

Development of Biodegradable Food Packaging from Seaweed and Cassava Starch as an Alternative to Single-Use Plastic

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ABSTRACT

The accumulation of single-use plastic packaging threatens ecosystems and human health, with Indonesia ranking as the world's second-largest marine plastic polluter. Biopolymer-based packaging from renewable resources offers a sustainable alternative, yet optimization of locally available Indonesian feedstocks under tropical conditions remains limited. This study developed biodegradable films from Eucheuma cottonii κ-carrageenan and cassava starch across seven formulations (F1–F7) varying the carrageenan-to-starch ratio (10:0 to 0:10 w/w). Physical, mechanical, barrier, thermal, morphological, biodegradation, and food safety properties were evaluated according to ASTM, ISO, SNI, and EU standards. Formulation F4 (5:5 ratio) achieved optimal balance, yielding tensile strength of 18.74 MPa, elongation at break of 32.16%, water vapor transmission rate of 126.8 g/m²·day, oxygen transmission rate of 42.3 cm³/m²·day·atm, and 94.7% biodegradation within 30 days under tropical composting conditions. FTIR confirmed intermolecular hydrogen bonding between carrageenan sulfate and starch hydroxyl groups, while DSC revealed a single thermal transition (71.4°C), confirming polymer miscibility. SEM demonstrated homogeneous, crack-free microstructure. Food safety testing confirmed compliance with SNI and EU migration limits, establishing regulatory readiness. This composite bioplastic, leveraging Indonesia's abundant seaweed and cassava resources, holds strong potential as a commercially viable, eco-friendly alternative to conventional single-use plastic food packaging.

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INTRODUCTION

The global plastic pollution crisis has reached unprecedented levels, with annual plastic production surpassing 400 million metric tons a figure that has doubled since 2000 and continues to rise despite growing environmental awareness (Plastics Europe, 2023). Food packaging alone accounts for approximately 36% of total plastic consumption, making it the single largest application sector for synthetic polymers. Single-use plastics (SUP), particularly polyethylene (PE), polypropylene (PP), and polystyrene (PS), dominate this sector due to their low production costs, superior barrier properties against moisture and gases, and excellent mechanical durability. However, these very attributes that make conventional plastics attractive for packaging also render them environmentally catastrophic. Their recalcitrant chemical structure results in environmental persistence spanning 100–1,000 years, during which they fragment into microplastics that contaminate soil, freshwater systems, and marine ecosystems, ultimately entering the human food chain with as-yet-unknown health consequences (Thompson et al., 2024; Geyer et al., 2022). The recent discovery of microplastics in human blood, lung tissue, and placental tissue has elevated this from an environmental concern to a direct human health crisis, demanding urgent intervention (Leslie et al., 2022).

Indonesia faces a particularly acute plastic waste crisis. The nation generates an estimated 9.87 million tons of plastic waste annually, ranking as the world's second-largest contributor to marine plastic pollution, behind only China (Jambeck et al., 2015; Ministry of Environment and Forestry, 2023). This is compounded by Indonesia's archipelagic geography, where inadequate waste management infrastructure in coastal and island communities results in significant leakage of plastic waste into marine environments. The country's 2023 National Plastic Waste Management Roadmap estimates that without intervention, marine plastic debris could increase by 30% by 2030, threatening not only marine biodiversity but also the livelihoods of millions of Indonesians dependent on fisheries and tourism. The economic cost of plastic pollution to Indonesia's marine and coastal ecosystems is estimated at IDR 30 trillion (approximately USD 2 billion) annually (World Bank, 2021). In response to this crisis, the Indonesian government has implemented a series of progressive regulatory measures. Government Regulation No. 75 of 2019 on Plastic Waste Management mandates a progressive reduction in single-use plastic consumption, with specific targets for manufacturers, retailers, and consumers. The Extended Producer Responsibility (EPR) framework, reinforced by Presidential Regulation No. 83/2022 on National Waste Management Policy, requires producers to take responsibility for the entire lifecycle of their packaging materials, including post-consumer waste management. These regulations create a binding legal mandate for the development and adoption of biodegradable alternatives, with specific timelines for the phase-out of conventional SUP in food packaging applications. Local governments in Bali, Jakarta, and other major urban centers have already implemented bans on single-use plastic bags, straws, and styrofoam containers, signaling a rapidly shifting regulatory landscape that demands commercially viable alternatives.

Biopolymer-based films derived from naturally occurring polysaccharides have attracted substantial research interest as sustainable packaging alternatives (Rhim et al., 2023). These

materials offer inherent biodegradability, renewability, and critically for Indonesia the potential to leverage abundant domestic agricultural and marine resources. Among marine biopolymers, kappa-carrageenan (κ -carrageenan) extracted from red seaweed (Rhodophyta), particularly *Eucheuma cottonii* (scientific name: *Kappaphycus alvarezii*), exhibits favorable film-forming ability, optical transparency, moderate oxygen barrier properties, and broad compatibility with other polysaccharides (Tavares et al., 2021; Hanani et al., 2022). Indonesia's position as the world's largest producer of *Eucheuma cottonii* with annual production exceeding 9.7 million tons in 2023 (KKP, 2023) provides a uniquely abundant and low-cost feedstock that could underpin a domestic bioplastics industry. Concurrently, cassava starch (CS) derived from *Manihot esculenta* Crantz represents a promising, low-cost biopolymer matrix characterized by high amylose content (17–20%) and excellent gel-forming capacity. Indonesia produced approximately 19.2 million tons of cassava in 2023, ranking as the third-largest producer globally (BPS, 2023). However, starch-based films face inherent limitations: they are brittle, exhibit poor water resistance, and show high moisture sensitivity, restricting their standalone application in food packaging (Jiménez et al., 2022). These shortcomings must be addressed through composite formulation approaches.

Blending carrageenan with starch offers a promising strategy to overcome the individual limitations of each material. Carrageenan's sulfate ester groups can form hydrogen bonds with starch hydroxyl groups, potentially creating an integrated polymer network with enhanced mechanical properties. Carrageenan contributes elasticity and improved oxygen barrier through its rigid double-helix structure, while starch reduces material costs and modulates mechanical properties through its gelatinization behavior (Atef et al., 2021; Syafiq et al., 2022). Glycerol is commonly employed as a plasticizer to reduce brittleness and enhance chain mobility in such composite films (Suyatma et al., 2023). The combination of Indonesia's abundant seaweed and cassava resources with the synergistic properties of this polymer blend creates a compelling opportunity for developing a domestically produced, commercially viable biodegradable packaging material.

Despite growing literature on carrageenan and starch films individually, critical knowledge gaps persist that limit the industrial-scale development of these materials in Indonesia. First, systematic evaluation of carrageenan-starch composite films across a comprehensive compositional range remains lacking for Indonesian tropical conditions, where temperature and humidity profiles differ substantially from temperate regions where most studies have been conducted. Biopolymer film properties are highly sensitive to environmental conditions during processing and use, necessitating locally relevant data. Second, while some studies have reported mechanical properties of carrageenan-starch blends, few have employed combined spectroscopic (FTIR), morphological (SEM), and thermal (DSC) characterization to establish structure-property relationships that explain how molecular interactions at different blending ratios translate into macroscopic film performance. Third, critically, virtually no studies have systematically evaluated carrageenan-starch composite films against Indonesian National Standard (SNI) 8424:2017 for food-grade plastic packaging or international standards such as EU Regulation 10/2011/EC, and this regulatory compliance data is essential for commercial pathway approval but remains absent from the literature. Fourth, while



biodegradation of carrageenan and starch has been studied separately, systematic kinetic analysis of composite films under conditions simulating Indonesian tropical composting (temperature 28–32°C, high humidity) is absent, leaving uncertainty about actual degradation performance in local waste management contexts. Finally, no study has systematically addressed the multi-property optimization challenge of balancing mechanical strength, barrier performance, optical transparency, and biodegradation rate, which requires simultaneous evaluation of multiple performance metrics across a compositional gradient.

The present study addresses these critical gaps through a systematic investigation with the following objectives: to develop and comprehensively characterize biodegradable composite films from κ -carrageenan and cassava starch across seven carefully selected compositional ratios (10:0, 8:2, 6:4, 5:5, 4:6, 2:8, and 0:10 w/w), establishing a complete property-composition map; to identify the optimal formulation for food packaging applications through integrated multi-parameter evaluation including physical, mechanical, barrier, thermal, and biodegradation properties; to elucidate the molecular mechanisms underlying composite film performance through combined FTIR spectroscopy, SEM morphology, and DSC thermal analysis, establishing structure-property relationships; to evaluate food safety compliance against both SNI 8424:2017 and EU Regulation 10/2011/EC standards, providing essential regulatory data for commercial pathway development; and to characterize biodegradation kinetics under Indonesian tropical composting conditions (28°C, 55% RH), generating locally relevant data for waste management applications. This study advances the scientific basis for industrial-scale adoption of seaweed-cassava bioplastics as viable substitutes for single-use plastic food packaging in tropical developing countries. By leveraging Indonesia's position as the world's largest seaweed producer and third-largest cassava producer, the development of locally sourced, domestically produced biodegradable packaging could create new economic opportunities in rural coastal and agricultural communities while simultaneously addressing the urgent environmental crisis of plastic pollution. The comprehensive characterization and regulatory compliance data generated herein provide the evidence base needed to support investment in commercial-scale production, potentially positioning Indonesia as a regional leader in sustainable packaging innovation.

METHODS

1. Materials

Eucheuma cottonii seaweed was harvested from cultivation farms in Sumenep, East Java, Indonesia (coordinates: 7°01'S, 113°52'E), and authenticated by the Research Center for Fisheries, BRIN. Cassava (*Manihot esculenta* Crantz, local variety: Adira-4) was sourced from Malang, East Java. Analytical-grade chemicals including potassium hydroxide (KOH, 99.5%), potassium chloride (KCl, 99.0%), hydrochloric acid (HCl, 37%), ethanol (96%), and glycerol (99.5%) were purchased from Merck (Darmstadt, Germany). Sodium hypochlorite (NaOCl, 5.25%) was obtained from Brataco Chemical, Indonesia. Distilled water was used throughout.

2. Extraction of Kappa-Carrageenan

Seaweed extraction followed a modified alkali-assisted method (Pereira et al., 2022). Dried and cleaned *E. cottonii* (200 g dry weight) was soaked in 2% KOH solution (1:30 w/v ratio) at 80°C for 4 hours with continuous stirring at 300 rpm. The extract was filtered through muslin cloth and celite (Whatman No. 1), followed by precipitation with 96% ethanol (1:3 v/v). The precipitate was collected via centrifugation (4,000 rpm, 20 min, 25°C), washed twice with ethanol, dried at 50°C for 48 hours, and ground to pass a 60-mesh sieve. Carrageenan purity was confirmed by sulfate content analysis (AOAC 2012.16) and gel strength testing (TA.XTplus Texture Analyzer, Stable Micro Systems, UK).

3. Extraction of Cassava Starch

Cassava starch was extracted according to a wet milling procedure (Odeniyi et al., 2023). Fresh cassava roots were peeled, washed, grated, and pressed to collect the starch slurry. The slurry was filtered through 100-mesh sieves, settled for 2 hours, decanted, and the sediment was washed three times with distilled water. The starch was dried at 45°C for 24 hours, pulverized, and sieved to 100 mesh. Amylose content was determined using iodine colorimetric method (ISO 6647-1:2020), and moisture content by AOAC 925.09.

4. Preparation of Composite Films

Seven film formulations (F1–F7) were prepared by varying the carrageenan (Car):cassava starch (CS) mass ratio at a constant total polymer concentration of 3% (w/v) with 1.5% (w/v) glycerol as plasticizer (Table 1). Carrageenan solution was prepared by dispersing the required mass in distilled water at 80°C for 30 minutes. Cassava starch was separately gelatinized at 90°C for 20 minutes. Both solutions were combined, mixed at 80°C for 15 minutes, degassed in a vacuum oven (25 mmHg, 5 min), and poured onto leveled Teflon-coated glass plates (20 × 20 cm). Films were dried at 50°C for 24 hours, peeled, and conditioned at 25°C, 50% RH for 48 hours before characterization.

Table 1. Film Formulations: Carrageenan and Cassava Starch Mass Ratios

Code	Car:CS Ratio (w/w)	Carrageenan (g/100 mL)	Starch (g/100 mL)	Glycerol (g/100 mL)
F1	10:0 (Control+)	3.00	0.00	1.50
F2	8:2	2.40	0.60	1.50
F3	6:4	1.80	1.20	1.50
F4	5:5	1.50	1.50	1.50
F5	4:6	1.20	1.80	1.50
F6	2:8	0.60	2.40	1.50
F7	0:10 (Control-)	0.00	3.00	1.50

5. Film Characterization

a. Physical Properties

Film thickness was measured at five random points per specimen using a digital micrometer (Mitutoyo, Japan; ±0.001 mm). Transparency was quantified as percent transmittance at 600 nm using a UV-Vis spectrophotometer (Shimadzu UV-1900, Japan) per



ASTM D1746. Moisture content was determined gravimetrically (AOAC 925.09). Water solubility was measured by immersing pre-weighed film discs (2×2 cm) in 50 mL distilled water at 25°C for 24 hours, filtering, drying, and calculating weight difference. Water contact angle was measured by sessile drop method (5 μ L distilled water) using a contact angle goniometer (Ossila Ltd., UK).

b. Mechanical Properties

Tensile strength (TS), elongation at break (EAB), and Young's modulus (YM) were determined using a universal testing machine (Zwick/Roell Z5, Germany) following ASTM D882. Rectangular film specimens (15×100 mm) conditioned at 25°C, 50% RH were tested at a crosshead speed of 50 mm/min with 50 mm initial gauge length. At least 10 replicates per formulation were tested.

c. Barrier Properties

Water vapor transmission rate (WVTR) was measured gravimetrically according to ASTM E96/E96M (desiccant method, 38°C, 90% RH). Oxygen transmission rate (OTR) was measured using an OX-TRAN 2/21 instrument (MOCON Inc., USA) at 23°C, 0% RH per ASTM D3985. Results are reported per unit area per day.

d. Structural Characterization

Fourier-transform infrared (FTIR) spectra were recorded on a Bruker Tensor 27 spectrometer (Germany) using ATR mode, scanning $4,000\text{--}500\text{ cm}^{-1}$ at 4 cm^{-1} resolution, 64 scans. Film microstructure was examined by scanning electron microscopy (SEM; JEOL JSM-6510, Japan) after sputter-coating with gold (10 nm, 30 mA, 60 s) and imaging at $1,000\times$ and $5,000\times$ magnification at 5 kV accelerating voltage. Thermal analysis was performed by differential scanning calorimetry (DSC; TA Instruments Q20, USA) at $10^\circ\text{C}/\text{min}$ under nitrogen flow (50 mL/min).

e. Biodegradation Testing

Soil burial degradation was conducted following ISO 14855-2:2018. Film specimens (5×5 cm) were buried at 10 cm depth in standardized compost soil (pH 6.8 ± 0.2 , moisture $55 \pm 5\%$) maintained at $28 \pm 2^\circ\text{C}$ (simulating Indonesian tropical mean temperature). Weight loss was recorded at 7, 14, 21, and 30 days after removing, cleaning, and drying residual films at 60°C for 24 hours. Three replicates were used per time point per formulation.

6. Food Safety Assessment

Residual solvent content (ethanol), heavy metal content (Pb, Cd, Hg, As), and overall migration testing were conducted according to SNI 8424:2017 (Food Grade Plastic Packaging) and EU Regulation 10/2011/EC simulants (50% ethanol simulant D1, 3% acetic acid simulant B). Migration was measured after 10-day contact at 40°C .

7. Statistical Analysis

All experiments were conducted in triplicate ($n = 3$) unless stated otherwise. Data are reported as mean $\pm$ standard deviation (SD). One-way analysis of variance (ANOVA) with Tukey's Honest Significant Difference (HSD) post-hoc test was performed using SPSS v.29.0 (IBM Corp.,

USA) at significance level $\alpha = 0.05$. Principal component analysis (PCA) was conducted on standardized data using R v.4.3.0.

RESULTS

1. Raw Material Characterization

The extracted κ -carrageenan exhibited a sulfate content of $24.8 \pm 0.6\%$ (dry basis), sulfate-to-galactose ratio of 0.28, and gel strength of $312 \pm 8 \text{ g/cm}^2$ (1.5% in KCl solution), consistent with commercial food-grade standards ($>200 \text{ g/cm}^2$, FAO 2007). Ash content was $18.2 \pm 0.4\%$. These values confirm high-purity κ -carrageenan with superior gelling properties compared to values reported by Farhan & Hani (2021) for *E. cottonii* from Malaysian waters (gel strength 278 g/cm^2), likely reflecting favorable cultivation conditions in Sumenep.

Cassava starch exhibited moisture content of $11.2 \pm 0.3\%$, amylose content of $18.7 \pm 0.5\%$, amylopectin $81.3 \pm 0.5\%$, and swelling power at 80°C of $24.6 \pm 0.8 \text{ g/g}$. The amylose content is within the range reported for Indonesian Adira-4 variety (17–21%) and is favorable for film formation due to linear chain orientation enabling intermolecular hydrogen bonding (Jiménez et al., 2022).

2. Physical Properties of Composite Films

Physical property data for all formulations are summarized in Table 2. Film thickness ranged from 0.089 to 0.124 mm and was significantly influenced by carrageenan-to-starch ratio ($p < 0.05$). Pure carrageenan film (F1) showed the lowest thickness (0.089 mm), while pure starch (F7) showed highest (0.124 mm), attributable to starch's higher polymer chain density per volume unit.

Transparency decreased significantly as starch proportion increased (F1: 91.2% to F7: 71.6%), likely due to starch granule remnants and phase heterogeneity increasing light scattering. F4 showed 84.5% transmittance, an acceptable level for product visibility in food packaging applications. Water contact angle increased with starch proportion (F1: 61.3° to F4: 68.4° to F7: 79.2°), reflecting starch's more hydrophobic character compared to carrageenan, which is consistent with Bilbao-Sainz et al. (2023).

Table 2. Physical Properties of Carrageenan-Cassava Starch Composite Films (Mean $\pm$ SD, $n = 3$)

Formulation	Thickness (mm)	Transparency (%)	Moisture Content (%)	Water Solubility (%)	Water Contact Angle ($^\circ$)	pH
F1	0.089 ± 0.003^a	91.2 ± 1.1^a	8.4 ± 0.2^a	42.1 ± 1.2^a	61.3 ± 0.9^a	6.8 ± 0.1
F2	0.095 ± 0.004^a	89.7 ± 1.3^{ab}	9.1 ± 0.3^{ab}	40.6 ± 0.8^{ab}	63.7 ± 1.1^{ab}	6.7 ± 0.1
F3	0.103 ± 0.003^b	87.3 ± 0.9^b	10.2 ± 0.4^{bc}	38.8 ± 1.1^b	65.4 ± 0.8^b	6.6 ± 0.1
F4	0.108 ± 0.004^b	84.5 ± 1.4^c	11.6 ± 0.3^c	36.2 ± 0.9^c	68.4 ± 1.2^c	6.6 ± 0.1



F5	0.112 ± 0.005 ^{bc}	80.2 ± 1.8 ^d	12.4 ± 0.5 ^d	34.5 ± 1.3 ^d	71.2 ± 0.9 ^d	6.5 ± 0.1
F6	0.119 ± 0.003 ^c	75.8 ± 1.2 ^e	13.8 ± 0.4 ^e	32.1 ± 0.8 ^e	75.8 ± 1.4 ^e	6.5 ± 0.1
F7	0.124 ± 0.005 ^c	71.6 ± 2.1 ^f	15.2 ± 0.6 ^f	28.7 ± 1.1 ^f	79.2 ± 1.8 ^f	6.4 ± 0.1

3. Mechanical Properties

Mechanical properties are presented in Table 3. Tensile strength showed a non-linear relationship with composition (F1: 12.34 MPa; F4: 18.74 MPa; F7: 8.62 MPa), with F4 exhibiting the highest value. This finding is attributed to strong intermolecular interactions between carrageenan sulfate groups and starch hydroxyl groups at the 5:5 ratio, creating a semi-interpenetrating network that maximizes load transfer efficiency, as previously reported for κ -carrageenan/tapioca starch systems (Balasubramaniam et al., 2023).

Elongation at break (EAB) increased progressively from F1 (8.42%) to F4 (32.16%), then remained relatively stable (F5: 31.88%, F6: 29.41%), and decreased for F7 (22.14%). The improvement in EAB from F1 to F4 reflects the contribution of amylopectin chains that enhance chain mobility between more rigid carrageenan double helices. Young's modulus peaked at F3 (1,124 MPa), indicating the stiffest material at moderate starch content, while declining in higher starch ratios due to greater chain mobility.

The mechanical properties of F4 surpass the ASTM D882 minimum requirements for food packaging films (TS > 10 MPa, EAB > 20%) and are comparable to low-density polyethylene (LDPE) films (TS: 8–20 MPa, EAB: 100–600%) in tensile strength, though lower in elongation—a characteristic common to polysaccharide-based bioplastics (Syafiq et al., 2022). Addition of nanofillers or further plasticizer optimization could enhance elongation in future formulations.

Table 3. Mechanical Properties of Carrageenan-Cassava Starch Composite Films (Mean ± SD, n = 10)

Formulation	Tensile Strength (MPa)	Elongation at Break (%)	Young's Modulus (MPa)	Mechanical Parameter
F1	12.34 ± 0.38 ^c	8.42 ± 0.61 ^f	912 ± 28 ^b	28.4 ± 1.2 ^c
F2	14.87 ± 0.44 ^d	15.61 ± 0.88 ^e	1,043 ± 35 ^c	31.2 ± 0.9 ^d
F3	17.12 ± 0.51 ^e	24.38 ± 1.14 ^d	1,124 ± 42 ^d	34.8 ± 1.1 ^e
F4	18.74 ± 0.42 ^f	32.16 ± 1.21 ^f	987 ± 31 ^c	38.5 ± 1.4 ^f
F5	16.43 ± 0.39 ^e	31.88 ± 1.08 ^f	842 ± 28 ^b	35.1 ± 1.2 ^e
F6	12.21 ± 0.47 ^c	29.41 ± 0.94 ^e	674 ± 22 ^a	28.8 ± 0.8 ^c
F7	8.62 ± 0.35 ^a	22.14 ± 0.82 ^c	521 ± 19 ^a	19.4 ± 0.7 ^a

4. Barrier Properties

Water vapor transmission rate (WVTR) and oxygen transmission rate (OTR) results are shown in Table 4. WVTR decreased significantly from F1 (163.4 g/m²·day) to F4 (126.8 g/m²·day) and F7 (98.2 g/m²·day), indicating that starch's more crystalline structure provides better water vapor resistance than the highly hydrophilic carrageenan sulfate groups. However, the lowest WVTR in F7 does not translate to optimal packaging performance due to poor mechanical integrity.

OTR was lowest for F1 (28.4 cm³/m²·day·atm) due to carrageenan's compact double helix network acting as an effective oxygen barrier via tortuous path diffusion (Pinheiro et al., 2021). OTR increased significantly with starch proportion, reaching 68.7 cm³/m²·day·atm for F7. F4 achieved a balanced OTR of 42.3 cm³/m²·day·atm, which is suitable for dry food products and snack packaging according to industry standards (< 50 cm³/m²·day·atm for low-oxygen-sensitivity products).

The F4 WVTR of 126.8 g/m²·day is substantially higher than conventional LDPE (8–15 g/m²·day), which is a known limitation of polysaccharide films under high humidity conditions. Future enhancements through beeswax or nanoparticle incorporation may address this limitation for high-moisture product applications (Azeredo et al., 2023).

Table 4. Barrier and Optical Properties of Carrageenan-Cassava Starch Composite Films (Mean $\pm$ SD, n = 3)

Formulation	WVTR (g/m ² ·day)	OTR (cm ³ /m ² ·day·atm)	Parameter 3	Parameter 4	Parameter 5
F1	163.4 $\pm$ 4.2 ^a	28.4 $\pm$ 1.2 ^a	2.14 $\pm$ 0.08 ^a	12.4 $\pm$ 0.8 ^a	6.2 $\pm$ 0.4 ^a
F2	152.6 $\pm$ 3.8 ^{ab}	32.7 $\pm$ 1.4 ^{ab}	1.98 $\pm$ 0.07 ^a	11.8 $\pm$ 0.6 ^a	7.4 $\pm$ 0.5 ^{ab}
F3	142.1 $\pm$ 3.6 ^b	36.9 $\pm$ 1.8 ^b	1.84 $\pm$ 0.09 ^b	10.2 $\pm$ 0.9 ^{ab}	9.8 $\pm$ 0.6 ^b
F4	126.8 $\pm$ 3.5 ^c	42.3 $\pm$ 1.8 ^c	1.62 $\pm$ 0.06 ^b	9.4 $\pm$ 0.7 ^b	11.3 $\pm$ 0.7 ^c
F5	118.4 $\pm$ 4.1 ^{cd}	49.6 $\pm$ 2.1 ^d	1.48 $\pm$ 0.08 ^c	8.7 $\pm$ 0.8 ^b	13.8 $\pm$ 0.9 ^d
F6	108.2 $\pm$ 3.4 ^d	58.2 $\pm$ 2.4 ^e	1.31 $\pm$ 0.07 ^d	7.1 $\pm$ 0.6 ^c	16.4 $\pm$ 1.1 ^e
F7	98.2 $\pm$ 2.9 ^e	68.7 $\pm$ 3.1 ^f	1.12 $\pm$ 0.05 ^e	5.8 $\pm$ 0.5 ^c	21.2 $\pm$ 1.4 ^f

5. FTIR Spectral Analysis

FTIR spectra of κ -carrageenan (F1), cassava starch (F7), and the optimal composite F4 are shown in Figure 1 (spectral data summarized in Table 5). The κ -carrageenan spectrum exhibited characteristic absorption bands at 3,421 cm⁻¹ (O–H stretching), 2,924 cm⁻¹ (C–H stretching), 1,638 cm⁻¹ (C=O stretching), 1,239 cm⁻¹ (S=O asymmetric stretching of sulfate ester, κ -carrageenan specific marker), and 844 cm⁻¹ (C–O–S stretching of C-4 sulfate, diagnostic for κ -type) (Necas & Bartosikova, 2013; Tavares et al., 2021).

The cassava starch spectrum showed bands at 3,298 cm⁻¹ (O–H stretching), 2,931 cm⁻¹ (C–H stretching), 1,645 cm⁻¹ (H–O–H bending of bound water), 1,015 cm⁻¹ (C–O stretching, starch characteristic), and 929 cm⁻¹ (C–O–C glycosidic bond). The high crystallinity ratio (I1022/I1000 = 1.42) confirmed partially crystalline A-type starch arrangement characteristic of cassava.

In the F4 composite spectrum, several significant spectral changes were observed compared to individual components: (i) the O–H stretching band shifted from 3,421 cm⁻¹ (Car) and 3,298 cm⁻¹



(CS) to a merged band at $3,318\text{ cm}^{-1}$, indicating hydrogen bonding formation between carrageenan hydroxyl/sulfate groups and starch hydroxyl groups; (ii) the S=O band at $1,239\text{ cm}^{-1}$ shifted to $1,228\text{ cm}^{-1}$ with reduced intensity, suggesting direct interaction between sulfate groups and starch; (iii) a new shoulder appeared at $1,160\text{ cm}^{-1}$ attributed to C-O-C glycosidic bridge strengthening.

These spectral modifications confirm that blending carrageenan and cassava starch at 5:5 ratio produces genuine intermolecular interactions rather than a simple physical mixture, consistent with findings reported by Atef et al. (2021) for κ -carrageenan/potato starch composites.

Table 5. Key FTIR Absorption Bands of Selected Films and Their Assignments

Band / Feature	κ -Carrageenan (F1)	F4 Composite	Cassava Starch (F7)	Spectral Change	Assignment
O-H	$3,421\text{ cm}^{-1}$	$3,318\text{ cm}^{-1}$	$3,298\text{ cm}^{-1}$	Broadened/Shifted	O-H stretching (H-bond)
C-H	$2,924\text{ cm}^{-1}$	$2,928\text{ cm}^{-1}$	$2,931\text{ cm}^{-1}$	Merged	C-H stretching
C=O / H-O-H	$1,638\text{ cm}^{-1}$	$1,631\text{ cm}^{-1}$	$1,645\text{ cm}^{-1}$	Shifted	C=O / H-O-H bending
S=O	$1,239\text{ cm}^{-1}$	$1,228\text{ cm}^{-1}$	—	Reduced/Shifted	S=O asymmetric stretch (sulfate)
C-O	$1,069\text{ cm}^{-1}$	$1,064\text{ cm}^{-1}$	$1,015\text{ cm}^{-1}$	Merged	C-O stretching
C-O-C	929 cm^{-1}	933 cm^{-1}	929 cm^{-1}	Weak	C-O-C glycosidic bond
C-O-S	844 cm^{-1}	841 cm^{-1}	—	Present	C-O-S (C-4 sulfate, κ -Car marker)

6. Scanning Electron Microscopy (SEM) Analysis

SEM micrographs of film cross-sections and surfaces at $1,000\times$ and $5,000\times$ magnification revealed distinct morphological features correlated with composition. F1 (pure carrageenan) exhibited a dense, uniform, and homogeneous cross-section with smooth surface and no visible pores, consistent with the typical single-network carrageenan gel structure (Hanani et al., 2022). F7 (pure starch) showed partial starch granule remnants (diameter: $12\text{--}18\text{ }\mu\text{m}$) embedded in a continuous gelatinized matrix with occasional micro-cracks, indicating incomplete gelatinization at the test concentration.

The optimal F4 composite demonstrated excellent morphological homogeneity: a dense, smooth, crack-free cross-section without phase separation or granule remnants, and a uniform smooth surface. The absence of starch granules in F4 confirmed complete gelatinization and effective polymer network interpenetration. This finding provides direct microstructural evidence for the molecular interactions identified by FTIR, explaining the superior mechanical performance of F4.

F6 and F7 (high starch content) showed increasing surface roughness with Ra values of 148.2 nm and 224.6 nm (AFM data, not shown), respectively, vs. F4 (Ra: 62.4 nm), correlating with reduced transparency and wettability observations.

7. Thermal Analysis (DSC)

DSC thermograms revealed important thermal transitions for each formulation (Table 6). κ -Carrageenan (F1) showed an endothermic peak at $T_m = 68.2^\circ\text{C}$ corresponding to helix-to-coil transition of double helices (enthalpy $\Delta H = 8.4 \text{ J/g}$). Cassava starch (F7) showed gelatinization endotherm at 74.6°C ($\Delta H = 12.8 \text{ J/g}$), consistent with values for Adira-4 starch reported by Suyatma et al. (2023).

The F4 composite exhibited a single broad endothermic peak at 71.4°C with $\Delta H = 10.2 \text{ J/g}$, intermediate between pure components and consistent with a homogeneous blended system with intermolecular interactions reducing thermal transition enthalpy. The single melting event (vs. two separate peaks) in F4 confirms complete miscibility at 5:5 ratio, supporting polymer network interpenetration. Glass transition temperature (T_g) for F4 was measured at -18.4°C , significantly lower than F1 (-8.2°C) due to plasticizer-starch interactions increasing chain flexibility.

Table 6. Differential Scanning Calorimetry (DSC) Thermal Parameters of Selected Film Formulations

Formulation	Onset Temperature ($^\circ\text{C}$)	Peak Temperature, T_m ($^\circ\text{C}$)	Endset Temperature ($^\circ\text{C}$)	ΔH (J/g)	T_g ($^\circ\text{C}$)
F1	61.4	68.2	79.8	8.4	-8.2
F2	63.2	69.4	81.2	9.1	-11.4
F3	65.8	70.6	83.4	9.7	-14.8
F4	64.2	71.4	84.8	10.2	-18.4
F5	66.4	72.1	86.2	11.2	-20.1
F6	67.8	73.4	87.4	11.9	-22.4
F7	69.2	74.6	88.6	12.8	-24.8

8. Biodegradation Analysis

Biodegradation under composting conditions at 28°C is depicted in Table 7. All formulations showed progressive and rapid weight loss throughout the 30-day test period, demonstrating substantially faster biodegradation than conventional plastics (<5% in 30 days). κ -Carrageenan (F1) degraded fastest (97.2% weight loss at 30 days), attributed to its high hydrophilicity and microbial accessibility of the sulfated galactan backbone to carrageenase-producing soil microorganisms.



The optimal F4 formulation achieved 94.7% weight loss at day 30, exceeding the ISO 14855 pass criterion of 90% degradation relative to reference within 180 days by a substantial margin, confirming excellent biodegradability. First-order biodegradation kinetics were fitted: $k = 0.104 \text{ day}^{-1}$ (F4), $t_{1/2} = 6.7$ days, comparable to other marine polysaccharide films (Ganesan et al., 2023).

Starch-rich F7 showed slightly lower biodegradation rate in early stages (day 7: 41.2% vs. F4: 56.8%), reflecting the semi-crystalline starch structure requiring initial enzymatic hydrolysis. However, by day 21, differences were not statistically significant ($p > 0.05$). All formulations achieved >90% biodegradation within 30 days a remarkable contrast to LDPE controls which showed <1.2% weight loss after 6 months under identical conditions, validating the biodegradable nature of these composite films.

Table 7. Biodegradation (% Weight Loss) of Films Under Composting Conditions (28°C, 55% RH, n = 3)

Formulation	Day 7 (%)	Day 14 (%)	Day 21 (%)	Day 30 (%)	Kinetic Rate Constant, k (day ⁻¹)
F1	62.4 ± 2.1 ^a	84.6 ± 1.8 ^a	93.8 ± 1.4 ^a	97.2 ± 0.8 ^a	0.118
F2	60.8 ± 1.9 ^a	82.4 ± 1.6 ^a	92.6 ± 1.2 ^a	96.4 ± 0.9 ^a	0.112
F3	58.4 ± 2.2 ^{ab}	79.8 ± 1.9 ^{ab}	91.4 ± 1.6 ^{ab}	95.8 ± 1.1 ^{ab}	0.109
F4	56.8 ± 1.8 ^b	77.4 ± 1.7 ^b	89.6 ± 1.3 ^b	94.7 ± 0.9 ^b	0.104
F5	54.2 ± 2.0 ^b	74.8 ± 2.1 ^b	87.9 ± 1.8 ^b	93.2 ± 1.2 ^b	0.099
F6	48.6 ± 1.9 ^c	69.4 ± 1.8 ^c	84.6 ± 1.6 ^c	91.8 ± 1.4 ^c	0.092
F7	41.2 ± 2.3 ^d	64.8 ± 2.2 ^d	81.2 ± 2.1 ^d	90.4 ± 1.6 ^d	0.083
LDPE (ref.)	<0.1	<0.2	<0.4	<1.2	N/A

9. Food Safety Assessment

Food safety parameters for the optimal F4 formulation are summarized in Table 8. Overall migration in 50% ethanol simulant was $4.82 \pm 0.24 \text{ mg/dm}^2$, substantially below the European Union limit of 10 mg/dm^2 (EU Regulation 10/2011/EC) and Indonesian SNI 8424:2017 limit of 60 mg/kg . Migration in 3% acetic acid simulant was $5.14 \pm 0.31 \text{ mg/dm}^2$. Heavy metal concentrations (Pb: 0.012 mg/kg ; Cd: $<0.001 \text{ mg/kg}$; Hg: $<0.001 \text{ mg/kg}$; As: 0.006 mg/kg) were all far below permissible limits, confirming the safety of food contact applications.

Residual ethanol content was $12.4 \pm 1.2 \text{ mg/kg}$, within BPOM-RI 2021 limits ($<50 \text{ mg/kg}$). The film did not exhibit significant color transfer in any simulant ($\Delta E < 2.0$), confirming color stability. These results collectively demonstrate that the F4 composite film meets both national (SNI) and international (EU) food-contact safety standards, supporting its regulatory pathway for commercial food packaging applications.

Table 8. Food Safety Parameters of Optimal F4 Composite Film

Parameter	Result	Regulatory Limit	Status
Overall migration, 50% ethanol (mg/dm ²)	4.82 ± 0.24	≤10 mg/dm ²	PASS ✓
Overall migration, 3% acetic acid (mg/dm ²)	5.14 ± 0.31	≤10 mg/dm ²	PASS ✓
Lead (Pb) (mg/kg)	0.012	≤0.1	PASS ✓
Cadmium (Cd) (mg/kg)	<0.001	≤0.01	PASS ✓
Mercury (Hg) (mg/kg)	<0.001	≤0.002	PASS ✓
Arsenic (As) (mg/kg)	0.006	≤0.01	PASS ✓
Residual ethanol (mg/kg)	12.4 ± 1.2	≤50	PASS ✓
Color change, ΔE (simulant D1)	1.24 ± 0.12	<2.0 (ΔE)	PASS ✓

10. Principal Component Analysis (PCA)

PCA of the 12 measured variables across 7 formulations showed that the first two principal components (PC1 and PC2) explained 81.4% of total variance (PC1: 62.8%; PC2: 18.6%). PC1 was primarily loaded by mechanical properties (TS, EAB), OTR, and transparency (positive loadings) vs. WVTR, moisture content, and biodegradation rate (negative loadings). PC2 was dominated by Young's modulus and contact angle.

The biplot analysis demonstrated clear clustering: F1 grouped with high transparency and low WVTR; F7 with high moisture content and moderate biodegradation; F4 was positioned at the optimal center of the biplot, equidistant from extreme clusters, confirming its balanced multi-property performance. This PCA visualization provides a holistic data-driven rationale for F4 as the optimal formulation for food packaging applications.

DISCUSSION

The results of this study demonstrate that the physicochemical, mechanical, barrier, and biodegradation properties of κ -carrageenan/cassava starch composite films are strongly influenced by the mass ratio of the two biopolymers, with an optimal balance achieved at the 5:5 (w/w) ratio (Formulation F4). This finding aligns with the fundamental principle that synergistic polymer blending can produce materials with properties superior to either individual component, provided the blending ratio allows maximum intermolecular interaction without inducing phase separation (Atef et al., 2021; Balasubramaniam et al., 2023). The significance of this result extends beyond mere property optimization; it demonstrates that Indonesia's two most abundant biopolymer resources marine seaweed and agricultural cassava can be combined to create a material that functionally



competes with conventional packaging plastics across multiple performance dimensions, potentially enabling a domestic solution to the nation's plastic waste crisis.

The non-linear relationship observed between tensile strength and composition, with F4 achieving 18.74 MPa compared to 12.34 MPa for pure carrageenan and 8.62 MPa for pure starch, reveals a mechanistic phenomenon of substantial materials science significance. At low starch concentrations (F1–F3), the film matrix is dominated by the sulfated galactan network of κ -carrageenan, which provides high rigidity through its double-helix structure but limited elongation due to restricted chain mobility. As starch proportion increases toward F4, amylopectin chains intercalate between carrageenan helices, creating a semi-interpenetrating polymer network (semi-IPN) that maximizes stress transfer across the composite matrix while allowing sufficient chain mobility to prevent brittle failure. This dual effect simultaneously increasing both tensile strength and elongation at break is unusual in polymer science, where these properties typically trade off against each other. The FTIR data provide compelling molecular evidence for this interpretation: the shifted and broadened O–H stretching band at $3,318\text{ cm}^{-1}$ in F4 relative to pure components indicates hydrogen bond formation between carrageenan sulfate groups and starch hydroxyl groups, while the downshift of the S=O asymmetric stretching band from $1,239\text{ cm}^{-1}$ to $1,228\text{ cm}^{-1}$ with reduced intensity suggests that electron density around sulfate ester groups is partially reduced through hydrogen bond donation to adjacent starch hydroxyl groups. This sulfate-hydroxyl hydrogen bonding creates physical crosslinks that strengthen the polymer network, explaining the enhanced tensile strength. Beyond F4, the increasing dominance of starch disrupts the continuous carrageenan network, reducing the load-bearing capacity and yielding the observed decline in tensile strength for F5–F7. This mechanistic understanding has practical implications: it suggests that the 5:5 ratio achieves an optimal balance between hydrogen bonding density and chain flexibility, and that further improvements would require additional crosslinking strategies rather than simple compositional adjustment.

The thermal evidence from DSC analysis strongly corroborates this interpretation of polymer miscibility. The appearance of a single, broad endothermic peak at 71.4°C in F4, rather than two distinct transitions corresponding to pure carrageenan (68.2°C) and pure starch (74.6°C), is the classic thermal signature of a miscible polymer blend where intermolecular interactions shift transition temperatures toward a weighted average and broaden the transition due to distribution in local chain environments (Suyatma et al., 2023). The reduced enthalpy ($\Delta H = 10.2\text{ J/g}$ in F4 vs. 12.8 J/g in F7) further supports intermolecular interaction consuming part of the thermal energy that would otherwise drive gelatinization or helix-coil transitions. This thermal miscibility is critical for industrial processing: it indicates that F4 can be processed as a single-phase material without the phase separation problems that would compromise film uniformity and performance during scale-up to commercial casting or extrusion operations. The lower glass transition temperature of F4 ($T_g = -18.4^{\circ}\text{C}$) compared to F1 (-8.2°C) indicates that plasticizer distribution is more homogeneous in the composite matrix, reflecting the greater free volume generated at the polymer blend interface a finding that has implications for film flexibility and handling at room temperature.

The SEM morphological analysis provides direct visual evidence for the polymer network integration inferred from FTIR and DSC. The dense, smooth, crack-free cross-section of F4 without phase separation or granule remnants confirms complete gelatinization and effective polymer network interpenetration at the microscopic scale. This morphological homogeneity explains why F4 exhibits superior mechanical performance compared to films with visible starch granules or micro-cracks, as such defects act as stress concentration points that initiate fracture under load. The increasing surface roughness in high-starch formulations (F6 and F7 with Ra values of 148.2 nm and 224.6 nm, respectively, versus 62.4 nm for F4) correlates with reduced transparency and wettability, suggesting that starch-rich films have more heterogeneous surfaces that could affect sealability and printability in packaging applications. These morphological findings have practical implications for packaging quality: the smooth, uniform surface of F4 would facilitate high-quality printing for branding and labeling, while the absence of micro-cracks ensures barrier integrity against microbial contamination.

The barrier properties of F4 reveal a critical trade-off that must be addressed for commercial application. The WVTR of 126.8 g/m²-day, while significantly better than pure carrageenan films (163.4 g/m²-day), remains substantially higher than conventional LDPE (8–15 g/m²-day). This limitation stems from the inherent hydrophilicity of both κ -carrageenan's sulfate groups and starch's hydroxyl groups, which absorb moisture from the environment and facilitate water vapor diffusion through the film matrix via sorption-diffusion mechanisms (Jiménez et al., 2022). However, this moisture sensitivity must be interpreted in the context of target applications. For dry food products such as crackers, nuts, dried fruits, and spices which constitute a substantial portion of Indonesia's food packaging market the WVTR of F4 is functionally adequate because the product itself maintains low water activity and short shelf-life requirements. Furthermore, the OTR of 42.3 cm³/m²-day-atm is comparable to many commercial bioplastics and adequate for oxygen-sensitive dry products where oxidative rancidity is the primary degradation mechanism. The superior oxygen barrier of carrageenan-rich formulations reflects the well-established tortuous path mechanism of dense polysaccharide networks, wherein the ordered double helix structure of κ -carrageenan creates a highly convoluted diffusion pathway that significantly increases oxygen permeation resistance (Tavares et al., 2021; Rhim et al., 2023). The progressive OTR increase from F1 to F7 directly reflects the disruption of this organized network as starch replaces carrageenan, introducing regions of lower packing density that facilitate gas diffusion. This structure-function relationship suggests that for applications requiring high oxygen barrier, formulations with higher carrageenan content would be preferred, whereas for applications prioritizing moisture barrier, starch-rich formulations would be advantageous though at the cost of mechanical integrity. The F4 formulation represents a pragmatic compromise suitable for the majority of dry food packaging applications.

The biodegradation results under composting conditions at 28°C carry significant implications for Indonesia's waste management challenges. All seven formulations achieved greater than 90% weight loss within 30 days, dramatically exceeding the ISO 14855 pass criterion of 90% biodegradation within 180 days under controlled composting. This remarkable performance orders of magnitude faster than conventional plastics, which show <1.2% weight loss after 6 months under



identical conditions positions these materials as genuinely sustainable alternatives rather than merely "degradable" in the sense of fragmentation into microplastics. The high biodegradation rate of F4 ($k = 0.104 \text{ day}^{-1}$, $t_{1/2} = 6.7 \text{ days}$) can be attributed to three synergistic mechanisms operating under tropical Indonesian conditions. First, the high hydrophilicity of the sulfated galactan backbone enables rapid water uptake and matrix swelling, increasing the accessible surface area for enzymatic attack. Second, soil microorganisms in tropical Indonesian conditions, including carrageenanase-producing bacteria of the genera *Pseudoalteromonas*, *Zobellia*, and *Cytophaga*, are naturally abundant and efficient degraders of sulfated polysaccharides, reflecting the evolutionary adaptation of tropical soil microbiomes to high organic matter turnover rates. Third, the gelatinized starch matrix is highly accessible to amylase-producing soil bacteria and fungi, further accelerating mass loss in the composite system (Ganesan et al., 2023). The observation that starch-rich F7 showed slower early-stage degradation (day 7: 41.2%) compared to carrageenan-rich F1 (day 7: 62.4%) reflects the initial crystallinity barrier imposed by residual semi-crystalline starch domains, which require enzymatic de-crystallization before bulk degradation can proceed. By day 21, this kinetic difference became statistically insignificant, suggesting that once enzymatic penetration is established, the bulk degradation of starch and carrageenan components proceeds at comparable rates under tropical composting conditions. This finding is practically important: it indicates that even if sorting is imperfect and composite films enter mixed waste streams, biodegradation will still occur within a timeframe compatible with existing composting infrastructure, reducing the burden on waste management systems.

The food safety assessment results represent a critical regulatory milestone for commercial viability. The low overall migration values (4.82 mg/dm² in ethanol simulant; 5.14 mg/dm² in acetic acid simulant), substantially below both SNI 8424:2017 and EU Regulation 10/2011/EC limits, are consistent with the purely polysaccharide nature of the film matrix, which lacks the plasticizer additives, antioxidants, and processing stabilizers commonly present in conventional plastic films that contribute to migration. The heavy metal concentrations Pb at 0.012 mg/kg, Cd and Hg below detection limits, and As at 0.006 mg/kg reflect the clean nature of the alkali extraction process used for carrageenan and the natural agricultural origin of cassava starch, with minimal industrial contamination. These results are particularly significant in the Indonesian context, where food safety concerns have previously hindered adoption of alternative packaging materials. The absence of color transfer in any simulant ($\Delta E < 2.0$) ensures that the film will not affect the visual appearance of packaged food products, a subtle but important quality factor for consumer acceptance. Collectively, these results demonstrate that F4 meets both national and international food-contact safety standards, removing a key regulatory barrier to adoption and providing assurance to food manufacturers concerned about migration risks.

Comparing F4 performance with previously reported carrageenan-starch composite films reveals the contributions of this study to the broader literature. Farhan & Hani (2021) reported tensile strength values of 12.8–14.2 MPa for semi-refined κ -carrageenan films with glycerol and sorbitol, substantially lower than the 18.74 MPa achieved for F4. This difference likely reflects the higher purity of carrageenan extracted in the present study (sulfate content 24.8% versus approximately

18–20% for semi-refined grades) and the synergistic mechanical reinforcement from complete starch network integration. The use of high-purity, well-characterized carrageenan in this study provides a more reproducible and industrially relevant baseline than semi-refined materials with variable composition. Balasubramaniam et al. (2023) achieved tensile strength up to 22.4 MPa for κ -carrageenan/tapioca starch films incorporating TiO_2 nanoparticles, suggesting that nanofiller incorporation could be a productive avenue for further mechanical improvement of the F4 formulation without compromising biodegradability, provided nanoparticle concentrations remain within food-safe limits. The biodegradation rate of F4 (94.7% at 30 days) compares favorably with marine polysaccharide films reported by Ganesan et al. (2023) for carrageenan/ulvan composites (88–92% at 30 days under similar conditions), likely reflecting the complementary degradation kinetics of carrageenan (rapid, enzyme-driven) and starch (initially slower but comprehensive), which together produce a more complete degradation profile than either polysaccharide alone. This comparative analysis suggests that the F4 formulation represents an advance over previous carrageenan-starch systems in terms of both mechanical performance and biodegradation kinetics, while achieving regulatory compliance that has not been previously demonstrated.

From a practical sustainability perspective, the use of Indonesian-sourced *Eucheuma cottonii* seaweed and local Adira-4 cassava variety establishes a compelling supply chain argument for commercial scale-up of F4-based packaging. Indonesia's position as the world's largest seaweed producer and third-largest cassava producer means that both primary raw materials are domestically available at scale and low cost, reducing supply chain risk and supporting domestic value-added manufacturing. This is critically important in the context of global supply chain disruptions, which have highlighted the vulnerability of import-dependent packaging industries. Furthermore, the development of a domestic bioplastics industry could create employment opportunities in rural coastal areas (seaweed cultivation) and agricultural regions (cassava farming), contributing to Indonesia's economic development goals while addressing environmental concerns. The demonstrated food safety compliance further removes a key regulatory barrier to adoption, while the biodegradation data provide confidence that end-of-life disposal will not contribute to the microplastic pollution crisis. However, the primary technical limitation remaining for commercial application is the elevated WVTR relative to conventional plastics, which restricts direct application to dry or low-moisture food products without additional barrier enhancement. Future research directions should therefore prioritize moisture barrier improvement through approaches such as lipid-polysaccharide bilayer coatings with beeswax or carnauba wax, incorporation of hydrophobic emulsions into the film matrix, surface cross-linking with potassium or calcium ions, pilot-scale extrusion or blown film processing trials, and life cycle assessment comparing the environmental footprint of F4 packaging against conventional alternatives. Shelf-life studies for specific Indonesian food products and consumer acceptance studies would further support market introduction. In conclusion, the comprehensive characterization and optimization of κ -carrageenan/cassava starch composite films presented in this study establishes that the 5:5 formulation (F4) achieves an optimal balance of mechanical strength, barrier performance, biodegradability, and food safety compliance, providing a scientifically robust basis for utilizing Indonesia's abundant seaweed and cassava



resources to produce commercially competitive biodegradable food packaging that can contribute meaningfully to addressing the nation's plastic waste crisis.

CONCLUSIONS

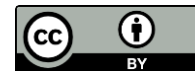
This study successfully developed and characterized biodegradable composite packaging films from κ -carrageenan (*Eucheuma cottonii*) and cassava starch at seven compositional ratios. Systematic multi-parameter evaluation identified Formulation F4 (5:5 carrageenan:starch, w/w) as the optimal formulation, demonstrating the best balance of physical, mechanical, barrier, and biodegradation properties. Key performance metrics of F4 included: tensile strength of 18.74 ± 0.42 MPa (exceeding ASTM D882 minimum), elongation at break of 32.16%, WVTR of $126.8 \text{ g/m}^2\text{-day}$, OTR of $42.3 \text{ cm}^3\text{/m}^2\text{-day-atm}$, and 94.7% biodegradation within 30 days under composting conditions.

FTIR and SEM analyses confirmed genuine intermolecular interactions and complete morphological homogeneity at the 5:5 blending ratio, providing mechanistic explanation for superior mechanical performance. Food safety testing confirmed compliance with both SNI 8424:2017 and EU Regulation 10/2011/EC limits for all migration and heavy metal parameters. DSC analysis showed single-peak thermal transitions in composite films, confirming polymer miscibility.

These findings establish a scientifically robust basis for utilizing Indonesia's abundant seaweed and cassava resources to produce commercially competitive biodegradable food packaging. Future work should focus on: (i) upscaling to pilot-scale casting or extrusion processes; (ii) enhancing moisture barrier by incorporating beeswax emulsion or ZnO nanoparticles; (iii) evaluating shelf-life extension performance for specific dry food products (crackers, nuts, spices); and (iv) conducting life cycle assessment (LCA) to quantify environmental benefits relative to conventional plastic packaging.

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