

Sustainable biocomposite films from coffee and durian agro-wastes with polyvinyl alcohol matrix

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Abstract. This study investigated the development of a biocomposite packaging material using coffee bean skin waste, polyvinyl alcohol, and the addition of durian seed starch. The inclusion of cellulose fibers from coffee bean skin waste can enhance the properties of polymeric materials, while PVA can increase the homogeneity and flexibility of the composite sheet. Furthermore, the addition of durian seed starch, which has a high starch content, was explored to reduce water absorption and enhance biodegradation of the biocomposite packaging. The objective of this study was to investigate the impact of varying the PVA to coffee bean skin waste ratio, the glycerol percentage, and the addition of durian seed starch on the tensile strength, elongation, modulus of elasticity, water absorption, and biodegradation of the biocomposite packaging. The selected biocomposite packaging with an additional 20% durian seed starch showed a tensile strength of 10.91 ± 0.36 MPa, elongation of $322.17 \pm 17.49\%$, modulus of elasticity of 3.39 ± 0.21 MPa, water absorption of $104.97 \pm 21.32\%$, and biodegradation based on weight loss of $6.61 \pm 0.77\%$ which shows good physical properties. The biocomposite packaging developed in this study demonstrates the potential to utilize waste materials from the food and agriculture industry to create sustainable and biodegradable packaging solutions.

1 INTRODUCTION

Conventional plastics, while offering advantages such as flexibility, light weight, and water resistance in packaging applications, face environmental challenges due to their difficulty in natural decomposition. This issue has caused the development of composite packaging derived from biodegradable biopolymers. Composites, comprising a matrix and fiber, show enhanced mechanical properties compared to individual materials. The term "bio" indicates that these materials are biodegradable and originate from renewable natural resources. Agricultural wastes, including polysaccharides like cellulose and starch, are known for creating biodegradable packaging. This also feeds the growing need for sustainable and biodegradable packaging solutions, particularly those utilizing plant-based materials [1].

Polyvinyl alcohol (PVA) is a synthetic polymer that can increase the homogeneity of a composite sheet. The characteristics of PVA with its chemical resistance, ability to form sheets, and its good biocompatible properties are able to increase the strength and flexibility of packaging. In addition, PVA interactions with starch also tend to be strong because both PVA and starch contain many hydroxyl groups so that they are able to form intermolecular and intramolecular hydrogen bonds. However, the use of high cellulose fibers can reduce PVA rate of solubility. When lower cellulose is added to the PVA matrix, PVA will be more easily exposed to the medium causing faster degradation than composites with higher cell. The addition of high cellulose will also increase absorption because the hydrogen bonds in cellulose molecules tend

to form intramolecular hydrogen bonds[2]. Therefore, increasing the percentage of cellulose in the composite will lead to an increase in degradation properties compared to pure PVA.

Cellulose microfibrils, derived from sources such as Ampel bamboo pulp, have demonstrated potential as reinforcing fillers in polyvinyl alcohol composites, improving their mechanical characteristics [3]. In addition, the integration of natural fibers, such as pineapple leaf fiber, banana fiber, or bamboo fiber, into polypropylene composites can enhance biodegradation, demonstrating partial degradation ranging from 5–15% depending on fiber content [4]. Similarly, agro-extracted cellulose fibers have been shown to significantly improve the mechanical and barrier properties of polylactic acid and polyhydroxybutyrate biocomposites, making them suitable for various biodegradable.

Cellulose is one of the most abundant fillers which is efficient for enhancing the properties of bio-based polymers, has good mechanical and barrier properties, and its hydrophilic characteristics make it compatible with starch. The incorporation of cellulose into polyvinyl alcohol matrices is particularly effective, as PVA, being a water-soluble and biodegradable polymer, benefits from the improved mechanical and barrier properties conferred by cellulose while maintaining its degradability [5] [6]. Despite these advantages, natural biopolymer-based films and their derivatives often exhibit poor mechanical characteristics and hygroscopic behavior, necessitating innovations in bio-composite film development to achieve commercial viability [7]. In this research the cellulose was obtained from coffee bean skin. Coffee bean

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skin is composed of heterogeneous components known as lignocellulosic waste consisting of cellulose, hemicellulose, and lignin [8].

Starch can be completely degraded (100%) in 32 days compared to pure PVA (54%) in 45 days [9]. When PVA was mixed with starch, its water absorption decreased compared to pure PVA due to hydrogen bonds between PVA and starch which could affect the number of free -OH groups. The addition of starch in high quantities will increase the amount of polymer so that the cavities in the gel that are formed are smaller and the matrix will also be thicker and denser so that it can reduce water absorption. This reduction in water absorption is crucial for improving the hydrolytic stability and barrier properties of the biocomposite films, extending their potential applications [10].

Durian seed starch has been investigated as a viable natural polymer for enhancing bioplastic properties due to its high starch content, offering potential improvements in tensile strength, elasticity, and thermal resistance while reducing water absorption of biopolymer [11]. Durian seed starch contains a starch content of $87.82 \pm 0.63\%$ with more higher amylose content ($22.87 \pm 0.33\%$) than cassava starch ($18.18 \pm 0.11\%$) [12]. Durian seed starch also has a smaller granule size ($1.09 \pm 0.15 \mu\text{m}$) than cassava starch ($1.36 \pm 0.17 \mu\text{m}$), where the smaller particle size can increase the rate of starch hydration. This characteristic, coupled with its notable amylose content, positions durian seed starch as a promising filler for biodegradable composites, offering enhanced mechanical integrity and reduced hygroscopicity [13].

However, mixing cellulose with starch can cause the surface of the composite sheet to become less homogeneous resulting in lower mechanical strength. This structural inhomogeneity often arises from challenges in achieving uniform dispersion of highly crystalline cellulose within the starch matrix, leading to stress concentration points that compromise overall material integrity [14]. To mitigate these issues, surface modification of cellulose or the incorporation of compatibilizers can enhance interfacial adhesion and dispersion, leading to more robust and homogeneous biocomposites. The addition of glycerol as a plasticizer can increase the stability of the resulting composites because they are able to form hydrogen bonds with polymers. Therefore, this research was carried out by making biocomposite packaging sheets from PVA-cellulose as a basis to obtain good mechanical characteristics by adding glycerol to reduce friability and increase plasticity and durian seed starch to reduce water absorption and increase biodegradation.

2 MATERIAL AND METHODS

The materials used to make the biocomposite packaging in this study were Palembang local durian seeds from Tom Durian, robusta and arabica coffee bean skin waste (75:25) from PT Cipta Harum Lestari, polyvinyl alcohol (PVA), food grade "Unicarb FCH5000 PT Niraku Jaya Abadi", glycerol, water and distilled water. The materials used in the analysis

were 1 N, 72%, distilled water, organic growing medium "Tanah Subur", 96% ethanol, 80% ethanol, 52% perchloric acid, anthrone reagent, pure amylose, 95% ethanol, NaOH 1N, acetic acid 1N, and iodine solution.

The equipment used to make biocomposite packaging in this study were a knife, cutting board, container, filter cloth, wet and dry blender, centrifuge "HERMLE", 50 ml falcon tube, tray, baking paper, cabinet dryer, 80 mesh sieve, measuring cylinder, herb grinder, beaker glass, volumetric flask, hotplate magnetic stirrer, thermometer, aluminum foil, spatula, stir bar, table scales, analytical scales Ohaus, staves and clamps, film maker, and mica plastic. The equipment used in the analysis were evaporating dish, ovens, desiccators, analytical scales Ohaus, table scales, beakers glass, measuring cylinder, trays and clamps, Whatman No.1 filter paper, erlenmeyer, vacuum pumps, condensers, rulers, scissors and cutters, texture analyzer TA.XTplus, mini tensile grip, tissue, container, cabinet dryer, centrifuge, magnetic stirrer, volumetric flask, test tube, spectrophotometer, waterbath, and volumetric pipette.

2.1 Procedures

2.1.1 Preliminary Research

Preliminary research aims to make durian seed starch and fine coffee bean skin waste as well as raw material analysis. Preliminary research includes the process of refining coffee bean skin waste and extraction of durian seed starch [15]. The analysis was carried out for fine coffee bean skin waste were lignocellulosic test and moisture content [16]. The analysis carried out for durian seed starch included analysis of starch content, amylose content, amylopectin content, and moisture content. The coffee bean skin waste was grinded using a herb grinder and sieved using an 80 mesh sieve.

The durian seed starch was prepared by peeling the durian seed skin and slicing it thinly with a thickness of ± 2 mm, then soaking it in a solution of 10% for 1 hour and cleaned with water, then the durian seeds were soaked again with solution for 1 hour and cleaned again until there was no mucus. The seeds were crushed in a blender with the addition of distilled water (1:2) for 5 minutes. The mixture was filtered with a filter cloth and the filtrate obtained was centrifuged with a centrifuge at 5000 rpm for 10 minutes, the precipitate obtained was then washed with distilled water twice using a centrifuge. The starch precipitate obtained was then dried in a cabinet dryer at 50°C for 6 hours, then pulverized with a dry blender and sifted through an 80 mesh sieve [12]. The resulting finely powdered durian seed starch was then stored in an airtight container for subsequent use in biocomposite formulations.

2.1.2 Research Stage I

Research stage I aims to determine the best ratio of PVA: coffee bean skin waste (100:0, 90:10, 80:20, 70:30, 60:40, 50:50) and the concentration of glycerol

(2% and 4%) and determining the effect on tensile strength, elongation, and modulus young as well as determining of selected treatment. Formulations of biocomposite packaging are shown in Table 1.

Table 1. Formulation of biocomposite packaging

Ingredient	Content
Polyvinyl alcohol solution (mL)	100, 90, 80, 70, 60, 50
Coffee bean skin waste (%)	0, 10, 20, 30, 40, 50
Aquades (mL)	0, 10, 20, 30, 40, 50
Glycerol (%)	2, 4

The manufacture of biocomposite packaging was carried out where 100 g of PVA was weighed, then 1000 ml of distilled water was heated on a hotplate magnetic stirrer to a temperature of 80°C and covered with aluminum foil, then PVA was added little by little and stirred until homogeneous, the PVA solution was left for 24 hours until no bubbles formed[17]. The coffee bean skin waste was weighed according to the treatment, then the volume of the PVA solution was measured using a 100 ml measuring cylinder according to the treatment. The mixture was then put into a 100 ml beaker glass and distilled water was added until the mixture reached 100 ml and glycerol was added according to the treatment, then stirred until homogeneous. Then, it was poured and printed on a film maker based on mica plastic as much as 45 g and dried in a cabinet dryer at 50°C for 24 hours. The biocomposite packaging sheets obtained were analyzed in the form of tensile strength, elongation, and modulus young and then the best formulation was selected.

2.1.3 Research Stage II

This stage focused on optimizing the selected biocomposite formulation from Stage I by incorporating varying concentrations of durian seed starch to evaluate its impact on water absorption and biodegradation characteristics. The effect durian seed starch ratio (0%, 10%, 20%, 30%, 40%) on the tensile strength, elongation, modulus young, water absorption and biodegradation. Research stage II involved the process of making biocomposite packaging sheets by adding durian seed starch and parameter testing. The formulation of the biocomposite packaging can be seen in Table 2.

Table 2. Formulation of biocomposite packaging with the addition of durian seed starch

Ingredient	Content
Polyvinyl alcohol solution (mL)	90
Coffee bean skin waste (%)	10
Aquades (mL)	10
Glycerol (%)	2
Durian seed starch (%)	0, 10, 20, 30, 40

2.1.4 Paramater Analysis

The moisture content procedure used for the analysis of durian seed starch and coffee bean skin waste was the oven drying method by AOAC (2005) with a modification, lignocellulosic test for coffee bean skin waste was carried out using the Chesson-Data method with modifications[18]. Tests for starch and amylose content were carried out at the BB-Postharvest Laboratory, Bogor, West Java, Indonesia, amylopectin content were obtained from calculating the difference between starch and amylose content. Physical properties of the biocomposite were analyze for the following parameter; tensile strength, elongation, and modulus young tests [19], [20], the water absorption test and Biodegradation testing [20],[21] also done following previous study. The biocomposite characterization also involved optimization of mechanical properties, such as tensile strength and elongation, which are crucial for assessing the material's suitability for packaging applications.

2.1.5 Experimental Design

The experimental design of Research Stage I was a Completely Randomized Two-Factor Design with 2 (two) replications with each replication being 2 (two) duplications. The factors on research stage I were the ratio of PVA: coffee bean skin waste and the concentration of glycerol. The experimental design of the Research Stage II was a Completely Randomized One-Factor Design with 3 (three) replications with each replication being 2 (two) duplications. The factor used was the concentration of the addition of durian seed starch. Statistical tests for the research stage I and II were carried out using ANOVA using the SPSS 26th version software.

3. RESULT AND DISCUSSION

The durian seed starch used is in the form of flour and is white in color. The results of the durian seed starch analysis can be seen in Table 3.

Table 3. Results analysis of durian seed starch

Parameter	Result (%)
Starch content*	66±0,54
- Amylose*	28.89±0.01
- Amylopectin	37.11±0.53
Moisture content	11.86±0.07

*Tested in BB-Postharvest Laboratory

The starch content extracted from durian seed was found to be 66.00 ± 0.54%, indicating a relatively high starch yield, which highlights its potential as an alternative starch source. Within this starch fraction, the amylose and amylopectin contents were determined to be 28.89 ± 0.01% and 37.11 ± 0.53%, respectively. This composition suggests a balanced ratio of amylose

to amylopectin, with a slightly higher proportion of amylopectin. Such a profile influences the functional properties of the starch, such as gelatinization, viscosity, and retrogradation behavior, making it suitable for various food and industrial applications. The amylose content measured in this study was observed to be greater, while the amylopectin content was lower [23], [24]. The proportion of amylose in starch contributes to the robustness of the sheet matrix, whereas the amylopectin content enhances the sheet's elastic properties. Additionally, the moisture content was measured at $11.86 \pm 0.07\%$, which falls within the acceptable range for dried starch and ensures its stability during storage. Durian seed starches has been used as a various polymer addition in making biocomposite, therefore it is important to understand its chemical and functional characteristics [12]. The processing of durian seeds into different forms, such as whole durian seed flour, demucilaged durian seed flour, and durian seed starch, significantly influences their chemical and functional properties, whereas the durian variety has minimal impact.

The coffee bean skin waste used is in the form of a fine grind. The results of the analysis of coffee bean skin waste can be seen in Table 4.

Table 4. Results analysis of coffee bean skin waste

Parameter	Result (%)
Hemicellulose content	24.97 ± 0.94
Cellulose content	22.10 ± 0.91
Lignin content	35.56 ± 3.59
Moisture content	10.83 ± 0.24

The analysis of fiber components in the sample showed a significant presence of structural polysaccharides. Lignin was the most abundant, with a content of $35.56 \pm 3.59\%$, followed by hemicellulose at $24.97 \pm 0.94\%$ and cellulose at $22.10 \pm 0.91\%$. The high lignin content may contribute to the rigidity and resistance to biodegradation of the material, which is an important consideration for applications in biocomposites or bioenergy. The moisture content was relatively low at $10.83 \pm 0.24\%$, indicating good storage stability and minimal microbial growth risk during storage. The hemicellulose and lignin content obtained in this coffee bean skin is lower than with 16.68% hemicellulose content, 28.58% lignin content and 23.77% cellulose content [8]. Cellulose, hemicellulose, and lignin are polysaccharides characterized by hydrophilic -OH groups in their structures, which facilitate the formation of hydrogen bonds with water molecules in the air, leading to moisture absorption. In this study, the coffee bean skin waste will act as the matrix of biocomposite film.

There was a significant interaction between the ratio of PVA:coffee bean skin waste and glycerol in affecting the tensile strength of the biocomposite packaging ($p \leq 0.05$). These interactions can be seen in Figure 1.

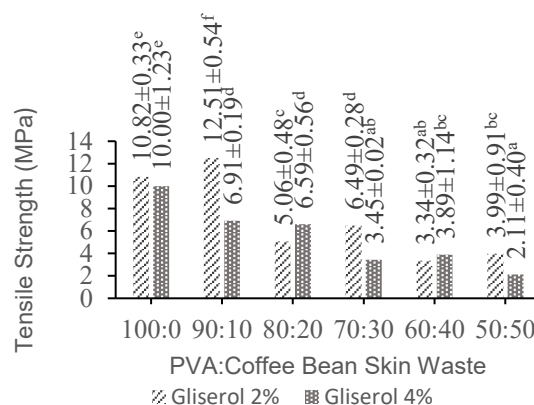


Figure 1. Graph of tensile strength of the biocomposite PVA:coffee bean skin waste with glycerol Note: Differences in superscripts show significant differences ($p \leq 0.05$)

The data clearly show that increasing glycerol concentration from 2% to 4% leads to a decrease in tensile strength across all PVA:coffee bean skin waste formulations. Glycerol acts as a plasticizer, reducing the intermolecular forces between polymer chains and increasing chain mobility. While this enhances flexibility and reduces brittleness, it also compromises mechanical strength. For example, at a 90:10 PVA:coffee waste ratio, tensile strength dropped from 12.03 ± 0.54 MPa (2% glycerol) to 6.99 ± 0.13 MPa (4% glycerol). This trend is consistent at all tested ratios, indicating that higher glycerol levels reduce the film's ability to resist tension.

Increased glycerol concentrations are associated with a reduction in tensile strength, which can be attributed to glycerol's interference with internal hydrogen bonds [26]. This disruption weakens the intermolecular attractions between polymer chains, leading to a decrease in both tensile strength and stiffness. Conversely, a higher polyvinyl alcohol content enhances tensile strength while reducing breakage, as the increased intermolecular hydrogen bonds require more energy to break. The reduction in tensile strength is a result of the replacement of intermolecular bonds between PVA molecules with bonds between PVA and glycerol.

An increase in coffee bean skin waste concentration up to 10% correlates with enhanced tensile strength; however, exceeding this threshold leads to a decline in the tensile strength. The initial enhancement is likely due to the straight, long polymer chains of cellulose, which helps forming a strong adhesion and robust hydrogen bond formation within the composite matrix. Increasing coffee bean waste percentage beyond this limit result in decreased tensile strength due to the semi-crystalline nature of cellulose, comprising both crystalline and amorphous regions [8]. While crystalline cellulose particles act as effective fillers that bond with PVA to create strong composites, the substantial lignin content (an amorphous component) in coffee bean skin waste can cause agglomeration and non-uniform dispersion, thereby negatively affecting tensile strength [27]. This also may be due to high levels of lignin, hemicellulose, and extractives that reduces the

polymer matrix's capacity to create a strong interfacial bond.

There was a significant interaction between the ratio of PVA:coffee bean skin waste and glycerol in affecting the elongation of the biocomposite packaging ($p \leq 0.05$). These interactions can be seen in Figure 2.

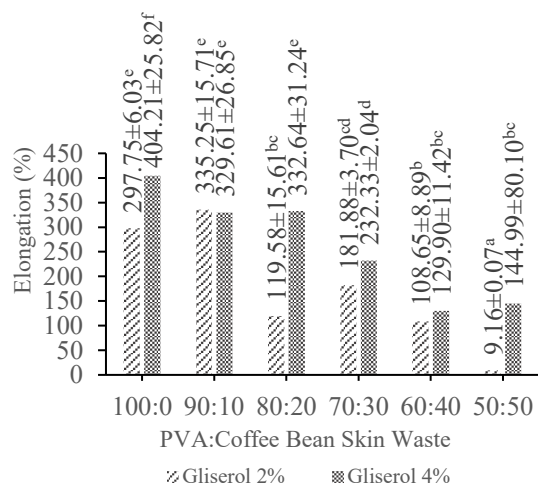


Figure 2. Graph of elongation of the biocomposite PVA:coffee bean skin waste with glycerol
 Note: Differences in superscripts show significant differences ($p \leq 0.05$)

Glycerol significantly influences the elongation at break of PVA-based biocomposite films by acting as a plasticizer. The data show that increasing glycerol concentration from 2% to 4% consistently results in a higher elongation percentage across all PVA:coffee bean skin waste ratios. For instance, at a 100:0 (pure PVA) ratio, elongation increased from $297.75 \pm 6.03\%$ to $402.52 \pm 25.82\%$ when glycerol concentration was increased from 2% to 4%. This trend is also evident at other compositions, such as 90:10 ($333.25 \pm 26.87\%$ to $352.76 \pm 36.71\%$) and 80:20 ($119.58 \pm 1.15\%$ to $352.64 \pm 31.24\%$).

This increase in elongation is attributed to the plasticizing effect of glycerol, which reduces intermolecular hydrogen bonding within the PVA matrix and enhances polymer As a result, films become more pliable and flexible, which allows them to undergo greater deformation before fracturing. However, this enhancement in flexibility frequently leads to a reduction in tensile strength, as demonstrated by the mechanical tests [28]. In conclusion, elevated glycerol concentrations improve film flexibility and elongation. However, the optimization of these concentrations is crucial to achieve a balance between the mechanical strength and flexibility of the biocomposite, especially when incorporating fillers such as coffee bean skin waste. The elongation value is expected to decrease with the incorporation of higher concentrations of coffee bean husk waste. This is because the cellulose macromolecular chains create extensive hydrogen bond networks, resulting in rigid structures, and the elongation value diminishes due to reduced deformation at break which reducing elasticity but increasing strength.[29].

There was a significant interaction between the ratio of PVA:coffee bean skin waste and glycerol in affecting the modulus young of the biocomposite packaging ($p \leq 0.05$). These interactions can be seen in Figure 3.

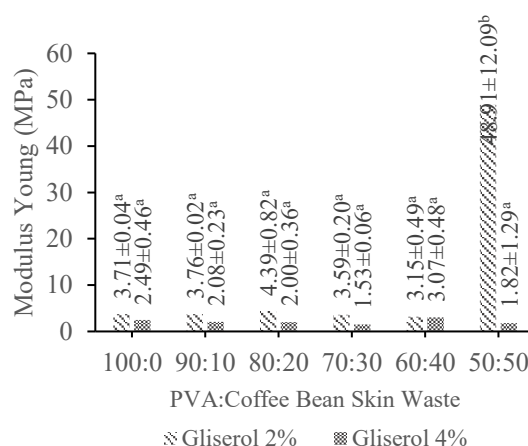


Figure 3. Graph of modulus young of the biocomposite PVA:coffee bean skin waste with glycerol
 Note: Differences in superscripts show significant differences ($p \leq 0.05$)

Glycerol, as a plasticizer, plays a crucial role in modifying the mechanical behavior of PVA-based biocomposite packaging. One key mechanical property affected is Young's modulus, which reflects the stiffness or rigidity of the film. Generally, an increase in glycerol concentration leads to a decrease in Young's modulus, indicating that the films become more flexible and less stiff. Glycerol interferes with the polymer chains, reducing the material's resistance to elastic deformation [30]. Glycerol presence disrupts the inter- and intramolecular hydrogen bonds within cellulose chains, replacing them with weaker polymer-glycerol bonds, which subsequently lowers the film's density and mechanical rigidity [26].

The data show that at lower coffee bean skin waste content (100:0 to 40:60 PVA:coffee ratios), increasing glycerol from 2% to 4% significantly reduced the modulus. For example, at 100:0 (pure PVA), Young's modulus decreased from 3.71 ± 0.04 MPa (2%) to 2.49 ± 0.46 MPa (4%), and at 80:20, from 4.39 ± 0.82 MPa to 2.00 ± 0.36 MPa. This trend occurs because glycerol molecules insert themselves between PVA chains, disrupting intermolecular hydrogen bonding and enhancing chain mobility, thus lowering the film's stiffness. However, a notable exception occurs at the 50:50 ratio, where the modulus for 4% glycerol (48.91 ± 12.09 MPa) is drastically higher than that for 2% glycerol (1.82 ± 1.29 MPa). This anomaly could be due to phase separation, filler aggregation, or interfacial interactions between the coffee skin particles, PVA, and excess glycerol—creating a more brittle, rigid structure at high filler and plasticizer concentrations. Increasing cellulose content enhances the stress at break and stiffness of composites, leading to a significant increase in the elastic modulus. Higher cellulose content, relative to pure PVA, positively reinforces the composite, increasing its breaking stress. However, this increase in cellulose simultaneously reduces the deformation at break, resulting in a more brittle composite due to decreased PVA deformability.

Based on the 12 formulations that have been tested, the selected formulation for biocomposite packaging from research stage I was PVA: Coffee bean skin waste

with a ratio of 90:10 and a concentration of 2% glycerol. This formulation was chosen because in addition to obtaining packaging that is strong and elastic enough, the resulting packaging is still able to utilize waste coffee bean skins and glycerol.

There was a significant effect of the addition of durian seed starch on the tensile strength of the biocomposite packaging ($p \leq 0.05$). This result can be seen in Figure 4. Addition of durian seed starch at varying concentrations has a significant impact on the tensile strength of the biocomposite material. The incorporation of starch into biocomposite films has a notable influence on their tensile strength, which reflects the film's ability to withstand pulling forces without breaking. The results show that adding starch up to a certain concentration (10–20%) slightly increases the tensile strength, indicating improved mechanical reinforcement. In the result the tensile strength improved from 9.73 ± 0.59 MPa (0% starch) to 10.91 ± 0.36 MPa (20% starch).

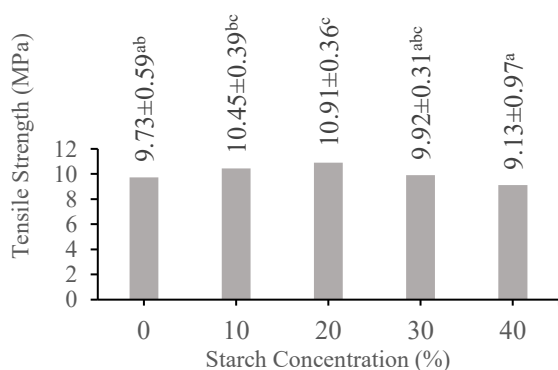


Figure 4. Graph of the tensile strength of the biocomposite with the addition of durian seed starch
 Note: Differences in superscripts show significant differences ($p \leq 0.05$)

Addition of durian seed starch at varying concentrations has a significant impact on the tensile strength of the biocomposite material. The incorporation of starch into biocomposite films has a notable influence on their tensile strength, which reflects the film's ability to withstand pulling forces without breaking. The results show that adding starch up to a certain concentration (10–20%) slightly increases the tensile strength, indicating improved mechanical reinforcement. For example, the tensile strength improved from 9.73 ± 0.59 MPa (0% starch) to 10.91 ± 0.36 MPa (20% starch). This enhancement is likely due to better hydrogen bonding between starch and the polymer matrix, which helps distribute stress more effectively during tension.

The observed increase in tensile strength can be attributed to the chemical similarities between starch and cellulose, which facilitate strong intermolecular interactions and enhance adhesion through hydrogen bonding. These hydrogen bonds, formed between hydroxyl groups from starch and both hydroxyl and carboxyl groups from cellulose, contribute to the enhanced strength of the composite. Furthermore, the incorporation of durian seed starch leads to an increase in thickness due to the higher concentration of

dissolved solids, resulting in tighter cross-linking within the starch polymer matrix. This increased cross-linking necessitates greater force to induce sheet breakage. However, at higher starch concentrations (30–40%), tensile strength begins to decline, with the lowest strength observed at 40% starch (9.13 ± 0.97 MPa). This reduction may be attributed to poor dispersion, particle agglomeration, or weak interfacial adhesion, which can create stress concentration points and compromise the structural integrity of the biofilm [30], [31]. Although statistical variation among the samples is relatively small, the data suggest that optimal starch content ranges between 10–20% for maintaining or improving tensile strength in the biocomposite.

There was a significant effect of the addition of durian seed starch on the elongation of the biocomposite packaging ($p \leq 0.05$). This result can be seen in Figure 5.

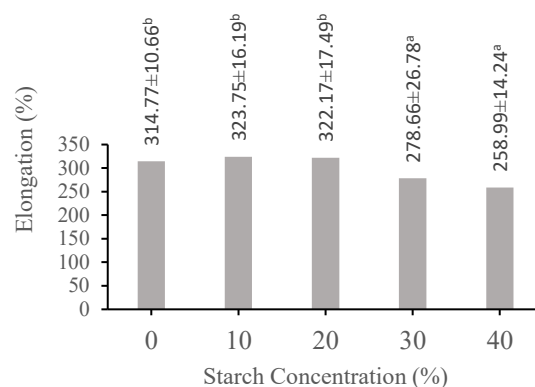


Figure 5. Graph of elongation of the biocomposite with the addition of durian seed starch

Note: Differences in superscripts show significant differences ($p \leq 0.05$)

The results showed that adding durian seed starch to a concentration of 10% could increase the percent elongation of the biocomposite packaging but will decrease the percent elongation after crossing the concentration limit. Despite a decrease in the percent elongation value when durian seed starch concentration was increased to 20%, this reduction was not statistically significant compared to a 10% concentration. This outcome is attributed to the effective hydrogen bonding between the hydroxyl groups of starch and the hydroxyl and carboxyl groups of cellulose, which enhances the flexibility of the composite sheet, thus promoting increased elongation. However, at much higher concentrations, the sheet structure becomes denser, increasing its propensity for breakage or damage. Consequently, an excess of starch and cellulose can lead to phase separation and poor particle dispersion, limiting the synergistic interaction between cellulose and starch. This phenomenon, where excessive filler content hinders rather than helps, is consistent with observations in other biocomposites, where non-homogeneous dispersion and agglomeration of fillers, lead to reduced tensile strength and elongation at break.

There was a significant effect of the addition of durian seed starch on the biodegradation of the biocomposite packaging ($p \leq 0.05$). This result can be seen in Figure 6.

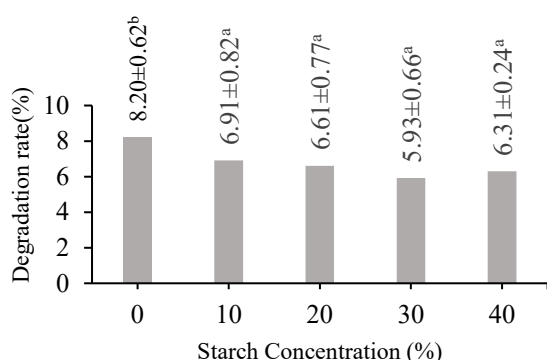


Figure 6 Graph of the biodegradation of the biocomposite with the addition of durian seed starch
 Note: Differences in superscripts show significant differences ($p \leq 0.05$)

The results showed that the higher the addition of durian seed starch, the lower the percentage of weight loss. Research by Jannah et al. indicated that durian seed starch films, when supplemented with glycerol, could degrade by up to 38.9% within 5 days and achieve complete degradation in soil by the 7th day. This accelerated decomposition is attributed to the hydroxyl groups in starch, which facilitate hydrolysis upon absorbing soil moisture. This observation aligns with a separate study by Cano et al., demonstrating that starch can fully degrade within 32 days, whereas pure PVA degrades only 54% over 45 days. Notably, PVA-starch mixtures exhibit a higher biodegradation rate than pure PVA alone [9]. Consequently, the quantity of polyvinyl alcohol added and the duration of the biodegradation test are crucial factors influencing the degradation of the resulting packaging. However, the rate of degradation can also be influenced by the specific chemical structure and molecular size of the starch, with certain types dramatically increasing the rate of dissolution [32].

The addition of durian seed starch to PVA biocomposites containing coffee bean skin waste led to a reduction in biodegradation. This observation aligns with research by Russo et al., which suggests that the slow degradation of PVA, a synthetic polymer, can impede starch degradation. High concentrations of PVA significantly decrease starch solubility, as PVA encapsulates starch particles dispersed within its continuous phase. Furthermore, the intermolecular mixing of the three components and potential phase separation among them can contribute to a reduced rate of starch hydrolysis in the biocomposite mixture. Conversely, the higher biodegradation observed in treatments without added durian seed starch can be attributed to the lignocellulosic components of coffee bean skin waste. These components possess substantial amorphous regions with less organized structures, making them more susceptible to enzymatic attack by

microorganisms and consequently more readily decomposed, as amorphous polymers degrade more easily than crystalline polymers. Other research has tried to incorporate certain nanofillers, such as silver nanoparticles or graphene oxide, into starch-based composites, to significantly reduce biodegradation rates and extend material longevity in various environmental conditions [33].

4. Conclusion

Biocomposite packaging can be formulated using a blend of polyvinyl alcohol, coffee bean skin waste, durian seed starch, and glycerol. This study identified an optimal formulation comprising a specific ratio of PVA to coffee bean skin waste, supplemented with 2% glycerol and 20% durian seed starch. Elevated concentrations of coffee bean skin waste were observed to enhance tensile strength and modulus of elasticity up to a certain threshold, while concurrently diminishing percent elongation. Conversely, increased glycerol concentrations led to a reduction in both tensile strength and modulus of elasticity, but resulted in an elevated percent elongation. Augmenting the concentration of durian seed starch enhanced both tensile strength and percent elongation up to a specific limit. However, it also resulted in a decreased biodegradation rate compared to biocomposites without durian seed starch; this outcome might be influenced by factors such as the duration of biodegradation testing and the absence of soil immersion. The addition of durian seed starch did not exert a statistically significant influence on the modulus of elasticity or water absorption properties.

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References

- 1.H. Zhang and S. S. Sablani, *Current Opinion in Food Science* **42**, 61 (2021). <https://doi.org/10.1016/j.cofs.2021.05.003>.
- 2.M. T. Taghizadeh and N. Sabouri, *International Nano Letters*, **3**, (2013). <https://doi.org/10.1186/2228-5326-3-51>.
- 3.K. W. Prasetyo, Fakhruzy, and W. B. Kusumaningrum, *IOP Conference Series Earth and Environmental Science* **572**, 12042 (2020). <https://doi.org/10.1088/1755-1315/572/1/012042>.
- 4.S. K. Chattopadhyay, S. Singh, N. Pramanik, U. K. Niyogi, R. K. Khandal, R. V. S. Uppaluri, and A. K. Ghoshal, *Journal of Applied Polymer Science* **121**, 2226 (2011). <https://doi.org/10.1002/app.33828>.
- 5.V. Chanthavong, M. Prabhakar, D. W. Lee, and J. Song, *Journal of Inorganic and Organometallic Polymers and Materials* **34**, 1861 (2023). <https://doi.org/10.1007/s10904-023-02928-x>.

- 6.A. S. Elgharbawy, A.-G. M. E. Demerdash, W. A. Sadik, M. A. Kasaby, A. H. Lotfy, and A. I. Osman, *Polymers* **16**, 1356 (2024). <https://doi.org/10.3390/polym16101356>.
- 7.M. Amini, M. Rasouli, M. Ghoranneviss, M. Momeni, and K. Ostrikov, *Scientific Reports* **12**, (2022). <https://doi.org/10.1038/s41598-022-23284-9>.
- 8.E. Melyna and A. P. Afridana, *EKUILIBIUM* **7**, 14 (2023). <https://doi.org/10.20961/equilibrium.v7i1.68556>.
- 9.A. Cano, M. Cháfer, A. Chiralt, and C. González-Martínez, *Polymer Degradation and Stability* **132**, 11 (2016). <https://doi.org/10.1016/j.polymdegradstab.2016.04.014>.
- 10.S. Jayarathna, M. Andersson, and R. Andersson, *Polymers* **14**, 4557 (2022). <https://doi.org/10.3390/polym14214557>.
- 11.R. Riyanto, S. Hasibuan, D. A. Tanjung, Z. Noer, and V. Sisca, *RASAYAN Journal of Chemistry* **15**, 2280 (2022). <https://doi.org/10.31788/rjc.2022.1547064>.
- 12.S. Baraheng and T. Karrila, *Food Bioscience* **30**, 100412 (2019). <https://doi.org/10.1016/j.fbio.2019.100412>.
- 13.W. G. Abera, R. Kasirajan, and S. L. Majamo, *Biomass Conversion and Biorefinery* **14**, 20419 (2023). <https://doi.org/10.1007/s13399-023-04207-8>.
- 14.E. S. Madivoli, P. G. Kareru, J. Gichuki, and M. M. Elbagoury, *Scientific Reports* **12**, (2022). <https://doi.org/10.1038/s41598-022-23305-7>.
- 15.Z. L. Chew, Y. L. Kua, S. Gan, K. W. Tan, and T. Z. E. Lee, *Waste and Biomass Valorization* **15**, 2299 (2023). <https://doi.org/10.1007/s12649-023-02294-2>.
- 16.S. S. Arya, R. Venkatram, P. R. More, and P. V. P., *Journal of Food Science and Technology* **59**, 429 (2021). <https://doi.org/10.1007/s13197-021-05032-5>.
- 17.M. M. Rahman, S. Sultana, M. Z. Islam, Md. K. U. Sarker, Md. E. Halim, and Md. A. A. Shaikh, *Results in Materials* **23**, 100601 (2024). <https://doi.org/10.1016/j.rinma.2024.100601>.
- 18.S. Nielsen, *Food Analysis*, (4th ed. Corr. 3rd printing, 2014)
- 19.T. Jamal, E. S. Zainudin, S. M. Sapuan, R. A. Ilyas, and K. Abdan, *Polymers* **14**, 388 (2022). <https://doi.org/10.3390/polym14030388>.
- 20.Z. H. Kamaruddin, R. Jumaidin, R. A. Ilyas, M. H. Selamat, R. H. Alamjuri, and F. A. B. M. Yusof, *Polymers* **14**, 514 (2022). <https://doi.org/10.3390/polym14030514>.
- 21.M. Hasan, D. A. Gopakumar, N. G. Olaiya, F. Zarlaida, A. Alfian, C. Aprinasari, T. Alfatah, S. Rizal, and H. P. S. A. Khalil, *International Journal of Biological Macromolecules* **156**, 896 (2020). <https://doi.org/10.1016/j.ijbiomac.2020.04.039>
- 22.C. Stanciu and C. Teacă, *BioResources* **19**, 5895 (2024). <https://doi.org/10.15376/biores.19.3.5895-5915>.
- 23.P. Leemud, S. Karrila, T. Kaewmanee, and T. Karrila, *Journal of Food Measurement & Characterization* **14**, 388 (2019). <https://doi.org/10.1007/s11694-019-00301-6>.
- 24.A. K. Rashwan, H. A. Younis, A. M. Abdelshafy, A. I. Osman, M. R. Eletmany, M. A. Hafouda, and W. Chen, *Environmental Chemistry Letters* **22**, 2483 (2024). <https://doi.org/10.1007/s10311-024-01753-z>.
- 25.M. D. Teli and A. Jadhav, *International Journal of Science and Research (IJSR)* **6**, 1370 (2017). <https://doi.org/10.21275/art20164365>.
- 26.R. Bidari, A. A. Abdillah, R. A. B. Ponce, and A. L. Charles, *Polymers* **15**, 338 (2023). <https://doi.org/10.3390/polym15020338>.
- 27.F. Sarasini, F. Luzi, F. Dominici, G. Maffei, A. Iannone, A. Zuorro, R. Lavecchia, L. Torre, A. Carbonell-Verdu, R. Balart, and D. Puglia, *Polymers* **10**, 1256 (2018). <https://doi.org/10.3390/polym10111256>.
- 28.E. Karaoğul, E. Altuntaş, T. Salan, and M. H. Alma, in *IntechOpen eBooks* (IntechOpen, 2019). <https://doi.org/10.5772/intechopen.81727>.
- 29.T. Jamal, S. M. Sapuan, and K. Abdan, *Scientific Reports* **11**, (2021). <https://doi.org/10.1038/s41598-021-93094-y>.
- 30.X. Zhai, W. Wang, H. Zhang, Y. Dai, H. Dong, and H. Hou, *Carbohydrate Polymers* **239**, 116231 (2020). <https://doi.org/10.1016/j.carbpol.2020.116231>.
- 31.M. P. Guarás, M. Menossi, A. T. Nicolini, V. A. Álvarez, and L. N. Ludueña, *Journal of Materials Science* **58**, 5456 (2023). <https://doi.org/10.1007/s10853-023-08354-1>.
- 32.S. Phattarateera, X. Li, C. Amphong, V. Limsamran, and P. Threepopnatkul, *Carbohydrate Polymer Technologies and Applications* **6**, 100340 (2023). <https://doi.org/10.1016/j.carpta.2023.100340>.
- 33.A. Gamage, P. Thiviya, S. Mani, P. G. Ponnusamy, A. Manamperi, P. Evon, O. Merah, and T. Madhujith, *Polymers* **14**, 4578 (2022). <https://doi.org/10.3390/polym14214578>.