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SUSTAINABLE ALTERNATIVES FOR PACKAGING: EXPLORING THE HIGROSPICITY OF BIODEGRADABLE MATERIALS BASED ON PLA, PBAT, AND RICE HUSK

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Abstract - The present study aims to develop a sustainable material using biodegradable polymers combined with agroindustry residues, to find more sustainable options to reduce the negative environmental impact of single-use food packaging. Different formulations containing polylactic acid (PLA), polybutylene adipate-co-terephthalate (PBAT) in the form of blends (BL), and composites (CO) reinforced with 10 and 20% of rice husk (RH) were prepared to identify the most appropriate formulation to produce a material biodegradable with the specific properties required for its application in packaging. The samples were characterized using moisture absorption, water absorption, and contact angle measurements. The results showed that PLA, PBAT, BL1, and BL2 behave similarly. Furthermore, it was observed that samples containing lower RH content (10%wt) showed lower hygroscopicity and hydrophilicity compared to the other composites. Concluding, as well as the PLA/PBAT blends, the composite materials containing rice husk are potential substitutes for single-use food packaging.

Keywords: PLA, PBAT, Rice Husk, Biodegradable Packaging, Sustainability.

Introduction

The growing concern about environmental pollution caused by the accumulation of plastic waste has driven the search for sustainable alternatives for single-use food packaging. Faced with these concerns, it is essential to seek sustainable solutions and adopt ecologically responsible practices to minimize the negative impact caused by single-use packaging waste on our planet ^[1]. Therefore, the development of biodegradable materials for packaging has gained prominence as a promising solution to replace non-biodegradable conventional polymers ^[2, 3].

One of the alternatives is using biodegradable polymers, such as polylactic acid (PLA) and polybutylene adipate-co-terephthalate (PBAT), which come from renewable sources and have characteristics suitable for packaging applications ^[4, 5]. PLA is a polymer obtained from renewable sources, such as corn starch, and is biodegradable in industrial composting ^[3]. Conversely, PBAT is a copolymer combining polybutadiene and adipate terephthalate properties, being flexible and biodegradable in composting ^[4].

These biodegradable materials have shown promise as alternatives to conventional polymers in packaging, since they have physical and mechanical properties suitable for various applications, such as food transport and storage, protective packaging, and packaging for electronic products, among others ^[5]. In addition, these materials are biodegradable; they can decompose naturally in the environment, reducing the accumulation of plastic waste and minimizing the environmental impact. However, in addition to the biodegradable polymer options already available on the market, it is essential to emphasize that the selection of materials to develop new formulations depends on the specific properties, availability, and cost of the raw materials used. In this context, developing new

formulations of sustainable materials by combining biodegradable polymers with residues, such as rice husks (RH) is interesting. Rice husk is rich in cellulose, lignin, and other organic components, which makes it a residue with a solid potential to produce sustainable materials for food packaging. In addition, the use of agroindustry residues contributes to the reduction of the environmental impact, since the use of a certain percentage by mass of rice husk in the formulation of the biodegradable material will help to reduce the amount of agricultural waste improperly discarded, promoting a more sustainable approach ^[6].

Therefore, the purpose of the present study is to develop a sustainable material using PLA/PBAT combined with rice husks (RH), aiming to replace single-use food packaging. The mixture of biodegradable polymers with agroindustry residues can be an exciting strategy for developing eco-friendly packaging, considering the availability, properties, and costs of the raw materials, as well as the compatibility and desired performance for the specific application.

Experimental

Materials

For the formulation of the samples, two biodegradable polymers were selected, PLA (Ingeo[™] Biopolymer 4043D) and PBAT (Ecoflex® F Blend C1200); and as filler, rice husks (RH) provided by local producers. Different formulations were produced to determine the most suitable formulation for getting the most cost-effective biodegradable material, as specified in Table 1.

Samples	PLA (%)	PBAT (%)	RH (%)
PLA	100	-	-
PBAT	-	100	-
BL1	90	10	-
BL2	80	20	-
CO1	80	10	10
CO2	70	10	20
CO3	70	20	10
CO4	60	20	20

Table 1 – Formulation of samples.

The materials were mixed and homogenized in a HAAKE Rheomix OS Polylab mixing chamber, to obtain blends (BL1 and BL2) and composite materials (CO1, CO2, CO3, and CO4). Then, the mixtures were compressed in a hydraulic press at 170 °C and, subsequently, the trays were made by thermoforming.

Characterization Methods

To compare the hygroscopicity of the thermoformed polymeric plates, according to the ASTM D5229 standard, a moisture absorption test was carried out in controlled relative humidity chambers at a constant temperature of 30 °C, using 75% relative humidity saturated saline solution. The water absorption content was evaluated through the immersion test in distilled water, according to ASTM D570, to determine the degree of hydrophilicity of the samples. And a wettability test was performed based on the ASTM D7334 standard to complement the barrier study of biodegradable materials. In this test, the images were acquired through a contact angle measurement system with a digital optical microscope, and the contact angle calculations were performed using the Surftens software. The statistical analysis of the variance (ANOVA) of obtained results has been carried out using the software Origin. A one-way ANOVA and a Tukey's test were used to check for statistical differences among groups ($p \le 0.05$).

Results and Discussion

Moisture Absorption

The humidity test was carried out in a controlled environment to analyze the behavior of the samples in the long term, where measurements were carried out every seven days until the increase in mass was considered substantially saturated, totaling 35 days, as shown in Fig. 1.



Figure 1 – Result of the average moisture absorption content of the analyzed samples in the long term.

In Fig. 1 it is possible to observe that, in the first seven days, moisture absorption was faster. According to Ighalo *et al.*, this is because the driving force of mass transfer is initially very high ^[7]. However, all samples achieved stability in moisture absorption after 14 days of testing, with values below 0.5%. An analysis of variance (ANOVA) was performed to evaluate the measurements found for the samples after 35 days of testing. Thus, statistically, it was verified that there are no significant differences between the PLA, PBAT, BL1, and BL2 samples. Regarding the PLA/PBAT/RH composites, it was found that CO2 (70|10|20) absorbed more moisture, with a value of 0.45%, followed by CO4(0.39%), CO3(0.35%) and CO1(0.33%). The greater amount of absorbent material present in the CO2 and CO4 composite formulation can explain this. However, it is important to point out that even with this higher moisture absorption, the value remained below 0.5%, which is considered satisfactory for application in packaging according to ISO 187.

Water Absorption

The hydrophilic nature of rice husk makes water absorption a critical aspect of producing materials for use in packaging ^[8,9]. To analyze the behavior of the samples in the long term, immersion tests were carried out, where measurements were carried out every seven days until the increase in mass was considered substantially saturated, totaling 42 days, as shown in Fig. 2.



Figure 2 – Result of the average water absorption content of the analyzed samples in the long term.

As predicted, the result showed that the PLA/PBAT/RH samples had a higher water absorption content. Because, the rice husk is composed of different types of components, with cellulose being one of the main constituents. In addition to cellulose, rice husk contains lignin, hemicellulose, silica, proteins, and lipids in varying amounts ^[8]. Although its composition varies, rice husk has strong hydrogen bonds due to its basic cellulose constitution, being highly hydrophilic ^[9]. Therefore, when incorporated into a polymeric matrix, rice husk can act as a water retention agent and, in addition, increase the porosity of the composite material. This can allow for greater penetration and retention of water in the matrix, resulting in greater water absorption ^[10].

From the analysis of variance (ANOVA), it can be concluded that there are no significant differences between the samples PLA, BL1, and BL2. In the case of PLA/PBAT/RH composites, the results showed that CO2 and CO4 are statistically equal, as well as CO1 and CO3, which means that these composites have similar behavior in terms of water absorption.

However, it is important to emphasize that the CO1 and CO3 samples showed minor water absorption compared to the other composites, suggesting more significant interfacial interaction and homogeneity of the samples. Suggesting that these samples are more suitable for applications where water resistance is an important property.

Contact Angle

The contact angle test was carried out to study the effect of incorporating fibers into the polymeric matrix, specifically on the surface's hydrophobicity changes. For such, after the deposition of the drop of water on the surface, images were captured at 3 and 180 seconds to analyze the surface wettability of the composite material. Figure 3 shows the deposited water droplet pictures on the analyzed samples, and Fig. 4 shows the results of contact angles.



Figure 3 – Images of water droplets on the surface of the samples in 3 and 180s, respectively: (a) PLA; (b) PBAT; (c) BL1; (d) BL2; (e) CO1; (f) CO2; (g) CO3 e (h) CO4.



Figure 4 – Comparative result of the contact angle of the analyzed samples.

In Fig. 3, comparing the behavior of water droplets under the surface of the samples, it is possible to suggest that the PBAT is more hydrophobic than the other samples, as less water droplet scattering is seen.

Regarding the composite materials, the results indicate that the CO2 and CO4 composites have greater hydrophilicity, which may be due to a higher rice husk content (20%wt) and, consequently, a lower homogeneity in the sample than the other analyzed composites. On the other hand, the CO1 and CO3 composites showed lower hydrophilicity, highlighting CO3.

It is noteworthy that despite the addition of rice husk, the composites showed good contact angles. This feat can be explained by the Cassie-Baxter phenomenon; when a textured surface comes into contact with a liquid, the liquid does not completely wet the surface, forming small air pockets trapped between surface irregularities. In this case, textured surfaces appear more hydrophobic than smooth surfaces ^[11].

Conclusions

The results obtained showed that the PLA, PBAT, BL1, and BL2 samples showed similar behavior. Furthermore, the CO1 and CO3 composites showed lower hygroscopicity and hydrophilicity when compared to the other composites. Concluding that, as well as biodegradable polymer mixtures, composite materials with a biodegradable polymeric matrix combined with rice husk are potential substitutes for single-use food packaging due to their barrier properties. However, further studies are necessary to investigate the mechanical properties, biodegradability, and potential for industrial-scale production.

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