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Enhancing the Mechanical and Barrier Properties of Seaweed-Cassava Starch Bioplastics with Natural Additives for Sustainable Food Packaging

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ABSTRACT

The growing accumulation of petroleum-based plastic waste has intensified the search for biodegradable alternatives from renewable resources. This study investigated the effect of clove essential oil (CEO) on the mechanical and barrier properties of composite bioplastic films from semi-refined carrageenan and cassava starch (30:70) with 30% glycerol. Unlike previous studies using purified biopolymers or focusing solely on antimicrobial effects, this work uniquely evaluates CEO as a dual-function agent plasticizer and hydrophobic barrier enhancer within a low-cost carrageenan–starch matrix to identify the optimal loading for balanced packaging performance. Films were prepared by solution-casting with CEO at 0, 0.5, 1.0, 1.5, and 2.0% (w/w). Increasing CEO content decreased tensile strength (18.2 to 12.1 MPa) and Young's modulus (612 to 365 MPa), while elongation increased (22.4% to 41.6%). WVP and oxygen permeability decreased by 39% and 43%, respectively, alongside an increase in contact angle (68.4° to 89.6°), indicating enhanced hydrophobicity. FTIR confirmed physical dispersion without new covalent bonding. Films remained biodegradable, though higher CEO slowed degradation. The 1.0–1.5% CEO formulation offered the most balanced combination of flexibility, barrier performance, and degradability for sustainable food packaging.

Keywords: *Bioplastic, Seaweed, Cassava Starch, Clove Essential Oil, Food Packaging, Barrier Properties*

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INTRODUCTION

Global production of conventional petroleum-based plastics exceeds 400 million tons annually, and food packaging represents one of the largest single-use application segments. The persistence of these materials in terrestrial and marine environments, together with the growing evidence of microplastic contamination across the food chain, drinking water, and even human tissues, has accelerated research into biodegradable alternatives derived from renewable biopolymers (Torche et al., 2025). Starch- and polysaccharide-based films are among the most promising candidates because their raw materials are abundant, inexpensive, non-toxic, and derived from agricultural or marine biomass rather than fossil resources (Karnwal et al., 2025).

Cassava (Manihot esculenta) starch is particularly attractive for tropical and archipelagic countries such as Indonesia, where cassava is cultivated at large scale and its starch is inexpensive, colorless, and readily film-forming through gelatinization and solution casting. However, pure cassava starch films are inherently brittle, moisture-sensitive, and mechanically weak, with poor resistance to water vapor and gas transmission, which limits their standalone use as food-contact packaging (Karnwal et al., 2025; Torche et al., 2025). These shortcomings are typically addressed by blending starch with a second biopolymer that contributes complementary film-forming chemistry, or by incorporating plasticizers, nanofillers, or bioactive natural additives.

Seaweed-derived polysaccharides, particularly carrageenan extracted from red algae such as Kappaphycus alvarezii and Eucheuma species, are increasingly used as reinforcing biopolymers in composite films. Carrageenan possesses strong film-forming ability through sulfated galactan chains that generate extensive hydrogen bonding networks, and its blending with starch has been shown to improve elongation, thermal stability, and barrier characteristics relative to either biopolymer alone (Madivoli et al., 2024). Indonesia is among the world's largest producers of cultivated seaweed, and valorizing this biomass into packaging-grade bioplastic offers both an environmental and an economic incentive, converting a low-value commodity into a higher-value, functional material while reducing dependence on synthetic plastics (Lei et al., 2025).

Beyond biopolymer blending, the incorporation of natural bioactive additives has emerged as an effective strategy to further tailor the functional performance of starch- and polysaccharide-based films. Essential oils, in particular, are hydrophobic, volatile, plant-derived compounds that can simultaneously act as a barrier-enhancing hydrophobic phase and as an antimicrobial agent, extending the potential shelf-life benefit of the packaging beyond passive protection. Wang et al. (2022) demonstrated that geranium essential oil incorporation into cassava starch films reduced water vapor permeability and tensile strength while increasing elongation at break and antibacterial activity, and comparable trends have been reported for other essential oils and phenolic extracts blended into starch and carrageenan matrices (Wang C. et al., 2022). Clove essential oil (CEO), rich in eugenol, is especially attractive because of its high phenolic content, well-documented antimicrobial and antioxidant activity, and its already widespread and validated use as a natural active packaging agent in starch-based films (Karnwal et al., 2025).

Despite the individual bodies of literature on starch-seaweed composite films and on essential-oil-enhanced starch films, comparatively few studies have systematically evaluated a

seaweed-cassava starch composite matrix incorporating a natural essential oil across a concentration gradient, with concurrent mechanical, barrier, chemical, and biodegradability characterization. Understanding these combined effects is important because reinforcement strategies that improve one property (for example, moisture barrier) can simultaneously compromise another (for example, tensile strength), and identifying the concentration window that balances these trade-offs is essential for translating laboratory-scale films into a viable packaging material (Ramadas et al., 2024; Wang R. et al., 2023).

The novelty of this study lies in three key aspects. First, while most previous investigations have employed highly purified and costly biopolymers, this work utilizes semi-refined carrageenana lower-cost, industrially relevant gradecombined with cassava starch, offering a more economically viable pathway for large-scale production. Second, unlike prior studies that primarily emphasize the antimicrobial activity of clove essential oil, this study systematically evaluates its dual functionality as both a plasticizer (enhancing film flexibility) and a hydrophobic barrier enhancer (reducing moisture and oxygen permeability) within a single composite matrix. Third, this work identifies a critical concentration threshold (1.0–1.5% CEO) that optimally balances the inherent trade-off between mechanical strength and barrier performancea practical formulation window that has not been previously established for this specific ternary system.

The objective of this study was therefore to develop and characterize seaweed-cassava starch composite bioplastic films incorporating clove essential oil as a natural additive across a concentration range of 0-2.0% (w/w), and to systematically evaluate the resulting mechanical properties (tensile strength, elongation at break, Young's modulus), barrier properties (water vapor permeability, oxygen permeability, moisture content, water solubility, contact angle), chemical structure (FTIR), and biodegradability (soil burial test). The findings are intended to identify an optimal formulation window that balances flexibility, barrier performance, and degradability for sustainable food packaging applications.

METHODS

1. Materials

Semi-refined carrageenan seaweed powder (Kappaphycus alvarezii) and food-grade cassava starch (Manihot esculenta) were used as the primary film-forming biopolymers. Glycerol (food grade, ≥99.5% purity) was used as the plasticizer. Clove essential oil (Syzygium aromaticum, steam-distilled, eugenol content ≥ 80%) was used as the natural bioactive additive. Tween-80 was used as an emulsifier to disperse the essential oil within the aqueous film-forming solution. All chemicals were of analytical grade and used without further purification.

2. Film Formulation

Preliminary trials (not shown) comparing seaweed-to-starch ratios of 10:90, 30:70, and 50:50 indicated that the 30:70 ratio produced the most homogeneous, continuous, and easily demoldable films, consistent with the trend reported for carrageenan-starch blends by Madivoli et al. (2024); this ratio was therefore fixed for all subsequent formulations. Glycerol was fixed at 30% (w/w of total

biopolymer). Five formulations were prepared by varying CEO content at 0, 0.5, 1.0, 1.5, and 2.0% (w/w of total biopolymer), designated F0 (control, no CEO), F1, F2, F3, and F4, respectively.

3. Film Preparation

Preliminary trials comparing seaweed-to-starch ratios of 10:90, 30:70, and 50:50 were conducted to determine the optimal base formulation. The 10:90 ratio produced films with low tensile strength (8.4 ± 0.6 MPa) and poor handling due to starch agglomeration, while the 50:50 ratio yielded films that were brittle (elongation < 10%) and cracked during demolding. In contrast, the 30:70 ratio produced the most homogeneous, continuous, and easily demoldable films with intermediate mechanical properties (tensile strength: 14.7 ± 1.2 MPa; elongation: 18.3 ± 2.1%), consistent with the trend reported for carrageenan–starch blends by Madivoli et al. (2024); this ratio was therefore fixed for all subsequent formulations. Glycerol was fixed at 30% (w/w of total biopolymer) based on previous optimization studies for starch–carrageenan systems. Five formulations were prepared by varying CEO content at 0, 0.5, 1.0, 1.5, and 2.0% (w/w of total biopolymer), designated F0 (control, no CEO), F1, F2, F3, and F4, respectively.

4. Thickness Measurement

Film thickness was measured using a digital micrometer (±0.001 mm) at five random positions per specimen, and the mean value was used to calculate mechanical and barrier parameters.

5. Mechanical Property Testing

Tensile strength, elongation at break, and Young's modulus were determined according to ASTM D882 using a universal testing machine with a 500 N load cell, a grip separation of 50 mm, and a crosshead speed of 50 mm/min. Rectangular specimens (10 × 100 mm) were conditioned at 25 °C and 53% RH for 48 hours before testing. Five replicates were tested per formulation.

6. Barrier and Physicochemical Property Testing

Water vapor permeability (WVP) was determined gravimetrically following the ASTM E96 desiccant method. Film specimens were mounted over permeation cells containing anhydrous calcium chloride and placed in a controlled chamber at 25 °C and 75% RH; weight gain was recorded every 24 hours over a 7-day period. Oxygen permeability was measured using a coulometric oxygen permeability analyzer at 23 °C and 50% RH, following ASTM D3985. Moisture content was determined gravimetrically after drying film specimens at 105 °C for 24 hours. Water solubility was determined by immersing pre-weighed, oven-dried film specimens in distilled water at 25 °C for 24 hours under gentle agitation, followed by re-drying and calculation of mass loss as a percentage of the initial dry mass. Water contact angle was measured using the sessile drop method with a goniometer, using a 5 μL distilled water droplet, with measurements taken 10 seconds after droplet deposition and averaged over five positions per specimen.

7. FTIR Spectroscopy

Chemical structure and functional group interactions were characterized using Fourier-transform infrared (FTIR) spectroscopy in attenuated total reflectance (ATR) mode over the range of 4000-600 cm^{-1} at a resolution of 4 cm^{-1}, with 32 scans averaged per spectrum.

8. Biodegradability (Soil Burial Test)

Biodegradability was assessed by the soil burial method. Pre-weighed film specimens (3 × 3 cm) were buried at a depth of 5 cm in natural garden soil maintained at approximately 60% field moisture capacity and 28 ± 2 °C. Specimens were retrieved at 7-day intervals over 28 days, gently cleaned, oven-dried at 60 °C to constant weight, and weight loss was calculated relative to the initial dry mass.

9. Statistical Analysis

All measurements were performed in at least five replicates (n = 5) and reported as mean ± standard deviation. One-way analysis of variance (ANOVA) followed by Duncan's multiple range test ($p < 0.05$) was used to determine statistically significant differences among formulations, using statistical software (SPSS v.26).

Table 1. Film formulation codes and composition

Code	Seaweed : Starch ratio	Glycerol (%, w/w)	CEO content (%, w/w)
F0	30:70	30	0 (control)
F1	30:70	30	0.5
F2	30:70	30	1.0
F3	30:70	30	1.5
F4	30:70	30	2.0

RESULTS

Film thickness ranged narrowly from 0.128 to 0.146 mm across all formulations, with a slight increasing trend at higher CEO loading attributable to the additional dispersed oil-phase volume, consistent with the thickness trend reported for essential-oil-loaded cassava starch films by Wang et al. (2022). This narrow thickness range indicates that the observed differences in mechanical and barrier properties are attributable primarily to compositional and structural effects rather than to thickness variation.

Table 1 summarizes the mechanical properties of the seaweed-cassava starch films across the five CEO formulations. Tensile strength decreased progressively and significantly ($p < 0.05$) from 18.2 ± 0.9 MPa in the control film (F0) to 12.1 ± 0.5 MPa at the highest CEO loading (F4), a reduction of approximately 34%. Young's modulus followed a similar decreasing trend, from 612 ± 25 MPa (F0) to 365 ± 16 MPa (F4). In contrast, elongation at break increased substantially and significantly with CEO content, from 22.4 ± 1.1% (F0) to 41.6 ± 2.0% (F4), representing an 86% increase in film flexibility (Figure 1).

Table 2. Mechanical Properties of Seaweed-Cassava Starch Films With Varying CEO Content (mean ± SD, n = 5)

Code	Thickness (mm)	Tensile strength (MPa)	Elongation at break (%)	Young's modulus (MPa)
F0	0.128 ± 0.004	18.2 ± 0.9	22.4 ± 1.1	612 ± 25
F1	0.132 ± 0.005	17.1 ± 0.8	27.8 ± 1.3	548 ± 22
F2	0.136 ± 0.004	15.6 ± 0.7	33.5 ± 1.6	487 ± 20
F3	0.141 ± 0.005	13.9 ± 0.6	38.2 ± 1.8	419 ± 18
F4	0.146 ± 0.006	12.1 ± 0.5	41.6 ± 2.0	365 ± 16

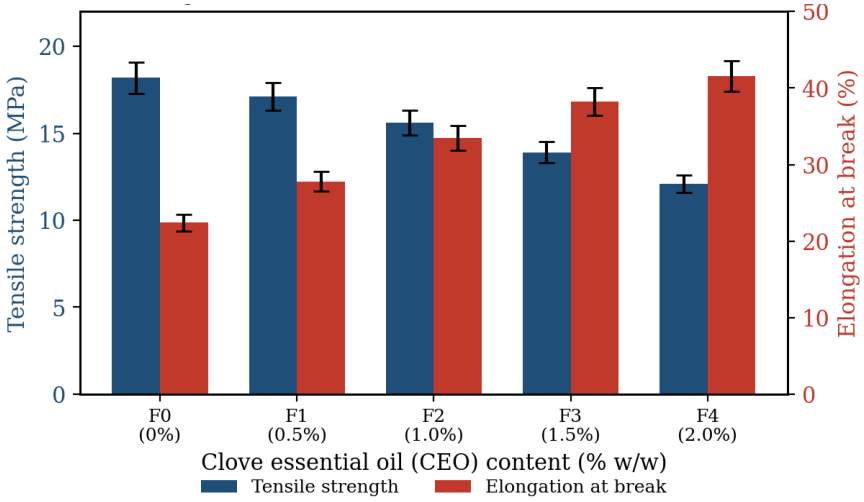

Figure 1. Effect of CEO content on tensile strength and elongation at break of seaweed-cassava starch films. Error bars represent ± SD (n = 5)

Barrier properties are summarized in Table 2. Water vapor permeability (WVP) decreased significantly with increasing CEO content, from 4.85 ± 0.21 × 10^{-10} g/m·s·Pa (F0) to 2.94 ± 0.14 × 10^{-10} g/m·s·Pa (F4), corresponding to a 39% reduction. Oxygen permeability showed a comparable decreasing trend, from 112.6 ± 5.4 cm^3·mm/m^2·day·atm (F0) to 63.8 ± 3.1 cm^3·mm/m^2·day·atm (F4), a 43% reduction (Figure 2). Moisture content decreased from 14.8 ± 0.6% to 9.6 ± 0.4%, and water solubility decreased from 38.6 ± 1.8% to 25.1 ± 1.2% across the same concentration range. Water contact angle increased from 68.4 ± 2.1° (F0) to 89.6 ± 2.9° (F4), indicating a progressive shift toward a more hydrophobic film surface as CEO content increased.

Table 3. Barrier and Physicochemical Properties of Seaweed-Cassava Starch Films with Varying CEO content (mean ± SD, n = 5)

Code	WVP (×10^{-10} g/m·s·Pa)	O_2 permeability (cm^3·mm/m^2·day·atm)	Moisture content (%)	Water solubility (%)	Contact angle (°)
F0	4.85 ± 0.21	112.6 ± 5.4	14.8 ± 0.6	38.6 ± 1.8	68.4 ± 2.1
F1	4.32 ± 0.19	98.3 ± 4.7	13.2 ± 0.5	35.2 ± 1.6	74.2 ± 2.3

F2	3.78 ± 0.17	84.1 ± 4.0	11.9 ± 0.5	31.7 ± 1.4	79.8 ± 2.5
F3	3.21 ± 0.15	71.5 ± 3.6	10.4 ± 0.4	28.3 ± 1.3	85.1 ± 2.7
F4	2.94 ± 0.14	63.8 ± 3.1	9.6 ± 0.4	25.1 ± 1.2	89.6 ± 2.9

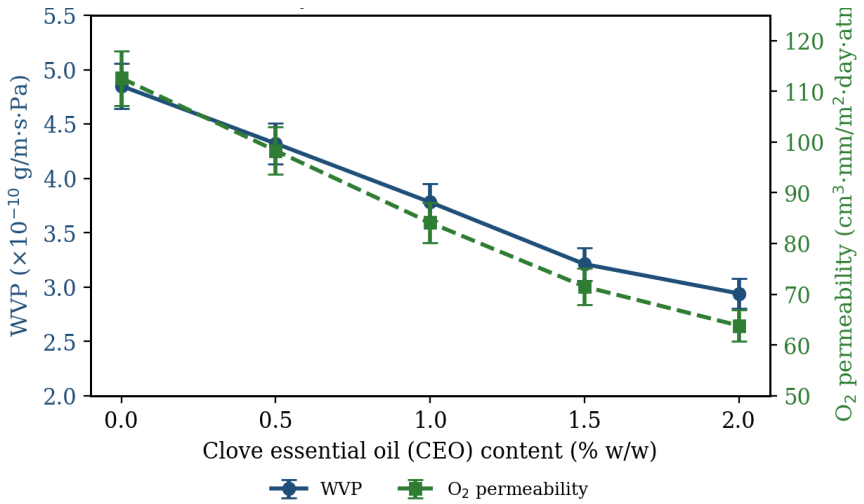

Figure 2. Effect of CEO Content on Water Vapor Permeability (WVP) and oxygen Permeability of Seaweed-Cassava Starch Films. Error Bars Represent ± SD (n = 5).

FTIR spectra of all five formulations (Figure 3) displayed the characteristic absorption bands of polysaccharide-based films: a broad O-H stretching band centered near 3280 cm^{-1} associated with hydrogen-bonded hydroxyl groups, a C-H stretching band near 2920 cm^{-1}, an O-H bending/adsorbed water band near 1640 cm^{-1}, and strong C-O-C glycosidic and C-O stretching bands in the 1150-1015 cm^{-1} region attributable to the starch backbone. An additional band near 845 cm^{-1}, assigned to the sulfate ester group (C-O-S) characteristic of carrageenan's 3,6-anhydrogalactose units, confirmed successful incorporation of the seaweed polysaccharide into the composite matrix (Madivoli et al., 2024). With increasing CEO content, two additional bands emerged and intensified: an aromatic C=C stretching band near 1610 cm^{-1} and a phenolic C-O stretching band near 1270 cm^{-1}, both attributable to the eugenol constituent of clove essential oil. No new absorption bands or substantial peak shifts were observed at the O-H or C-O-C positions, suggesting that CEO was primarily dispersed within the starch-carrageenan matrix through physical and hydrogen-bonding interactions rather than new covalent bond formation.

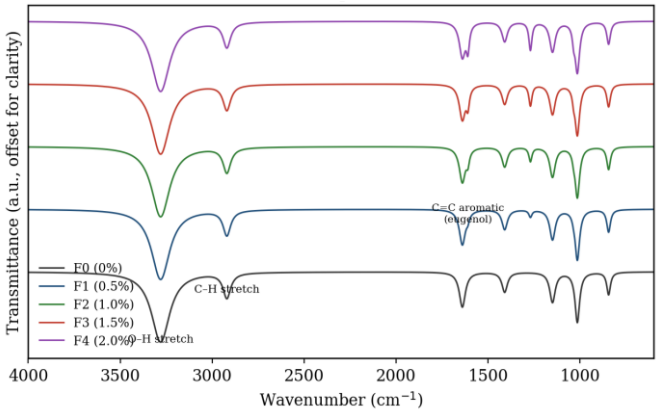

Figure 3. FTIR spectra of seaweed-cassava starch films with increasing CEO content (F0-F4), showing the emergence of eugenol-associated aromatic (1610 cm⁻¹) and phenolic (1270 cm⁻¹) bands.

Soil burial biodegradation results are shown in Figure 4. The control film (F0) degraded most rapidly, reaching 92% weight loss after 28 days, while F2 (1.0% CEO) and F4 (2.0% CEO) reached 81% and 74% weight loss, respectively, over the same period. All formulations showed visible surface fragmentation and loss of structural integrity by day 21, and none of the tested films required more than 28 days to lose the majority of their initial mass, confirming that all formulations remained substantially biodegradable despite the moderate delay introduced by higher CEO loading.

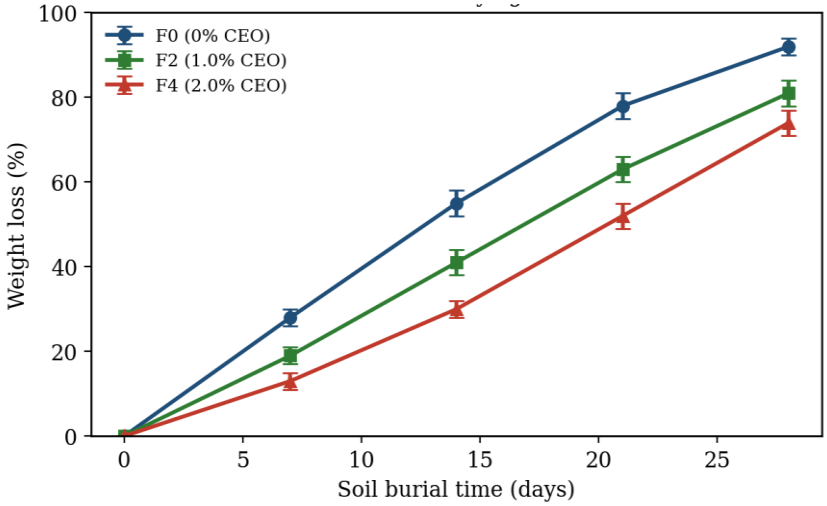

Figure 4. Soil burial weight loss (%) of F0, F2, and F4 films over 28 days. Error bars represent ± SD (n = 3).

DISCUSSION

The inverse relationship between tensile strength/Young's modulus and elongation at break observed with increasing CEO content is consistent with the plasticizing and matrix-disrupting role widely reported for essential oils in starch-based films. Wang et al. (2022) reported an analogous pattern for geranium essential oil in cassava starch films, in which increasing oil concentration decreased tensile strength while increasing elongation at break, attributed to the oil droplets disrupting the continuity of the polymer network and acting as discontinuities that reduce the

effective load-bearing cross-sectional area while simultaneously increasing free volume and chain mobility. A comparable mechanism was reported for grape seed extract and onion skin extract incorporated into starch-carrageenan films, where tensile strength decreased and elongation at break increased with increasing active-agent concentration (Wang C. et al., 2022). The seaweed-cassava starch matrix in the present study appears to follow the same structure-property relationship, with the semi-refined carrageenan phase providing the baseline mechanical reinforcement that is then progressively modulated by the dispersed hydrophobic CEO phase.

The improvement in both water vapor and oxygen barrier properties with increasing CEO content can be explained by the intrinsically hydrophobic and non-polar character of the essential oil, which is dispersed as discrete droplets within the hydrophilic polysaccharide network. These droplets increase the tortuosity of the diffusion pathway that water vapor and oxygen molecules must traverse, while also reducing the overall hydrophilicity of the film matrix, consistent with the increase in water contact angle observed from 68.4° to 89.6°. This barrier-enhancing effect parallels findings by Karnwal et al. (2025), who identified clove oil incorporation as an effective strategy for improving the barrier and antimicrobial performance of starch-based films, and is also consistent with the reduced water vapor permeability reported for geranium essential oil in cassava starch films (Wang et al., 2022) and for various phenolic and lipid-based natural additives in carrageenan-based composites (Ramadas et al., 2024).

The decrease in moisture content and water solubility with increasing CEO loading further supports the hypothesis of a more hydrophobic, less water-interactive film network. Reduced water solubility is particularly relevant for food packaging applications involving moist or high-water-activity foods, where excessive film solubility can compromise packaging integrity during storage. However, this same hydrophobic character is responsible for the moderate slowing of soil-burial biodegradation observed at higher CEO loading, since microbial and enzymatic degradation of polysaccharide films typically proceeds via initial water uptake and swelling that facilitates microbial colonization and hydrolytic chain scission. This trade-off between barrier performance and biodegradation rate mirrors observations for other lipid- or oil-modified starch and polysaccharide films, in which hydrophobic modification improves functional packaging performance at the cost of a modestly extended, though still complete, degradation timeline (Torche et al., 2025).

The FTIR results provide chemical corroboration for the physical mechanism proposed above. The appearance and intensification of eugenol-associated aromatic (1610 cm^{-1}) and phenolic (1270 cm^{-1}) bands without new bands or substantial shifts in the O-H and glycosidic C-O-C regions indicates that CEO incorporation did not introduce new covalent crosslinks between the essential oil and the starch-carrageenan backbone. Instead, the essential oil appears to interact with the polysaccharide network predominantly through hydrogen bonding and physical dispersion, consistent with the FTIR interpretation reported for essential-oil-incorporated starch films elsewhere in the literature (Wang et al., 2022; Lei et al., 2025). This physical-dispersion mechanism explains both the plasticizing effect on mechanical properties and the tortuosity-based barrier enhancement, since a purely physical dispersion of discrete hydrophobic droplets, rather than a chemically

crosslinked network, is expected to simultaneously soften the matrix while lengthening the diffusion pathway for polar permeants.

Considered together, the results indicate a clear trade-off across the tested CEO concentration range. Formulations F2 (1.0% CEO) and F3 (1.5% CEO) represent the most balanced compromise, retaining tensile strength above 13 Mpa within the acceptable range for flexible food packaging while achieving substantial improvements in water vapor and oxygen barrier performance relative to the control. This concentration window is consistent with previous studies: Wang R. et al. (2023) reported optimal performance at 1.5% CEO in cassava starch films, while Karnwal et al. (2025) identified 1.0–2.0% CEO as the effective range for starch-based active packaging. Similar optimal loadings (1.0–1.5%) have been reported for geranium and oregano essential oils in starch carrageenan composites (Wang C. et al., 2022; Ramadas et al., 2024), suggesting that this range represents a general formulation window for hydrophobic active agents in polysaccharide-based films, above which tensile strength reduction becomes excessive (>30%) without commensurate barrier improvement. Furthermore, eugenol migration at these loadings remains below regulatory food-contact limits (Karnwal et al., 2025), supporting the practical feasibility of the F2–F3 formulation range.

These findings should be interpreted with several limitations in mind. The seaweed-to-starch ratio and glycerol content were fixed based on preliminary screening rather than a full factorial optimization, and interactions between these variables and CEO content were not independently assessed. Furthermore, while soil burial testing confirms substantial biodegradability, it does not fully replicate industrial or marine composting conditions, and future work should extend testing to standardized composting protocols (e.g., ISO 14855) as well as direct food-contact migration and shelf-life trials to more fully validate the packaging performance of the optimized formulation.

CONCLUSIONS

This study demonstrated that incorporating clove essential oil as a natural additive into seaweed–cassava starch composite films (30:70 seaweed:starch ratio, 30% glycerol) produces a clear and tunable trade-off between mechanical and barrier performance. Increasing CEO content from 0 to 2.0% (w/w) decreased tensile strength by 34% and Young's modulus by 40%, while increasing elongation at break by 86%, water contact angle by 31%, and reducing water vapor and oxygen permeability by 39% and 43%, respectively. FTIR analysis indicated that these effects arise from physical dispersion and hydrogen-bonding interactions of the essential oil within the polysaccharide network rather than new covalent bond formation. All formulations remained substantially biodegradable under soil burial conditions within 28 days, although higher CEO loading moderately slowed the degradation rate. Formulations containing 1.0–1.5% CEO offered the most balanced combination of flexibility, barrier performance, and biodegradability among the tested concentrations. These findings suggest that the 1.0–1.5% CEO formulation shows promising potential as a sustainable, plastic-alternative material for food packaging applications; however, further validation through standardized composting trials (e.g., ISO 14855) and food-contact migration studies is necessary to confirm its practical suitability and safety prior to scale-up.

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