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CMC-GELATIN COMPOSITE FILM CHARACTERISTICS WITH THE ADDITION OF CINNAMON ESSENTIAL OIL (*CINNAMOMUM VERUM*)

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ABSTRACT

Bioplastic films based on carboxymethyl cellulose (CMC) and gelatin have gained considerable attention as sustainable alternatives to synthetic packaging materials, particularly for food applications. This study aimed to characterize composite films composed of 1.5% CMC and 1.5% gelatin, plasticized with 20% glycerol (relative to total polymer), and incorporated with cinnamon essential oil (CEO) at concentrations of 0% (A), 0.1% (B), 0.3% (C), and 0.5% (D), fabricated using the casting method. Physical, mechanical, and barrier properties were evaluated, including thickness, tensile strength, elongation at break, water vapor transmission rate (WVTR), water solubility, and antimicrobial activity. Results showed that CEO concentration significantly influenced elongation at break and WVTR. Elongation increased from 43.80% (A) to 61.59% (D), while tensile strength decreased from 11.00 MPa (A) to 6.24 MPa (D). WVTR decreased substantially from 47.13 g/m²/24h (A) to 19.63 g/m²/24h (D), indicating improved moisture barrier properties with higher CEO. Film thickness remained consistent across all treatments (~0.15–0.18 mm). Water solubility values ranged between 33.82% and 44.27%. Antimicrobial activity was not detected in any treatment under the test conditions applied. These findings suggest that the incorporation of cinnamon essential oil can effectively modulate the functional properties of CMC-gelatin composite films, with treatment D (0.5%

CEO) showing the most promising barrier characteristics for potential food packaging applications.

KEYWORDS: CMC; gelatin; cinnamon essential oil; bioplastic film; food packaging; casting method.

1. INTRODUCTION

The increasing global demand for sustainable and environmentally friendly packaging has driven extensive research into biopolymer-based films as viable alternatives to petroleum-derived plastics [1]. Bioplastic films derived from natural polysaccharides and proteins offer biodegradability, renewability, and biocompatibility, making them attractive candidates for food packaging applications [1]. Among the various biopolymers investigated, carboxymethyl cellulose (CMC) and gelatin have emerged as promising film-forming materials due to their complementary functional properties. CMC is an anionic cellulose derivative obtained by carboxymethylation of cellulose, widely utilized in the food and pharmaceutical industries for its excellent film-forming ability, water solubility, and non-toxicity [2]. However, CMC films alone tend to be brittle and exhibit relatively poor mechanical properties, limiting their practical application. Gelatin, a protein derived from the hydrolysis of collagen found in animal bones and skin—including fish byproducts—is recognized for its superior film-forming capacity, transparency, and good mechanical properties [3]. When CMC and gelatin are combined, synergistic interactions between the polysaccharide and protein matrices can result in composite films with enhanced mechanical and barrier properties [6].

Plasticizers are commonly incorporated into biopolymer films to reduce brittleness and improve flexibility. Glycerol, a polyol widely used in the food industry, is one of the most effective plasticizers for hydrophilic polymer films due to its ability to disrupt intermolecular hydrogen bonding within the polymer matrix, thereby increasing chain mobility and elongation at break [7]. In the present study, glycerol was employed at 20% of the total polymer weight, based on established protocols for CMC-gelatin composite film preparation [8,9]. A significant challenge associated with natural biopolymer films is their susceptibility to microbial contamination and moisture, which can compromise food safety and shelf life. To address these limitations, the incorporation of natural antimicrobial agents such as essential oils has been increasingly explored [14]. Cinnamon essential oil (CEO), derived from the bark of *Cinnamomum verum*, is recognized for its potent antimicrobial, antifungal,

and antioxidant properties, primarily attributed to its major bioactive component, cinnamaldehyde [5]. The hydrophobic nature of CEO can also potentially improve the moisture barrier properties of hydrophilic biopolymer films by creating a more tortuous path for water vapor diffusion [5,14].

Previous studies have reported that incorporation of essential oils into biopolymer matrices can significantly affect the physical, mechanical, and functional properties of resulting films, often leading to trade-offs between antimicrobial activity and mechanical integrity [15,16]. However, the specific behavior of CMC-gelatin composite films incorporated with CEO at different concentrations requires further investigation, particularly in the context of film-forming compositions relevant to fish product packaging. The objective of this study was therefore to characterize the physical, mechanical, and barrier properties of CMC-gelatin composite films incorporating cinnamon essential oil at four concentration levels (0%, 0.1%, 0.3%, and 0.5%), prepared by the casting method, and to evaluate the effect of CEO concentration on these functional characteristics.

2. MATERIALS AND METHODS

2.1. Materials

Carboxymethyl cellulose (CMC, food grade), gelatin (fish-derived, food grade), glycerol (pro-analysis), and cinnamon essential oil (*Cinnamomum verum*, steam-distilled) were obtained from commercial suppliers. All chemicals used for analyses were of analytical grade. Distilled water was used throughout the study.

2.2. Preparation of CMC-Gelatin Composite Films

Films were prepared using the casting method following a procedure adapted from Fakhouri et al. [9] with modifications. CMC solution (1.5% w/v) and gelatin solution (1.5% w/v) were each prepared separately by dissolving the respective polymers in distilled water at 60°C with continuous stirring for 30 minutes. The two solutions were then combined and mixed homogeneously using a magnetic stirrer. Glycerol was added as a plasticizer at a concentration of 20% based on the total dry weight of the polymers (CMC + gelatin). Cinnamon essential oil was incorporated at four concentration levels: 0% (treatment A, control), 0.1% (treatment B), 0.3% (treatment C), and 0.5% (treatment D), calculated on a v/v basis relative to the total film-forming solution. The film-forming solutions were homogenized using an ultrasonic probe for 2 minutes and then degassed under vacuum. Solutions were cast onto leveled acrylic plates (20 cm × 20 cm) and dried in a forced-air oven

at 45°C for 24 hours. The dried films were peeled from the plates and conditioned at 23 ± 2°C and 50 ± 5% relative humidity for 48 hours prior to analysis.

2.3. Film Thickness

Film thickness was measured using a digital micrometer (Mitutoyo, Japan) at five random positions for each film sample. The average of five measurements was recorded and expressed in millimeters [10]. Thickness measurements were conducted in accordance with ASTM D6988-13.

2.4. Tensile Strength and Elongation at Break

Tensile strength (TS) and elongation at break (EAB) were determined according to ASTM D882-18 using a universal testing machine (Lloyd Instruments, UK). Film specimens were cut into rectangular strips (10 mm × 100 mm) and subjected to uniaxial tensile testing at a crosshead speed of 50 mm/min with an initial gauge length of 50 mm. Tensile strength (MPa) was calculated by dividing the maximum load at failure by the initial cross-sectional area of the sample, while elongation at break (%) was calculated as the percentage increase in gauge length at the point of fracture relative to the original gauge length. At least five replicates were measured per treatment.

2.5. Water Vapor Transmission Rate (WVTR)

Water vapor transmission rate was measured gravimetrically according to ASTM E96/E96M-16 (desiccant method). Film samples were mounted on cups containing silica gel desiccant (0% RH), which were then placed in a desiccator maintained at 25°C and 75% relative humidity using a saturated NaCl solution. The cups were weighed at regular 24-hour intervals for 5 days. WVTR was expressed as g/m²/24h, calculated from the slope of the linear portion of the weight gain versus time plot, divided by the film area.

2.6. Water Solubility

Water solubility was determined following the method of Gontard et al. [12] with minor modifications. Film discs (2 cm diameter) were weighed after drying at 105°C for 24 hours (initial dry weight, W_1), then immersed in 50 mL of distilled water at 25°C for 24 hours with gentle agitation. Undissolved film pieces were recovered by filtration, dried again at 105°C for 24 hours, and reweighed (final dry weight, W_2). Solubility (%) was calculated as: Solubility (%) = $[(W_1 – W_2) / W_1] \times 100$.

2.7. Antimicrobial Activity

Antimicrobial activity of the films was evaluated against Staphylococcus aureus (ATCC 25923) and Escherichia coli (ATCC 25922) using the agar disc diffusion method according to the Clinical and Laboratory Standards Institute [13] guidelines. Film discs (6 mm diameter) were placed on Mueller-Hinton agar plates previously inoculated with a bacterial suspension (0.5 McFarland standard). Plates were incubated at 37°C for 24 hours. The diameter of the inhibition zone (mm) was measured using a digital caliper. Experiments were conducted in triplicate.

2.8. Statistical Analysis

All data were analyzed using one-way analysis of variance (ANOVA) followed by Duncan's Multiple Range Test (DMRT) at a significance level of 5% ($\alpha = 0.05$) using SPSS software version 26.0 (IBM, USA). Results are reported as mean ± standard deviation. Different superscript letters indicate significant differences among treatments.

3. RESULTS AND DISCUSSION

3.1. Film Thickness

The thickness of CMC-gelatin composite films with varying concentrations of cinnamon essential oil is presented in Table 1. Film thickness values ranged from 0.15 ± 0.00 mm (treatment B and C) to 0.18 ± 0.70 mm (treatment D), with no statistically significant differences among the four treatments ($p > 0.05$).

Table 1. Thickness of CMC-Gelatin Composite Films with Different Concentrations of Cinnamon Essential Oil.

Treatment	Thickness (mm)
0% CEO	0.16 ± 0.28^{a}
0.1% CEO	0.15 ± 0.00^{a}
0.3% CEO	0.15 ± 0.70^{a}
0.5% CEO	0.18 ± 0.70^{a}

Note: Values are means ± SD (n=5). Different superscript letters indicate significant differences ($p < 0.05$, DMRT).).

The uniformity in thickness across all treatments suggests that the incorporation of CEO at the concentrations tested did not substantially alter the volume or distribution of solid components within the film matrix during the casting and drying process. Similar findings

have been reported for recent CMC-gelatin and related active film systems, where relatively stable thickness values were attributed to controlled casting procedures and consistent solid content [3,4]. The slight numerical increase in thickness observed at 0.5% CEO (treatment D) may be attributable to the physical incorporation of oil droplets within the polymer network, contributing marginally to overall film thickness, though this difference was not statistically significant [4].

3.2. Tensile Strength

The tensile strength values of the CMC-gelatin composite films are summarized in Table 2. Tensile strength decreased from 11.00 ± 2.62 MPa (treatment A) to 6.24 ± 1.36 MPa (treatment D) with increasing CEO concentration, although differences among all treatments were not statistically significant ($p > 0.05$).

Table 2. Tensile Strength of CMC-Gelatin Composite Films with Different Concentrations of Cinnamon Essential Oil.

Treatment	Tensile Strength (MPa)
0% CEO	11.00 ± 2.62[a]
0.1% CEO	10.72 ± 1.08[a]
0.3% CEO	8.42 ± 1.51[a]
0.5% CEO	6.24 ± 1.36[a]

Note: Values are means ± SD (n=5). Different superscript letters indicate significant differences ($p < 0.05$, DMRT).

The observed decrease in tensile strength with increasing CEO concentration is consistent with findings reported for other essential oil-incorporated biopolymer films [15,16]. This phenomenon is primarily attributed to the disruption of the continuous polymer network by the incorporation of hydrophobic CEO droplets, which act as discontinuities or stress concentration points within the film matrix [16,17]. The hydrophobic oil droplets interfere with intermolecular hydrogen bonding and electrostatic interactions between CMC and gelatin chains, reducing the cohesive strength of the film. Additionally, the incorporation of dispersed oil droplets may create micro-defects or heterogeneities in the polymer matrix, further compromising mechanical resistance under tensile loading [16].

The lack of statistical significance across treatments, despite the numerical decrease, may reflect the high variability inherent in tensile testing of relatively thin biopolymer films and the relatively narrow range of CEO concentrations employed [11]. Notwithstanding, the trend

is consistent with recent studies on CMC-based and gelatin-based films incorporating active compounds or essential oils [3,4].

3.3. Elongation at Break

Elongation at break (EAB) of the composite films increased significantly with increasing CEO concentration (Table 3), ranging from 43.80 ± 0.20% (treatment A) to 61.59 ± 3.09% (treatment D).

Table 3. Elongation at Break of CMC-Gelatin Composite Films with Different Concentrations of Cinnamon Essential Oil.

Treatment	Elongation at Break (%)
0% CEO	43.80 ± 0.20[a]
0.1% CEO	45.16 ± 2.12[a]
0.3% CEO	55.87 ± 0.77[b]
0.5% CEO	61.59 ± 3.09[c]

Note: Values are means ± SD (n=5). Different superscript letters indicate significant differences ($p < 0.05$, DMRT).

The significant increase in elongation at break with higher CEO concentrations can be attributed to the plasticizing effect of the oil on the polymer matrix. The incorporation of CEO may contribute to increased free volume and chain mobility within the biopolymer network, facilitating greater deformation before fracture [4,16]. Additionally, the disruption of strong inter-chain interactions by embedded oil droplets reduces the rigidity of the polymer matrix, enabling greater extensibility [4,16].

These findings align with recent reports on composite biopolymer films in which active oil or bioactive incorporation increased extensibility while modifying tensile behavior [4,16]. The simultaneous decrease in tensile strength and increase in elongation is a characteristic behavior of plasticized polymer systems, reflecting the classic ductility-strength trade-off in film mechanics [7]. From a food packaging perspective, a higher elongation at break is desirable for films intended to wrap irregularly shaped food products, as it reduces the risk of tearing during application.

3.4. Water Vapor Transmission Rate (WVTR)

WVTR decreased significantly with increasing CEO concentration, from 47.13 ± 0.00 g/m²/24h in the control (treatment A) to 19.63 ± 5.55 g/m²/24h in treatment D (Table 4), indicating substantially improved moisture barrier properties.

Table 4. Water Vapor Transmission Rate (WVTR) of CMC-Gelatin Composite Films.

Treatment	WVTR (g/m²/24h)
0% CEO	47.13 ± 0.00^{c}
0.1% CEO	31.42 ± 0.00^{b}
0.3% CEO	25.52 ± 2.77^{ab}
0.5% CEO	19.63 ± 5.55^{a}

Note: Values are means ± SD (n=3). Different superscript letters indicate significant differences ($p < 0.05$, DMRT).

The marked reduction in WVTR with increasing CEO concentration can be explained by the hydrophobic nature of the essential oil. CMC and gelatin are inherently hydrophilic polymers with a high affinity for water molecules, resulting in elevated moisture vapor transmission in unmodified films [14]. The incorporation of hydrophobic CEO droplets into the film matrix creates a more tortuous pathway for water vapor diffusion, effectively reducing the permeability of the film to moisture [5,14]. This is consistent with the tortuous path model for diffusion in composite systems, wherein dispersed hydrophobic domains force diffusing molecules to navigate around the barrier phase, increasing the effective diffusion length [25]. These results are in agreement with previous reports on essential-oil-containing biopolymer films, including systems enriched with cinnamon oil, in which moisture barrier performance improved as hydrophobic domains were incorporated into the matrix [17,18,19]. From a practical standpoint, the significantly lower WVTR observed in treatments C and D represents a substantial improvement in moisture barrier performance, which is critically important for food packaging applications aimed at extending product shelf life by limiting moisture exchange between the food and its environment.

3.5. Water Solubility

Water solubility values of the composite films are presented in Table 5. Solubility ranged from 33.82 ± 0.69% (treatment B) to 44.27 ± 1.71% (treatment D), with significant differences detected among treatments.

Table 5. Water Solubility of CMC-Gelatin Composite Films with Different Concentrations of Cinnamon Essential Oil.

Treatment	Water Solubility (%)
0% CEO	40.24 ± 2.92^{bc}
0.1% CEO	33.82 ± 0.69^{a}
0.3% CEO	38.30 ± 2.24^{ab}
0.5% CEO	44.27 ± 1.71^{c}

Note: Values are means ± SD (n=3). Different superscript letters indicate significant differences ($p < 0.05$, DMRT).

The non-monotonic trend in water solubility across CEO concentrations suggests complex interactions between CEO and the CMC-gelatin matrix. The lowest solubility observed at treatment B (0.1% CEO) may reflect an optimal level of oil incorporation that enhances the hydrophobic character of the film surface without significantly disrupting polymer chain packing. At low concentrations, CEO may partially fill the free volume between polymer chains, reducing water accessibility [16,20].

Conversely, the higher solubility observed at treatment D (0.5% CEO) may be attributed to the disruptive effect of higher oil concentrations on the cohesive structure of the polymer network. Higher essential oil concentrations may also weaken the cohesive structure of the polymer network and facilitate water penetration into the film matrix [16,20]. Similar non-linear relationships between essential oil concentration and film solubility have been reported for other protein- and polysaccharide-based films [19,20].

From a food packaging perspective, water solubility of biopolymer films must be considered in relation to the intended application. For moisture-sensitive food products, lower solubility is desirable to maintain film integrity. However, for applications requiring water-soluble packaging (e.g., single-use dissolvable sachets), higher solubility may be advantageous [14].

3.6. Antimicrobial Activity

None of the film treatments (A through D) exhibited measurable inhibition zones against Staphylococcus aureus or Escherichia coli under the disc diffusion test conditions employed (Table 6). This absence of detectable antimicrobial activity, even in films containing 0.5% CEO (treatment D), warrants careful consideration.

Table 6. Antimicrobial Activity (Inhibition Zone Diameter, mm) of CMC-Gelatin Composite Films

Treatment	*S. aureus* (mm)	*E. coli* (mm)
0% CEO	0 (no zone)	0 (no zone)
0.1% CEO	0 (no zone)	0 (no zone)
0.3% CEO	0 (no zone)	0 (no zone)
0.5% CEO	0 (no zone)	0 (no zone)

Note: Disc diffusion method; CLSI guidelines. Inhibition zone measured excluding disc diameter.

Several factors may explain the absence of antimicrobial activity. First, the overall CEO loading levels in the film may have been insufficient to produce detectable inhibition zones in the agar diffusion assay. The minimum inhibitory concentration (MIC) of cinnamon essential oil against *S. aureus* and *E. coli* in broth systems has been reported in the literature [21]; however, migration of oil from a solid film matrix to an agar medium is inherently limited compared to direct application of the oil in liquid form.

Second, the encapsulation of CEO within the hydrophilic CMC-gelatin matrix may restrict its release and diffusion into the surrounding agar. Hydrophilic polymers can effectively entrap hydrophobic molecules, limiting their bioavailability [22]. Third, interactions between CEO components and the polymer matrix, including reactions involving cinnamaldehyde and protein side chains, may reduce the effective concentration of bioactive molecules available for antimicrobial action [23]. Future studies may explore enhanced delivery systems such as nanoemulsification, encapsulation, or other controlled-release strategies to improve CEO bioavailability and antimicrobial efficacy in film matrices [24].

4. CONCLUSIONS

This study demonstrated that the incorporation of cinnamon essential oil (CEO) at concentrations of 0–0.5% significantly influenced the functional properties of CMC-gelatin composite films prepared by the casting method. The most notable effects were observed for elongation at break and water vapor transmission rate. Elongation at break increased significantly from 43.80% (control) to 61.59% (0.5% CEO), while WVTR decreased substantially from 47.13 to 19.63 g/m²/24h, indicating that CEO effectively enhanced film flexibility and moisture barrier performance. Tensile strength showed a decreasing trend with increasing CEO concentration (11.00 to 6.24 MPa), though differences were not statistically

significant. Film thickness remained consistent across all treatments. Water solubility exhibited a non-monotonic trend, with the lowest value at 0.1% CEO and the highest at 0.5% CEO. No antimicrobial activity was detected under the disc diffusion conditions applied, likely due to limited CEO migration from the film matrix.

Overall, treatment D (0.5% CEO) demonstrated the most favorable barrier properties, with significantly improved moisture resistance, making it the most promising formulation for food packaging applications where moisture management is critical. However, further research is recommended to optimize film formulations for improved antimicrobial efficacy, potentially through nanoencapsulation or higher CEO loading, and to assess the compatibility and safety of these films in direct food contact applications.

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