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# Synthesis and Characterization of Eco-friendly Bioplastics from Tapioca Starch, Chitosan, and Carrageenan with the Addition of Sugarcane Bagasse Microcellulose

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solvent casting;
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tapioca starch.

**ABSTRACT.** The increasing global demand for plastic has become a serious threat to the environment due to environmental pollution. Eco-friendly materials, such as bioplastics, have become a necessity. This study aims to synthesize tapioca starch-based chitosan carrageenan bioplastics with the addition of bagasse microcellulose. The bagasse microcellulose was obtained by hydrolysis. Bioplastics were produced using the solvent casting method, with fixed masses of tapioca starch, chitosan, and carrageenan of 4 g, 0.49 g, and 2.4 g, respectively. The mass of sugarcane bagasse microcellulose was varied as follows: 0 g (F0), 0.2 g (F1), 0.4 g (F2), 0.6 g (F3), 0.8 g (F4), and 1 g (F5). The test parameters in this study were density, moisture content, opacity, and surface morphology of bioplastics. The test results showed that adding bagasse microcellulose increased bioplastic density and opacity and reduced moisture content as the filler amount increased. The optimum formulation was obtained in F3, with a density of 1.459 g/mL, a moisture content of 14.652%, and an opacity of 2.373 /mm. Based on the characteristics produced, the tapioca starch-chitosan-carrageenan-sugarcane bagasse microcellulose bioplastic aligns with the direction of environmentally friendly bioplastic development in accordance with SNI 7188-7:2022.

## INTRODUCTION

Conventional petroleum-based plastics have many advantages and are widely used in almost all areas of people's lives, but about 79% of plastic products are not effectively recycled (Li *et al*., 2021). The increasing use of conventional plastics poses a major threat to the survival of humans and other living organisms due to the environmental pollution they cause (Podkościelna *et al*., 2022). Based on the findings of the Indonesian Environmental Forum (WALHI) Investigation Team in the West Nusa Tenggara (NTB) region and the Nusantara River Expedition (ESN), rivers in Mataram City are filled with plastic waste, resulting in microplastic pollution. The NTB and ESN Investigation Team conducted an inventory of plastic waste piles in channels and rivers in Mataram City and found that the rivers had become garbage dumps for sachet waste, cracker bags, Styrofoam, baby diapers, and clothing waste (https://walhintb.org/). In response to this case, the Mataram City Government has issued a policy to reduce the use of single-use plastics as a strategic step in overcoming environmental problems due to plastic waste through Mayor's Regulation No. 2 of 2023 on restricting the use of single-use plastics. This policy aims to reduce the consumption of single-use plastics, such as shopping bags, straws, and plastic packaging, which are one of the main contributors to environmental pollution. In response to this plastic crisis, there is an urgent need to find more sustainable alternatives. Bioplastics are emerging as a promising solution. The development of bioplastics not only offers a solution to the problem of plastic waste but also provides economic benefits. The use of local resources and the development of new industries through bioplastics can create jobs and support sustainable economic growth.

A lot of research has been conducted to develop environmentally friendly, biodegradable plastics to replace petrochemical-based plastics (Chowdhury *et al*., 2023; Li *et al*., 2022). In recent years, there has been an increase in research on the development of bioplastics and biocomposites (Farias da Silva and Soares, 2021). Bioplastics

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are a type of plastic produced from renewable natural resources such as starch, cellulose, protein, and vegetable oils, and are designed to be more environmentally friendly and to be decomposed naturally by microorganisms (Liu *et al*., 2023). These materials are abundantly available in a variety of agricultural resources. Their composition can vary among agricultural resources that produce bioplastics with different properties. Cellulose and starch are important biomacromolecules in bioplastic research and production due to their abundance in nature (Chandrasekar *et al*., 2023; Czaikoski *et al*., 2020). Starch is a natural polymer found in plants. Starch consists of two types of molecules, namely linear amylose and branched amylopectin. The ratio of amylose to starch amylopectin significantly affects the structure and thermal stability of the resulting bioplastics (Ghasemlou *et al*., 2022). Some research on starch-based bioplastics includes corn starch (Yang et al., 2022), cassava/tapioca (González *et al*., 2023; Meite *et al*., 2022), potato husk starch (Imoisili and Jen, 2023; Tien *et al*., 2023), and banana peel starch (Shafqat *et al*., 2021; Verma *et al*., 2024). The starch used in this study is tapioca starch. Tapioca starch was chosen as the main ingredient because it is readily available, affordable, non-toxic, and forms a good film. However, pure starch-based bioplastics tend to be fragile and less water-resistant, so they need to be combined with other supporting materials.

Research on tapioca starch-based bioplastics has been carried out extensively, including tapioca starch bioplastics which have a tensile strength value between 6 – 9 MPa (Waryat and Yuliasih, 2018); tapioca starch bioplastics with variations *plasticizer* has a tensile strength value between 0.33 – 1.1 MPa (González *et al*., 2023); tapioca starch with the addition of sweet orange peel pectin, sorbitol and chitosan has a tensile strength of 1,675 – 3,764 MPa (Gustiyani and Muryeti, 2023); Cassava Starch Bioplastic with the addition of *whey* cheese and glycerol have tensile strength of 0.00023 – 0.00646 MPa (Masahid *et al*., 2023). The bioplastics produced from the modified tapioca flour still do not meet the SNI 7818:2014 criteria for the quality requirements of biodegradable plastic bags. Therefore, in this study, tapioca starch bioplastics were supplemented with chitosan, carrageenan, and bagasse cellulose.

Chitosan is a naturally occurring amino polysaccharide obtained through chitin deacetylation and is the second most commonly used natural polymer. Its non-toxic, biocompatible, and biodegradable properties have prompted extensive research into several applications (Harugade *et al*., 2023). Chitosan is often used as an additive in starch mixtures to manufacture biodegradable plastics. The goal is to improve the physical and mechanical properties of the resulting bioplastics. These bioplastics can form hydrogen bonds between amylose and amylopectin chains in starch (Asjun *et al*., 2023; Hidayat *et al*., 2020; Kusumastuti *et al*., 2020; Rasak *et al*., 2024).

In addition to chitosan, another ingredient used to make bioplastics is carrageenan. Carrageenan gel can form a thin layer, making it suitable as a bioplastic material (Farhan and Hani, 2017). Carrageenan exhibits strong gelling properties, excellent water solubility and absorption, and biodegradability, which together lead to its widespread use in food and packaging applications (Pires *et al*., 2023). However, its limitations include a poor water barrier and low mechanical strength, which reduce its protective function in food packaging. Therefore, carrageenan must be combined with other materials. This study added cellulose extracted from bagasse to improve the water resistance of tapioca chitosan starch bioplastics.

Bagasse (*Saccharum officinarum*) is a by-product of the sugar-making process and of making sugarcane ice, which is solid and rich in fiber. The use of sugarcane bagasse waste in Indonesia remains suboptimal (Fadilla *et al*., 2023). Sugarcane bagasse waste contains 45 – 50% cellulose, 25 – 30% hemicellulose, 25% lignin, and 2.4 – 9% ash (Karp *et al*., 2013). Cellulose can be isolated from bagasse using the delignification method, followed by conversion to microcellulose via hydrolysis, which involves a common reaction as shown in Reaction 1.

$$(C_6H_{10}O_5)_n + H_2O \xrightarrow[HCl,75°C]{\text{Hidrolisis}} (C_6H_{10}O_5)_m + \text{oligosakarida} \qquad \text{(Reaction 1)}$$

The combination of cellulose and fiber in bioplastic production improves the physicochemical properties of bioplastics (Azmin et al., 2020). Therefore, the researcher uses raw materials to produce bioplastics from tapioca starch, with the addition of chitosan, carrageenan, and bagasse microcellulose to improve their quality. The quality parameters of the bioplastics tested in this study are density, moisture content, opacity, and bioplastic surface morphology.

## RESEARCH METHODS

The tools used in this study are a glass tool, a plastic petri dish, a spatula, a digital scale (Kern), a hot plate magnetic stirrer (Thermo Scientific), a UV-Vis Spectrophotometer (Shimadzu 1900i), a Fourier Transform



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Infrared (FTIR) Spectrophotometer (Perkin Elmer Spectrum Two with ZnSe ATR), and a Scanning Electron Microscopy (SEM) (JEOL JCM-7000). The materials used in this study include tapioca flour (Rose Brand), bagasse (sugarcane ice waste in West Lombok), chitosan (CV. Chimultiguna), kappa carrageenan refined (Nurashop), aquades, glycerol (Cliger Shop), NaOH (Merck), $H_2SO_4$ (Merck), $CH_3COOH$ (Merck), and NaOCl (Sigma Aldrich).

**Cellulose Isolation of Bagasse and Formation of Cellulose Microcrystalline**

Cellulose bagasse was isolated in several stages (Katakojwala and Mohan, 2019; Nasution *et al*., 2024). The bagasse was separated from the skin, cut into small pieces, washed, and dried in an oven at 55 °C for 24 hours. The dried bagasse was blended and sifted through a 100-mesh sieve. This powder was stored in a tightly closed container before the cellulose extraction process was carried out. The isolation process was carried out by weighing 25 g of bagasse powder, adding 500 mL of 2 N NaOH, stirring, and heating to 70 °C for 2 hours using a hotplate stirrer. The mixture was filtered, the residue was neutralized with distilled water, and the residue was then reacted with 1 N $H_2SO_4$ at 70 °C for 2 hours using a hotplate stirrer. This mixture was filtered, and the residue was neutralized using distilled water. After neutralization, the residue was bleached using 500 mL of 1.75% NaOCl solution at 60 °C for 1 hour using a magnetic stirrer. The fibers were filtered and washed with distilled water. After that, the fibers were ovened at 60 °C for 1 hour. The obtained alpha-cellulose was placed in a beaker and hydrolyzed with 2.5 N HCl at 70 °C for 15 minutes. The hydrolyzed mixture is then added to the beaker, stirred using a spatula, and left for 24 hours. The obtained microcrystalline cellulose was rinsed with distilled water and oven-dried at 60 °C for 1 hour to reduce moisture content.

**Bioplastic Synthesis**

Bioplastics were produced using methods reported by Mustariani *et al*. (2025) and Tien *et al*. (2023) with modifications. The ingredients with fixed amounts are tapioca starch 4 g, chitosan 0.49 g, carrageenan 2.4 g, glycerol 1.5 mL, and distilled water 250 mL. Sugarcane bagasse microcellulose was prepared in 5 formulations, namely F0, F1, F2, F3, F4, and F5, with amounts of 0, 0.2, 0.4, 0.6, and 0.8 g, respectively. All ingredients were dissolved separately in distilled water and heated at 70 °C while constantly stirring until fully dissolved. After that, all the solutions were mixed and heated for 20 minutes at 70 °C, with glycerol added as a plasticizer. The solution was molded in a Petri dish and left at room temperature for up to 24 hours, then dried in an oven at 55 °C for 24 hours. The bioplastic that has formed is removed from the mold and stored in a sealed plastic container lined with filter paper.

**Test Parameters**

The test parameters in this study include density, solubility, moisture content, and bioplastic opacity. The density test was carried out using the ASTM D792-91 standard on bioplastics cut to 2 × 2 cm (Maulida *et al*., 2016). The moisture content of bioplastics was calculated from the reduction in bioplastic mass after drying at 105°C to a constant weight (Chandrasekar *et al*., 2023). Bioplastic opacity was measured using a UV-Vis spectrophotometer at 600 nm, and absorbance data were converted to opacity values according to Equation 1.

$$\text{Opacity} = \frac{A_{600}}{t} \quad (1)$$

$A_{600}$ is the absorbance at 600 nm, t is the thickness of bioplastics in mm (Ranote *et al*., 2024; Socaciu *et al*., 2020). Furthermore, the bioplastic surface morphology was examined using SEM (Hossain *et al*., 2023).

## RESULTS AND DISCUSSION

**Cellulose Insulation of Bagasse**

Bagasse cellulose was successfully isolated from bagasse waste, as seen in Figure 1. The isolation process went through four stages, namely (1) delignification to dissolve lignin and part of hemicellulose, (2) mild acid hydrolysis to dissolve hemicellulose, (3) bleaching to remove lignin/pigment by oxidizing it, and (4) conversion to microcellulose using the HCl hydrolysis method. This hydrolysis process aims to break the cellulose chains in the amorphous region, leaving behind crystalline cellulose fragments. The $H^+$ ions of HCl will break the glycosidic (β-1,4-glycosidic) bonds in the amorphous (irregular) region of the cellulose structure, while the crystalline part remains relatively intact so that a shorter, harder, and more stable cellulose particle is formed, namely microcrystalline cellulose. This reaction corresponds to Reaction 1.



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**Figure 1**. Sugarcane bagasse microcellulose produced.

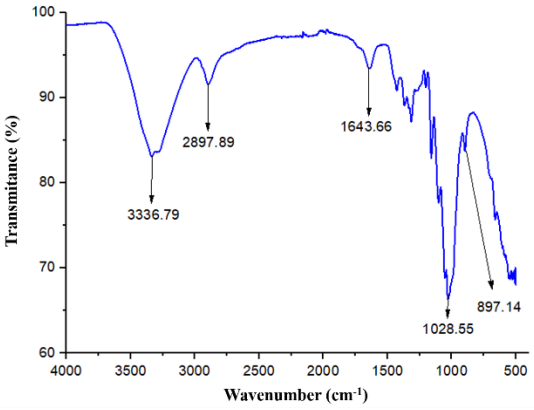



**Figure 2**. FTIR spectra of microcellulose FTIR is isolated from bagasse.

The microcellulose produced has a yield value of 15.95%. Figure 2 shows the FTIR spectra of microcellulose isolated from bagasse. Absorption peak 3336.79 $cm^{-1}$ indicates the presence of a stretch of the –OH (hydroxyl) group. A wide peak indicates the presence of intra- and intermolecular hydrogen bonds in the polysaccharide chain. Absorption peak 2897.89 $cm^{-1}$ indicates the presence of C–H strain vibrations of the methyl and methylene (–CH2–) groups in the cellulose glucose ring. Absorption peak 1643.66 $cm^{-1}$ indicates the presence of C–C bending vibrations in the structure of the glucose ring. Absorption peak 1028.55 $cm^{-1}$ shows the presence of a typical sign of the presence of primary alcohol in the $CH_2OH$ group in cellulose. Absorption peak 897.14 $cm^{-1}$ shows the presence of a C–O–C (β-1,4-glycosidic) bond, which is the typical peak of the main marker of natural cellulose (Harini and Mohan, 2020). No significant peaks were found at a wavelength of about 1730 cm-1, indicating that lignin and hemicellulose have been largely eliminated, thereby confirming that the delignification and bleaching process is successful.

**Characteristics of Bioplastics from Tapioca Starch, Chitosan, and Carrageenan with the Addition of Sugarcane Bagasse Microcellulose**

Bioplastics have been successfully made from tapioca starch, carrageenan, chitosan, and sugarcane bagasse microcellulose. The addition of sugarcane bagasse microcellulose yields bioplastics with varying physical appearances. The resulting bioplastics resemble conventional plastics. The bioplastics made from tapioca starch, chitosan, and carrageenan, with the addition of bagasse microcellulose, undergo tests of density, moisture content, opacity, and surface morphology. The average results for the density, moisture content, and opacity of bioplastics, with three replicates, are shown in Table 1.

**Table 1.** Average data on the characteristics of bioplastics from tapioca starch, chitosan, and carrageenan, with the addition of sugarcane bagasse microcellulose.

| Characteristics | Formulation | | | | | |
|---|---|---|---|---|---|---|
| | F0 | F1 | F2 | F3 | F4 | F5 |
| Density (g/mL) | 1.388 ± 0.06 | 1.410 ± 0.096 | 1.415 ± 0.031 | 1.459 ± 0.095 | 1.461 ± 0.008 | 1.453 ± 0.033 |
| Moisture content (%) | 16.419 ± 1.031 | 16.363 ± 0.436 | 16.028 ± 0.349 | 14.652 ± 0.342 | 14.482 ± 0.882 | 14.197 ± 0.265 |
| Opacity (/mm) | 2.294 ± 0.146 | 2.330 ± 0.076 | 2.346 ± 0.208 | 2.373 ± 0.045 | 2.379 ± 0.009 | 2.383 ± 0.044 |

**Bioplastic Density**

Bioplastic density measurements are important for determining density levels and homogeneity, and for understanding their relationship to material strength and function. A graph of the density of tapioca chitosan carrageenan starch bioplastics with the addition of bagasse microcellulose is shown in Figure 3.



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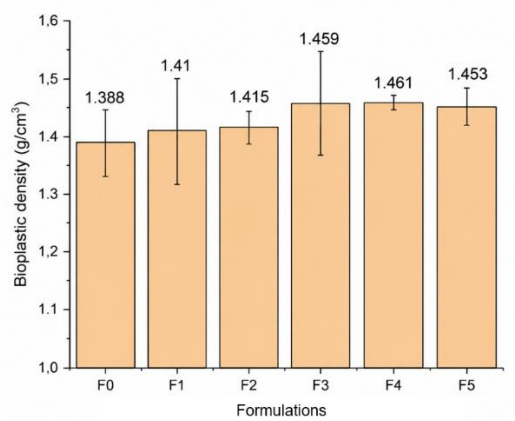



**Figure 3.** Graph of the bioplastic density of tapioca chitosan starch carrageenan with variations in the addition of sugarcane bagasse microcellulose.

Figure 3 shows that the bioplastic density increases with increasing concentration of sugarcane bagasse microcellulose. In the control formulation (F0) without the addition of sugarcane bagasse microcellulose, a bioplastic density of 1.388 ± 0.06 g/cm³ is shown. When tapioca chitosan starch bioplastics are supplemented with bagasse microcellulose, the bioplastic density increases to 1.410 – 1.461 g/cm$^3$. This increase in density is partly due to the microcellulose nature of bagasse, which has a crystalline structure and high intrinsic density, allowing it to fill cavities or pores between polymer chains, reduce empty space, and increase the total density of bioplastics. The hydrogen bond between the hydroxyl group of microcellulose and the active group in chitosan, carrageenan, and starch also strengthens interactions between polymer chains, resulting in a more compact and denser film. In the F5 formulation, there is a decrease in density, partly due to the amount of microcellulose added exceeding the optimal composition, which causes particle agglomeration, increasing the porosity of bioplastics and reducing the density of bioplastic structures.

The bioplastic density values obtained in this study are close to those reported for Ethiopian sweet potato starch and Ethiopian banana fiber by Majamo and Amibo (2024), which range from 1.64 to 1.73 g/mL. However, it is still higher than the bioplastics from potato flour and dragon fruit peel pectin reported by Tien *et al*. (2023), which range from 1.02 – 1.63 g/mL. (Ranote *et al*., 2024) also reported that the addition of cellulose fillers treated with acid and graphene oxide increased the bioplastic density of tapioca starch from 1.31 to 1.35 – 1.44 g/mL.

**Moisture content of Bioplastics**

Measuring the moisture content of bioplastics is important for determining their resistance. High moisture content can cause bioplastics to expand easily, become brittle, or degrade during storage, affecting their durability and performance in use. Data on the moisture content of tapioca-chitosan-carrageenan-cellulose starch bioplastics can be seen in Figure 4.

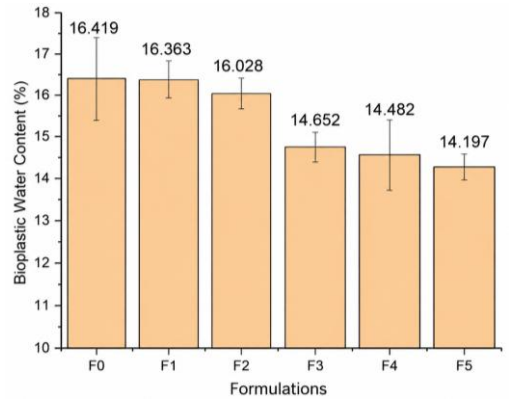



**Figure 4.** Graph of the moisture content of tapioca chitosan starch bioplastics with variations in the addition of sugarcane bagasse microcellulose.

Figure 4 shows that the moisture content of bioplastics decreases with increasing bagasse microcellulose concentration. The control formulation (F0) without the addition of sugarcane bagasse microcellulose shows a


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bioplastic moisture content of 16.419%. When tapioca chitosan starch bioplastics were supplemented with bagasse microcellulose, the bioplastics' moisture content decreased, ranging from 14.197 – 16.363%. This decrease in moisture content is partly due to the added bagasse microcellulose, which can fill voids and interact strongly with the starch, chitosan, and carrageenan matrix present in bioplastics through hydrogen bonding (Singha *et al*., 2023). Filling the blanks in these bioplastics can increase their density, as evidenced by the bioplastic density data in Figure 3, which has also increased. This is in line with the research by Singha *et al*. (2023), which showed that adding bagasse fibers reduced the moisture content of PVA-based biodegradable films and sweet orange peel from 36.71% to 32.32%.

The water content of the bioplastics in this study, in accordance with the quality requirements of bioplastics under the Indonesian National Standard (SNI) 06-3735-1995, namely good bioplastics with a water content of no more than 16%, is found in formulations F3, F4, and F5. The moisture content values for bioplastics F0, F1, and F2 are still above 16%, so they do not meet the bioplastic quality requirements under SNI. The moisture content of bioplastics that meet the SNI quality requirements in this study ranges from 14.197 – 14.652%. Low water content helps the film resist mold growth, which can damage its appearance and mechanical properties. The metabolic activity of microorganisms is accelerated by high water content (Singha *et al*., 2023).

**Bioplastic Opacity**

Opacity is the ability of a material to block or absorb light. The higher the opacity of bioplastics, the more opaque the material and the less light passes through the bioplastic. Bioplastic opacity data for tapioca starch–chitosan–carrageenan–cellulose bagasse are shown in Figure 5.

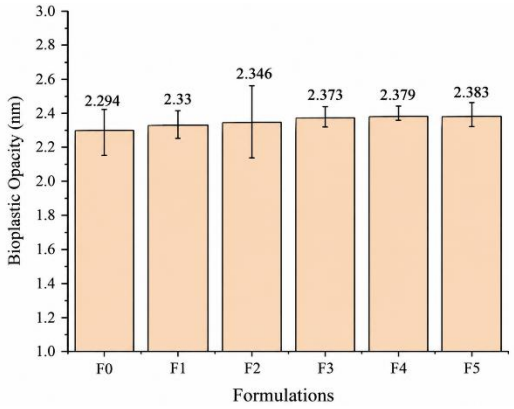



**Figure 5**. Graph of bioplastic opacity of tapioca chitosan starch carrageenan with variations in the addition of sugarcane bagasse microcellulose.

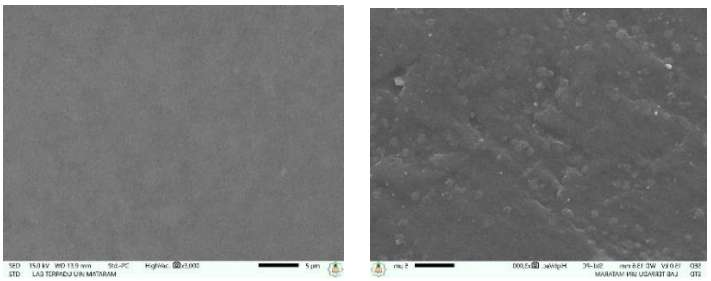
Figure 5 shows that the opacity of bioplastics increases with increasing concentration of bagasse microcellulose. The control formulation (F0), without the addition of bagasse microcellulose, shows a bioplastic opacity of 2.294 /mm. When tapioca chitosan starch bioplastic carrageenan was added with bagasse microcellulose, the opacity of the bioplastics ranged from 2.330 to 2.383 /mm. This increase in opacity is likely due to the added bagasse microcellulose, a crystalline material that can increase the crystallinity of bioplastics. The crystalline part of the polymer has a more regular, denser structure, resulting in greater light scattering than the amorphous region. This makes the film slightly more opaque. In addition, the addition of filler can also increase the inhomogeneity and roughness of the structure, resulting in reduced light transmission. Although the increase is not large, this trend suggests that as microcellulose content increases, the film's transparency decreases, so it may need to be adapted for packaging applications that require a certain level of clarity.

**Bioplastic Surface Morphology**

This bioplastic morphological characterization was carried out to determine the homogeneity level and distribution of bagasse microcellulose in bioplastics. Morphological characterization was performed on the control formulation (F0), without bagasse microcellulose, and on F4, the optimal formulation containing 0.8 g of bagasse microcellulose.



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(a) (b)

**Figure 6**. The results of the surface morphology test were (a) control bioplastics and (b) bioplastics with the addition of F4 bagasse microcellulose (right) at 3000× magnification.

Figure 6 shows that the addition of microcellulose from bagasse in F4 results in a rougher, more heterogeneous surface, indicating that the bagasse microcellulose is not fully dispersed. This morphology is common in starch-based bioplastics that are given cellulose fillers; the more microcellulose, the more protrusions and agglomeration areas, which can change mechanical and physical properties, such as increasing tensile strength, decreasing solubility, and increasing opacity. This is consistent with the opacity data obtained, which shows an increase in opacity as the number of fillers added increases.

## CONCLUSION

Microcellulose was successfully extracted from bagasse with a yield value of 15.95%. The resulting microcellulose was used as a filler in bioplastics based on tapioca chitosan and carrageenan starch. The results of the density, moisture content, and opacity tests indicated that F4 was the best formulation when 0.8 g of bagasse microcellulose was added. The optimum bioplastic produced had a density of 1.461 g/mL, a moisture content of 14.482%, and an opacity of 2.379/mm. The addition of sugarcane bagasse microcellulose reduced the moisture content of bioplastics and increased their density.

## CONFLICT OF INTEREST

There are no conflicts of interest in this article.

## AUTHOR CONTRIBUTION

YY: Methodology, Conceptualization, Manuscript Draft Writing; SS: Manuscript Review and Editing; MK: Data Analysis, Software; BMS: Data Retrieval; NA: Data Retrieval.

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## DECLARATION OF GENERATIVE AI

No declaration of generative AI use was provided by the authors.

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