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Potential biodegradable materials containing oat hulls, TPS, and PBS by thermoplastic injection

Samuel Camilo da Silva[1]*, Fabiola Azanha de Carvalho[1] and Fabio Yamashita[1]

[1]Laboratório de Tecnologia, Departamento de Ciência e Tecnologia de Alimentos, Universidade Estadual de Londrina – UEL, PR, Brasil

**samuelcamiloutfpr@outlook.com*

Abstract

Fossil-origin plastics have raised great concerns due to their non-biodegradable nature. Biodegradable polymers can be an alternative for these materials', however they have higher cost. The use of agro-industrial waste in blends with biopolymers can provide cheaper materials' with improved properties. This study aims to develop low-cost biodegradable materials by extrusion and thermoplastic injection using oat hulls, polybutylene succinate (PBS), and starch. Six formulations with different concentrations of oat hulls (0-56% w/w) were extruded in a single-screw extruder, and then the materials were produced by thermoplastic injection. The extrusion aligned the oat hull fibers, making the material dimensionally stable. The oat hulls enhanced stiffness and reduced material density compared to non-hull counterparts. Besides that, the oat hulls are a low-cost agro-industrial byproduct, and it was possible to produce biodegradable materials with up to 56% hulls and only 20% PBS. These biodegradable materials are environmentally friendly and non-toxic.

Keywords: *biodegradable materials, blending, extrusion, natural fibers, mechanical properties.*

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1. Introduction

The escalating production of fossil-origin materials, currently amounting to 350 million tons annually, has sparked growing concerns due to their non-biodegradable nature and significant impact on the ecosystem[1].In response to these challenges, biodegradable polymers have emerged as an alternative to fossil-origin materials. According to ASTM standards[2], biodegradable polymers undergo decomposition in natural aerobic environments (composting) through the metabolic activity of microorganisms capable of metabolizing their molecular structures. Despite their potential, biodegradable polymers still face limitations compared to synthetic polymers, particularly mechanical properties, water vapor barriers, and production costs, making large-scale adoption challenging[3-5]. The utilization of blends, are well known for reducing material costs and improving specific properties[5-7]. Extensive research has investigated the incorporation of different components, such as starch[8,9] and fibers[10,11], particularly agro-industrial residues or byproducts[3,12].

Polybutylene succinate (PBS) is a biopolymer from the family of polyesters that can be processed through injection or extrusion to produce rigid or flexible materials comparable to polypropylene/polyethylene. PBS presents excellent biodegradation properties in soil and water[13]. Additionally, starch, which is widely available from diverse sources (corn, potato, and cassava), can also be effectively processed through injection or extrusion to produce biodegradable materials at a lower cost compared to petroleum-derived or aliphatic polyesters[7,14-18]. A significantly underutilized agro-industrial residue with high potential is oat hulls, a byproduct of oat milling processing, yielding approximately 25-36% during milling[19,20]. Despite their versatility, oat hulls are primarily adopted as biomass for generating electricity and steam[21], overlooking its potential for other valuable applications, such as in biodegradable blends. The low density and weight of these natural fibers can be attractive in applications where lighter and more rigid materials are desired, such as in the coating of cups, straws, spoons, single-use trays for some foods, in the automotive industry, tissue engineering, and marine industry, among others[10,22,23].

Combining oat hull, PBS, and starch can ensure biodegradability, lower cost, and reasonable material properties. This could guarantee the production of a new material with many possible applications, such as single-use trays, food packaging, among others, using a by-product considered to be waste in the oats industry, while making it cheaper and more environmentally friendly. To do this, it is necessary to study the behavior of oat hulls at different concentrations to understand its effect as a fiber-reinforced agent. This study aims to develop low-cost biodegradable materials by extrusion and thermoplastic injection using oat hulls, PBS, and starch.

2. Materials and Methods

2.1 Material

The materials used in this study included corn starch (14% moisture) (APTI™,Brazil), glycerol (technical grade glycerin, Dinâmica, Brazil), polybutylene succinate (PBS) (TK-BIO®, China) (Table 1), and oat hull (SL-Alimentos, Brazil). Before material production, oat hulls were grounded (Mill An 11, Brazil) and sieved (28 *mesh*) to enhance their homogeneity.

2.2 Production of biodegradable materials by thermoplastic injection

Preliminary tests were conducted using different concentrations of PBS, starch, and oat hulls to evaluate their processability in a pilot single-screw extruder (model EL-75, BGM, Brazil) and pilot injector AX16-lll (AX-Plasticos, Brazil). Three formulations (T20: 20 PBS %-w/w; 44,8 Starch %-w/w; 24 Glycerol %-w/w; 11,2 Oat hulls %-w/w; T60: 20 PBS %-w/w; 22,4 Starch %-w/w; 24 Glycerol %-w/w; 33,6 Oat hulls %-w/w; T100: 20 PBS %-w/w; 0 Starch %-w/w; 24 Glycerol %-w/w; 56 Oat hulls %w/w.) were produced in a pilot single screw extruder (90/120/120/110°C) using a temperature profile from heating zone 1 to zone 4 at a screw speed of 40 RPM. After extrusion, they were processed in the pilot injector to produce a dog bone-shaped specimen type IV[24]. The temperature profile was set at 120/120/110°C from the feeder to the nozzle. Based on the preliminary tests, it was decided to set the concentration of PBS and glycerol (plasticizer) constant and vary the concentrations of starch and oat hulls, as detailed in Table 1. All material was manually mixed and processed as described in the preliminary tests.

2.3 Mechanical properties

Mechanical tensile tests (Young's modulus, tensile strength, and elongation at break) were performed according to ASTM 638-14[24] using a universal testing machine (EMIC, model DL 2000, Brazil). Before the analysis, the specimens were conditioned at room temperature, maintaining a relative humidity of 53 ± 2% for one week as per ASTM 638-14[24] standard procedures.

2.4 Scanning electron microscopy (SEM)

The microstructure images of the samples were captured using a scanning electron microscope (Philips, model FEI Quanta 200, USA). Before imaging, the specimens were subjected to cryogenic fracture by immersion in liquid nitrogen and metalized with a thin layer of gold using a metallizer [Bal-Tec, model SCD-050, Germany]. All samples were analyzed using a 20 kV voltage accelerator and 2000x magnification.

2.5 X-ray diffraction (XRD)

The crystallinity of the biodegradable materials was determined using X'PertPRO equipment (Panalytical, Philips, Netherlands) with copper Ka radiation (α = 1.5418 A) operating at room temperature at 40 kV. The relative crystallinity index (RCI) was calculated as the ratio between the area of the crystalline region and the sum of crystalline and amorphous regions (Equation 1)[9,25].

$$RCI = \frac{C_a}{C_a + A_a} \quad (1)$$

Where C_a is the crystalline area, and $A_a$is the amorphous area.

2.6 Density

Density was calculated by weighing the mass and measuring the volume using a digital caliper (Starrett, Brazil). Five specimens from each formulation were placed in a desiccator for one week under a relative humidity of 53%.Then, the specimens were weighed and measured to obtain thickness, length, and width.

2.7 Linear Contraction Index (LCI)

After production, ten specimens from each formulation were measured using a digital caliper (Starrett, Brazil). These specimens were subsequently placed in desiccators at a relative humidity of 53% and a temperature of 25°C for one week. The linear contraction index (LCI) was determined using Equation 2[26].

$$LCI(\%) = \left(\frac{Lcm - Lcp}{Lcm}\right) x100 \quad (2)$$

Where Lcm is the initial specimen length and Lcp is the length after one week.

2.8 Moisture sorption isotherms

Approximately 0.5 to 0.8 g of the biodegradable material was placed in an Aquasorp isotherm generator (Decagon Devices, USA), and the experimental moisture sorption data was fitted by the Guggenhein-Anderson-de-Boer (GAB)

Table 1. Formulation of biodegradable material containing PBS, starch, and oat hulls,

Formulation	PBS (%- w/w)	Starch (% - w/w)	Glycerol (% -w/w)	Oat Hulls (% - w/w)
PBS	100	0	0	0
F0	20.0	56.0	24.0	0
F20	20.0	44.8	24.0	11.2
F40	20.0	33.6	24.0	22.4
F60	20.0	22.4	24.0	33.6
F80	20.0	11.2	24.0	44.8
F100	20.0	0	24.0	56.0

model (Equation 3) using the Nonlinear Regression Module (Statistica Software 7.0,StatSoft, USA).

$$X_w = \frac{C.k.m_o.a_w}{(1-k.a_w)(1-k.a_w+C.k.a_w)} \quad (3)$$

Where aw = water activity, X_w = equilibrium moisture content (kg/kg dry solid), m_0 = monolayer moisture content (kg/kg dry solid), and C and k = GAB constants.

2.9 Color

The CIELabcolor parameters (L*,a*, and b*) of five specimens of each formulation were measured with a colorimeter (Minolta CR 400, Japan) with a visual angle of 10°, according to ASTM D2244-09B[27].

2.10 Statistical analysis

All data obtained were evaluated by analysis of variance (ANOVA) and Tukey's test at a 5% significance level ($p < 0.05$) using Statistica software, version 7.0 (StatSoft, USA).

3.Results and Discussions

3.1 Mechanical properties

The results of the mechanical properties of the biodegradable materials are shown in Table 2.

The F0 formulation had the highest tensile (TS) strength (1.8 MPa), approximately double that of F100 (0.9 MPa). The higher the oat hull concentration, the lower the TS, probably due to the limited interfacial adhesion between the oat hulls and the polymeric matrix. The interaction between the polymeric matrix (PBS) and the oat hull is difficult due to the hydrophilic nature of the hull[28]. According to Mochane et al.[10], the hydrophilic characteristics of natural fibers may adversely affect interfacial adhesion, mainly when the polymeric matrix is hydrophobic, as the PBS, resulting in poor mechanical properties[29,30]. Other factors, such as fiber length, fiber type, extraction process, and moisture absorption, can decrease the TS[30].

According to Aydemir and Gardner[31], in blends of polyhydroxybutyrate and polylactic acid reinforced with cellulose nanofibrils, increasing the fiber concentration did not improve the mechanical properties, and there was a greater agglomeration and clogging of the fibers in the matrix structure, enhancing the stiffness and ductility of the material. Ayu et al.[32] produced sheets of PBS, starch, and empty fruit buncher fiber (EFB), and according to the authors, adding fiber fillers up to 8 wt% decreased tensile and flexural strength due to a lack of interfacial adhesion and poor dispersion of the fibers in the PBS matrix. As the literature indicates, incorporating natural fibers in blends to improve mechanical properties can present some difficulties due to fiber dispersion, polar and non polar phases, fiber concentration, and extrusion process

The F0 material had the highest flexibility, possibly due to its high thermoplastic starch content, which increases its elasticity[33]. Thermoplastic starch (TPS), when in the presence of a plasticizer such as glycerol, has its flexibility increased by lowering the glass transition temperature (Tg),which can be beneficial for some applications such as food packaging and films[34]. The material with 20% oat hull content (F20) had approximately 7% lower elongation than F0. Calabia et al.[35] produced sheets of PBS and cotton fiber and reported a decrease in the elongation at break of approximately 8% in formulations containing fiber. This decrease in elongation was attributed to the reduction in PBS chain movement, leading to higher stiffness in the materials. Similarly, Yang et al.[36] produced injected materials using bamboo fiber and polypropylene, and they found that as the fiber concentration increased, the elongation at break (%) decreased. This decrease resulted from difficulties maintaining fiber dispersion in the blend, which adversely affected material flexibility. The same occurred in this study, where the material became more stiff and less flexible.

3.2 Scanning electron microscopy

The images of the F0 material (Figure 1a, Figure 1b) showed starch granules (circular shape) and a plasticized superficial area, characteristic of blends containing thermoplastic starch[37,38]. Cracks were also observed due to the poor compatibility between PBS and starch, forming a heterogeneous phase (immiscible blend). Similar characteristics can create a strong bond, resulting in dimensional stability and improved mechanical and barrier properties[39,40].

The F20 (Figure 2a, Figure 2b), F40 (Figure 3a, Figure 3b), F60 (Figure 4a, Figure 4b), F80 (Figure 5a, Figure 5b), and F100 (Figure 6a, Figure 6b) materials' surface and fracture images showed oat hull fibers (cylindrical shape), most of which were agglomerated and aligned, possibly due to extrusion orientation. Many cavities and pores were also observed in the formulations containing oat hulls, which might explain the decrease in the mechanical properties of these materials. Similar observations were reported by Calabia et al.[35] for PBS and cotton fiber composites and Ayu et al.[32] for sheets containing empty fruit bunches, PBS,

Table 2. Mechanical properties of biodegradable materials produced by thermoplastic injection,

Formulation	Tensile Strength (MPa)	Elongation at Break (%)	Young's Modulus (MPa)
F0	1.8 ± 0.1^{a}	20.2 ± 1.3^{a}	13.8 ± 2.8^{c}
F20	1.6 ± 0.1^{ab}	13.3 ± 0.9^{b}	15.9 ± 6.6^{c}
F40	1.4 ± 0.1^{bc}	11.1 ± 1.3^{c}	22.6 ± 8.2^{bc}
F60	1.2 ± 0.1^{cd}	08.3 ± 2.2^{c}	24.5 ± 10^{b}
F80	1.1 ± 0.1^{df}	7.4 ± 0.6^{de}	17.9 ± 5.2^{c}
F100	0.9 ± 0.1^{f}	6.5 ± 0.4^{e}	29.6 ± 6.6^{a}

[a,b,c,d,e]Means at the same column with different letters represent a significant difference ($p \leq 0.05$) between theformulations, according to Tukey's test.

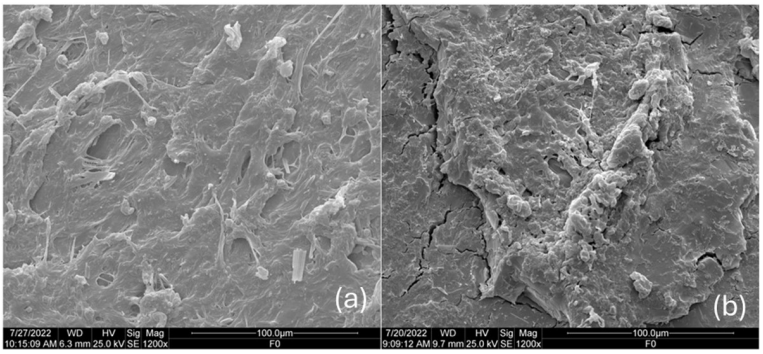

Figure 1. (a) F0 Formulation (100.0 µM); (b) F0 Fractured (100.0 µM).

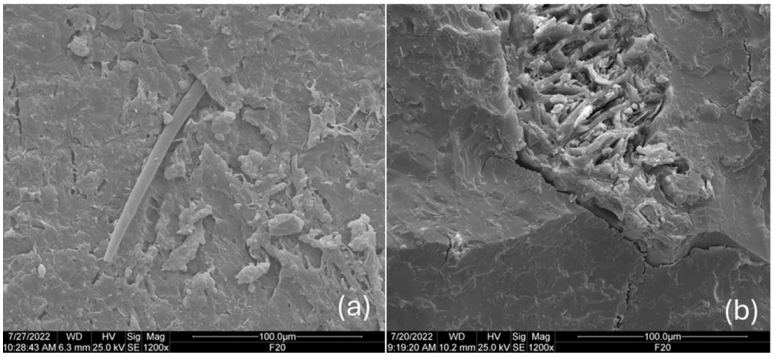

Figure 2. (a) F20 Formulation (100.0 µM); (b) F20 Fractured (100.0 µM).

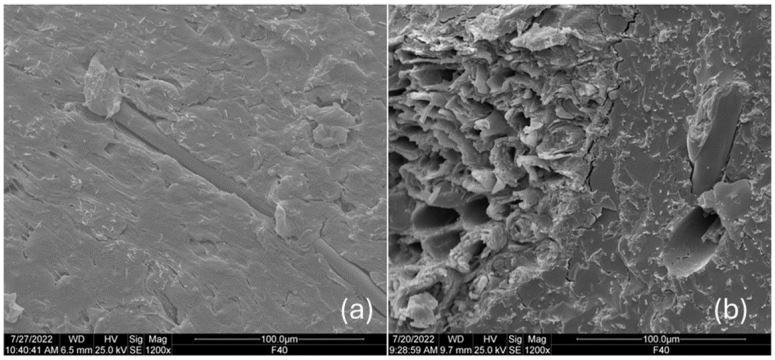

Figure 3. (a) F40 Formulation (100.0 µM); (b) F40 Fractured (100.0 µM).

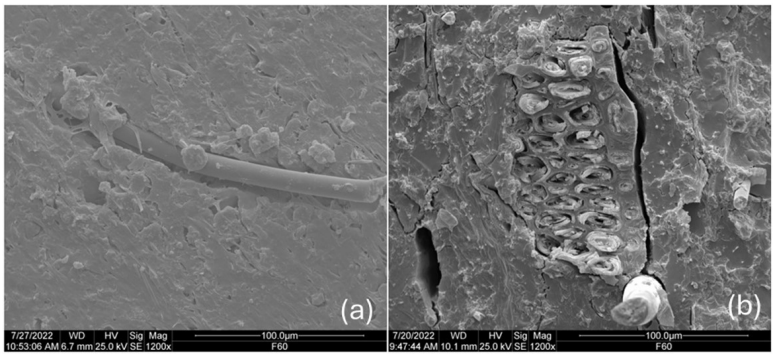

Figure 4. (a) F60 Formulation (100.0 µM); (b) F60 Fractured (100.0 µM).

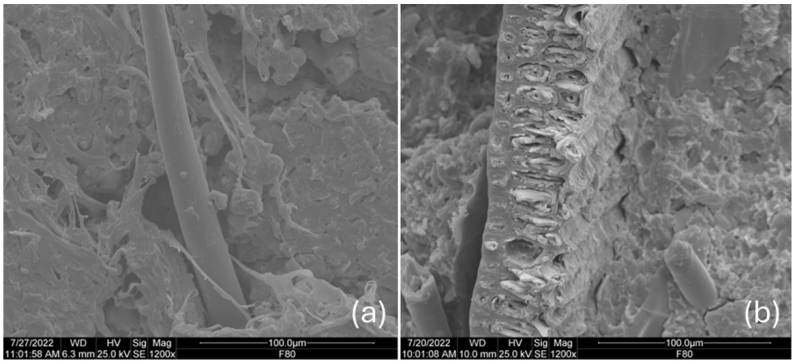

Figure 5. (a) F80 Formulation (100.0 µM); (b) F80 Fractured (100.0 µM).

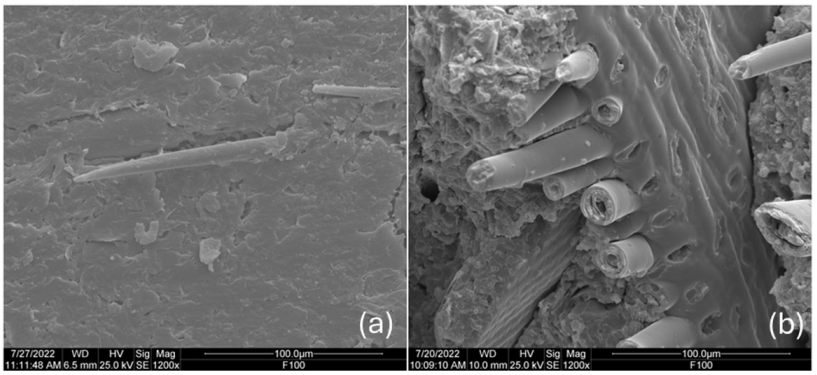

Figure 6. (a) F100 Formulation (100.0 µM); (b) F100 Fractured (100.0 µM).

and starch. In both studies, it was possible to observe the presence of long fibers and voids in the structure, indicating weak interfacial adhesion.

Mechanical and barrier properties highly depend on morphology status[10,32]. Some methods can improve the interfacial surface between the components, such as treatments with silanes, alkalis, or acetones, reducing incompatibility[12]. However, the work aimed to produce biodegradable and nontoxic materials, and chemical or surface cleaning methods can harm the environment.

3.3 X-ray diffraction (XRD)

The X-ray diffractograms and the respective relative crystallinity index (RCI) of the biodegradable materials are presented in Figure 7.

Two peaks (19.9° and 22.3°) were identified in all blend formulations, and the oat hull concentration did not influence the crystallinity of the materials. Hu et al.[41] produced blends containing PBS and different types of cellulose, and all diffractograms showed reduced peaks, suggesting that the natural fibers used had low crystallinity. Liu et al.[38] produced materials with starch, PBS, and ionic liquid; varying starch content did not modify the PBS crystalline phase.

PBS showed peaks at approximately 19.6°, 22.3°, and 28.8°, and peaks near 19.5° and 22.5° are characteristic of the crystalline phase of PBS[42,43]. PBS also showed a larger peak at 22.3° when compared to the other formulations because

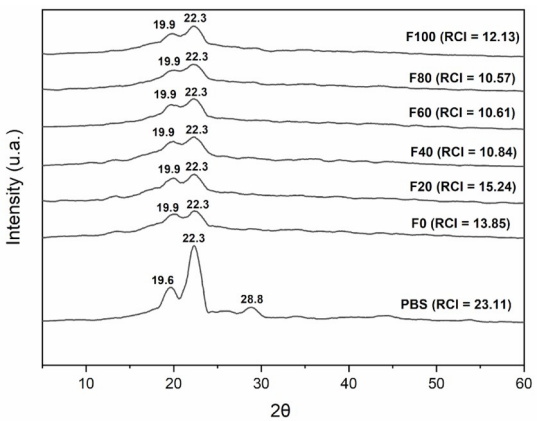

Figure 7. X-ray diffractograms and relative crystallinity index (RCI) of the biodegradable materials.

the addition of starch, oat hulls, and glycerol impaired the crystallinity properties of PBS during the production/extrusion process of the materials. This same behavior was observed by Xu et al.[43] for blends produced with PBS and corn starch, possibly as starch particles obstruct PBS segments. The low relative crystallinity indexes (RCIs) of the biodegradable materials containing starch, ranging from 10.57% to 15.24%, can be attributed to the destruction of the semicrystalline

structure of starch during the extrusion process, leading to the formation of higher amorphous zones[8,44].

3.4 Linear Contraction Index (LCI)

The linear contraction indexes (LCIs) of the biodegradable materials are presented in Table 3. F0 (1.56%) and F20 (1.07%) had the highest LCI values. LCI of the other materials was not significantly different and ranged from 0.70 to 0.78%. Materials produced by injection molding can contract as they change from melting to solid under atmospheric pressure[45].

Increasing the fiber concentration resulted in a lower contraction of the injected material because the fibers can act as fillers, i.e., occupying spaces in the blend structure and making it less capable of shrinking or expanding[46,47]. This can be advantageous since it avoids the appearance of marks on thicker parts of the materials, provides good geometrical stability,avoids deformation after injection, and reduces material shrinkage[47]. The SEM analysis of the materials (item 3.2) with oat hulls showed that the fibers were oriented and agglomerated in a cylindrical shape, suggesting that these aligned and clogging fibers decreased the materials' LCI. The LCI can also be influenced by temperature, pressure, injection flow rate, equipment design, and material composition[45,48].

Table 3. Linear Contraction Index (LCI) of the biodegradable materials.

Formulation	LCI (%)
F0	1.56 ± 0.06^{a}
F20	1.07 ± 0.06^{b}
F40	0.78 ± 0.04^{c}
F60	0.75 ± 0.05^{c}
F80	0.74 ± 0.03^{c}
F100	0.70 ± 0.04^{c}

*a,b,c Means with different letters represent a significant difference ($p \leq 0.05$) between theformulations, according to Tukey's test.

Table 4. Density of the biodegradable materials produced by injection extrusion.

Formulation	Density (g 100 g^{-1})
F0	1.34 ± 0.02^{b}
F20	1.32 ± 0.04^{b}
F40	1.35 ± 0.04^{b}
F60	1.32 ± 0.04^{b}
F80	1.35 ± 0.03^{b}
F100	1.28 ± 0.02^{a}

a,b Means with different letters represent a significant difference ($p \leq 0.05$) between the formulations, according to Tukey's test .

3.5 Density

F100 had the lowest density, and the other formulations did not show significant differences (Table 4) because of the lower density of the PBS compared to thermoplastic starch (TPS). Additionally, the high oat hull content in the F100 formulation contributed to decreased material density, as fibers have lower densities than TPS. One of the main advantages of adding natural fibers in polymeric structures is to reduce the material density[10,49,50]. This can lead to applications where lighter and less flexible materials are desired, such as food packaging, single-trays, cups, and the automotive industry[22,23]. Aslan et al.[51] produced composites of polypropylene and sisal fiber, fiberglass, and carbon fiber, and increasing sisal fiber concentration reduced the material's densities. The density of the material decreases with the addition of natural fibers in the blend in most cases.

3.6 Moisture sorption isotherms

The Guggenheim-Anderson-de-Boer (GAB) parameter values of the moisture sorption isotherms of the biodegradable materials are presented in Table 5.

The m_0 of the F0 material (16.11 g 100 g^{-1}) was the highest among all formulations, mainly due to its higher proportion of thermoplastic starch (TPS) and the absence of oat hull because TPS is known for its high hydrophilicity[8]. As the oat hull content increased (F20, F40, F60, F80, F100), the m_0 values decreased compared with F0 material due to the reduction of TPS concentration in the blend. The F100 material had no starch in its composition and had an intermediary m_0 value between F0 and F80 because the oat hull is less hydrophobic than PBS but more hydrophobic than TPS. The k and C values were similar for all formulations.

3.7 Color

The CIELabcolor parameters L*, a*, and b* of the biodegradable materials are presented in Table 6.

F0 had the highest luminosity (62.69), and the formulations containing oat hulls had lower values, ranging between 41 and 45. The decrease in luminosity can be attributed to the opaque nature of oat hulls, which can significantly influence the color parameters. For the a* parameter F20 presented the highest value (4.09). The b* parameter of the F0 material

Table 5. GAB model parameters of the moisture sorption isotherms of the biodegradable materials.

Formulation	m_0(g 100 g^{-1})	k	C	R^2
F0	16.11	0.78	10.000	0.97
F20	4.66	1.01	10.000	0.99
F40	4.56	1.01	10.000	0.98
F60	2.43	1.06	10.000	0.99
F80	2.31	1.06	10.000	0.99
F100	4.03	1.02	10.000	0.99

Formulations containing F - Oat Hulls in different concentrations.

Table 6. CIELabcolor parameters of the biodegradable materials.

Formulation	L*	a*	b*
F0	62.69 ± 0.27	-2.71 ± 0.13	9.79 ± 0.26
F20	42.82 ± 0.47	4.09 ± 0.25	2.57 ± 0.31
F40	41.54 ± 0.74	3.17 ± 0.09	0.54 ± 0.29
F60	43.49 ± 1.91	2.62 ± 0.22	1.05 ± 0.92
F80	45.29 ± 3.50	2.47 ± 0.29	2.09 ± 1.97
F100	42.69 ± 2.03	2.84 ± 0.22	1.66 ± 1.29

The L* parameter is luminosity; a* red/green coordinate; b* yellow/blue coordinate.

(9.79) was higher than that of the others because it did not contain an oat hull in its formulation, only PBS and starch, resulting in a yellowish color.

In this study, all the materials with oat hulls had a brownish color. The oat hulls are composed of pentosans (30-35%) and protein (4%)[20], which are components that can contribute to the Maillard reaction during the extrusion process at high temperatures (90-110°C). This Maillard reaction could explain the color alteration observed in the materials with oat hulls.

4. Conclusions

Biodegradable materials of oat hulls, starch, and polybutylene succinate presented excellent processability at the extrusion, to produce the pellets, and at the thermoplastic injection, to produce the dog bone-shaped specimens. The extrusion aligned the oat hull fibers, making the material dimensionally stable, i.e., without the shrinkage observed in materials without fiber. Furthermore, incorporating oat hulls enhanced stiffness and reduced material density compared to non-hull counterparts. These materials' are adequate to produce lighter and stiffer materials for commercial uses, such as fast-food packaging and single-use utensils like trays, spoons, and cups. Besides that, the oat hulls area low-cost agro-industrial byproduct, and it was possible to produce biodegradable materials with up to 56% hulls and only 20% PBS. Their potential for large-scale manufacturing is highlighted by their compatibility with existing equipment and processes in the plastics industry. For future projects, new studies using FTIR spectroscopy can be carried out to better understand the interactions between oat hulls, TPS and PBS.

5. Author's Contribution

- **Conceptualization –** Fabio Yamashita.
- **Data curation –** NA.
- **Formal analysis –** Samuel Camilo da Silva; Fabiola Azanha de Carvalho.
- **Funding acquisition –** Fabio Yamashita.
- **Investigation –** Samuel Camilo da Silva; Fabiola Azanha de Carvalho.
- **Methodology –** Fabio Yamashita.
- **Project administration –** Fabio Yamashita.
- **Resources –** Fabio Yamashita.
- **Software –** NA.
- **Supervision –** Fabio Yamashita.
- **Validation –** Samuel Camilo da Silva; Fabiola Azanha de Carvalho.
- **Visualization –** Samuel Camilo da Silva; Fabiola Azanha de Carvalho; Fabio Yamashita.
- **Writing – original draft –** Samuel Camilo da Silva; Fabio Yamashita.
- **Writing – review & editing** – Fabio Yamashita.

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