Development of eucalyptus fiber masterbatch for polyethylene additives

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Abstract: Growing concerns about sustainability and environmental impact have driven the development of alternative materials, such as polymer composites reinforced with natural fibers. However, a gap remains between scientific advances and large-scale industrial applications of these composites. This study aimed to develop eucalyptus fiber masterbatches as an alternative to optimize the production of high-density polyethylene (HDPE) composites. To obtain the masterbatches, high fiber concentrations (30%, 40%, and 50%) were incorporated into HDPE using twin-screw extrusion. From the 50% concentrate, composites containing 10% fiber were subsequently produced. Optical microscopy analysis showed good fiber dispersion and distribution in the polymer matrix, with an estimated average length of 7.48 mm. Mechanical testing revealed that, although fiber addition reduced tensile strength, it increased the elastic modulus, indicating structural reinforcement. In terms of impact strength, no changes were observed in the composite produced from the masterbatch, confirming adequate fiber distribution and the absence of agglomerates. Overall, the results demonstrate the feasibility of using eucalyptus fiber as reinforcement in HDPE composites produced from fiber concentrates.

Keywords: Polymer composites. Eucalyptus fiber. Polyethylene. Additives. Masterbatch.

1.Introduction

Growing concern for sustainability and minimizing environmental impacts has driven the development of alternative materials, such as polymer composites reinforced with natural fibers. These materials have gained prominence due to their economic, environmental, and technical advantages, such as low cost, biodegradability, good specific strength, and wide availability of raw materials [1].

In this context, eucalyptus fiber stands out as a promising option, mainly due to its abundance in Brazil, its high cellulose content, and its good mechanical properties [2–3]. However, for these fibers to be efficiently incorporated into polymer matrices, an appropriate beneficiation process is required. This process includes steps such as drying, grinding, screening, and, in some cases, physical and chemical treatments, intended to increase fiber compatibility.

One potentially efficient way to incorporate natural fibers into polymer matrices in industrial processes is through masterbatch production. This approach uses a carrier polymer to incorporate high concentrations of fibers, which can then be added directly during processing when a relatively low final concentration of additives, in this case, natural fibers, is required. Because they are pre-dispersed in the carrier polymer, they do not require an additional dispersive mixing step [4]. This technique can promote mix homogenization, improve reinforcement dispersion, and provide greater control over the properties of the final composite [5–6].

Considering this scenario, this study obtained composites with high concentrations of eucalyptus fiber in the form of masterbatch and evaluated the influence of this dispersion method on the mechanical properties of HDPE composites.

2.Materials

In this study,high-density polyethylene (HDPE JV060U), manufactured and supplied by Braskem, with MFI equal to 7.0 g/10 min (190°C, 2.16 kg), melting temperature of 140°C and density of 0.957 g/cm³ was used [7]. The eucalyptus fiber was donated by the company DEXCO,containing particles in various particle sizes (range of 149 μm ≤ X ≥ 710 μm).

3.Methods

3.1. Masterbatch preparation

Eucalyptus fibers were initially dried in an oven at 110 °C for 4 h. The fibers and pure polymer were then processed in a co-rotating twin-screw extruder (model DR.16.AX, AX Plásticos), with a screw diameter (D) of 16 mm and a length-to-diameter ratio (L/D) of 40. The temperature profile adopted was: Z1: 150 °C, Z2: 155 °C, Z3: 160 °C, Z4: 165 °C, Z5–6: 170 °C, Z7: 175 °C, and Z8–9: 175 °C. Formulations containing 30% and 40% fiber by mass were extruded at 120 rpm, while the formulation with 50% fiber was extruded at 150 rpm, with the aim of improving fiber incorporation efficiency during this stage. The melt flow index (MFI) of the masterbatches was determined using a standardized plastometer at the Materials Development Laboratory of UFBA – Camaçari, at 190 °C with a load of 2.16 kg, in accordance with ISO 1133.

3.2. Preparation of the composite from masterbatches

After analyzing the melt flow index, the masterbatch containing 50% fiber was selected for compound preparation, resulting in an HDPE composite with 10% fiber (Figure 1).

Figure 1. Masterbatch with 50% eucalyptus fiber

The chosen masterbatch was dried at 100 °C for 5 h and subsequently processed with virgin polymer in the same extruder (model DR.16.AX, AX Plásticos), under identical conditions (D = 16 mm, L/D = 40, screw speed of 150 rpm, and the same temperature profile). For comparison, pure polymer was also extruded under the same processing conditions.

3.3. Characterization of the composite obtained from the masterbatch

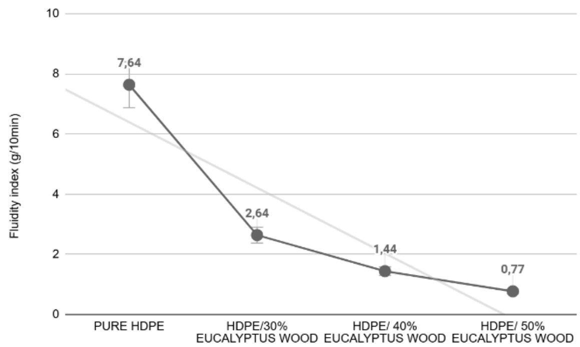
The degree of fiber dispersion in the composite was qualitatively assessed by optical microscopy. Thin films of the material were prepared by hot pressing and subsequently analyzed using a RoHS digital microscope at the Materials Development Laboratory of UFBA – Camaçari Campus. After this step, the material for obtaining the test specimen was prepared. The composites were oven-dried at 100 °C for 5 h and then subjected to hot pressing in a heated hydraulic press (SOLAB, model SL-11). Upper and lower molds were preheated to 200 °C and filled with an average mass of 115 g. The pressing cycle consisted of 25 min of heating followed by 40 min of cooling under a pressure of 5 tons. After cooling and demolding, the specimens were laser-cut to the dimensions specified in ASTM D638, at a cutting speed of 4 mm/s. Tensile strength tests were performed at the SENAI CIMATEC Mechanical Testing Laboratory using a universal testing machine (Emic DL 2000), with data acquisition and processing performed by Tesc software, according to ISO 527, at a test speed of 5 mm/min. Impact strength was determined using an Instron CEAST 9050 impact machine, equipped with a 2.7 J hammer in Izod configuration, in accordance with ISO 180. Prior to testing, the specimens were notched at a 45° angle and 2.5 mm depth. For each formulation, five specimens were tested in both tensile and impact strength assays.

4. Results and Discussion

4.1. Obtaining and characterizing masterbatches

In the first stage, the fiber processing conditions were evaluated with HDPE in a twin-screw extruder. During the extrusion process, for the formulation containing 50% eucalyptus fiber, the screw rotation speed had to be adjusted from 120 to 150 rpm to improve the efficiency of fiber incorporation into the polymer. Figure 2 shows the melt flow index results of the concentrates.

Figure 2. Melt flow index (MFI) of eucalyptus fiber masterbatches.



The graph shown demonstrates a significant reduction in the flow rate as the amount of wood in the formulation increases. This behavior was expected, as incorporating a rigid phase into the polymer hinders the molecular mobility of the chains, increasing viscosity and decreasing its flowability [8]. Despite the low melt flow rate, the masterbatch with 50% fiber was selected. According to Manrich (2005) [5], the viscosity of the polymer matrix is directly linked to dispersion efficiency due to the intensity of the forces transferred during the mixing process. In situations where the mixture begins with a low melt flow rate, dispersion may occur. Subsequently, with the increase in temperature and shear during extrusion, the increase in melt flow rate resulting from these parameters contributes to good distribution and a good level of mixing.

4.2. Characterization of the composite obtained from the masterbatch

Using optical microscopy, it was possible to observe the distribution and dispersion of wood particles in the polymer matrix after the concentrate was dispersed in the HDPE. The images obtained were processed and analyzed using Image J software, which enabled the segmentation and quantification of the particles present in the matrix. The analyzed sample contained a total of 108 particles, corresponding to the eucalyptus fibers incorporated into the matrix. The distribution revealed a predominance of small fibers, suggesting good dispersion of the lignocellulosic reinforcement in the composite. Based on the fiber area analysis, an estimated average length of 7.48 mm was calculated, considering the square root of the area as an approximation of the observed fiber length.

Furthermore, the volumetric proportion of the fibers in the sample was estimated based on the ratio between the sum of the particle areas and the total area of the analyzed image. The results indicated that the fibers occupied approximately 7.072% of the image area, which is close to the proportion initially intended. In terms of particle size distribution, it was observed that 90.74% of the particles had an area of less than 50 mm², while only 0.93% of the fibers exceeded 10,000 mm². This confirms a predominance of small particles, a desirable characteristic to ensure better interfacial interaction between the HDPE matrix and the eucalyptus fibers.

Figure 3. Optical microscopy of the composite with eucalyptus particles and HDPE matrix obtained from master-batch (image magnified 120x).

Figure 4. Analysis of the proportion of the dispersed phase in the HDPE JV060U matrix.

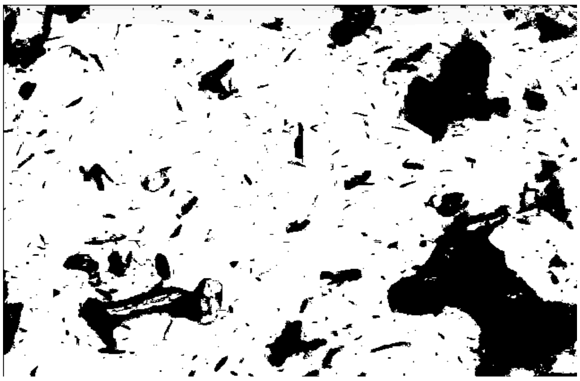


Table 1 presents the results of the mechanical properties under tension and impact resistance of the composite with 10% fiber.

Table 1. Mechanical properties of the composite

Material	Tensile Strength (Mpa)	Modulus of elasticity (Mpa)	Impact resistance (KJ/m²)
HDPE Pure	16.40 ± 1.5	1140.46 ± 134.2	5.80 ± 0.5
HDPE/10% Fiber	7.08 ± 4.7	1454.88 ± 144.3	5.64 ± 0.4

The results indicate a reduction in the composite's tensile strength compared to the pure polymer. For a formulation with natural fiber, without the use of a compatibilizer, this decrease in strength may be associated with several factors. One of the main factors is poor interfacial adhesion between the polymer matrix and the natural fibers or the accumulation of empty balls in the specimen, since this study used press molding [9].

The modulus of elasticity represents the stiffness of a material when subjected to elastic deformation. It was observed that the presence of fiber resulted in an increase in material stiffness. A considerable increase in the modulus of elasticity when fiber was added was observed in other studies, with its maximum values also varying depending on the type of fiber used [10]. The increase in the elastic modulus can result from good fiber distribution in the matrix, as well as its orientation and interaction between phases [11].

Impact strength is a fundamental property in assessing the toughness of materials, representing their ability to absorb energy under rapid loading before fracturing. The results demonstrated that there was no significant variation in this property for the composite obtained from the masterbatch. These results indicate that, under certain conditions, the presence of natural fiber can act as a reinforcing element, contributing to energy absorption and hindering crack propagation in the material. Another factor that influences this property is the formation of agglomerates. Agglomerates can act as stress concentrators, promoting crack formation, which reduces the energy absorbed upon impact [12]. The results obtained in this work for impact strength indicate that no agglomerates formed.

5. Conclusion

The study demonstrated the feasibility of producing a masterbatch with 50% eucalyptus fiber for HDPE additives. Microscopic analysis of the dispersion in the composite obtained from the concentrate revealed good fiber dispersion and a proportion close to the expected. Regarding mechanical performance, the results demonstrated the need to evaluate the interface between the fiber and the matrix to better elucidate the tensile strength results, which showed a significant reduction. The modulus of elasticity is influenced by the presence of fiber, resulting in a material with less deformation. Impact resistance was unchanged, indicating good fiber distribution in the composite, without agglomeration.

Finally, this method simplifies the composite preparation process and can bring economic benefits for large-scale production, with broad prospects for technological application.

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References

[1] SATYANARAYANA, Kestur G.; ARIZAGA, Gregorio GC; WYPYCH, Fernando. Biodegradable composites based on lignocellulosic fibers—An overview. Progress in polymer science, v. 34, n. 9, p. 982-1021, 2009.
[2] PAIVA, Jane MF; TRINDADE, Wanderson G.; FROLLINI, Elisabete. Compósitos de matriz termofixa fenólica reforçada com fibras vegetais. Polímeros, v. 9, p. 170-176, 1999.
[3] SORDI, Julia Casteli de; SCHNEIDER, Elisangela Edila; NACHTIGALL, Sonia Marli Bohrz. Compósitos poliméricos com fibras naturais. Salão de Iniciação Científica (15.: 2003: Porto Alegre). Livro de resumos.[recurso eletrônico]. Porto Alegre: UFRGS, 2003., 2003.
[4] RABELLO, Marcelo. Estrutura e Propriedades de Polímeros. E-Guide, p. 341, 2023.
[5] MANRICH, Silvio. Processamento de termoplásticos: rosca única, extrusão e matrizes, injeção e moldes. Artliber Ed., 2005.
[6] CANEDO, Eduardo Luís. Fenômenos de transporte. Grupo Gen-LTC, 2000.
[7] Polietileno de Alta Densidade JV060U. Braskem, 2018.
[8] STARK, Nicole; BERGER, Mark J. Effect of species and particle size on properties of wood-flour-filled polypropylene composites. Proceeding of functional fillers for thermoplastic and thermosets. December, p. 8-10, 1997.
[9] DE CARVALHO, Marcelo Sousa; AZEVEDO, Joyce Batista; BARBOSA, Josiane Dantas Viana. Effect of the melt flow index of an HDPE matrix on the properties of composites with wood particles. Polymer Testing, v. 90, p. 106678, 2020.
[10] BALASURIYA, P. W.; YE, Lin; MAI, Y.-W. Mechanical properties of wood flake–polyethylene composites. Part I: effects of processing methods and matrix melt flow behaviour. Composites Part A: Applied Science and Manufacturing, v. 32, n. 5, p. 619-629, 2001.
[11] KARMARKAR, Ajay et al. Mechanical properties of wood–fiber reinforced polypropylene composites: Effect of a novel compatibilizer with isocyanate functional group. **Composites Part A: Applied Science and Manufacturing**, v. 38, n. 2, p. 227-233, 2007.
[12] BARBOSA, Josiane Dantas Viana et al. Development and characterization of WPCs produced with high amount of wood residue. Journal of Materials Research and Technology, v. 9, n. 5, p. 9684-9690, 2020.