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BIODEGRADABLE FILMS FROM COCOA HUSK-DERIVED CELLULOSE WITH KAPPA AND IOTA-CARRAGEENAN FORMULATIONS

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Abstract

The extensive use of conventional plastic packaging has raised environmental concerns due to its non-biodegradable nature, encouraging the development of biodegradable materials from renewable resources. Cocoa pod husks, an abundant agricultural by-product, are a promising source of cellulose for biodegradable film production. Kappa (κ-) and iota (ι-) carrageenans are widely used as matrix materials to improve the film-forming properties. This study investigated the characteristics of biodegradable packaging films produced from cocoa pod husk-derived cellulose combined with κ- and ι-carrageenan. Films were produced using the solution casting method with eight different formulations of cellulose, κ-carrageenan, and ι-carrageenan. The films were characterized by (FTIRr-transform infrared spectroscopy (FTIR), X-Ray Diffraction (XRD), thickness, tensile strength, elongation at break, Young's modulus, swelling, and water vapor transmission rate (WVTR). Cellulose incorporation generally increased film thickness and reduced tensile strength compared to carrageenan-based films, while lowering the WVTR in selected formulations. Films containing higher proportions of ι-carrageenan exhibited the greatest elongation and the highest water absorption. The swelling values ranged from 159.50% to 473.23%, whereas the WVTR ranged from 1.14 to 5.43 g/m²/24 h. Increasing the cellulose content generally reduced the WVTR, reflecting the improved water vapor barrier properties associated with higher crystallinity. All cellulose-containing films satisfied the minimum tensile strength requirement specified in JIS Z 1707, although their elongation remained within the poor-to-moderately good category. Overall, biodegradable films formulated from cocoa pod husk cellulose with κ- and ι-carrageenan demonstrate promising potential as sustainable and environmentally friendly food packaging materials.

Keywords: bioplastics, mechanical properties, polysaccharide-based films, water vapor transmission

Film Biodegradabel Berbasis Selulosa Kulit Kakao dengan Formulasi Kappa dan Iota-Karagenan

Abstrak

Penggunaan kemasan plastik konvensional menimbulkan permasalahan lingkungan karena sulit terdegradasi sehingga mendorong pengembangan material biodegradabel yang

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berasal dari sumber daya terbarukan. Salah satu bahan yang berpotensi dimanfaatkan adalah selulosa dari kulit buah kakao sebagai hasil samping pertanian. Untuk meningkatkan kemampuan pembentukan film, selulosa diformulasikan dengan kappa (κ-) dan iota (ι-) karagenan. Penelitian ini bertujuan mengkaji karakteristik film kemasan biodegradabel berbasis selulosa kulit buah kakao yang diformulasikan dengan κ- dan ι-karagenan. Film dibuat menggunakan metode solution casting dengan delapan variasi formulasi selulosa, κ-karagenan, dan ι-karagenan. Karakterisasi meliputi analisis FTIR (*Fourier Transform Infrared*), XRD (*X-Ray Diffraction*), ketebalan, kuat tarik, elongasi saat putus, modulus Young, *swelling*, dan laju transmisi uap air (*water vapor transmission rate*/WVTR). Hasil penelitian menunjukkan bahwa penambahan selulosa umumnya meningkatkan ketebalan film dan menurunkan kuat tarik dibandingkan film berbasis karagenan, serta menurunkan nilai WVTR pada beberapa formulasi. Film dengan dominasi ι-karagenan menghasilkan nilai elongasi tertinggi, tetapi juga memiliki kemampuan menyerap air paling besar. Nilai *swelling* berkisar antara 159,50% hingga 473,23%, sedangkan WVTR berada pada rentang 1,14–5,43 g/m^2/24 jam. Makin tinggi kandungan selulosa, makin rendah nilai WVTR, yang menunjukkan kemampuan penghambatan terhadap transmisi uap air makin baik dan berkaitan dengan meningkatnya kristalinitas film. Seluruh film yang mengandung selulosa memenuhi persyaratan minimum kuat tarik menurut JIS Z 1707 meskipun nilai elongasinya masih tergolong rendah hingga cukup baik. Hasil penelitian ini menunjukkan bahwa formulasi selulosa kulit buah kakao dengan κ- dan ι-karagenan berpotensi dikembangkan sebagai material bioplastik yang ramah lingkungan untuk aplikasi kemasan pangan.

Kata kunci: bioplastik, film polisakarida, laju transmisi uap air, sifat mekanis

INTRODUCTION

Food packaging plays a crucial role in the global food industry by protecting food products, maintaining their quality and safety, and extending their shelf life during storage and distribution (Suvarna *et al*., 2022). Proper packaging design reduces food waste and ensures product stability throughout the supply chain. Nevertheless, synthetic plastics such as polyethylene, polypropylene, and polyethylene terephthalate are still the most widely used packaging materials because of their low cost, lightness, and ease of processing. The extensive reliance on petroleum-based materials has resulted in a continuous increase in the generation of plastic waste. In Indonesia, plastic waste has increased markedly over the past three years, from approximately 5 million tons to 6.5 million tons annually (KLHK, 2022). Furthermore, synthetic plastics raise environmental concerns because they originate from non-renewable resources and persist in the environment due to their resistance to natural degradation processes (Chawla *et al*., 2021).

After use, synthetic plastic packaging is commonly disposed of through landfilling or incineration, both of which pose serious environmental challenges to the ecosystem. Suleman *et al*. (2022) reported that insufficient recycling and ineffective waste management of plastic packaging lead to global economic losses estimated at USD 80–120 billion annually. Increasing awareness of these negative impacts has encouraged consumers to seek safe, bio-based, and environmentally friendly packaging materials. Consequently, the food industry faces growing pressure to develop alternative packaging systems that reduce environmental burdens. Consequently, research and industrial interest in biodegradable packaging materials has increased significantly (Petkoska *et al*., 2021).

Studies on biodegradable packaging materials have expanded rapidly in recent years, driven by increasing concerns regarding food safety, human health, and environmental protection (Mahcene *et al*., 2021). Biodegradable packaging contributes to sustainable packaging concepts by reducing dependence on fossil-based resources, lowering post-consumer waste, and minimizing food waste (Zhao *et al*., 2021). A distinctive feature of biodegradable packaging is its close integration with food products, allowing safe disposal without generating

persistent environmental residues (Barbosa *et al*., 2021). Therefore, the development of biodegradable packaging based on biomass-derived polymers has gained considerable attention. Recent reviews have emphasized that biopolymer-based materials, including cellulose, starch, chitosan, alginate, and carrageenan, are promising alternatives to conventional plastics because of their renewable origins and biodegradability (Cheng *et al*., 2024; Nurdiani *et al*., 2024; Arshad *et al*., 2025; Fransiska *et al*., 2025).

Cellulose is one of the most abundant natural polysaccharides and has attracted attention as a raw material for biodegradable packaging applications (Petkoska *et al*., 2021). It can be obtained from wood and agricultural residues, providing opportunities for waste utilization (Gabriel *et al*., 2020). Indonesia, particularly Lampung Province, is a major cocoa-producing region, with an annual production of approximately 56 thousand tons (BPS, 2022). In cocoa processing, beans are the primary product, whereas cocoa pod husks (CPH) are generally discarded as waste. Cocoa pod husks can account for up to 75% of the total fruit weight. Recent studies have confirmed that cocoa pod husks are rich in structural polysaccharides, with cellulose content typically ranging from approximately 26 to 44%, along with hemicellulose, lignin, and minor ash fractions, depending on the extraction conditions and cultivar (Amrillah *et al*., 2024). These findings further support CPH as a promising cellulose source for biodegradable packaging materials

Despite these advantages, cellulose-based films have limitations related to poor water resistance and limited flexibility, which restrict their direct applications. Although cellulose is non-toxic and possesses good mechanical strength (Shahidi & Hossain, 2022), formulation strategies are required to improve its processability and performance. Hydrocolloid-based materials are known for their high water absorption capacity and good compatibility with natural polymers (Jayakody *et al*., 2023). One of the most important natural sources of hydrocolloids is red seaweed, which contains carrageenan and is widely used as a stabilizer and film-forming agent in biodegradable packaging (Bhat *et al*., 2020). Carrageenan consists of three main fractions: kappa (κ), iota (ι), and lambda (λ). A recent review by Jabeen *et al*. (2025) highlighted that κ- and ι-carrageenan are the most widely applied fractions in food, coating, and biodegradable packaging applications because of their gel-forming ability, whereas λ-carrageenan does not. Mathew *et al*. (2024) reported that carrageenan-based films, particularly when modified or combined with other polymers, exhibit improved mechanical, thermal, and functional properties, supporting the use of κ–ι carrageenan combinations to overcome the inherent brittleness of κ-carrageenan films and enhance overall film performance. Sulistiana *et al*. (2024) demonstrated that combining κ- and ι-carrageenan improves the mechanical properties of biodegradable packaging materials.

In a previous study, biodegradable films based on κ-carrageenan and cocoa pod husk-derived cellulose demonstrated satisfactory film formation but exhibited limited functional performance, particularly in terms of elongation at break and water vapor barrier properties (Fadhallah *et al*., 2025). However, existing reports predominantly focus on single carrageenan fractions or empirical blending approaches, providing limited mechanistic understanding of how κ- and ι-carrageenan differentially regulate the crystalline-amorphous balance, polymer chain mobility, and moisture transport behavior in cellulose-based films derived from agricultural residues. In particular, the comparative roles of κ- and ι-carrageenan in tuning the balance between film flexibility and water vapor barrier properties have not been fully elucidated. Therefore, this study aimed to investigate the characteristics of biodegradable packaging films produced from cocoa pod husk-derived cellulose combined with κ- and ι-carrageenan.

MATERIALS AND METHOD

Isolation of Cellulose from Cocoa Pod Husk

Cocoa pod husk waste was collected from cocoa farmers in Sukoharjo Village (Lampung, Indonesia). It was initially washed with tap water to remove physical contaminants,

cut into approximate 3×3 cm cubes, and dried in a laboratory oven (Memmert, Germany) for 6 h at 60°C to achieve a moisture content of 5%±1%. The dried material was then ground using a blender (Waring 8010 BU) for 2 min and sieved through an 80-mesh screen to obtain a fine powder. The isolation procedure, adapted from Sena *et al.* (2021), began by subjecting 250 g of cocoa pod husk powder to delignification with 1 L of 12% w/v sodium hydroxide (NaOH) solution and stirring for 120 min. The resulting mixture was filtered, and the residue was repeatedly washed with water until a neutral pH was reached. The cellulose residue was then bleached by mixing with 300 mL of 3% hydrogen peroxide (H_2O_2) and heating for 1 h at 100°C. The bleached cellulose was filtered, washed again to achieve a neutral pH, dried in an oven for 3 h at 60°C, and pulverized using a grinder.

Biodegradable Films Preparation

Biodegradable films were fabricated using the casting method based on the procedure reported by Fadhallah *et al.* (2025). Cellulose powder, κ-carrageenan, ι-carrageenan (IndoGum, mesh-80), and glycerol (100% purity, OneMed) were prepared according to the formulations listed in Table 1. Cellulose powder isolated from cocoa pod husks was first dispersed in 30 mL of distilled water in a glass beaker and stirred until a homogeneous suspension was formed. Glycerol was then added under continuous stirring. Separately, κ-carrageenan and ι-carrageenan powders were pre-mixed in a separate container until evenly blended and subsequently added gradually to the cellulose suspension. The mixture was then adjusted to a final volume of 100 mL with distilled water and heated at 60 °C for 30 min under continuous stirring on a magnetic hot plate (Cimarec+, US). The resulting film-forming solution was immediately cast onto 10 cm diameter Petri dishes and allowed to cool and set at 25 °C for 1 d. To ensure complete film formation, the cast films were dried in a dehydrator at 50°C for 2 h. Following drying, the films were conditioned at room temperature for an additional 2 h before being carefully peeled from the Petri dishes, yielding film packaging ready for further characterization.

FTIR and XRD Analysis

Fourier-transform infrared (FTIR) spectroscopy was used to investigate the chemical structures and functional groups of the samples. Potassium bromide (KBr) pellets were prepared, and the FTIR portable instrument (Agilent 630, US) was set to a resolution of 2 cm^{-1} within the wavenumber range of 500–4,000 cm^{-1}. A sample of 1×1 cm was placed in the instrument, and infrared scanning was performed 15 times. The resulting infrared spectra were recorded, graphically presented, and interpreted. Subsequently, X-Ray Diffraction (XRD) analysis was conducted to determine the crystallinity level of the film samples using an X-Ray Diffraction instrument (PANalytical,

Table 1 Formulation for cellulose, κ-carrageenan, and ι-carrageenan films

Code	Cellulose (g)	κ-carrageenan (g)	ι-carrageenan (g)	Distilled water (mL)	Glycerol (mL)
T1	0	10	0	87.5	2.5
T2	0	0	10	87.5	2.5
T3	8	2	0	87.5	2.5
T4	8	1	1	87.5	2.5
T5	8	0	2	87.5	2.5
T6	5	5	0	87.5	2.5
T7	5	2.5	2.5	87.5	2.5
T8	5	0	5	87.5	2.5

X'Pert PRO, Netherlands). Samples were cut into a 2 cm diameter and were tested using Cu Kα radiation (λ=1.54060 ˚A) under operational conditions of 40 kV and 35 mA with a scanning speed of 1∘/min. The XRD data were visualized using Origin software (version 95E), and the crystallinity index (CI) was estimated using the Segal method, as described by Salem *et al*. (2023).

Mechanical Testing

Mechanical testing of the film samples was essential to determine their physical strength and structural integrity for simulated packaging applications. The evaluated parameters included the thickness, tensile strength, elongation at break, and Young's modulus. The film thickness was measured using a digital micrometer (RoHS, China) at room temperature. The testing procedure was based on the ISO 527-1 standard (ISO for Standardization, 2019). A Universal Testing Machine (Zwick Roel) was used, configured with a 25 kg load cell, a crosshead speed of 1 mm/s, and a grip separation of 60 mm. Samples were cut to a size of 7×2 cm and mounted vertically onto the machine grips under controlled conditions of 65% relative humidity (RH) and room temperature (25–27 °C). Upon testing, the monitor displayed the resulting force (kgf) and the change in length (mm). The tensile strength (MPa) was calculated by dividing the force by the cross-sectional area (mm2) of each sample. The elongation at break (%) was determined by dividing the change in the sample length at the break by the initial length. Young's modulus (MPa) was expressed as a relative stiffness index calculated from the tensile strength and elongation at break values. All results are presented as mean ± standard deviation.

Barrier Properties (Water Absorption and WVTR)

The barrier properties of the biodegradable films were assessed by measuring water absorption and Water Vapor Transmission Rate (WVTR) in triplicate. The initial weight (W_0) of a 2×2 cm sample was recorded, and the sample was then immersed in 20 mL of distilled water at room temperature for 24 h. After the immersion period, the sample was removed, drained for 5 min, wiped with tissue to remove surface water, and the final weight (W_1) was measured. The water absorption (%) was calculated by dividing the difference between the final and initial weights by the initial weight and multiplying the result by 100 (ASTM, 2022).

For WVTR testing, which measures the film's permeability to water vapor, a crucial factor where a lower WVTR indicates better protection against sample weight loss, the procedure followed the ASTM E96 standard (ASTM, 2017). Circular film samples (7 cm diameter) were used to seal a 6 cm diameter Petri dish containing 10 g of silica gel. The sample was secured tightly using rubber bands and petroleum jelly (Vaseline) on the dish as a lid, ensuring a hermetic seal to ensure that water vapor transmission occurred only through the film sample. The initial weight (W_0) of the dish assembly was recorded using an analytical scale (0.0001 precision), and it was stored at room temperature (25–27 °C) for 24 h (T). The final weight (W_1) was recorded the following day. The WVTR ($g/m^2/24h$) was calculated by dividing the change in the weight of the dish assembly (final weight minus initial weight) by the product of the total observation time (T) and the transmission area of the sample (A). All WVTR results are presented as the mean followed by the standard deviation.

Data Analysis

The FTIR and XRD data were analyzed descriptively. The mechanical and barrier property data were statistically analyzed using IBM SPSS® version 25. Analysis of variance (ANOVA) was performed to determine the effects of treatments, followed by Duncan's Multiple Range Test (DMRT) at a 5% significance level to evaluate the differences among treatments.

RESULTS AND DISCUSSION

The visual appearances of the films are shown in Figure 1, where differences in transparency, color, and surface texture can be observed across the formulations. The absence of cellulose resulted in more transparent and smoother films, which is consistent with

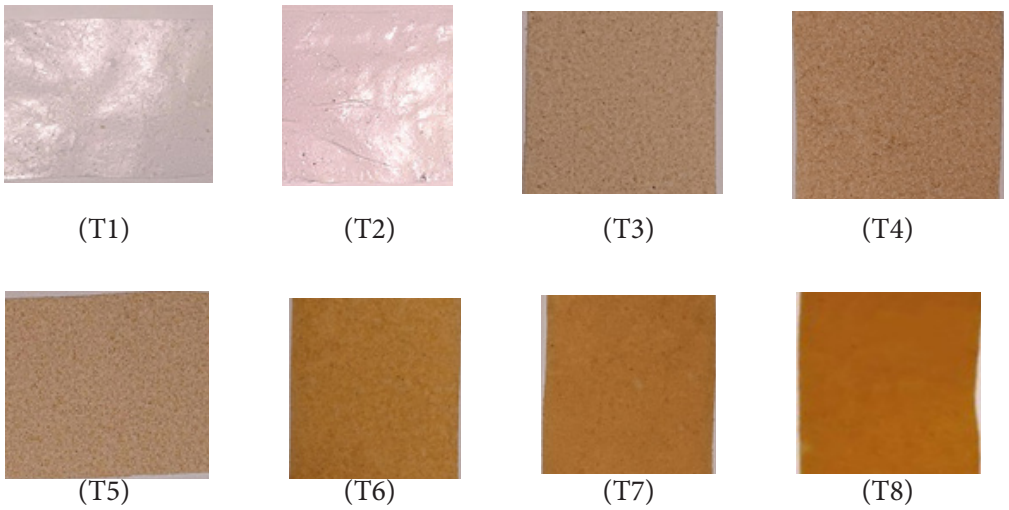

Figure 1 Visual appearance of cellulose- and carrageenan-based biodegradable films with different formulations

a more homogeneous carrageenan matrix and reduced light scattering in smoother, well-packed films (Mathew *et al.*, 2024). In contrast, the films containing cellulose were more opaque and had a slightly rougher surface because the dispersed cellulose fibers/particles increased light scattering and reduced transparency (Dmitrenko *et al.*, 2023). At certain compositions, the films appeared more uniform, and films with higher carrageenan content showed a smoother appearance with a consistent brownish color, suggesting better matrix continuity and stronger interfacial cohesion, as carrageenan acts as a binding phase around dispersed particles in the composite film (Dmitrenko *et al.*, 2023). Overall, the results indicate that the film appearance varied with the formulation.

FTIR Analysis

The FTIR spectra of all the films exhibited the typical vibrational features of polysaccharide-based materials, confirming the successful formation of carrageenan/cellulose composite films (Figure 2). The most prominent feature across all formulations was a broad O–H stretching band observed at 3260–3332 cm^{-1}, which reflects the high density of hydroxyl groups inherent to both cellulose and carrageenan, as well as the changes in the hydrogen-bonding environment within the composite network (Oh *et al.*, 2005). In polysaccharide composites, variations in the O–H stretching region are commonly interpreted as indicators of changes in hydrogen bonding environments and enhanced polymer–polymer compatibility, particularly in systems involving cellulose and sulfated galactans, such as carrageenan (Bhatia *et al.*, 2024). In addition, C–H stretching vibrations appearing at 2885–2922 cm^{-1} were consistently detected in all samples and were attributed to the polysaccharide backbone, in good agreement with previously reported spectra of cellulose-based films (Fadhallah *et al.*, 2025).

More specific structural information was obtained from the fingerprint region, which indicated carrageenan incorporation and its contribution to the composite structure. The presence of sulfate ester groups was confirmed by the S=O stretching band detected at 1252-1267 cm^{-1}, indicating that the carrageenan molecular structure remained intact after blending with cellulose. Variations in transmittance within this region were observed among the formulations, suggesting differences in sulfate group environments depending on the cellulose content and κ-/ι-carrageenan ratio. Formulations containing higher carrageenan proportions, particularly T8, exhibited a more pronounced sulfate-related band, indicating a stronger contribution of carrageenan functional groups within the

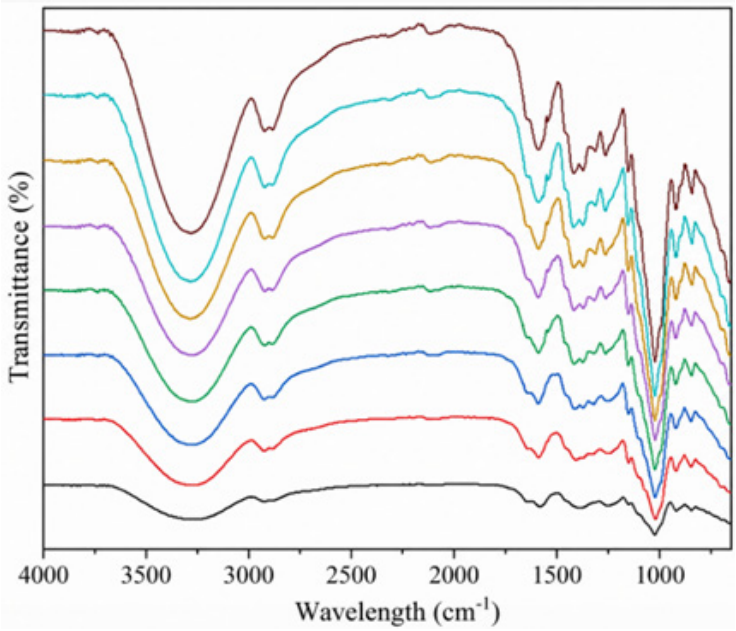

Figure 2 FTIR spectra of biodegradable films prepared from cocoa pod husk cellulose, κ-, and ι-carrageenan (T1: ▬, T2: ▬, T3: ▬, T4: ▬, T5: ▬, T6: ▬, T7: ▬, T8: ▬)

composite matrix. Similar sulfate-associated absorption behavior in carrageenan-based composite films has been reported in previous studies, where the sulfate ester region was influenced by the polymer composition and intermolecular interactions within the film network (Dmitrenko *et al.*, 2023; Bhatia *et al.*, 2024). Absorption bands within the range of ~1234–1260 cm^{-1} are widely recognized as characteristic of carrageenan and are frequently used for its identification and differentiation among carrageenan types (Olatunji, 2020; Renaka *et al.*, 2025).

Furthermore, distinct bands observed at 916–924 cm^{-1} and 842–849 cm^{-1} were assigned to 3,6-anhydro-D-galactose and D-galactose-4-sulfate, respectively, both of which are diagnostic features of κ-carrageenan and are closely related to its helix-forming capabilities. In parallel, pure ι-carrageenan has been reported to exhibit characteristic absorption bands at approximately 805, 845, and 905 cm^{-1}, corresponding to C–O–SO_3 stretching vibrations and sulfated 3,6-anhydrogalactose units (Polícia *et al.*, 2024). The slight shifts observed in the O-H stretching region (3260-3332 cm^{-1}) and fingerprint bands among the formulations indicate intermolecular interactions and changes in hydrogen bonding between the cellulose and carrageenan chains. These spectral variations suggest differences in polymer compatibility and structural organization depending on the kappa/iota carrageenan ratio. Similar FTIR peak shifts associated with hydrogen-bond rearrangement and polymer–polymer interactions have also been reported for carrageenan/cellulose-based composite films and other polysaccharide biopolymer systems (Dmitrenko *et al.*, 2023; Mathew *et al.*, 2024). The detection of absorption bands within this wavenumber region in the present study confirms the successful incorporation of ι-carrageenan alongside κ-carrageenan into the cellulose-based composite films.

XRD Analysis

The X-ray diffraction (XRD) patterns of the biodegradable films prepared from cocoa pod husk-derived cellulose and κ/ι-carrageenan showed clear differences in structural organization depending on the formulation (Figure 3). In the cellulose-containing films, diffraction peaks at 2θ ≈ 15–16° and 22–23° were observed, which are widely reported for cellulose extracted from agricultural residues and indicate the presence of ordered microfibril domains (Abdo *et al.*,

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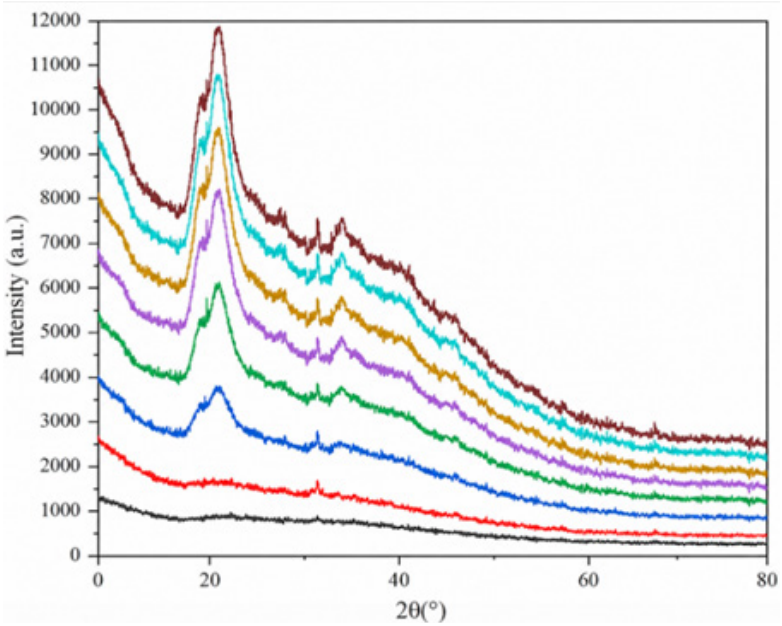

Figure 3 XRD pattern of biodegradable films prepared from cocoa pod husk cellulose, κ-, and ι-carrageenan (T1: ▬, T2: ▬, T3: ▬, T4: ▬, T5: ▬, T6: ▬, T7: ▬, T8: ▬)

2023). The retention of these peaks suggests that the native crystalline structure of cocoa pod husk cellulose was preserved after film preparation. In contrast, carrageenan-based films exhibited broad diffraction halos in the range of 2θ ≈ 15–25°, reflecting their predominantly amorphous nature. These profiles are characteristic of carrageenan matrices, which lack long-range order unless reinforced with other polymers or fillers, as reported in previous composite film studies (Park *et al*., 2024). The dominance of the amorphous features is consistent with the random coil configuration typical of sulfated polysaccharide networks.

To quantitatively assess the structural ordering, the crystallinity index (CI) was calculated using the Segal method. The CI values varied substantially among the formulations, ranging from approximately 0.9% to 46.3% (Table 2), indicating that the crystallinity was strongly influenced by the cellulose content and carrageenan type. Films composed solely of carrageenan showed very low CI values (10.4% for κ-carrageenan and 0.9% for ι-carrageenan), confirming their amorphous character and aligning with literature reports on unreinforced carrageenan films (Park *et al*., 2024). The incorporation of cocoa pod husk-derived cellulose increased the crystallinity index, particularly at higher cellulose loading (8 g), where CI values reached ~45–46% for κ-, ι-, and κ/ι-carrageenan formulations. These results indicate that cellulose remained the primary contributor to crystalline domain formation, while κ/ι-carrageenan blending appeared to affect the overall structural ordering of the amorphous phase, as inferred from the XRD patterns, rather than suppressing cellulose crystallinity, as observed in cellulose-rich biocomposites (Abdo *et al*., 2023; Ramadas *et al*., 2024).

At a lower cellulose content (5 g), CI values decreased (25.1–34.1%), with κ/ι-carrageenan films showing the lowest crystallinity, reflecting increased amorphous character owing to the flexible and elastic nature of ι-carrageenan. Similar trends have been reported for biodegradable polysaccharide composites, where the incorporation of flexible polymers increases the amorphous fraction and enhances chain mobility (Ramadas *et al*., 2024). Overall, these findings demonstrate that the crystalline–amorphous balance of the films can be effectively tuned through formulation design, consistent with the intermolecular interactions identified by FTIR analysis (Figure 2), which

are known to interfere with cellulose chain packing, thereby modulating the crystalline–amorphous organization.

Mechanical Properties

The mechanical properties of the biodegradable films, including thickness, tensile strength, and elongation at break, were significantly affected by the formulation of cellulose, κ-, and ι-carrageenan (Table 2).

The film thickness differed significantly among the formulations, with values ranging from 0.165 to 0.618 mm. According to the Japanese Industrial Standard (Japanese Standard Association, 2019), which recommends a maximum thickness of 0.25 mm for biodegradable food packaging films, only T1 satisfied the thickness requirement, whereas all other formulations exceeded this limit. The observed variation in thickness reflects the combined influence of cellulose, κ-carrageenan, and ι-carrageenan on the rheological behavior of the film-forming solutions and drying process. Cellulose contributed to the thickness development primarily by increasing the total solid content and promoting denser film structures during solvent evaporation. κ-Carrageenan further enhanced the thickness by forming strong and compact gel networks, which increased the solution viscosity and limited polymer chain rearrangement during drying. In contrast, ι-carrageenan formed softer and more elastic gel structures with higher water-binding capacity, resulting in swollen matrices before drying and consequently thicker films after solvent removal. These formulation effects are consistent with the FTIR results (Figure 2), which showed intensified hydrogen bonding between the cellulose hydroxyl groups and carrageenan sulfate groups, leading to reduced matrix shrinkage during film formation. Similar behavior was reported by Dmitrenko *et al.* (2023), who observed that increasing the nanocellulose content in carrageenan films increased the thickness from approximately 0.08 to 0.14 mm owing to the enhanced polymer network density and water retention. Dhilipkumar *et al.* (2025) also reported that carrageenan-rich films, particularly those containing elastic gel-forming fractions, exhibited increased thickness owing to gel swelling during casting.

The tensile strength was significantly influenced by the film formulation, with values ranging from 0.724 to 2.888 MPa (Table 2). All films exceeded the minimum tensile strength requirement specified by the Japanese Industrial Standards (JIS, >0.392 MPa), confirming adequate mechanical integrity for biodegradable packaging materials. Carrageenan-only films (T1 and T2) exhibited the highest tensile strength, which can be attributed to the formation of compact and cohesive gel networks, particularly in κ-carrageenan-rich systems. In contrast, the

Table 2 Mechanical properties of biodegradable films prepared from cocoa pod husk cellulose, κ-, and ι-carrageenan

Code	Crystallinity index (%)	Thickness (mm)	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)
T1	10.18	0.165±0.057^{c}	2.888±0.313^{a}	6.487±2.085^{b}	0.472±0.126^{a}
T2	0.92	0.307±0.256^{bc}	2.529±1.681^{ab}	15.704±6.732^{a}	0.234±0.263^{a}
T3	44.69	0.537±0.071^{ab}	1.251±0.343^{bc}	2.995±3.147^{b}	0.444±0.163^{a}
T4	40.76	0.560±0.066^{ab}	0.838±0.204^{c}	1.953±0.919^{b}	0.499±0.239^{a}
T5	43.93	0.515±0.038^{ab}	0.724±0.296^{c}	1.831±0.987^{b}	0.531±0.381^{a}
T6	31.50	0.432±0.208^{ab}	1.643±0.304^{abc}	7.592±2.320^{b}	0.226±0.059^{a}
T7	25.09	0.618±0.145^{a}	0.922±0.672^{c}	6.591±2.859^{b}	0.142±0.073^{a}
T8	34.10	0.472±0.019^{ab}	1.586±0.247^{abc}	7.093±3.023^{b}	0.259±0.139^{a}

Values are expressed as mean ± standard deviation (n = 3).

Different letters in the same column indicate significant differences (p<0.05)

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cellulose-containing films (T3–T8) exhibited lower tensile strength despite their higher crystallinity indices (Table 2), indicating that the addition of cellulose did not act as an effective reinforcing phase in this system. This behavior suggests that the reinforcing effect of cellulose is limited by factors such as dispersion quality and interfacial compatibility with the carrageenan matrix, causing the cellulose domains to function as stress concentrators rather than load-bearing elements. Similar reductions in tensile strength upon cellulose incorporation have been reported in polysaccharide-based films when cellulose is present as micro-scale fillers without surface modification (Asadzadeh *et al*., 2023; Ramadas *et al*., 2024). Overall, these results demonstrate that the tensile strength of the present films is governed by a balance between gel network rigidity and matrix continuity, rather than by crystallinity alone.

The elongation of the biodegradable film exhibited significant differences among the formulations, ranging from 1.831% to 15.704%. According to the JIS classification, elongation values below 10% are considered poor, whereas values between 10-50% are categorized as moderately good (Japanese Standard Association, 2019). Based on this criterion, only T2 achieved moderately good elongation performance, whereas all other formulations remained within the poor-elongation category. The superior elongation of T2 can be attributed to the dominant contribution of ι-carrageenan, which promotes the formation of elastic gels and enhances polymer chain mobility. While cellulose and κ-carrageenan increased rigidity through crystalline reinforcement and compact gel networks, ι-carrageenan disrupted ordered cellulose packing and increased the amorphous fraction, as confirmed by the lower crystallinity index obtained from X-ray diffraction analysis (Table 2). FTIR results further supported the presence of intermolecular interactions between cellulose and carrageenan chains, which may have contributed to changes in polymer organization and increased film extensibility. Dhilipkumar *et al.* (2025) reported elongation values exceeding 15% for carrageenan-based films dominated by elastic gel structures, whereas an increase in cellulose content reduced elongation due to restricted segmental mobility. Cheng *et al.* (2022) also highlighted that ι-carrageenan-rich systems consistently exhibit higher elongation than κ-carrageenan films because of their softer and more flexible gel networks. From an application standpoint, T2 was the most suitable formulation among the tested samples for applications requiring moderate flexibility.

Young's modulus did not differ significantly among the formulations ($p > 0.05$), indicating that variations in cellulose, κ-carrageenan, and ι-carrageenan ratios did not lead to statistically distinguishable differences in film stiffness. This result suggests that, within the formulation range studied, Young's modulus was not primarily governed by the polymer composition alone. In hydrophilic polysaccharide films, stiffness is often more strongly influenced by residual moisture and plasticizer effects than by compositional changes. Bound water acts as an internal plasticizer, reducing the intermolecular cohesion and homogenizing the elastic response across the samples. Eslami *et al.* (2023) showed that variations in the water and plasticizer contents can substantially reduce Young's modulus and mask the compositional effects in polysaccharide-based films. From an application perspective, the films can be considered moderately flexible materials suitable for packaging or coating applications, where uniform elasticity is preferred over high stiffness.

Barrier Properties

The swelling of the biodegradable films varied significantly among the formulations ($p<0.05$), with values ranging from 159.50% to 473.23% (Table 3). T2 exhibited the highest swelling, followed by T1 and T8, whereas T3 exhibited the lowest value. The pronounced swelling in carrageenan-rich formulations can be attributed mainly to the presence of ι-carrageenan, which forms highly hydrated and elastic networks owing to its flexible backbone and high sulfate group density, as also reported by Ramadas *et al.* (2024) and Wang *et al.* (2023). Conversely, the limited swelling observed in T3 indicates a stronger

Table 3 Swelling and WVTR values of biodegradable films prepared from cocoa pod husk cellulose, κ-, and ι-carrageenan

Code	Swelling (%)	WVTR (g/m2/24h)
T1	356.49±44.32^{b}	4.48±2.68^{ab}
T2	473.23±79.95^{a}	5.35±1.01^{a}
T3	159.50±12.19^{f}	1.14±1.05^{b}
T4	213.51±2.58^{ef}	2.13±1.69^{ab}
T5	226.48±1.57^{de}	3.86±2.30^{ab}
T6	252.02±19.77^{de}	5.43±2.75^{a}
T7	288.99±7.81^{cd}	4.93±1.88^{ab}
T8	348.39±24.61^{bc}	5.36±1.94^{a}

Values are expressed as mean ± standard deviation (n = 3).
Different letters in the same column indicate significant differences (p<0.05)

structural contribution from cellulose, whose hydrogen-bonded and partially ordered domains restrict chain mobility and suppress excessive volumetric expansion in aqueous media (Grzybek *et al.*, 2024). Formulations T4–T7 exhibited intermediate swelling behavior, suggesting a more effective balance between cellulose reinforcement and carrageenan gelation. This interpretation is supported by FTIR and XRD analyses (Figure 2 and 3), which reveal abundant hydrophilic functional groups and mixed amorphous–crystalline structures governing water uptake. In practical terms, formulations with high swelling may be advantageous for applications requiring rapid hydration or disintegration, whereas lower-swelling films provide improved dimensional stability under moist conditions.

The WVTR values of the films ranged from 1.14 to 5.43 g/m²/24h (Table 3), confirming that water vapor transport was significantly affected by the formulation (p<0.05). The highest WVTR was observed for T6, T8, and T2, whereas lower values were recorded for T3. These results suggest that the vapor permeability is influenced by the structural arrangement and compactness of the polymer matrix. Films containing higher cellulose proportions tended to exhibit lower WVTR values. In contrast, formulations containing higher carrageenan fractions showed relatively higher WVTR, which may be associated with the hydrophilic nature of carrageenan. Similar effects of polysaccharide composition and amorphous structure on WVTR have been reported for cellulose- and carrageenan-based biodegradable films (Argel-Pérez *et al.*, 2024; Wang *et al.*, 2023).

The WVTR behavior was also consistent with the crystalline index results (Table 2), corresponding to the lowest WVTR value, indicating that a higher structural order reduced vapor diffusion through the film matrix. Conversely, films with lower crystallinity tended to exhibit higher WVTR owing to the presence of more amorphous regions that facilitate water vapor transport. FTIR analysis (Figure 2) further suggested intermolecular interactions between the cellulose and carrageenan chains, which may contribute to the differences in structural organization and barrier performance among the formulations. Low-WVTR films are potentially suitable for moisture-sensitive products, whereas high-WVTR formulations may be advantageous for applications requiring controlled moisture exchange, such as fresh produce packaging.

CONCLUSION

Biodegradable films based on cocoa

pod husk-derived cellulose combined with κ- and ι-carrageenan were successfully prepared in this study. The film characteristics were primarily determined by the relative contributions of cellulose, κ-carrageenan, and ι-carrageenan. Cellulose improved the water vapor barrier properties of the films, κ-carrageenan contributed to higher tensile strength and rigidity, and ι-carrageenan promoted greater flexibility accompanied by higher water absorption. These results confirm that incorporating cocoa pod husk-derived cellulose with κ- and ι-carrageenan is a promising strategy for developing biodegradable films with improved functional properties for sustainable food packaging applications.

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