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Research Article

Characteristics of Different Chitosan Types on κ-Carrageenan Polyelectrolyte Complex (PEC) Bioplastics as Food Packaging

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Abstract

Bioplastic represents an eco-friendly alternative to synthetic plastic and can be derived from natural polysaccharides like carrageenan. κ-carrageenan is noted for its gel-forming properties, making it a common raw material for bioplastics. Mechanical properties values of κ-carrageenan bioplastics are usually below standard. The addition of materials such as chitosan can enhance those properties. Chitosan-carrageenan can form polyelectrolyte complex (PEC) through electrostatic interactions without toxic crosslinking agents. Polymer's molecular weight is a crucial factor influencing PEC formation. Chitosan's molecular weight varies based on the raw material and extraction process. This study aims to identify the most suitable type of chitosan for food packaging bioplastics using polyelectrolyte complex (PEC) method. Three types of commercial chitosan with different molecular weights were evaluated (shrimp, crab, and fish scale chitosan). Japanese Industrial Standards (JIS) were used for characterization assessment of bioplastics such as thickness, tensile strength, water resistance, water vapor transmission, and biodegradation rate as well as additional tests including Fourier Transform Infrared Spectroscopy (FTIR), Scanning Electron Microscopy (SEM) analysis, and Total Plate Count (TPC) on fish fillets. The findings indicated that crab chitosan-κ carrageenan PEC bioplastic exhibited optimal results with a thickness of 0.178 mm, tensile strength of 18.053 MPa, elongation at break at 211.73%, water resistance of 63.94%, water vapor transmission (WVT) of 0,001456 g/m2/day, biodegradation rate of 3.358% over 7 days, and the lowest TPC in fish fillets after 24 h, increasing from 4.39 log CFU/g to 7.45 log CFU/g. The molecular weight of chitosan was shown to significantly influence the PEC bioplastics' characteristics.

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1. Introduction

The proliferation of synthetic plastic production, primarily driven by its application in food packaging such as shopping bags, food wrappers, and other related materials has raised significant environmental concerns due to its reliance on fossil fuel-based raw materials characterized by high strength and environmental stability (Ncube *et al*., 2021; Qasim *et al*., 2021). These attributes contribute to widespread plastic pollution, underscoring the urgent need for eco-friendly alternatives that offer comparable strength and stability. Bioplastics have emerged as a promising solution in this context. Nowadays, bioplastics have been developed to meet the needs of food packaging with new innovations that are safer than conventional plastics, such as bioplastics that can detect food spoilage with bioindicators (Rahmadhia *et al.*, 2022; Wang *et al*., 2022) and bioplastics that have antibacterial, antifungal, and antioxidant activity to inhibit the deterioration of food product quality (Abang *et al*., 2023). Effective bioplastics for food packaging must adhere to stringent criteria, including biodegradability, derivation from renewable sources, barrier properties, non-toxicity, and mechanical strength equivalent to conventional plastics, as delineated by the Japanese Industrial Standards (JIS).

Extensive research has explored the utilization of bioplastics as food packaging materials. Bioplastic raw materials commonly used are derived from polysaccharides such as carrageenan, which is extracted from seaweed. Indonesia is one of the largest seaweed producers in the world, with a production of 9.12 million tons in 2021 (Jaya *et al*., 2023). This abundance of seaweed allows for the sustainable production of carrageenan, ensuring it does not compete with public foodstuffs. The use of carrageenan in bioplastics offers several advantages, including low carbon footprint, biodegradability, and the potential to reduce environmental pollution caused by traditional plastics. κ-carrageenan is common bioplastic raw material derived from seaweed that has been widely used due to its high gel strength among other types of carrageenan (Adam *et al*., 2022; Fransiska *et al*., 2021). The higher gel strength of κ-carrageenan is due to the lower ester sulfate in κ-carrageenan compared to other types of carrageenan (Lim *et al*., 2021). κ-carrageenan can enhance the mechanical properties of bioplastics, such as improving tensile strength and thermal stability. Bioplastics based on κ-carrageenan are hydrophilic and have low mechanical strength so it requires additives such as chitosan to enhance their physical properties (Sedayu *et al*., 2019; Sudhakar *et al*., 2021). Chitosan is derived from fishery by-products and has been used extensively due to its compatibility, environmental degradability, antibacterial properties, and non-toxicity (Chen *et al*., 2023; Maliki *et al*., 2022; Roy *et al*., 2024). Chitosan as a reinforcement material in bioplastic has been shown to improve mechanical characteristics by filling in the empty spaces between the matrix such as water resistance, tensile strength, and heat stability of corn starch bioplastic (Tan *et al*., 2022) and enhancing the mechanical and barrier properties of tapioca starch bioplastic (Shapi'i *et al*., 2022).

However, combining the two materials using conventional methods often fails due to difference in charges that can lead to the frequent use of hazardous chemical crosslinkers to achieve stability. Hazardous chemical crosslinkers include genipin, glutaraldehyde, epichlorohydrin, and others that have mutagenic and toxic properties (Sapula *et al*., 2023). The synergistic interaction between the sulfate groups of κ-carrageenan and the amine groups of chitosan has been shown to form stable bonds in a hydrogel system (Kołodziejska *et al*., 2021; Qureshi *et al*., 2024), which can be adopted to improve the mechanical properties of bioplastic. The electrostatic interaction-based self-crosslinking method through the formation of polyelectrolyte complexes (PECs) is a promising approach to avoid the use of hazardous crosslinkers while maintaining stability (Ismillayli *et al*., 2020). The formation of stable electrostatic interactions between oppositely charged polyelectrolytes in an aqueous medium leads to the formation of a precipitated solid phase known as PEC. Compared to products without PEC, PEC exhibits several advantages, including improved mechanical properties such as uniform component distribution, enhanced tensile strength, increased resistance to oxygen transmission, and other benefits (Li *et al*., 2021; Ushimaru *et al*., 2021).

The properties of polyelectrolyte complex (PEC) bioplastics are influenced by several factors, including temperature, pH, and molecular weight of the material (Carvalho *et al*., 2021). Molecular weight of the material significantly impacts thickness, tensile strength, water sensitivity, oxygen transmission, and other characteristics. Chitosan molecular weight is generally categorized into three categories: low (<100 kDa), medium (100–1000 kDa), and high (>1000 kDa) (Gonçalves *et al*., 2021). The molecular weight of chitosan is influenced by the glucosamine unit in the polymer chain, where each glucosamine unit contains one amine group. Lower molecular weight chitosan has fewer amine groups, which can affect the quality of bioplastics. Research by Liu *et al.* (2020) demonstrated that high molecular weight chitosan (110 kDa) can improve mechanical properties, barrier performance, water resistance, and maintain the quality and stability of packaged products. In contrast,

low molecular weight chitosan is less commonly used in bioplastic production without PEC due to its lower amine groups, resulting in substandard quality.

However, the PEC method can utilize low molecular weight chitosan due to its high stability and solubility in industrial applications (Alemu *et al.*, 2023). Chitosan from shrimp shells, crab shells, and fish scales has different molecular weights, enabling the production of food packaging bioplastics with distinct characteristics. Tilapia fillets products dominate Indonesia's tilapia exports and successfully penetrate into the United States market (Dai *et al.*, 2020). Exported tilapia fillets are typically packaged in non-biodegradable plastic materials to protect the product from the external environment. Therefore, tilapia fillets were used as samples to be wrapped with bioplastic in this study. Numerous studies have characterized bioplastics derived from various forms of chitosan extracted from shrimp, crab, and fish shells through the formation of polyelectrolyte complexes (PEC) with kappa carrageenan. However, the application of these bioplastics as food packaging materials, specifically for fish fillets, remains underexplored.

The primary objective of this study is to investigate the properties of polyelectrolyte complex (PEC) bioplastics produced using κ-carrageenan and various types of chitosan derived from shrimp, crab, and fish scales. Each type of chitosan has a different molecular weight that can lead to varying electrostatic interactions with κ-carrageenan and ultimately affect the bioplastic properties. The outcome of this study will provide insights into the optimal type of chitosan with different molecular weights to produce bioplastics that meet bioplastic standards without relying on hazardous chemical crosslinkers. This research aims to contribute to the development of sustainable and eco-friendly food packaging materials that meet bioplastic standards.

2. Materials and Methods

2.1 Materials

2.1.1 The equipment

The equipment used in this research are analytical scale (PA313, Ohaus Pioneer, US); micrometer (device); pH meter (WD3541903, Oakton, Singapore); hot plate magnetic stirrer (Cimarec, Thermo Fisher Scientific, US); waterbath; oven; silica gel; measuring glass; stative clamp; thermometer; and 25×15×7 cm^3 glass mould.

2.1.2 The materials

The main materials in this research are three different types of chitosan with different molecular weight (Mw) and were obtained from Chimultiguna. Shrimp chitosan has a medium molecular weight of 143.2 kDa, while crab chitosan and fish scale chitosan have low molecular weights of 87.34 kDa and 75 kDa, respectively. κ-carrageenan with molecular weight of 788.65 kDa was obtained from Kappa Carrageenan Nusantara. Other materials used were distilled water; NaOH; and HCl from SAP Chemicals.

2.1.3 Ethical approval

This study does not require ethical approval because it does not use experimental animals.

2.2 Methods

2.2.1 Experimental design

The experimental design in this study consists of one independent variable, namely the addition of different chitosan. There were four treatments of chitosan addition to polyelectrolyte complex (PEC) bioplastics (A0 = no chitosan, A1 = shrimp chitosan (Mw 143.2 kDa), A2 = crab chitosan (Mw 87.34 kDa), A3 = fish scale chitosan (Mw 75 kDa)) with five replications.

2.2.2 Gel permeation chromatography (GPC)

The molecular weight of chitosan was determined using gel permeation chromatography (GPC) to assess its molecular properties according to Gomes *et al.* (2021). The mobile phase (GPC) was prepared by dissolving 0.15 M ammonium acetate and 0.2 M acetic acid in deionized water, adjusting the pH to 4.5, and filtering the solution through a 0.45 µm filter to remove particulates. For sample preparation, chitosan samples were dissolved in the GPC buffer to achieve a concentration of 1 g/L, ensuring complete dissolution through stirring and filtering the solution through a 0.45 µm syringe filter to remove undissolved particles. The GPC system was calibrated using Novema Precolumn (10 mm, 8 × 50 mm), Novema 30 Å (10 mm, 8 × 300 mm), and two Novema 1000 Å (10 mm, 8 × 300 mm) columns. The chitosan sample solution (1 g/L) was loaded into the injector, and the sample was allowed to elute through the columns and detectors. The collected data were used to calculate the polydispersity index (PDI).

2.2.3 Optimization of κ-carrageenan and chitosan volume ratio

The volume ratio between κ-carrageenan and chitosan in this study refers to Ismillayli *et al.* (2020) with modifications because the method in that study

requires 24 h of stirring without stopping to dissolve chitosan, so to shorten the time, the researchers modified the concentration of ingredients. Table 1 shows the combination of several concentrations between κ-carrageenan and chitosan from the initial experiment conducted in this study. All samples in this initial experiments were conducted using shrimp chitosan with the same temperature and pH treatment to observe the PEC formed between κ-carrageenan and chitosan. Based on the initial experiment conducted in this study, the concentration combination between κ-carrageenan and chitosan to form polyelectrolyte complex (PEC) indicated by the formation of solid precipitation is 0.75% (w/v) chitosan and 1.25% (w/v) κ-carrageenan.

ually (20 ml/min) with stirring at 500 rpm. Then, the temperature of the mixture was kept constant at 40°C for 2 h. The pH of the solution mixture was measured using a pH meter and adjusted to pH 5. Before molding, the mixture was cooled in a waterbath at 25°C for 15 minutes. The precipitation formed is PEC which was poured into a 25x15x7 cm^3 glass mold and dried at 35°C for three days.

2.2.5 Bioplastic characterization

The bioplastic samples were tested with a series of characterization tests, including thickness measurement, tensile strength, water vapor transmission, swelling test, biodegradation rate, Fourier-Transform Infrared Spectroscopy (FTIR) for polyelectrolyte

Table 1. Volume ratio of κ-carrageenan and chitosan that can form polyelectrolyte complex (PEC).

	Carrageenan 1.25% (w/v)	Carrageenan 1.5% (w/v)
Chitosan 0.38% (w/v)	No PEC formation	PEC coarcervate
Chitosan 0.5% (w/v)	No PEC formation	PEC coarcervate
Chitosan 0.75% (w/v)	**PEC precipitation**	PEC coarcervate
Chitosan 1% (w/v)	Materials flocculation	Materials flocculation

Description: The bolded word (PEC precipitation) indicates a feasible concentration combination between κ-carrageenan 1.25% (w/v) and chitosan 0.75% (w/v) to form polyelectrolyte complex (PEC) in this study.

2.2.4 Preparation of polyelectrolyte complex (PEC) bioplastic

The polyelectrolyte complex (PEC) bioplastics fabrication follows Al-Zebari *et al.* (2019) and Ismillayli *et al.* (2020) with modificaiton. Chitosan solution was prepared by dissolving 1.5 g of chitosan (0.75% w/v) in 200 mL of 0.01 M HCl (±pH 2) at 60℃ for 4 h and stirring of 600 rpm. HCl was chosen as a chitosan solvent because it is a strong acid that dissolves the entire chitosan structure. The positively charged amino groups of chitosan can form complexes with negatively charged substances. Under acidic conditions, these amino groups protonate, increasing chitosan's solubility and interaction with other polymers such as alginate or carrageenan, which are used to form polyelectrolyte complexes. κ-carrageenan solution was prepared by dissolving 4.4 g κ-carrageenan (1.25% w/v) in 350 mL of distilled water at 45℃ for 2 h and stirring of 400 rpm. Both solutions were stored for 24 h at room temperature (± 32℃) before use. Before mixing, the pH of the chitosan solution was increased from ± pH 2 to ± pH 4 by using 5 M NaOH and the κ-carrageenan solution was heated to 60°C for 1 h with 300 rpm of stirring. The addition of chitosan solution to the κ-carrageenan solution was done grad-

complex (PEC) characterization, and Scanning Electron Microscope (SEM) for morphological examination. Subsequently, these bioplastics were employed as packaging material for tilapia fillets over a 24 h period, followed by total plate count (TPC) testing of the fillets.

2.2.6.1 Thickness

Bioplastic thickness test was conducted using a screw micrometer with a precision of 0.1 mm. Measurements were taken at five points on each bioplastic sample, and the average thickness was calculated. Subsequently, the bioplastic thickness was compared against the standard thickness specified for food packaging suitability which should not exceed 0.25 mm according to Japanese Industrial Standard (JIS Z1707:2019).

2.2.6.2 Tensile strength

The tensile strength test was carried out following the method described by Putra *et al.* (2019), using a tensile strength testing device to pull the bioplastic until it fractured. Tensile strength was determined based on the maximum force recorded at the breaking point. According to the Japanese Industrial Standard,

the standard for tensile strength in bioplastics is 3.92 MPa. Samples were cut to specified dimensions in a dogbone shape measuring 6x1.5 cm² and tested using the tensile strength testing device. JIS Z1707:2019 is specifically developed for the assessment of bioplastics, offering methodologies that are more pertinent than the general ASTM standards applicable to conventional plastics. This standard facilitates a thorough evaluation of bioplastics, encompassing aspects such as biodegradability, environmental impact, and essential mechanical properties. In contrast, ASTM D638 and D882 predominantly concentrate on mechanical strength, which may not adequately reflect the distinctive characteristics of bioplastics.

2.2.6.3 Elongation at break

Elongation at break is a crucial mechanical property used to assess the ductility and toughness of materials. It measures the amount of deformation a material can withstand before breaking. This test was carried out with the same method as tensile strength test described by Putra *et al.* (2019). The tensile strength testing device showed stress and strain value which can be calculated using the appropriate equation.

Elongation at break = Stress/Strain%.....(i)

2.2.6.4 Water vapor transmission

Water vapor transmission testing was carried out according to the method described by Sunardi *et al.* (2020). Bioplastic samples measuring 5x5 cm² were weighed before (W0) and after (W1) testing. Each sample was placed in a desiccator with silica gel at the bottom and a glass cup containing distilled water. After allowing the samples to stand for one day, the transmission rate was calculated using the appropriate equation.

Water Vapor Transmission = ΔW/(t.A).....(ii)

Where :

ΔW = difference between initial weight (W0) and final weight (W1) (grams)

t = time (day)

A = area of bioplastic surface(cm²).

2.2.6.5 Swelling test

The test was conducted according to the standard swelling test specified by the Japanese Industrial Standard (JIS) (1975). According to JIS, good water resistance for bioplastics is ≤70%. The water resistance test involved weighing the initial weight of 2x2 cm² bioplastic (W0), immersing it in a glass beaker containing distilled water for 10 minutes at room temperature, and then cleaning and weighing the sample to obtain the final weight (W1). The results were calculated using the appropriate equation (Ghanbari *et al.*, 2018).

Swelling Test = (W1-W0)/W0×100%.....(iii)

Where :

W0 = initial weight of bioplastic (grams),

W1 = final weight of bioplastic (grams).

2.2.6.6 Biodegradation rate

The biodegradation rate test was conducted by burying biodegradable plastic samples measuring 2x2 cm² in soil for seven days. The samples were weighed initially before burial and weighed again after burial.

2.2.6.7 Fourier-transform infrared spectroscopy (FTIR)

The FTIR analysis of bioplastics involves exposing the bioplastic samples to infrared radiation using specialized instrumentation. The molecules within the sample absorb the infrared rays, inducing oscillation and rotation of their constituent atoms. Each functional group within the bioplastic molecule absorbs infrared radiation at distinct frequencies, thereby generating unique wavelength peaks in the FTIR spectrum. Identification of specific functional groups is achieved through comparison of these wavelength patterns with established reference spectra.

2.2.6.8 Scanning electron microscopy (SEM)

SEM analysis was performed on PEC κ-carrageenan bioplastics incorporated with the optimal type of chitosan to examine their morphological structure, following the method outlined by Ismillayli *et al.* (2020). This technique involves directing an electron beam onto the sample surface to produce high-resolution three-dimensional images. Bioplastic samples, cut into 10x10 mm², were affixed to a copper substrate using conductive glue and coated with a thin layer of gold in a vacuum environment. Images were generated from signals emitted during the interaction of the electron beam with the sample, processed by the SEM detector.

2.2.6.9 Total plate count (TPC)

The total plate count (TPC) assay was conducted on tilapia fillets before and after storage in bio-

plastic packaging for 24 h. The fillets were cut into 3x3 cm^2 pieces and wrapped in bioplastic, which was then sealed using a hot seal tool. The samples were then diluted with Buffered Pepton Water (BPW) to a 10^{-4} dilution. Then, 1 ml of the resulting suspension was spread onto a petri dish containing Nutrient Agar (NA). The petri dishes were incubated at 37°C for 24 h in an inverted position. After incubation, the number of bacterial colonies was counted and multiplied by the dilution factor. The maximum colony count limit for the product, as specified by the Standar Nasional Indonesia (SNI), is 10^5 CFU/gram.

2.3 Analysis Data

The data obtained from the study will first undergo normality and homogeneity tests. Subsequently, analysis of variances (ANOVA) was conducted at a significance level of 99%%. If significant effects were observed from the treatments, the analysis proceeded with the Duncan Multiple Range test, also at a 99% significance level, to determine differences among treatments. All the analyses were performed using IBM SPSS Statistics 20.

3. Results and Discussion

3.1 Results

3.1.1 Mechanical properties

The results of this study demonstrated that polyelectrolyte complex (PEC) bioplastics composed of κ-carrageenan and crab chitosan (A2) with a molecular weight of 87.34 kDa exhibited the most favorable outcomes among the tested treatments in Table 2 and Figure 1. The strong ionic bond between chitosan polycations and κ-carrageenan polyanions can be seen in Figure 2, and PEC A2 bioplastics lead to enhanced mechanical properties, including higher thickness value in Figure 3, higher tensile strength in Figure 4, higher elongation at break in Figure 5, and resistance to water and oxygen. The molecular weight of 87.34 kDa in the κ-carrageenan and chitosan combination optimizes the formation of these ionic bonds, resulting in bioplastics with impermeable properties. This impermeability significantly reduces the water vapor transmission (0.001456 g/m^2/day) in Figure 6 and water resistance (63.94%) of PEC A2 bioplastics in Fig

Table 2. Characteristics of PEC bioplastics with different types of chitosan.

Treatment	Chitosan Type	Characteristic	Color
A0	Control	Rigid, brittle, inflexible, rather thick	Transparent
A1	Shrimp	Pliable, flexible, thick	Transparent
A2	Crab	Somewhat stiff, thickest	Transparent
A3	Fish Scale	Pliable, flexible, thinnest	Transparent

Description: A0 (Control), A1 (PEC with 0.75% (w/v) shrimp chitosan), A2 (PEC with 0.75% (w/v) crab chitosan), and A3 (PEC with 0.75% (w/v) fish scale chitosan).

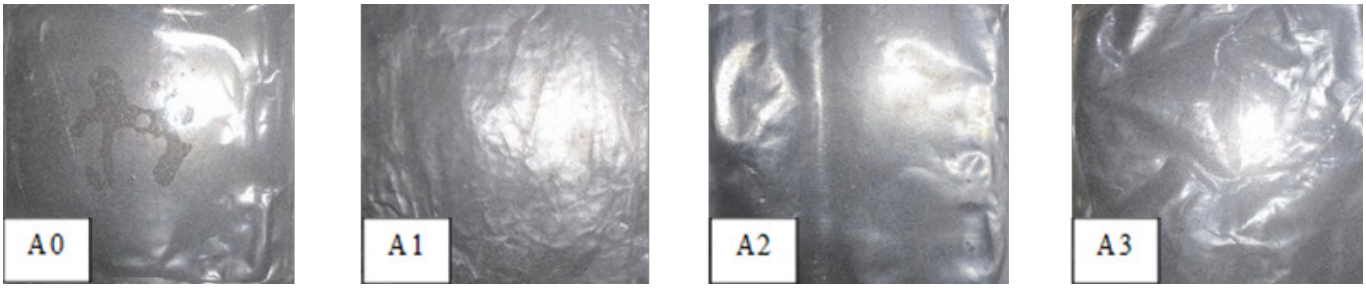

Figure 1. Polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3).

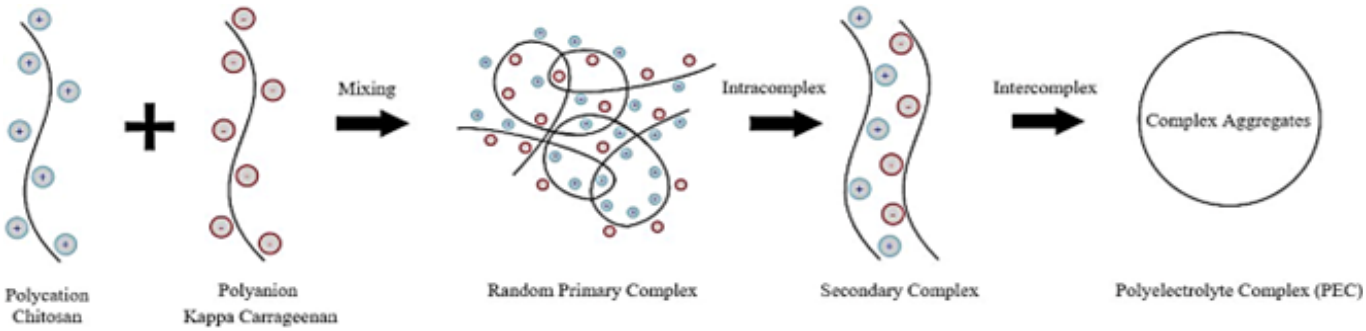

Figure 2. Formation process of polyelectrolyte complex (PEC) from K-carrageenan and chitosan (Meka *et al.*, 2017).

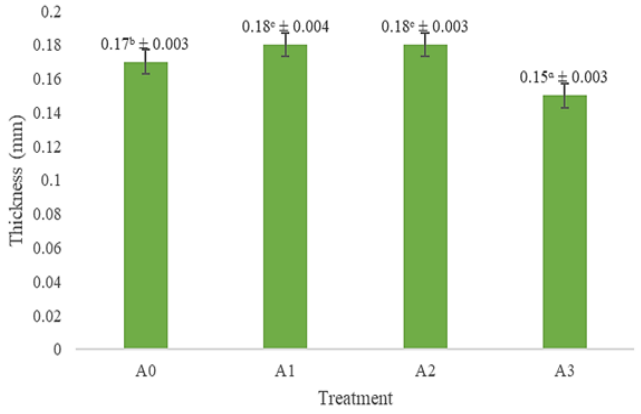

Figure 3. Thickness of polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3). Different superscript letters indicate the comparison between treatments is very significantly different (P < 0.01).

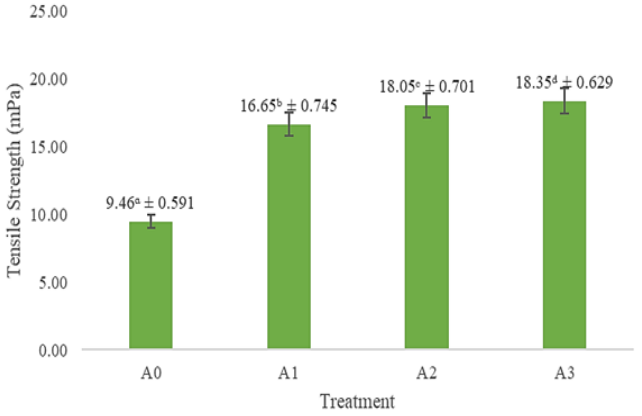

Figure 4. Tensile strength of polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3). Different superscript letters indicate the comparison between treatments is very significantly different (P < 0.01).

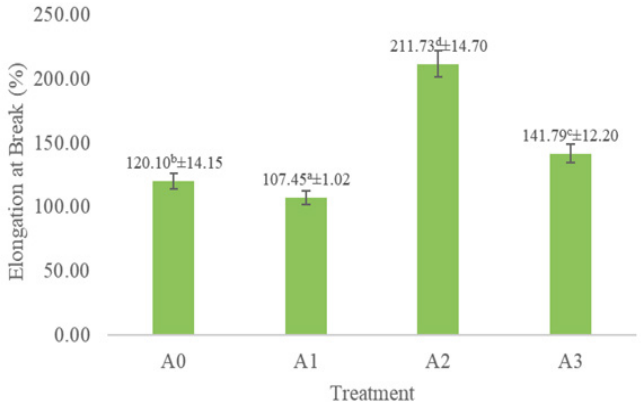

Figure 5. Elongation at break of polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3). Different superscript letters indicate the comparison between treatments is very significantly different (P < 0.01).

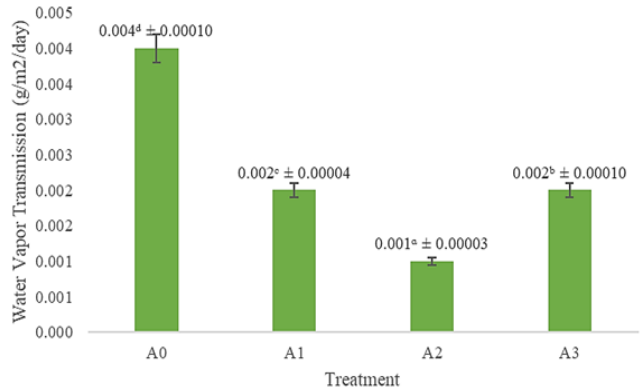

Figure 6. Water vapor transmission (WVT) of polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3). Different superscript letters indicate the comparison between treatments is significantly different (P < 0.01).

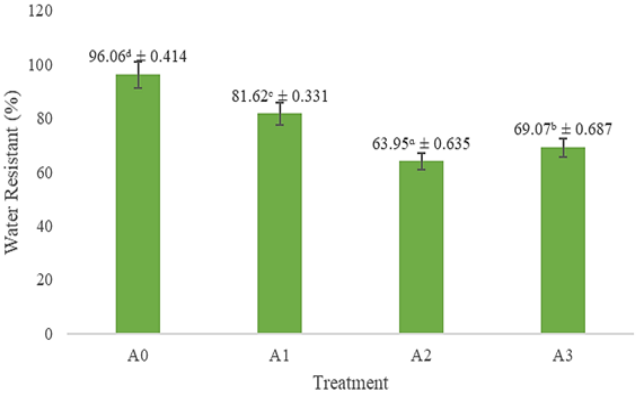

Figure 7. Water resistance of polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3) with swelling test. Different superscript letters indicate the comparison between treatments is significantly different (P < 0.01).

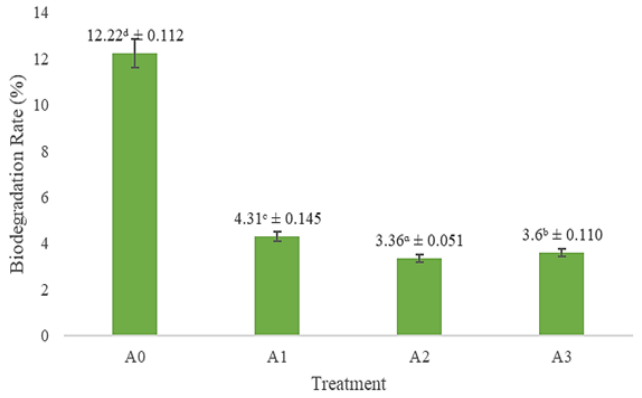

Figure 8. Biodegradation rate of polyelectrolyte bioplastic (PEC) from κ-carrageenan without chitosan (A0), with 0.75% (w/v) shrimp chitosan (A1), 0.75% (w/v) crab chitosan (A2), and 0.75% (w/v) fish scale chitosan (A3). Different superscript letters indicate the comparison between treatments is very significantly different (P < 0.01).

ure 7. The lower degradation rate of 3.358% in soil, as seen in Figure 8, is attributed to the reduced access of microorganisms to the bioplastic surface due to the impermeability, as reported by Adhikari *et al.* (2016). Figure 9 shows that due to ionic bonds from PEC form an impermeable layer while non-ionic bonds form a permeable layer (Hubbe *et al.*, 2021).

The test results in each treatment showed very significant differences ($P < 0.01$). The A2 bioplastic displayed superior mechanical characteristics in nearly all assessments, including a thickness of 0.178 mm, an elongation at break of 211.73%, a water vapor transmission of 0.001456 g/m²/day, a water resistance of 63.94%, a degradation rate of 3.358%, and the lowest Total Plate Count (TPC) of 2.88×10^7 CFU/g after 24 h in Table 3. The tensile strength test results indicated that the PEC bioplastic made from κ-carrageenan and fish scale chitosan (A3) achieved the highest value of 18.348 MPa. However, this result was not significantly different from that of the A2 bioplastic, which had a tensile strength of 18.053 MPa, as determined by the Duncan Multiple Range test.

3.1.2 Polyelectrolyte Complex (PEC) formation

The formation of an ionic bond between the amine group of chitosan and the sulfate group of κ-carrageenan was evaluated through Fourier Transform Infrared (FTIR) absorption peaks at 1034 cm^{-1}, 1224 cm^{-1}, 1525 cm^{-1}, 1636 cm^{-1} as seen in Figure 10. The Morphological structure of biolastic was evaluated using Scanning Electron Microscopy (SEM), which shows electrostatic interactions from the irregular and fibrous surface structure leading to the formation of a polyelectrolyte complex. Figure 11 shows the irregular and fibrous surface structure of A2 bioplastic.

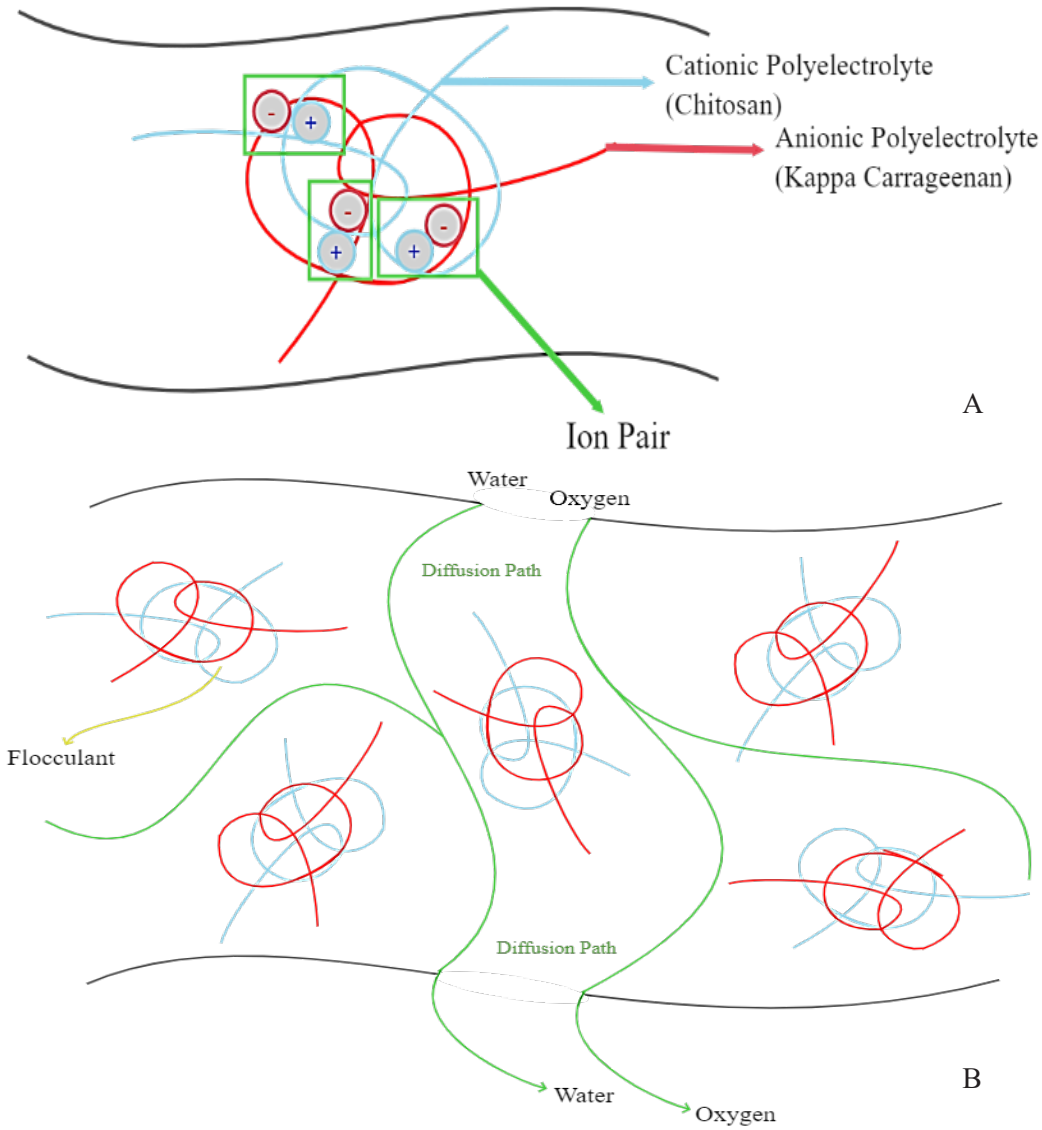

Figure 9. a) Ionic bonds form impermeable layer, b) Non-ionic bonds (flocculants) form permeable layer (Hubbe *et al.*, 2021).

Table 3. Total plate count (TPC) of tilapia fish fillet before and after 24 h of storage.

Treatment	Time	Log CFU/g
No Plastic	0 h	4.4±0.02
	24 h	7.73^{c}±0.01
A0	0 h	4.41±0.01
	24 h	7.68^{c}±0.02
A1	0 h	4.4±0.02
	24 h	7.56^{b}±0.02
A2	0 h	4.4±0.01
	24 h	7.42^{a}±0.10
A3	0 h	4.4±0.01
	24 h	7.5^{b}±0.03

Description: A0 (Control), A1 (PEC with 0.75% (w/v) shrimp chitosan), A2 (PEC with 0.75% (w/v) crab chitosan), and A3 (PEC with 0.75% (w/v) fish scale chitosan). ANOVA test results showed significant differences ($P < 0.01$). Different superscript letters indicate the comparison between treatments based on Duncan Multiple Range test.

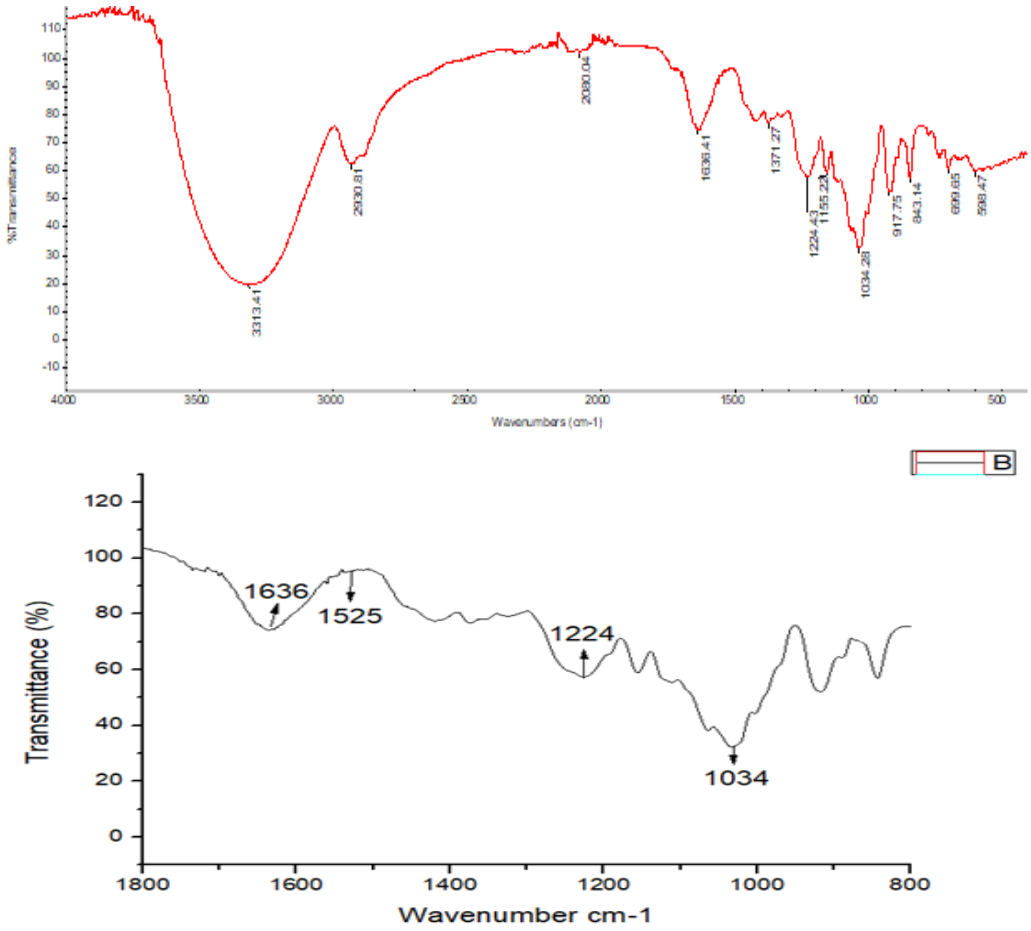

Figure 10. Fourier-transform infrared spectroscopy (FTIR) of κ-carrageenan-crab chitosan polyelectrolyte complex (PEC) bioplastic (A2).

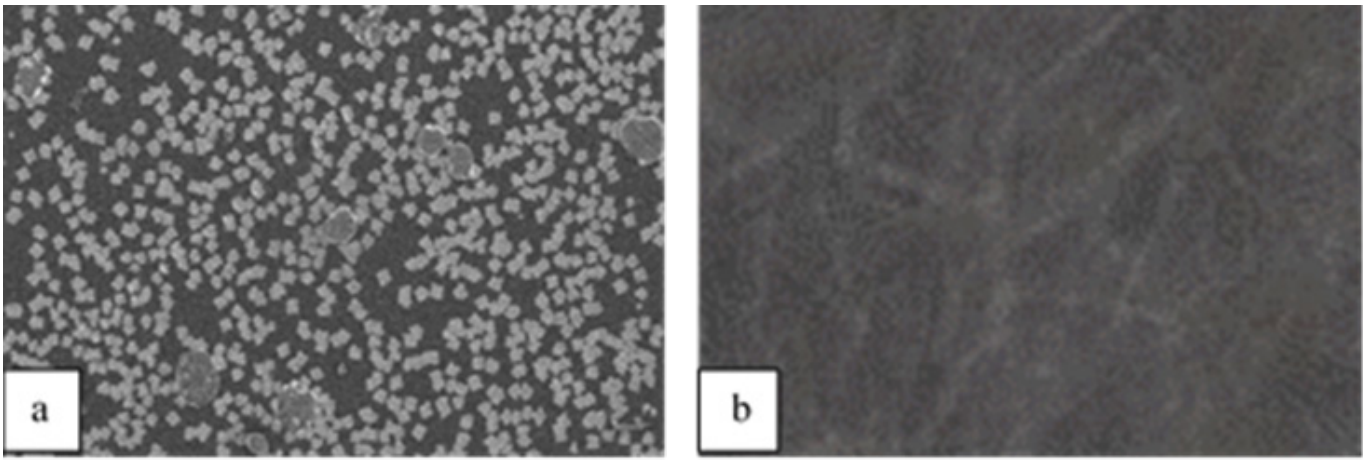

Figure 11. Scanning electron microscope (SEM) of κ-carrageenan-crab chitosan polyelectrolyte complex (PEC) bioplastic (A2) with magnification of 10.000x (a) and 3.000x (b).

3.2 Discussion

3.2.1 Mechanical properties of different chitosan on κ-carrageenan pec bioplastics

This study involved the production of bioplastics with dimensions of 25x15 cm². Each sample utilized κ-carrageenan of consistent molecular weight, while incorporating chitosan of varying molecular weights. Shrimp chitosan has a molecular weight of 143.2 kDa, which is categorized in the medium molecular weight category, whereas crab chitosan has a molecular weight of 87.34 kDa and fish scale chitosan has a molecular weight of 75 kDa, which are categorized in the low molecular weight category. The bioplastics exhibited similar physical characteristics, necessitating a series of mechanical tests to identify the most suitable type of chitosan for producing PEC bioplastics.

Chitosan has a pKa of 6-6.5, indicating that it will dissolve in acidic conditions with a pH below 6-6.5. The protonation process of chitosan occurs because hydrochloric acid (HCl) is a strong acid capable of donating H+ ions (protons). The H+ from HCl moves to the amine group (NH_2) in chitosan, converting it into a protonated ammonium form (NH_3). K-carrageenan, with a pKa of 1.5-2.5, ionizes at a pH above 1.5-2.5 and degrades below this range (Ismillayli *et al.*, 2020). The sulfate groups ionize in neutral pH water (Sariyer *et al.*, 2020). Polyelectrolyte κ-carrageenan is formed from the interaction of the sulfate group () with water molecules, resulting in the ionization of the sulfate group into SO_4.

After a brief period of mixing, the primary complex forms rapidly due to the interaction of two solutions with different charges. PEC formation shows the initial complex lacks stable ionic bonds because the electrostatic forces are too strong, necessitating further mixing to achieve stability. Continued mixing for one to two hour from the initial mixing phase will result in the formation of a secondary complex. This secondary complex will have more stable electrostatic bonds. Meka *et al.*, 2017 demonstrated that there are five phase of pH in the formation of PEC, phase I with the very low pH (<pH 2) there will be no PEC formation, phase II with neutral pH (pH 4-7) forms stable PEC in the form of precipitates, phase III with alkaline pH (>pH 7) forms coacervate (separate liquid phase), phase IV with further increase in ionic bonding forms more PEC in the form of denser precipitates, and phase V with salt addition degrades PEC and there will be no PEC formed. Additional treatments, such as the addition of salt, variation in mixing time, and pH adjustments, can cause the secondary complexes to form complex aggregates (Neitzel *et al.*, 2021).

Salt plays a crucial role in the formation of polyelectrolyte complexes (PECs), as it enables the production of final products with tailored characteristics for specific applications. Salt can be introduced through the addition of salt or formed during the mixing process of oppositely charged solutions, resulting in ionic interactions (Meng *et al.*, 2020). In this study, salts were formed through the reaction process of producing bioplastics without the addition of salt. Increasing salt concentrations can lead to increased polymer flexibility, thereby reducing aggregation. However, further increases in salt concentration can cause secondary aggregation, resulting in macroscopic flocculation or agglomeration of large-sized polymers (Meka *et al.*, 2017). The formation of such macroscopic flocculation is undesirable in PEC manufacturing for bioplastic applications, as it can result in unstable PEC structures and a more brittle end product with compromised mechanical characteristics.

The polyelectrolyte complex (PEC) formation process in this study involves the reaction between chitosan polycation and carrageenan polyanion, resulting in the formation of hydrogel (Mokhtari *et al.*, 2021).

The properties of chitosan, including its molecular weight, can significantly influence polycation formation (Liu *et al.*, 2020). The molecular weight of chitosan affects the solubility, viscosity, and charge density of chitosan solutions. Chitosan with a lower molecular weight tends to have fewer amine groups, resulting in higher solubility and weaker polycation properties (Hosseini-Ashtiani *et al.*, 2021). Low molecular weight chitosan typically forms a weak polyelectrolyte complex when combined with κ-carrageenan polyanion. Conversely, if the molecular weight of chitosan is too high, the resulting polycations are less stable due to lower solubility that can lead to aggregation and clumping (Suryani *et al.*, 2022). Thus, the molecular weight of chitosan impacts the formation of polyelectrolyte complexes with κ-carrageenan polyanions and can affects the characteristics of the resulting bioplastics (Andonegi *et al.*, 2020).

Chitosan with a higher molecular weight exhibits greater resistance to acid, necessitating the use of a stronger acid at a higher concentration for complete dissolution without clumping. In this study, shrimp chitosan with the highest molecular weight of 143.2 kDa among the three types displayed complete insolubility and clumping in HCl acid solvent at pH 2. In contrast, crab chitosan and fish scale chitosan with lower molecular weights dissolved completely without clumping in the same acid solvent. This indicates that higher molecular weight with higher amine groups reduces the solubility of chitosan. However, chitosan with too low molecular weight can be degraded in acidic solvents due to its high solubility that can lead in weakened polycation properties (Gonçalves *et al.*, 2021). Chitosan with a higher degree of deacetylation (DD) tends to have a higher molecular weight which generally results in improved mechanical properties. The DD indicates the extent to which the acetyl groups are removed from chitin, transforming it into chitosan. A higher DD have more free amine groups ($-NH_2$) that are available, which increases intermolecular interactions such as hydrogen bonding. This contributes to stronger and more rigid material characteristics (Zhang *et al.*, 2023).

Favian and Nugraheni (2023) explained that the interaction between the positive charge of polycation and the strong negative charge of polyanion forms a complex that can increase viscosity, thereby increasing the thickness of the resulting bioplastic. Bioplastic A2 which has the highest thickness value demonstrates that the sulfate group on κ-carrageenan and the amine group on crab chitosan form strong ionic bonds. In contrast, A3 bioplastic with the lowest thickness value of 0.153 mm indicates a weaker ionic bond between the amine group of fish scale chitosan and the sulfate group of κ-carrageenan. These characteristics are due to the synergistic effect of ionic and hydrogen bonds between κ-carrageenan and chitosan molecules. κ-carrageenan with sulphate groups can be used as an ionic cross-linking agent to stabilize polycation chitosan in acidic conditions (Kołodziejska *et al.*, 2021).

The formation of the polyelectrolyte complex between the amine groups of chitosan and the sulfate groups of kappa carrageenan occurs through physisorption. This process does not involve the breaking or forming of chemical bonds like those in chemisorption but rather the formation of weak ionic bonds. The resulting complex is generally reversible, unlike chemisorption, which is irreversible. The ionic bonds in physisorption can be disrupted by changes in pH or ionic strength, which is characteristic of physisorption processes (Sims *et al.*, 2021). The presence of ionic bonds forming a polyelectrolyte complex can create an impermeable layer on bioplastics. The ionic bond between chitosan polycations and κ-carrageenan polyanions forms a matrix polyelectrolyte complex, whereas non-ionic bonds between the two polyelectrolytes act as plasticizers. High ionic bond densities in the polyelectrolyte complex result in stiffer bioplastics, which are inelastic and impermeable to moisture. In contrast, lower ionic bond densities lead to the formation of flocculants or clumps due to insufficient ionic bonds, resulting in more permeable and flexible bioplastics with higher elasticity but lower resistance to oxygen and water (Hubbe *et al.*, 2021).

3.2.2 PEC bioplastic as food packaging

The total plate count (TPC) results of tilapia fillets packaged with each PEC bioplastic sample demonstrated that all samples exceeded the SNI threshold of 5 log CFU/g. Notably, PEC A2 bioplastics exhibited the lowest TPC value of 7.45 log CFU/g after 24 h of storage at room temperature. The storage of tilapia fillets for 24 h was selected because it exceeds the 10 h threshold beyond which the product is prone to increased bacterial growth. Tilapia undergoes a decay process, characterized by the pre-rigor phase within 0-6 h of storage and the rigor mortis phase between 6-10 h at room temperature. The higher ionic bond density between chitosan polycations and κ-carrageenan polyanions in PEC bioplastics significantly reduces the permeability of these bioplastics to oxygen and water. This impermeable structure acts as a barrier layer, preventing bacterial penetration into the tilapia fillets. Xue *et al.* (2023) highlighted the importance of impermeable properties in bioplastics for

maintaining product quality and safety in food packaging applications.

3.2.3 Electrostatic interaction from PEC formation

The formation of an ionic bond between the amine group of chitosan and the sulfate group of κ-carrageenan is evident from the Fourier Transform Infrared (FTIR) absorption peaks at 1034 cm^{-1}, 1224 cm^{-1}, 1525 cm^{-1}, 1636 cm^{-1}.Wavelength 1034 cm^{-1} shows an aliphatic C–N stretching (amines) and 1224 cm^{-1} shows an aromatic C–N stretching (amines) which indicates the presence of sulfate groups from ammonium sulfate as a result of ionic interaction between the amine group of chitosan and the sulfate group of κ-carrageenan. In addition, at wavelength 1525 cm^{-1} and 1636 cm^{-1} are primary amide bending (amides) indicates the presence of NH^{3+} which forms a polyelectrolyte complex with κ-carrageenan. This bond is a key component of the polyelectrolyte complex (PEC) formed electrostatically between chitosan and κ-carrageenan, as demonstrated by Ismillayli *et al.* (2020) at a wavelength of 1652 cm^{-1}. Additionally, Scanning Electron Microscopy (SEM) results show electrostatic interactions leading to the formation of a polyelectrolyte complex showing the irregular and fibrous surface structure of A2 bioplastic. Ismillayli *et al.* (2020) showed that irregular and fibrous surface is a result of complex aggregates formed through ionic interactions between amide groups of chitosan and sulfate groups of κ-carrageenan.

4. Conclusion

The best type of chitosan in the production of κ-carrageenan bioplastic using polyelectrolyte complex (PEC) method in this study was crab chitosan. PEC κ-carrageenan-crab chitosan has the best mechanical characteristics which include thickness, tensile strength, water resistance, water vapor transmission (WVT), and biodegradation rate, and has the lowest TPC result in tilapia fish fillet after 24 h. This study demonstrates the potential for the utilization of other types of chitosan with a molecular weight comparable to crab chitosan, as well as the application of other food products besides tilapia fillets.

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Authors' Contributions

All authors have contributed to the final manuscript. The contribution of each author as follow, Dhena; collected the data, drafted the manuscript, and designed the figures. Amin and Laksmi; devised the main conceptual ideas and critical revision of the article. All authors discussed the results and contributed to the final manuscript.

Conflict of Interest

The authors declare that they have no competing interests.

Declaration of Artificial Intelligence (AI)

The author(s) acknowledge the use of ChatGPT, Perplexity, DeepL, and Grammarly for language refinement, summarization, and translation in preparing this manuscript. All AI-generated content was rigorously reviewed, edited, and validated to ensure accuracy and originality. Full responsibility for the manuscript's final content rests with the author(s). To ensure transparency and support the review process, a comprehensive delineation of the tool's application is furnished in the "Introduction" or "Materials and Methods" section of this manuscript in compliance with the publisher's ethical guidelines.

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