

Preparation and Characterization of Red Seaweed (*Eucheuma cottonii*)-Based Bioplastic Reinforced with Polyvinyl Acetate (PVAc)

Mahreni^{1*}, Nanda Yunia Erma¹, Nur Aisyah Novitasari¹

¹ Department of Chemical Engineering, Universitas Pembangunan Nasional Veteran Yogyakarta, Indonesia

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Abstract

Conventional petrochemical-based plastic waste has become a serious environmental problem due to its resistance to decomposition and suboptimal management. This study aims to produce and characterize carrageenan-based bioplastic from red algae (*Eucheuma cottonii*) by examining the effects of varying concentrations of sorbitol as a plasticizer and polyvinyl acetate (PVAc) as a reinforcement on mechanical properties, water resistance, and biodegradability. The study was conducted in several stages, first involving the extraction of red algae with 0.1N NaOH. The extraction product was then mixed with a plasticizer, reinforcement, and emulsifier to form a homogeneous mixture. After forming a homogeneous emulsion, the mixture was molded and oven-dried at 60°C for 12 hours. The results showed that increasing the sorbitol concentration decreased tensile strength but increased elongation and accelerated biodegradation. Conversely, adding PVAc increased tensile strength and water resistance but reduced elongation and slowed degradation. The resulting carrageenan-based bioplastic has the potential to serve as an environmentally friendly alternative with properties that can be tailored for sustainable applications.

Keywords Biodegradable, Bioplastic, Carrageenan, Reinforced, Surfactant

INTRODUCTION

Indonesia is an archipelagic nation with the world's second-longest coastline and abundant marine resources, including red seaweed (*Eucheuma cottonii*). However, the country faces serious environmental challenges regarding the accumulation of conventional plastic waste, which accounts for 19.21% of total national waste generation (Menhik, 2025). Petrochemical-based plastics take between 10 and 500 years to degrade, contributing to long-term environmental pollution and microplastic accumulation in aquatic ecosystems (Jumaidin et al., 2017). Consequently, researchers are developing eco-friendly plastic alternatives derived from renewable resources. One promising natural raw material for bioplastic production is carrageenan, a sulfated polysaccharide extracted from red seaweed cell walls. Kappa-carrageenan derived from *Eucheuma cottonii* features a linear structure dominated by D-galactose and 3,6-anhydrogalactose units with a sulfate content of less than 28%, enabling the formation of strong, stable gels (Kuznetsova et al., 2020). However, pure carrageenan films have limited mechanical properties and water resistance, preventing them from fully replacing conventional plastics (Ili Balqis et al., 2017).

While previous studies have examined the individual effects of sorbitol or PVAc on the properties of carrageenan-based bioplastics (Krishnan et al., 2024), research into the interaction between the two within a carrageenan- PVAc-sorbitol composite system remains limited, especially concerning their simultaneous influence on mechanical properties (tensile strength, elongation), water resistance (water uptake), and biodegradability. Therefore, this study aims to

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Corresponding author's email: mahreni@upnyk.ac.id

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analyze the effects of varying concentrations of sorbitol (0–50%) and PVAc (0–25%) on these properties, with the goal of developing a carrageenan-based bioplastic formulation that meets application requirements while helping reduce conventional plastic waste.

LITERATURE REVIEW

To enhance the performance of carrageenan-based bioplastics, additives such as plasticizers and reinforcing agents are required. Sorbitol is widely used as a plasticizer because its multiple hydroxyl groups interact with the polysaccharide matrix via hydrogen bonding, thereby increasing the material's flexibility and elongation (Hardiningtyas et al., 2024). Studies (Ili Balqis et al., 2017) indicate that carrageenan films containing sorbitol have a more compact structure, lower water uptake, and higher water vapor permeability than those using glycerol or PEG-300 as plasticizers. Nevertheless, excessive sorbitol concentrations can reduce tensile strength by weakening intermolecular bonds within the polymer matrix (Mahreni et al., 2021).

On the other hand, polyvinyl acetate (PVAc) is a biodegradable synthetic polymer that can serve as a reinforcing agent to enhance mechanical strength, interfacial adhesion, and water resistance. Research by Maryam et al. (2019) indicates that adding 2–10% PVAc can increase the structural density of bioplastics and improve their mechanical properties. However, the interaction between PVAc and carrageenan within the bioplastic system requires further optimization, particularly regarding phase compatibility and its impact on hydrophilicity and biodegradability.

RESEARCH METHOD

Materials and Tools

This study used an analytical balance, oven, magnetic stirrer, heating stove, glass beaker, measuring cylinder, stirring rod, thermometer, and silicone mold. The materials used included red algae (*Eucheuma cottonii*), sodium hydroxide, sorbitol, polyvinyl acetate (PVAc), distilled water (aquadest), carboxymethyl cellulose (CMC), and isopropyl alcohol (IPA).

Research Methods

Red algae were washed, soaked in clean water for 24 hours, and dried at 105°C to constant weight. The dried algae were then soaked in distilled water for 30 minutes at a 1:10 w/v ratio, filtered, and homogenized using a blender. Extraction was performed using 0.1 N NaOH at 70–80°C for 90 minutes. The extract was filtered, and the filtrate was precipitated with isopropyl alcohol (IPA) at a 1:2 w/v ratio for 30 minutes at 5–10°C. The resulting carrageenan fibers were dried at 50–60°C to constant weight.

The carrageenan fibers (5 g) were mixed with sorbitol (10%, 20%, 30%, 40%, and 50%), PVAc (5%, 10%, 15%, 20%, and 25%), and CMC (6%), all based on the carrageenan weight, with 100 mL of distilled water. The mixture was stirred and heated at 60°C until homogeneous, poured into silicone molds, and dried at 60°C for 12 hours.

Characterization

The resulting bioplastic was characterized for tensile strength, elongation, water uptake, biodegradability, functional groups using FTIR, and surface morphology and composition using SEM-EDS. Tensile strength and elongation were measured using a Universal Testing Machine (UTM) according to ASTM D882, with tensile strength calculated from the maximum force relative to the initial cross-sectional area and elongation from the increase in sample length.

For water uptake, 2 × 2 cm bioplastic samples were weighed (W_0), immersed in distilled water for 3 minutes, dried with tissue, and reweighed (W_1) following Islamiyah et al. (2022). The water uptake percentage was calculated using Equation 1.

$$\text{Water Uptake Rate (\%)} = \frac{(W_1 - W_0)}{W_0} \times 100 \% \quad (1)$$

Biodegradability was evaluated using 2×2 cm bioplastic samples following [Suwardi et al. \(2020\)](#). Samples were weighed initially (W_0), buried in soil at a depth of 8 cm for 7 days, then cleaned and reweighed (W_1). The biodegradation rate was calculated from the sample weight loss using Equation 2.

$$\text{Weight Loss (\%)} = \frac{(W_0 - W_1)}{W_0} \times 100 \% \quad (2)$$

Based on the percentage of sample weight loss, the time required for the bioplastic to biodegrade completely (100%) can be calculated ([Triani et al., 2022](#)). Each formulation was prepared and characterized using one measurement ($n = 1$). Therefore, standard deviations were not calculated, and the reported values represent the measurements obtained from the individual formulations.

$$\text{Perfect Degradation Time (days)} = \frac{100\%}{\text{Weight Loss (\%)}} \times \text{test time} \quad (3)$$

To minimize measurement variability, we kept sample dimensions, testing conditions, and measurement procedures consistent throughout characterization. Tensile strength and elongation measurements followed ASTM D882, while water uptake and biodegradability tests used fixed sample dimensions and specified immersion and soil-burial conditions. The calculated properties were based on the measured initial and final sample weights or dimensions.

Statistical Analysis

Regression analysis evaluated the relationship between sorbitol or PVAc concentration and the measured bioplastic properties. Each concentration was represented by one measurement; therefore, no statistical analysis of replicate variability or standard deviation was performed. We fitted linear and exponential regression models to the observed data trends, using R^2 , regression equations, and corresponding figures to describe trends in tensile strength, elongation, water uptake, and Perfect Degradation Time.

FINDINGS AND DISCUSSION

Effect of Sorbitol Variation on Tensile Strength and Elongation

Figure 1 shows how sorbitol concentration affects the tensile strength and elongation of bioplastics. Increasing sorbitol concentration from 0% to 2.3% decreased tensile strength from 34 MPa to 17 MPa. Sorbitol reduces the energy required for polymer movement and acts as an internal plasticizer by competitively hydrogen-bonding with polymer chains ([Azizati et al., 2022](#)). Its hydroxyl groups interact with the hydroxyl groups of starch/cellulose, weakening intermolecular hydrogen bonds and reducing matrix cohesiveness ([Larasati et al., 2024](#)). Consequently, the bioplastic becomes less rigid and more easily deformed under tensile loads ([Desramadhani et al., 2023](#)).

Increasing sorbitol concentration increased elongation from 14% to 37%. Sorbitol reduces polymer network density and increases polymer chain mobility, resulting in greater flexibility under tensile stress ([Azizati et al., 2022](#)). It also reduces crystallinity and hydrogen bond density, further increasing material flexibility while contributing to lower tensile strength ([Gurunathan et](#)

al., 2025). Regression analysis showed that tensile strength followed an exponential trend, $y = 33.435e^{-0.295x}$ ($R^2 = 0.9807$), while elongation followed $y = 15.862e^{0.3912x}$ ($R^2 = 0.9775$). The high R^2 values indicate a consistent relationship between sorbitol concentration and the bioplastic's mechanical properties.

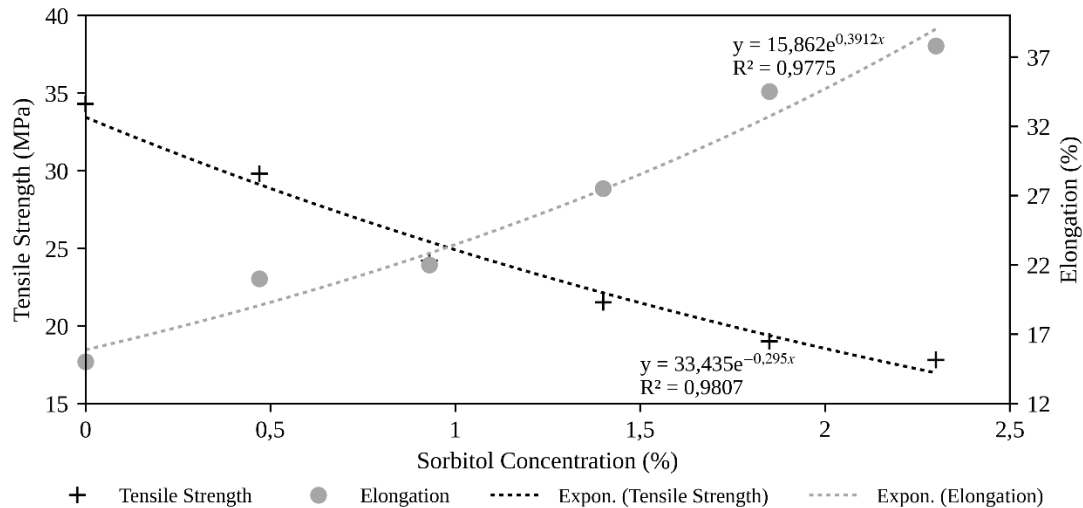


Figure 1. Relationship between sorbitol concentration and tensile strength and elongation

Bioplastics containing carrageenan, PVAc, and CMC with 0.46% sorbitol exhibited a tensile strength of 29.86 MPa and an elongation of 20.90%, compared with 15.88 MPa and 145.33%, respectively, for pure carrageenan with 0.6% sorbitol (Maryuni et al., 2018). The higher tensile strength is attributed to the reinforcing effects of PVAc and CMC, which strengthen intermolecular interactions within the composite matrix and improve load distribution (Kalsum et al., 2020). The lower elongation may also be attributed to the lower sorbitol concentration (0.46%), which provides insufficient plasticization to substantially soften the matrix, whereas 0.6% sorbitol in pure carrageenan resulted in much higher elongation (145.33%).

Effect of Polyvinyl Acetate Variation on Tensile Strength and Elongation

Figure 2 shows that increasing PVAc concentration increased tensile strength from 8.4102 to 21.9637 MPa, while decreasing elongation from 43.7378% to 33.1289%. PVAc acts as a reinforcing agent by strengthening intermolecular interactions within the carrageenan matrix (Xu et al., 2025). Hydrogen bonding between carrageenan and vinyl-based polymers can enhance film tensile strength (Shahbazi et al., 2016), consistent with reported changes in -OH and C=O peaks in PVA-carrageenan composites (Ezhilarasi, 2024).

The decrease in elongation indicates increased matrix rigidity as PVAc concentration increases, resulting from restricted polymer chain mobility (Kramár et al., 2020; Olonisakin et al., 2025). Regression analysis showed linear relationships for both tensile strength ($y = 11.876x + 8.4479$; $R^2 = 0.9818$) and elongation ($y = -8.9023x + 43.651$; $R^2 = 0.9939$), indicating consistent changes in mechanical properties with increasing PVAc concentration. Compared with ZnO nanoparticles used as a reinforcing agent in a CMC matrix, which achieved a maximum tensile strength of 15.12 MPa, the present study achieved 21.96 MPa at 1.17% PVAc (Vyas et al., 2025). This result further supports PVAc's effectiveness as a reinforcing agent for polysaccharide-based bioplastics. The mechanical properties were evaluated against SNI 7818:2014 for biodegradable

plastic bags, which specifies a minimum tensile strength of 3.44 MPa and elongation of 100%, as well as biodegradability requirements (Badan Standardisasi Nasional, 2014).

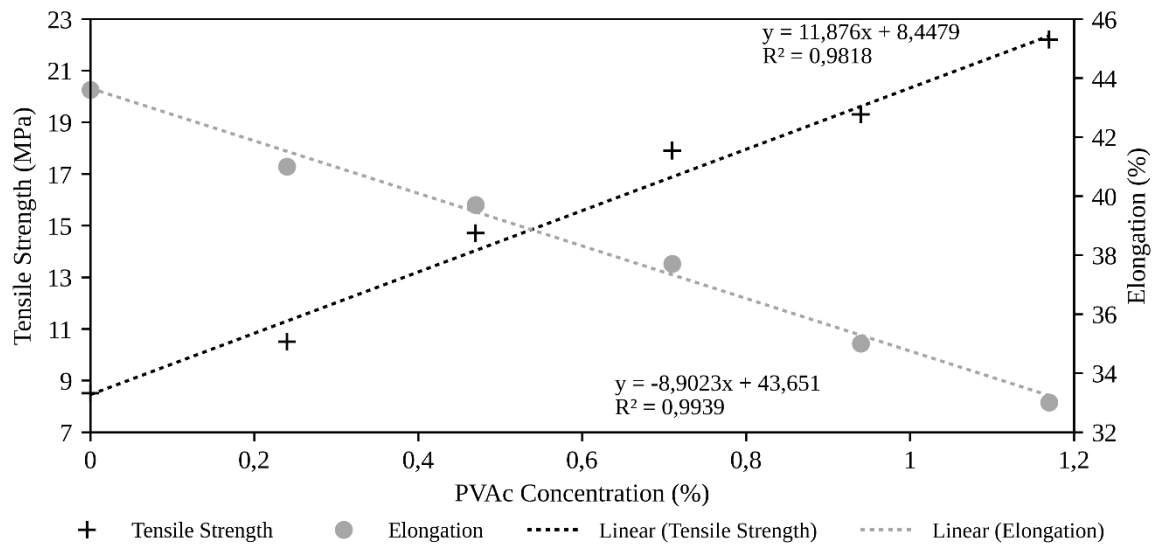


Figure 2. Graph showing the relationship between PVAc concentration, tensile strength, and elongation

Based on the minimum tensile strength requirement of 3.44 MPa set by SNI 7818:2014, all bioplastic samples met the standard, with tensile strength ranging from 17.84 MPa (P6, 2.30% sorbitol) to 34.33 MPa (P1, 0% sorbitol) for sorbitol variations and from 8.41 MPa (R1, 0% PVAc) to 21.96 MPa (R6, 1.17% PVAc) for PVAc variations. However, none met the minimum elongation requirement of 100%, with the highest values of 43.74% (R1) and 37.68% (P6), indicating the need for higher plasticizer concentrations or more flexible polymer combinations (Badan Standardisasi Nasional, 2014). For comparison, polyethylene (PE), polypropylene (PP), and polyvinyl chloride (PVC) have tensile strengths of 8–30, 31–41, and 40–60 MPa, respectively (Adetunji & Erasmus, 2024). The highest tensile strengths in this study, particularly R6 (21.96 MPa) and P1 (34.33 MPa), approach the ranges of PE and the lower range of PP, although their elongation remains lower than that of PE, which can reach 100–1000%.

Effect of Sorbitol Variation on Water Uptake and Biodegradability

Figure 3 shows that increasing sorbitol concentration from 0% to 2.5% increased water uptake from 260.28% to 311.69%. This is attributed to sorbitol's role as a hydrophilic plasticizer, which increases matrix flexibility and permeability, thereby enhancing water absorption and microbial accessibility (Rosadi et al., 2024). Sorbitol's hydroxyl (–OH) groups can also form hydrogen bonds with water, promoting matrix hydration and degradation (Afdal et al., 2022). Soil degradation generally involves water diffusion and swelling followed by enzymatic degradation and weight loss (Dirpan et al., 2023).

Increased water uptake was accompanied by a decrease in Perfect Degradation Time (PDT), from approximately 24 days without sorbitol to 15 days at 2.5% sorbitol. This indicates that higher sorbitol concentrations facilitate biodegradation by increasing water availability and microbial accessibility, while sorbitol may also serve as an organic carbon source for microorganisms (Faldo et al., 2025). Partial degradation refers to the decomposition or loss of

structural integrity without complete mineralization into CO₂ and H₂O (Gadhi et al., 2020).

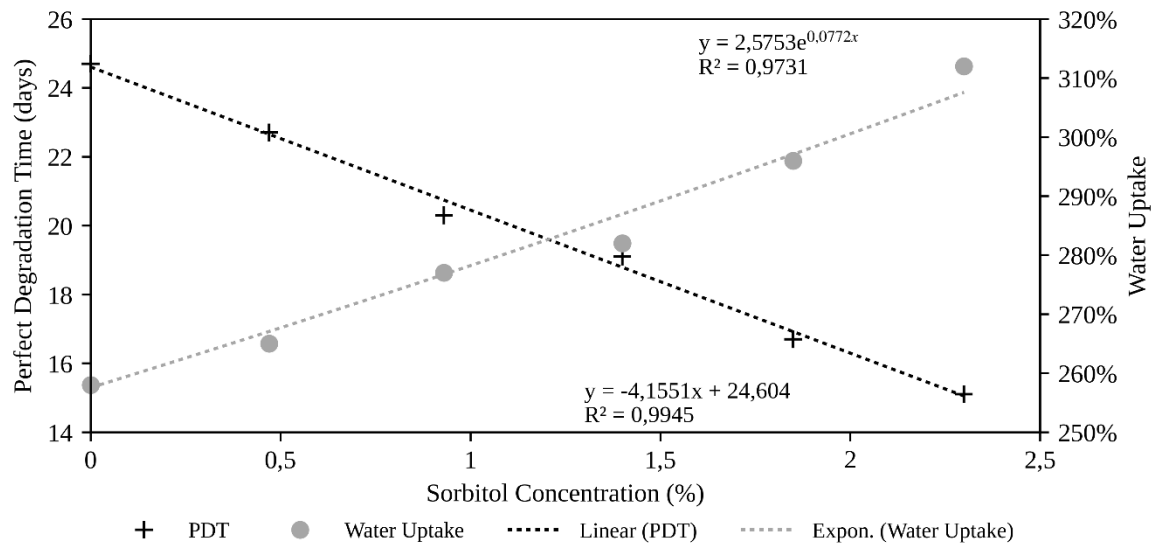


Figure 3. Graph showing the relationship between sorbitol concentration, biodegradability, and water uptake

Regression analysis showed an exponential relationship between sorbitol concentration and water uptake ($y = 2.5753e^{0.0772x}$; $R^2 = 0.9731$) and a linear relationship with PDT ($y = -4.1551x + 24.604$; $R^2 = 0.9945$), indicating that water uptake increased while degradation time decreased consistently as sorbitol concentration increased. The carrageenan-based bioplastic containing PVAc, CMC, and 2.3% sorbitol showed 311.69% water uptake and a 15-day degradation time. In contrast, Rosadi et al. (2024) reported 87.52% water uptake and a biodegradation rate of 93.42/day for pure carrageenan with 3% sorbitol. Thus, the present sample exhibited approximately 3.5 times higher water uptake. This difference may be attributed to the presence of hydrophilic CMC, whose $-\text{COO}^-$ groups increase water affinity, as well as the less dense composite matrix that facilitates water penetration. Despite its high water uptake, the sample showed a relatively longer degradation time, likely because PVAc forms a coating around carrageenan particles, requiring microorganisms to degrade the PVAc before accessing the carrageenan matrix.

Effect of Polyvinyl Acetate Variation on Water Uptake and Biodegradability

Figure 4 shows that increasing PVAc concentration increased the Perfect Degradation Time (PDT) from 12 days at 0% PVAc to 23 days at 1.2%, indicating slower biodegradation. This is attributed to the increasing PVAc content, a vinyl polymer whose carbon-carbon (C-C) backbone resists microbial and enzymatic degradation (Amann & Minge, 2011).

Increasing PVAc concentration also reduced water uptake. This is attributed to PVAc's hydrophobic acetate groups, which limit water diffusion into the polymer matrix (Asni et al., 2015). In contrast, carrageenan contains hydrophilic hydroxyl and sulfate groups that promote water absorption; therefore, adding PVAc reduces the matrix's overall water affinity (Amann & Minge, 2011). Regression analysis showed a linear relationship between PVAc concentration and water uptake ($y = -1.129x + 3.7835$; $R^2 = 0.9696$) and an exponential relationship with PDT ($y = 11.379e^{0.5113x}$; $R^2 = 0.9523$), confirming that PVAc addition reduced water uptake while increasing degradation time. The observed trend is consistent with Xu et al. (2025), who reported that PVAc can improve film structural integrity while reducing substrate accessibility to

microorganisms. Nevertheless, the bioplastics in this study degraded within approximately 12–22 days even at the highest PVAc concentration, substantially faster than conventional plastics.

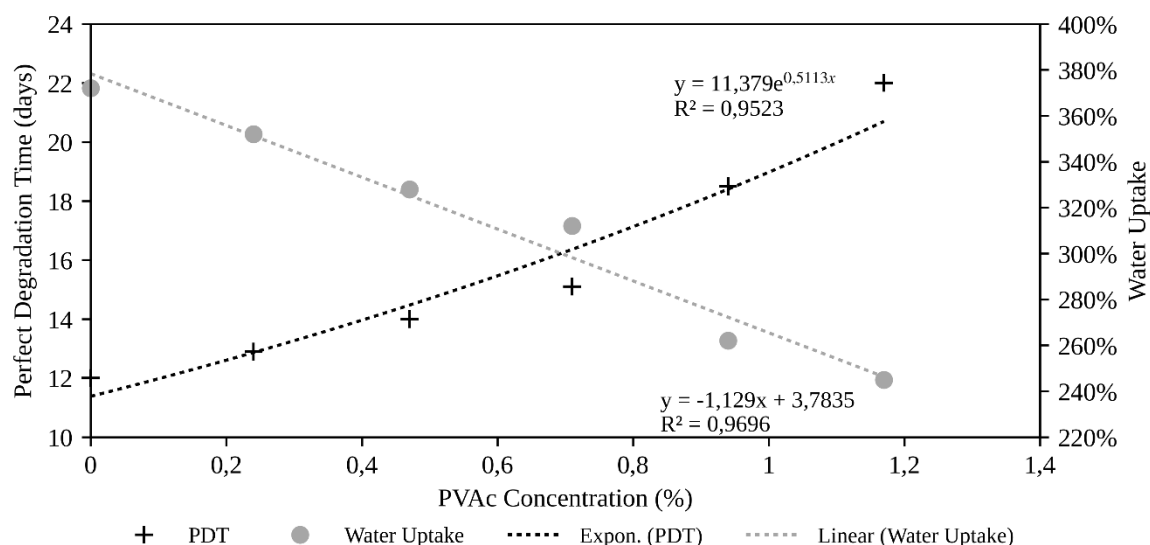


Figure 4. Graph showing the relationship between PVAc concentration, biodegradability, and water uptake

According to SNI 7188.7:2016, bioplastic products should achieve at least 90% degradation within 180 days under aerobic composting conditions ([Badan Standardisasi Nasional, 2016](#)). The bioplastics in this study degraded completely within 15–25 days, exceeding this requirement. However, their high water uptake limits applications requiring high water resistance. Overall, this study's novelty lies in systematically evaluating sorbitol and PVAc as a plasticizer and reinforcing agent, respectively, within the same carrageenan-based bioplastic system. Unlike previous studies that primarily investigated sorbitol or PVAc individually, this study evaluates their respective effects on mechanical properties, water uptake, and biodegradability within the same formulation framework, providing a broader basis for optimizing carrageenan-based bioplastic formulations.

FTIR Test Results

Figure 5 shows the FTIR results, with an absorption band at 3369 cm^{-1} indicating O-H groups, which are abundant in polysaccharides. This broad absorption reflects an extensive network of intramolecular and intermolecular hydrogen bonds, characteristic of hydrophilic polymers such as carrageenan. The absorption band at 2916.71 cm^{-1} indicates C-H groups commonly found in polysaccharide polymers like carrageenan. An absorption band around 1640 cm^{-1} indicates O-H bending vibrations resulting from water molecules physically bound to the polymer ([Nandiyanto et al., 2019](#)). Absorption bands at $1223\text{--}1225\text{ cm}^{-1}$ indicate sulfate ester groups (S=O) derived from iota-, kappa-, and lambda-type carrageenans. Absorption peaks at 1065 cm^{-1} and 1036 cm^{-1} identify C-O-C stretching vibrations associated with the glycosidic bonds of the polysaccharide chains. The band at 921 cm^{-1} represents the characteristic vibration of 3,6-anhydrogalactose, a major component of carrageenan. The peak at 843 cm^{-1} is a characteristic marker for the galactose-4-sulfate group, confirming the presence of kappa-carrageenan ([Giarni et al., 2025](#)).

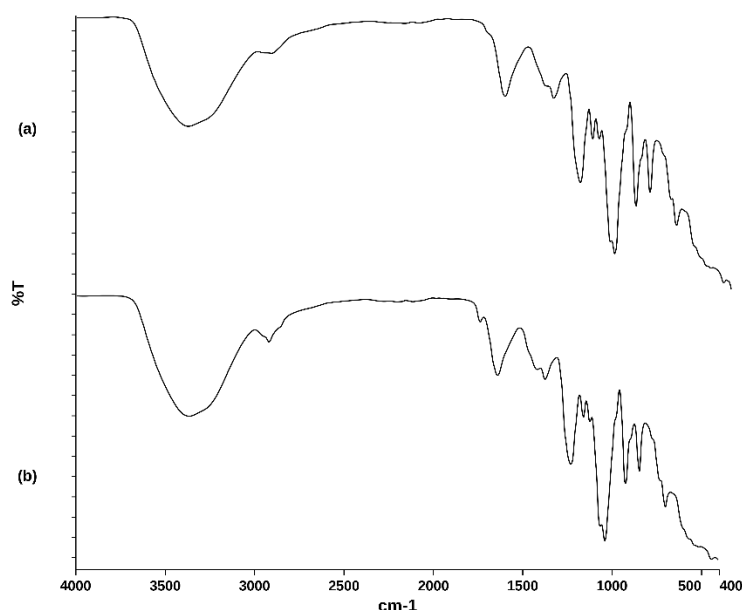


Figure 5. FTIR test results for (a) sample P1 and (b) sample P2

The main difference between P1 and P2 is an absorption peak at 1735 cm^{-1} in P2, corresponding to the C=O stretching vibration of the PVAc ester carbonyl group, consistent with the reported PVAc range of $1720\text{--}1740\text{ cm}^{-1}$ (Kaya et al., 2014). Shifts in the -OH and C=O peaks have also been reported in PVA-carrageenan composites, indicating intermolecular interactions (Ezhilarasi, 2024). The absence of new peaks beyond those associated with the constituent components suggests that carrageenan, sorbitol, PVAc, and CMC interact primarily through physical interactions such as hydrogen bonding (Sasria et al., 2025). The lower %T of the O-H band in P2 is associated with the addition of sorbitol, while the consistent kappa-carrageenan fingerprint peaks indicate that its chemical structure remained unchanged (Sasria et al., 2025).

SEM Test Results

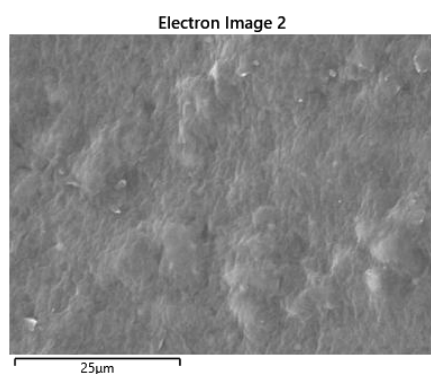


Figure 6. Surface morphology of P1 Sample

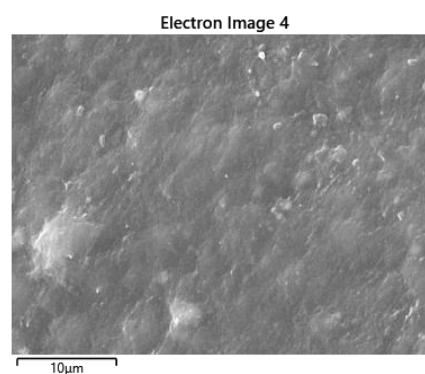


Figure 7. Surface morphology of P2 sample

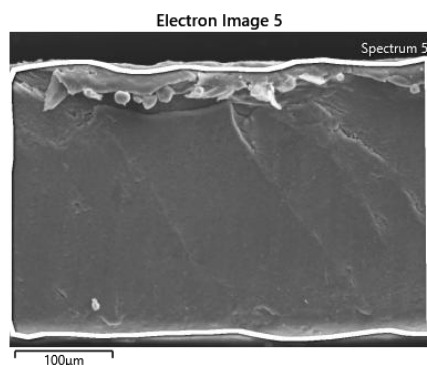


Figure 8. Cross-sectional morphology of P1 sample

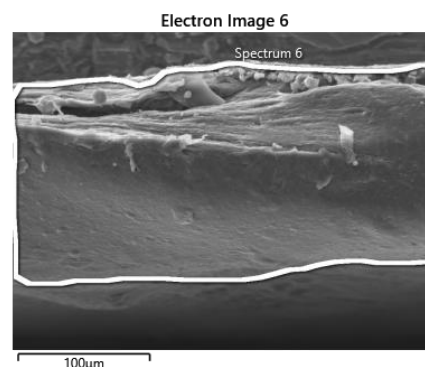


Figure 9. Cross-sectional morphology of P2 sample

Figures 6–9 show SEM images of the surface and cross-sectional morphology of samples P1 and P2. Sample P1 exhibited a relatively flat but wavy surface with localized agglomeration, attributed to the absence of sorbitol and the resulting strong intermolecular interactions between polymer chains (Sasria et al., 2025). Its cross-section showed a relatively dense, layered structure with a more uniform inner matrix, indicating CMC's contribution to matrix homogeneity (Suryanto et al., 2025). In contrast, P2 exhibited a rougher surface with more evenly distributed protrusions and a more irregular cross-section with visible gaps. Adding sorbitol weakens intermolecular interactions and produces a looser polymer matrix (Kaya et al., 2014). The absence of large cracks also indicates CMC's contribution to maintaining matrix integrity (Mahardika et al., 2025).

Table 1. EDS elemental composition of sample P1

Element	Wt%	Wt% Sigma
C	37.33	3.95
N	0.00	5.40
O	48.68	3.29
Na	4.82	0.60
Mg	0.04	0.20
P	0.02	0.19
S	5.35	0.51
K	3.46	0.38
Ca	0.31	0.18
Fe	0.00	0.23
Total	100.00	

Table 2. EDS elemental composition of sample P2

Element	Wt%	Wt% Sigma
C	39.15	3.26
N	0.00	5.29
O	45.97	2.74
Na	3.99	0.55
Mg	0.14	0.21
P	0.00	0.22
S	6.21	0.55

Element	Wt%	Wt% Sigma
K	3.75	0.42
Ca	0.76	0.25
Fe	0.02	0.30
Total	100.00	

EDS analysis of P1 showed that oxygen (O) and carbon (C) were the dominant elements at 48.68% and 37.33%, respectively, consistent with the polysaccharide-based carrageenan matrix. Sulfur (S) at 5.35% indicates sulfate groups in kappa-carrageenan, while Na (4.82%) may originate from residual Na⁺ ions from the 0.1 N NaOH extraction and K (3.46%) from the red algae (Suryanto et al., 2025). For P2, the carbon content increased to 39.15%, while oxygen decreased to 45.97%. Sulfur also increased from 5.35% to 6.21%, indicating differences in the distribution of carrageenan sulfate groups. The C and O compositions remained relatively similar between the samples, suggesting that CMC helped maintain matrix homogeneity and prevent significant phase separation (Suryanto et al., 2025).

The SEM results are consistent with Maryuni et al. (2018), who reported that adding sorbitol produces a more irregular surface morphology in carrageenan-based bioplastics. The observed agglomeration in P1 and more distributed protrusions in P2 are also consistent with the effects of plasticizer addition reported by Sasria et al. (2025). The relatively homogeneous cross-sectional morphology further supports CMC's role in maintaining matrix structure (Suryanto et al., 2025). Overall, the SEM and EDS results support the structural differences between P1 and P2 observed after sorbitol addition, consistent with the FTIR results.

CONCLUSIONS

Carrageenan-based bioplastics from *Eucheuma cottonii* were successfully produced using sorbitol and PVAc. Sorbitol increased elongation, water uptake, and biodegradation rate, while PVAc increased tensile strength and water resistance but reduced elongation and biodegradation rate. All samples met the SNI minimum tensile strength requirement and completely degraded within 15–25 days.

LIMITATIONS & FURTHER RESEARCH

This study was limited by using a single measurement for each formulation (n = 1), which prevented evaluation of experimental variability and calculation of standard deviations. In addition, sorbitol and PVAc concentrations were varied separately, so their interaction effect was not statistically evaluated, and none of the samples met the minimum elongation requirement of 100%. Further research should employ replicated experiments and a factorial experimental design or response surface methodology to optimize sorbitol and PVAc composition and improve the balance among mechanical strength, flexibility, water resistance, and biodegradability.

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