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Effect of Stearic Acid on Barrier and Mechanical Properties of Edible Films Based on Carboxymethyl Cellulose (CMC), Konjac Glucomannan (KGM), and κ-Carrageenan (κCarr)

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ABSTRACT: The development of edible films using natural polysaccharides presents a sustainable alternative to synthetic packaging materials. This study aimed to enhance the barrier properties of edible films composed of carboxymethyl cellulose (CMC), konjac glucomannan (KGM), and κ-carrageenan (κCarr) by incorporating stearic acid (SA). Films were prepared by blending the biopolymers with SA at varying concentrations (0.1–0.5% w/w) and characterized for their structural, physical, and mechanical properties. Fourier-transform infrared (FTIR) spectroscopy confirmed molecular interactions between SA and the polysaccharide matrix, evidenced by reduced O–H absorption bands and intensified $–CH_2–$ peaks. SA incorporation increased film thickness and moisture content but reduced tensile strength, elongation at break, solubility, and water vapor permeability (WVP). Although the WVP of SA-modified films did not meet the Japanese Industrial Standard at the tested concentrations, the observed trend suggests that higher SA levels could further improve barrier performance. The optimal formulation (0.5% SA) demonstrated enhanced hydrophobicity, acceptable water activity, and moderate tensile strength and opacity. These findings indicate that stearic acid can effectively modify the functional properties of polysaccharide-based edible films, advancing their potential as eco-friendly food packaging materials. Further optimization of SA concentration is recommended to achieve industrial moisture barrier standards.

Keywords: edible film; carboxymethyl cellulose; konjac glucomannan; κ-Carrageenan; stearic acid

1. Introduction

Polysaccharide-based polymers offer an environmentally friendly alternative to petroleum-derived materials in food packaging. With growing concerns about the adverse effects of synthetic polymers on food quality, human health, and ecosystems (Bhatia et al., 2024)., biodegradable options have become increasingly preferred. These polysaccharide materials can be categorized into starch-based and non-starch-based types. However, starch-based bioplastics face limitations in packaging applications due to their inherent hydrophilicity and poor mechanical strength, which compromise their performance in moisture-resistant or high-stress applications (Jayarathna et al., 2022). This underscores the need for safer, more sustainable alternatives to conventional plastics. Edible films represent one such promising solution (Jaya, 2014).

Edible films are thin packaging layers that serve as barriers against mass transfer while potentially acting as carriers for functional additives. This innovative technology supports sustainable food production by reducing packaging waste, extending product shelf life, and providing an eco-friendly alternative to conventional synthetic packaging materials (Larasati et al., 2024). As demonstrated by (Istiani et al., 2023), hydrocolloid-based edible films effectively protect food products from oxygen and carbon dioxide permeation while maintaining excellent mechanical properties. These films can be developed using various biopolymer combinations, with carboxymethyl cellulose (CMC), konjac glucomannan (KGM), and κ-carrageenan representing particularly promising components. Our preliminary research indicates that combining these three biopolymers synergistically enhances the film's mechanical properties beyond what can be achieved with individual components alone.

Carboxymethyl cellulose (CMC), a cellulose derivative modified through carboxymethylation, features a primary structure of β-(1,4)-D-glucopyranose polymer backbone (Haleem et al., 2014). While CMC demonstrates excellent flexibility and film-forming capabilities, it exhibits limited resistance to water vapor transmission (Yildirim-Yalcin et al., 2022). Konjac glucomannan (KGM), composed of β-1,4-glucosidic bonds linking D-glucose and D-mannose units in a 1.6:1 molar ratio, forms viscous solutions in cold water (Cahyarani Heksa et al., 2024). Although KGM

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demonstrates excellent water absorption and significantly increases viscosity, the films often show diminished elasticity and mechanical strength (Wu et al., 2019). Carrageenan, a sulphated polyglactin extracted from red algae (*Rhodophyceae*), contains 15-40% sulphate esters. Its molecular structure consists of alternating α-1,3 and β-1,4 glycosidic bonds connecting D-galactose and 3,6-anhydrogalactose units (Adinda Larasati et al., 2024). While capable of forming stable gels and transparent films, carrageenan-based materials tend to be more brittle and rigider compared to those derived from CMC or KGM.

Previous internal studies revealed that composite films of carboxymethyl cellulose (CMC), konjac glucomannan (KGM), and κ-carrageenan (κCarr) exhibited a water vapor permeability (WVP) of 1. 44×10^{-10}g.$Pa^{-1}\cdot s^{-1}\cdot m^{-1}$, a value exceeding the permissible limits set by the (Japanese Industrial Standard (JIS), 1975). Study by (Thakur et al., 2016) demonstrates that stearic acid (SA) incorporation can significantly reduce WVP to 3.55×10^{-11} $g\cdot Pa^{-1}\cdot s^{-1}\cdot m^{-1}$.The use of stearic acid to enhance the water barrier qualities, specifically to reduce water vapor permeability (WVP) value of edible films based on CMC, KGM, and κCarr is supported by this research. Stearic acid (SA) is a food-grade, non-toxic, and cost-effective compound that exhibits dual functionality in both organic and inorganic systems. Its molecular structure consists of a long nonpolar hydrocarbon chain, which confers solubility in organic solvents, and a polar carboxylic group capable of forming coordinate bonds with metal cations (Bangar et al., 2024). Therefore, this study aims to analyse the effects of SA incorporation on edible films composed of CMC, KGM, and κCarr blends.

2. Materials and Methods

2.1. Materials

The basic components used were CMC from CP Kelco which has a 2% solution viscosity of 313 cPs, konjac glucomannan flour from PT Bumi Mina Tani Indonesia which has a purity of 96%, and κ-carrageenan flour synthesized by (Maulidia et al., 2024). Stearic acid from PT Wilmar Nabati Indonesia was used as the lipid component. Plasticizer was glycerol (85% purity) from Merck KgaA EMSURE ACS Ltd and emulsifier was Tween© 20. Each substance and solvent that was used was of laboratory standard.

2.2. Film Preparation

The procedure for making the edible film involved dissolving a 1% (w/v) solution of the hydrocolloid base mixture—comprising CMC, KGM, and κ-carrageenan (in a ratio of 60:30:10% dry weight)—in distilled water. A magnetic stirrer was used to agitate the mixture. The stirring occurred at 500 rpm for 2 hours at 70°C. Separately, stearic acid at concentrations of 0.1, 0.2, 0.3, 0.4, and 0.5% (w/w) was dissolved in tween 20 (0.2% w/w) at 70°C. After 2 hours of stirring, the stearic acid solution was combined with the hydrocolloid mixture and stirred for an additional hour until fully mixed. Next, it was added in the form of plasticizer, glycerol, 0.5% w/w, and stirring was done for an hour. Once the film-forming solution was homogeneously mixed, it was subjected to ultrasonic treatment (Ultrasonic cleaner Delta D68H; 80kHz) in a water bath for 15 minutes. 20 mL solution were placed to Petri dishes for the casting process, and they were then dried for 24 hours at 45°C in an oven.

2.3. Characterization

The characterization of the edible film included tests for FTIR analysis, thickness, water vapor permeability, moisture content, solubility, water activity, tensile strength, elongation, as well as color and opacity.

2.3.1. FTIR Analysis

FTIR spectroscopy was used to test the films using an Agilent Cary 630 FTIR spectrophotometer equipped with MicroLab software. With wavenumbers spanning from 4000 to 650 cm^{-1}, the spectra were measured at room temperature (30°C). Origin 2025 software was used to examine the generated spectra.

2.3.2. Thickness

Film thickness was measured using a digital micrometer SYNTEX 251767, 0–25 mm range with an accuracy of 0.001 mm. The film sample was placed between the spindle and anvil surfaces of the micrometer and gently compressed until it was flattened. Measurements were taken at five different points on each sample (D = 8 cm), and the average value was used.

2.3.3. Water Vapor Permeability

A modified technique based on ASTM E96, which was taken from (Basha et al., 2011), was used to test water vapor permeability (WVP). Film samples were preconditioned at 50% relative humidity (RH) before testing. Then, 20 grams of silica gel (0% RH) were placed inside an aluminium cup and the film sample was sealed over the cup using plasticine. The sealed cup was then placed in a desiccator containing a saturated sodium chloride (NaCl) solution to maintain a controlled environment at 75% RH. Over the course of seven days, the cup was weighed every 24 hours. To guarantee data reliability, a linear regression was used on the weight increase data, with an acceptance criterion of $R^2 > 0.99$. The weight growth (Δm) over time (Δt) was used to compute the WVP value, which was then represented in units of g/Pa·s·m using the following equation (1).

$$\mathrm{WVP} = \frac{(W/_t)}{A} \times \frac{FT}{P_{in}-P_{out}} \qquad (1)$$

Where W/t is weight loss per unit time, slope of the straight line (g/d), A is the test area (m^2), FT is film's thickness (mm), P_{in} is water vapor partial pressure (Pa) inside the cup and P_{out} is water vapor partial pressure (Pa) outside the cup.

2.3.4. Moisture Content, Solubility, Water Activity

The moisture content (MC) can be found by measurement using a gravimetric technique applied. 1 × 3 cm film strips used to determine their original mass. They were then dried in an oven set at 105°C for 24 h until reach constant weight. The moisture content was then calculated using the following formula in which W_0 is beginning dry weight (g) and W_1 is final dry weight (g).

$$\mathrm{MC}\ (\%) = \frac{W_0 - W_1}{W_0} \times 100 \tag{2}$$

Film solubility was tested alongside moisture content using a modified method from (Thakur et al., 2019). Samples were soaked in 10 mL of distilled water at room temperature for 24 hours without stirring. After that, the undissolved particles were dried for a further twenty-four hours at 105°C until its weight remained constant. Equation (3) was used to calculate solubility, where X_i represents the film sample's original weight and X_f represents its weight following drying.

$$\mathrm{FS}(\%) = \frac{X_i - X_f}{X_i} \times 100 \tag{3}$$

2.3.5. Tensile Strength and Elongation

Tensile strength (TS) and elongation at break (EAB) were measured following (*ASTM D882 TENSILE AND ELONGATION*, 2018) use Trapezium X software (v1.5.1, 2017) on a universal testing machine (Shimadzu EZ-LX, Kyoto, Japan). Film samples (10 × 40 mm) were conditioned at 25°C and 50% RH before testing. Each was mounted between grips, and the maximum load was recorded to calculate TS (MPa) and EAB (%) using equations (4) and (5)

$$\mathrm{EAB}\ (\%) = \frac{L - L_0}{L_0} \times 100 \tag{4}$$

$$\mathrm{TS} = \frac{F}{T \times W} \tag{5}$$

where F is maximum load (N), T is film thickness (mm), W is film width (mm), L_0 is initial length (mm), L is final length at breakage point of film (mm).

2.3.6. Color and Opacity

Film color was measured using a FRU WR-10QC colorimeter at five random points, with results expressed in L*, a*, b* values. Color difference (ΔE) was measured using these parameters according to (Bhatia et al., 2024) in which ΔL*, Δa*, and Δb* reflect variations in color between the standard and the film samples (L* = 100, a* = 3.54, and b* = 4.02).

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \tag{6}$$

Using a UV-Visible spectrophotometer (Thermo Scientific) with a previously developed procedure, film opacity was assessed (Tran et al., 2021). A quartz cuvette containing a 10 x 40 mm film strip was used to measure absorbance at 560 nm. Next, opacity was computed using equation (7), where T is the thickness of the film.

$$\mathrm{Opacity} = \frac{\mathrm{Abs}_{560}}{T} \tag{7}$$

3. Results and Discussion

In this study, the characterizations tested were FTIR, thickness, water vapor permeability, moisture content, solubility, and water activity, tensile and elongation, and color and opacity.

3.1. FTIR Analysis

FTIR spectroscopy analysis of the composite film (Figure 1.) revealed that stearic acid (SA) incorporation modified the chemical structure through two primary mechanisms: enhanced hydroxyl group interactions, evidenced by increased O-H stretching vibration intensity (at 3400 cm^{-1}) (Nandiyanto et al., 2019), and elevated alkyl chain presence, demonstrated by intensified $-CH_2-$ asymmetric/symmetric stretching vibrations (at 2925-2850 cm^{-1}) (Bangar et al., 2024).

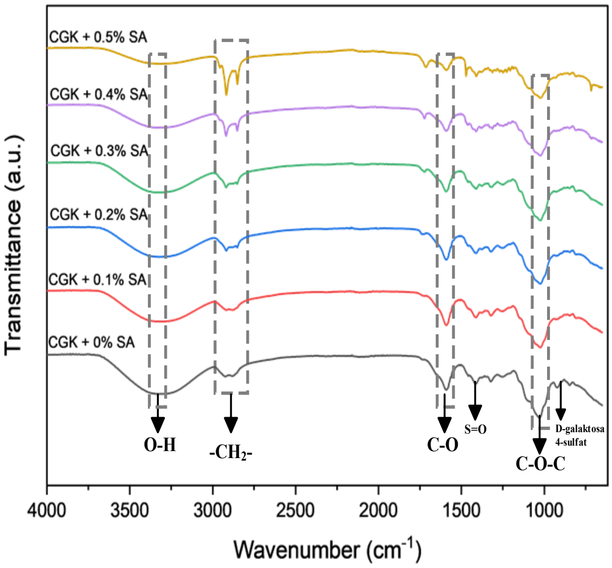

Figure 1. FTIR result of edible film with various stearic acid (SA)

Additional characteristic peaks confirmed the preservation of base components, including C-O stretching of carboxyl groups (at 1050 cm^{-1}), C-O-C glycosidic linkages (at 1150 cm^{-1}) (Ren et al., 2025), and κ-carrageenan's D-galactose-4-sulfate vibrations (at 850 cm^{-1}) (Maulidia et al., 2024), collectively verifying successful SA integration into the CMC-KGM-κCarr matrix through both hydrogen bonding and hydrophobic interactions.

3.2. Thickness

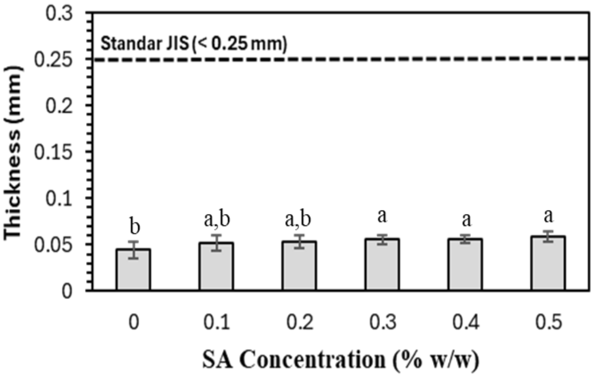

Figure 2. Thickness of edible film with various stearic acid (SA). Within each column, values sharing the same superscript letter indicate no significant difference according to Duncan's multiple range test ($p > 0.05$)

The film thickness is a key factor influencing several other characteristics, including the film's permeability and mechanical strength (Thakur et al., 2016). Based on Figure 2., the addition of stearic acid (SA) significantly increases film thickness and has meet the standard (Japanese Industrial Standard (JIS), 1975) of <0.25 mm. This may be attributed to a reduction in O–H groups with increasing SA concentration, as indicated by the FTIR results in Figure 1. Film thickness is also influenced by the homogeneity of the film. This is influenced by the pace at which the emulsion is stirred. Increased stirring speeds facilitate more even ingredient dispersion inside the film matrix, especially when it comes to SA distribution—thereby reducing pore formation and fatty acid agglomeration (Schmidt et al., 2013).

3.3. Water Vapor Permeability

The addition of stearic acid (SA) is expected to reduce water vapor transmission; however, at concentrations of 0.1–0.5% w/w relative to a 1% base solution, the resulting values do not meet the (Japanese Industrial Standard (JIS), 1975) requirement of less than 7 $g/m^2 \cdot 24h$. Moreover, the inconsistency in the film matrix due to the presence of SA droplets—classified as lipids—can increase water vapor transmission, thereby affecting the film's permeability, particularly at SA concentrations between 0.3–0.5% (Bhatia et al., 2024).

The identical findings were reported by (Oyeyinka et al., 2017), who observed that incorporating SA at concentrations up to 10% (based on a 3% dry starch solution) with Bambara groundnut starch resulted in a WVP value of 1.44×10^{-3} $g \cdot mm \cdot h^{-1} \cdot kPa^{-1} \cdot m^{-2}$, which is 36 times lower than the WVP value shown in Figure 3. (Nzama et al., 2024) also reported that using 2% (w/w dry starch) into starch nanocrystal resulting in 1.44 x 10^{-5} g. $Pa^{-1}.h^{-1}.m^{-1}$ which is greater than WVP value in this study. Likewise, (Thakur et al., 2016) reported that 0.9% SA combined with ι-carrageenan yielded a WVP of 3.5×10^{-11} g/Pa·s·m. These results imply that WVP is significantly affected by the hydrophilic–hydrophobic balance of the film's constituents (Bhatia et al., 2024).

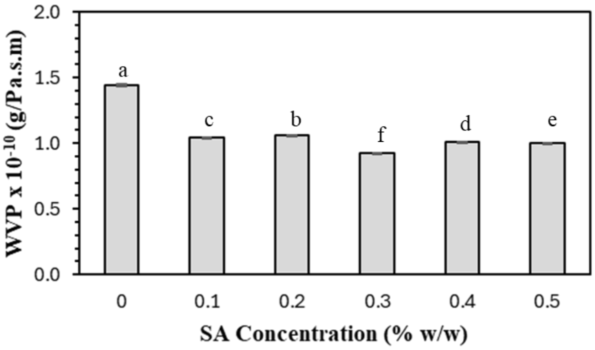

Figure 3. WVP value of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test)

While the hydrocolloid-based film (comprising CMC, glucomannan, and κ-carrageenan) with stearic acid (SA) concentrations up to 0.5% did not meet the standard water vapor permeability (WVP) requirements, our results suggest that higher SA concentrations may potentially enhance the moisture barrier properties and achieve the desired WVP levels.

3.4. Moisture Content, Solubility, Water Activity

To prevent microbial development and maintain the quality of packaged foods, the amount of moisture in the packing is essential (Zhou et al., 2021). Figure 4 illustrates that adding SA tends to raise the edible film's moisture content, however this difference is not statistically significant when compared to the control at the highest SA concentration of 0.5%. This could be attributed to the uneven dispersion of SA within the film matrix, affecting the overall homogeneity of the film (Schmidt et al., 2013). These findings contrast with those of (Zhou et al., 2021), who observed that increasing the concentration of Camellia oil in a KGM–κ-carr–Camellia oil film formulation up to 6% w/w reduced the water content to 5.7%.

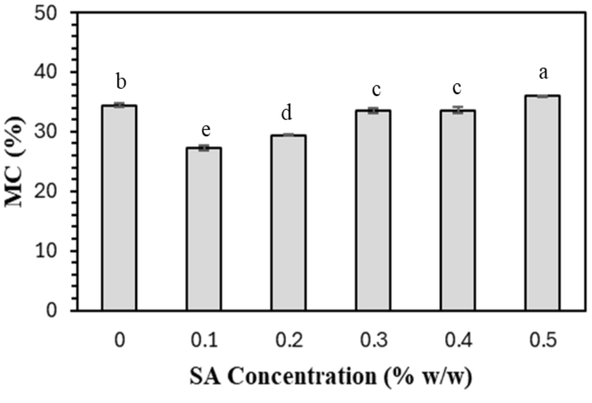

Figure 4. Moisture content of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test)

Similarly, (Oyeyinka et al., 2017) reported that the addition of SA up to 10% had no significant impact on water content. Additionally, the presence of plasticizers such as glycerol and emulsifiers like tween 20 may contribute to the water content of the film, as both compounds contain hydroxyl groups (O–H) as shown in Figure 1. and exhibit hydrophilic characteristics (Larasati et al., 2024).

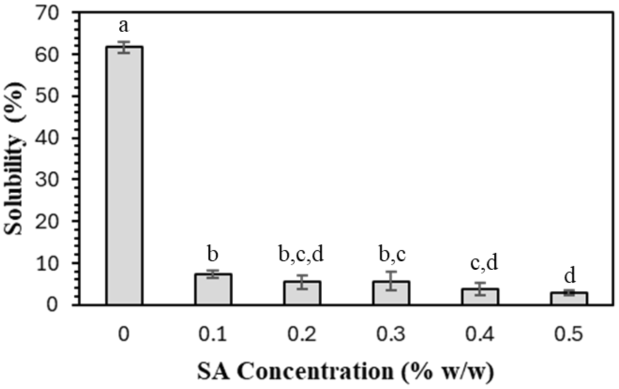

Figure 5. Solubility of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test)

The solubility of an edible film reflects its tendency to be either hydrophilic or hydrophobic. As shown in Figure 5., the solubility of the CMC–KGM–κ-carr edible film with the addition of stearic acid (SA) ranges from 2.78% to 7.29%. These values are comparable to those reported by (Zhou et al., 2021), who observed a decrease in solubility from 21.1% to 8.7% with increasing concentrations of camellia oil (CO), a lipid-based additive.

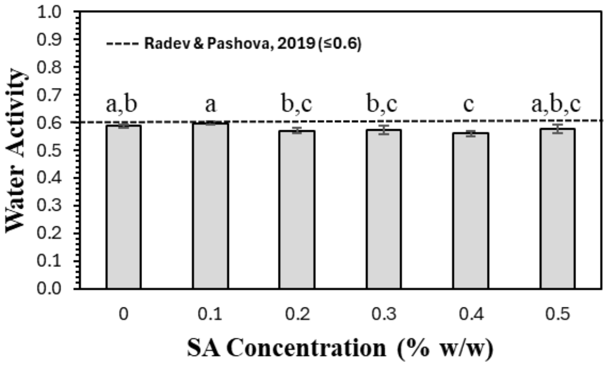

Figure 6. Water activity of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test)

However, the solubility values in the present study are significantly lower than those reported by (Oyeyinka et al., 2017), where the addition of SA to starch-based films resulted in solubility values ranging from 37.73% to 46.53%. In Figure 5., a decreasing trend in solubility is observed with the addition of SA, compared to the control. This aligns with previous studies indicating that increasing the concentration of hydrophobic agents typically reduces the water solubility of edible films. Therefore, the CMC–KGM–κ-carr film formulated with SA can be considered to exhibit more hydrophobic characteristics.

Based on Figure 6., water activity (Aw) shows a decreasing trend with the addition of stearic acid. The resulting values comply with the established standard of Aw < 0.6 (Radev & Pashova, 2019). A lower water activity value enhances the film's resistance to microbial spoilage, thereby extending its shelf life even during prolonged storage.

3.5. Tensile Strength and Elongation

The tensile strength of the CMC:KGM:κ-carr edible film with the addition of stearic acid (SA) ranged from 2.37 to 5.03 MPa, as shown in Figure 7. The addition of SA tends to reduce the tensile strength of the film. This reduction may be attributed to the presence of SA, a lipid component, which can form hydrogen bonds that replace the original intermolecular bonds among CMC, KGM, and κ-carrageenan molecules. As a result, the film structure becomes less dense and more irregular, leading to decreased mechanical strength and increased brittleness (Indriyati et al., 2020).

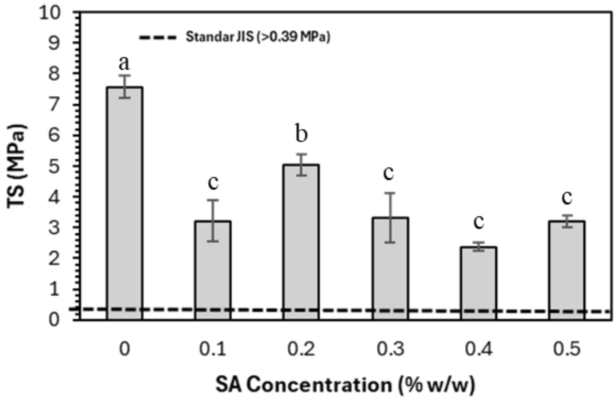

Figure 7. Tensile Strength (TS) of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test)

The study by (Liu et al., 2025) reported that while SA coatings significantly enhanced hydrophobicity, this improvement was accompanied by reduced transparency, diminished mechanical performance, and altered color properties. Similarly, (Oyeyinka et al., 2017) reported that incorporating SA into starch-based films disrupted the starch matrix, resulting in a less cohesive and mechanically weaker structure. The use of emulsifiers and plasticizers, such as tween 20 and glycerol, also contributed to the reduction in tensile strength (TS) while increasing elongation at break (EAB) due to their hydrophilic nature and plasticizing effect (Ziani et al., 2008). Despite the decrease, the TS values observed in this study are higher than those reported for κ-carr–SA films by (Praseptiangga et al., 2023), which ranged from 2.52 to 3.15 MPa.

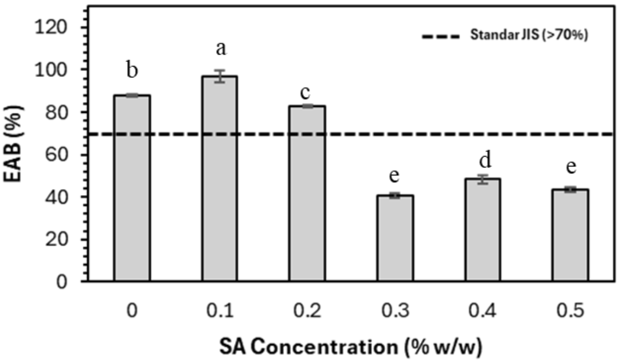

Figure 8. Elongation of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test).

The elongation of the CMC:KGM:κ-carr edible film with the addition of stearic acid (SA) ranged from 40.56% to 96.83%, as shown in Figure 8. These values indicate that most of the formulations met the packaging standard for elongation, which is greater than 70%, as defined by the (Japanese Industrial Standard (JIS), 1975). Although (Anker et al., 2002), as cited in (Wei et al., 2015), reported that lipids such as acetem could enhance elongation, the present study observed a different trend: the elongation values decreased when SA was added. This discrepancy may be due to the distinct chemical characteristics of stearic acid, which is more hydrophobic and tends to form aggregates within the hydrophilic film matrix. These aggregates can disrupt the continuity of the polymer network, thereby reducing the flexibility and extensibility of the film.

3.6. Color and Opacity

Based on Table 1. and Table 2., the addition of SA to the edible film formulation increases whiteness and decreases transparency. This is reflected in the elevated L* values (approaching 100), along with a rise in opacity from 5.739% in the control to approximately 9.5% at 0.5% SA concentration.

Table 1. Color analysis of of edible film with various stearic acid (SA)

Sample	L* Value	a*	b*	ΔE
CGK + 0% SA	99.6898	4.01	5.176	1.55
CGK + 0.1% SA	98.4628	3.924	5.046	2.06
CGK + 0.2% SA	98.4738	3.956	5.608	2.47
CGK + 0.3% SA	98.5285	3.886	5.734	2.50
CGK + 0.4% SA	98.6205	3.882	4.898	1.85
CGK + 0.5% SA	99.3716	3.736	5.246	1.64

While the control film appeared more transparent with a slight yellowish tint (indicated by a positive b* value), the addition of SA resulted in a more opaque and whiter appearance, although the brightness (L*) remained consistently high. Physical appearance of the edible film shown in Figure 9.

Figure 9. Physical appearance of edible film with various stearic acid (SA)

The decrease in film transparency upon lipid incorporation can be explained by increased light scattering, which arises from the different refractive indices of the dispersed lipid phase and the continuous hydrocolloid matrix (Thakur et al., 2017). Increasing stearic acid (SA) concentrations introduce more lipid particles into the film matrix, thereby amplifying this scattering effect. This phenomenon manifests as both decreased transparency and increased whiteness. Notably, films containing 0.5% SA demonstrated the most pronounced effects, exhibiting the highest opacity and most distinct whitish appearance among all tested formulations, as quantitatively confirmed by the opacity measurements presented in Table 2.

Table 2. Opacity of of edible film with various stearic acid (SA). Significant differences ($p < 0.05$) are denoted by distinct letters within columns (Duncan's test)

Sample	Opacity (%)
CGK + 0% SA	5.738 ± 0.0413^{c}
CGK + 0.1% SA	3.385 ± 0.0157^{f}
CGK + 0.2% SA	5.571 ± 0.0107^{e}
CGK + 0.3% SA	5.627 ± 0.0155^{d}
CGK + 0.4% SA	6.833 ± 0.0393^{b}
CGK + 0.5% SA	9.526 ± 0.0127^{a}

4. Conclusions

The incorporation of stearic acid (SA) into CMC-KGM-κCarr edible films enhanced barrier properties but concurrently altered their physical characteristics. SA addition resulted in increased film thickness (0.061 mm at 0.5% SA) and moisture content (35.95%), while reducing tensile strength (2.53 MPa), elongation (14.09%), solubility (2.78%), and WVP (0.99×10^{-10} g·Pa^{-1}·s^{-1}·m^{-1}). FTIR analysis confirmed these modifications, showing attenuated O–H group absorption and intensified $-CH_2-$ group peaks, indicative of hydrophobic interactions. Optimal performance was achieved with 0.5% SA, yielding balanced properties, such as water activity (0.577), optical characteristics (99.37% color value, 9.5% opacity), and mechanical parameters. These results demonstrate SA's dual role as both a barrier enhancer and a structural modifier in polysaccharide-based films.

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Statement
In preparing this work, the authors utilized ChatGPT 4.0 and QuillBot to assist with language enhancement and proofreading. The authors subsequently reviewed and revised the content as necessary and take full responsibility for the final version of the publication.

CRediT authorship contribution statement
Lintang Dian Widyasti: Writing –review & editing, Writing –original draft, Visualization, Investigation, Formal analysis.
Siti Nurkhamidah: Validation, Resources, Conceptualization, Supervision.
Wahyu Meka: Validation, Resources, Conceptualization, Supervision.

Declaration of competing interest
The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability
The data that has been used is confidential.

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