Evaluation of the influence of Eucalyptus fiber particle size on the mechanical behavior of injection molded PBS composites

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Abstract: The growing demand for sustainable and high-performance materials has driven the development of polymer composites reinforced with vegetable fibers. In this context, Polybutylene Succinate (PBS) stands out as a promising matrix due to its biodegradability, thermal processability and compatibility with lignocellulosic fillers. The production of PBS-based composites has gained both academic and industrial relevance, especially when associated with reinforcements of natural origin that can improve mechanical properties and reduce costs. In this scenario, eucalyptus, widely cultivated in Brazil and abundant in processing residues, emerges as a viable source of fibers for the development of structural biocomposites, expanding the potential application of PBS in different industrial sectors. The composites were produced with 30% by mass of eucalyptus fibers, with two different particle size ranges (FN- >710 µm to <149 µm and FP - 210 µm to < 149µm), using a two-step process: extrusion and injection molding. The specimens obtained were evaluated for mechanical and morphological properties. The results showed that the addition of fibers promoted an increase in the modulus of elasticity and hardness of the material, with better performance observed in composites prepared with sieved particles (PBS/FP), due to greater granulometric uniformity and better wettability of the fibers by the matrix. Despite the good interfacial adhesion observed in the formulation with 30% reinforcement, the load transfer efficiency was still limited. However, the additional sieving step proved to be advantageous for the final performance of the material. Based on these results, future studies will be carried out to prioritize strategies for compatibilization and optimization of fiber dispersion in the matrix, with a focus on improving the matrix-reinforcement interface - a critical factor for the performance of polymer composites.
Keywords: Eucalyptus fiber. Polymeric biocomposites. Polybutylene Succinate. Injection molding. Mechanical properties.

1. Introduction

The Polybutylene Succinate (PBS) is an emerging biopolymer, derived from renewable sources, environmentally safe and has several special properties, such as good transparency and excellent processability, comparable to that of Polypropylene (PP), has a shiny appearance and flexibility, but also has some disadvantages, such as fragility and cost [1].

Its processing versatility and compatibility with lignocellulosic fillers make it particularly suitable for applications that require sustainability without compromising mechanical performance, reducing total material cost while altering desired properties and degradation rates. When combined with eucalyptus fibers - a widely available and low-cost material in the national scenario - PBS can create materials with improved characteristics, capable of meeting the demands of various industrial sectors, including the agro-industrial packaging sector [2].

However, an interaction between the polymer matrix and the fibers is a determining factor for the final properties of the composite. Especially for PBS, some studies have been carried out with Canabrava Fiber, the results of which are very promising.

In this work, eucalyptus fiber is being evaluated for the production of a new composite, with improved mechanical properties. The Eucalyptus is a widely cultivated species in the country and has become the main source of raw material for the production of plywood and furniture, due to its rapid growth, adaptability to different soils and climatic conditions and excellent mechanical properties [3].

Its processing generates by-products such as sawdust and chips, which can be reprocessed and used, for example, as a reinforcing material in polymer composites.

In this context, this work aims to evaluate the effect of the particle size of eucalyptus fibers (FN: X ≤ 710µm and FP: X ≤ 210 µm) on the mechanical properties of composites injected with PBS, using a fixed filler content of 30%, seeking to verify whether the variation in the particle size distribution of the fibers positively or negatively influences the properties of the material, especially in terms of mechanical resistance, rigidity and tenacity, with a focus on evaluating its viability and performance when applied to packaging aimed at the agro-industrial sector.

2. Experimental

2.1 Materials

BioPBSTM, sold by PTT-MCC Biochem, was used as a polymeric matrix, with a melt flow index (MFR) equal to 22 g/10 min (190°C, 2.16 kg), melting temperature of 115°C and density of 1.26 g/cm3 [4]. The Eucalyptus fiber was donated by the company DEXCO, as residue from the production of plywood panels, containing particles in different particle sizes (range of 149 µm ≤ X ≥ 710 µm), in which part of this material was mechanically sieved (Bertel brand) on a 70 mesh sieve (210 µm) and the other part remained unchanged.

2.2 Preparation of the composites

All materials were dried in a vacuum oven to remove moisture. The powdered Eucalyptus fiber, in its global form, that is, without processing (FN), and sieved with a 70 mesh mesh (FP) were dried at 110°C in an oven with air circulation for a period of 4 hours; while for PBS, the drying condition followed the manufacturer's technical sheet which suggests a temperature of 80°C for 5 h.

Three samples were prepared: pure PBS (100% PBS), PBS/FN (70% PBS/30% global eucalyptus fiber) and PBS/FP (70% PBS/30% Eucalyptus fiber in 70 mesh), which were chosen based on other studies carried out by the research group with PBS and natural fibers [5].

Subsequently, the materials were extruded in a twin-screw extruder, model DR.16.AX (AX Plásticos), co-rotating, with a screw diameter (D) of 16 mm and a length/diameter ratio (L/D) of 40, using a temperature profile of Z1:90/ Z2:100/ Z3-4:125/ Z5-6:130/ Z7-8:125/ Z9:120°C and screw speed of 120 rpm.

The mixture obtained was pelletized and dried for 8 h at 80 °C and then subjected to an injection molding process, with a temperature profile of 150/160/170/180ºC, holding pressure of 700 bar for 12 s, injection pressure of 800 bar and injection flow of 40 cm3/s, specimen format in the ASTM D638 standard.

2.3 Characterization

2.3.1 Mechanical Properties

The tensile test was carried out on a universal testing machine EMIC DL-2000 (INSTRON), with a 9 kN load cell and a speed of 5 mm/min, in accordance with ISO 527-1 with an atmosphere of 23 ±2 °C and 50 ±10% relative humidity; the Izod impact test was carried out on an INSTRON impact instrument, in accordance with the ISO 180 standard, whose sample indentation was 2 mm deep; Finally, SHORE D hardness was measured using the Bareiss HPE II Shore D equipment.

For all tests, at least five specimens were used for each sample. All data were subjected to Tukey's test to check for significant differences between means using OriginPro Graphing & Analysis 6.4. Means indicated by equal letters and in the same column do not differ at the 5% significance level (Tukey, $p < 0.05$).

2.3.2 Microscopia Eletrônica de Varredura

The impact fracture surface of the specimens of all samples was metallized with carbon and characterized by a Jeol JSM-5800 LV scanning electron microscope, with a supply voltage of 3–20 kV, to analyze the morphology and adhesion of the polymer matrix to the fibers.

3. Results and Discussions

The particle size classification of the eucalyptus fiber was obtained using the method of weighing the residual mass retained in ASTM standardized sieves as illustrated in Figure 1.

The results show that the fiber used has a heterogeneity in size distribution. For this study, two different fiber samples were evaluated: one with the original size distribution, resulting from the primary processing of fiber waste (FN: X ≤ 710 µm) and the second, sieved (FP: X ≤ 210 µm).

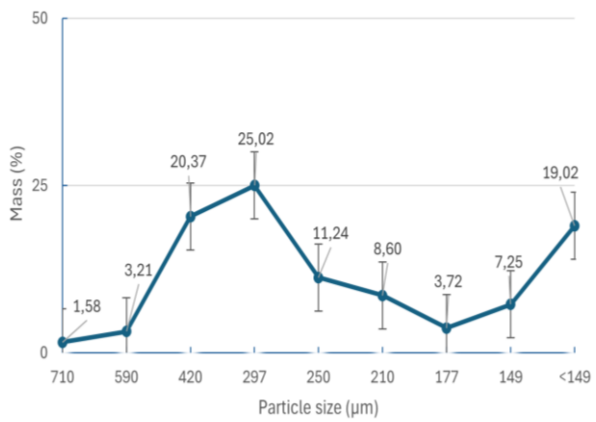


Figure 1. Particle size distribution for eucalyptus fiber.

ISSN: 2357-7592
XI INTERNATIONAL SYMPOSIUM ON INNOVATION AND TECHNOLOGY
Quantum Technologies: The information revolution that will change the future - 2025

The stress-strain curves, Figure 2, reveal significant differences depending on the mechanical properties of the composites. It is noted that the addition of eucalyptus fiber caused a significant increase in the elastic modulus for both composites, Table 1.

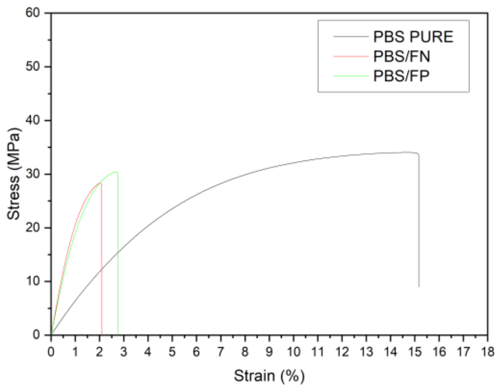


Figure 2. Stress x Strain curve for the samples.

For PBS/FN, the heterogeneous distribution of particles may have better filled the spaces between the different sizes of the fiber, creating a more interconnected stress network, reducing the porosity of the composite (reducing voids) and improving the packing density, making it denser and more rigid[6].

According to [7], the orientation of the fibers in different positions, for both composites, may also have contributed to the high modulus of elasticity presented, since fibers in the central position are fibers that are mainly aligned along the direction of flow and traction, where, in the presence of tension, this is applied directly to the fibers, resulting in materials with greater stiffness.

On the other hand, this gain in modulus of elasticity and hardness for PBS/FN, Table 1, was accompanied by the compromise of other mechanical properties, such as ductility and tensile strength.

The presence of particles with irregular sizes made it difficult to disperse the fiber in the matrix, creating clusters that acted as stress concentrators, favoring the propagation of cracks and reducing the final strength of the material, Figure 3. Furthermore, small cracks in the matrix were observed for PBS/FN, which suggests low efficiency of stress transfer from the matrix to the reinforcing agent [8].

For fibrous composites, there is a critical fiber length to obtain good properties, because the ends of the fibers are stress concentration points that weaken the material. The reduction in the size of the dispersed phase up to a certain limit increases the interfacial area and improves the transfer of stresses imposed on the system, thus increasing its resistance [9].

Samples	Tensile strength	Elongation at break	Modulus of elasticity	Impact resistance	Hardness
	Mpa	%	Mpa	J/m	Shore D
PBS PURE	33,69 ± 1,41^{a}	14,01 ± 0,47^{a}	702,13 ± 13,56^{a}	25,83 ± 1,70^{a}	63,37 ± 1,77^{a}
PBS/FN	28,40 ± 0,73^{b}	1,93 ± 0,06^{b}	2705,57 ± 54,17^{b}	18,47 ± 0,44^{b}	72,28 ± 0,76^{b}
PBS/FP	30,16 ± 0,59^{c}	2,53 ± 0,08^{c}	2348,23 ± 59,06^{c}	14,93 ± 1,05^{c}	72,21 ± 1,04^{b}

Table 1. Mechanical properties, impact resistance and hardness of the samples.

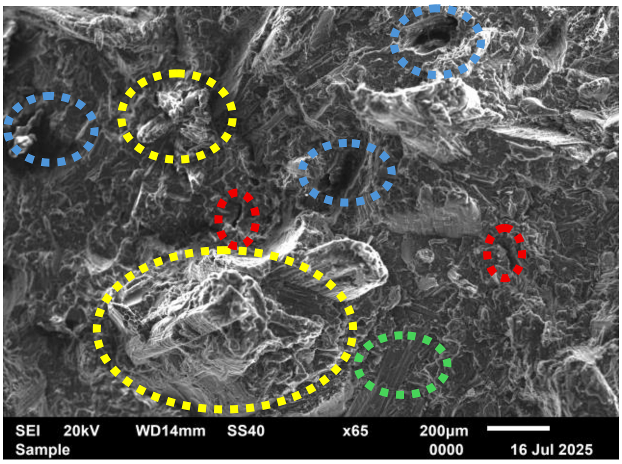
For PBS/FP, particle size uniformity guaranteed a greater surface area [10], promoting better wettability and regions with good adhesion between fiber-matrix, becoming more effective in stress distribution [11].

Despite the difference in polarity and low compatibility between the predominantly hydrophobic PBS polymer matrix and the hydrophilic components of the eucalyptus fibers, a partial transfer of tensions between the phases is observed, represented through extractions of fibers and regions with detachment, as shown in Figure 3.

From the SEM images, it is possible to verify that most of the failure of the composite materials (PBS/FN and PBS/FP) occurred due to the extraction without fracture of fibers from the matrix.

In other words, even with the absence of gaps between the fiber and the composite matrix, the interfacial interactions were not strong enough to resist fiber withdrawal during impact, resulting in low energy consumption (in magnitude).

PBS/FN



PBS/FP

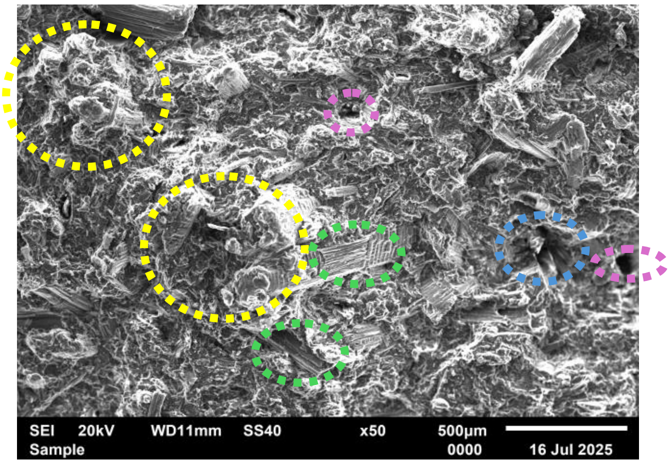


Figure 3. SEM of PBS/FN and PBS/FP composites. Fiber agglomerates (dashed yellow); Crack in the polymer matrix (red dashed lines); Fiber extraction (blue dashed lines); Fiber detachment (green dashed lines); Pores or voids (pink dashed lines).

In general, the loss in tensile strength and the marked reduction in the failure deformation of the composites compared to pure PBS is due to the process of adding vegetable fibers, itself, which limits the movement of the polymer chains, as well as their elongation, becoming a zone of structural weakness where the material can fail under tension [12].

Furthermore, the added fiber content of 30% did not allow for a homogeneous distribution in the polymer matrix, limiting the tensile property in the longitudinal direction to a maximum value. These results can be confirmed using rupture elongation and impact resistance data.

4. Conclusions

The incorporation of eucalyptus fiber into PBS promoted a significant gain in the modulus of elasticity and hardness for both composites compared to pure PBS, especially PBS/FN. However, its tensile strength and elongation at break value were lower compared to PBS/FP, due to the particle size uniformity that allowed a better surface area and, consequently, better wettability.

This study made it possible to verify that, although larger particles (PBS/FN) cause a more compact filling, smaller particles (PBS/FP) demonstrated more efficient general mechanical properties, showing that the dispersion of these particles is a critical point, since the natural propensity of fillers is to form aggregates.

Another relevant point refers to the concentration of fiber added (30%), which despite being considered high by some authors, for these composites, good matrix-fiber interfacial adhesion was observed, but still insufficient for a fully effective transfer of stress, being reflected in some limitations in the mechanical performance of the composites.

Finally, tests with the particle fraction of $X \leq 210$ µm (FP) showed promise, since this particle size range contributed to better performance of the composite compared to the fiber without sieving. Although the sieving separation process involves an additional cost, the benefits obtained in terms of interfacial interaction and mechanical properties justify its adoption.

As future perspectives, we highlight the need to investigate strategies to improve the dispersion of particles in the matrix, as well as the use of compatibilizing agents or surface treatments that favor adhesion in the matrix-fiber interface, aiming to improve the performance of the developed composites.

Agradecimentos

The authors would like to thank the Materials Characterization Laboratory at SENAI CIMATEC for the availability of equipment, DEXCO for donating the fibers and UFS for injecting the test specimens.

Referências

[1] Zeng, J. B.; Li, Y. D.; Zhu, Q. Y.; Yang, K. K.; Wang, X. L.; Wang, Y. Z. A novel biodegradable multiblock poly(ester urethane) containing poly(l-lactic acid) and poly(butylene succinate) blocks. Polymers. 2009; 50: 1178–1186.

[2] Thurber, H.; Curtzwiler, GW. Suitability of poly(butylene succinate) as a coating for cardboard convenience food packaging. In: International Journal of Biobased Plastics. 2020; 2: 1–12.

[3] IBÁ. Relatório Anual 2023: Indústria Brasileira de Árvores. Brasília, 2023.

[4] PTT MCC Biochem, 2023. BioPBS ™ FZ71PM / FZ71PB Technical Data Sheet Injection molding for general purpose.

[5] Horiuchi, L. N.; Barbosa, R. M.; Azevedo, J. B.; Villen, F. G.; Viseras, C.; Fialho, R. L. L. Eco-friendly composite materials of polybutylene succinate with clay minerals, lignin and canabrava fiber. Applied Clay Science. 2024; 262.

[6] Valente, B.F.A.; Silvestre, A.J.D.; Neto, C.P.; Vilela, C.; Freire, C.S.R. Effect of the Micronization of Pulp Fibers on the Properties of Green Composites. *Molecules*. **2021**; *26*: 5594.

[7] Qaiss, Abouelkacem & Bouhfid, Rachid & Essabir, Hamid. Effect of Processing Conditions on the Mechanical and Morphological Properties of Composites Reinforced by Natural Fibres. 2015.

[8] Phiri, Mohau Juiz; Mofokeng, Julia Puseletso, Phiri, Mapoloko Mpho, Mngomezulu, Mfiso, Tywabi-Negva, Zikhona. Chemical, thermal and morphological properties of polybutylene succinate-waste pineapple leaf fibres composites. Heliyon. 2023; 9 (11): 21238.

[9] BUCKNALL, C.B. Toughened Plastics. Applied Science Publishers, London (1977).

[10] Rocha, T. M. S.; Miranda, L. F. R.; Parchen, C. F. A. Influência da composição granulométrica das partículas de resíduos de madeira nas propriedades de compósitos minerais: parte 2: gesso-madeira. Associação Nacional de Tecnologia do Ambiente Construído. 2019; 19 (4): 195-206.

[11] SANTOS, Eylisson André dos. Avaliação mecânica e microestrutural de compósitos de matriz de poliéster com adição de cargas minerais e resíduos industriais. 2007. 103 f. Dissertação (Mestrado em Tecnologia de Materiais; Projetos Mecânicos; Termociências) - Universidade Federal do Rio Grande do Norte, Natal, 2007.

[12] P. Noorunnisa Khanam, M.A. AlMaadeed. Improvement of ternary recycled polymer blend reinforced with date palm fibre. Materials & Design. 2014; 60: 532-539, ISSN 0261-3069.A.C.O. CARNEIRO. Aproveitamento energético de resíduos da produção de compensados de eucalipto. Ciência Florestal. 2018; 28 (1): 428-439.