Production of LLDPE and Sisal Composites via 3D Printing

Mariana Souza[1]*, Vinícius Oliveira[1], Lucas Horiuchi[1], Ana Paula Gonçalves[1], Paulo Romano Correia[1], Rodrigo Polkowski[1]
[1]TRL9 Lab; Salvador, Bahia, Brazil

3D printing is a processing technique that presents an interesting possibility for obtaining various materials, with advantages such as lower cost and design adaptation. Various materials are used for printing, including polymers. However, some difficulties can be faced due to the characteristics of the processed materials, such as their mechanical propertie (tensile strength and strain at maximum load). In this context, composite materials formed by linear low-density polyethylene (LLDPE) and sisal fibers in natura and chemically treated with sodium hydroxide solutions were obtained via extrusion (initially on a twin-screw printer to obtain the masterbatch, a single-screw printer to manufacture the filaments and a 3D printer to obtain the test specimens) with the aim of developing new materials and contributing to aspects related to LLDPE processing in the context of additive manufacturing. The specimens were mechanically evaluated through tensile testing, revealing that the control formulation exhibited greater tension at maximum force than the composites, as well as greater strain at maximum load. Comparing the materials with fiber addition without treatment and with treatment with sodium hydroxide at different content, it was observed that the treatment did not lead to significant differences in the evaluated mechanical properties. Thus, although the processing was successful, resulting in intact specimens, fiber treatments under the evaluated conditions were not sufficient to obtain improved mechanical properties.
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Additive manufacturing, also known as 3D printing or rapid prototyping, has been around for decades. However, it was initially an expensive and unviable technology for the general market. However, with advances in the 21st century, costs have dropped significantly, enabling the introduction of 3D printing into various sectors [1].

3D processing has undergone significant expansion in recent years, being widely used worldwide and increasingly applied to mass customization and production of any type of design in areas such as agriculture, healthcare, the automotive and aerospace industries [2].

Aspects associated with 3D printing such as less material waste, ease of manufacturing, lower post-processing rate and energy efficiency make the process sustainable for industrial use [3].

Since the origin of additive manufacturing (AM) there has been a growing interest in this technology for polymers and their composites, driven by advantages such as high efficiency, resolution and customization, however this type of processing requires certain conditions of these materials such as physical and rheological properties, providing limited application of AM in several industries [4]. Several polymers are used in 3D printing processes, such as PLA and PETG [5,6].

Polyolefins and polyolefin elastomers are widely used in various applications, however, they present challenges for use in 3D printing such as excessive softness, shrinkage, warping and low mechanical properties, which limits their application [7]. Other critical issues related to fused deposition (FDM) polyolefin 3D printing are low bed adhesion and low interlayer adhesion. However, the addition of filler can help reduce warpage and increase the strength of printed parts [8].

Low-density polyethylene (LLDPE) is a polymer characterized by medium crystallinity, presenting moderate rigidity and mechanical strength [9].

Some studies have used materials composed of LLDPE obtained by 3D printing [7,10], however, compared to other polymers, the number of studies is still incipient and, therefore, works that propose to investigate possibilities for this material can

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Address for correspondence: Mariana Souza. TRL9 LAB Testing and Technical Analysis. Salvador, Bahia, Brazil. Zipcode:40210-630. E-mail: mariana.costa@trl9.tech.

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contribute with new paths in terms of additive manufacturing of different polymers, expanding the range of uses.

Regarding the addition of vegetable fibers to the LDPE matrix, the study developed by Wearn [11] addresses that components such as lignin and hemicellulose present in these fibers can hinder their adhesion to the polymer matrix, emphasizing that the removal of these compounds in adequate proportions from coconut fibers reduces their hydrophilicity and facilitates coupling to the LDPE. Still according to Wearn [11] the most commonly used chemical treatment for vegetable fibers is alkaline extraction using a sodium hydroxide solution, which is responsible for removing some components from the surface of the fibers, such as lignin and hemicellulose.

Some studies have already proposed to evaluate the effect of mercerization treatment of lignocellulosic biomass in composites involving low-density polyethylene (LDPE) such as castor cake [12], coconut fiber [11], in addition to publications involving linear low-density polyethylene (LLDPE) and natural fibers such as areca fiber [13] and ground hemp fibers [14].

However, the 3D printing process of composites formed by natural fibers and an LLDPE matrix is a field that can be contributed to, aiming to collaborate with the expansion of printing possibilities for this polyolefin, in addition to adding value to a lignocellulosic residue, in this case using residues from sisal processing.

Fiber is the main commercial product, corresponding to 4% of the weight of the sisal leaf, and among the co-products are mucilage, juice and short fibers, also called loofah, corresponding to, respectively, 15; 80 and 1%. In terms of production volume, Brazil is the main world producer, with Bahia ranking first [15].

Therefore, this work aims to produce composites of LLDPE and sisal fibers in natura and chemically treated via 3D printing, with the materials obtained being mechanically evaluated by tensile test.

Materials and Methods

The composites were prepared by mixing LLDPE (Braskem, ML3602U resin) and sisal fibers, totaling 04 formulations composed of both natural fibers and fibers chemically treated with sodium hydroxide solution at different concentrations (5 and 12.5%). For the treated fibers, a 1:20 ratio was used, with a 5% fiber content in each of the composites (Table 1).

The extrusion to obtain the masterbatch occurred in a co-rotating twin-screw extruder (DR.16.40.AX from AX Plásticos), with the temperature profile was 100/150/170/180/190/190/195/200/195 °C and screw rotation speed to 120 rpm. After extrusion, the materials were granulated. Then, the production of the filaments occurred, using a Filmaq 3D single-screw extruder, speed 18.5 rpm, temperature 185 ºC.

The production of the specimens of tensile tests, prepared in accordance with ASTM D638 type IV, were carried out on a 3D printer (Prusa Research MK3S+), through the following printing parameters: nozzle diameter: 0.6 mm, printing and printing bed temperature, respectively, 190 and 123 °C; printing speed: 30 mm/s; rectilinear pattern, orientation: 0% and infill density 100%.

The specimens were tested on an EMIC universal testing machine model DL200MF with a 2 kN load cell and a test speed of 50 mm/min, with five

Table 1. Formulation of LLDPE/sisal composites.

Formulation	LLDPE (%)	Natural fiber (%)	Treated fiber 5%	Treated fiber 12.5%
F1	100.0	–	–	–
F2	95.0	5.0	–	–
F3	95.0	–	5.0	–
F4	95.0	–	–	5.0

replicates being prepared for each formulation. The properties analyzed were tensile strength (MPa) and strain at maximum load (%). The tensile tests results were analyzed by the Tukey test ($p<0.05$) to compare means.

Results and Discussion

The 3D-printed specimens are shown in Figure 1. The images of the composites demonstrate that it was possible to obtain specimens without the presence of macroscopic defects, requiring future scanning electron microscopy analyses and dimensional analysis of the test specimens to identify aspects such as the distribution of matrix fibers and variation in dimensions, respectively.

In terms of processability, the adjustments made during printing, in addition to the development of a PETG substrate for subsequent deposition of the composites, were also essential for printing. The PETG surface enabled layer-by-layer deposition of the material, minimizing the shrinkage effect characteristic of LLDPE, with this surface being removed immediately after the 3D printing process. Figure 2 shows the test specimens obtained via 3D printing after performing a tensile test.

The results of the tensile strength (MPa) and strain at maximum load (%) of the different formulations are presented in Table 2.

When evaluating the mechanical properties of the specimens printed via 3D processing, it was observed that the pure polymer differs statistically from the composites in relation to the properties evaluated, except for the strain at maximum load (%) in which the composite F2 is statistically equal to the control formulation – F1, while the composites did not present any significant difference between them.

Evaluating the composites exclusively, it can be stated that the addition of fibers, whether made with raw material or with material treated with sodium hydroxide solution at different concentrations, did not contribute to mechanical changes in the materials under the conditions evaluated in the present study. Therefore, for these samples, mercerization did not contribute to greater fiber adhesion and dispersion to the matrix reflecting similar results, regardless of the chemical treatment used.

Figure 1. Tensile specimens obtained via 3D printing.

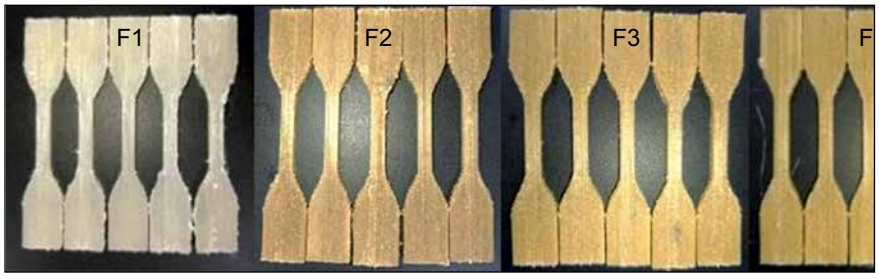

Figure 2. Test specimens obtained via 3D printing after performing a tensile test.

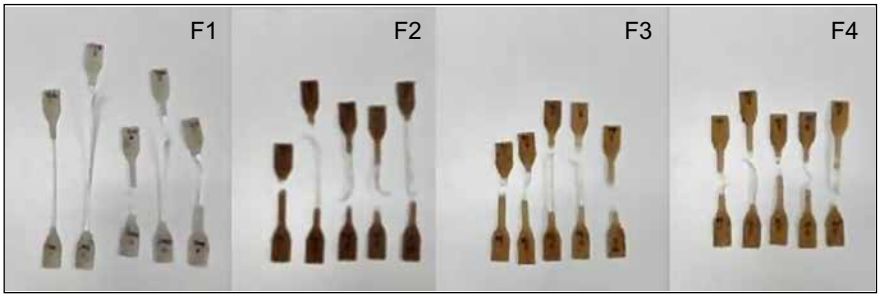

Table 2. Mechanical performance of composite materials.

Formulation	Tensile strength (MPa)	Strain at maximum load (%)
F1	14.83± 0.77a	31.92±2.96a
F2	13.56±0.44b	26.88±1.20ab
F3	13.50±0.50b	26.56±0.72b
F4	13.38±0.36b	28.88±1.04b

Means followed by the same letter do not differ statistically from each other using the Tukey test at 95% confidence interval.

Rocha and colleagues (2020) evaluated the mechanical properties of composites formed by LDPE/PHB and castor bean cake treated via mercerization with 5 and 10% calcium hydroxide solutions. They observed that the chemical treatments contributed to improved properties due to better adhesion properties between interfaces. The authors also revealed that the lower concentration solution probably contributed to better results, attributing the inferior performance of the materials added with fibers treated with higher concentrations of sodium hydroxide to the compromised integrity of the fibers.

The evaluation of coconut fibers treated with 10% sodium hydroxide solution [11] revealed that in terms of tensile properties, the composites with different contents, 5 e 10%, presented maximum stress values inferior compared to pure LDPE. Although it is expected that the removal of components such as lignin and hemicellulose through alkaline treatment would contribute to increased adhesion between filler and matrix [11].

In terms of the strain at maximum load parameter, it was observed that materials F3 and F4, which correspond to the composites in which fibers treated with sodium hydroxide solution at different concentrations were added, presented a lower capacity to deformation than pure LDDPE. A decrease in deformation in composites involving lignocellulosic fibers is a possible effect.

A material produced by Lemos [16] based on PLA and lignocellulosic fibers evaluated through tensile testing showed a decrease in mechanical strength when compared to the control material. This result was attributed to the presence of short fibers dispersed in the matrix, which may have caused a discontinuity, thus hindering the distribution and transfer of the applied stress, leading to a reduction in the maximum stress supported by the biocomposites. Regarding elongation, a decrease in this parameter was observed, which was intensified by the increase in the content of added natural fiber.

There was an increase in stiffness of the fiber composite produced by Wearn [11], reflected in a higher tensile modulus since, according to the authors, the addition of coconut fiber makes the material more rigid due to the stress it is capable of withstanding. Another point raised is that coconut fibers, when compared to other lignocellulosic materials, have a high lignin content, thus reducing the average tensile strength and increasing the material's stiffness.The addition of fiber also reduced the ductility of the samples since there was a lower deformation rate of the composites up to the maximum tensile stress [11].

In evaluating mechanical properties, Lemos and Martins (2014) [16] explored how the insertion of a rigid phase, such as wood fibers, can increase the rigidity of the polymer blend. They emphasized, through the evaluated references, that a relevant point for materials of this nature is the surface treatment, since this modifies the lignin content through the use of alkaline solutions of sodium hydroxide andhypochlorite. Therefore, the surface chemical treatment of the fibers can substantially affect the thermal, mechanical, and adhesion properties between the polymer matrix and the fibers, which are dependent on factors such as reagent concentrations and temperature. In the present work, only the sodium hydroxide

concentration parameter at room temperature was evaluated, and the study can be expanded in the future to investigate different chemical treatment temperatures and their effect, for example, aiming to present new perspectives for the addition of natural fibers in LLDPE, thus contributing to the aggregation of value of an agro-industrial residue, in addition to collaboration for the field of composite production via 3D printing.

Conclusion

Composite materials composed of LLDPE and sisal fibers were obtained through 3D printing, demonstrating the potential of additive manufacturing for processing various materials. The mercerized (chemically treated) fibers were obtained, however, the necessary reinforcing effect was not observed in the materials evaluated, requiring further studies to improve the characteristics of these composites.

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