



Research Article

## The Effect of Chitosan and Glycerol Variations on Galactomannan Extract from Coconut Pulp as a Raw Material for Making Bioplastics

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### ABSTRACT

This study aims to develop galactomannan-based bioplastics derived from coconut pulp with the addition of chitosan and glycerol and to evaluate their chemical structure, morphology, mechanical properties, water resistance, and biodegradability. This bioplastic is made using a simple solution casting method. The polymer and plasticizer are dissolved until homogeneous, and poured into a mold. Galactomannan extracts were synthesized into bioplastics with 3 composition variations. Fourier transform infrared spectroscopy of chitosan gave characteristic absorption bands of O–H, C–H, N–H bending, and C–O–C, with a deacetylation degree of 81%. SEM images of formulation A showed a fairly impermeable surface with tiny pores. Fourier Transform Infrared Spectroscopy of galactomannan gave –OH bands indicating the presence of extensive intra- and intermolecular hydrogen bonds within the polymer matrix. Mechanically, formulation A delivered the highest tensile strength (0.2892 MPa), while formulation C gave the best elongation (46%) and water resistance (87%). All formulations were very simple and degraded in soil and exceeded the SNI limit for biodegradation, although only elongation and biodegradation met SNI 7188.7:2016. These results indicate that coconut dregs galactomannan combined with chitosan and glycerol has the potential to be a raw material for biodegradable bioplastics.

**Keywords:** Bioplastic, Coconut Pulp Galactomannan, Chitosan, Glycerol

### 1. INTRODUCTION

Indonesia is one of the world's largest coconut producers, with its utilization primarily focused on copra, oil, and coconut milk. However, this high production rate presents its own challenges in the form of coconut flesh byproducts or waste, which are often underutilized. If left to accumulate, this coconut flesh can trigger new environmental problems due to organic decomposition. However, coconut flesh has enormous potential to be transformed into value-added products. This material is rich in carbohydrate components and polysaccharides (such as galactomannans) that have excellent film-forming properties, making it a very promising natural candidate as a raw material for future polymer materials [1][2].

The development of alternative materials from renewable sources such as coconut meat has become increasingly urgent amid the current pollution crisis. Plastic waste has become one of the most concerning environmental threats in Indonesia. According to data from the Ministry of Environment, Indonesia produces approximately 5.4 million tons of plastic waste annually, accounting for 14% of the total national waste production [3]; [4]. Although the government has attempted to reduce this figure through a policy of charging for plastic bags, its implementation has not been fully effective. The Association of Micro, Small, and Medium Enterprises (MSMEs) believes this policy merely limits the use of plastic without providing a real alternative to single-use plastic [5]. Therefore, a more comprehensive approach is urgently needed. Harnessing the potential of coconut meat polysaccharides for the synthesis of environmentally friendly bioplastics offers a dual solution: addressing the growing synthetic plastic waste crisis while maximizing the value of coconut industry byproducts [6][7]

Bioplastic is a type of plastic produced from natural sources or organic materials, such as plants, algae, starch, lignin, or microorganisms.[8] [9] [10]. Bioplastics can be classified into two main categories: biobased bioplastics and biodegradable plastics. Biobased bioplastics are designed from natural sources and contain organic components.[11]. The raw materials can be plants such as corn, wheat flour, sugar cane, algae, and coconut pulp. Meanwhile, bioplastic is a type of plastic that can decompose more quickly in nature using oxygen, heat, microbes, and other natural factors.[12].

Coconut pulp is a byproduct of coconut milk production that has not been optimally utilized. Coconut pulp is primarily composed of 93% carbohydrates, consisting of 61% galactomannan, 26% mannose, and 13% cellulose. Galactomannan, one of the primary polymers in coconut pulp, has promising applications in bioplastic production. This polymer has a structure consisting of manopyranose and galactopyranose units, which allows galactomannan to form a film layer in bioplastic production.[13].

According to research by [14] Regarding galactomannan-based bioplastics, which will involve the extraction of coconut pulp with a mixture of polyvinyl alcohol (PVA). The flexibility and compatibility of GM using biopolymers and other bioactive compounds further expand its application, positioning GM-based films and coatings as promising solutions for advancing sustainable packaging technology in the food industry. Furthermore, these bioplastics exhibit

good biodegradability, making them an environmentally friendly choice compared to conventional plastics, as they can be degraded by microorganisms in a short time.

[15] research results showed that the addition of beeswax and glucomannan had no significant effect ( $p > 0.05$ ) on thickness, water vapor permeability, biodegradability, tensile strength, and elongation. However, the addition of beeswax and glucomannan had a significant effect ( $p > 0.05$ ) on water resistance. The addition of beeswax and glucomannan also increased ( $p < 0.05$ ) the results of water resistance and tensile strength, while decreasing ( $p < 0.05$ ) the results of thickness, water vapor permeability, biodegradability, and elongation of bioplastics. It can be concluded that beeswax and glucomannan are able to provide strong bioplastic characteristics and have protective properties for products with a longer shelf life at room temperature.

Bioplastics, which are made from a single component, often tend to be more fragile than conventional plastics. Therefore, the manufacture of bioplastics requires the addition of additional materials such as plasticizers and fillers. One effective type of plasticizer is glycerol [16] ;[17] ; [18]. The addition of glycerol plays a crucial role in producing more elastic and flexible bioplastics, making them more suitable for various applications, such as packaging. Furthermore, the addition of fillers in bioplastic production is crucial because they can improve various mechanical properties, such as strength, gas resistance, melting resistance, thermal stability, and good biodegradability [19] ; [20] [21].

Based on the description above, research is needed to investigate the effects of varying chitosan and glycerol on galactomannan extract derived from coconut pulp to form bioplastics. This research is needed to create bioplastics with improved mechanical and functional properties, such as tensile strength, water resistance (swelling), and biodegradability. Furthermore, analysis of the bioplastic's morphological structure will be conducted using Scanning Electron Microscopy (SEM) techniques to determine the distribution and relationships between components in the bioplastic. This research is expected to produce plastics with the best characteristics.

## 2. EXPERIMENTAL SECTION

The research was quantitative, using an experimental method, and used two variables: an independent variable and a dependent variable. In this study, the independent variable was a variation of galactomannan from coconut pulp mixed with chitosan and glycerol. The

dependent variables were tensile strength and elongation, water resistance (swelling), biodegradation, and morphology.

### 2.1. Materials

Coconut dregs are used to produce galactomannan compounds derived from restaurant waste; glycerol is used as a plasticizer; chitosan is used commercially; methanol p.a. (97%) and distilled water are used as solvents; and 1% ( $\text{CH}_3\text{COOH}$ ) is used for the synthesis of chitosan from shells.

### 2.2. Instrumentation

All solutions were prepared using standard laboratory glassware, including beakers, volumetric flasks, and Erlenmeyer flasks. The mixture was homogenized using a magnetic stirrer, while the mass of each ingredient was measured using an analytical balance. Filtration was performed using filter paper and a funnel, and liquid transfer was performed using a volumetric pipette. Samples were molded into trays and dried in Petri dishes, then covered with aluminium foil to avoid contamination.

The functional groups of the samples were analyzed using FTIR spectroscopy (Bruker Tensor 27, Germany). Surface morphology was examined using an SEM (JEOL JSM-6510LV, Japan). Mechanical properties were evaluated through tensile tests using a universal testing machine (Instron 3365, Instron Corp., USA).

### 2.3. Procedure

#### 2.3.1. Galactomannan from Coconut Pulp

Galactomannan from coconut pulp refers to [22]. The coconut pulp was washed with running water to remove coconut milk and impurities, then dried in the sun for 2 days. The dried sample was then ground and sieved using a 40-mesh sieve. A total of 50 g of coconut pulp powder was extracted with 350 mL of distilled water at 55°C for 3 hours with moderate stirring. The extraction result was then filtered, and the filtrate was added with 97% ethanol at a ratio of 1:3 (filtrate: ethanol) to precipitate the galactomannan. The mixture was left at 10°C for 12 hours. The precipitate formed was then dried in the sun for about 24 hours, ground, and sieved using a 100-mesh sieve to obtain galactomannan with a uniform size.

#### 2.3.2. FTIR Analysis of Chitosan

FTIR analysis of chitosan was performed to identify functional groups in the sample. The chitosan sample was mixed with potassium bromide (KBr) and then pressed to form thin

pellets. The sample pellets were then analysed using an FTIR instrument at specific wavenumbers to obtain an infrared absorption spectrum. The resulting spectrum was then used to identify the characteristic functional groups of chitosan based on the position of their absorption bands [23].

### 2.3.3. Scanning Electron Microscopy (SEM) Analysis

The morphology and surface characteristics of the synthesized nanoparticles were analyzed using Scanning Electron Microscopy (SEM). The dried samples were carefully dispersed on aluminum stubs using conductive carbon tape. Before imaging, the samples were sputter-coated with a thin layer of gold to enhance conductivity and prevent electron charging.

### 2.3.4. Bioplastic Synthesis

The manufacture of bioplastics was carried out based on [24] Regarding galactomannan-based bioplastics resulting from coconut pulp extraction with a mixture of polyvinyl alcohol (PVA). Variations in bioplastic composition can be seen in Table 1. The table below explains samples A, B, and C with variations 1 and 2 in the materials used in this study.

**Table 1.** Variations in bioplastic-forming materials

| No | Sample | Galactoman (g) | Glycerol plasticizer (mL) | Chitosan (g) |
|----|--------|----------------|---------------------------|--------------|
| 1  | A      | 5              | 2                         | 1            |
| 2  | B      | 5              | 2.5                       | 2            |
| 3  | C      | 5              | 3                         | 3            |

The chitosan solution was prepared by dissolving chitosan in 2% acetic acid until a homogeneous solution was obtained. The mixture was then stirred using a magnetic stirrer until all the chitosan was completely dissolved. The resulting chitosan solution was then used for subsequent analysis or formulation[25].

Bioplastic is made by dissolving 5 g of galactomannan in 50 mL of distilled water, then stirring at 65°C until thickened. Next, glycerol is added according to variations, then heated and stirred until homogeneous. After that, a 1%, 2%, or 3% chitosan solution is added and heated again until a gelatinized solution is formed. The mixture is poured into a mold, dried for 2 days, left for 24 hours at room temperature, then removed from the mold and stored in an airtight container.

### 2.3.5. Bioplastic Characteristics Test

The characteristics of bioplastics were analysed through tensile strength, elongation, swelling, biodegradation, and morphology tests. Tensile strength and elongation tests were

conducted using UTM according to ASTM D882, while swelling was assessed based on the sample's ability to absorb water.

### 2.3.6. Biodegradation Test

The biodegradability of the bioplastic films was evaluated using a soil burial test, adapted from the standard guidelines of ASTM D5988, and the procedures described by [26]. For sample preparation, the bioplastics were cut into uniform dimensions of 2 cm × 2 cm, and their initial dry weights ( $W_0$ ) were accurately measured using an analytical balance. The degradation test was conducted in containers filled with natural compost soil, serving as the degradation medium (pH ~6.5–7.0). To simulate an active environmental condition for microbial degradation, the soil moisture content was maintained at 50–60% via periodic water spraying, and the samples were kept at an ambient room temperature of 25–28°C.

The testing duration was set for a monitoring period of 30 days. At predetermined intervals (days 7, 14, 21, and 30), the buried samples were carefully retrieved from the soil. The retrieved films were gently brushed to remove adhering soil particles, rinsed with distilled water, and subsequently dried in an oven at 60°C until a constant final weight ( $W_t$ ) was achieved. The primary measurement parameter for biodegradation was the weight loss percentage, calculated using the following equation:

$$\text{lose weight (\%)} = \frac{W_0 - W_t}{W_0} \times 100$$

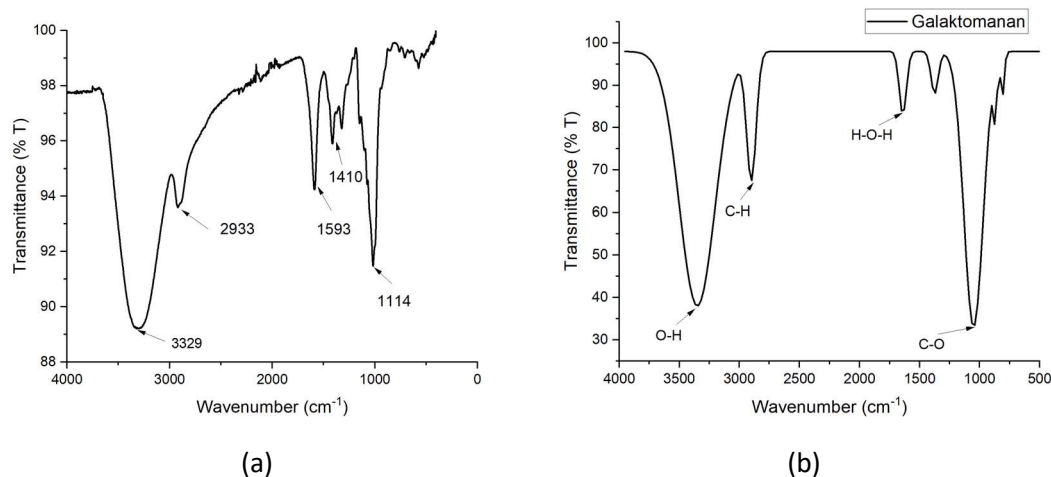
All weight measurements were performed in triplicate to ensure reproducibility. Furthermore, the surface morphology of the bioplastics before and after the degradation process was observed using Scanning Electron Microscopy (SEM). This qualitative parameter was utilized to evaluate the surface structure, filler distribution, and the emergence of pores and cracks caused by microbial activity (Zamora et al., 2025).

## 3. RESULTS AND DISCUSSION

### 3.1. FT IR Chitosan Results

FTIR spectrum of the sample in Figure 1(a) shows the presence of typical functional groups of chitosan through a broad absorption band at 3329  $\text{cm}^{-1}$  attributed to O-H and N-H stretching vibrations due to the presence of interchain hydrogen bonds. The band at 2933  $\text{cm}^{-1}$  indicates aliphatic C-H stretching vibrations, while the band at 1593  $\text{cm}^{-1}$  is related to N-H bending vibrations of amine groups, confirming the presence of free amine groups as the main characteristic of chitosan. The band at 1410  $\text{cm}^{-1}$  indicates C-H deformation, and the

strong band at  $1114\text{ cm}^{-1}$  indicates C-O-C/C-O stretching vibrations in the polysaccharide structure. Overall, the appearance of these bands is consistent with the FTIR characteristics of chitosan and indicates that the analyzed sample contains the main functional groups of chitosan [27].



**Figure 1.** FT IR (a) Chitosan Results and (b) Galaktomanan

The deacetylation degree (DD) of chitosan of 81% indicates that most of the acetyl groups in chitin have been released during the deacetylation process, so that the sample characteristics are suitable for chitosan. Based on SNI 7949:2013, the minimum deacetylation degree of chitosan is 75%; thus, the results obtained have met the required chitosan quality standards.[28].

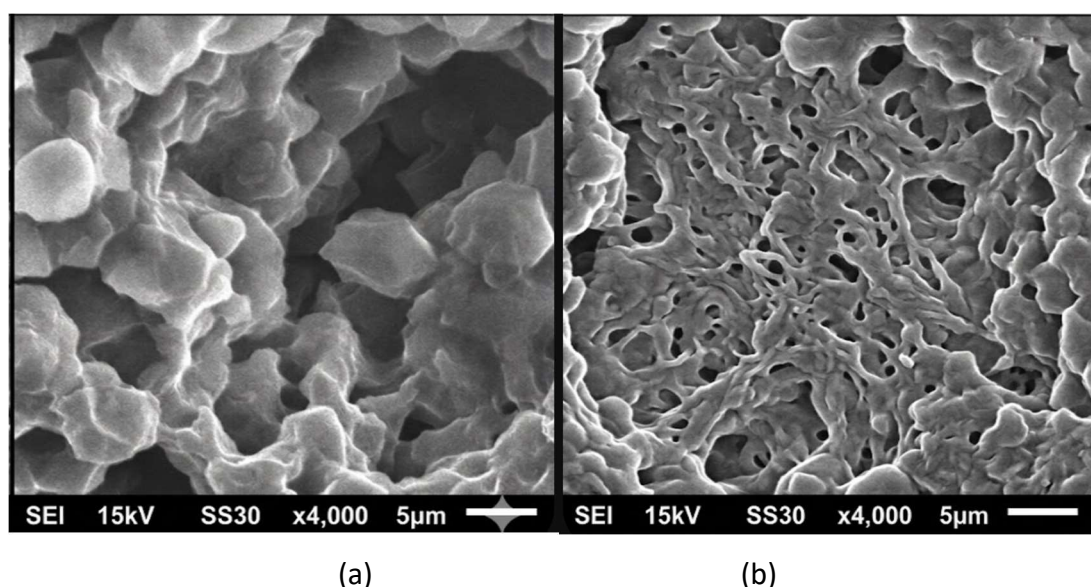
The FTIR spectrum of the galactomannan sample (Figure 1(b)) shows typical absorption bands common to galactomannan polysaccharides. The broad and intense absorption band at around  $3350\text{ cm}^{-1}$  is attributed to the stretching vibrations of the hydroxyl (–OH) groups, indicating the presence of extensive intra- and intermolecular hydrogen bonding within the polymer matrix. The absorption peak at  $2900\text{ cm}^{-1}$  corresponds to the C–H stretching vibrations of the aliphatic groups (CH and CH<sub>2</sub>) in the sugar ring. A minor band near  $1640\text{ cm}^{-1}$  is assigned to the H–O–H bending vibration, which is associated with the presence of bound water molecules due to the highly hygroscopic nature of the polysaccharide. In the fingerprint region, a prominent and sharp absorption band at  $1050\text{ cm}^{-1}$  is attributed to a combination of C–O and C–C stretching vibrations, representing the typical glycosidic bond (C–O–C) of the pyranose ring. Furthermore, the specific anomeric configuration of galactomannan is confirmed by two weak but distinctive bands at lower wavenumbers. The absorption band at  $870\text{ cm}^{-1}$  is a characteristic marker for the  $\beta$ -glycosidic bond in the D-manopyranose main

chain, while the peak at  $810\text{ cm}^{-1}$  indicates the presence of an  $\alpha$ -glycosidic bond referring to the D-galactopyranose branch chain.

These spectral features are in good agreement with previous characterization studies of galactomannan from various botanical sources [29]. For example, a similar study by [30]. Nearly identical absorption profiles were reported for guar gum and locust bean gum, particularly highlighting the structural conservation of the anomeric vibrations at  $\sim 870\text{ cm}^{-1}$  and  $\sim 810\text{ cm}^{-1}$ . The agreement of these fingerprint regions with existing literature confirms that the extraction process successfully isolated galactomannan while maintaining its native macromolecular structure without degradation or modification of unwanted functional groups.

### 3.2. Bioplastic Morphology Results

Morphological analysis was conducted to evaluate the surface structure, the presence of pores, cavities, and cracks in the galactomannan–chitosan composite bioplastic film plasticized with glycerol. Characterization was carried out using a scanning electron microscope (SEM) at several magnification levels to observe the topographic details and uniformity of the film surface. The formulation tested morphologically was the sample with the best mechanical properties (variation A: 5 g galactomannan, 2 mL glycerol, 1 g chitosan).



**Figure 2.** Results of SEM analysis at 4000 $\times$  magnification (a) Variation A, (b) Variation C

SEM images show that bioplastics have small pores and a relatively dense structure, which is consistent with reports that the addition of fillers or supporting components can increase the matrix density and produce a smoother and more compact surface [31]. In some areas,

voids/empty spaces are visible, which can be attributed to the entrapment of air bubbles during the mixing and film formation process, a phenomenon also observed in other chitosan-composite films when there are dispersion imperfections or the presence of separate phases [32].

Additionally, cracks and dark areas were observed, indicating inhomogeneity of the solid phase. Chitosan is known to have limited solubility under certain conditions, and this property can affect film homogeneity and structure formation, potentially resulting in agglomerated areas and morphological defects.[33]. Non-uniform structure, the presence of pores or cracks in biopolymer films, has been reported to affect mechanical properties (tensile strength, elongation) and functional performance, including water absorption behavior and degradation properties [34].

Morphological analysis of bioplastics was conducted to determine the surface structure, cracks, and smoothness of the bioplastic material. In this study, morphological testing was conducted using a scanning electron microscope (SEM). The bioplastic sample with the highest tensile strength was variation A, which consisted of 5 grams of galactomannan, 2 mL of glycerol, and 1 gram of chitosan. SEM observations were conducted at various magnification levels to examine the details of the surface structure and cracks in the composite bioplastic. Figure 2 shows that the resulting bioplastic has a surface morphology with small pores. This indicates a higher particle density. Empty spaces and cavities caused by air bubbles were also observed. These air bubbles appeared due to the imperfect mixing of galactomannan, glycerol, and chitosan during the mixing process [35].

Black cracks were observed on the surface of the bioplastic, suspected to be caused by incompletely dissolved chitosan clumps. Chitosan is difficult to dissolve in water, disrupting the homogeneity of the mixture and increasing viscosity. This condition causes cracks on the surface of the resulting bioplastic and reduces its elasticity. This characteristic is common in bioplastic materials and makes them less elastic. This characteristic is common in bioplastic materials and can affect the mechanical properties and rate of biodegradation, such as the tensile strength of the bioplastic.

The morphology of the bioplastic sample of variation C exhibits a significantly different topographic profile than that of variation A. SEM images of variation C show a looser, less compact surface structure, dominated by the presence of large pores (macropores) and

widely distributed voids. This decrease in matrix density indicates a weakening of the intermolecular interactions and cross-linking density between galactomannan and chitosan within the polymer network.

In contrast to variation A, which has a dense and rigid structure, the presence of large voids in variation C contributes to an increase in free volume within the bioplastic matrix. These voids reduce the spatial barriers between polymer chains, providing greater mobility for the polymers to slide and stretch when subjected to tensile forces [36]. This visual phenomenon is highly consistent with the results of mechanical tests, where variation C recorded a drastic increase in elongation percentage, with a consequent decrease in the material's load-bearing capacity (tensile strength).

Furthermore, the hollow and discontinuous surface topography of the C variation plays a crucial role in determining the physicochemical and biological performance of the material. Morphologically, these macro-voids act as primary capillary pathways that facilitate water diffusion into the film matrix. This open matrix structure not only increases the affinity for water absorption but also significantly expands the contact surface area for hydrolytic microorganisms [37].

### 3.3. Bioplastic Characteristics Test Results

The bioplastic characteristic test in this study examined the mechanical properties, water resistance, and biodegradation of galactomannan-based bioplastics with the addition of chitosan and glycerol. Three composition variations (A, B, C) were compared with the Indonesian National Standard (SNI) 7188.7:2016 reference, with the research results as in Table 2.

**Table 2.** Bioplastic Characteristics Test Results

| Variation | Tensile Strength (Mpa) | Elongation (%) | Water resistance (%) | Biodegradation (%) |
|-----------|------------------------|----------------|----------------------|--------------------|
| A         | 0,2892                 | 16             | 62                   | 90                 |
| B         | 0,2143                 | 26             | 51                   | 95                 |
| C         | 0,1679                 | 46             | 87                   | 97                 |

Based on Table 2, the highest tensile strength is found in variation A at 0.2892 MPa, but still far below the SNI standard of 24.7–30.2 MPa. The lowest tensile strength is in variation C at 0.1679 MPa, associated with the excessively high chitosan content, which makes the solution inhomogeneous, stiffer, and more porous. This reduces tensile strength because

chitosan is difficult to dissolve, increases viscosity, disrupts homogeneity, and produces a stiff and porous plastic [38].

The bioplastic from this research resulted in increased strength but decreased the percentage of elongation because the increasing number of hydrogen bonds made the distance between molecules closer together.[38]. The highest elongation was 46% in variation C, according to the SNI range of 21–220%, while the lowest was 16% in variation A. Elongation is inversely proportional to tensile strength; increasing chitosan increases hydrogen bonding and intermolecular density, thus changing strength and elongation[39].

Water resistance testing showed that the addition of chitosan and glycerol increased the ability to absorb water due to its hydrophilic properties, but excess glycerol can reduce water resistance [40] ; [41]. This reinforcement effect has been empirically proven to significantly increase the Tensile Strength value of variation A to reach the highest value of 0.2892 MPa. However, due to the stiffness of the formed microstructure, the flexibility of the matrix decreases drastically, resulting in the lowest percentage of elongation at break (Elongation), which is only 16%. Microscopic phenomena can explain why the Tensile Strength value of Variation C decreases sharply to only 0.1679 MPa. However, this reduction in cross-linking density provides a larger free space (free volume) for the polymer chains to slide and stretch each other when given external mechanical force, which directly triggers an exponential increase in the elongation value to reach 46%.

Biodegradation data show that all of these bioplastic formulations are highly degradable ( $\geq 90\%$  in 7 days) and exceed the SNI limit. The addition of chitosan and glycerol strengthens the structure and water resistance but tends to slow the degradation rate, despite the high final degradation percentage [42] ; [43]. The optimal formulation depends on the target: if very fast degradation is desired, a composition similar to variation A can be chosen, whereas if better water resistance is needed but still biodegradable, galactomannan–chitosan–glycerol bioplastics show low tensile strength, but elongation, durability, and water resistance are quite good. Variations in the composition of chitosan and glycerol greatly affect the balance between mechanical strength and water absorption capacity, so formulation optimization is necessary to meet all international standard parameters.

This research resulted in a higher chitosan variation, a denser structure, and slower degradation. Variation C, with the highest chitosan, had the longest degradation time;

chitosan makes the bioplastic molecular bonds tighter and relatively resistant to microorganisms, thus slowing degradation. Glycerol can accelerate, but if excessive, it actually inhibits it. In general, increasing glycerol can accelerate degradation, but at too high a concentration it actually reduces the effectiveness of biodegradation because glycerol is difficult for microorganisms to break down. This is seen in variation C (the highest glycerol), which actually degraded the longest. Variation A: highest tensile strength, lowest elongation, lowest water resistance, and fastest degradation. Variation C had the lowest tensile strength, highest elongation and water resistance, and the longest degradation time but the highest percentage of biodegradation, indicating a compromise between strength, flexibility, water resistance, and degradation speed.

In line with these results, several studies have shown that the interaction and distribution of chitosan in a starch matrix can increase tensile strength but also change elongation, and that films with smooth surfaces and compact structures tend to have better mechanical properties and lower water barrier porosity [44]. Thus, the morphology observed in galactomannan–chitosan films has the potential to correlate with the tensile strength and elasticity values measured in this formulation.

#### 4. CONCLUSION

This study demonstrated that varying the composition of coconut pulp galactomannan-based bioplastics with the addition of glycerol and chitosan significantly impacted the mechanical properties, water resistance, and biodegradability of the bioplastic. Formulation A produced the highest tensile strength of 0.2892 MPa and the best biodegradation performance, with a degradation rate reaching 90% in 3 days, exceeding the Indonesian National Standard (SNI) requirements ( $\geq 60\%$  in 1 week). Meanwhile, formulation C demonstrated the highest elongation (46%) and the best water resistance (87%), indicating better flexibility and water stability. However, of all tested parameters, only the elongation and biodegradation values met the SNI 7188.7:2016 standard.

Morphological analysis using SEM on formulation A revealed a surface with small pores, reflecting high particle density, but voids caused by air bubbles due to inhomogeneous mixing were still present. Furthermore, the presence of incompletely dissolved chitosan agglomerates caused dark-colored cracks that reduced the material's homogeneity and

elasticity. These morphological characteristics are commonly found in bioplastics and have the potential to influence the rate of biodegradation and mechanical properties, including the tensile strength of bioplastics.

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## AUTHOR CONTRIBUTIONS

Yuna Salza Yasmina contributed to the research data collection. Husnawati Yahya contributed to the editing of the manuscript, and Khairun Nisah contributed to the data analysis, article preparation, and review.

## CONFLICT OF INTEREST

The author declares that he has no conflict of interest related to the content of this article.

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