Development of Natural Polymer-Based Edible Films from Carrageenan and Pectin: Mechanical and Physical Properties

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Abstract. This study aimed to evaluate the effect of pectin addition on the physical and mechanical characteristics of carrageenan-based edible films as an environmentally friendly food packaging material. Pectin addition was carried out at various concentrations of 0–7%, and characterization included tensile strength, elongation at break, thickness, water content, water solubility, film color, and surface morphology analysis using SEM. The results showed that pectin addition increased tensile strength from 24.12 MPa to 38.12 MPa and elongation at break from 59.14% to 70.79%, indicating improvements in mechanical properties and film flexibility. Film thickness, moisture content and water solubility also increased with pectin addition, indicating the hydrophilic nature of pectin in the film matrix. Color analysis showed a decrease in brightness and a slight increase in reddish and yellowish hues, but the color changes remained within acceptable limits ($\Delta E < 3$). SEM analysis showed a denser film surface and indicated aggregation. Overall, the addition of pectin to carrageenan edible film can improve the mechanical and functional quality without films sacrificing visual appearance, thus having the potential to be a biodegradable food packaging material that supports sustainability.

1 Introduction

The problem of environmental pollution caused by synthetic plastic waste has become a global concern. Conventional plastics widely used in the food packaging industry are non-biodegradable, resulting in long-term negative impacts on the ecosystem. Edible film has been developed as an environmentally friendly packaging alternative. Edible film is a thin, edible layer made from natural, biodegradable materials that can be used as a substitute for plastic in food packaging [1]. One of the advantages of edible film is its ability to protect food products from external influences such as oxygen, moisture, and microorganisms, while also being safe for consumption with the food product.

Edible films are produced from edible biopolymers and additives such as plasticizers. Polysaccharide-based films have attracted attention due to their beneficial characteristics, including their diversity of sources, suitability for thermal treatment, and their function as efficient oxygen and carbon dioxide barriers compared to proteins or lipids [2]. Carrageenan is a polysaccharide extracted from red seaweed, and is a potential ingredient in the manufacture of edible films. Carrageenan has good gel-forming ability, is transparent, and flexible, making it suitable for use as a film matrix. There are three main types of carrageenan based on the number and position of sulfate groups on the galactose/anhydrogalactose chain: κ- (kappa), α- (iota), and β- (lambda) carrageenan containing one, two, and three sulfate groups per disaccharide repeat unit [3]. Of the three types, κ-carrageenan has the best gel-forming properties and can therefore be used in film formation. Despite its potential, carrageenan tends to become brittle during the drying process due to its low mechanical stability and the formation of a double helix in its matrix. To improve film characteristics, carrageenan can be combined with pectin. The addition of pectin offers dual benefits, particularly in improving the mechanical and functional properties of the material [4]. Pectin is a natural polysaccharide found abundantly in the cell walls of green plants, particularly in fruits such as apples, lemons, and oranges. Pectin is widely used in the food industry because it is biocompatible, non-toxic, and anionic, useful as a gelling agent and stabilizer. Furthermore, pectin is an interesting new biopolymer with potential applications in the pharmaceutical and healthcare sectors due to its unique characteristics. Pectin can be a good drug delivery agent due to its chemical composition and functional groups present on its surface.

The combination of pectin and κ-carrageenan has been known for its excellent film-forming properties. This composite material has been reported to produce transparent materials [5]. Furthermore, pectin contains a high concentration of carboxyl groups, while carrageenan is rich in hydroxyl groups. This combination of functional groups can improve the properties of films made from both materials, making it a promising blend for the development of edible films. This can result in the formation of hydrogen and ionic bonds between the two polymers, resulting in the formation of a stable cross-linked network with smaller intermolecular distances. Such a polymer network with

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smaller intermolecular distances can exhibit better compactness. This approach can overcome the limitation of low mechanical strength in carrageenan gels. Thus, this study aims to develop and evaluate the physical and mechanical characteristics of carrageenan and pectin-based edible films. The results are expected to contribute to the development of environmentally friendly, applicable and sustainable packaging in the food industry.

2 Materials and Method

2.1 Materials

Pectin was supplied by PT Reinaldoi (Surabaya, Indonesia) and the average particle size of pectin was 70 nm. Kappa-carrageenan (molar mass 401.32 g/mol with CAS number 11114- 20-8 SIGMA-Aldrich, USA). Polyethylene glycol (PEG) 400 and HCl were supplied by PT Indrasari (Semarang, Indonesia).

2.2 Preparation of the CAR/PEC gel solution

The gel solution was prepared with 5 gr CAR and different proportions of PEC (based on 0, 1, 3, 5, 7 gram). Different proportions of CAR/PEC were added to a beaker of 100 mL distilled water and then slowly added 4 mL PEG 400. The mixed solution was stirred at a high speed (530 rpm) at 70 °C for 20 min, and then stirred at a low speed (200 rpm) for 20 min to obtain the transparent gel solution.

2.3 Preparation of the CAR/PEC film

The mixed solution was put directly onto the tray and dried at 55°C until the CAR/PEC film with different ratios of PEC formed. CAR/PEC-0, CAR/PEC-1, CAR/PEC-3, CAR/PEC-5, and CAR/PEC-7 represented different carrageenan-based film with 0, 1, 3, 5, and 7 gram of pectin, respectively.

2.4 Characterization of CAR/PEC film

2.4.1 Tensile Strength and Elongation at Break

The tensile strength and elongation at break were measured at 27 °C using a Testometric Machine M350-10CT (Testometric Co., Ltd., Rochdale, Lancs, UK) according to the American Society for Testing and Materials (ASTM) ID: D882-12. All samples were tested with the same size (5 cm × 10 cm) and equilibrated at 28 °C and 87% RH in a desiccator containing silica crystals to extract the saturated water solution for 48 h before testing. The cross-head speed was maintained at 1 mm/s.

2.4.2 Moisture Content

The water content of the sample is calculated by cutting the sample, then weighing 1 gram, then putting it into a cup. Next, the cup containing the sample is heated at a temperature of 110 ° C for 3 hours in an oven. The sample is cooled in a desiccator for 30 minutes and then weighed, this last step is repeated until the weight is constant. The water content is calculated using the following equation:

$$MC\ (\%)=\frac{W_1-W_2}{W_1}\times 100 \quad (1)$$

where W_1 is the initial sample weight and W_2 is the final sample weight.

2.4.3 Water Solubility

The solubility of the emulsion film was measured by cutting and weighing the sample, then the sample was soaked in 50 ml of distilled water at room temperature for 24 hours with periodic stirring. Furthermore, the wet specimen was dried again at 105 ^{o}C for 24 hours and weighed. The solubility was calculated by the following equation:

$$WS\ (\%)=\frac{W_d-W_f}{W_f}\times 100\% \quad (2)$$

where W_d is the weight of the dry sample and W_f is the weight of the final sample.

2.4.4 Thickness

The thickness of the edible film was tested using a pre-calibrated micrometer screw, with measurements taken at 3 random points on the edible film.

2.4.5 Color

The color of the formed edible film was analyzed using a WR-10QC colorimeter. Color analysis included brightness (L*), redness (a*), yellowness (b*), and total color difference (ΔE).

2.4.6 SEM Analysis

Morphological analysis was conducted to determine the morphology in the form of shape, surface structure, and texture of a sample. The morphology of the sample was analyzed using SEM JEOL JSM-6510LA.

3 Result and Discussion

3.1 Mechanical Properties

The effect of pectin addition on tensile strength and elongation at break is shown in Table 1. The results showed that the addition of pectin to the carrageenan edible film increased the tensile strength value from 24.12 ± 0.07 MPa (CAR/PEC0) to 38.12 ± 0.12 MPa (CAR/PEC5). This increase indicates that the addition of pectin can strengthen the film polymer network. This can occur because the hydrogen interaction between the hydroxyl groups of carrageenan and the carboxyl groups of pectin forms a tighter intermolecular bond and increases the cohesiveness of the film network [6]. However, at the highest pectin concentration (CAR/PEC7), the tensile strength decreased slightly to 35.12 ± 0.18 MPa. This can be attributed to the presence

of pectin agglomeration at high concentrations, which causes an inhomogeneous distribution of pectin within the carrageenan matrix, resulting in the formation of weak points in the film structure. Previous research also reported that increasing pectin concentration above the optimum point can lead to an excess of the dispersed phase, thus affecting the mechanical integrity of the biopolymer film [7].

Table 1. Mechanical properties of CAR/PEC film

Sample	Tensile Strength (MPa)	Elongation at Break (%)
CAR/PEC0	24.12 ± 0.07	59.14 ± 0.29
CAR/PEC1	27.20 ± 0.09	63.21 ± 0.61
CAR/PEC3	33.00 ± 0.19	67.32 ± 1.32
CAR/PEC5	38.12 ± 0.12	70.79 ± 0.60
CAR/PEC7	35.12 ± 0.18	66.04 ± 0.23

Furthermore, the elongation at break value also increased from 59.14 ± 0.29% in CAR/PEC0 to 70.79 ± 0.60% in CAR/PEC5, indicating increased film flexibility with the addition of pectin. Pectin acts as a natural plasticizer that can increase the mobility of polymer chains in the carrageenan film matrix by reducing excessively rigid intermolecular interactions, resulting in a more elastic film [8]. The elongation at break value slightly decreased in CAR/PEC7 (66.04 ± 0.23%) which may be caused by increased excessive intermolecular bonds at high pectin concentrations, thereby reducing the flexibility of the polymer chains. Recent research has shown that a balance between tensile strength and film flexibility is necessary to produce films with optimal mechanical performance as packaging materials [9].

3.2 Physical Properties

The results of the carrageenan/pectin edible film thickness test with varying pectin concentrations are shown in Figure 1. It can be seen that the addition of pectin concentration to the film formulation causes an increase in film thickness. The film thickness in the CAR/PEC0 sample (without pectin) was recorded at around 0.11 mm, while the thickness increased to around 0.23 mm in the CAR/PEC7 sample with the highest pectin concentration. This increase in thickness occurs because the addition of pectin increases the total amount of solids in the film-forming solution, so that during the drying process, more solid residues are formed and produce a thicker film [10]. In addition, pectin has the ability to form a gel and bind with carrageenan through hydrogen interactions, resulting in a denser and thicker matrix structure in the film. The addition of pectin also increases the viscosity of the film-forming solution, which contributes to the increase in thickness due to the formation of a thicker film layer during the casting and drying processes. The higher the pectin concentration, the higher the viscosity and soluble solids remaining after water evaporation, thus providing a greater thickness to the resulting film.

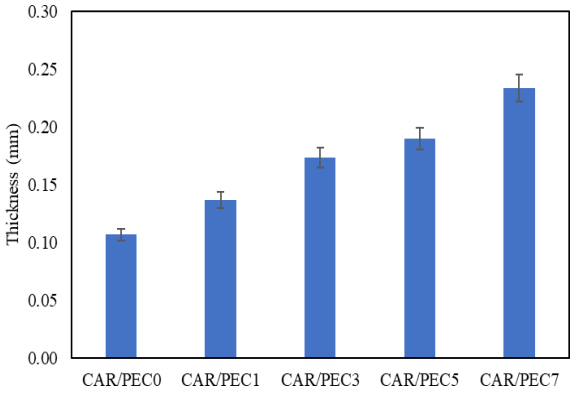

Fig. 1. Thickness of CAR/PEC film

The results of the moisture content test of carrageenan edible film with the addition of pectin are shown in Table 2. It can be seen that the moisture content increased with the addition of pectin, from 11.16 ± 0.26% (CAR/PEC0) to 14.55 ± 0.29% (CAR/PEC5). However, at the highest pectin addition (CAR/PEC7), the moisture content decreased slightly to 13.36 ± 0.38%. This increase in moisture content can be attributed to the hydrophilic nature of pectin which has many hydroxyl and carboxyl groups, thereby increasing the ability of the film matrix to bind water [11]. The addition of pectin increases the water retention capacity of the film network, which can also help maintain film flexibility. However, at very high pectin concentrations (CAR/PEC7), a decrease in moisture content can occur due to increased intermolecular interactions (intra- and inter-molecular hydrogen bonds) which limit the space for free water binding in the film matrix [6].

Table 2. Moisture content and water solubility of CAR/PEC film

Sample	Moisture Content (%)	Water Solubility (%)
CAR/PEC0	11.16 ± 0.26	48.32 ± 0.59
CAR/PEC1	12.63 ± 0.43	52.01 ± 0.23
CAR/PEC3	13.67 ± 0.38	54.55 ± 0.50
CAR/PEC5	14.55 ± 0.29	58.30 ± 0.18
CAR/PEC7	13.36 ± 0.38	53.31 ± 0.40

The water solubility of edible films also increased with the addition of pectin, from 48.32 ± 0.59% (CAR/PEC0) to 58.30 ± 0.18% (CAR/PEC5). At CAR/PEC7, the

solubility value also decreased to 53.31 ± 0.40%. The increase in film solubility with the addition of pectin is related to the hydrophilic nature of pectin which increases the film's affinity for water, thus increasing the amount of film that dissolves in water during the test [12]. However, the decrease in solubility at high pectin concentrations (CAR/PEC7) may occur due to the formation of a denser polymer network and stronger intermolecular bonds at high pectin concentrations, thereby reducing the water-soluble fraction of the film. This indicates the existence of an optimum concentration of pectin in the film formulation to produce solubility that meets application requirements.

Based on Table 3, increasing the concentration of pectin in CAR/PEC edible films showed a decrease in the L* (brightness) value and an increase in the a* (redness), b* (yellowness), and ΔE (total color change) values. The L* value decreased from 89.77 in CAR/PEC0 to 88.53 in CAR/PEC7, indicating that the film darkened with increasing pectin concentration. The decrease in the L* value may be due to increased film thickness at higher pectin concentrations, which reduces light transmission through the film, making it appear more opaque and dark [13]. Pectin also has a natural yellowish color that can increase the b* value of the film, as seen in the increase in the b* value from 10.73 in CAR/PEC0 to 11.23 in CAR/PEC7.

Table 3. Color parameter of CAR/PEC films

Sample	L*	a*	b*	ΔE
CAR/PEC0	89.77 ± 0.34	0.03 ± 0.05	10.73 ± 0.08	0.00 ± 0.00
CAR/PEC1	89.46 ± 0.41	0.05 ± 0.04	10.89 ± 0.07	0.35 ± 0.05
CAR/PEC3	89.08 ± 0.39	0.18 ± 0.06	11.01 ± 0.05	0.76 ± 0.06
CAR/PEC5	88.72 ± 0.42	0.37 ± 0.03	11.14 ± 0.06	1.18 ± 0.07
CAR/PEC7	88.53 ± 0.45	0.48 ± 0.04	11.23 ± 0.04	1.41 ± 0.09

The increase in the a* value also occurred, which may be attributed to the interaction between carrageenan and pectin during film formation, which affects the film's optical characteristics. The addition of pectin increases the total solids and film thickness, allowing the natural pectin pigment to be more trapped in the film matrix and producing a more intense color. The increase in the ΔE value indicates a more pronounced color difference compared to the control as the pectin concentration increases. According to visual standards, ΔE value between 1–2 is usually still difficult to detect by the human eye [14]. The resulting color change was still within a visually insignificant range, making it acceptable for food packaging applications requiring a transparent appearance.

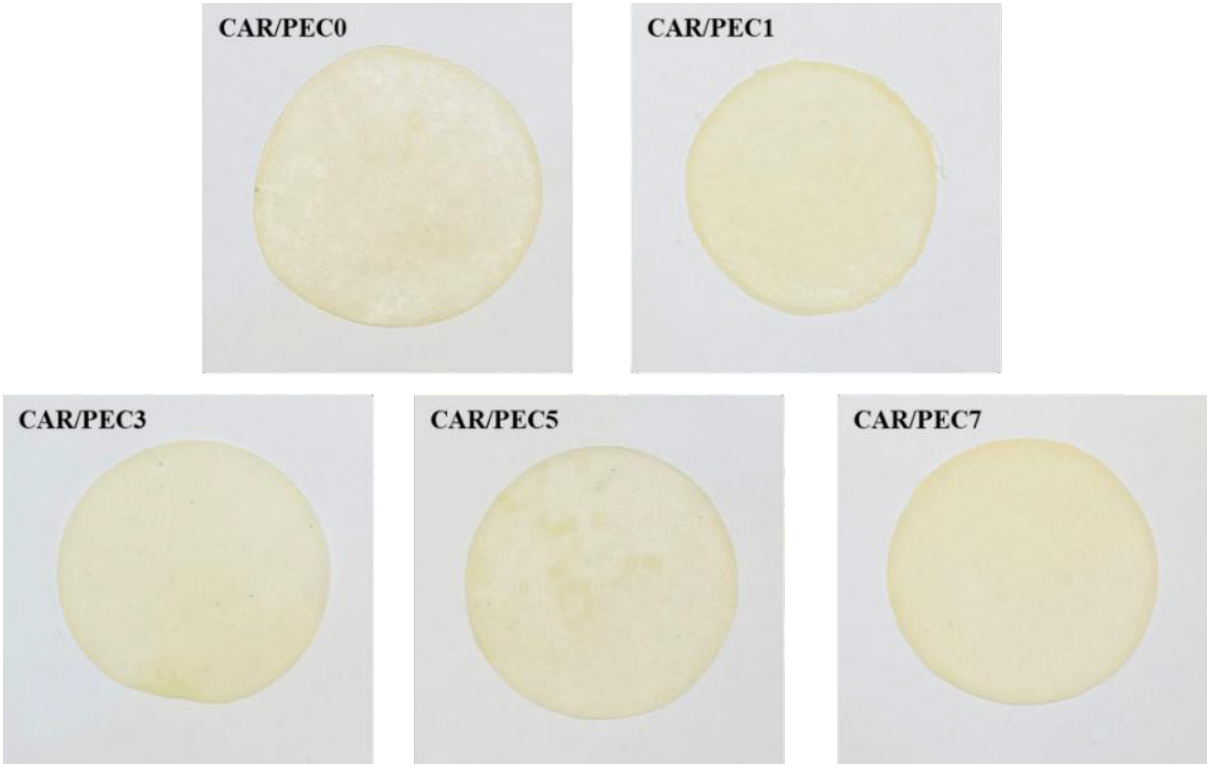

Fig. 2. Visual appearance of CAR/PEC films

3.3 SEM Analysis of CAR/PEC Film

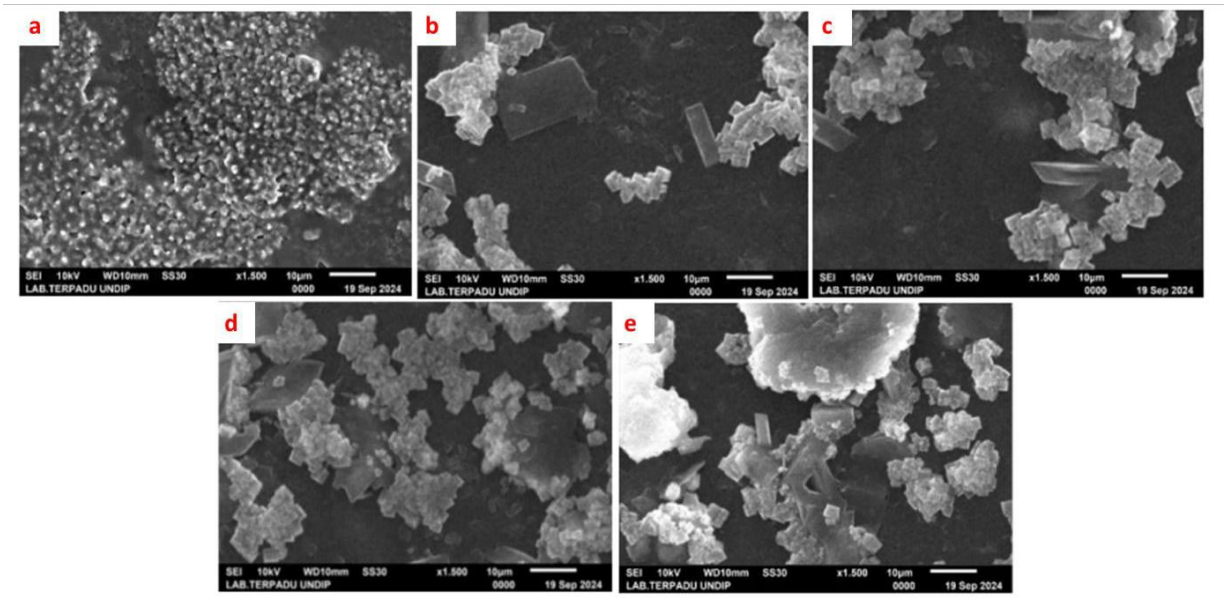

Fig.3. SEM of (a) CAR/PEC0; (b) CAR/PEC1; (c) CAR/PEC3; (d) CAR/PEC5; (e) CAR/PEC7

Based on Figure 3a, film without pectin exhibited a more homogeneous surface compared to capsules with added pectin. The capsule morphology became more heterogeneous when pectin was added. Figure 3 (b, c, d, and e) shows a lattice-like pattern scattered throughout the film. This indicates the presence of aggregates or pectin-rich areas that were not completely mixed within the kappa-carrageenan matrix. This is in line with the research of Bai et al. (2023) which stated that film with high pectin concentrations showed white spots as a sign of pectin aggregates and made the film surface rough [15]. The addition of pectin increases electrostatic repulsion interactions because kappa-carrageenan has negatively charged sulfate groups, while pectin also has negatively charged carboxylate groups. These two molecules tend to repel each other electrostatically. This electrostatic repulsion can cause clumping, resulting in a less homogeneous solution.

4 Conclusions

In summary, the addition of pectin to carrageenan edible film improves the mechanical properties of the film by increasing the tensile strength and elongation at break up to a certain concentration, indicating an increase in the strength and flexibility of the film. The addition of pectin also reduces the film thickness, increases the water content and water solubility indicating the hydrophilic nature of pectin in the film matrix. SEM analysis shows aggregation on the film surface. Meanwhile, color analysis shows a decrease in brightness and a slight increase in redness and yellowness, but the color changes remain within the visually acceptable range. These results indicate that the addition of pectin can improve the quality of carrageenan edible film as an environmentally friendly food packaging material with good mechanical properties, flexibility, and visual appearance.

The authors gratefully acknowledge the financial support of Diponegoro University through the Riset Profesor 2025 research scheme. The authors are also grateful to the Faculty of Chemical Engineering, Diponegoro University, Chemical Engineering Laboratory for the convenience in carrying out this research.

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