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Research Paper

Physical Properties of Biodegradable Chitosan-Cassava Starch Based Bioplastic Film Mechanics

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Abstract
Petroleum-derived plastics are widely used but pollute the environment significantly. The development of biodegradable plastics is urgently needed to be replaced. The mechanism for making bioplastic films from cassava starch-chitosan/glycerol uses a double-screw extruder process. The film took into account the multi-hydroxyl capacity of starch by combining glycerol (in a ratio of 3:1 w/w) and chitosan (at concentrations of 0.5, 1.0, and 1.5% (w/w). The impact of chitosan involvement on the characteristics of the bioplastic material was studied, including physical, thermal, mechanical, and biodegradability properties. The findings showed that using chitosan as a filler in cassava starch bioplastics resulted in bioplastic films with high compressive capacity and water resistance. The resulting biopolymer's contact angle was increased by including C−O functional groups in the molecule, as evidenced at a wavelength of 1028 cm^{-1} of the FTIR spectra. The contact angle was increased from θ = 65.3059 ± 2.7936° to θ = 68.6047 ± 3.2391°. An increase in tensile strength was also observed, indicating increased stiffness compared to chitosan-free bioplastics. The best bioplastic blend was the formulation of cassava starch and glycerol containing 0.5% chitosan. Bioplastic has physical properties of density 0.8625 ± 0.0277 g/mL; contact angle 68.6046 ± 3.2391°; water uptake 11.0660 ± 0.3709%; tensile strength 2.0181 ± 0.0594 MPa; elongation 54.2243 ± 3.2623%; thermal 137.5°C; moisture content 4.9464 ± 0.1172%; and the fastest biodegradation rate. The bioplastic synthesized in this study is readily biodegradable in the natural environment, making it highly sustainable and more environmentally friendly, and it can be a viable substitute to reduce the use of petroleum-based bioplastic.

Keywords
Bioplastic Film, Chitosan, Twin-Screw Extruder, Physical Properties, Spectroscopy, Thermal Properties

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1. INTRODUCTION

The bioplastic synthesized in this research effortlessly undergoes decomposition in the natural surroundings, rendering it highly sustainable, more environmentally friendly, and offering a viable substitute for reducing the use of disposable plastics. To produce biodegradable plastic, renewable biopolymer materials such as starch, cellulose, collagen, casein, animal protein, or lipids are directly extracted from biomass, either with or without modification (Abdullah et al., 2020a). Biodegradable plastics or bioplastics can be used like conventional plastics but will be more easily degraded by microorganism activity after being discharged into the environment (Muthusamy and Pramasivam, 2019).

Starch is one of the natural polymers with properties close to synthetic polymers, affordable price, sufficient availability, and good novelty. However, bioplastics from starch have two weaknesses, there are low mechanical strength (tensile strength) and poor water resistance (more hydrophilic) (Baklagina et al., 2018). Starch can be mixed with other hydrophobic biopolymers, such as chitosan, to reduce its hydrophilic properties (Agusman et al., 2022). Chitosan, as an essential natural biopolymer with broad applications due to its biocompatibility and biodegradability, is a linear polysaccharide of a shellfish shell (crustacean) consisting of randomly distributed β-(1→4)-linked D-glucosamine (deacetylation unit) and N-acetyl-D-glucosamine (acetylation unit) (Mawazi et al., 2024). Chitosan is derived from the deacetylation of chitin and serves

structural element in the exoskeletons of crustaceans, such as prawns and crab shells of prawns and crab (Agusman et al., 2022; Utami et al., 2021). Chitosan is used as a bioplastic raw material because of its non-toxic, biofunctional, biodegradable, and antimicrobial properties (Utami et al., 2021). The plasticizers reportedly used in starch-based bioplastic formulations are glycerol and sorbitol. Glycerol may function as a plasticizer to diminish intermolecular tensions and enhance the mobility of polymer chains by minimizing the likelihood of discontinuities and brittle regions (Cacique et al., 2017; Mukuze et al., 2019; Van Chien et al., 2024) and chitosan is applied to reduce starch hydrophilic properties (Agusman et al., 2022).

Chitosan is hydrophobic nature makes it suitable as a raw ingredient for bioplastics. Chitosan is anticipated to decrease the percentage of starch in bioplastic compositions (Cacique et al., 2017; Van Chien et al., 2024) and add value to crab and shrimp canning industry waste. Recent publications on starch film formulation reported that the addition of chitosan improved various characteristics of starch-based films (Albar et al., 2024; Cacique et al., 2017; Deng et al., 2022; Hao et al., 2023; Luchese et al., 2018; Othman et al., 2024; Utami et al., 2021; Van Chien et al., 2024). Chitosan's antibacterial qualities make it a profitable material for producing bioplastics, making it a viable choice for packaging food products (Barik et al., 2024; Jin et al., 2024; Zhou et al., 2024). Investigating the bioplastic characteristics of Cassava starch in different chitosan concentrations is a fascinating subject. It will offer insights into the benefits and drawbacks of potential future applications of food shelf life. In this study, the performance of bioplastics made from Cassava starch was improved by adding chitosan from crab shells. The reinforcing effects of chitosan on the mechanical and physical as well as antimicrobial properties of starch bioplastics were studied.

2. EXPERIMENTAL SECTION

2.1 Materials

Materials Cassava starch was a commercial product supplied by PT. Surya Pati Kencana (Pati, Central Java, Indonesia). Chitosan from crab shells was provided by the Research Center for Marine and Fisheries Product Processing and Biotechnology, the Ministry of Marine Affairs and Fisheries, Republic of Indonesia (Jakarta, Indonesia). Glycerol was a commercial product from PT. Wilmar Nabati Indonesia (Jakarta, Indonesia). Salt agar (SA) was purchased from Merck (Darmstadt, Germany).

2.2 Bioplastic Preparation

Bioplastic preparation refers to the publication of Abdullah et al. (2019) with some modifications. Bioplastics are produced through the combination of Cassava starch and glycerol along with the inclusion of chitosan derived from crab shells, all of which are mixed with Cassava starch. The glycerol ratio of 3:1 (w/w) achieved by using a blender (Phillips HR2118/01) for approximately 3 minutes. Chitosan, with a moisture content of 1.12%, ash content of 77.73%, volatile content of 1.83%, and carbon content of 19.33%, was included in the mixture at concentrations of 0.5, 1.0, and 1.5% (w/w). Bioplastics were prepared using a twin screw plastic production Masterbatch Extruder (Nanjing Tenda Machinery Co. Ltd) at a temperature of 110–145°C. The twin screw extruder was run at a speed of 50 rpm, and the resulting pellets were cut into sizes of 3-5 mm through a pelletizer. Bioplastic sheets were molded by compressing the mixture (± 15 g) using a load of 40 kgf/cm^2 at a temperature of 145°C for 5 minutes.

2.3 Bioplastic Characterization

The bioplastics were characterized by conducting measurements of various physical properties, i.e. density (ASTM D792-08, 2008), moisture content, and surface hydrophobicity (ASTM D5946-04). The mechanical properties, such as tensile strength and elongation, were determined using ASTM D882-18 (2018). Functional group analysis was performed using spectrophotometry, while microstructure properties were examined using microscopy (ASTM E 2015, 1991). Thermal properties were assessed using differential scanning calorimetry (DSC), and biodegradability analysis was conducted according to ASTM G21.

2.3.1 Physical Properties

The density of bioplastic was determined following the guidelines of ASTM D792-08 (2008). The percentage of water of the bioplastic was determined by measuring the weight of the 3×3 cm films and exposing them to heat at 105°C for 3 hours until a stable weight was achieved. The moisture content was determined by calculating the percentage of the difference between the weight of the sample after it was wetted and the initial weight of the dry sample, using the ASTM D570 standard. Contact angle measurements (ASTM D5946-04) were employed to evaluate the hydrophobicity of the film surface. The interaction between the bioplastic samples and water in a dynamic environment by means of water uptake was measured by spraying water on the surface of the film sample (1×1 cm^2) within a chamber using an automatic micro syringe (5 μL) as a modification analysis (Cravigan et al., 2020).

2.3.2 Mechanical Properties

The mechanical properties of bioplastics were evaluated by measuring their tensile strength and elongation using a universal testing instrument (UCT-5T, Orientec Co. Ltd, Japan). The tests were conducted according to the ASTM D882-18, 2018 standard, with an initial gauge separation of 100 mm and a crosshead speed of 50 mm/min. The measurements were performed on three samples of bioplastics, and the results were reported as average ± standard deviation (n=3) the samples were stored at 25°C and 55% relative humidity for 24 hours prior to testing.

2.3.3 Functional Group Analysis

The chemical structure was examined using FTIR analysis following the manual process of the Thermo Scientific Nicolet

iS5 ATR-FTIR spectrometer, Madison, WI, USA. A Fourier Transform Infrared (FTIR) spectrometer was utilised with a scanning resolution of 4 cm^{-1} inside the wavenumber range of 4000 cm^{-1} to 400 cm^{-1}. The spectra integration was performed by conducting 16 scans.

2.3.4 Microstructure Properties

The Jeol Scanning Electron Microscope analysis procedure (Jeol JSMIT300 Scanning Electron Microscope, Tokyo, Japan) and (ASTM E 2015, 1991) were used to examine the cross-sectional morphology of the bioplastics. Before the measurement, using a sputtering technique, the sample's surface was covered with a thin layer of gold (~10 nm). The analysis was performed at an acceleration voltage of 20 kV. SEM image analysis was then performed using ImageJ software to extract data from binary images (black and white) in APS-ImageJ by performing a thresholding procedure followed by binarization by adjusting the brightness and contrast to obtain an image resolution that can be measured by the number of porosity pixels and by dividing it by the number of areas observed in the image and then multiplied by 100%. The particle size distribution value is expressed as the number of pores (count) per unit length of the membrane (AlMarzooqi et al., 2016; Saraf et al., 2019).

2.3.5 Thermal Properties

The thermal properties of the bioplastics were examined using differential scanning calorimetry (DSC) using the manual approach outlined in the (DSC 214 Polyma Netzsch, Germany). Measurements were performed within a temperature range of 25-250°C, using a heating rate of 10°C per minute, under a nitrogen atmosphere. Prior to DSC measurement, bioplastic samples were subjected to oven-drying at a temperature of 105°C for a duration of 30 minutes.

2.3.6 Biodegradability Analysis

The degradation of bioplastic was assessed using *Aspergillus niger* in on a salt agar (SA) medium, following the guidelines of ASTM G21. The salt agar was dissolved in 100 mL of distilled water and then placed into a petri plate. *Aspergillus niger* was applied and distributed evenly using a single-use sterile spreader on all sides of the sample (1.5×1.5 cm), which was positioned on the SA media and thereafter kept in an incubator at 37°C and 90% relative humidity for a duration of ten days. The microbial degradability of the bioplastic samples was evaluated by monitoring the proliferation of *A. niger* over a period of ten days. ImeJ was employed to quantify the area occupied by *A. niger* on the surface.

2.4 Statistical Analysis

A completely randomized design was employed for the experimental with the variable of 0.0, 0.5, 1.0, and 1.5% (w/w) chitosan in a 3:1 (w/w) Cassava starch-glycerol mixture; three replicates were performed. Data are presented as the mean ± standard deviation. The data, with the exception of biodegradability data, were subjected to one-way analysis of variance (ANOVA) followed by Tukey's post hoc test to assess the significance of differences between the mean values. Statistical significance was defined as a condition where the probability (P) is less than 0.05. The Statistical analysis was conducted using the IBM SPSS Statistics V21 (IBM, USA). Qualitative analysis was conducted on the biodegradability data.

3. RESULT AND DISCUSSION

3.1 Bioplastic Sheet from Chitosan-Cassava Starch

Bioplastic sheets made from chitosan-Cassava starch with different chitosan content are shown in Figure 1 different concentrations of chitosan lead to different bioplastic appearance. The pure-modified starch bioplastic exhibited higher transparency compared to the inclusion of chitosan. The inclusion of chitosan in the bioplastic renders it opaque, as demonstrated by Abdullah et al. (2020b). They observed that adding less than 50% chitosan led to a hazy appearance. Similarly, Agusman et al. (2022) verified that the incorporation of chitosan resulted in darker films. The appearance of bioplastics is also related to the mixing process carried out by the hot-press method at high temperatures, resulting in an inhomogeneous surface, which the SEM results can confirm.

3.2 Physical Properties

Table 1 shows the physical and mechanical properties of chitosan-cassava starch bioplastic, i.e., moisture content, density, and contact angle. Moisture content causes the material to swell, soften, and degrade. The composition, structure, and method of bioplastic processing affect the moisture content. For bioplastics to have a low moisture content and not readily absorb water, they must be modified using other polymers (Marichelvam et al., 2019). The addition of 1.5% chitosan significantly reduced the moisture content of the bioplastics.

The density of bioplastics in the study was determined by measuring the (mass per unit volume of the particle using a pycnometer. The graph illustrates a positive correlation between the chitosan concentration and the observed variable. The bioplastic density result was in the range of 0.86-0.91 g/mL, which indicated proportional to material mass. When the material mass is more significant, a higher density value is obtained (Kedir et al., 2024). Based on Maleki and Milani (2020), the density of some commercial chitosan was in the range of 0.20-0.38 g/mL. The increase in density value is related to the addition of water (Agusman et al., 2021). The interparticle spacing is influenced by both the distance and shape of the particles, as well as the interparticle forces. A particle distance that is greater than the particle size will result in low density and high compressibility. In contrast, the opposite will occur if the particle distance is smaller (closer) than the particle size, causing no space between particles (Brown et al., 2024).

The contact angle and water uptake of chitosan-Cassava starch bioplastics with different chitosan contents are shown in Table 1. The value increased with the addition of chitosan, indicating that the bioplastic had hydrophobic characteristics.

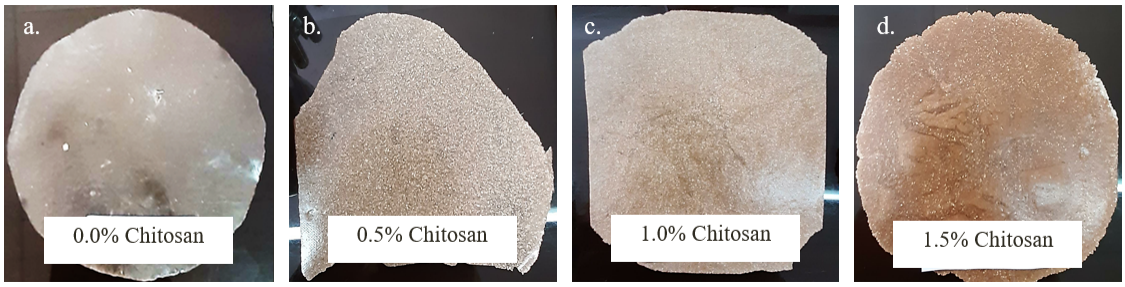

Figure 1. Digital Photo of 15 g Chitosan-Cassava Starch Bioplastic After Molded with 40 kgf/cm^2 Load at 145°C for 5 Minutes

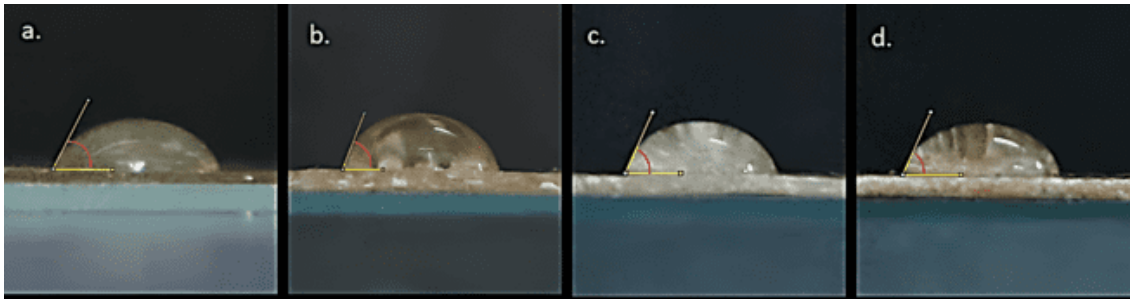

Figure 2. Contact Angle of Chitosan-Cassava Starch Bioplastic at Different Chitosan Concentrations: (a) 0.0%, (b) 0.5%, (c) 1.0%, and (d) 1.5%

Table 1. The Physical Parameters (Density, Contact Angle, and Water Uptake) and Mechanical Properties (Tensile Strength and Elongation) of Bioplastics Made from Chitosan-Cassava Starch with Varying Chitosan Concentration

Parameters	Chitosan (%)			
	0.0	0.5	1.0	1.5
Physical Properties				
Moisture Content (%)	8.0097 ± 0.3311	4.9464 ± 0.1172	4.9464 ± 0.1172	3.7486 ± 0.2920
Density (g/mL)	0.9048 ± 0.0487	0.8625 ± 0.0277	0.9114 ± 0.0184	0.9143 ± 0.0293
Contact angle (°)	65.3059 ± 2.7936	68.6046 ± 3.2391	66.6380 ± 3.3633	65.5558 ± 3.1399
Water uptake (%)	8.6620 ± 0.4436	11.0660 ± 0.3709	9.8642 ± 0.5249	9.5525 ± 0.1592
Mechanical Properties				
Tensile strength (MPa)	3.2607 ± 0.3457	2.0181 ± 0.0594	4.6111 ± 0.2023	8.8730 ± 0.0450
Elongation (%)	85.4867 ± 3.2406	54.2243 ± 3.2623	24.4600 ± 1.1069	6.6266 ± 3.7662

The contact angle of the chitosan-Cassava starch bioplastics indicates the hydrophobic nature of the bioplastics by quantifying their level of wetness. A substance is considered hydrophilic if its contact angle value is near to 0°. Alternatively, if the contact angle approaches 180°, the material can be classified as hydrophobic (Marichelvam et al., 2019).

The contact angle measurements, as shown in Figure 2, were further validated through a water uptake test, the results of which are presented in Table 1. As the concentration of chitosan increases, the water uptake value decreases, approaching 180°, indicating that the material becomes hydrophobic. Chitosan retards the swelling process because it is difficult to dissolve in water owing to its crystal structure composed of intramolecular and intermolecular hydrogen bonds (Vlacha et al., 2016). Moreover, bioplastic surface roughness and pore structure significantly influenced the contact angle results. The measured contact angle decreased with increasing porosity, pore diameter, and material permeability (Krainer and Hirn, 2021).

3.3 Mechanical Properties

Mechanical properties of chitosan-Cassava starch, i.e., tensile strength and elongation, are shown in Table 1. The tensile strength of chitosan-Cassava starch bioplastics increased with an increasing chitosan concentration except for 0.5% of chitosan. The bioplastic containing 1.5% chitosan achieved the maximum tensile strength of 8.8730 MPa. The research findings indicate that the incorporation of chitosan can facilitate

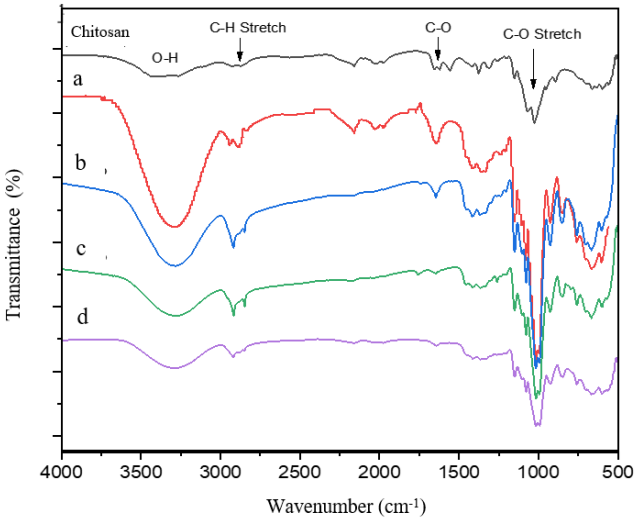

Figure 3. FTIR Spectra of Chitosan-Cassava Starch Bioplastic at Different Chitosan Concentrations: (a) 0.0%, (b) 0.5%, (c) 1.0%, and (d) 1.5%

the formation of intermolecular hydrogen bonds and amino groups in bioplastic molecules, resulting in an augmentation of the quantity of hydrogen bonds between bioplastic molecules (Ginting et al., 2016). Increases in hydrogen bonds can make bioplastics stronger and more breaking-resistant (Ginting et al., 2018). Furthermore, chitosan possesses a linear molecular structure, which predisposes it to form a crystalline phase due to its capacity to organize polymer molecules in an orderly manner (Baklagina et al., 2018). However, increasing the chitosan content reduced the elongation at the break value.

3.4 Functional Group Analysis

The spectra illustrate the prevailing functional groups observed in starch, O−H alcohol, C−H alkanes, and C−C bonds from Cassava starch. The presence of O−H groups overlapping with N−H is indicated by a distinct absorption peak at wave number 3361 cm^{-1}. Moreover, the existence of C−H was identified by an absorption peak at a wave number of 2877 cm^{-1}. In addition, the presence of C−O was confirmed by an absorption peak at a wave number of 1028 cm^{-1}. The FTIR spectra obtained following the formation of bioplastics exhibited a discernible alteration in the absorption peak pattern. Similarly, a reduction in the stretching of −OH was observed as the chitosan levels increased, it was evidenced by the diminished slope of the spectral peaks. The spectral absorption exhibited broadening near these specific wavelengths. This phenomenon can be explained by the number O−H functional groups resulting from the incorporation of Cassava starch. The finding of Syuhada et al. (2020) confirmed that a novel compound could be generated through physical mixing without the introduction of additional functional groups.

In Figure 3, the spectra of bioplastics composed of chitosan and Cassava starch with varying concentrations of chitosan are presented. The FTIR spectrum have spectra on the same functional groups, i.e. O−H, CH, and C−O., obtained from the formation of bioplastics is shown with apparent changes in the absorption peak pattern. A reduction in -OH stretching is observed with increasing chitosan content, which was evidenced by a decrease in the slope of the spectrum peak and a broadening is seen near this specific wavelength. This phenomenon can be associated with the augmentation of the O−H functional group produced by cassava starch. This group shows that the material has hydrophilic properties from the formation of hydrogen bonds so that it can increase its solubility in water, as mentioned by Mason et al. (2005). Another functional group that clearly changes its spectrum peak pattern is the C−O ester group. The spectral peak becomes more robust with increasing chitosan content. This group is also an indicator of hydrophilic properties, which indicates that the product will have biodegradable properties that can be easily decomposed. As mentioned by, the bond in the C−O group allows water molecules to bind. It can cause microorganisms to enter the biodegradable plastic membrane matrix, which causes environmental degradation. Overall, from the FTIR spectra profile, bioplastic with 1.5% chitosan content is the best because it has the weakest O−H bond and the strongest C−O, which shows lower hydrophilic properties but better degradable potential.

3.5 Microstructure Properties

The micrographs in Figure 4 show the cross-sectional morphology of chitosan-Cassava starch bioplastics at various chitosan concentrations: 0% (Figure 4a), 0.5% (Figure 4b), 1% (Figure 4c) and 1.5% (Figure 4d). The surface of Figure 4a, which represents a starch bioplastic without chitosan, displayed the highest level of homogenity compared to the other samples. The observed result may be attributed to the advantageous interaction between starch and chitosan. The higher chitosan content in starch bioplastics (Figure 4b, 4c, and 4d) formed an inhomogeneous morphology structure owing to the insoluble chitosan in the solvent, as shown by Hasan et al. (2020); Yang et al. (2023). The chitosan particles in cassava chitosan-starch bioplastics exhibited a well-defined size distribution, as confirmed by SEM microscopy. The particle sizes, ranging from 12.0 to 14.5 μm, were consistent across different chitosan concentrations, as shown in Figure 5 and Table 2.

Table 2. Particle Size Distribution Based on SEM Micrographs of Chitosan-Cassava Starch Bioplastic

Chitosan Concentrations	Particle Size Distribution
0.0%	14.06 ± 3.46
0.5%	14.5 ± 4.03
1.0%	12.06 ± 3.86
1.5%	9.20 ± 6.25

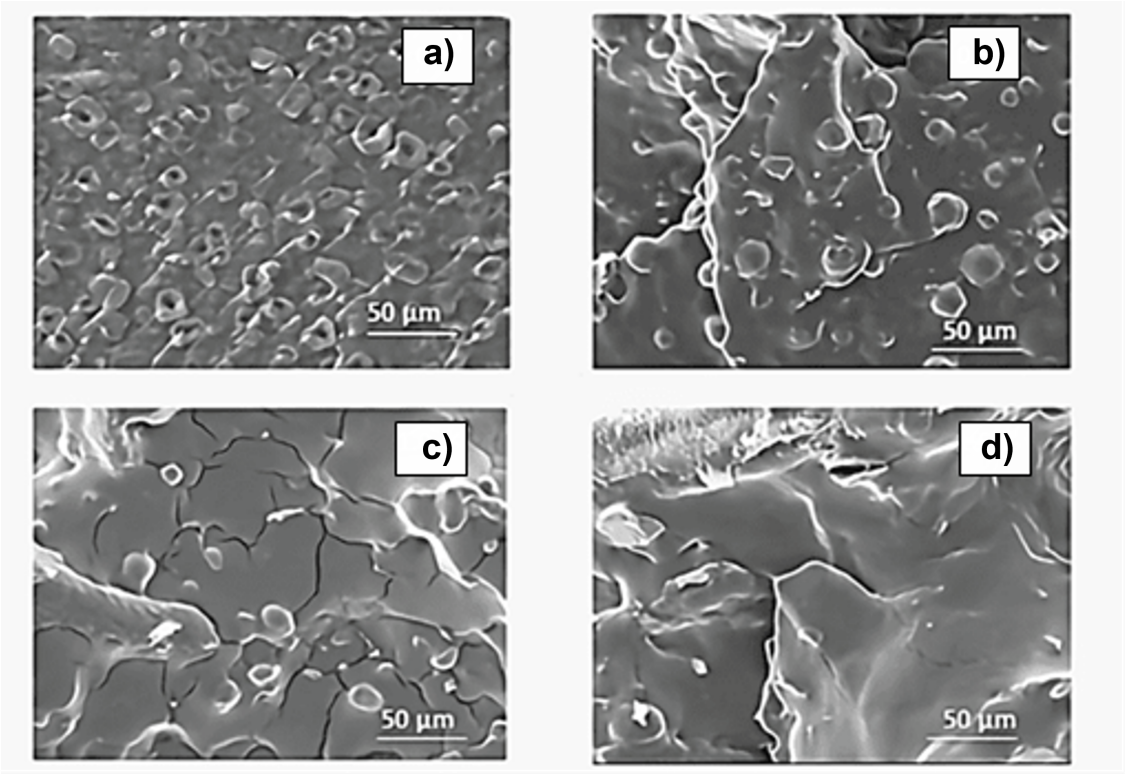

Figure 4. Scanning Electron Microscope (SEM) Micrographs of Chitosan-Cassava Starch Bioplastic at Different Chitosan Concentrations: (a) 0.0%, (b) 0.5%, (c) 1.0%, and (d) 1.5%

3.6 Thermal Properties

Examination of the DSC curves (Figure 6) showed that chitosan had a melting peak at 91.7°C, while starch-based bioplastics without chitosan did not show a clear peak. The melting point of cassava starch reported 172.87°C (Ibrahim et al., 2020). Chitosan incorporation elevated the melting point of the starch-based bioplastic from 137.5 to 150.6°C. It can be proven from bioplastic with 0.5% chitosan that had a melting temperature of 137.5°C and increased when the chitosan concentration was 1.0%. However, bioplastic with 1.5% chitosan presented two peaks at 138.5 and 148.6°C; the melting point of bioplastics increased with increasing chitosan levels. At 1.5%, the chitosan melting point could not be determined because two peaks were detected, attributed to impurities. However, bioplastics melting point is higher than pure chitosan's reported (Contessa et al., 2023).

A general drop in melting point is observed with increasing chitosan concentration. The increased melting point indicates that the crystalline structure of cassava starch is disrupted by chitosan, which prevents the starch from forming a well-ordered melt. The chitosan-starch blends have several melting peaks (Figure 6), particularly at 1.5% chitosan concentrations. The curve suggests a non-uniform melting tendency, which could be brought on by phase separation or the inch crystalline areas in the mixture. Compared to pure starch, the mixes' lower melting temperatures and broader melting peaks allude to a decrease in crystallinity. Chitosan reduces the crystallinity of starch granules by acting as a plasticizer and upsetting the hydrogen bonding network inside them. Concentration determines how chitosan affects melting behaviour. A lesser melting point drop and fewer distinct peaks are observed at lower concentrations (0.5%), where the effect is less noticeable. Broader melting peaks and a more noticeable melting point decrease result from the breakdown of starch crystallinity that occurs when chitosan content rises. The DSC results show that the thermal characteristics of cassava starch change when chitosan is added. The reduced crystallinity, multiple melting peaks, and observed melting point depression indicate that chitosan alters the structure of starch and affects how it melts.

3.7 Biodegradability Analysis

The results in Figure 7 show that the biodegradability of chitosan-cassava starch bioplastic tended to decrease as the chitosan content increased, indicating that the inclusion of chitosan enhances the degradation of the bioplastics by introducing a crystalline structure and antibacterial properties (Paradika, 2017). ASTM 5336 states that bioplastics (PLA from Japan and PCL from the UK) degrade for up to 60 days (100%) (Vlacha et al., 2016). After a 10-day observation period, the area covered by *A. niger* in the bioplastics without chitosan addition reached 96.45%. With the addition of 0.5, 1.0, and 1.5% chitosan, the areas covered by the fungus decreased to 90.39, 70.94, and

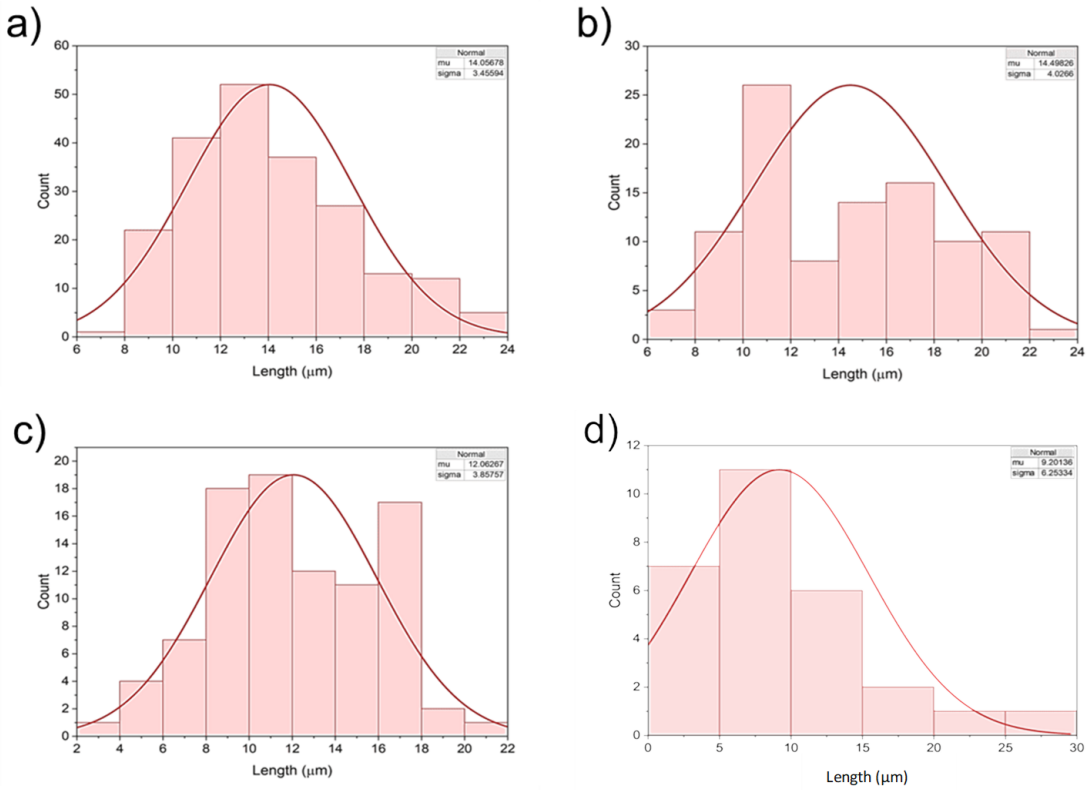

Figure 5. Particle Size Distribution Based on SEM Micrographs of Chitosan-Cassava Starch Bioplastic at Different Chitosan Concentrations: (a) 0.0%, (b) 0.5%, (c) 1.0%, and 1.5%

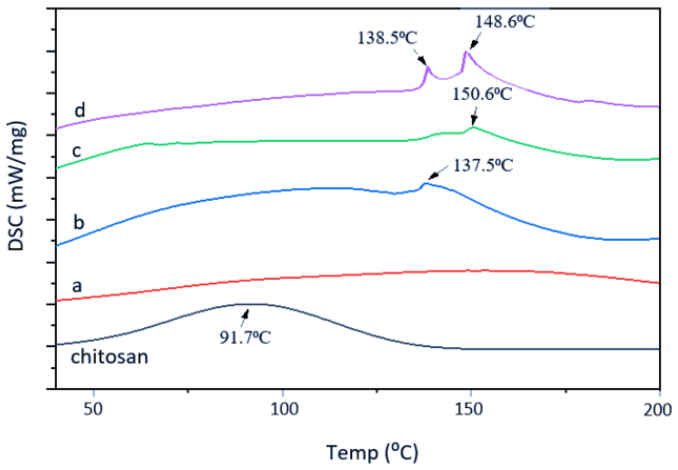

Figure 6. Differential Scanning Calorimetry (DSC) Curves of Chitosan-Cassava Starch Bioplastic at Different Chitosan Concentrations: (a) 0.0%, (b) 0.5%, (c) 1.0%, and (d) 1.5%

50.53%, respectively. As mentioned in (Nissa et al., 2019), *A. niger* growth on bioplastics without chitosan and with chitosan at a concentration of 0.5 and 1.0% is included in score 4 with a growth area of 60-100%. In contrast, bioplastics with 1.5% chitosan had a score of 3.

Overall, the density of starch-based bioplastics has increased in line with the enhancement of the chitosan. This density level is related associated to the wettability of the surface characteristics. A higher density of bioplastics means a closer distance of each particle and may cause difficulties in water leakage into the space between particles. The correlation between the density of bioplastics and water contact angle data is evident. The inclusion of 0.5% chitosan in starch-based bioplastic resulted in an increased in the water contact angle compared to the control. This can be attributed to the formation of hydrogen bonds between chitosan and starch, which may limit the availability of hydrophilic groups and weaken the interaction with water molecules. The addition of chitosan to a starch-based bioplastic, leads to the formation of hydrogen bonds between the NH_2 and OH groups of chitosan (Luchese et al., 2018). Increasing chitosan up to 1.0-1.5% will decrease the water contact angle, which is related to the cross-sectional morphology of chitosan-starch, even when the mixture is homogenous; however, cracks exist.

The reduction in water uptake was ascribed to the bioplastics hydrophobic characteristics of chitosan, which diminished the inherent hydrophilic nature of starch molecules. Furthermore, it was proposed that the hydrogen bonding interaction

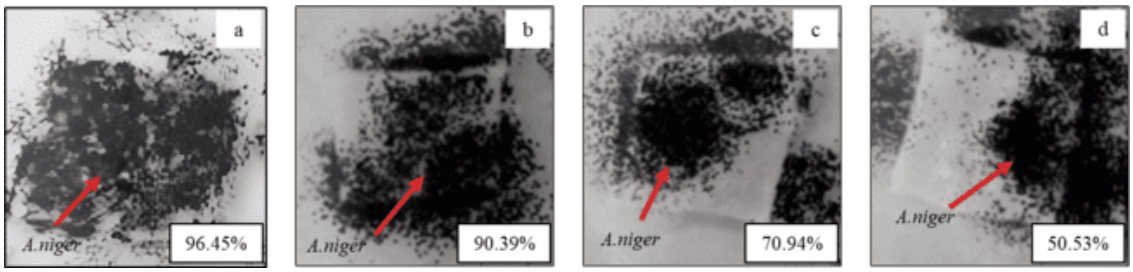

Figure 7. Biodegradability of Chitosan-Cassava Starch Bioplastics with (a) 0%, (b) 0.5%, (c) 1%, (d) 1.5% Chitosan Content

between starch and chitosan acted as a barrier, impeding the penetration of water molecules through the prototype (Pooja et al., 2023). The addition of chitosan to bioplastics can lead to changes in their thermal properties, as observed by DSC, including alterations in melting temperature, hydrophobicity, and permeability. These effects demonstrate the potential of chitosan to influence the thermal behavior and overall performance of bioplastic materials. The effects of chitosan on the thermal properties of bioplastics result from its influence on the crystalline structure, surface properties, and barrier characteristics of the material (Abdullah et al., 2020a).

The augmentation of starch-based bioplastics with chitosan presents the opportunity to improve their biodegradability and mechanical characteristics. However, the specific impact of chitosan on biodegradability may be contingent on the composition of the bioplastic and the conditions specific to the biodegradation test. The degradation of bioplastics is subject to diverse factors, such as environmental conditions, moisture levels, and microbial activity. The composition of bioplastics, particularly the starch concentration, has been proven to influence their degradability, with a higher starch concentration correlating with an elevated rate of degradation in the bioplastic (Pooja et al., 2023).

4. CONCLUSIONS

A twin-screw extruder process physically uses bioplastics from cassava-chitosan/glycerol. The density, tensile strength, and melting point of the bioplastics increased with increasing chitosan content. Moisture content, contact angle, water uptake, and elongation at rest decreased with increasing chitosan content. SEM micrographs show that the higher the chitosan levels in the bioplastic, the rougher and inhomogeneous the surfaces of the bioplastics. The FTIR spectrum shows changes in the absorption of the −OH group, where higher chitosan levels widen the absorption of the −OH group, but the overall type of functional group remains. The best mixture of bioplastic is cassava starch/glycerol containing 0.5% chitosan. It has physical properties of 0.8625 ± 0.0277 g/mL density, 68.6046 ± 3.2391° contact angle, 11.0660 ± 0.3709% water uptake, 2.0181 ± 0.0594 MPa tensile strength, 54.2243 ± 3.2623% elongation, 137.5°C thermal; and 4.9464 ± 0.1172% moisture content. In summary, the addition of chitosan to starch bioplastics results in an almost heightened density, increased resistance to water, and improved elongation properties. These effects highlight the potential of chitosan to positively alter the attributes of starch bioplastics, making positioning material for various applications.

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