition of CH. It was observed that the main elements found in the husk were magnesium and calcium, and that toxic elements such as cadmium, nickel and lead were not present, or they were below the detection limit of the equipment, as is the case of mercury. Therefore, these results showed that it was suitable to use this CH as a filler in polymers, since the products produced with these composites will not show metal toxicity.

One of the biggest challenges with the use of this type of wastes in thermoplastic polymers reinforcement is related to their degradation temperature. CH is an organic waste, and processing at high temperatures can lead to its degradation. TGA analysis of CH showed that at 195 °C (Fig. S3), the higher processing temperature applied in this work (180 °C for injection molding and 195 °C for extrusion), the mass loss was only 9.6%. This loss was attributed to moisture and volatile compounds present in the husk, and showed that this waste was stable at all processing temperatures, that is, it will not degrade in the mixing processes and production of the products by injection or extrusion.
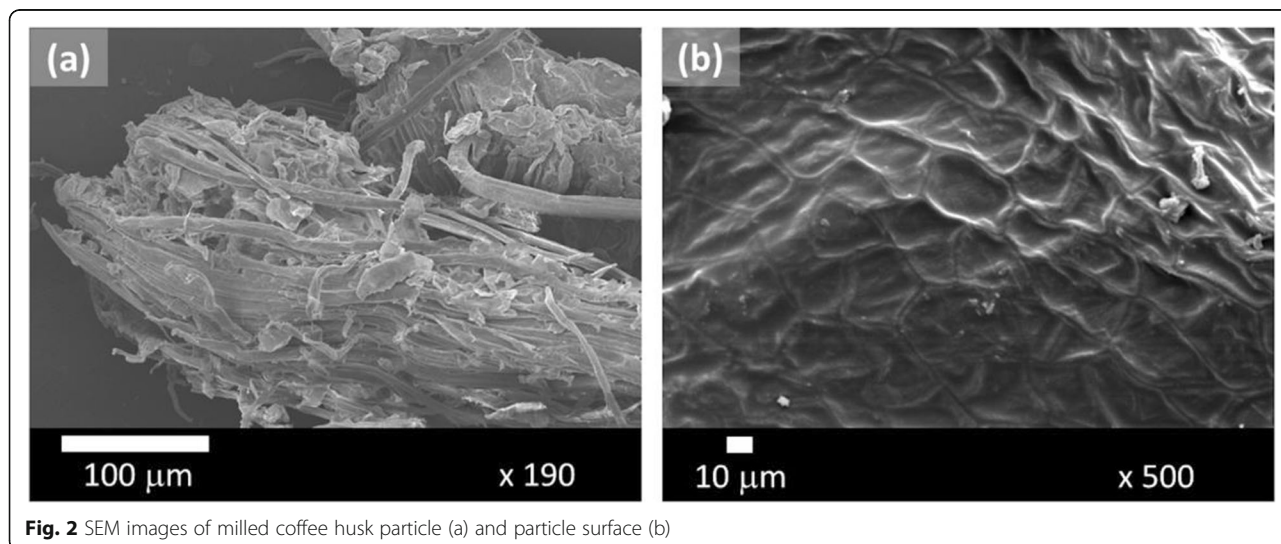

Fig. 2 SEM images of milled coffee husk particle (a) and particle surface (b)

Table 3 Chemical composition of coffee husk used in this work

Parameter	Result[a]
Cadmium	Not detected
Calcium	78
Calcium - CaO	109
Cupper	4
Total Iron	57
Magnesium	213
Magnesium - MgO	353
Mercury	< 0.2
Nickel	Not detected
Lead	Not detected

[a]Units: mg 100 g^{-1}, except for Hg which is mg kg^{-1}

Composites processing

In Fig. 3, it is presented the torque variation along time of a PE-E/40 composite. During mixing process, the polymeric matrix was first added to the mixing chamber and allowed to melt for 2 min, followed by the addition of CH. The initial mixing torque increased by the shearing action toward HDPE. The temperature increasing caused by the friction between polyethylene pellets summed to the processing temperature, led the polymer to melt, and as a consequence, the torque values decreased and stabilized. A second torque peak raised was caused by the addition of 40 wt% CH to the molten polymer. After 3 min, it was reached the torque stabilization, indicating that the filler was dispersed and homogenized in the mixture. It was noticed that stabilization torque value increased with CH addition. This is a well-known behavior on polymer/organic fillers composites, where it is expected an increase on viscosity since fillers make more difficult the material to flow, due to decrease of polymer chains mobility [12]. However, despite there was not lubricant or other kind of additive within the formulation, the addition of CH to the polymer matrix did not prevent the melt processing of composites, even though stabilization torque increased by about 21% compared to that of the virgin polymer at the beginning of the mixing process. Similar results were observed during processing of composites with 20 wt% of CH.

Materials and composites characterization
Thermal behavior of composites

Table 4 lists the thermal properties and crystallization degree (X_c) of pure polymers and composites. It was noticed that melting temperature of composites was slightly different than that of virgin polymers, and shifts to lower values by 2 °C approximately. This variation was attributable to CH inclusion in the polymer matrix. Nevertheless, this variation was not significant, and suggested that CH addition did not affect negatively the composites processing temperature. More notable variations were observed in the ΔH_m values of composites, which were lower than those of pure polymer resins. This was expected, since the area of the melting peak will be smaller due to the addition of CH. However, variation on crystallinity did not follow the same trend.

Crystallization degree was calculated using Eq. (2) for a theoretical enthalpy of fusion of 290 J g^{-1} of polyethylene 100% crystalline [18]. This equation considers the amount (wt%) of each component in the mixture. In
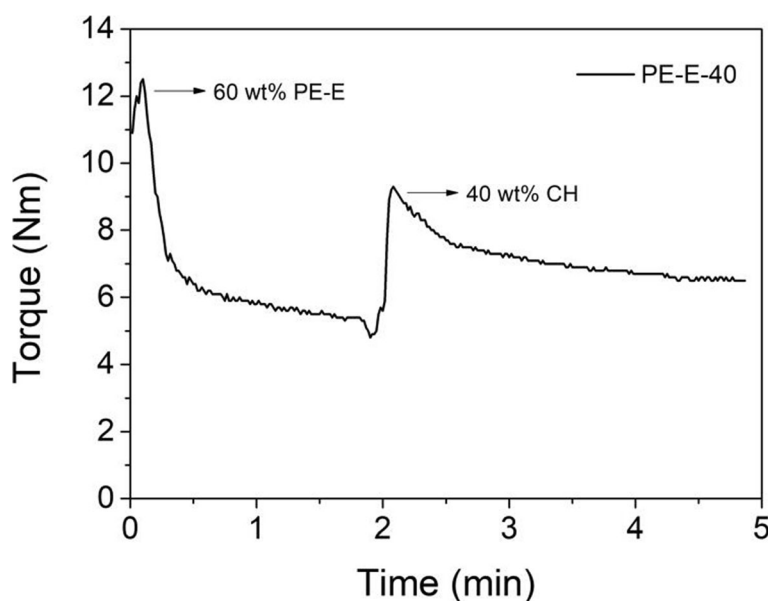

Fig. 3 Mixing sequence of PE-E-40 composite

Table 4 Thermal behavior and crystallization degree of composites

Sample	T_m (°C)	ΔH_m (J g^{-1})	X_c (%)
PE-E	123.23	41.0	14.1
PE-E-20	123.92	30.0	12.9
PE-E-40	120.31	32.8	18.9
PE-I	122.57	23.3	8.0
PE-I-20	120.52	22.9	9.9
PE-I-40	120.16	21.7	12.5

general terms, adding CH to polyethylene matrix caused an increase in composites crystallinity, suggesting that some CH particles acted as nucleating agents that enhance crystals growth, and hence, a reduction in the amorphous portion within the polymeric matrix. This has a direct effect on composites properties, particularly, in mechanical properties, since they are influenced by their crystallization degree [12, 18].

Composites morphology

Figure 4 shows SEM images of different composites containing 20 wt% CH, where it was noticed the distribution of CH along polyethylene matrix. In PE-E-20 composites, there was observed some voids around the fillers that separated them from the matrix, which could be attributed to the evaporation of water trapped in the filler, since the drying time for CH used in this work was short

(4 h). Nevertheless, some CH particles bonded to the polymeric matrix with a fibrillar structure were observed, as a sign of physical filler/matrix interaction. In the case of injection molded composites, the interaction was similar to the extruded ones. However, CH fibers similar to those observed in Fig. 2a were better exposed due to the fact that during injection molding fibers went through conditions of high shearing stress. That allowed the polyethylene matrix to interact better with the fibers, which were well embedded. Even though the matrix wetted better the CH fibers in injected molded samples, there were still noticed some voids left, which was to be expected since no compatibilizer agent was present.

Some interesting findings are observed in Fig. 5, where the SEM images of PE-I-40 composite revealed CH fibers exfoliation. This phenomenon is common in organic fillers, and occurs by the action of high shearing effects during composites melt processing [19, 20]. The formation of a spiral-like structure allowed CH particles to increase the surface interaction area with the polymer matrix, as observed in Fig. 5a, where the spirals are filled with the matrix letting some to get inside this structure during the melting stage. Some regions revealed the polymer matrix completely filled the exfoliated spiral, as noticed in Fig. 5b, giving rise to a better interaction between the two components. This could positively affect the mechanical properties of the eco-composites. It is important to note that no compatibilizing agent was
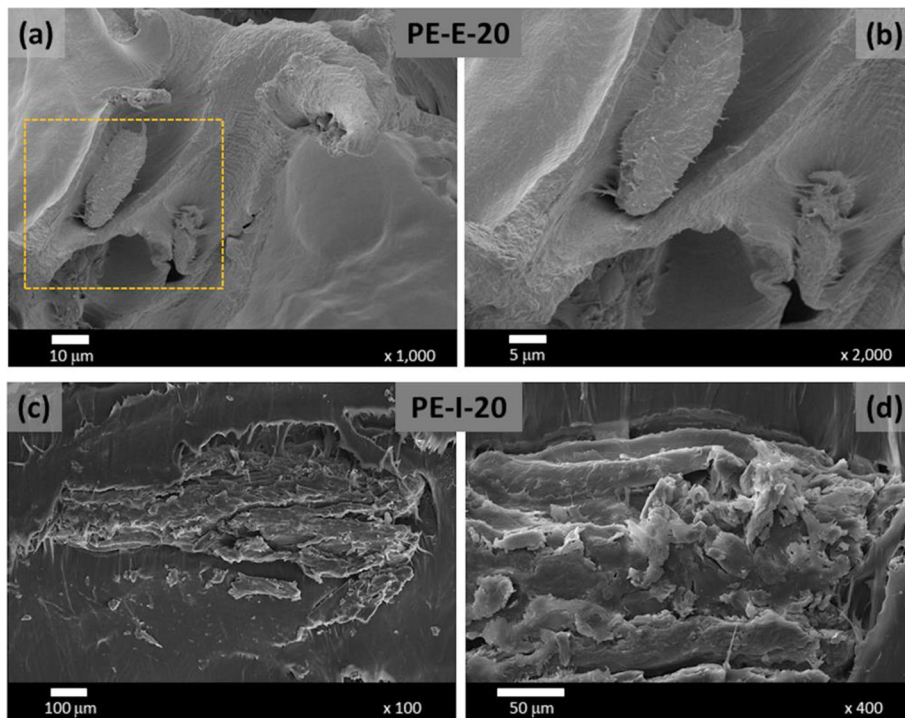

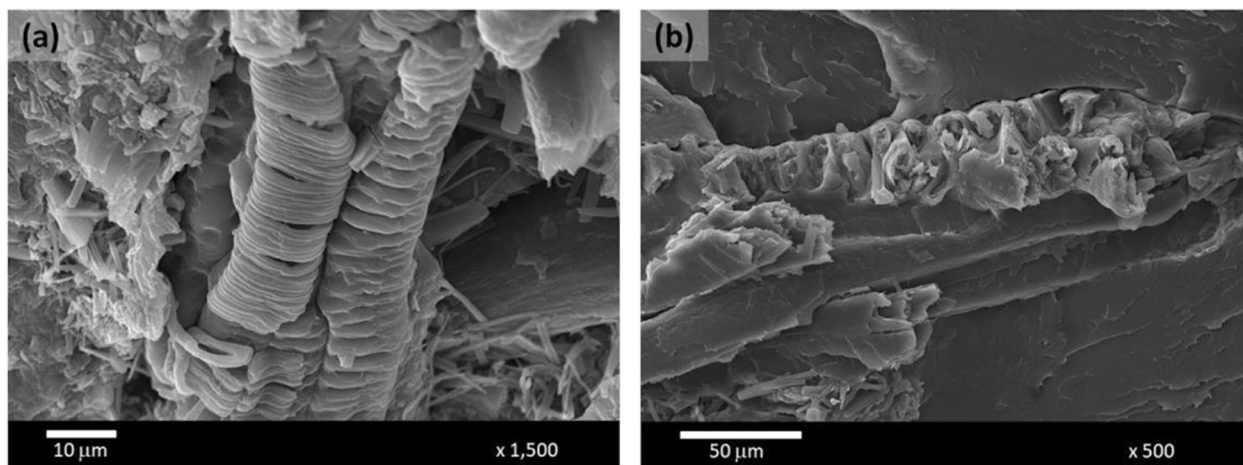**Fig. 4** SEM images of PE-E-20 (a, b) and PE-I-20 (c, d) eco–composites

Fig. 5 Coffee husk spiral-like structure formed by exfoliation during melt mixing process of PE-I-40 partially filled with polyethylene (a) and fully filled with polyethylene (b)

used in this work, so these types of interactions are important to improve the contact between the organic filler (CH) and the polymeric matrix.

Mechanical properties

In Fig. 6, it is presented the representative behavior of tensile deformation when CH was added to the polyethylene matrix. When 20 wt% CH was added to PE-I matrix, tensile strength and ductility of composite decreased by 78 and 87%, respectively, compared to the polymer without CH. Additionally, the amplification in Fig. 6 shows that increasing CH content in composites led to a major decreasing in mechanical properties, since tensile strength decreased by 85% and ductility by 93%,

when 40 wt% CH was added. This behavior was due to a combinatorial effect between the increasing of matrix crystallinity, fillers addition, and the lack of compatibilizer agents. As a consequence, the lack of interface formation leads to poor stress transference from the PE matrix to the CH filler. Similar results were reported in different works [12, 21, 22], where addition of different natural fillers to polymeric matrices led to decreasing the mechanical properties. Some authors used compatibilizer agents which improved the mechanical performance of composites. In their work, Yang and coworkers [21] added maleated PP and maleated PE to LDPE and HDPE matrices, and obtained small increases in the tensile strength.
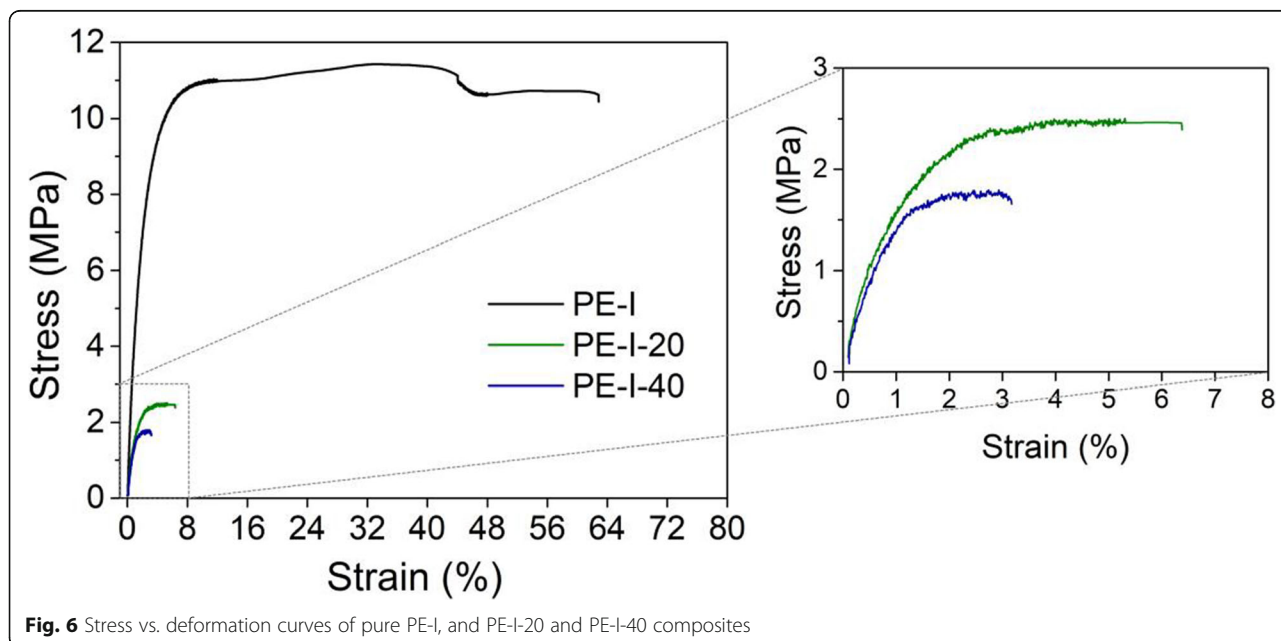

Fig. 6 Stress vs. deformation curves of pure PE-I, and PE-I-20 and PE-I-40 composites

Nevertheless, compatibilizer agents help improving mechanical performance of composites. It is found that particle size distribution of fillers has a strong effect on tensile strength. In their works, Yang et al. [21] and Zhang et al. [22] reported values of tensile strength for uncompatibilized composites between PE and different natural fillers as high as the virgin polymers, since the particle size was close to 80–120 mesh (0.125–0.18 mm). In this work, we added CH with a particle size of maximum 60 mesh (0.25 mm), which decreased the contact surface area between the matrix and the filler. On the other hand, it is known that the different polarity of a natural filler and an apolar polymeric matrix, as well as the chemical composition of the filler affect to the adhesion between filler and matrix [23].

Table 5 summarizes the mechanical behavior of virgin materials and eco-composites. Young modulus exhibited a similar behavior in both kind of composites. Addition of CH gradually increased composites modulus, and hence their stiffness. This is in good agreement with crystallinity results reported in Table 4, where higher degree of crystallization leads to more rigid materials, since polymer mobility is reduced.

Regarding elongation at break resistance of composites, it was observed that adding CH to polyethylene matrix caused a prompter rupture of materials. This was explained from the increase in materials rigidity, which made them to become more brittle. Additionally, the lack of compatibilizer agent led to poor interphase between the polymeric matrix and the filler. This is observed in the work of Zhang et al. [22], where addition of compatibilizer agent to composites of HDPE with CH fillers improves in almost 100% the elongation at break of composites.

Shore D hardness was also measured to contrast with tensile tests, and results showed they were in good agreement. Adding CH to both polyethylene resins used in this work gave as a result harder material.

Despite the decrease in some mechanical properties, stiffer materials were obtained. In addition, tensile resistance and ductility obtained, will allow obtaining different products that can be used in multiple applications that do not demand high ductility.

Obtained products using eco-composites

PE/CH eco-composite products were obtained with the highest CH content (40 wt%) in order to verify the processability of the mixtures with the higher waste content at an industrial scale. As mentioned before, no pre-treatments to CH or compatibilizers for the mixture were used for neither, extruded nor injection molded products. Although the use of compatibilizers is highly recommended in order to improve matrix-filler compatibility between natural fillers and polyolefins such as polyethylene, in this work it was decided not to use compatibilizers in order to use the least number of reagents and produce more sustainable composites. Nevertheless, the products obtained do not exhibit considerable visual defects, and, according to mechanical properties presented previously, they are suitable to be used in different applications.

The whole extrusion and injection processes to obtain the eco-compounds are illustrated in Fig. 7. In both processes, the raw materials were pre-mixed and then molded at temperatures lower than CH degradation.

Three types of products were obtained through the extrusion process, extruded profiles for floors and construction uses (Fig. 8). Extruded profiles can be used for residential or commercial floors, such as terraces, walkways, docks, patios, etc., as well as for the manufacture of other products such as park benches, furniture or fences (Fig. 8b and c). They also have multiple uses in the construction sector, such as in the manufacture of structures, lightweight walls, enclosures, among others (Fig. 8a and b).

Similar to what was observed during the mixing process in the torque rheometer, an increase in the viscosity of the mixture was identified during the industrial manufacture of the extruded profiles. Usually, the products manufacture from polymeric matrix with some type of filler presents a restriction related to the increase of composite viscosity when the filler is added. However, with the processing temperature used in this work (195 °C), an adequate mixture flow could be checked, since no volume or surface defects were found in the extruded products.

Table 5 Mechanical properties of pure polymers, and PE-E/CH and PE-I/CH composites

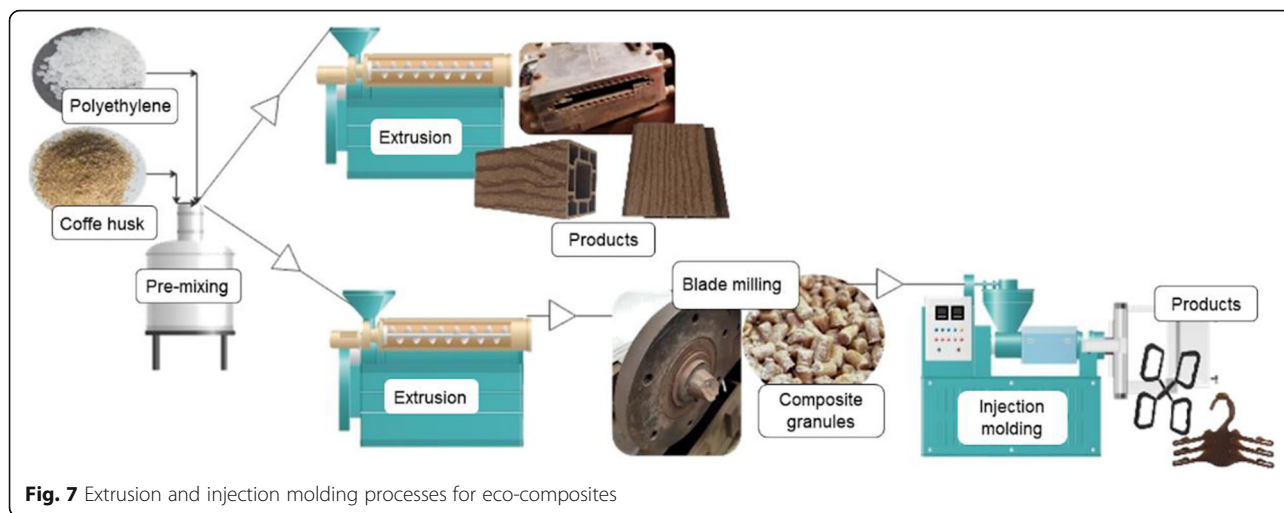| Sample	Young Modulus (MPa)	Tensile strength (MPa)	Elongation at break (%)	Hardness (Shore D)
PE-E	950 ± 1.5	9.9 ± 0.2	37.1 ± 0.1	62 ± 0.6
PE-E-20	980 ± 1.4	3.3 ± 0.1	4.8 ± 0.2	68 ± 0.7
PE-E-40	1020 ± 1.2	2.9 ± 0.1	3.7 ± 0.2	75 ± 0.6
PE-I	350 ± 1.1	11.4 ± 0.1	62.9 ± 0.1	41 ± 0.9
PE-I-20	390 ± 1.5	2.5 ± 0.1	8.1 ± 0.3	49 ± 0.5
PE-I-40	420 ± 1.3	1.7 ± 0.1	3.9 ± 0.3	55 ± 0.4

Fig. 7 Extrusion and injection molding processes for eco-composites

On the other hand, two injected molded products were obtained using PE-I-40 eco-composite. These products were bag handles (Fig. 9a) and clothes hanger (Fig. 9b). There are many important concerns about the use of natural fiber eco-composites in conventional injection molding, including the low effective flow within the molds and the weak interface between the natural filler and the polymer [24]. However, injected products were obtained without any visible surface defects such as voids, bubbles or burn marks, which indicates that the given temperature and speed conditions are suitable for this type of composite.

The physical characteristics of all manufactured products are presented in Table 6. All products obtained met the physical parameters of the initial design.

Products manufactured by injection and extrusion of eco-composite have wide uses, and could replace products made exclusively with polyethylene in some

applications. There were no operational problems during the extrusion or injection processes even using a high amount of filler (40 wt%). Furthermore, even without the addition of fiber-polymer compatibilizing agents, it is possible to obtain products without significant defects. This indicates that the developed eco-composite could be produced in conventional facilities and used in production of common products, which would generate interesting economic and environmental benefits.

Environmental assessment of the products
Life cycle approach
Considering that raw materials extraction, use and final disposal of products causes important environmental and economic consequences, a comparative study was made between different Life Cycle Analysis (LCA) of products in which natural fibers were mixed with thermoplastics polymers, such as HDPE and polypropylene
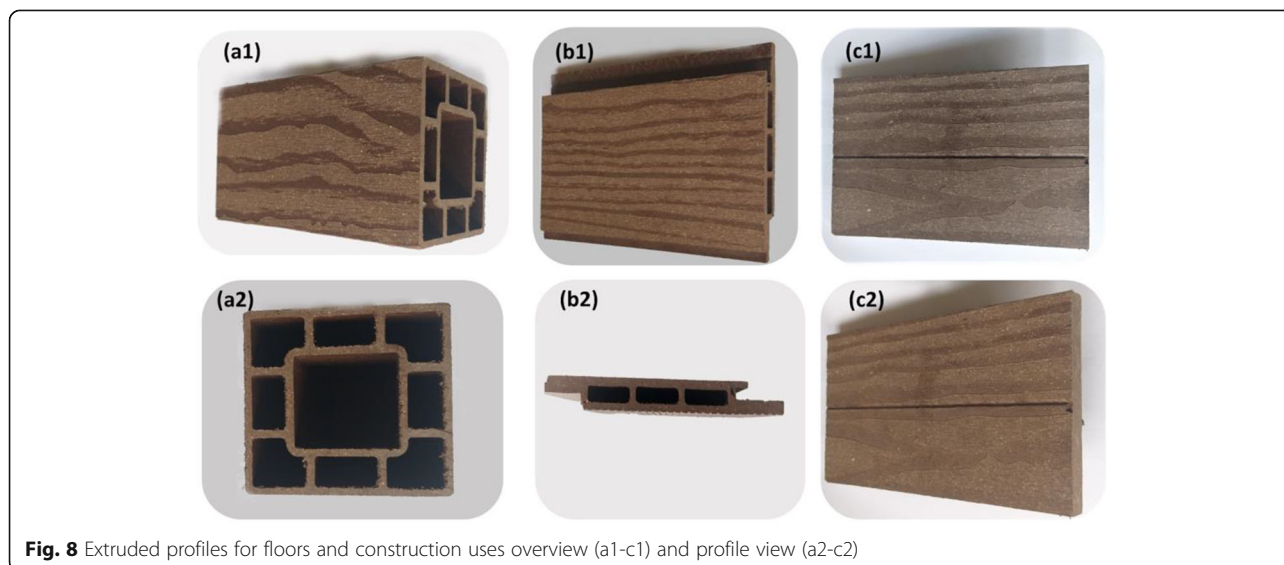

Fig. 8 Extruded profiles for floors and construction uses overview (a1-c1) and profile view (a2-c2)
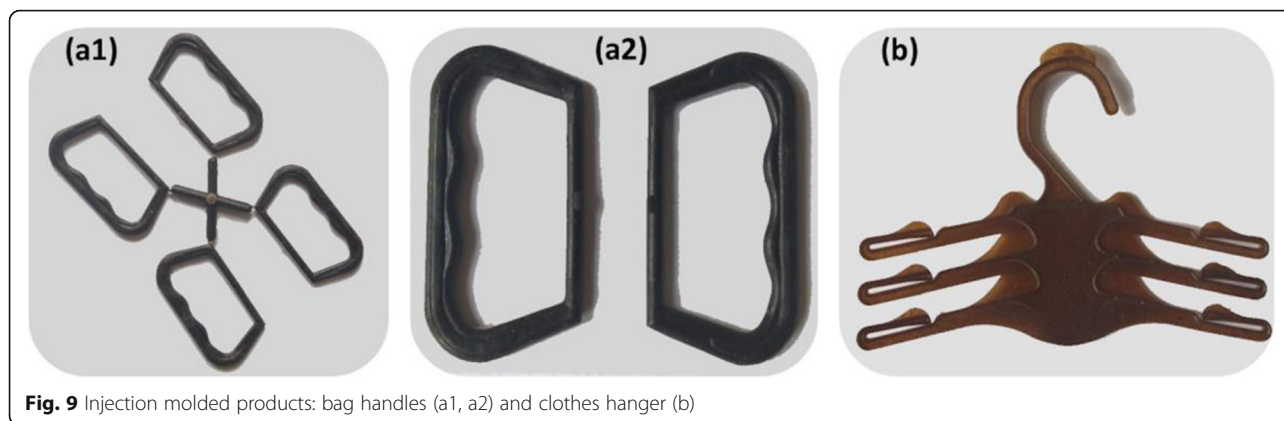
Fig. 9 Injection molded products: bag handles (a1, a2) and clothes hanger (b)

(PP) (Table 7). Although PP has not been used in this work, this polymer has been included as it is another thermoplastic polymer highly used in processes similar to LDPE. Some important comparisons were made based on the LCAs studied. Products or composites made 100% in thermoplastic were compared with both composites with natural fillers (mainly natural fibers from wastes) and those with conventional fillers (inorganic commercial fillers), which are dominant reinforcement in polymer matrix currently.

Saraiva et al. [25] compared LCA for virgin HDPE and for HDPE/sponge gourds residue composites (90/10 and 70/30 wt%, matrix/filler composition). It was found that 70/30 composites had the lowest Global Warming Potential (GWP), with 0.61 kg CO_2eq, followed by 90/10 composites (0.75 kg CO_2eq) and virgin HDPE (0.84 kg CO_2eq). Likewise, Vidal et al. [26] compared the LCA for both virgin HDPE and recycled HDPE/cotton composite (72/20 wt%), as well as for virgin PP and recycled PP/cotton composite (73/20 wt%). Environmental impact analyses found that the GWP of virgin thermoplastics was higher than that of recycled fiber composites by approximately 60–70%. The same occurred for Acidification Potential (AP) and Eutrophication Potential (EP).

On the other hand, regarding the impact of the substitution of conventional fillers for natural ones, Luz et al. [27] compared environmental impact of PP composites filled with 20 wt% talc (conventional filler) and sugar cane bagasse (natural fiber). The LCA showed lower GWP values when natural fiber was used as filler than when the conventional one (talc) did, achieving a considerable decrease of approximately 90%. The same occurred for other environmental impacts such as Ozone Depletion Potential (ODP), AP and EP, with decreases of 6, 3 and 2%, respectively. In another work, Corbiere-Nicollier et al. [28] analyzed the life cycle for standard pallets with the same service requirements made of PP/fiberglass and PP/china reed composites, finding lower environmental impacts mainly in terms of energy consumption and GWP for products made with natural fibers. This was justified due to three important factors: (a) the substitution of glass fiber production by the natural fiber production; (b) the indirect reduction in the use of polypropylene linked to the higher proportion of natural fiber used; and (c) the reduced pallet weight, which reduces fuel consumption during transport. Other authors agree with this result when targeting equal strength equivalence, since they observed that the use of natural fibers to replace conventional fibers such as fiberglass, allows to obtain composites with similar properties and a better environmental performance [31, 32].

As mentioned above, natural fibers have advantages that encourage their use as fillers of polymeric matrices, such as low cost, density and weight, less abrasive damage to equipment, biodegradability and minimum health risks [9, 33]. As a result, several authors have pointed out that the use of natural fibers in the composites production has positive impacts on both the economic and environmental perspectives, enabling the production of new renewable raw materials for new green products on the market [34, 35].

Table 6 Physical characteristics of products obtained using eco-composites

Characteristic	Extruded profile A	Extruded profile B	Extruded profile C	Clothes hanger	Bag handle
Thickness (mm)	5	8	10	4	3
Width (mm)	80	180	120	165	30
Length (mm)	160	200	200	200	70
Weight (g)[a]	200	260	350	80	50

[a]For 1 m of extruded profiles products and for unit of product for injected ones

Table 7 Life Cycle Analysis comparison of different thermoplastic/natural fiber composites

Product type/ composite type	System boundaries	Functional unit	Polymer		Natural Fiber		Other	Manufacture Process	Impact Categories				Ref.
			Type	%	Type	%	%		GWP (kg CO_2-eq)	ODP (kg CFC11-eq)	AP (kg SO_2-eq)	EP (kg $(PO_4)_3$-eq)	
Food packaging	Cradle to grave	1 packaging[a]	HDPE	100	Sponge gourds residue	0	0	Extrusion, injection and other	0.84	0.54	–	–	[25]
		1 packaging[a]	HDPE	90	Sponge gourds residue	10	0	Extrusion, injection and other	0.75	0.55	–	–	
		1 packaging[a]	HDPE	70	Sponge gourds residue	30	0	Extrusion, injection and other	0.61	0.65	–	–	
Composites	Cradle to grave	1 kg composite	HDPE (recycled)	72	Recycled cotton (cotton linter)	20	8	Extrusion	0.61	–	4.03E-03	2.67E-04	[26]
		1 kg composite	PP (recycled)	73.2	Recycled cotton (cotton linter)	20	6.8	Extrusion	0.70	–	4.25E-03	3.63E-04	
		1 kg composite	PP (recycled)	50	Rice husks	45	5	Extrusion	0.71	–	4.87E-03	9.34E-04	
		1 kg	HDPE (virgin)	100	–	0	0	Extrusion	1.88	–	2.14E-02	1.30E-03	
		1 kg	PP (virgin)	100	–	0	0	Extrusion	1.99	–	2.03E-02	1.26E-03	
Automotive parts	Cradle to grave	1 m^2 surface area covered	PP	80	Sugarcane bagasse	20	0	Extrusion, injection	7.23E-06	4.18E-09	1.71E-06	1.67E-06	[27]
		1 m^2 surface area covered	PP	80	Talc	20	0	Extrusion, injection	7.07E-05	4.46E-09	1.77E-06	1.71E-06	
Transport pallet	Cradle to grave	Standard pallet (same service requirements)	PP	58	Glass fiber	42	0	Not specified	75.3	–	0.653	0.0682	[28]
			PP	47	China reed	53	0	Not specified	40.4	–	0.411	0.0632	
Wood plastic composite	Cradle to Gate	1 kg virgin composite	HDPE	68	Wood	29	3	Not specified	2.18	1.10E-09	6.10E-03	6.20E-04	[29]
			HDPE	39	Wood	58	3	Not specified	1.73	9.50E-10	4.40E-03	4.90E-04	
		1 kg recycled composite	HDPE	68	Wood	29	3	Not specified	0.76	6.60E-10	1.40E-03	1.90E-04	
			HDPE	39	Wood	58	3	Not specified	0.78	6.60E-10	1.50E-03	2.00E-04	
Biocomposites	Cradle to grave	Coffee jar lid (53 mm diameter)	HDPE	100	Banana fiber (untreated)	0	0	Extrusion, injection	0.04	–	–	–	[30]
			HDPE	60	Banana fiber (untreated)	40	0	Extrusion, injection	0.03	–	–	–	
			PLA	100	Banana fiber (untreated)	0	0	Extrusion, injection	0.06	–	–	–	
			PLA	60	Banana fiber (untreated)	40	0	Extrusion, injection	0.04	–	–	–	

[a]Used to transport ten mangoes from the production area to the end-consumer

These studies show that using natural fibers as fillers in composites, and the replacement of traditional fillers by natural fibers, reduces the environmental impacts related to the different processes of the products' life cycle and allows obtaining products that can be made available to the market. Additionally, it makes possible the valorization of agro-industrial wastes. In this same sense they point out the PE/CH eco-composites produced in

this work. The production of products with CH in their composition will reduce the environmental impact of pure polyethylene products, and makes CH valorization possible, avoiding the final disposal of this waste in landfills, which would increase the emission of greenhouse gases.

Polyethylene substitution environmental benefits

The successful production of PE/CH eco-composites by extrusion and injection molding technology can reduce the adverse environmental impacts of the plastics industry. The CH utilization to reduce the amount of conventional petrochemical polymers used, valorizing an abundant waste, contributes to save material costs while providing an eco-composite that meets the circular economy criteria and can offer a green niche for the plastics industry.

Considering the Colombian case, in 2018 the apparent consumption of polyethylene in extrusion and injection molding processes was approximately 400 kt [36]. According to our research presented in this work, it was possible to produce composites and products with up to 40 wt% CH, that is to say, there is the possibility of replacing approximately up to 160 kt of polyethylene with CH per year in various products manufactured in Colombia.

On the other hand, it is calculated that in Colombia the production of CH was approximately 5200 tons for the same year [37]. Therefore, considering the above data, all the production of this agro-industrial waste could be valorized in the plastic sector, thus avoiding its disposal in landfills or open dumps, which are the usual practices used today in Colombia.

Environmental problems associated with open dumps are well known. Some of them are local air pollution, greenhouse gas emissions, vector borne diseases and inappropriate land use [38]. Greenhouse gases, such as methane and carbon dioxide, are emitted in significant quantities due to decomposition of organic matter. According to the USEPA model, if the 5200 tons of CH generated in Colombia in 2018 are disposed in landfills, a total of 5,350,000 m^3 of greenhouse gases would be generated, being the total emissions of methane and carbon dioxide 3,200,000 and 2,150,000 m^3, respectively. Approximately 60% of these greenhouse gases emissions would be generated in the first 20 years of biomass disposal, and then continue to be generated downwards for more than 50 years.

Additionally, in Colombia it is estimated that the percentage of plastic materials consumed in different uses is 54% in packaging, 22% in building and construction, 7% in agricultural products, 7% in general consumer goods and 11% in industrial products and other applications [36]. Considering that the apparent consumption of polyethylene in Colombia is approximately 400 kt per year, according to the eco-composites produced in this work, if only 3.25% of these thermoplastic products substitute 40% of polyethylene with CH, all CH produced each year in the country could be used. This implies a reduction in the emission of 5,350,000 m^3 of greenhouse gases, according to previous calculations.

This economic and environmental opportunity to take advantage of the CH as raw material in the manufacture of polyethylene products, could have specific applications in subsectors such as construction and general consumer goods, considering that currently products manufactured in wood-polymer composites, such as produced in this work, are being positioned in the local and global market [39].

Conclusions

In this work, polyethylene eco-composites filled with 20 and 40 wt% CH were produced and characterized by different thermal, morphological and mechanical techniques. The industrial processability of the composites was verified by producing five products by extrusion and injection molding processes. Additionally, the environmental impacts of eco-composites were considered.

SEM images showed that CH particles were embedded in the polyethylene matrices. However, due to the non-use of compatibilizer agent, some gaps were observed at the polymer-filler interphase. Interestingly, it was also observed that during composite processing, the exfoliation of some CH fibers took place, which allowed a better fiber-matrix surface contact area. The mechanical behavior of virgin materials and eco-composites showed that both elongation at break and tensile strength decreased when CH was incorporated into the polymer, probably due to different aspects, such as the differences in chemical structure and polarity between the filler and the polymer matrix, a high particle size of the filler and the absence of a compatibilizer agent. However, the Young modulus and hardness values of the eco-composites increased compared to virgin polymers, indicating that the addition of CH to LDPE and HDPE results in a stiffer and harder material.

The industrial processability of the eco-composites with the highest CH content (40 wt%) was verified through the production of five products, three extruded profiles for different uses, and a clothes hanger and a bag handle obtained by injection. In all cases, the products met the physical parameters of the initial design, and did not present significant defects.

According to many life cycle studies, composites that use natural fibers, such as CH, provide the reduction of environmental impacts such as AP, EP, and GWP caused by the life cycle of products manufactured from them. In Colombian case, if only 3.25 wt% of PE products were

produced with the eco-composites developed in this work, all the CH produced in the country would be used, and the emission of about 5,350,000 m^3 of greenhouse gases would be avoided. This alternative for waste valorization brings a sustainable perspective to developing countries due to its positive economic and environmental implications.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s42834-021-00107-6.

> **Additional file 1: Table S1** Extruder screw profile, barrel sections temperatures and geometry. **Table S2** Screw profile and sections temperatures. **Table S3** Setting of injection molding machine parameters. **Fig. S1** Schematic diagram of the extruder screw sections. **Fig. S2** Schematic diagram of the reciprocating screw extruder sections (a) feeding zone, (b) melt zone, and (c) metering zone, (d) diameter = 35 mm, (l) length = 1575 mm. **Fig. S3** TGA analysis of rice husk (heating rate of 10 °C min^{-1}, temperature range of 25–900 °C, and synthetic air atmosphere flow of 100 cm^3 min^{-1}).

Authors' contributions
Leyla Y. Jaramillo: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – Original Draft, Writing – Reviewing & Editing, Visualization, Funding acquisition; Mauricio Vasquez-Rendón: Conceptualization, Methodology, Validation, Formal analysis, Writing – Original Draft, Writing – Reviewing & Editing, Visualization; Sergio Upegui: Methodology, Investigation, Funding acquisition; Writing – Original Draft; Juan C. Posada: Methodology, Investigation, Funding acquisition; Manuel Romero-Sáez: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – Original Draft, Writing – Reviewing & Editing, Visualization, Funding acquisition. All authors read and approved the final manuscript.

Availability of data and materials
All data generated or analyzed during this study are available with the authors.

Declarations

Competing interests
The authors declare they have no competing interests.

Author details
^{1}GITIMA Research Group, Tecnológico de Antioquia, Medellin 050036, Colombia. ^{2}Quality, Metrology and Production Research Group, Instituto Tecnológico Metropolitano, Medellin 050036, Colombia.

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