Evaluation of the Tensile Properties of LLDPE/Sisal Composites Produced by 3D Printing

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This study evaluates the use of sisal residues as reinforcement in linear low-density polyethylene (LLDPE) composites produced by extrusion and 3D printing, with or without the compatibilizer Polybond™ 3029. The proposal is justified by the high volume of waste generated by the sisal industry, about 98% of the plant, corresponding to approximately 15 million tons annually worldwide, according to the FAO, and by the intrinsic advantages of plant fibers, such as low cost, low density, and wide availability. Three formulations containing 5% sisal residue (either in natura or treated with NaOH) and 17.5% Polybond™ were prepared and compared to a control consisting of pure LLDPE. Mechanical tests, conducted according to ASTM D638, revealed that the composite with Polybond showed tensile strength similar to that of the pure polymer but superior to the other formulations, indicating better fiber–matrix adhesion. The elastic modulus showed no statistically significant differences, and the elongation at break remained close to that of pure LLDPE. These results indicate that using untreated sisal residue can reduce production costs without compromising the material's ductility. It is concluded that incorporating sisal residues into LLDPE, especially with the addition of Polybond, adds value to an agricultural byproduct and fosters the development of sustainable composites with potential for diverse applications, such as packaging, consumer goods, and lightweight structures, including the possibility of customized production via 3D printing.
Keywords: Composites. Sisal. LLDPE. 3D Printing.

The increasing environmental concerns and the growing demand for sustainable materials have spurred interest in valorizing agricultural residues through the development of composite materials [1].

Currently, vegetal fibers, such as sisal, offer several advantages: low density, low cost, non-abrasiveness, and high modulus of elasticity. More importantly, they are abundant in nature, as they can be obtained through various cultivation methods. Both manufacturing methods and material selection play a crucial role in the development of polymer composites [2].

According to the FAO (Food and Agriculture Organization), the residual biomass after the extraction of sisal fiber accounts for about 98% of the plant, which amounts to approximately 15 million tons of waste per year worldwide [3].

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Incorporating sisal residues as reinforcement into thermoplastics like linear low-density polyethylene (LLDPE) represents a compelling opportunity to create value-added composites with enhanced mechanical performance for various applications such as food packaging, grocery bags, industrial films, and tubes [4].

The processing route of compounding by extrusion followed by additive manufacturing (3D printing) enables precise control over fiber dispersion and part geometry. Recent studies on 3D-printed vegetal fiber composites, such as PLA reinforced with sisal strands have demonstrated notable improvements in tensile properties often outperforming conventional vegetable fiber composites [5].

This suggests that extrusion combined with layer-by-layer deposition can effectively integrate vegetable fibers into thermoplastics, enhancing interfacial bonding and mechanical integrity.

A critical factor in achieving robust tensile properties in such composites is the use of compatibilizers, such as Polybond, to improve adhesion between hydrophilic fibers and hydrophobic polymer matrices [6]. Using

compatibilizers or coupling agents is an effective approach to improving the interface between components [7]. These agents possess functional groups with affinity for both the fiber surface and the polymer matrix. One end of the coupling molecule reacts with groups on the reinforcement surface, while the other bonds with the functional polymer phase.

Polyethylene-grafted maleic anhydride (PE-g-MA) is a commonly used coupling agent to enhance compatibility between plant fibers and polymer matrices, as the maleic anhydride (MA) moiety interacts with the hydroxyl groups present in the fiber structure [7]. Although specific data on Polybond in LLDPE–sisal systems remain limited, literature indicates that compatibilizer-enabled surface modifications (e.g., alkali) substantially boost tensile strength, modulus, and impact resistance in sisal-reinforced composites [8].

Therefore, the addition of Polybond in LLDPE/sisal composites is expected to reinforce the fiber-matrix bond, which translates into superior tensile performance obtained by extrusion followed by 3D printing.

In summary, the combination of sisal residue valorization, extrusion compounding, Polybond compatibilization, and additive manufacturing fosters the development of sustainable LLDPE-based composites. These materials promise enhanced tensile properties and customisable geometries, with broad potential in automotive, consumer goods, and lightweight structural applications.

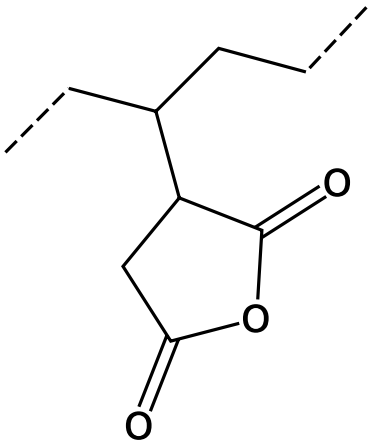
Materials and Methods

Materials

Sisal residue was supplied by the cooperative APAEB SISAL (Community Association for Sisal Production and Marketing) located in Valente. The polymer used to prepare the composites was LLDPE ML3602U from Braskem, with a density of 0.937 and a melt flow index (190 °C/2.16 kg) of 5.0.

Compatibilizing agent used was POLYBOND™ 3029 (SI Group). POLYBOND™ corresponds to HDPE modified with maleic anhydride (HDPE-g-MA) and is represented by the chemical structure in Figure 1.

Figure 1. Chemical structure of POLYBOND™



Fonte: Chloé Épinat, 2014.

Preparation of Composites

The sisal residue was oven-dried at 80 °C for 24 h and manually pre-mixed with the polymer. The composite pellets were prepared from three different formulations containing 5% sisal residue either in natura or subjected to alkaline treatment (5% (w/v) NaOH, 2 h, stirrer), and mixed with 17.5% POLYBOND™ 3029, as described in Table 1. In addition to these formulations, a control sample was produced, consisting of 100% LLDPE polymer.

Table 1. Composite formulations.

Sample	Descriptive
LLDPE	Pure LLDPE
LLDPE/5SN	LLDPE composite with 5% untreated sisal residue
LLDPE/5ST	LLDPE composite with 5% sisal residue treated with 5% NaOH
LLDPE/5SC	LLDPE composite with 5% sisal residue and Polybond compatibilizer

The composites were produced using a twin-screw extruder (L/D = 40 mm, AX-Plásticos, model DR1640, Brazil). The temperature profile and screw rotation speed were set to 100/150/170/180/190/190/195/200/195 °C and 120 rpm, respectively. After extrusion, the composites were granulated.

Filament Preparation

Filaments were produced using a Filmaq 3D screw extruder, operating at 185 °C and 18.5 rpm.

3D Printing

Prior to printing, the filaments obtained in the previous step were dried at 60 °C in a dryer. A Prusa Research MK3S+ printer was then used to fabricate the composite specimens under the following parameters: nozzle diameter: 0.6 mm; infill: 100%; print speed: 30 mm/s; printing temperature: 190 °C; bed temperature: 123 °C; orientation: 0°; infill pattern: rectilinear, layer height: 0.3 mm; substrate printing temperature: 255 °C.

Figure 2 shows the experimental scheme for sample preparation and Figure 3 shows the printed test specimens.

Tensile Testing

Test specimens were prepared according to ASTM D638, Type IV, and evaluated using an EMIC universal testing machine (model DL200MF) equipped with a 2 kN load cell. The tests were conducted at a crosshead speed of 50 mm/min, pulling each specimen until failure. Five specimens from each formulation were tested, and the following properties were determined: tensile strength (MPa), elongation at break (%), and tensile modulus (MPa).

Statistical Analysis

The Tukey test was applied to assess significant differences between mean values using the Statistica 7.0 software. Means followed by the same letters in the same column did not differ significantly at the 5% significance level (Tukey, $p < 0.05$).

Figure 2. Scheme for sample preparation.

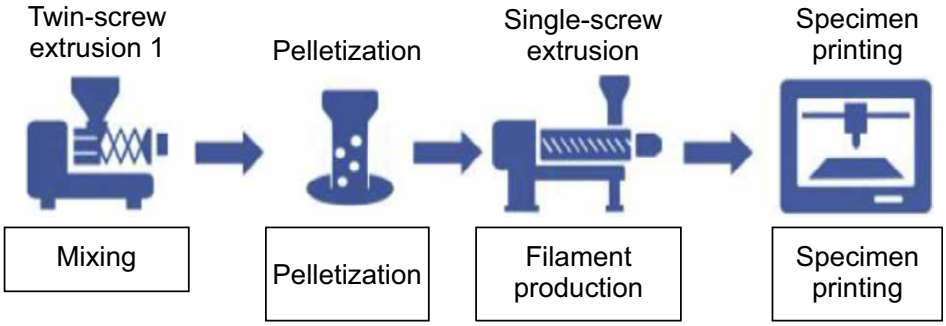
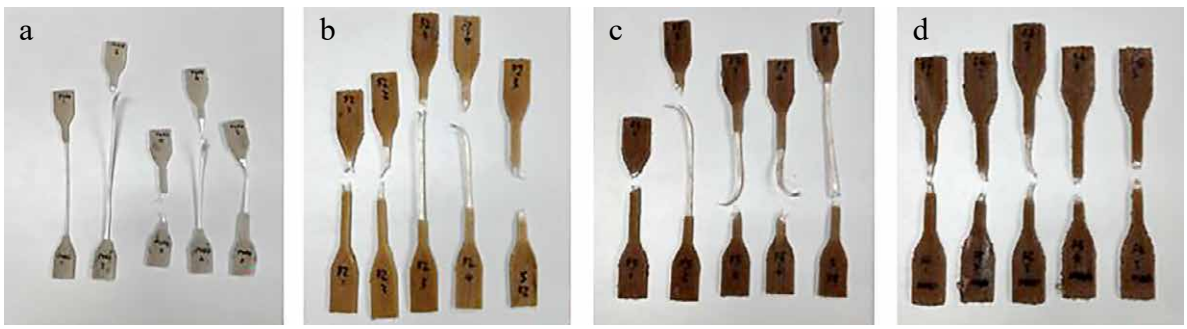


Figure 3. Printed test specimens.



Results and Discussion

The stress–strain curves of the pure polymer and the composites are shown in Figure 4.

Based on the curve profile of the pure polymer, an elastic behavior followed by plastic deformation was observed, which is characteristic of a ductile material. A similar performance was observed for all three composites.

From Figure 4, it can be seen that the addition of sisal residue reduced the deformation of the LLDPE.

This behavior was expected, as it is associated with the higher stiffness and lower intrinsic elongation of the fibers compared to the polymer matrix. The fibers restrict the mobility of the polymer chains and introduce interfaces that can act as stress concentration points. As a result, the composite exhibits lower ductility and fractures at lower deformation levels compared to the pure matrix.

As previously mentioned, LLDPE exhibits ductile fracture, characterized by high deformation prior to failure, localized necking formation, and molecular alignment in the direction of the applied load. The fracture surface shows a smooth and elongated appearance, as illustrated in Figure 5.

Regarding the fracture mechanism, one hypothesis is that, in the plastic regime, molecular chains slide, the crystallites deform, and some partially break down. Thus, failure occurs when the elongated fibrillar structure can no longer sustain the load, leading to rupture by chain scission. In the case of a ductile matrix, there is no rapid crack propagation as observed in brittle fracture [9].

For the evaluated tensile properties, a Tukey test was performed to verify whether there were differences between the means of the pure polymer and the composites. Regarding the maximum stress values, shown in Table 2, it was observed that the pure polymer (LLDPE) and the composite (LLDPE/5SC), referring to the LLDPE composite containing 5% sisal residue with the compatibilizing agent POLYBOND, did not differ statistically from each other. However, they exhibited superior mechanical behavior compared to the other composites with untreated and NaOH-treated fibers.

This is possibly due to the improved adhesion at the fiber-matrix interface, resulting in more efficient load transfer and, consequently, enhanced mechanical properties. Maleic anhydride (MA),

Figure 4. Stress-strain curve of samples.

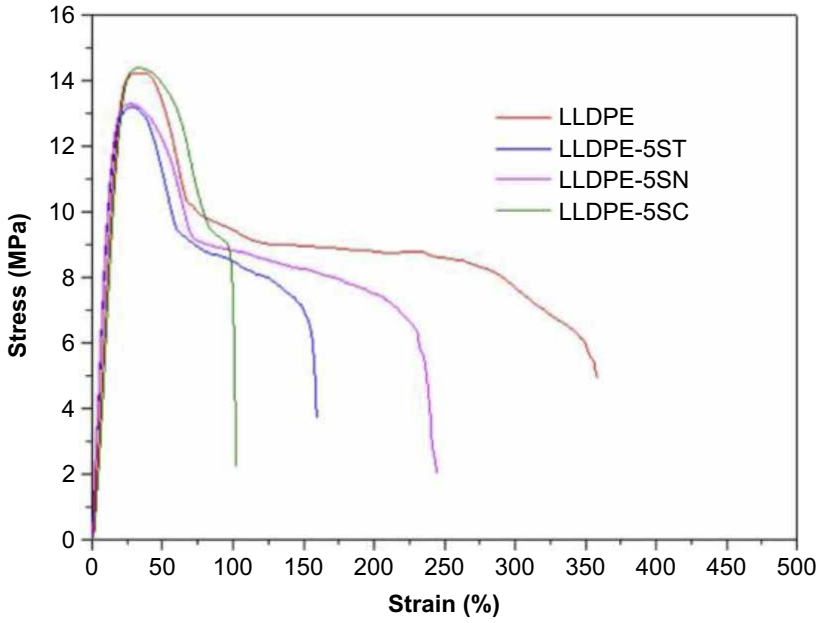


Figure 5. Test specimens after the tensile test.

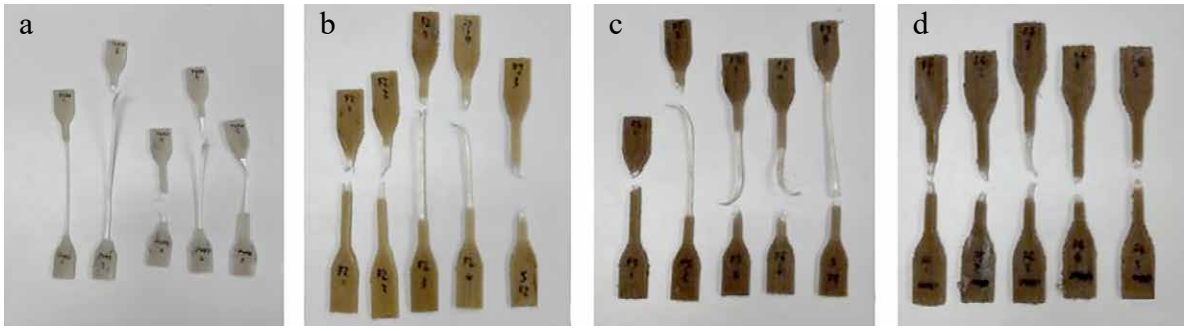


Table 2. Tensile strength (MPa) of pure polymer and composites.

Sample	Tensile strength (MPa)
LLDPE	14.82^{a}±0.77
LLDPE/5SN	13.56^{b}±0.43
LLDPE/5ST	13.50^{b}±0.50
LLDPE/5SC	14.57^{a}±0.30

present in POLYBOND™ 3029, reacts with the hydroxyl groups on the fiber surface. Through the formation of ester bonds between the MA groups and the fibers, the surface hydrophilicity is reduced, which in turn enhances the compatibility of the fibers with hydrophobic polymers [10].

Although NaOH removes impurities and increases fiber roughness, this treatment does not necessarily ensure strong chemical adhesion with the apolar LLDPE matrix. Moreover, even with such treatment, if the fibers are poorly dispersed, no significant improvement in tensile strength will be observed. Therefore, one possible reason why the NaOH chemical treatment did not improve the composite's maximum stress is the still limited interfacial adhesion between the fibers and the polymer matrix. Treatments often improve the adhesion between matrix and fiber; however, there is controversy in the literature regarding the effect of such treatments on the mechanical properties of the fibers and their reinforcing capability. Even when a more significant improvement is observed after chemical treatment, this enhancement is often within the variability of the results. In some cases, similar overall gains can be achieved by simply washing and drying the fibers prior to incorporation into the polymer matrix, which has the added benefits of being more economically viable and environmentally friendly than certain chemical treatments [11].

As observed in Table 3, an increase in the modulus of elasticity of the composites compared to the polymer matrix was expected, since the fiber interferes with the mobility of the polymer chains, leading to an increase in the modulus of elasticity [12]. However, based on the statistical evaluation using Tukey's test, the samples exhibited similar performance, with the exception of the LLDPE/5SC composite.

Mokoena and colleagues. (2004) studied composites of linear low-density polyethylene and short sisal fibers and observed an increase in the tensile strength of the polymer by adding 40% fiber, reaching approximately 22 MPa; with 10% sisal fiber, the tensile strength was about 17 MPa [13].

Table 3. Tensile modulus (MPa) of pure polymer and composites.

Sample	Tensile modulus (MPa)
LLDPE	83.28^{ab} ±4.65
LLDPE/5SN	108.22^{a}±16.69
LLDPE/5ST	100.67^{ab} ±17.24
LLDPE/5SC	82.22^{b}±7.51

Composites reinforced with vegetable fibers, such as sisal in LLDPE, may exhibit a modulus of elasticity similar to that of the pure polymer due to limited interfacial adhesion, low fiber volume fraction, and the presence of short or poorly dispersed fibers, factors that compromise efficient load transfer and the enhancement of the material's stiffness.

Strain at break is the measure of how much a material can elongate or deform before breaking during a tensile test. Expressed as a percentage, this property indicates the material's ductility, being high in ductile materials and low in brittle materials. Table 4 shows the deformation data of pure polymers and composites.

Table 4. Elongation at break (%) of pure polymer and composites.

Sample	Elongation at break (%)
LLDPE	81.87[a]±49.17
LLDPE/5SN	46.00[a]±22.30
LLDPE/5ST	36.83[a]±25.76
LLDPE/5SC	25.60[a]±8.85

Composites reinforced with vegetable fibers generally exhibit lower strain at break compared to the pure polymer, due to the higher stiffness of the fibers and the restriction of polymer chain mobility, resulting in reduced ductility. Table 4 shows the expected result of the decrease in this property when comparing the pure polymer to the composite.

Composites exhibit lower elongation at break than the pure polymer primarily due to the presence of rigid fibers that limit the material's elongation capacity. Vegetal fibers, such as sisal, have higher stiffness than the polymer matrix, which reduces the mobility of the polymer chains and consequently the plastic deformation before fracture. Moreover, the fiber-matrix interface can act as a stress concentration point, facilitating crack initiation. Imperfect adhesion may cause premature failures, while fiber breakage or slippage also contributes to the overall reduction in the composite's ductility. Thus, the combination of these factors results in lower strain at break in composites compared to the pure polymer.

The Tukey test showed that there was no difference between the pure polymer and the composites, nor among the different treatments applied to the composites. In this context, untreated sisal residue, for example, can be added to the polymer, reducing the material cost while maintaining the same strain at break as the polymer matrix.

Mokoena and colleagues (2004) studied composites of linear low-density polyethylene and short sisal fibers and evaluated the elongation at break, which was found to be lower (around 12%) compared to this work (46%). This difference may be related to the fiber content added as well as the type of linear low-density polyethylene used to prepare the formulations.

Preliminary Economic Assessment

For a preliminary economic assessment, the price of the composite components (untreated sisal waste and pure polymer) should be considered. Considering the commercial price per kilogram of each component, the composite containing 5% sisal (R$5.00) fiber and LLDPE (R$12.96) had a reduction of approximately R$0.40/kg, being around 3.1% cheaper than the pure polymer (LLDPE). It is worth noting that the absolute cost reduction or percentage reduction tends to increase as the fiber content increases. Thus, for example, a composite containing 50% sisal fiber would imply an absolute reduction of 3.98, corresponding to a percentage reduction of 30.72% when compared to the pure polymer.

Regarding sustainability, quantified sustainability metrics, such as carbon footprint reduction, are relevant. Reported information indicates that, on average, 1.205 kg of carbon dioxide equivalent is emitted into the atmosphere

when producing 1 kg of linear low-density polyethylene (LLDPE) [14], and the use of non-renewable energy in the production of sisal fiber is approximately 4.2 GJ/t of fiber, and greenhouse gases are approximately 270 kg of CO_2/t of fiber [15]. In a simplified way, it can be estimated that with 5% sisal, a modest reduction in emissions of approximately 0.02–0.05 kgCO_2e per kg of composite (or ~2–4%) was obtained.

Conclusion

For tensile strength, the sample prepared with POLYBOND™ 3029 showed superior performance compared to the other composites, possibly due to better adhesion at the fiber-matrix interface, resulting in more efficient load transfer. Based on the strain at break results, it was observed that the pure polymer and the composites exhibited the same mechanical performance. Thus, sisal residue can be added to LLDPE without any prior treatment, which reduces the material cost and adds value to a residue that can be used for multiple applications.

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