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Comparative Analysis of the Characteristics of Biodegradable Plastics Based on Cassava Starch, Durian Seed Starch, and Purple Sweet Potato Starch with Chitosan and Glycerol Modification

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ABSTRACT

Conventional plastics made from synthetic polymers have non-biodegradable properties that cause environmental problems. This study aims to compare the characteristics of biodegradable plastics based on cassava starch, durian seed starch, and purple sweet potato starch. Film production was carried out using a blending method with varying chitosan concentrations (0; 1; and 2%). Characterizations carried out included tensile strength, elongation, swelling, air solubility, and biodegradation tests. The results showed that the type of starch and chitosan concentration significantly affected the mechanical and physical properties of biodegradable plastics. Purple sweet potato starch plastic had the highest tensile strength, the lowest swelling and air solubility values, but the slowest biodegradation rate. Cassava starch plastic showed balanced mechanical and degradation characteristics. Meanwhile, durian seed starch plastic had the lowest tensile strength, the highest swelling and air solubility values, but the fastest biodegradation rate.

I. INTRODUCTION

Plastic is widely used as a food packaging material because it is relatively inexpensive and can protect products from damage. However, its use can have serious environmental impacts due to its non-biodegradability[1]. As a synthetic polymer, plastic takes a very long time to degrade, even hundreds of years. Furthermore, when plastic is burned, the resulting carbon emissions can also pollute the environment [2].

The difficult degradation of plastic results in the accumulation of plastic waste, increasing the risk of pollution [2]. Furthermore, the accumulation of plastic waste has the potential to release toxic chemicals that worsen environmental conditions. To address this problem, a packaging material is needed that is environmentally friendly, readily available, inexpensive, abundant in nature, and has comparable strength to conventional plastics, namely biodegradable plastic. Biodegradable plastic is a biopolymer with the potential to replace conventional plastics and can be made from starch, cellulose, chitosan, gelatin, or other biopolymers using appropriate methods [3].

Recent research has focused on the development of biodegradable plastics as a substitute for conventional plastics. One potential raw material for biodegradable plastics is starch. Various studies have been conducted to develop starch-based biodegradable plastics because it is readily available in the environment, making it a relatively inexpensive raw material, and it can form films well [3]. Research by Prameswari et al., 2022, shows that starch has significant potential for producing environmentally friendly plastics. This development demonstrates the significant potential for creating environmentally friendly packaging that is not only safe for use in food products but also able to reduce the environmental impact of conventional plastic waste.

This research seeks to utilize starch from various potential plant species, such as cassava starch, durian seed starch, and purple sweet potato starch. According to research byIsmaya et al. (2021), cassava

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starch has the potential to be used as a primary ingredient in biodegradable plastic production due to its high amylose content, which allows it to form films with good mechanical strength. The use of cassava as a raw material for biodegradable plastics was developed due to the ease of starch isolation and its relatively high starch content, reaching 90% [4].

Durian seed starch is an agro-industrial waste product rich in amylopectin and can be used as an additive in biodegradable plastics to increase flexibility. Durian seed starch was chosen for biodegradable plastic production due to its abundant availability and underutilization. It also contains a high carbohydrate content, particularly starch, of approximately 43.6%, making it a potential alternative to other biodegradable plastics [5]. Meanwhile, purple sweet potatoes were chosen due to their natural origin, easy access, abundant availability, relatively low cost, and ease of soil degradation. Purple sweet potatoes contain 31.67% starch [6]. Purple sweet potato starch not only has functional properties as a film former but also contains anthocyanin pigments, which have the potential to provide added value in the form of functional properties for smart packaging and aesthetics for biodegradable plastics.

However, using only starch as the main ingredient will result in a brittle, stiff, inelastic film that is unable to absorb water. To improve the mechanical properties and characteristics of biodegradable plastics, additives are needed. These additives include plasticizers, which increase film elasticity, and fillers, which strengthen the structure and improve the film's mechanical properties.

Plasticizers are additives that increase plasticity while reducing viscosity and friction during the biodegradable plastic manufacturing process. One of the most commonly used plasticizers is glycerol, a natural plasticizer with relatively high hygroscopic properties. According to Rahmawati et al. (2020), adding glycerol to starch-based films can increase elongation, although at too high a concentration it can reduce the film's tensile strength. The addition of glycerol as a plasticizer reduces internal hydrogen bonds, increasing the stability of the biopolymer chains, and improving the film's mechanical properties [7].

Fillers function as fillers to improve the mechanical properties and characteristics of films [7]. Chitosan has antimicrobial properties, can increase mechanical strength, and improve water vapor barrier properties. In research by Solekah et al., 2021, the addition of chitosan to starch-based plastics can increase the tensile strength of films. According to Khodijah & Tobing (2023), the addition of chitosan can trigger the formation of hydrogen bonds between the polymer chains and the starch structure, thereby strengthening molecular interactions, ultimately increasing the film's tensile strength.

Several previous studies have reported that the use of cassava starch, sweet potato starch, and durian seed starch, separately with the addition of chitosan or glycerol, can produce biodegradable plastics with quite good characteristics. The focus of this research is to characterize the mechanical, physical, and degradation properties of the resulting biodegradable plastics. It is hoped that this will provide an alternative formulation for environmentally friendly biodegradable plastics that offer added functional and aesthetic value.

II. MATERIALS AND METHOD

Biodegradable plastic is made using a blending method, which involves mixing all the ingredients and then heating them at 90°C. The manufacturing process consists of three stages. In the first stage, starch is mixed with a solution of chitosan, CMC, and glycerol, then stirred slowly. In the second stage, the mixture is heated while stirring using a hot plate and magnetic stirrer to a temperature of 90°C, so that the solution becomes homogeneous and air bubbles disappear. Next, in the third stage, the solution is dried in an oven at 90°C and stored at room temperature (24°C). The independent variable in this study is the variation in chitosan concentration (0%, 1%, and 2%). Biodegradable plastic testing is carried out through tensile strength tests, elongation tests, water solubility tests, swelling tests, and biodegradation tests.

III. RESULT AND DISCUSSION

Characterization Results of Biodegradable Plastic

Based on the research conducted, films with various chitosan variations were obtained to determine the optimum composition used in the manufacture of biodegradable plastics and the comparison of biodegradable plastics based on purple sweet potato starch, cassava starch, and durian starch. The resulting plastics were then analyzed for tensile strength, elongation, water solubility, swelling, and biodegradation. The following data shows the results of the biodegradable plastic analysis that was conducted:

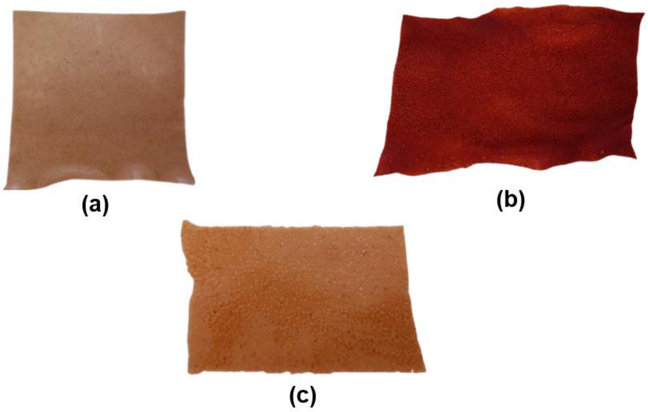

Figure 1. Results of Biodegradable Plastic After Drying (a) Cassava Starch Film, (b) Purple Sweet Potato Starch Film, (c) Durian Starch Film

The results of the study showed that cassava starch, durian seeds, and purple sweet potatoes successfully formed a thin biodegradable plastic film with a purplish to brownish color depending on the composition. The film using durian starch produced a sheet-shaped film that was elastic, slightly sticky, easy to remove, with a certain thickness and a light yellow color originating from the remains of other non-starch components (protein or fat). The results of the film using cassava starch showed a thin, transparent sheet that was translucent and elastic. The white color typical of pure starch indicates good starch isolation [8]. The results of the film using purple sweet potatoes produced a purple film originating from anthocyanin compounds that function as natural dyes and antioxidants in the film [9].

Tensile Strength and Elongation Test

The maximum stress of biodegradable plastic when resisting an applied force during testing is known as tensile strength. The tensile strength value of biodegradable plastic describes the magnitude of its tensile strength when the bioplastic is subjected to a load [10]. Tensile strength measurements determine the magnitude of the force required to achieve maximum tensile strength per unit surface area of the film to stretch or elongate [11].

Table 1. Characteristics of the Strength of Biodegradable Plastic with Chitosan Variation

Chitosan Variation	Tensile Strength	Elasticity	Mechanical Strength
1 gram	No tensile strength	Moderately elastic	Very easy to tear
2 grams	Tensile strength	Not elastic	Easy to tear
3 grams	Very high tensile strength	Not elastic	Not easy to tear

Based on the data obtained, variations in chitosan affect the tensile strength, elasticity, and strength of the film. The greater the chitosan content in the sample, the greater the tensile strength. The increased tensile strength is due to chitosan forming hydrogen bonds between molecules, making the film denser, stronger, and more difficult to break. Hydrogen bonding interactions between chitosan, starch, and glycerol can occur if free OH groups are present, allowing for bonding between compounds. The higher the chitosan concentration, the more hydrogen bonds there are in the film, requiring greater energy to break them. This, in turn, increases the tensile strength of biodegradable plastics [12].

However, increased tensile strength can also decrease the film's elongation. Elongation testing is performed to determine the percentage of film elongation during tensile strength testing. This decrease is related to the presence of voids resulting from the cleavage of bonds between polysaccharides by glycerol, and elasticity due to the reduced distance between molecules. This weakens the intermolecular bonds in the plastic, resulting in a lower tensile strength [13].

This research aligns with research (Ekariski & Yudhistira, 2017)[14], which found that varying the amount of chitosan used increased the film's tensile strength. Hydrogen bonds form between the $NH3^+$ in chitosan and the hydroxyl groups in starch. The amine groups in chitosan react with acetic acid to form $NH3^+$, which can form a film. Hydroxyl bonds, which are easily separated during gelatinization, bind to the $NH3^{+}$ in chitosan, increasing the number of hydroxyl bonds.

The addition of chitosan can decrease the elongation percentage of the film by producing a thicker film with a higher concentration of dissolved components, resulting in stiffer and denser films. The hydrogen bonds formed between chitosan and amylose and amylopectin strengthen the film structure but reduce its elasticity. The greater the concentration of chitosan added, the more hydrogen bonds are formed, so that the elasticity of the film decreases [15].

Table 2. Strength Characteristics of Biodegradable Plastics Based on Starch Variations

Starch Variation	Tensile Strength	Elasticity	Mechanical Strength
Purple Sweet Potato	Very high tensile strength	Not elastic	Not easy to tear
Cassava	Moderate tensile strength	Less elastic	Easy to tear
Durian Seed	No tensile strength	Less elastic	Easy to tear

Based on the data obtained, purple sweet potato has a stronger tensile strength than cassava starch and durian seed starch. The tensile strength of purple sweet potato demonstrates the highest structural integrity. This strength is driven by the starch structure, which is ideal for forming a dense film matrix, supported by interactions with chitosan, and reinforced by non-starch components [16]. Purple sweet potato contains anthocyanins, which can influence the crystal structure and intermolecular interactions, thereby increasing film density [9]. Starch with a higher amylose content tends to form stronger films. Purple sweet potato has a higher amylose ratio than cassava starch or durian seed starch, resulting in higher tensile strength.

High strength indicates a very high cross-linking density between polymer chains. This can be caused by the high amylose content in sweet potato starch (forming strong crystals) or the interaction of anthocyanins with the starch chains, which restricts molecular movement. Very strong tensile strength is often correlated with low elasticity. The highly structured chitosan and starch matrix restrict the movement of the polymer chains, resulting in a rigid film. This research aligns with research [17], which found that the use of sweet potato starch produced a film with excellent mechanical strength. The addition of chitosan increased mechanical strength but made the material less elastic, resulting in a decrease in elongation.

Based on the data obtained, it can be analyzed that cassava starch has a lower tensile strength than purple sweet potato starch, but still superior to durian seed starch. The cassava starch isolation process tends to produce starch with high purity with minimal residual fat or protein [4]. Cassava starch has a relatively balanced polysaccharide structure and amylose and amylopectin content, suitable for forming a strong and elastic polymer matrix. Cassava starch easily gelatinizes, forming a homogeneous, dense film with strong intermolecular bonds. The molecular structure of cassava starch-based films tends to be more homogeneous and evenly distributed, resulting in good stress distribution during tensile and elongation tests, making them resistant to damage [18].

Based on the data obtained, durian starch has a lower tensile strength than purple sweet potato starch and cassava starch. Durian seeds are waste that contain significant amounts of non-starch compounds such as protein, fiber, or fat [5]. Durian seeds are food waste that cannot be consumed directly due to their hazardous cyclopropane fatty acid content, requiring several processing steps, such as cooking and steaming [19]. Durian seeds contain fiber and other components that are less homogeneous enough to form a film. These components act as interfering factors and can hinder the formation of hydrogen bonds between the starch, chitosan, and glycerol, causing the film matrix to become brittle and reduce tensile strength. The presence of non-starch impurities can act as blockers, preventing chitosan from interacting effectively with the hydroxyl groups in starch. As a result, the strengthening effect of chitosan is not optimal, and the film remains weak.

Despite high starch content, the exact composition is lower or less than ideal for film formation. If amylopectin content is too dominant and amylose is less stable, the film matrix density will decrease. The matrix becomes less structured, resulting in fewer hydrogen bonds, resulting in lower film tensile strength. Low tensile strength indicates a less dense film structure. This is caused by low amylose content or the presence of residual components that interfere with the formation of strong hydrogen bonds between starch and chitosan, making the matrix prone to failure when pulled [20].

Swelling Test

Biodegradable plastics based on purple sweet potato, cassava starch, and durian seed starch exhibit swelling test characteristics influenced by the hydrophilic properties of the starch and its plastic formulation. The swelling test measures the extent to which the film absorbs and retains water without dissolving after being immersed in water for a specified period [21]. The swelling value is influenced by the amylose content, which is more hygroscopic than the branched structure of starch and amylopectin, the size of the starch granules, the fiber, protein, and lipid content, the homogeneity of the film matrix, and the hydrogen interactions within the polymer structure. The higher the affinity of the starch for water, the higher the swelling test results. In general, all starch-based films have a relatively high swelling percentage due to their natural hydrophilic nature, but there are differences based on the starch structure [20].

Table 3. Swelling Test Results of Biodegradable Plastics Based on Starch Variations

Starch Variation	Initial Weight (g)	Final Weight (g)	Swelling (%)
Purple Sweet Potato	0,603	1,020	69,15
Cassava	0,538	1,203	123,60
Durian Seed	0,337	0,911	170,33

Each starch produced different swelling test results, purple sweet potato starch produced the lowest swelling test value of 69.15%, which indicates that this film absorbs the least water, followed by cassava starch which has a swelling test value that is almost 2 times greater than purple sweet potato, namely 123.60%, and the highest swelling test value is in durian seed starch at 170.33%, which means this film absorbs the most water.

(a) (b) (c)

Figure 2. Biodegradable Plastic After Soaking (a) Purple Sweet Potato Starch Film, (b) Cassava Starch Film, (c) Durian Seed Starch Film

Purple sweet potatoes produced the most water-stable films, with the lowest swelling values, indicating the densest, stiffest, and strongest polymer matrix [16]. The dense hydrogen bonds between the starch and chitosan chains significantly limit water penetration. This film exhibited very high tensile strength, indicating a very dense and rigid polymer matrix with strong hydrogen bonds. This dense structure limits free space (pores) and prevents water molecules from penetrating and being trapped, resulting in a low swelling test percentage.

Purple sweet potatoes contain high levels of amylopectin starch, resulting in a more branched structure. Purple sweet potatoes also contain anthocyanins and phenolic compounds that can act as cross-links with the starch chains, stabilizing some of the polymer network, strengthening the bonds and inhibiting water penetration. Purple sweet potato starch granules are large to medium in size, facilitating gelatinization, resulting in a smoother gel and more even water distribution [22].

Cassava starch produced a swelling test value nearly twice that of purple sweet potato, at 123.60%. Cassava starch has a higher water absorption capacity than purple sweet potato, but still lower than durian seed starch, indicating its matrix is quite hydrophilic and has more free space (pores) to accommodate water compared to purple sweet potato. Cassava starch has a relatively pure and regular starch structure and a very high amylopectin content, resulting in many exposed hydroxyl groups, allowing the film to absorb water very quickly due to its highly hydrophilic nature and its ability to bind water. Cassava starch granules are very fragile, so when soaked, the matrix expands rapidly. The polymer bonds in the cassava starch structure are less dense; in the absence of strong cross-linking, water readily enters the film matrix.

In theory, durian seed starch typically has a harder time absorbing water due to its high amylose content and denser film structure, making it difficult for water to penetrate. Durian seed starch contains significant amounts of protein and fat, which act as barriers to water penetration. Starch granules are harder and less homogeneous, so they don't break down quickly when soaked, resulting in slower water absorption. However, in this experiment, the lower initial film weight resulted in faster water saturation. With a small film mass, even a small amount of water absorption drastically increased the swelling percentage. A very high swelling value indicates that this film has the loosest network structure and low interpolymer cohesiveness. This aligns with the tensile strength test results, which showed the durian seed film to be the weakest. This loose structure allows water molecules to enter and become trapped within the matrix.

Durian seed starch has a less cohesive structure, resulting in the lowest tensile strength and the presence of non-starch impurities. A less dense matrix creates more voids, allowing the film to absorb and retain greater amounts of water, resulting in a high swelling percentage. A high amylose content can provide a dense structure. However, if the mixing or heating process is not optimal, the amylose fails to form a dense crystalline film, resulting in the film becoming brittle and absorbing more water. An inhomogeneous film can also cause a sharp increase in the swelling percentage [20].

Water Solubility Test

Biodegradable plastics based on purple sweet potato, cassava starch, and durian seed starch have water solubility test characteristics influenced by the composition of the starch and plasticizer used, the content of lipids, proteins, and other compounds, the presence of chitosan, the film structure, and the level of hydrophobicity of the material. Water solubility testing is conducted to determine how much the film will dissolve or lose mass when immersed in water for a certain period of time. Naturally, starch (amylose and amylopectin) is a polysaccharide with many hydroxyl groups, making it hydrophilic and readily soluble in water.

(a) (b) (c)

Figure 3. Biodegradable Plastic Soaking Process in Water Solubility Test (a) Purple Sweet Potato Starch Film, (b) Durian Seed Starch Film, (c) Cassava Starch Film

(a) (b) (c)

Figure 4. Biodegradable Plastic After Drying (a) Purple Sweet Potato Starch Film, (b) Durian Seed Starch Film, (c) Cassava Starch Film

Table 4. Results of Water Solubility Test of Biodegradable Plastic from Starch Variations

Starch Variation	Initial Weight (g)	Final Weight (g)	Water Solubility Test (%)
Purple Sweet Potato	0,867	0,371	57,20
Cassava	0,637	0,248	61,06
Durian Seed	0,525	0,130	75,23

Purple sweet potato starch had the lowest solubility percentage, at 57.20%, indicating that this film was the most stable in water and tended not to dissolve immediately upon immersion. This very high matrix density creates a very dense, compact, and strong structure, requiring water to penetrate the polymer matrix, resulting in a slower dissolution process [16]. Purple sweet potato starch has a relatively higher amylose content, which allows it to form a more compact and robust film structure, thus reducing solubility. Figure 4.3 shows that the film color may fade due to anthocyanin dissolution, but the film mass does not disappear rapidly. The presence of anthocyanin pigments can also form additional interactions with the film matrix, increasing structural stability [9].

Cassava starch had a higher solubility test value than purple sweet potato starch, at 61.06%, but still lower than durian seed starch. Cassava starch has a high amylopectin content, which is more soluble in water. Cassava film structures are less dense than purple sweet potatoes, as they have lower tensile strength test values. Cassava starch contains highly pure starch, which is highly soluble or dispersible in water. Durian cassava starch has the highest solubility, facilitating the formation of strong hydrogen bonds with chitosan, creating a fairly effective barrier against water.

Durian seed starch has the highest solubility, at 75.23%, meaning this film is the least stable and loses the most mass when immersed in water. Durian seed starch has a low amylose content, and non-starch ingredients can interfere with the formation of a strong film structure [5]. Very high solubility indicates the loosest film structure and weakest hydrogen bonds. The film matrix becomes looser, allowing water to more easily penetrate the structure and dissolve it. This is consistent with previous tensile strength test results, which indicated that the film lacked tensile strength because its low density and cohesiveness made it easy for water to penetrate and dissolve its components.

The difference in solubility percentage between the three starches is usually small, but stronger films exhibit slightly better water resistance due to their denser structure. Water solubility testing is important for assessing film stability, as films that dissolve too easily exhibit low stability and decompose rapidly under wet conditions.

Biodegradation Testing

As a natural polymer, starch is a food source readily digested by microbes. Biodegradable plastics based on purple sweet potato starch, cassava starch, and durian seed starch have varying biodegradation test characteristics, influenced by the composition and structure of the base material[23]. Biodegradation testing is conducted by burying the film in the soil and observing weight loss (mass), physical changes (color, shape, brittleness), the appearance of mold or microbial growth, and the time it takes for the film to break or disintegrate [24]. In general, all types of starch show weight loss after biodegradation, and all three are biodegradable due to their high starch content. While all starch-based films will degrade much faster than conventional plastics, the specific rate may vary slightly depending on the starch structure and components.

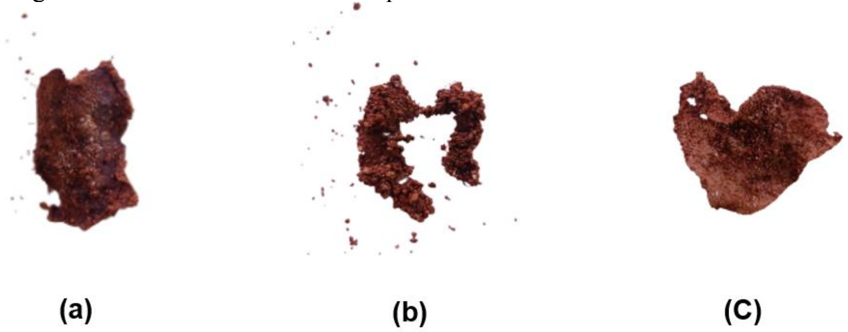

Figure 5. Biodegradable Plastics After Burying in the Soil for 7 Days: (a) Purple Sweet Potato Starch Film, (b) Durian Seed Starch Film, (c) Cassava Starch Film

Table 5. Results of Biodegradation Test of Biodegradable Plastics from Starch Variations

Starch Variation	Initial Weight (g)	Final Weight (g)	Biodegradation (%)
Purple Sweet Potato	1,623	1,192	26,55
Cassava	1,426	1,023	28,26
Durian Seed	0,850	0,493	42,00

Purple sweet potato starch had the lowest degradation percentage at 26.55%, indicating a slower biodegradation rate among the three samples. The lowest degradation rate was associated with the strongest film mechanical properties. The very high matrix density and strong polymer bonds make the film more difficult for amylase enzymes produced by microorganisms to penetrate, thus slowing the degradation process. Cassava starch achieved a biodegradation test result of 28.26%, degrading slightly faster than purple sweet potato.

Cassava starch-based plastic also has good biodegradation characteristics, as cassava starch is readily digested by soil microorganisms [25]. This film degraded by a significant percentage in a relatively short time in the soil burial test, especially if it did not contain many binders or fillers that resist degradation. Well-isolated cassava starch, with minimal impurities, provides a highly pure starch substrate for amylase enzymes. The amylose and amylopectin structures of cassava starch are highly susceptible to attack by microbial enzymes, allowing for rapid degradation in soil.

Durian seed starch had the highest mass loss percentage at 42.00%, indicating the fastest biodegradation rate during the test period. This high degradation rate is consistent with the film's weakest mechanical properties and its high water absorption. Its loose structure and low cohesiveness facilitate water and microbial enzymes' penetration into the matrix and the breakdown of starch chains. The presence of non-starch impurities provides an additional nutrient source attractive to microbes, making the matrix more susceptible to degradation [5].

IV. CONCLUSION

Based on the research results, it can be concluded that the type of starch and the addition of chitosan significantly influence the characteristics of the resulting biodegradable plastic. Purple sweet potato starch-based biodegradable plastic has the highest tensile strength and the best water resistance, but shows a slower biodegradation rate due to its very dense matrix structure. Cassava starch-based plastic shows relatively balanced mechanical properties, solubility, and biodegradation, making it a potential alternative environmentally friendly packaging material. Meanwhile, durian seed starch-based plastic has a less homogeneous film structure, low tensile strength, and high water absorption and solubility, but experiences the fastest biodegradation. The addition of chitosan increases tensile strength but decreases film elasticity. Thus, the selection of starch type and the composition of additives need to be adjusted to the intended use of the biodegradable plastic.

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