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Additional Information

“Development of compatibilized polyamide 1010/coconut fibers composites by reactive extrusion with modified linseed oil and multi-functional petroleum-derived compatibilizers”

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Abstract. This work reports the preparation and characterization of fully biobased polymer composites with coconut fibers (CFs) as an alternative to wood-plastic composites, typically based on petroleum-derived materials. Polyamide 1010 (PA1010) was melt-extruded with 20 wt% of CFs and, after that, shaped into pieces by injection molding. Four different multi-functionalized compatibilizers were tested to increase the polymer-fiber interactions with the subsequent improvement on toughness. These consisted of two chemically modified vegetable oils, namely maleinized and epoxidized linseed oil, MLO, and ELO respectively, and two commercial additives derived from petroleum and based on glycidyl functionality, that is, low-functionality epoxy-based styrene-acrylic oligomer (ESAO) and polystyrene-glycidyl methacrylate random copolymer (PS-GMA). The addition of all four compatibilizers improved both the mechanical and thermomechanical properties of the composites, thus resulting in high-performance composite materials with relatively low water uptake. Furthermore, the

morphology of the obtained composites revealed an extraordinary embedment of the fibers into the biopolymer matrix, which plays a crucial role in improving toughness. Among all the tested compatibilizers, those derived from vegetable oils can be considered the most interesting due to they offer a complete sustainable solution.

Keywords: Bio-based polyamides; natural fibers; high-performance composites; polymer-matrix interaction; multi-functionalized vegetable oils.

1. Introduction.

In the last years, natural fibers are gaining an essential role in developing high environmentally-friendly composites to overcome the current ecological and environmental problems related to the recyclability of glass fiber-reinforced plastics (GFRP). Besides, by using natural fibers, it is possible to upgrade a wide variety of natural waste or by-products. For this reason, natural fibers such as hemp [1, 2], flax [3, 4], jute [5, 6] or sisal [7, 8] are becoming particularly attractive in technological sectors such automotive and transportation industries because of their lower cost and lower density, therefore leading to lightweight composites as realistic alternatives to glass-reinforced composites in many applications [9]. Moreover, one of the primary wastes from agriculture, agroforestry and other food-related industries are lignocellulosic materials that offer high potential as reinforcing materials as they are readily available and characterized by having a low weight, being fully bio-sourced and biodegradable (disintegrable in compost soil), showing low abrasive properties and, in most cases, they represent a cost-effective solution to other conventional reinforcing fibers. In particular, coir palms (*Cocos nucifera L.*) are abundantly growing in tropical countries, and their wide variety of products are being applied in both food and non-food products. Coconut palms are grown in more than 100 countries and autonomous regions of the world, which results in annual worldwide production of about 60.5 million tons in 2014 [10]. They represent an essential agribusiness in developing countries with a tropical climate [11]. A variety of uses have been proposed for the husks and shells, which remain after harvesting and processing of coconuts. For example, coconut husks and shells are widely used as an alternative energy source in some countries. In particular, the coconut husk is constituted of 30 wt% fiber and 70 wt% pith material [12]. This 30 wt% represents a colossal waste that does not have a great value, and it is discarded entirely. Coconut husks can be further processed into new useful fiber-like products known as coir when the fibers completely ground up. Nowadays, only 15 wt% of the husk fibers are recovered for different uses [13]. A much

more efficient utilization is possible, especially using shorter fibers (sometimes called fiber bits) that cannot be used for high-value products [10].

As already mentioned, an interesting approach is the use of coconut fibers as reinforcement in composites with a thermoplastic polymeric matrix, thus leading to the so-called "natural fiber-reinforced plastics" (NFRP) or "wood-plastic composites" (WPC) that are gaining a considerable interest due to the increasing environmental concerns as they positively contribute to lowering the carbon footprint and give rise to new solutions for a sustainable development in the frame of a new paradigm based on the Circular Economy [14-17]. The use of wood-plastic composites (WPCs) offers significant advantages from both economic and environmental standpoints, but they also are attractive from a technical point of view as they can be shaped by conventional manufacturing processes such as hot-press molding, extrusion, injection processes, and so on. [18, 19].

Typical polymer matrices in WPC include commodity polymers such as low- and high-density polyethylene (LDPE and HDPE) [20, 21], polypropylene (PP) [22, 23], polystyrene (PS) [24], polyvinyl chloride (PVC) [25] and some polyamides (PA)[26, 27]. Nevertheless, in accordance with the increasing environmental concern, new biopolymer matrices are being increasingly used in WPCs, such as polylactide (PLA), poly(ϵ -caprolactone) (PCL), poly(butylene succinate) (PBS), poly(butylene succinate-co-adipate) (PBSA), among others [28-30]. These polymers offer similar properties to those based on commodities but, also, they are disintegrable in controlled compost soil, that is, compostable. Additionally, to these aliphatic polyesters, new polymers are being synthesized from renewable resources. This is particularly noticeable in engineering plastics such as polycarbonate (PC), polyethylene terephthalate (PET), polyethylene furanoate (PEF), and polyamides (PAs) and their blends, which can be totally or partially obtained from renewable resources [31-33]. Polyamide 1010 (PA1010) is a new material from different standpoints; on the one hand, it is fully bio-derived from castor-

oil derivatives and, on the other hand, it has comparable properties, or even higher than those of petroleum-derived polyamide 12 (PA12). For these reasons, PA1010 offers interesting uses in engineering applications such as the automotive industry. PA1010 are flexible, high-impact resistance materials with improved heat resistance and low extraction [34, 35]. These balanced properties make PA1010 suitable for thermal management and complex parts in the automotive industry and also food packaging applications [36-38]. On the contrary, in the case of bio-PAs, the production cost may be higher, because the process of harvesting the plant material from the plant material is more expensive. In this sense, bio-PAs still present higher prices, in the range of US\$15,000–20,000 a ton, than conventional PA6 and PA66, that is, US\$2,000–4,000 a ton, or even specialty PA12, that is, US\$8,000–12,000 a ton, for this reason the use of loads and fibres is a great advantage for cost reduction [39]. In this sense, some authors have reported the use of PA1010 with some natural fibers such as hemp fiber [35, 40] or flax fibers [41].

One of the main drawbacks related to natural fiber thermoplastic reinforced plastics is the poor compatibility between the typical hydrophobic polymer matrix and the highly hydrophilic reinforcement, which are typically lignin, hemicellulose, and cellulose. This weak polymer-fiber interactions, together with some other drawbacks related to the high polar nature of the lignocellulosic fibers, such as poor fiber dispersion into the hydrophobic polymer matrix or high moisture gaining, do not allow to benefit from the excellent properties of the thermoplastic matrix and it leads to low-performance materials [42]. To overcome or, at least, minimize these effects, it is necessary to improve the polymer-fiber interaction, and this process can be achieved by different approaches. Some physical processes based on ultraviolet (UV) radiation and plasma treatments have given interesting results in different natural fiber reinforced plastics (NFRPs)[43, 44]. Conventional chemical processes on fibers such as esterification, acetylation, and so on, allow reducing the hydrophilicity of natural fibers with a positive effect on improved interactions between the polymer matrix and the

chemically modified fiber. In some cases, specific additives can be used, which contain functional molecules that can react with both the polymeric matrix and the natural fiber. In general, bi-functional copolymers give good results [45-47]. In this sense, Jeng *et al.* [48] have used polypropylene-*grafted*-maleic anhydride (PP-*g*-MA) to make PP compounds compatible with natural fibers with excellent results. Other approaches include the use of other functional molecules such as stearic acid [49], maleic anhydride [50] or silane and isocyanate [51], which can positively contribute to improved interface interactions. Recent researches are being focused on the development of highly environmentally friendly functional materials that could potentially help in increasing polymer-fiber interactions. The use of compatibilizers represents a powerful strategy to enhance interfacial adhesion between the lignocellulosic fillers/fibers and a wide range of polymer matrices [46, 52]. Multi-functionalized vegetable oils (VOs) represent a real solution to a range of new materials with different uses. For instance, epoxidized linseed oil (ELO) and maleinized linseed oil (MLO) have been successfully used as compatibilizers in polymer blends and polymer composites with polyester-type matrices such as PLA, PBS, PCL, PBSA, etc. [28, 53-55], due to the high reactivity of the oxirane and maleic anhydride groups towards hydroxyl groups in both aliphatic polyester and lignocellulosic fiber. Similar uses have been proposed for modified soybean oil and cottonseed oil [56, 57]. As an example, Garcia-Garcia *et al.* [58] recently reported that the use of vegetable oil derivatives does not compromise the overall biodegradation of poly(3-hydroxybutyrate) (PHB) aliphatic polyester.

These environmentally friendly additives derived from vegetable oils are gaining interest against petroleum-derived conventional chain-extenders or compatibilizers such as typical epoxy-based styrene-acrylic oligomers (ESAOs), commercially known as Joncryl[®]. These oligomers can quickly form new ester bonds through reaction of their epoxy groups with the hydroxyl terminal groups of the biopolymer chains during melt processing [59] and, obviously, these epoxy rings can also react with hydroxyl

groups of the lignocellulosic fillers/fibers to provide improved polymer-fiber interactions [60-62].

In this research work, highly environmentally friendly composite materials have been obtained with a PA1010 matrix and coir fiber as reinforcement. The effect of CFs on the mechanical, thermal, morphological, and thermomechanical properties was studied as a function of the different compatibilizers employed. The efficiency of the two bio-based compatibilizers obtained from linseed oil, namely ELO and MLO, respectively, was compared with two commercial petroleum-based compatibilizers widely used as chain extenders in polyester formulations, that is, the ESAO supplied as Joncryl® ADR 4300 and the polystyrene-glycidyl methacrylate random copolymer (PS-GMA) as Xibond™ 920.

2. Experimental.

2.1. Materials

Fully bio-based homopolyamide PA1010 commercial-grade NP BioPA1010-201 was provided, in pellets form, by NaturePlast (Iffs, France). According to the manufactured information, this PA1010 is a medium-viscosity injection-grade with a density of $1.05 \text{ g}\cdot\text{cm}^{-3}$ and a viscosity number (VN) of about $160 \text{ cm}^3\cdot\text{g}^{-1}$.

CF was supplied by BCK Barnacork S.L (Barcelona, Spain). The fibers present an average length of $5.2 \pm 2.2 \text{ mm}$ and an average diameter of $205 \pm 98 \text{ }\mu\text{m}$. **Figure 1** shows the optical images of CFs used to measure the average diameter and length together with the corresponding histogram profiles. As can be seen, both distributions have a high standard deviation due to the high heterogeneity of CFs.

MLO was VEOMER LIN supplied from Vandeputte (Mouscron, Belgium). It has a viscosity of 1000 cP at 20°C and an acid value comprised in the 105 and 130 mg KOH·g⁻¹ range. ELO was supplied by Traquisa S.L. (Barcelona, Spain). This has a molecular weight (M_w) of about $1037 \text{ g}\cdot\text{mol}^{-1}$, a density between 1.05 and $1.06 \text{ g}\cdot\text{cm}^{-3}$ at

20 °C, and a viscosity of 8–11 p (at 25 °C). Regarding the petroleum-derived compatibilizers, a low-functionality ESAO, Joncryl® ADR 4300, was obtained from BASF S.A. (Barcelona, Spain) in flakes form. Its average M_w is 5500 g·mol⁻¹, and it shows a glass transition temperature (T_g) of 56 °C. Its epoxy equivalent weight (EEW) is 445 g mol⁻¹, and its functionality (f) is ≤ 5 . PS-GMA was Xibond™ 920 kindly supplied by Polyscope (Geleen, The Netherlands). Its M_w is 50000 g·mol⁻¹, its T_g is 95 °C, and its glycidyl methacrylate (GMA) content is 20 m/m%. **Figure 2** shows the chemical structure of all four compatibilizers used in this study.

2.2. Manufacturing of PA1010/CF composites

In a previous stage, to remove any residual moisture which could affect processing, both PA1010 and coir fiber were dried at 60 °C for 45 h in a dehumidifying dryer MDEO from Industrial Marsé (Barcelona, Spain). After this stage, an initial mechanical mixing of the appropriate components, which are summarized in **Table 1**, was obtained in a zipper bag and then the different mixtures were subjected to a compounding process in a twin-screw co-rotating extruder from Construcciones Mecánicas DUPRA S.L. (Alicante, Spain). The screws had a diameter of 25 mm and a length (L) to diameter (D) ratio (L/D) of 24. The mixtures were subjected to the following temperature program during extrusion from the hopper to the extrusion die: 200 °C – 210 °C – 215 °C and 220 °C. **Figure 3** shows an image of the injection-molded composites together with the starting materials.

The compounded pellets corresponding to the different formulations were further processed by injection molding in a Meteor 270/75 from Mateu & Solé (Barcelona, Spain). The selected temperature profile in the injection molding machine was the following (from the feeding zone to the injection nozzle): 205 °C, 205 °C, 210 °C, and 210 °C. A clamping force of 75 tons was applied. Concerning the cavity

filling and cooling times, they were set to 1 s and 10 s, respectively. Standard samples for characterization were obtained by injection molding.

2.3. Mechanical characterization.

Different mechanical tests were used to assess the compatibilization effectiveness of different compatibilizers mentioned above on mechanical performance. In particular, tensile, hardness, and Charpy impact tests were carried out. Concerning tensile tests, they were carried out in a universal test machine LLOYD 30 K (Hampshire, United Kingdom) on dog bone-shaped samples following the recommendations of ISO 527-1:2012. The selected load cell was 30 kN, and the cross-head speed was set to 1 mm min⁻¹. Hardness values were measured by using a hardness tester durometer Brevetti AFFRI – Model ART. 13 durometer, (Induno Olona, Italy). The measurements were taken at 10 different points on injection-molded samples with a size of 80x10x4 mm³, according to ISO 868:2003. Toughness was also assessed on rectangular samples with dimensions 80x10x4 mm³ by the Charpy impact test with a 6J pendulum from Metrotec S.A. (San Sebastián, Spain) on notched samples (V-notch with a radius of 0.25 mm), following the specifications of ISO 179-1:2010. To obtain reliable results, all tests were carried out on at least 6 different samples of each material, and the main parameters from each mechanical test were obtained and averaged.

2.4. Thermal characterization.

Thermal characterizations were carried out by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). To evaluate the main thermal transitions of PA1010/CFs composites, a differential scanning calorimeter (DSC) Q200 from TA Instruments (Schwerzenbach, Switzerland) was used. Samples (average weight of 5 – 8 mg) were placed into standard sealed aluminum crucibles characterized by a total volume of 40 μ L. Then, samples were subjected to a dynamic thermal program scheduled in three different stages: a first heating step from 25 °C to 150 °C was

followed by a cooling down to 25 °C and then a second heating step was programmed from 25 °C up to 350 °C. The heating/cooling rate was set to 10 °C min⁻¹, and the selected atmosphere was nitrogen at a constant flow rate of 66 mL min⁻¹. In addition to the melt peak temperature (T_m), additional information about the melting enthalpy (ΔH_m) was collected to calculate the degree of crystallinity (χ_c) as indicated in **Equation 1**.

$$\chi_c(\%) = \left[\frac{\Delta H_m}{\Delta H_m^0 \cdot (1-w)} \right] \cdot 100 \quad \text{Equation 1}$$

Where ΔH_m (J·g⁻¹) corresponds to the melting enthalpy of the crystalline fraction in PA1010 and ΔH_m^0 (J·g⁻¹) stands for the melting enthalpy of a theoretically fully crystalline PA1010, that is, 244 J·g⁻¹ [63]. Finally, the term $1-w$ represents the weight fraction of PA1010 where w represents the weight fraction of all components except PA1010.

In addition to thermal transitions, thermal stability was assessed by TGA in a Seiko Exstar 6300 analyzer (Tokyo, Japan). Samples amounts of 5–7 mg were placed in standard alumina crucibles of 70 μ L and subjected to a dynamic heating program from 30 °C to 700 °C at a heating rate of 10 °C·min⁻¹ under nitrogen atmosphere at 60 mL/min.

2.5. Thermomechanical characterization.

Complementary to other thermal analysis techniques, dynamical thermomechanical properties were obtained in a dynamic mechanical thermal analyzer (DMTA) DMA1 from Mettler-Toledo (Schwerzenbach, Switzerland). The selected setup was dynamic flexural load in a single cantilever. Rectangular samples with dimensions of 20×6×2.7 mm³ were subjected to dynamic temperature heating program from -150 °C up to 150 °C at a constant heating rate of 2 °C·min⁻¹. The maximum flexural deformation in the cantilever was set to 10 μ m, and the selected frequency was 1 Hz.

2.6. Morphology characterization.

The morphology of the developed composites was studied by field emission electron microscopy (FESEM) in a microscope ZEISS SUPRA 25 from Oxford Instruments (Abingdon, UK) working at an acceleration voltage of 2 kV. To provide electrical conducting properties to fractured samples, these were covered with a thin platinum layer in a high vacuum sputter coater EM MED20 from Leica Microsystems (Milton Keynes, United Kingdom).

2.7. Water uptake characterization.

To evaluate the effect of natural fiber on the water uptake, injection-molded samples sizing 4x10x80 mm³ were immersed in distilled water at 24 ± 1 °C. The evolution of water absorption was followed for a whole period of 14 weeks. Samples were extracted weekly. The residual water was removed with a dry cloth, and then, samples were weighed on an analytical balance with a precision of ± 0.1 mg to give high accuracy. All measurements were performed in triplicate to obtain reliable data. The weight of the samples during the water uptake period was obtained using an ABT 220-SDM analytical balance from Kern&Sohn (Balingen-Frommern, Germany). The analysis was carried out according to ISO 62: 2008. To obtain the diffusion coefficient (D), the ISO 62:2008 allows the application of Fick's first law. The diffusion coefficient can be estimated according to **Equation 2**. In this sense, $W_t/W_s \leq 0.5$ is a linear plot of $\Delta m_t = f(\sqrt{t})$ that allows calculation of the diffusion coefficient from the slope (θ) [64, 65].

$$\frac{W_t}{W_s} = \frac{4}{d} \left(\frac{D t}{\pi} \right)^{\frac{1}{2}} \quad \text{Equation 2}$$

Where d is the initial thickness of the sample, D is the diffusion coefficient, and W_s is the saturation weight in the linear region. The slope is calculated from the plot representation of W_t/W_s versus $t^{1/2}$. To obtain the diffusion coefficient, **Equation 3** was used [66]:

$$D = 0.0625 \pi d^2 \theta^2 \quad \text{Equation 3}$$

where θ is the slope, and d is the initial thickness of the sample. Since this expression is only correct for a one-dimensional shape such as a film, new **Equation 4** considers different corrections to make this expression useful for three-dimensional samples:

$$D_c = D \left(1 + \frac{d}{h} + \frac{d}{w} \right)^{-2} \quad \text{Equation 4}$$

where D_c is the corrected diffusion coefficient (geometry), h is the total length, w is the width, and d is the sample thickness. The use of this equation assumes that the velocity of diffusion is equal in all directions. [65-67].

3. Results and discussion

3.1. Mechanical characterization of PA1010/CFs composites

Table 2 shows the mechanical results of PA1010/coir fiber composite with different compatibilizers, obtained from tensile tests. Concerning neat PA1010, the tensile modulus, E and σ_{\max} were 636 MPa and 41.2 MPa, respectively. About the elongation, it is important to remark two different values, both the elongation at break (ϵ_b) of 256% [68] and the elongation at the maximum tensile stress σ_{\max} , which was only 11.8%. These mechanical properties are typical of a strong and tough engineered material. It is important to note that the incorporation of a CF increases the tensile modulus. It is important to bear in mind that the tensile modulus represents the stress to the strain ratio in the linear region of a typical stress-strain diagram. As one can see in Table 2, the decrease in elongation at break is much more pronounced than the decrease in the tensile strength. Since the elongation is in the denominator of the tensile modulus definition ($E = \sigma / \epsilon$)_{linear region}, the overall result is an increase in the tensile modulus and, subsequently, the material becomes more rigid. In particular, reinforcement with 20 wt% CF yielded an increase in the tensile modulus of about two

times the value of neat PA1010, that is, 940 MPa. On the contrary, the maximum tensile strength was remarkably reduced down to values of 20.2 MPa, which represents about half the value of neat PA1010. Similarly, the elongation at break is coincident with the elongation at the maximum stress since the material lost certain cohesion. This poor material's cohesion is responsible for this dramatic decrease in elongation down to values of 3.1%. Similar findings were reported by L.H. Staffa *et al.* [69] with PP composites containing 30 wt% coir fiber with a decrease in elongation at break from >350% down to 3.69% with slight improvements achieved by using different compatibilizers. This is the typical behavior of fibers with weak interactions with the polymer matrix, as reported by other authors. For instance, Rozman *et al.* [70] reported that the addition of coir fiber to PP provided a dramatic reduction of elongation at break and, subsequently, an increase in Young's modulus of the obtained composites. This behavior has been reported in a wide variety of polymer-fiber composites with poor or absence of fiber-polymer interactions and is much more pronounced in polymers characterized by high elongation at break. Following this, Carbonell-Verdú *et al.* [71] reported this effect on bio-based high-density polyethylene (bio-HDPE) reinforced with 20 wt% slate fiber (SF). The modulus was increased from 373 MPa, for neat bio-HDPE, up to 1483 MPa in the composite containing 20 wt% SF. On the contrary, the elongation at break decreased dramatically from 520% for neat bio-HDPE down to 18.7% for the above-mentioned uncompatibilized composites. For this reason, it is necessary to improve polymer-fiber interactions. In this work, different bio-based and petroleum-derived compatibilizers were used with the main aim of overcoming this drawback related to poor polymer-fiber interactions.

The addition of 7.5 phr of MLO or ELO provided interesting changes in the mechanical behavior of PA1010/CFs composites. Both chemically modified vegetable oils contributed to slightly increasing tensile strength (σ_{max}) and in particular, an increase in the elongation at break (ϵ_b) of PA1010/CF composites. Following the definition of the tensile modulus, as the ϵ_b increases, the modulus decreases. The

addition of MLO and ELO led to E and σ_{\max} values of about 780 MPa and 22–23 MPa, respectively, which are improved properties compared to uncompatibilized PA1010/CF composite. It is worthy to note that both modified vegetable oils can provide different overlapping phenomena to binary systems such as chain extension, compatibilization, plasticization, branching, and, in some cases, some cross-linking. It is worthy to note the significant increase in ε_b that ELO can provide to the PA1010/CFs composites with an ε_b value of 7.1%, which is more than double the value of the uncompatibilized composite. This phenomenon could be related to two mechanisms, one related to the plasticization ELO can provide and, on the other hand, to an increase in polymer-fiber interaction that, in turn, is responsible for an increase in material's cohesion and this has a positive effect on cohesion-related properties such as tensile strength and elongation. Balart *et al.* [55] reported that the incorporation of 7.5% ELO into a PLA composite with hazelnut waste powder provided some plasticization and compatibilization between the polymer and the lignocellulosic filler. This was particularly observed by observation of completely embedded hazelnut particles in the PLA matrix after the ELO addition. As indicated previously, modified vegetable oils can provide some plasticization to the matrix [72, 73], but the increase in tensile strength suggests an improvement of the compatibility between the fiber and the surrounding biopolymer matrix.

Concerning the petroleum-derived compatibilizers, their effects were not so remarkable as those provided by the chemically modified vegetable oils. The amount of petroleum-derived compatibilizers was lower than that of MLO or ELO but previous works have demonstrated that vegetable oil contents of 7.5–10 phr provided optimum balanced properties [58, 73, 74], while the typical content of these two petroleum-derived compatibilizers is lower than 3 phr [59, 75]. Both petroleum-derived compatibilizers offered a slight increase in the tensile modulus, which is directly related to the compatibilizing effect as the elongation at break is also slightly increased up to values of 3.6%, but, in general, their effect on the tensile properties was not remarkable. As

shown in **Figure 2**, both petroleum-derived compatibilizers are copolymers of poly(styrene) which is not miscible with PA, nevertheless their functionality, *i.e.* glycidyl methacrylate (GMA) is highly reactive with condensation polymers such as polyamides and cellulosic fillers through the reaction with terminal hydroxyl groups in polyamide and hydroxyl groups in CF (mainly in hemicellulose and cellulose) as previously reported[57].

About the Shore D hardness values, all the tested PA1010/CFs composites, except that for the MLO-containing composite, offered similar values. Shore D hardness values increased from 70.2, for neat PA1010, up to values around 74-75 for all the composite pieces. This increase is directly related to the intrinsic hardness of the lignocellulosic components of CF. In the case of the PA1010/CG+MLO, the Shore D hardness was slightly lower than other composites, and this could be due to the plasticization that MLO can provide. Regarding impact strength, the trend is identical to the elongation at break as the impact strength is highly sensitive to the material's cohesion. Neat PA1010 is a tough material with a relatively high impact strength of 11.3 kJ m^{-2} , even on notched samples. It is worthy to note that the impact strength is directly related to two mechanical parameters, that is, the applied stress and the deformation before failure. As it has been described previously, the elongation at break for neat PA1010 was very high with relatively high tensile strength. Subsequently, the impact strength was also high. Identically to the elongation at break, the addition of 20 wt% CFs promoted a dramatic decrease in toughness down to values of $1.7 \text{ kJ}\cdot\text{m}^{-2}$. The effect of both linseed oil derived compatibilizers, resulted in improved toughness up to values of $2.4 \text{ kJ}\cdot\text{m}^{-2}$ and $2.0 \text{ kJ}\cdot\text{m}^{-2}$ for MLO and ELO-compatible composites, respectively. This is directly related somewhat to the compatibilization attained, as observed with the elongation at break. Regarding ESAO and PS-GMA, both produced similar results with no remarkable improvement in toughness. Nevertheless, both petroleum-derived compatibilizers resulted in stiffer materials.

3.2. Morphology of PA1010/CF composites

Figure 4 shows the morphology of an individual CF. The average diameter calculated from the optical images was $205 \pm 98 \mu\text{m}$, and **Figure 4a** shows a short fiber with a diameter of $78 \mu\text{m}$, but it is worthy to note that typical CFs are quite heterogeneous with diameters changing from 90 to $400 \mu\text{m}$. The cross-section of this fiber is shown in **Figure 4b**, where the typical tubular formations (see white circle) can be observed [76]. This structure is responsible for the lightness of the CFs together with its excellent thermal insulation properties as air is located into these tubular formations, thus contributing to good insulation properties [77]. Indeed, CFs have not only been used in polymer matrices but also in concrete for construction and building materials due to the excellent insulating properties [78]. CFs are among the thickest fibers commercially available. Depending on the variety, single fibers can reach average diameters above $200 \mu\text{m}$ [79].

Figure 5 gathers the FESEM images corresponding to the fracture surfaces of the injection-molded pieces after the impact test. Regarding neat PA1010, the fracture is typical from an impact test on notched samples. As the material is rather tough, the surface was very rough, and this effect is visible by the presence of crack fronts (white arrows) and the coalescence of microcracks during crack growth (see white rectangles in **Figure 5a**). Despite this material is very ductile and allows substantial plastic deformation, the conditions of the impact test (notched samples) promoted stress concentration with lower energy absorption, and, subsequently, with lower evident deformation. Nevertheless, the high roughness observed is representative for deformation as completely brittle materials typically show a very smooth surface. Uncompatibilized composites with 20 wt% CFs (**Figure 5b**) showed a remarkably different surface morphology after the failure by the impact. The relative high diameter of CFs, with an average diameter of $95 \mu\text{m}$ but with some bigger fibers of up to $200 \mu\text{m}$, promoted a stress concentration effect due to the loss of material continuity. This led to

a more brittle fracture surface (see the white arrows indicating micro-crack formation and a smoother fracture surface). Even after the achievement of high polymer-filler interaction, one assumes that the short fiber and its high diameter did not contribute to improving mechanical properties. K. Mustapha *et al.* [80] established a threshold at about 10 wt% short CFs on a polyolefin matrix. Above this threshold content value, mechanical properties suffered a dramatic decrease. L.H. Staffa *et al.* [69] also demonstrated the poor compatibility of composites based on a PP matrix and 30 wt% CFs. A SEM analysis reported some evidence of pulled-out fibers for the uncompatibilized PP/CFs composites. It was also observed that the use of PP-*g*-MA copolymer improved the interaction, but mechanical properties were only slightly improved due to the high content of short CFs in these composites.

The interaction between the lignocellulosic fibers and the PA1010 matrix is not deficient since PAs are more polar than PP, LDPE or HDPE, widely used as matrices in these composites. As it can be seen in **Figure 5b**, the uncompatibilized PA1010/CFs presented some embedded fibers in the cross-section direction (circles) while some longitudinal fibers were pulled-out (see the white elliptical shape). The addition of compatibilizers resulted in a better fiber dispersion with a clear cross-section of CFs that were rather good embedded in the PA1010 matrix. One can expect somewhat improved interactions for all four compatibilizers since the mechanical properties were slightly improved, as shown in previous **Table 2**, specifically for the bio-based compatibilizers derived from linseed oil, although both petroleum-derived compatibilizers also provided improved stiffness. As it can be seen in **Figure 5c** and **Figure 5d**, the normal/perpendicular fibers appeared well embedded in the PA1010 matrix (white circles) while some debonding occurred with CFs in the longitudinal direction (parallel to fracture surface).

In the case of ELO (**Figure 5d**), CFs were fully embedded in the PA1010 matrix. The above-reported brittleness is also evident by the presence of micro-crack formation

(white arrows). Similar behavior can be observed for both petroleum-derived compatibilizers. **Figure 5e**, shows the compatibilized composite with EASO and the morphology of its fracture surface was similar, with perfectly embedded perpendicular CFs (white circles) and some debonded CFs in the longitudinal direction (white elliptical shapes). Moreover, an identical morphology is detectable by using PS-GMA as a compatibilizer, thus indicating its effectiveness (**Figure 5f**).

Figure 6 shows zoomed FESEM images showing the fracture surface of a perpendicular CF and its surrounding area. These zoomed images allow precise observation of the individual microfibrils. The diameter of these hollow microfibrils can be as high as 12 μm , as reported by F.Z. Semlali Aouragh Hassani *et al.*[79]. The diameters of the CFs used in this study were about 5 to 10 μm , and it plays an essential role in composites. As can be seen for the uncompatibilized PA1010/CF composites in **Figure 6a**) the lumen of these microfibrils was full as the biopolymer chains could enter these macro-tubes. Nevertheless, the morphology and shape of the individual microfibrillar tubes were lost during processing. The fiber-polymer interaction is considered to be good enough as a tiny gap was observed. The fiber seems to be fully embedded in the PA1010 matrix. Both vegetable oil-derived compatibilizers produced different morphologies. As expected, these molecules increased the free volume of the PA1010 matrix due to their plasticizing effect and, thus, the polymer chains could enter the tubular microfibrils more easily. It can be noted in **Figure 6b** and **Figure 6c** that the tubular shape of the microfibrils was not altered after processing these composites due to the plasticization provided by both MLO and ELO. In the case of MLO, it is possible to see some biopolymer tubes out of the tubular microfibrils that indicate deformation during the fracture test. Concerning the petroleum-derived compatibilizers, both ESAO (**Figure 6d**) and PS-GMA (**Figure 6e**) broke up the tubular morphology, but their compatibilizing effect was evident as the CFs were wholly embedded in the biopolymer matrix without any gap between the fibers and the surrounding PA1010 matrix. Similar findings have been reported by L. Quiles Carrillo

et al. [62] in PLA/almond shell flour (ASF) green composites with a decreased the gap between the dispersed particles and the surrounding biopolymer matrix.

3.3. Thermal properties of PA1010/CF composites

Figure 7 gathers a comparative plot of the DSC thermograms of the PA1010/CFs composites processed with the different compatibilizers, while **Table 3** shows a summary of the main thermal parameters obtained by DSC for these composites.

Neat PA1010 was characterized by melting temperature (T_m) of 203.9 °C and a degree of crystallinity (χ_c) of approximately 21%. The addition of CFs did not provide any relevant change in T_m , showing a similar value of 203.4 °C whereas the crystallinity was not profoundly affected, with a χ_c value of 20.7%. It has been widely reported the nucleating effect of lignocellulosic fillers, such as hemp or rice straw fibers, on different polymers and biopolymers with the subsequent increase in the crystallinity [81, 82]. However, it has been previously observed that CF itself shows a low nucleating effect in other natural filled composites [83, 84].

With the addition of MLO and ELO, crystallinity in the green composites remarkably increased up to values of about 28%. This increase can be related to two different phenomena. On the one hand, the increased polymer-particle interactions could favor the nucleating effect of the lignocellulosic CFs and, on the other hand, both chemically modified vegetable oils provided a plasticization that promoted chain mobility, thus favoring folding of polymer chains into a packed structure [28]. However, the effect of MLO and ELO on the T_m values, inducing a decrease by 1 °C and 2 °C, respectively, was neglected as it is within the typical deviation range. With the incorporation of both petroleum-derived compatibilizers, that is, and, a slight increase in T_m was observed for the composites containing PS-GMA while it remains almost constant for the ESAO-containing composites. About the χ_c values, the crystallinity of

PA1010/CF composites, both ESAO and PS-GMA induced a decrease down to values around 17%. This phenomenon is ascribed to the characteristic chain-extension effect of these additives, which could restrict the formation of more packed and ordered regions. According to this, Qian *et al.* [85] reported that the addition of chain extenders, particularly epoxides, led to an apparent decrease in crystallinity in PA1010. A similar effect was also observed in our study for PLA/ASF composites compatibilized by ESAO [62].

Regarding degradation at high temperatures, that is, thermal decomposition, **Figure 8** shows a comparative plot of the characteristic TGA curve of neat PA1010 and the uncompatibilized and compatibilized PA1010/CFs composites as well as the raw CFs. The most relevant information obtained from TGA is gathered in **Table 4**. In particular, the temperature at which a 5 wt% mass loss occurs ($T_{5\%}$) and the maximum degradation rate temperature (T_{max}) are shown. CF is a lignocellulosic material, and it shows relatively low thermal stability compared to PA1010. In the graph, it can be observed an initial weight loss in the temperature range of 80–120 °C that is attributable to the residual water removal. At moderate temperatures above 220 -230 °C, degradation of hemicelluloses occurred, and this has a negative effect on the overall thermal stability, as both lignin and cellulose are much more thermally stable. In this sense, a degradation temperature so close to the melting temperature of the PA1010 could generate a slight degradation in the CF (less than 9 wt% due to hemicellulose initial degradation), which could result in a change of colour to a darker shade. As seen in **Figure 8**, hemicellulose degradation starts above 220 – 230 °C. In this context, it can be seen how the injection-molded samples (**Figure 3**) have a darker colour than the original colour of coconut fibers due to potential slight hemicellulose degradation. Moreover, another interesting phenomenon is usually observed when natural fibers are embedded into a polymer matrix in the melt state. Dominici *et al.* [86] reported a clear darkening effect by using yellowish coffee silver skin into a biobased polyethylene, using processing temperatures below 200 °C, and even in those mild temperature

conditions, a darkening effect was observed. Other authors have reported similar behaviour with lignocellulosic loads and a slight darkening effect [87]. Liminana et al.[88] also reported similar findings with almond shell flour that got darker when injection moulded with poly(butylene succinate) – PBS using a low temperature processing profile. Some authors have reported the relevance of phase change materials (PCMs) to provide thermal inertia and thermal stabilization effects on polymer blends and composites by using the involved heat absorption during the phase change[89, 90]. Although the thermal degradation of lignin starts at similar temperatures of hemicellulose, that is, at about 250 °C, its degradation rate is remarkably lower and is extended up to temperatures above 500 °C. The residual mass of CF was close to 30%, which is a relatively high value, as reported by Rosa *et al.* [91]. Rosa *et al.* [92] also reported similar results regarding coir husk fibers. This was characterized by a first weight loss up to 120 °C due to water evaporation while lignin removal was reflected in the amount of final mass, measured in the temperature range comprised between 350 °C and 500 °C. Hemicellulose removal can be observed by the disappearance of the shoulder at 275 °C, as shown by the first derivative of the TGA curves. As expected, the low $T_{5\%}$, which can be considered as the onset degradation, of CF contributed to a decrease in the overall thermal stability of the composites. Whereas neat PA1010 showed remarkable thermal stability and its characteristic $T_{5\%}$ was 420.3 °C, the $T_{5\%}$ value for the uncompatibilized PA1010/CF was remarkably lower, that is, 292.8 °C. Although PA1010 can withstand higher temperatures, the thermal stability of its composites with CF was compromised due to CF degrades at relatively low temperatures.

As it can be seen in **Table 4**, the T_{max} value of PA1010 was 466.6 °C. It is also worthy to note that the residual moisture in the neat PA1010 was very low since the weight loss in the 100–150 °C range was neglected. Similar thermal degradation profiles for different bioPAs have been described in the literature [68, 93, 94]. As PA1010 was the main component of the composites, the T_{max} values remained almost

constant, showing values of around 465 °C, independently of the compatibilizer used. The most visible effect of the different compatibilizers was observed by a slight increase in the $T_{5\%}$ values, specifically for ELO and PS-GMA, which provided higher values of 12 °C and 17 °C, respectively. This slight increase in thermal stability can be directly related to increased fiber-polymer interactions, which gives some evidence of partial compatibilization. In this regard, Chieng *et al.* [95] have reported that epoxidized vegetable oils can establish covalent links between lignocellulosic fillers and the PLA matrix. As indicated previously, this enhanced chemical interactions successfully act as a physical barrier that obstructs the removal of volatile products during decomposition. In our previous work [62], it was suggested that the increased thermal stability of the PLA-based composites was achieved by favoring the polymer-particle interactions by processing with ESAO. Concerning the residual mass, all the PA1010/CF composites showed residues of around 6–7%, mainly due to the presence of CF.

3.4. Thermomechanical properties of PA1010/CF composites

Figure 9 shows the evolution of DMTA curves of the neat PA1010 and the PA1010/CFs with different compatibilizers. In particular, **Table 5** summarizes some of the thermomechanical properties obtained from these curves. In this sense, in **Figure 9a** one can observe the evolution of the storage modulus (E') as a function of temperature. The dynamic thermomechanical behavior of PA1010 was characterized by an E' value in the of 1250–800 MPa range from -100 °C to 25 °C. Similar results were shown by other authors [68]. Above 50 °C, the storage modulus dropped down to a value of 200 MPa. This reduction is directly related to the alpha α -transition of PA1010 in which the amorphous phase of the biopolymer changes from the glassy to a rubbery state.

The addition of CF produced an increase in E' , below and above the α -transition. For the uncompatibilized PA1010/CF composite, a value of 1515 MPa at -100 °C was obtained, which represents 20% more than the neat biopolymer. In general, the addition of synthetic and natural fibers in a polymer matrix increases the stiffness

of the material. In this sense, other authors have shown that the incorporation of short natural fibers increases in an obvious way the values of E' of the obtained composites [96-98]. Concerning the presence of compatibilizers, it is worthy to note they provide increased rigidity on composites at temperatures below the glass transition region.

This increase observed in stiffness may be due to a direct improvement in the compatibility between the matrix and the dispersed fibers. Although the vegetable oil-derived compatibilizers provided additional plasticization, as mentioned above, the interaction of the glycidyl and maleic anhydride groups with both PA1010 and CFs led to improved cohesion between both components of the composite. This phenomenon increases the stiffness of the composite at low temperatures, significantly improving their potential uses for technical applications. In all cases, the improvement in stiffness was very similar since it changed from a value of 1500 MPa, for the uncompatibilized PA1010/CFs composite, up to about 1600 MPa after the addition of the different compatibilizers. In this sense, Michaz-Ul Haque *et al.* [99] reported that the addition of glycidyl methacrylate (GMA) to ethylene-vinyl acetate (EVA) composite filled with cellulose fibers increased the stiffness and the effectiveness of the fillers based on the E' values of the obtained composites.

About the dynamic damping factor ($\tan \delta$), it represents the ratio between the lost energy (E'') and the stored energy (E'), so that low values indicate low energy dissipation. **Figure 9b** shows the evolution of $\tan \delta$ as a function of temperature. It can be observed that the $\tan \delta$ peak for neat PA1010 was 52.6 °C. This value is representative of the α -transition of the biopolymer, which relates to its T_g . Moreover, two additional peaks were located at temperatures of -65 °C and -130 °C, which are attributed to the β - and γ -relaxations of PA1010. For instance, Pagacz *et al.* [100] reported that the peak around -130 °C reflects the γ -relaxation, which has been attributed to motions of methylene groups [101]. The peak located at about -80 °C represents the β -relaxation, typically attributed to non-hydrogen-bonded amide groups

[101] and, more specifically, to water-bound on carbonyl groups. Finally, the prominent peak above 50 °C is typically attributed to the dynamic glass transition of this type of polyamide [102, 103]. One can observe that the $\tan \delta$ peak of neat PA1010 was remarkably higher than the peaks seen for the PA1010/CFs composites. This observation suggests that all composites showed a higher elastic behavior, that is they performed as stiffer materials with higher E' values, and this contributes to lowering $\tan \delta$. In particular, the PA1010/CF + ELO showed the lowest $\tan \delta$ value, in terms of temperature value and peak intensity, thus indicating improved fiber-matrix interactions as outlined previously.

The addition of CF to PA1010 reduced its T_g by 7.5 °C, which could be related to the presence of some inherent low- M_w components in the fibers. The compatibilizing or coupling effect of MLO, ELO, and PS-GMA was evidenced by an increase in T_g up to values of 48.1 °C, 50.1 °C, and 54.2 °C, respectively, which are significantly higher than the T_g value for the uncompatibilized PA1010/CFs composite, that is, 45.1 °C. This increase in T_g is representative of improved fiber-matrix interaction as these interactions restrict the biopolymer chains' mobility and lead to an increased T_g . Other authors have reported similar results for PAs with other lignocellulosic fibers [68].

3.5. Evolution of the water uptake and water diffusion process

Since the here-developed PA1010/CFs composites contain a substantial amount of a lignocellulosic filler, they are characterized by extremely high hydrophilic behavior. Therefore, the study of their water absorption or uptake becomes indeed crucial for real applications. The study of the water uptake and diffusion of the PA1010/CFs composites is shown in **Figure 10**. Water absorption is a diffusion-controlled process and, subsequently, it follows Fick's Law. This fact can be observed in **Figure 10a**, which represents the water uptake during the first 24 h, and it is useful to calculate D and D_c that can be seen in **Table 6**. One can observe that the D_c value indicates the hydrophilic nature of the developed materials. Neat PA1010 is a polar polymer, and it

tends to absorb water as similar to other PAs (see **Figure 10b**). It resulted in an asymptotic value of about 1.69 wt%, which is remarkably higher compared to non-polar polymers such as polyolefins with values close to 0.1–0.3 wt% [104, 105]. Besides, it is essential to remark that water absorption in polymer composites also highly depends on the nature of the lignocellulosic filler [106].

The values of D_c were calculated considering the saturation mass. This resulted in a D_c value of $0.54 \cdot 10^{-8} \text{ cm}^2 \cdot \text{s}^{-1}$ for neat PA1010. The addition of a lignocellulosic filler, highly hydrophilic, promoted an increase in D_c up to values of $1.15 \cdot 10^{-8} \text{ cm}^2 \cdot \text{s}^{-1}$. In general, all four compatibilizers provided slightly lower D_c values, close to $1 \cdot 10^{-8} \text{ cm}^2 \cdot \text{s}^{-1}$, which indicates that in a first approach, at short immersion times, all four compatibilizers reduce the water diffusion rate. Nevertheless, as can be seen in **Figure 10b**, the saturation values for the uncompatibilized PA1010/CFs composite were around 2.18 wt% while all the compatibilized PA1010/CFs composites tended to stabilize water absorption at an average value of 2.68 wt%. This result indicates that, despite the reduction attained in water uptake observed for early immersion stages, the use of compatibilizers promotes the absorption of water in composite in the long term. Our previous work [62] also demonstrated the effect of different compatibilizers on the water uptake of PLA composites filled with 25 wt% ASF. It was observed a very low water saturation for neat PLA of 0.7 wt% after 14 weeks, while the uncompatibilized composite containing 25 wt% ASF reached water saturation at 4.8 wt%. This points out that the water uptake impairment of the PA1010/CFs composites is lower. This fact can be related to the entrance of the compatibilized polymer into the tubular-shaped microfibrils, which leads to exposure of the lignocellulosic walls that, in turn, contribute to slightly higher water uptake values for long periods.

On the other hand, the presence of compatibilizers, such as MLO or ELO, increases the biopolymer free volume, and this phenomenon enhances new paths for water from entering into the composite structure. Balart et al. [107] reported similar findings for PLA/hazelnut shell flour (HSF) composites. In particular, the uncompatibilized

composite with 20 wt% HSF showed an asymptotic water absorption of 3.5 wt% after 14 weeks, while these composites when compatibilized with different amounts of ELO reached an asymptotic water absorption of almost 4.5 wt%.

4. Conclusions.

This work demonstrated the feasibility of manufacturing highly environmentally friendly and wood-plastic composites with high performance based on a fully bio-based matrix of PA1010 and a reinforcement phase of 20 wt% CFs. It is important to bear in mind, that environmental concerns are related to both the origin (petroleum or natural resources) and the end-of-life (biodegradable or not). PA1010/CF composites include these concerns as the polymer matrix is fully obtained from renewable resources and the reinforcement/filler is a by-product of the coconut industry in the form of fiber. These composites were manufactured by melt compounding followed by injection molding to develop materials with wood appearance. Since the wood color palette is very wide, including yellow, orange, red woods, but also brown and dark woods, the herein developed materials show similar appearance to dark woods such as ebony and wenge wood. Mechanical characterization revealed the remarkable effect that CF delivers on the final properties of composites. In particular, the elongation at break is dramatically reduced from 256%, for neat PA1010, down to values of 3.1%, for the uncompatibilized composite. The incorporation of multi-functionalized vegetable oil derived compatibilizers, namely MLO and ELO, resulted in a noticeable improvement of all the mechanical properties, including toughness. The two tested commercial petroleum-derived compatibilizers based on GMA multi-functionality, that is, Joncryl® 4300 and Xibond™ 920, also provided some enhancement of the composite performance. The water uptake remained at relatively low values, thus leading to materials that could replace wood in different industrial applications.

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Figure captions

Figure 1. Optical images of coconut fiber (CFs) (a & c) at different magnifications and statistical distribution of the length (b) and diameter (d).

Figure 2. Schematic representation of the chemical structure of a) bio-based and b) petroleum-derived functional compounds used.

Figure 3. Image of the starting materials polyamide 1010 (PA1010) and coconut fibers (CFs) and of the obtained pieces after melt compounding and injection molding.

Figure 4. Morphology of a single coconut fiber (CF) obtained by field emission electron microscopy (FESEM): a) longitudinal (axis) direction taken at 250x and b) cross-section taken at 1000x. Scale markers of 10 μm .

Figure 5. Field emission electron microscopy (FESEM) images taken at 100x, showing the morphology of fractured images from impact tests corresponding to: a) neat polyamide 1010 (PA1010); b) PA1010/coconut fibers (CFs); c) PA1010/CF + maleinized linseed oil (MLO); d) PA1010/CF+ epoxidized linseed oil (ELO); e) PA1010/CF + epoxy-based styrene–acrylic oligomer (ESAO); f) PA1010/CF + polystyrene-glycidyl methacrylate random copolymer (PS-GMA).

Figure 6. Field emission electron microscopy (FESEM) images taken at 1000x, showing the morphology of the cross-section of the embedded coconut fibers (CFs) in polyamide 1010 (PA1010) corresponding to: a) PA1010/CF; b) PA1010/CF + maleinized linseed oil (MLO); c) PA1010/CF + epoxidized linseed oil (ELO); d) PA1010/CF + epoxy-based styrene–acrylic oligomer (ESAO); d e) PA1010/CF + polystyrene-glycidyl methacrylate random copolymer (PS-GMA).

Figure 7. Differential scanning calorimetry (DSC) thermograms of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styrene–acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA).

Figure 8. a) Thermogravimetric analysis (TGA) curves and b) first derivative (DTG) curves of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styrene–acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA).

Figure 9. Dynamic mechanical thermal analysis (DMTA) curves of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styrene–acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA): a) Storage modulus (E') and b) Dynamic damping factor ($\tan \delta$).

Figure 10. Water uptake of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styrene–acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA): a) Water absorption during the first 24 h; b) Evolution of the water uptake for 14 weeks.

Tables

Table 1. Summary of codes and compositions according to the weight content (wt %) of polyamide 1010 (PA1010) and coconut fibers (CFs) in which maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styren--acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA) were added as parts per hundred resin (phr) of PA1010/CFs composite.

Code	PA1010 (wt%)	CFs (wt%)	MLO (phr)	ELO (phr)	ESAO (phr)	PS-GMA (phr)
PA1010	100	0	0	0	0	0
PA1010/CF	80	20	0	0	0	0
PA1010/CF + MLO	80	20	7.5	0	0	0
PA1010/CF + ELO	80	20	0	7.5	0	0
PA1010/CF + ESAO	80	20	0	0	2.5	0
PA1010/CF + PS- GMA	80	20	0	0	0	2.5

Table 2. Summary of the mechanical properties of the polyamide 1010 (PA1010)/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styren—acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA).

Sample	E (MPa)	σ_{max} (MPa)	ϵ_b (%)	Shore D hardness	Impact strength (kJ m⁻²)
PA1010	636 ± 29	41.2 ± 2.1	256.5 ± 26.8	70.2 ± 1.1	11.3 ± 0.4
PA1010/CF	940 ± 26	20.2 ± 0.6	3.1 ± 0.2	74.2 ± 0.8	1.7 ± 0.3
PA1010/CF + MLO	785 ± 23	22.0 ± 1.0	4.2 ± 0.4	71.6 ± 0.5	2.4 ± 0.3
PA1010/CF + ELO	777 ± 20	23.3 ± 0.6	7.1 ± 0.5	75.0 ± 0.7	2.0 ± 0.1
PA1010/CF + ESAO	941 ± 32	19.2 ± 0.4	3.6 ± 0.2	75.0 ± 0.8	1.6 ± 0.2
PA1010/CF + PS-GMA	884 ± 22	18.9 ± 0.3	3.6 ± 0.3	74.1 ± 0.9	1.4 ± 0.2

Table 3. Main thermal properties of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styren--acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA,. in terms o: melting temperature (T_m), and normalized melting enthalpy (ΔH_m), and degree of crystallinity (χ_c).

Sample	T_m (°C)	ΔH_m (J·g⁻¹)	χ_c (%)
PA1010	203.9 ± 1.5	52.3 ± 1.9	21.4 ± 1.2
PA1010/CF	203.4 ± 1.2	40.4 ± 1.7	20.7 ± 0.8
PA1010/CF + MLO	202.5 ± 0.9	50.3 ± 1.6	27.7 ± 0.7
PA1010/CF + ELO	201.5 ± 1.2	51.9 ± 1.5	28.6 ± 0.8
PA1010/CF + ESAO	204.9 ± 1.1	31.9 ± 1.7	17.7 ± 0.8
PA1010/CF + PS-GMA	203.1 ± 1.1	31.4 ± 1.4	16.5 ± 0.9

Table 4. Main thermal degradation parameters of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styren—acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA) in terms of the temperature required for a mass loss of 5% ($T_{5\%}$), the maximum degradation rate temperature (T_{max}), and residual mass at 700 °C.

Sample	$T_{5\%}$ (°C)	T_{max} (°C)	Residual weight (%)
PA1010	420.3± 0.7	466.6± 1.0	1.1± 0.2
PA1010/CF	292.8± 1.2	463.5± 1.1	7.2± 0.8
PA1010/CF + MLO	290.5± 1.1	461.7± 0.9	7.3± 0.9
PA1010/CF + ELO	304.6± 1.7	469.9± 0.8	7.2± 0.8
PA1010/CF + ESAO	299.3± 1.3	459.7± 1.0	6.8± 0.2
PA1010/CF + PS-GMA	309.5± 1.4	461.3± 0.9	6.9± 0.2

Table 5. Main thermomechanical parameters of polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styren—acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA) in terms of: storage modulus (E') measured at $-100\text{ }^{\circ}\text{C}$ and $100\text{ }^{\circ}\text{C}$ and glass transition temperature (T_g).

Sample	E' (MPa) at $-100\text{ }^{\circ}\text{C}$	E' (MPa) at $100\text{ }^{\circ}\text{C}$	T_g ($^{\circ}\text{C}$)
PA1010	1245 ± 21	155 ± 2	52.6 ± 0.8
PA1010/CF	1515 ± 25	235 ± 3	45.1 ± 1.0
PA1010/CF + MLO	1610 ± 16	215 ± 3	48.1 ± 0.8
PA1010/CF + ELO	1585 ± 20	225 ± 1	50.1 ± 0.9
PA1010/CF + ESAO	1600 ± 17	240 ± 2	44.1 ± 1.1
PA1010/CF + PS-GMA	1610 ± 19	260 ± 2	54.2 ± 0.9

Table 6. Values of the diffusion coefficient (D) and the corrected diffusion coefficient (D_c) for polyamide 1010 (PA1010) and the PA1010/coconut fibers (CFs) composites processed with maleinized linseed oil (MLO), epoxidized linseed oil (ELO), epoxy-based styren--acrylic oligomer (ESAO), and polystyrene-glycidyl methacrylate random copolymer (PS-GMA).

Sample	$D \times 10^8 \text{ (cm}^2 \cdot \text{s}^{-1}\text{)}$	$D_c \times 10^8 \text{ (cm}^2 \cdot \text{s}^{-1}\text{)}$
PA1010	1.13± 0.08	0.54± 0.02
PA1010/CF	2.42± 0.09	1.15± 0.04
PA1010/CF + MLO	2.02± 0.07	0.96± 0.03
PA1010/CF + ELO	2.28± 0.06	1.08± 0.04
PA1010/CF + ESAO	2.17± 0.05	1.03± 0.05
PA1010/CF + PS-GMA	2.08± 0.06	0.99± 0.03