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Development of Sustainable Bioplastic Composite Films from Cocoa Pod Husk Waste Cellulose and Kappa-Carrageenan

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Abstract

Cocoa pod husk (CPH), typically considered agricultural waste, contains cellulose suitable for bioplastic production, offering a sustainable alternative to synthetic plastics. Its reinforcement with kappa-carrageenan is designed to improve the properties of cellulose-based bioplastics while reducing agricultural waste. This study evaluates the effects of cellulose from CPH waste and kappa-carrageenan formulations on bioplastic properties. The cellulose was isolated through a delignification and bleaching process, while the bioplastics were prepared by varying the ratios of cellulose and kappa-carrageenan in six different formulations. The resulting films were evaluated for their physical, mechanical, and barrier properties, as well as their stability and biodegradability. The ratio of cellulose to kappa-carrageenan significantly impacts the films' properties. Significant improvements in tensile strength were observed in P5 (2 g cellulose, 8 g kappa-carrageenan) and P6 (10 g kappa-carrageenan), increasing by 79% and 240%, respectively, as the cellulose concentration decreased and kappa-carrageenan increased. However, the significant drawback in barrier properties was found in water vapor transmission rate (WVTR), with the higher kappa-carrageenan and lower cellulose concentrations films resulting in increased WVTR values by 13% (P5) and 17% (P6). The bioplastic with P1 (8 g cellulose, 2 g carrageenan), P2 (6 g cellulose, 4 g carrageenan), P3 (5 g cellulose, 5 g carrageenan), and P4 (4 g cellulose, 6 g carrageenan) formulations completely degraded in 3 weeks, while those with higher kappa-carrageenan content degraded faster, with P5 completely degrading in 2 weeks and P6 in 1 week. This study implies a potential reduction in environmental impact by replacing conventional plastics with the development of biodegradable materials derived from agricultural waste and promoting sustainable agricultural practices by utilizing CPH.

Keywords: biodegradability; food packaging; mechanical properties

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INTRODUCTION

Plastic is one of the most extensively utilized packaging materials due to its flexibility, strength, transparency, shatter resistance, and cost-effectiveness. However, most plastics are derived from non-renewable petroleum and natural gas resources, leading to a significant environmental concern. Common plastics, including high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyethylene terephthalate (PET), polyvinyl chloride (PVC), polystyrene

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(PS), polypropylene (PP), and polycarbonate (PC), take 100 to 500 years to decompose naturally, posing a long-term waste problem (Dey et al., 2023; Kumari et al., 2023). With global plastic consumption increasing over 160 times in the last 70 years and production reaching 400 million tons in 2022, the volume of plastic waste continues to rise, further intensifying environmental pollution (Pilapitiya and Ratnayake, 2024). These synthetic plastics contribute to the carbon footprint and environmental degradation, as they are non-degradable and difficult to recycle (Chawla et al., 2021). It is estimated that the recycling and processing of plastic waste results in global losses of 80 to 120 billion USD annually (Suleman et al., 2022). In response, government initiatives focus on limiting single-use plastics and encouraging sustainable practices. The development of bioplastics from renewable materials is emerging as a viable and sustainable alternative to reduce dependence on petroleum-based plastics (Singh et al., 2022).

Bioplastics, derived from renewable biomass, provide an environmentally friendly alternative to conventional plastics due to their biodegradability by soil microorganisms. Ideal materials for bioplastic production are renewable, environmentally safe, biodegradable, abundantly available, and relatively inexpensive (Pooja et al., 2023). These materials should also possess properties similar to conventional plastics, such as strength, flexibility, and water resistance. Cellulose is a promising candidate for bioplastic production as it can enhance both the hydrophobicity and rigidity of bioplastics (Asim et al., 2021). As one of the most abundant polysaccharides in nature, cellulose is derived from trees and agricultural plant waste (Gabriel et al., 2020; Petkoska et al., 2021) and offers several advantages, including its non-toxic nature, high durability, and mechanical strength (Shahidi and Hossain, 2022). According to El Halal et al. (2015), cellulose has significant potential as a future bioplastic material due to its low cost and natural degradability, making it a promising solution to environmental issues related to plastic waste. Agricultural biomass waste, such as cocoa pod husks (CPHs), is particularly rich in cellulose and presents a valuable raw material for bioplastic production. Incorporating valuable agricultural waste sources into biodegradable packaging material (Utami et al., 2021) aligns with sustainable agriculture practices, supporting rural economies and resource efficiency.

Cocoa (*Theobroma cacao* L.) is one of the significant global crops. Indonesia was the world's third-largest cocoa producer in 2021, contributing 728,046 tonnes. While global cocoa production has steadily increased, Indonesia's peak production of 844,626 tonnes occurred in 2010 (FAO, 2024). Despite the high production, the primary utilization of cocoa focuses on the beans for chocolate products, leaving the husks underutilized. CPH constitute approximately 75% of the cocoa fruit, yet they are often discarded or minimally utilized. Traditionally, they are used as animal feed (Carta et al., 2020) and organic fertilizers (Iremiren and Ipinmoroti, 2014), yet these uses are not optimized, as most husks are merely collected or discarded around cocoa plantations, potentially causing foul odors (Ouattara et al., 2020). With a cellulose content of 35.4%, hemicellulose at 37%, and lignin at 14.7% (Daud et al., 2013), CPHs are a valuable resource for bioplastic production. The high cellulose content can increase the tensile strength of bioplastics due to the long and straight polymer chains. However, this may inversely affect the elongation percentage, making the bioplastics less elastic and compact (Abe et al., 2021). The addition of kappa-carrageenan is proposed to enhance the mechanical properties of bioplastics.

Kappa-carrageenan, extracted from *Eucheuma cottonii*, a type of red algae, acts as a stabilizer and gel-forming agent, providing strong gel properties. It can form a robust polymer matrix, enhancing the tensile strength and compactness of the film (Dogaru et al., 2020; Bhatia et al., 2024). Bhat et al. (2020) reported that kappa-carrageenan exhibits better gelling properties than other fractions, such as iota and lambda. However, the higher kappa-carrageenan concentration decreases elasticity, making the bioplastic more rigid and limiting its usage (Tasende and Manríquez-Hernández, 2016). Therefore, combining cellulose and kappa-carrageenan not only enhances the mechanical properties but also supports sustainable agriculture initiatives by converting agricultural by-products into value-added products, which can contribute to economic sustainability in developing regions (Nugroho, 2024).

Previous studies have investigated bioplastics made from cellulose or kappa-carrageenan combined with other materials. The study by Maulida et al. (2020) examined the impact of incorporating CPH as fillers for bioplastics on mechanical properties, specifically focusing on tensile strength and elongation at break. This

research sheds light on how the addition of CPH influences the performance of bioplastics in terms of their ability to withstand tensile forces and elongation before breaking. Hasan et al. (2021) explored the reinforcement of cellulose isolated from straw waste in chitosan-based biodegradable films. They found that a formulation of 1% cellulose, 84% chitosan, and 15% castor oil significantly increased tensile strength by 400% compared to samples without cellulose. Yahaya et al. (2023) examined the combination of kappa-carrageenan and cellulose nanofiber. They found that increasing cellulose content enhanced both tensile strength and elongation of the bioplastic while concurrently reducing its elasticity and water vapor permeability.

Recently, Saputri et al. (2024) varied the cellulose from the banana bunch to cassava starch-based films and significantly improved the tensile strength and lowered the water vapor absorption. Those studies were limited in focusing on either cellulose or kappa-carrageenan mixed with various other biopolymers rather than examining the synergistic benefits of combining cellulose, specifically from CPH with kappa-carrageenan. Also, no studies have explored the formulation of cellulose derived from CPH in combination with kappa-carrageenan for producing bioplastics with optimal characteristics. The integration of these findings underscores the necessity for further research into the specific bioplastic formulation of cellulose from CPH and kappa-carrageenan to maximize the mechanical properties and environmental benefits of bioplastics. Therefore, this study aims to evaluate the effects of cellulose from CPH waste and kappa-carrageenan formulations on bioplastic properties.

MATERIALS AND METHOD

Materials and equipment

CPH waste (moisture content 71.28%wb, cellulose yield 21.29%wb), which consists of a mix of Criollo and Forastero types, was collected from local cocoa farmers in Sukoharjo I Village, Pringsewu Regency, Province of Lampung, Indonesia. It was then packaged in a plastic sack before being transferred to the laboratory for further processing. Other materials used include a food-grade kappa-carrageenan (Indogel SGP-168M, 60 mesh), which was purchased from an Indonesian seaweed manufacturer, acetic acid 1%, glycerol 5% w/v (OneMed), NaOH, H_2O_2, distilled water, and silica gel. Equipment used in this study includes an MTS Landmark Servohydraulic Test System (model Landmark 370.10 100 kN, United States), a digital micrometer (RoHS, China), a digital scale (JOIL, Indonesia), a glass plate 20 cm x 20 cm, an electrical powder grinder, a 250-mesh nylon filter, petri dish, and other chemical glassware.

Methods and data analysis

CPH flour preparation was carried out at Laboratory of Agricultural Product Processing and the cellulose isolation at Laboratory of Analysis of Agricultural Products (Faculty of Agriculture, Universitas Lampung). The analysis of fourier transform infra-red (FTIR) was carried out at Integrated Laboratory and Innovation Technology Center (Faculty of Mathematics and Natural Science). Several analyses for the swelling test, water vapor transmission rate (WVTR), and storage stability were conducted at Laboratory of Analysis of Agricultural Products (Faculty of Agriculture, Universitas Lampung), while the measurement of mechanical properties including thickness, tensile strength, elongation at break, and young's modulus at Material Laboratory (Faculty of Engineering, Universitas Lampung).

This study employed a randomized complete block design (RCBD) in four replications with kappa-carrageenan and cellulose levels from CPH formulation. The formulations are P1 (8 g cellulose, 2 g carrageenan), P2 (6 g cellulose, 4 g carrageenan), P3 (5 g cellulose, 5 g carrageenan), P4 (4 g cellulose, 6 g carrageenan), P5 (2 g cellulose, 8 g carrageenan), and P6 (10 g carrageenan). Using a constant total of 10 g of film-forming materials, CPH's cellulose and kappa-carrageenan combination was used to observe each other impact on the bioplastic properties. Also, this helped to identify the optimal balance for desired performance in sustainable packaging applications. Observed parameters in this study were FTIR, thickness, swelling test, tensile strength, elongation at break, young's modulus, storage stability, and biodegradation test. The resulting data were statistically analyzed using one-way ANOVA and the Duncan post-hoc test at 5%, assisted by IBM SPSS version 25 software. The FTIR, stability test, and biodegradation test data were analyzed and discussed descriptively.

Cellulose isolation

The cellulose isolation of CPH waste was initially started by preparing CPH powder, which

was prepared based on Sena et al. (2021) with modification. CPH were washed, cut into approximately 5 cm, and dried at 65 °C in an oven for 24 hours. The dried husks were ground for 5 minutes and sieved using an 80-mesh sieve. The remaining residue was reground until a fine powder was obtained. Cellulose isolation was carried out through the delignification and bleaching process. The delignification process started by immersing the CPH powder in 12% (w/v) NaOH solution for 2 hours at room temperature. The mixture was filtered using a 250-mesh nylon sieve, and the residue was washed until the filtrate was clear. The obtained cellulose was then bleached with 10% (v/v) H_2O_2 solution for 2 hours at 100 °C. After filtration and washing, the cellulose was dried at room temperature for 24 hours, followed by oven drying at 65 °C for 6 hours. The dried cellulose was then ground into a fine powder.

Preparation of bioplastic composite films

The bioplastics were prepared based on the modified method of Rendón-Villalobos et al. (2022). Cellulose with varying masses was placed in beakers. Subsequently, 45 ml of 1% acetic acid was added and heated for 30 minutes at 70 °C to hydrolyze the cellulose. Afterward, 100 ml of distilled water, 5 ml of glycerol, and varying masses of kappa-carrageenan were added (Table 1). The mixture was reheated for 30 minutes at 70 °C with constant stirring using a digital magnetic stirrer (Cimarec Thermo Fisher, US). The mixture was then poured onto a 20 cm x 20 cm glass plate and dried at room temperature for 48 hours.

FTIR analysis

The analysis of FTIR was carried out following Dmitrenko et al. (2023). Potassium bromide (KBr) pellets were prepared before analysis. The FTIR portable instrument (Agilent Technologies Cary 630, US) was set to a resolution of 2 cm^{-1} with a wavenumber range of 650 to 4,000 cm^{-1}. Bioplastic samples were cut into 1 cm x 1 cm pieces and placed in the sample holder. Infrared scans were performed 32 times to obtain more accurate spectra. The resulting infrared spectra were then analyzed to identify the chemical structure and changes in the functional groups of the samples.

Thickness and swelling test

Bioplastic thickness and swelling test conducted following ASTM D6988 (ASTM, 2021) and ASTM D570 (ASTM, 2022), respectively. The thickness was measured using a digital micrometer with a precision of 1 μm at 5 different points on each sample. The final thickness value was determined by averaging the measurements from the 5 points. A swelling test was employed to measure the samples' water absorption ability. Bioplastic samples (2 cm x 2 cm) were weighed to obtain the initial weight (W0). The samples were then immersed in 20 ml of distilled water at room temperature for 10 minutes. After immersion, they were drained for 5 minutes, and the surface was wiped with a tissue. The samples were then reweighed to obtain the final weight (W1). The swelling value (%) was calculated by dividing the weight change (ΔW) by the initial weight and multiplying it by 100%.

Mechanical properties

The observed mechanical properties of bioplastic samples include tensile strength, elongation at break, and young's modulus. The test followed the ASTM D882-02 standard (ASTM, 2010), using an MTS Landmark 370.10 100 kN universal testing machine. Bioplastic samples were cut into 19 mm x 120 mm sizes. Testing was performed at room temperature (25 °C) with a load cell of 100 kN, a crosshead speed of 0.010 kN $minute^{-1}$, and an initial grip separation of 20 mm.

The tensile strength value (MPa) was calculated by dividing the maximum strength values when the samples break (N) with the surface area of samples (mm^2). Elongation at break was calculated at the point when the samples break. The initial length (L0) was

Table 1. Formulation of bioplastic composite films

Formulation	CPH's cellulose (g)	Kappa-carrageenan (g)	Acetic acid 1% (ml)	Distilled water (ml)	Glycerol (ml)
P1	8	2	45	100	5
P2	6	4	45	100	5
P3	5	5	45	100	5
P4	4	6	45	100	5
P5	2	8	45	100	5
P6	0	10	45	100	5

measured before testing up to the grip, while the final length (L1) was measured after the sample broke. The elongation at break was calculated by dividing the length change (ΔL) by the initial length of the sample and multiplied by 100%. Young's modulus value (MPa), an indicator of material stiffness, was calculated from the tensile strength ratio to elongation at break value.

WVTR

The WVTR was measured using the gravimetric cup method according to ASTM E96 (ASTM, 2017). Bioplastic samples were cut into circles with a diameter matching the test cup. Silica gel (10 g) was placed in the cup and sealed with the bioplastic sample. The sample cup was then placed in a jar containing 40% (b/v) NaCl solution and conditioned at room temperature for 24 hours. The initial and final weights of the sample were recorded to calculate the WVTR (g m^{-2} day^{-1}).

Stability test

The stability test was conducted following Fiqinanti et al. (2022) to visually observe physical changes in the bioplastics. Bioplastic samples were cut into 3 cm x 3 cm from each treatment and placed on a glass plate at room temperature (25 °C). Visual observations were made weekly for 5 weeks, focusing on changes in sample integrity, color, and surface condition. The results of the observations were then descriptively described in detail.

Biodegradability test

The biodegradability test was conducted using the soil burial test method, according to Hidayati et al. (2019). Ultisol soil was collected from the surface at 25 and 50 cm depth from the Integrated Field Laboratory, Faculty of Agriculture, Universitas Lampung. Bioplastic samples were prepared (3 cm x 3 cm), then buried at a depth of 8 cm in pots filled with soil and incubated at room temperature. The soil was watered weekly. The initial weight of the bioplastic sample was measured. Visual observations and weight loss measurements were made periodically (every 1 week) to monitor the level of bioplastic degradation until the sample was completely degraded. The weight loss percentage reflects the bioplastic biodegradability and was calculated by comparing the weight difference at each observation period to the initial weight. This test was carried out without replication.

RESULTS AND DISCUSSION

FTIR analysis

The FTIR spectrum of bioplastic films with varying cellulose (isolated from CPH waste) and kappa-carrageenan ratios is presented in Figure 1. The O-H stretching peak detected at 3,265; 3,272; and 3,295 cm^{-1} is related to the polysaccharides hydroxyl group and the hydrogen bonding within the bioplastic films, which is also associated with water absorption (Dmitrenko et al., 2023). In P1, with the highest cellulose content, this peak is broad and intense (49.34% transmittance at 3,272 cm^{-1}), indicating extensive hydrogen bonding among the hydroxyl groups of cellulose. As carrageenan content increases across the formulations from P2 to P6, the intensity of this peak progressively decreases. For instance, in P6, which contains the highest carrageenan content, the O-H peak shows a lower intensity (43.51% transmittance at 3,265 cm^{-1}). This reduction indicates fewer hydrogen bonds, suggesting that carrageenan, with fewer hydroxyl groups than cellulose, forms less extensive hydrogen bonding networks. The gradual decrease in the O-H peak intensity highlights the diminishing role of cellulose's hydrogen bonding as carrageenan content increases.

The peak detected at 1,640 and 1,647 cm^{-1} in all samples indicates carbonyl group and O-H vibrations of absorbed water (Abdullah et al., 2021; Santos et al., 2023). In P1, this peak appears at 1,647 cm^{-1} with a moderate transmittance of 75.25%, signifying the presence of glycerol used as a plasticizer. As the carrageenan content rises, the intensity of this peak increases, reflecting enhanced interactions between glycerol and carrageenan. For instance, in P6, the C=O peak at 1,640 cm^{-1} has a transmittance of 69.72%, indicating stronger interactions between glycerol and carrageenan. This trend suggests that carrageenan interacts more robustly with glycerol than cellulose, enhancing the flexibility of the films by forming a more integrated matrix with the plasticizer.

The C-O-C and C-O stretching peaks detected at 1,028; 1,244; 1,252; and 1,267 cm^{-1} represent the polysaccharide backbone structures of cellulose and carrageenan (Gulzar et al., 2022). In P1, the C-O-C stretching peak at 1,252 cm^{-1} and the C-O stretching peak at 1,028 cm^{-1}, with transmittances of 71.23% and 31.16%, respectively, reflect the strong presence of cellulose's polysaccharide structure.

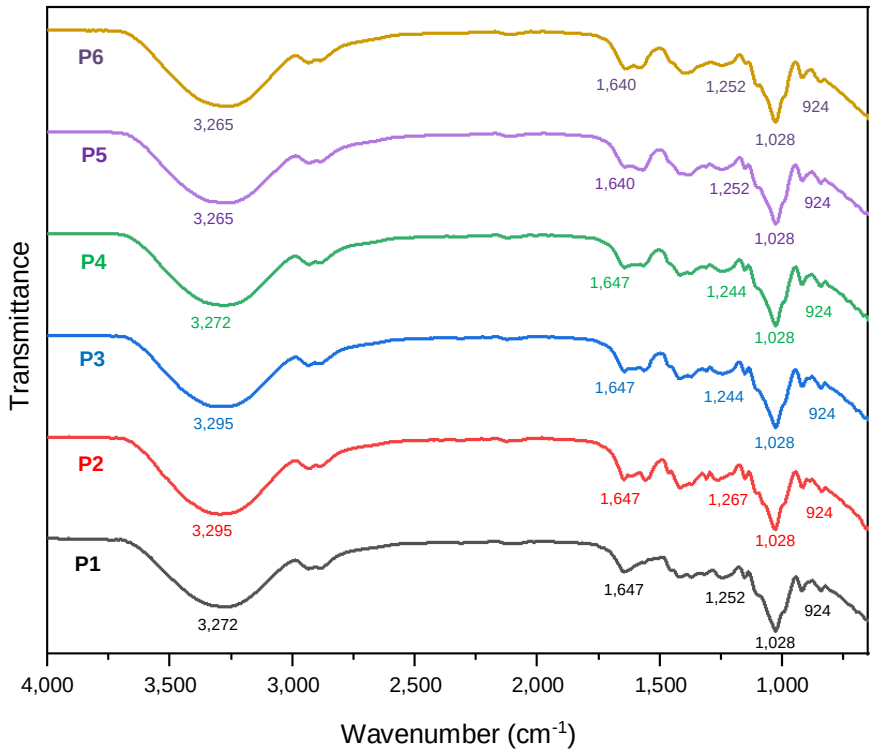

Figure 1. FTIR spectra of bioplastic

As carrageenan content increases in subsequent formulations, these peaks show significant contributions from both polysaccharides. For example, in P6, the C-O-C peak remains at 1,252 cm^{-1} with a transmittance of 72.23%, and the C-O peak at 1,028 cm^{-1} shows a transmittance of 32.05%. The slight variations in intensity across formulations indicate changes in the polysaccharide composition, with carrageenan increasingly contributing to these peaks.

The sulfate ester peaks detected at 924 cm^{-1} are characteristic of carrageenan, which corresponds to C-O-C stretching related to 3,6-anhydro-D-galactose (Santos et al., 2023) and provides a clear marker for its presence in bioplastic films. In P1, these peaks are minimal, reflecting the low carrageenan content. However, as the carrageenan content increases from P2 to P6, the intensity of the sulfate ester peaks rises significantly. For instance, in P6, the sulfate ester peak at 924.4 cm^{-1} has a transmittance of 64.17%, clearly indicating the dominance of carrageenan. The increasing prominence of these peaks confirms the growing concentration of carrageenan in the formulations, which correlates with the observed flexibility enhancements due to carrageenan's ability to interact effectively with glycerol. Overall, the results demonstrate a clear transition from cellulose-dominated features in P1 to carrageenan-dominated features in P6. This shift in chemical composition impacts the bioplastic films' physical properties, which is discussed later. Higher cellulose content potentially contributes to rigidity, while higher carrageenan content enhances flexibility due to its stronger interactions with glycerol.

Thickness

Thickness is a parameter that can affect bioplastic's mechanical and physical properties, such as water vapor and gas permeability, tensile strength, and elongation. Thicker bioplastics can offer better protection for packaged products (Arifin et al., 2023). The study shows a significant variation in bioplastic thickness based on the different ratios of CPH cellulose and kappa-carrageenan used (Figure 2).

It is revealed that the higher cellulose usage results in the lowest bioplastic thickness, while the highest kappa-carrageenan results in the thickest. This study agreed with a study by Favian and Nugraheni (2023) who reported that a 0.5% addition of carrageenan into a chitosan-based bioplastic blend significantly increased the thickness by 2.8 times. Additionally, Akshana et al. (2024) revealed that incorporating 0.5 to 2 g of cellulose did not significantly increase the thickness of starch/gelatin blend films. Kappa-carrageenan acts as a suspending agent, gelling agent, and thickener that can increase the viscosity of the dissolved solids in the bioplastic solution.

Varying carrageenan concentrations in biodegradable films affect film thickness, indicating that the concentration of carrageenan plays a role in determining film thickness (Rajasekar et al., 2023). The higher concentrations of carrageenan will increase the viscosity, leading to increased thickness due to the creation of more space in the film matrix (Hanry and Surugau, 2020). This is due to the repulsion between negative charges on the sulfate groups, which causes the molecular chains to become rigid. The hydrophilic nature of the polymer leads to water molecules binding around it, increasing the viscosity of the bioplastic solution mixtures (Heriyanto et al., 2018). The difference in total dissolved solids in each treatment causes differences in the resulting bioplastic thickness during casting.

Swelling

Swelling tests are crucial in determining a bioplastic's ability to absorb water, reflecting its degradation potential. Lower swelling values indicate reduced water absorption, while higher values suggest greater water uptake, aiding in faster material breakdown (Fadhallah et al., 2024). The study shows that the combination of cellulose from CPH and kappa-carrageenan significantly affects the swelling values of the bioplastics, ranging from 74.56 to 95.68% (Figure 3).

The swelling values of the bioplastic samples show a clear increasing trend from P1 to P6, indicating that the films absorb more water as the carrageenan content increases. In P1, with the highest cellulose content, the swelling percentage is the lowest at 74.56%. This suggests that cellulose, with its dense structure and strong hydrogen bonding, restricts water absorption, leading to lower swelling. This observation is supported by the FTIR results (Figure 1), where the O-H stretching peak at 3,272 cm^{-1} in P1 is broad and intense, indicating extensive hydrogen bonding among the cellulose's hydroxyl groups. These bonds limit water uptake, correlating with

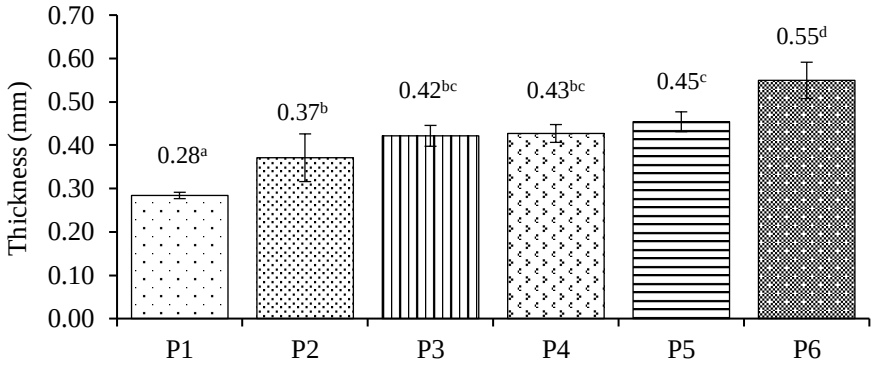

Figure 2. Thickness of bioplastic from various formulations

Note: Different superscript letter indicates significant differences at α = 5% of Duncan test

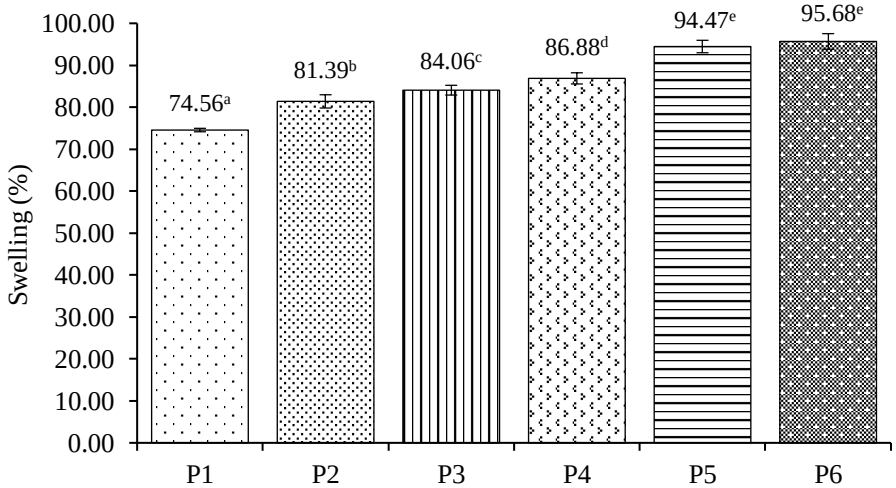

Figure 3. Swelling value of bioplastic from various formulations

Note: Different superscript letter indicates significant differences at α = 5% of Duncan test

the lower swelling observed in P1. Additionally, the result is consistent with findings by Adeleye et al. (2022), where cellulose from CPHs showed a swelling capacity of 36.36%, indicating low water absorption. Akshana et al. (2024) found higher cellulose and lower starch concentrations, resulting in the gradual decrease of water solubility of cellulose-starch blend films. Also, Etale et al. (2023) stated that cellulose's natural insolubility in water is related to the numerous hydrogen bonds in its crystalline and amorphous phases. This swelling value coincides with the thickness results (Figure 2), showing that P1 has the lowest thickness value and exhibits the lowest swelling value.

In contrast, the highest swelling value, 95.68%, was found in P6, with no significant difference from P5 at 94.47%. This trend aligns with de Lima Barizão et al. (2020) and Moustafa et al. (2021), who reported increased swelling as kappa-carrageenan concentration rose in starch-based films. Kappa-carrageenan's hydrophilic nature, supported by the FTIR data (Figure 1) showing dominant sulfate ester peaks at 924 cm^{-1} in P6 with the highest intensity, explains its ability to absorb large amounts of water, forming hydrogen bonds through its free hydroxyl (-OH) groups and sulfate ester structures, which integrate water into the double helix matrix (Muthukumar et al., 2021). This swelling value also coincides with the thickness value (Figure 2), showing P6 as the thickest film with the highest swelling value.

Mechanical properties

The observed properties, including tensile strength, elongation at break, and young's modulus, exhibit clear trends as the proportion of carrageenan increases (Table 2). The ratios of these components strongly influence the mechanical properties of bioplastics formulated from cellulose and kappa-carrageenan. Higher carrageenan content enhances tensile strength and elongation at break due to its ability to form a robust and flexible gel matrix, while higher cellulose content increases stiffness but reduces flexibility.

The tensile strength of the bioplastics ranged from 1.81 in P1 to 6.77 MPa in P6, demonstrating a significant increase with higher carrageenan content. This phenomenon aligns with the findings by de Lima Barizão et al. (2020) that the higher carrageenan ratio to starch enhances the tensile strength of bioplastics from 6.53 (25% carrageenan) to 17.29 MPa (50% carrageenan). Favian and Nugraheni (2023) also reported the same trend, where adding 0.5% carrageenan exhibits a 47% increase in tensile strength. Carrageenan's ability to form a robust and interlinked polymer matrix through hydrogen bonding and ionic interactions is primarily responsible for this increase. The firm gel network formed by carrageenan contributes to the bioplastic's ability to resist tensile forces before breaking. Conversely, the higher cellulose content led to lower tensile strength, consistent with the findings of Rohadi et al. (2022) who reported that a 0.5% increase in the concentration of cellulose filler extracted from Napier grass significantly reduced the tensile strength of bioplastic.

The elongation at break, which measures the bioplastic's ability to stretch before breaking, ranged from 3.66% in P1 to 43.88% in P5. The increase in elongation with higher carrageenan content can be attributed to carrageenan's ability to form hydrogen bonds with water, creating a flexible gel matrix that enhances the bioplastic's elasticity (Sofianto et al., 2022). This trend is consistent with Abdullah et al. (2021) who observed similar increases in elongation with higher carrageenan levels, which significantly rose from 13% (0% carrageenan) to 17% (10% carrageenan). In contrast, higher cellulose content resulted in lower elongation values, making the bioplastic more brittle and rigid. This effect is due to cellulose's linear and crystalline structure, which limits the bioplastic's ability to deform

Table 2. Mechanical properties of bioplastic

Sample	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)
P1	1.81 ± 0.37^{a}	3.66 ± 0.97^{a}	50.44 ± 8.51^{a}
P2	2.06 ± 0.66^{ab}	16.62 ± 6.93^{b}	14.05 ± 6.35^{b}
P3	2.33 ± 0.43^{ab}	23.47 ± 7.50^{bc}	13.61 ± 10.82^{b}
P4	2.35 ± 0.38^{ab}	30.43 ± 4.07^{cd}	7.87 ± 1.94^{b}
P5	3.24 ± 0.46^{b}	43.88 ± 3.45^{e}	7.37 ± 0.53^{b}
P6	6.77 ± 1.08^{c}	40.31 ± 5.23^{de}	17.26 ± 5.28^{b}

Note: Different superscript letters in the same column indicate significant differences at $\alpha = 5\%$ of the Duncan test

under stress (Shang et al., 2019; Zamanian et al., 2021). Kassab et al. (2019) revealed the same trend that the higher cellulose nanocrystal, from 0 to 1%, significantly reduced the strain value of kappa-carrageenan-based films from 27.5 to 23%. Abdullah et al. (2020) also reported that the elongation of microcrystalline cellulose-starch-based bioplastic with 10% cellulose is significantly lower (1.6%) than without cellulose (9.2%).

Young's modulus, which indicates the stiffness of the bioplastic, ranged from 7.37 in P5 to 50.44 MPa in P1, showing a general decrease with increasing carrageenan content. This decrease in stiffness corresponds to carrageenan's plasticizing effect, which disrupts the crystalline regions of cellulose, resulting in a more flexible material. The highest modulus observed in P1 is due to the higher cellulose content, which increases stiffness through strong hydrogen bonds between cellulose molecules, reinforcing the polymer matrix (Müller et al., 2009). This finding is supported by Abdullah et al. (2020), who reported that higher cellulose concentrations lead to an increase in young's modulus by 500%, from 0.2 (without cellulose) to 1.1 GPa (10% cellulose). However, in formulations with higher carrageenan content, such as P6, the modulus slightly increased compared to other high-carrageenan formulations. This may be due to a denser gel network, which, although less stiff than cellulose, still contributes to a degree of structural reinforcement (de Lima Barizão et al., 2020).

WVTR

The analysis of the WVTR in bioplastics formulated with varying ratios of cellulose from CPHs and kappa-carrageenan reveals a significant influence of the polymer composition on the resulting WVTR values. The observed WVTR values ranged from 22.14 to 25.82 g m^{-2} day^{-1} (Figure 4), demonstrating that higher concentrations of carrageenan lead to increased water vapor permeability, whereas higher cellulose content results in a lower WVTR. The formulation of these biopolymers significantly affects the material's ability to transmit water vapor, with specific trends aligning with their inherent properties.

Carrageenan, particularly kappa-carrageenan, is a hydrophilic biopolymer characterized by its ability to absorb water due to hydroxyl (-OH) groups and sulfate ester groups within its structure. These groups form hydrogen bonds with water molecules, forming a gel-like network that swells and increases the diffusion pathways for water vapor. Consequently, as the carrageenan content in the bioplastic increases, the matrix becomes more amorphous and less dense, creating space that further enhances WVTR. This is evident in the highest WVTR observed in sample P6 (25.82 g m^{-2} day^{-1}), which contains 10 g of carrageenan without cellulose. This trend is consistent with previous research showing increased WVTR with higher carrageenan content, as highlighted by studies such as Jacoeb et al. (2014), where WVTR increased from 231.23 (2% carrageenan) to 246.06 g m^{-2} day^{-1} (3% carrageenan).

In contrast, cellulose, with its semi-crystalline structure and low crystallinity index of 26.32% (Adeleye et al., 2022), contributes to bioplastic's hydrophobicity and barrier properties. The crystalline regions of cellulose are more ordered and compact, restricting the movement of water vapor through the matrix. The amorphous regions may still allow some permeability, but the overall effect of higher cellulose content is a reduced WVTR. This is supported by the lower WVTR observed in sample P1 (22.14 g m^{-2} day^{-1}), which

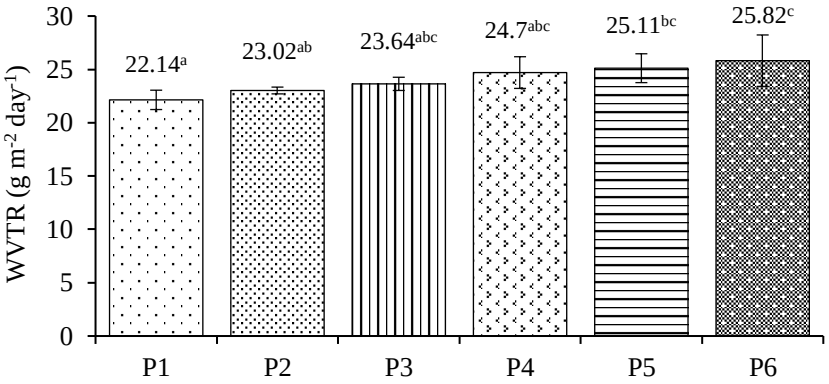

Figure 4. WVTR of bioplastic from various formulations

Note: Different superscript letter indicates significant differences at α = 5% of Duncan test

contains the highest amount of cellulose (8 g). This cellulose-rich matrix effectively impedes WVTR due to its limited pore space and more structured molecular arrangement, as evidenced by previous studies. Akshana et al. (2024) reported the same trend, which showed a significant decrease in WVTR of starch-based biodegradable films from $9x10^{-3}$ to $4x10^{-3}$ g m^{-2} day^{-1} as the cellulose from palmyra fruit concentration rise, from 0 to 2 g, respectively. Sunardi et al. (2020) revealed a significant reduction in the WVTR value of bioplastic film with the higher addition of nipa palm frond cellulose, from 0.061 (without cellulose) to 0.028 g cm^{-2} $hour^{-1}$ (0.6% cellulose). Another study discussed how cellulose alters the diffusion path of water molecules in bioplastic nanocomposites, creating a tortuous route that obstructs WVTR through the film. This suggests that incorporating cellulose can enhance the barrier properties of bioplastic films by reducing WVTR.

The interplay between the hydrophilic nature of carrageenan and the more hydrophobic properties of cellulose is crucial in determining the water vapor barrier properties of the bioplastic. While carrageenan increases the film's ability to absorb and transmit water vapor, cellulose contributes to a denser, less permeable structure. This balance is essential for optimizing the barrier properties of bioplastic films, depending on the desired application. However, it is essential to note that the WVTR values observed in this study exceed the maximum standard set by JIS z 1707 (7 g m^{-2} day^{-1}), suggesting that further formulation adjustments may be necessary to meet specific water vapor barrier requirements. This indicates that while the bioplastics demonstrate promising properties, additional refinement is required to align with industry standards.

Stability test

This test was conducted to determine the durability of bioplastics at room temperature. Visual observations were made until bioplastic changes were detected (Figure 5). The results showed that the bioplastics stored at room temperature did not undergo significant visual changes from week 1 to 4. However, in week 3, samples P5 and P6 exhibited small holes on the surface of the bioplastics.

The appearance of small holes in P5 and P6 by the third week can be correlated with the high carrageenan content in these formulations. Carrageenan is known to be more susceptible to water absorption due to its hydrophilic nature, which could lead to structural weaknesses over time, especially under ambient conditions where moisture is present. The formation of holes suggests that the water absorption might be causing localized swelling or dissolution of the carrageenan-rich regions, leading to structural failures in tiny perforations. By week 5, samples P4 and P2 began to show mold growth, indicated by black spots on the bioplastic surface. Additionally, by week 5, the bioplastics became slightly damp. Carrillo et al. (2023) reported the same phenomenon but with a faster formation of black spots, indicating mold growth on rich carrageenan films in 30 days. Richert et al. (2018) identified the black spot found in biodegradable films are related to the growth of *Aspergillus niger* and *Chaetomium globosum*.

Karaca et al. (2022) explained that bioplastic containing high moisture content, due to the material's intermolecular hydrogen bond, promotes mold growth and microbial activation noticed with the appearance of a black spot on the surface of the film. In this study, the delayed onset of mold growth in P4 and P2, which contain lower carrageenan content, indicates that cellulose-rich bioplastic may be more resistant to immediate moisture absorption but are not entirely immune to microbial growth over time. This microbial susceptibility could be exacerbated by the gradual accumulation of moisture within the bioplastic matrix, further supported by the dampness observed in the later weeks. This was due to the use of hydrophilic materials and the placement of the bioplastics at room temperature, leading to water vapor absorption from the environment. These conditions facilitate mold growth when bioplastics are stored in open spaces for extended periods.

Biodegradation

The biodegradability test was conducted to determine the time required for bioplastics to decompose or degrade naturally through microbial activity or biological processes in a specific soil environment. This test employed a soil burial method, where each bioplastic sample was placed in the soil until complete degradation was observed (Akhlaq et al., 2023). Table 3 presents the observation results of sample burial over 3 weeks. The outcomes from the soil burial test correlate strongly with the compositional differences among formulations P1 to P6. The higher the carrageenan content, the faster the degradation, as evidenced by the early and

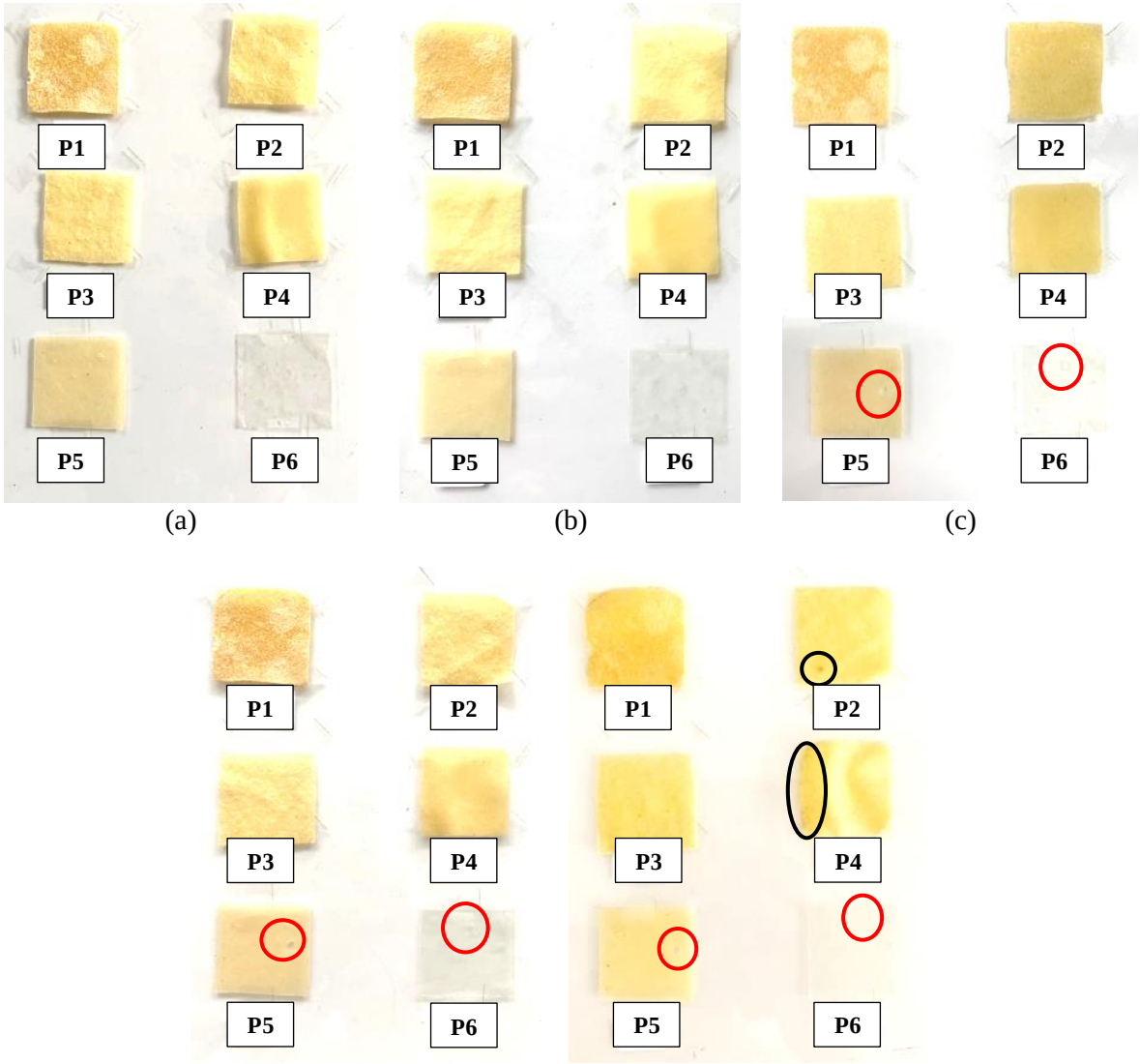

Figure 5. Visual observation on bioplastic during stability test at room temperature (a) 1, (b) 2, (c) 3, (d) 4 and (e) 5 weeks

Note: **O** = small hole, **O** = black spot formation

significant breakdown of samples P5 and P6. In contrast, formulations with higher cellulose content (e.g., P1) exhibited greater resistance to degradation. Higher concentrations of carrageenan accelerate degradation due to its hydrophilic nature and amorphous structure (Dmitrenko et al., 2023), which are more susceptible to microbial degradation. On the other hand, cellulose contributes to structural integrity, slowing down the degradation process, especially at higher concentrations. The crystalline regions of cellulose are less accessible to microorganisms (Adeleye et al., 2022), resulting in a slower degradation process that complements the rapid breakdown of carrageenan.

During the first week, the bioplastics softened and were covered with mold on the surface. By the second week, the bioplastics fragmented into small, brittle pieces that adhered to the soil. By the third week, complete degradation was observed, with no remnants of the bioplastics remaining in the soil. The rapid degradation of these bioplastics can be attributed to their composition, which includes natural raw materials such as cellulose and carrageenan. Other studies, such as Rumi et al. (2024), reported a more extended degradation period of 63 days for cellulose-based films made from low-quality cotton, while Bilo et al. (2018) found that cellulose-based bioplastics made from rice straw required 105 days for complete degradation. Additionally, Fadhallah et al. (2024) observed that bioplastics containing 15% carrageenan combined with starch degraded faster (11 days)

Table 3. Observation of the bioplastic biodegradation test for 3 weeks

Sample	Visual observation (with weight loss)		
	1 week	2 weeks	3 weeks
P1	 38.9%	 87.4%	 100.0%
P2	 59.2%	 92.8%	 100.0%
P3	 65.8%	 93.4%	 100.0%
P4	 87.8%	 95.5%	 100.0%
P5	 91.4%	 100.0%	–
P6	 100.0%	–	–

than those containing 10% carrageenan (15 days). The findings of this study indicate a relatively short degradation time of carrageenan-rich bioplastic. The rapid degradation observed is consistent with the hydrophilic nature of carrageenan, which accelerates the breakdown process in moist environments. This characteristic, combined with the amorphous regions of cellulose, which are more susceptible to microbial attack, contributes to the swift biodegradation observed in these samples.

The biodegradation process of bioplastics involves the breakdown of polymer chains by microorganisms into simpler units, resulting in organic compounds that are environmentally benign (Pooja et al., 2023). Microorganisms degrade the cellulose content in bioplastics through the cleavage of β-1,4-glycosidic bonds into simpler glucose units (Hidayati et al., 2019). The crystalline structure of cellulose (denser and more ordered) tends to degrade more slowly than its amorphous structure. Bioplastics are readily degraded due to hydroxyl (-OH) and carboxyl (-COOH) groups, which are naturally degradable. Other factors influencing the bioplastic degradation rate include moisture, temperature, and the quantity and type of microbes present in the soil (Rendón-Villalobos et al., 2022). Hydroxyl groups in cellulose and carrageenan facilitate hydrogen bonding with water, increasing microbial accessibility and accelerating degradation. When these factors are optimized, the biodegradation of bioplastics occurs more rapidly, reflecting the environmentally friendly potential of these materials.

CONCLUSIONS

This study successfully produced biodegradable films from cellulose derived from CPHs and kappa-carrageenan, demonstrating that varying ratios of these materials significantly affect bioplastic properties. Higher cellulose content enhances rigidity, while increased kappa-carrageenan improves tensile strength and elongation. FTIR analysis confirms the chemical interactions between cellulose, kappa-carrageenan, and glycerol, supporting these findings. The formulation of P5, which consists of 2 g of CPH's cellulose and 8 g of kappa-carrageenan, is recommended for bioplastic film packaging based on its mechanical, physical, and biodegradability properties. This tailored formulation offers a sustainable alternative to conventional plastics, addressing environmental concerns associated with petroleum-based plastics. Further research should focus on optimizing these bioplastic formulations to enhance commercial viability, specifically in sustainable packaging applications where tailored mechanical properties are essential.

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