Development of Processing Strategies to Enable 3D Printing of LLDPE With Sisal Fiber Reinforcement

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Linear low-density polyethylene (LLDPE) is a packaging material with superior properties compared to conventional low-density polyethylene. However, due to its low thermal stability, it cannot yet be used in high-performance applications. Due to its use as single-use packaging and low natural degradation in nature, the continued use of this material may delay the achievement of the UN's sustainable development goals. Therefore, sisal fibers can be used in LLDPE, reducing the carbon footprint of this material and utilizing agro- industrial residue. However, the use of these fibers reduces the processing temperature to approximately 200 °C due to their degradation and therefore brings even more challenges to the application of this material in 3D printing. This polymer exhibits significant thermal shrinkage due to crystallization, which hinders its application in 3D printing. To this end, this work aims to develop new techniques for printing a novel LLDPE/ sisal fiber composite. Printing temperatures between 175 and 190 °C were tested, and it was observed that temperatures below 190 °C caused problems due to the low polymer fluidity in the 3D printer's small extruder nozzle. However, this temperature was enough to guarantee the completion of the printing. Three substrate materials were analyzed: polyethylene terephthalate glycol (PETG), high-impact polystyrene (HIPS), and polylactic acid (PLA), at a printing bed temperature of 80 to 123 °C. The experiments enabled the printing of good-quality test specimens using HIPS and PETG as substrates, but only at a bed temperature of 123 °C. The test specimens produced enabled the feasibility of future mechanical characterization of the material, and the methods developed will enable the development of LLDPE in the literature. As future research, the generation of a customized g-code will be studied that will allow the use of the same substrate to manufacture multiple test specimens, thus reducing process waste.

Keywords: LLDPE. 3D printing. Composite. Sisal fibers.

Linear low-density polyethylene (LLDPE) is a semi-crystalline and flexible polymer used in packaging, cable sheathing, among other applications. Although LLDPE exhibits superior properties compared to non-linear LDPE, the low heat deflection temperature and limited mechanical strength of both materials hinder their use in structural applications [1,2].

The use of PE and its variations as single-use packaging exacerbate environmental problems and hinders companies' progress toward the sustainable development goals (SDGs) proposed by the UN [3] due to the low degradation rate in nature. To mitigate the environmental impact of using this polymer, the incorporation of sisal fibers into LLDPE can enhance sustainability by reducing its carbon footprint, in addition to providing a destination for industrial waste from fiber processing [4]. Because of this, 3D printing can provide applications with high added value to LLDPE, and possibly recycled LLDPE, such as automotive parts, in which its high toughness, low density, and low cost can favor the reduction of vehicle weight and manufacturing costs [5]. These benefits can be increased with the addition of sisal fibers due to their low cost, high production volume, and growing environmental disposal concern. The use of these fibers can provide an alternative to agro-industrial residues that would otherwise be used as biomass [6].

However, the addition of sisal fibers poses challenges for printing this material due to the decrease in thermal stability [7], in addition to the difficulties caused by high thermal shrinkage, which is amplified by the effects of crystallization due to

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the polymer's high crystallinity (above 50%) [8]. To enable printing of LLDPE, the literature has resorted to the manufacture of blends with other polymers such as polyolefin elastomers [9], but 3D printing the polymer in pure form remains a challenge.

Aiming to open a new research avenue for 3D printing of LLDPE with sisal fibers, this work aims to develop new techniques for printing a novel LLDPE/ sisal fiber composite based on the following guiding questions: (1) What are the problems observed when printing with LLDPE? (2) Which are the printing requirements considering the addition of fibers? (3) Which are the best substrate materials to prevent detachment from the print bed? (4) Which are the best printing parameters for LLDPE?

Materials and Methods

The materials used were LLDPE grade ML3602U from Braskem in the formulations (Table 1) described in Table 1. For incorporation of sisal fibers, the twin-screw extruder L/D = 40 mm, AXPlásticos (model DR1640, Brazil) using a temperature profile of 100/ 150/ 170/ 180/ 190/ 190/ 195/ 200/ 195 °C and 120 rpm was used. For filament production was used the Filmaq3D CV Model single-screw Extruder at the temperature of 185 °C. The substrate materials used were high-impact polystyrene (HIPS) filaments and natural-colored polylactic acid PLA from 3DFILA, in addition to black Polyethylene terephthalate glycol (PETG) from Prusa.

To print the test specimens, the Prusa MK3s+ 3D printer with a smooth PEI table was used, using the printing parameters described in Table 2. The G-code was generated through the Prusa Slicer software. For printing, test specimens were printed in accordance with ASTM D638 type 4.

Table 1. Formulations used in this study.

Formulation ID	LLDPE (%)	Sisal Fibers (%)
F0	100	0
F1	95	5

Results and Discussion

Table 2. 3D printing parameters used in this study.

Printing parameter	Value
Nozzle diameter	0.6 mm
Extrusion width	0.6 mm
Layer height	0.3 mm
Printing temperature PETG, HIPS	255 °C
Printing temperature PLA	210 °C
Printing temperature LLDPE	190 °C
Bed temperature	80-123 °C
N° of substrate layers	3
Type of substrate	Raft

Observation of Printing Problems with LLDPE With Sisal Fibers

It was observed that under processing conditions of 190 °C and printing bed temperature of 80 °C, it was not possible to guarantee adhesion. Figure 1 shows a test specimen with 5% sisal fibers content that detached from the printing bed during production, and which inspired this study.

Analysis of Printing Requirements Considering LLDPE Composite with 5% Sisal Fibers

Although the use of sisal fibers as reinforcement reduces the environmental impact of this plastic and, consequently, its carbon footprint, their addition limits the processing temperature to approximately 195 °C due to fiber degradation at high temperatures. Based on this, a printing test was performed with pure LLDPE (F0) to determine the lower possible printing temperature of the material

Figure 1. Detachment of the printing bed caused by the sample warping in composition with LLDPE with 5% sisal fibers.

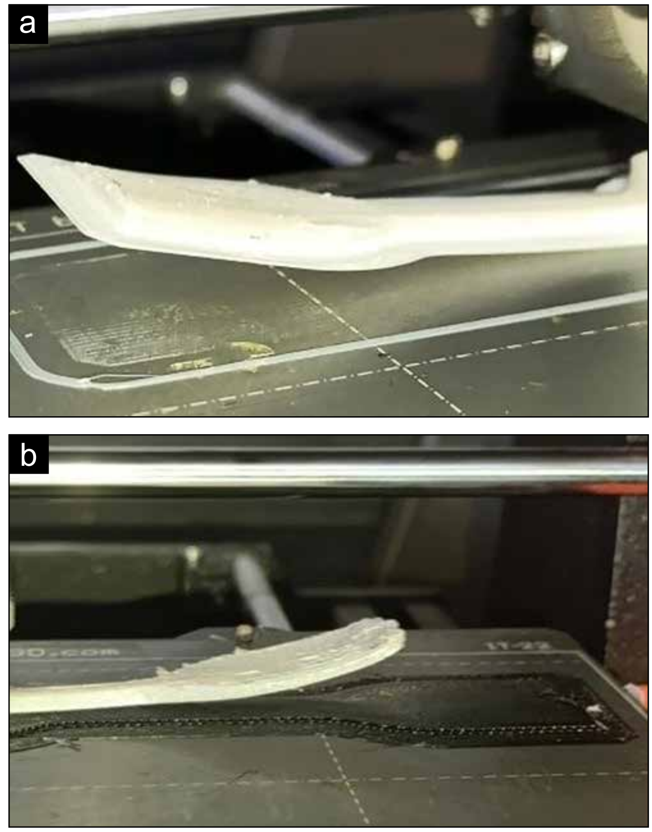
within the 190 °C limit. To this end, temperatures of 175, 180, 185, and 190 °C were tested. At 175 °C, the test specimens exhibited voids in their first layers due to the low fluidity of the polymer in the extruder nozzle, indicating that the material did not melt properly. Tests at 180 and 185 °C showed few voids in the first layers but still did not melt properly, resulting in extruder motor stalling with a characteristic clicking sound and the failure of the print. From this, it was defined that the best printing temperature that maintains the integrity of the fibers and guarantees the completion of the print is 190 °C, also been in in agreement with the literature [4].

<u>Analysis of the Best Suitable Substrate Material</u>

For the analysis, a raft-type interface was created with the bed temperature set at 80, 100 and 123 °C (maximum possible temperature) to reduce printing defects due to thermal contraction.Figure 2a shows that the HIPS substrate peeled off along with the sample at 80 °C, while with the PETG substrate (Figure 2b), the sample peeled off, but the substrate remained in full contact with the print bed.

To evaluate the 100 °C printing bed temperature, tests were also performed with PLA. It was expected that, due to the material's lower melting

Figure 2. Printing LLDPE with different substrate materials at a bed temperature of 80 °C [(A) HIPS (B) PETG].

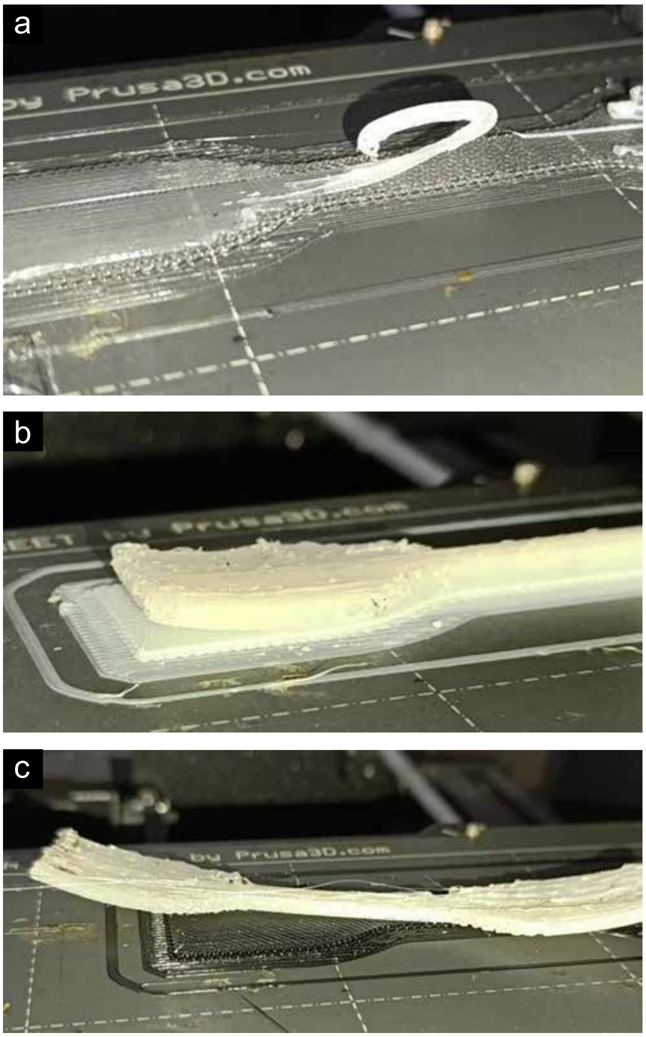


temperature and higher print bed temperature, LLDPE would be able to melt the substrate material and achieve better adhesion.

However, as can be seen in Figure 3a, LLDPE did not adhere satisfactorily to PLA, and testing with this substrate material was discontinued. Figure 3b shows the test with HIPS. Although the substrate material remained adhered to the print bed, the extruder nozzle lost its position due to thermal contraction of the specimen, and the print could not be completed.

To understand the behavior of polymers at the highest possible bed printing temperature with the intention of mitigating the thermal gradient between the printing layers and consequently the thermal contraction of the part, tests were carried out with a bed temperature of 123 °C. In Figure 4, it was observed that the HIPS material had satisfactory adhesion and was able to complete the

Figure 3. Printing LLDPE with different substrate materials at 100 °C bed temperature [(a) PLA (b) HIPS (c) PETG].



printing with low warping compared to previous tests, a result that allowed the production of test specimens for mechanical evaluation.

To validate this result with the addition of sisal fibers, Figure 5a shows the specimen printed with the HIPS substrate, and Figure 5b shows the specimen produced after cooling and removal of the substrate. The substrate had satisfactory adhesion to the specimen and was able to complete the printing.

Figure 6a shows the specimen produced with the PETG substrate with slight warping, but much lower than that of specimens produced with a lower bed temperature. Figure 6b shows the specimen after cooling and removal of the substrate.

Figure 4. Printing LLDPE with HIPS substrate at a bed temperature of 123 °C.

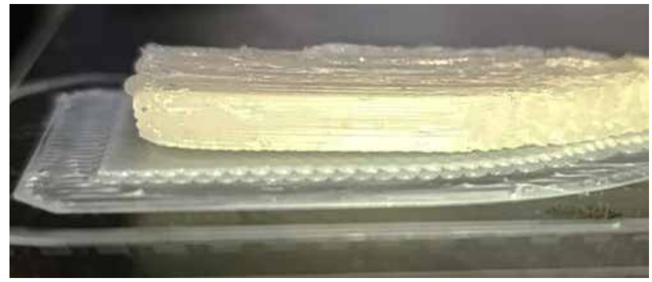


Figure 5. Printing LLDPE composite with HIPS substrate at 123 °C bed temperature (A) after printing (B) after cooling and detachment from substrate.

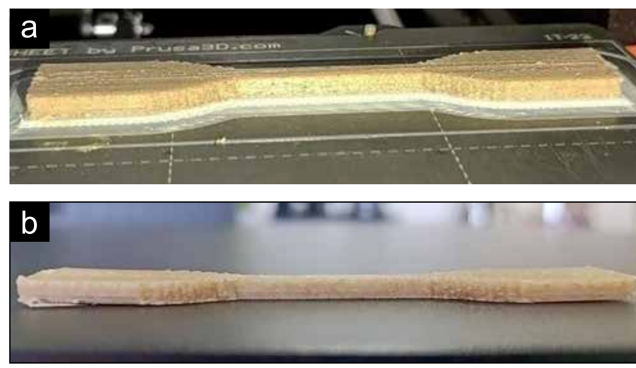
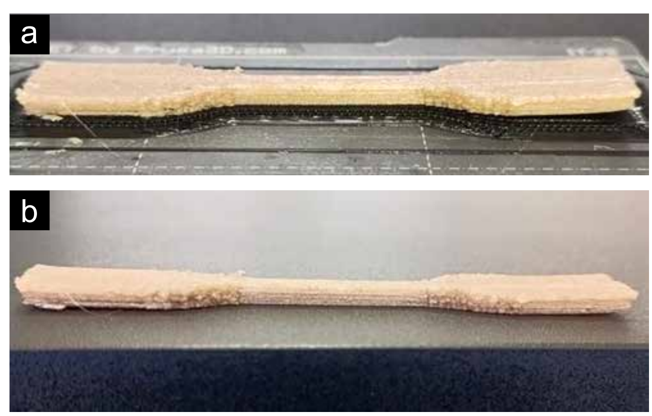


Figure 6. Printing of LLDPE with 5% sisal fibers with PETG substrate.



The feasibility of printing at such a high bed temperature can be explained by the high polymer temperature (above the cold crystallization temperature) [6], which neutralizes the thermal contraction caused by this phenomenon, in addition to reducing the effects of thermal contraction on the material due to the smaller temperature gradient between the print nozzle and the bed. After printing is complete, the entire part is cooled uniformly by natural convection. This is to ensure that all layers cool evenly after printing, and the crystallization process occurs gradually throughout the part, increasing its dimensional stability.

Based on the tests, it can be concluded that both HIPS and PETG polymers serve as suitable substrates for LLDPE printing maintaining good adhesion even with composite material, but only at 123 °C bed temperature.

Conclusion

After analyzing the technical requirements for 3D printing of pure LLDPE and a novel LLDPE with 5% sisal fibers composite, it was possible to determine that a printing temperature of 190 °C is the most suitable as it guarantees adequate fluidity for printing without compromising the integrity of the sisal fibers. Furthermore, three interface materials were tested, two of which successfully printed tensile specimens. Printing was possible using HIPS and PETG substrates using a raft support pattern, but only at a bed temperature of 123 °C. The developed printing method proved to be an important step forward in enabling literature experimentation with this material with 3D printing. Future research will develop custom G-code for substrate reuse between prints, thus reducing material waste.

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