Journal of Metals, Materials and Minerals, 35(3), e2236, 2025

Optimization and characterization of carrageenan/alginate ratio and seaweed waste to develop composite bioplastics from *Kappaphycus alvarezii*

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Received date:
17 December 2024
Revised date:
1 April 2025
Accepted date:
1 June 2025

Keywords:
Alginate;
Bioplastics;
Biopolymer;
Carrageenan;
Composites

Abstract

Carrageenan and alginate, derived from seaweed, offer potential as alternatives to traditional plastics. This study aims to assess the properties of biodegradable bioplastics made from a composite of carrageenan, alginate, and waste from *Kappaphycus alvarezii* seaweed carrageenan extraction. A mixture of carrageenan and alginate was added with a ratio of 4:6 and *Kappaphycus alvarezii* extraction waste was 0%, 20%, 40%, 60%, and 80% waste concentration, in the carrageenan-alginate-waste mixture. The process involves blending carrageenan, alginate, waste, glycerol, and distilled water until uniform at 90°C and 1000 rpm for 45 min, followed by molding, immersion in a 4% $CaCl_2$ solution, and drying at 60°C for 18 h. The casting method was used for producing the bioplastics, and their physicochemical properties were tested using standard methods. Findings reveal that incorporating waste enhances flexibility and biodegradability but diminishes tensile strength and water resistance. FTIR spectra indicate no chemical interaction among the constituents. Scanning electron micrographs demonstrate that waste addition up to 60% yields a uniform bioplastic surface, while at 80%, it leads to fractures. Interestingly, these bioplastics exhibit varied features that offer various applications by considering sustainability and environmental impact in materials development of seaweed-derived biopolymers as alternatives to conventional plastics.

1. Introduction

Kappaphycus alvarezii stands out as a significant red seaweed due to its considerable economic importance, primarily stemming from the production of k-carrageenan colloid. This substance has wide-ranging applications in food, pharmaceuticals, cosmetics, packaging, bioplastics, and composite materials [1]. The k-carrageenan content within the seaweed typically ranges from 25% to 35% of its dry weight, leaving the remainder as unused waste [2]. Research examining biodegradability, compatibility, and economic impact demonstrates seaweed's potential as a suitable material for biodegradable films and as a component in composite materials alongside other polymers, offering diverse application opportunities [3].

Carrageenan is a linear galactan sulfate hydrocolloid, which is composed of a series of β-D-galactopyranose bound at the C3 position (β-Gal, unit G) with α-D-galactopyranose bound at the C4 position (α-Gal, unit D). Unit D is sometimes also found in the form of 3,6-anhydro-α-D-galactopyranose (anGal, unit A) forming a disaccharide unit that makes up the carrageenan polymer chain [4]. Carrageenan gel can form a thin layer, so it has the potential to be used as a material for making bioplastics [5], but its mechanical quality, water resistance, and thermal properties do not meet the requirements. This deficiency can be overcome by adding filler, chemical modification, or mixing it with other biopolymers such as alginate.

Alginate is a polysaccharide obtained from brown algae (Phaeophyta), a linear copolymer of D-mannuronic acid and L-guronic acid which is arranged in a blockwise or alternating pattern along the polymer chain. The homopolymer groups of mannuronic acid (M block) and guluronic acid (G block) are alternated with M and G (MG block). The ratio of G/M alginate monomer units depends on the type, origin, age, and harvest time of seaweed [6]. Sodium-alginate was reported to increase compatibility and biodegradability and form fewer voids in starch/agar/LDPE composites [7]. Carrageenan and alginate can form compatible, degradable, non-toxic, synergistic composites in improving the mechanical properties of films and can form hydrogels with Ca^{2+} and K^{+} [8].

The carrageenan extraction process from K. alvarezii seaweed produces a solid residue of 60%w/w to 70%w/w, of which one-third is cellulose, 4.5%w/w lignin, and 4.5%w/w hemicellulose [9-11], which is considered waste that pollutes the environment and disturbs aesthetics. Sustainable and rational use of this waste in bioproducts is a useful innovation for reducing the effects of ecological hazards, useful in the development of economically valuable and sustainable materials such as nanocomposites [12], bioethanol [10], nanocrystalline cellulose [1], biofilm materials [13], and bioplastic film-reinforcing agents [14].

Preparation of k-carrageenan and alginate mixture films has been carried out by several researchers. Roh and Shin [15] found that the

DOI:10.55713/jmmm.v35i3.2236

best composition was achieved in the film with a ratio of alginate: carrageenan = 6:4. Edible film preparation from a mixture of k-carrageenan, i-carrageenan, and alginate was carried out by Paula *et al.* [4], who proved that k-carrageenan can increase the water resistance and tensile strength of edible films, while alginate makes bioplastics more uniform, homogeneous, and transparent. Biodegradable film from a mixture of starch/polyvinyl alcohol and alginate as lettuce packaging has been demonstrated by Brandelero *et al.* [16], while Farhan and Hani [5] have characterized edible packaging from Semi Refined Carrageenan (SRC) with glycerol and sorbitol plasticizers.

Seaweed hydrocolloid films are hydrophilic so their mechanical properties, water blocking (water resistance), and thermal properties are limited. Efforts to improve the quality of hydrocolloid bioplastics include, among other things, the addition of nanocrystalline cellulose, micro cellulose, nano clay, etc. which have been reviewed by Khalil *et al.* [14], while efforts to improve the characteristics of carrageenan-based biofilm properties have been reviewed by Sedayu *et al.* [17]. The addition of supporting materials and chemical engineering requires relatively expensive chemicals which will affect the production costs and selling prices of bioplastic products, so in this research, the strengthening of carrageenan/alginate composite bioplastics was carried out by adding waste from *Kappaphycus alvarezii* seaweed extraction as a biofiller.

Natural biofillers such as talc, clay, and eggshells are reported to be able to improve the mechanical properties of starch bioplastics [3], but research on adding fillers from seaweed waste is still rare. The application of seaweed waste for reinforcement has been carried out on starch-based bioplastics [13,18-20]. Research on the use of carrageenan waste as raw material for bioplastics has been carried out using sorbitol plasticizers [21] and glycerol [22]. This research aims to explore the potential of K. alvarezii seaweed extraction waste as a reinforcement for carrageenan-alginate composites. The resulting bioplastics can be developed into replacement products for conventional plastics to reduce plastic pollution. This study presents a novel contribution to the field of bioplastic development and sustainable materials science, particularly by utilizing marine biomass from *Kappaphycus alvarezii*. Some previous studies use pure, refined biopolymers in optimizing bioplastic composites [45-47], instead, this study is the first to optimize and characterize bioplastic composites based on varying carrageenan/alginate ratios using both primary and waste biomass from *Kappaphycus alvarezii*. Further, the research supports sustainable material development by adopting a zero-waste approach through the incorporation of seaweed waste a low-cost and underutilized byproduct—into the bioplastic formulation.

2. Experimental

2.1 Bioplastic making

Bioplastics were made using the method of Lim *et al.* (23). 146.25 mL of distilled water was heated using a hot plate stirrer to a temperature of 90°C. After that, a mixture of carrageenan and alginate was added with a ratio of 4:6 referring to Roh and Shin [15] and *Kappaphycus alvarezii* extraction waste with the formulation as in Table 1. The mixture was homogenized again for 45 min at a speed of 1000 rpm, and then 1.5 mL of glycerol was added and homogenized again for 15 min.

The hydrogel was poured into a 22 cm × 17 cm × 3.5 cm polypropylene mold and then dried in an oven at 60°C for 18 h. After that, the bioplastic is released from the mold and then soaked in 4% $CaCl_2$ crosslinker solution for 5 min [15], then dried at room temperature for 6 h [20].

2.2 Bioplastic characterization

2.2.1 Thickness and opacity

The thickness of bioplastics is determined according to ASTM D 6988-13. The opacity of bioplastics was determined using a lux meter [16]. The environmental light intensity value is measured with a lux meter (lux i), then the bioplastic is placed on top of the lux meter sensor and the light intensity (lux f) is measured.

$$\text{Opacity (\%)} = \frac{\text{Lux i-Lux f}}{\text{Lux i}} \times 100 \qquad (1)$$

2.2.2 Water resistance

This test was performed according to Yang *et al.* [24]. The bioplastic was cut to a size of 20 mm × 20 mm, weighed (M0) then placed in a container containing water (50 mL) and left for 24 h at 25°C. The remaining bioplastic was removed and dried at a temperature of 105°C, then weighed (M1).

$$\text{Solubility in water (\%)} = (M0 - M1)/M0 \times 100\% \qquad (2)$$

M0 = Initial sample weight (g)
M1 = Final sample weight (g)

Table 1. Formulation of Carrageenan/Alginate Composite Bioplastics and *K. alvarezii* Waste.

Formulation code	Mixture composition				
	Waste [g]	Carrageenan [g]	Alginate [g]	Glycerol [mL]	Aquades [mL]
F0	0	0.9	1.35	1.5	146.25
F20	0.45	0.72	1.08	1.5	146.25
F40	0.9	0.54	0.81	1.5	146.25
F60	1.35	0.36	0.54	1.5	146.25
F80	1.8	0.18	0.27	1.5	146.25

*Note: F20 means 20% waste concentration in the carrageenan-alginate-waste mixture, etc.

2.2.3 Mechanical properties (tensile strength and elongation)

Mechanical properties were determined according to ASTM D 882-18. The bioplastic sample was cut to a size of 1 cm × 15 cm, then clamped at both ends and pulled at a speed of 50 mm·min^{-1}.

$$\text{Tensile strength} = \text{Fmax} / \text{A} \tag{3}$$

Fmax= Maximum force (N)
A = Bioplastic surface area (cm^2)

$$\text{Elongation (\%)} = (\text{A} - \text{B})/\text{B} \times 100\% \tag{4}$$

A = Length of bioplastic after breaking
B = Initial length of bioplastic

2.2.4 Biodegradability

The biodegradation test was carried out based on the method of Behera *et al*. [25]. Bioplastic samples were cut into 2 cm × 2 cm pieces and placed in a desiccator for 24 h and weighed (W0). The samples were buried in the soil at a depth of 2 cm for 6 days, then washed using distilled water, then dried with tissue paper and dried in a desiccator for 24 h, then weighed (W1).

$$\text{Biodegradation (\%)} = (\text{W0} - \text{W1})/\text{W0} \times 100\% \tag{5}$$

W0 = Initial weight of sample (g)
W1 = Final weight of the sample (g)

2.3 Fourier transform infrared spectroscopy (FTIR)

The test was carried out with an infrared spectrometer (FTIR) at wave numbers 4000 cm^{-1} to 650 cm^{-1}.

2.4 Scanning electron microscopy (SEM)

SEM scanning was carried out to determine surface morphology such as structure formation, crack identification, roughness, and density of bioplastic structures [26].

2.5. Data analysis

All data obtained (thickness, opacity, water resistance, and tensile strength) were analyzed by parametric statistics using normality tests, homogeneity tests, and ANOVA tests. Determining significantly different variants was carried out by looking at the comparison values using the Tukey test at a confidence level of 95%. Statistical tests were carried out using SPSS 25 software.

3. Results and discussion

3.1 Sample characteristics

The sample preparation and extraction process of *Kappaphycus alvarezii* are illustrated in Figure 1. As shown in Figure 1(a), the seaweed exhibits a brown color, a smooth texture, and an elongated, branched thallus. The seaweed was thoroughly washed and sun-dried until its

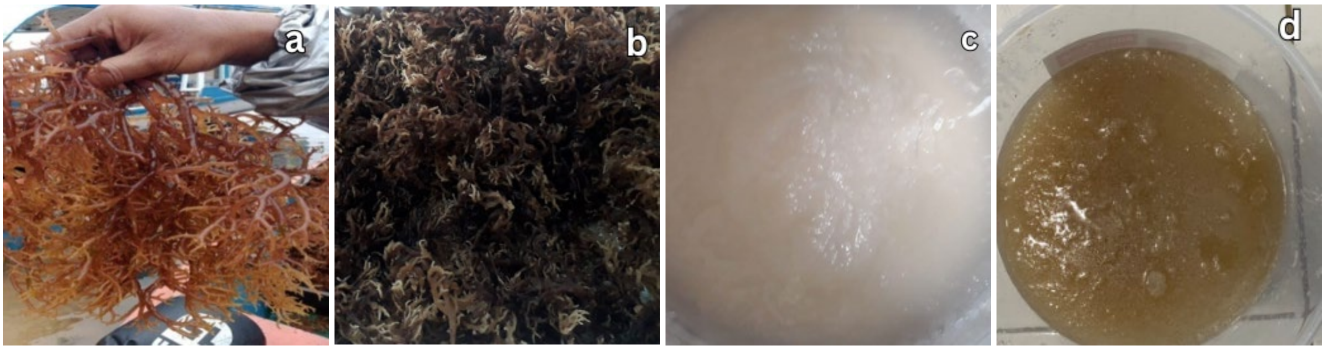

Figure 1. Research materials ((a) fresh *K. alvarezii*, (b) dry *K. alvarezii* (moisture content < 15%), (c) wet carrageenan, (d) wet carrageenan extraction waste

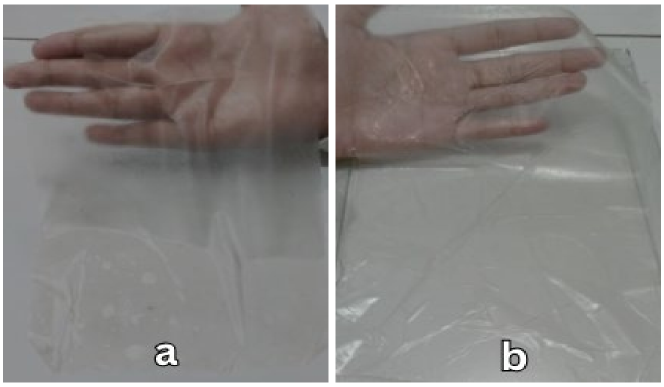

Figure 2. Carrageenan-alginate bioplastic with glycerol plasticizer (Note: (a) Without adding decarrageenan, (b) with adding 20% decarrageenan waste.

moisture content dropped below 15% [Figure 1(b)]. The extracted wet carrageenan appears white in color [Figure 1(c)], while the extraction residue of *K. alvarezii* is a thick, semi-solid, brownish material with no noticeable odor (Figure 1(d)). The bioplastics produced in this study are presented in Figure 2. In general, bioplastic without the addition of waste looks more transparent, and the surface is smoother and more flexible [Figure 2(a)], whereas the bioplastic produced with the addition of waste is white, transparent, and slightly opaque [Figure 2(b)]. This difference may be attributed to the fact that higher levels of waste result in thinner bioplastic films, allowing light to pass through more easily. With increased waste levels, the proportion of carrageenan and alginate in the bioplastic decreases, promoting greater mobility within the polymer matrix and allowing light to penetrate the material.

3.2 Physical and mechanical properties of bioplastics

Figure 3 presents the effect of decarrageenan waste concentration on the physical and mechanical properties of the bioplastics, including the following characteristics:

3.2.1 Opacity and thickness of bioplastics

All bioplastics show good texture, and relatively high levels of transparency (low opacity) between 13.27 ± 1.254% (L0) to 20.379 ± 0.948% (L80) [Figure 3(a)]. According to Paşcalau et al. (8), the combination of alginate and carrageenan produces a see-through and colorless film. As the concentration of waste increases, these bioplastics tend to become even more transparent. This result is in contrast to Sedayu et al. [17], where the high cellulose content in waste tends to increase the opacity of bioplastics. This is thought to be because the higher the concentration of waste added, the bioplastic becomes thinner, or the thickness of the bioplastic decreases so that light passes through more easily. At high waste levels, the amount of carrageenan and alginate is relatively small, so that chain mobility in the inter-molecular space in the alginate and carrageenan polymer matrix becomes easier so that light can penetrate the bioplastic.

The thickness of bioplastic without the addition of decarrageenan waste (L0) measured 107.78 ± 7.26 µm, whereas bioplastic with 80% decarrageenan waste (L80) had a thickness of only 83 ± 9.33 µm. As depicted in Figure 3(b), there is a tendency for bioplastic thickness to decrease with increasing waste concentration. Sudhakar *et al*., [48] attribute this variation in bioplastic thickness to the molding process and the formulation of the polymer mixture utilized in bioplastic production. Decreasing levels of carrageenan and alginate result in reduced dispersion of the glycerol plasticizer within the intermolecular spaces of the polymer matrix, thereby causing a decrease in bioplastic thickness [49]. It's worth noting that the bioplastic thickness observed in this study complies with the standard specified by the Japanese Industrial Standard, which mandates a thickness of less than 250 µm.

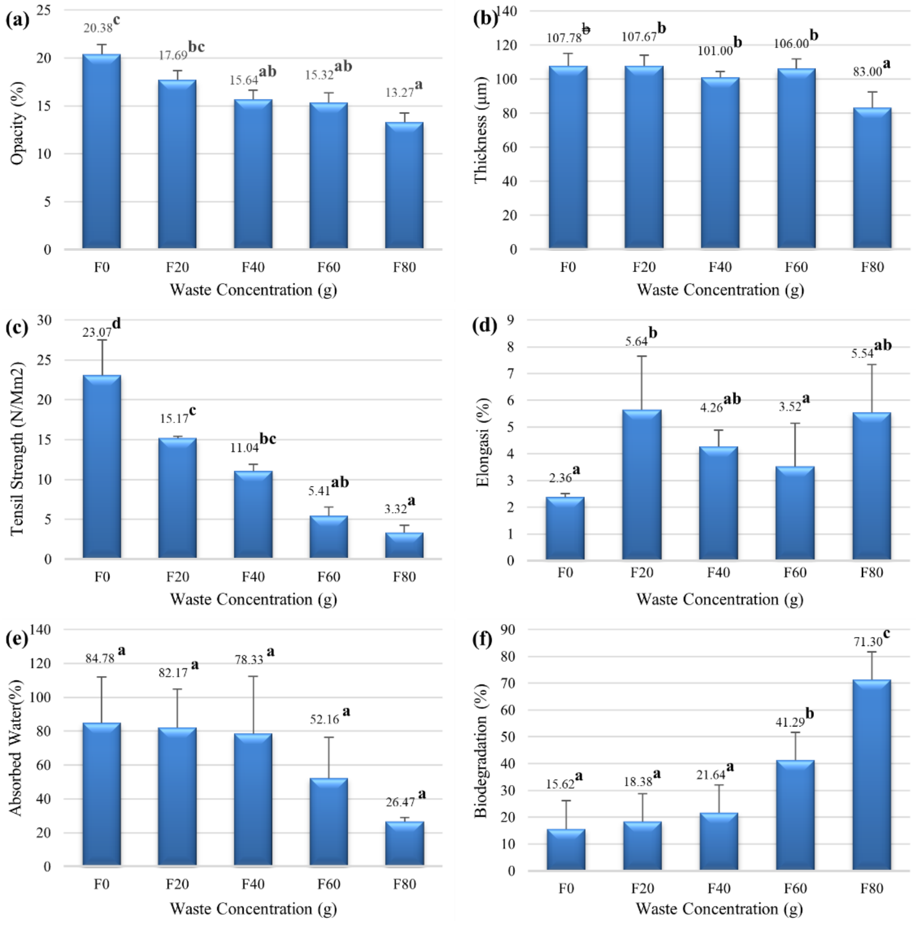

Figure 3. Effect of decarrageenan waste concentration on the opacity (a), thickness (b), tensile strength (c), elongation (d), absorbed water (e), and biodegradation (f) of bioplastics (Note: different superscript letters indicate significant differences ($p<0.05$)

3.2.2 Tensile strength and elongation of bioplastics

The addition of waste significantly reduces the tensile strength of bioplastics. The highest tensile strength was achieved by bioplastics without the addition of decarrageenan waste, namely 23.07 ± 4.41 $N \cdot mm^{-2}$, and decreased as the waste concentration increased. With the addition of 80% waste, the tensile strength of the bioplastic was 3.32 ± 0.94 $N \cdot mm^{-2}$ [Figure 3(c)]. This result is different from Jumaidin *et al.* [3], where the optimum tensile strength occurs at an addition of 30% to 40%, after which it decreases as the waste filler increases. This phenomenon of decreasing tensile strength is caused by the effect of matrix discontinuity due to the high concentration of waste which causes reduced stress transfer from the matrix to the waste [27]. The relatively high tensile strength of bioplastics without waste is thought to be caused by the similar hydrophilic character between carrageenan and alginate, resulting in good compatibility and increased adhesion, whereas waste is less compatible with carrageenan and alginate, resulting in lower tensile strength [28]. The tensile strength of pure alginate varies according to various references, namely 60 MPa [29], 122 MPa [30], or 34 MPa [31], while the tensile strength of carrageenan is 40.30 MPa [5] and 42.5 MPa [17]. The addition of glycerol will also reduce the tensile strength of bioplastics according to research by Jost *et al.* [29]. The same thing also happens to the thermoplastic chitosan [32]. Tensile strength is also influenced by the origin of seaweed and its treatment (eg M/G ratio, viscosity, composition), manufacturing method (casting or thermo mechanic, mixing time or thickness), and level of dryness (humidity, drying time). The tensile strength and elongation of bioplastics in this study were lower compared to commonly used plastics such as polyvinyl alcohol (44 MPa to 64 MPa), HDPE (22 MPa to 31 MPa), polypropylene (31 MPa to 38 MPa) and polystyrene (45 MPa to 83 MPa) [33].

The elongation of bioplastics treated with waste fluctuated but was higher than without the addition of waste (Figure 3(d)), the highest was achieved at 20% addition (L20), but at 40% waste treatment it decreased, and increased again at 60% and 80% treatment. The reduction in bioplastic elongation with the addition of 40% and 60% waste is thought to be due to the formation of new hydrogen bonds between carrageenan, alginate, and waste (as shown in the FTIR spectra) so that the mobility of the alginate and carrageenan polymer chains is reduced and increases resistance to deformation. Apart from that, it is suspected that the remaining carrageenan in the waste forms a good network with alginate and glycerol, thus strengthening the matrix [28]. This result is somewhat different from Jumaidin *et al.* [3], where maximum elongation occurs at 30% waste addition and decreases at higher waste additions. In applications such as food packaging, bioplastic material with good mechanical properties (tensile strength and elongation) is very necessary to maintain and protect the integrity of food ingredients from damage/rotting during storage and transportation.

3.2.3 Water resistance and biodegradability of bioplastics

Water resistance is one of the important properties of bioplastic materials. Water absorption capacity shows the ability of bioplastics to absorb water. The lower the water absorption capacity the more water resistant the bioplastic is. Decarrageenan waste reduces the water absorption capacity of bioplastics, or its water resistance becomes stronger, which virtually shows that the bioplastics are still intact when soaked in water for 24 h [Figure 3(e)]. Cellulose in waste acts as a reinforcement agent for bioplastics [14]. The addition of waste causes cross-linking to quickly form a cold-setting gel that is stable and insoluble in water [34]. In carrageenan-alginate composites, alginate molecules occupy a position on the outside of the film surface because alginate is more hydrophilic than carrageenan [35].

Decarrageenan waste increased the biodegradability of bioplastics by up to 36.4% [Figure 3(f)]. As more waste is added, the biodegradability of bioplastics also increases. Biodegradability shows the ease with which a material can be degraded in the environment. The reduced weight of bioplastics is an indicator of the biodegradation process by moisture and microorganisms during immersion in the soil [36]. The addition of decarrageenan waste to the carrageenan-alginate matrix causes a decrease in the weight of the composite, thus indicating a faster biodegradation process. According to Tajeddin *et al.* [37], the more cellulose content in bioplastics, the easier it is to degrade. Similar results were reported on the addition of agar, carrageenan, and Na-alginate in thermoplastic starch matrices [38]. Biodegradability is also influenced by soil type, type of microorganisms, and humidity. Water is a medium for most microorganisms such as bacteria.

3.3 Bioplastic microstructure (FTIR)

The FTIR spectra of carrageenan-alginate-waste composite bioplastics show the same pattern (Figure 4). This shows that bioplastics do not undergo chemical reactions/modifications due to the addition of waste. This result was also reported for the addition of coconut fiber to thermoplastic cassava starch [39]. The FTIR spectra of bioplastics did not show any new peaks originating from the thermal decomposition of alginate and carrageenan, because the process of making bioplastics in this study was carried out at temperatures below 100°C. In contrast to the research of Jumaidin *et al.* [40] which shows a new peak at around 1700 cm^{-1} originating from the carbonyl group (–C=O) of ketone, aldehyde, or carboxylic acid compounds caused by the thermal decomposition of carrageenan and alginate. The absence of new peaks indicates that no chemical reaction occurs between components and each component plays its role [4]. The peak at 3200 cm^{-1} to 3400 cm^{-1} indicates the stretching vibration of the hydroxyl group (–OH), both in the free position and bound inter and between molecules, which are hydrogen bonded to each other. The peak around 2900 cm^{-1} comes from the –C–H stretching vibration of the methylene ($–CH_2–$) and/or methyl ($–CH_3$) groups. Peak in the range of 1020 cm^{-1} to 1089 cm^{-1} is a typical peak of the –C–O– anhydrous glucose strain. The peak at 1300 cm^{-1} comes from the bending vibration of the methyl group ($–CH_3$).

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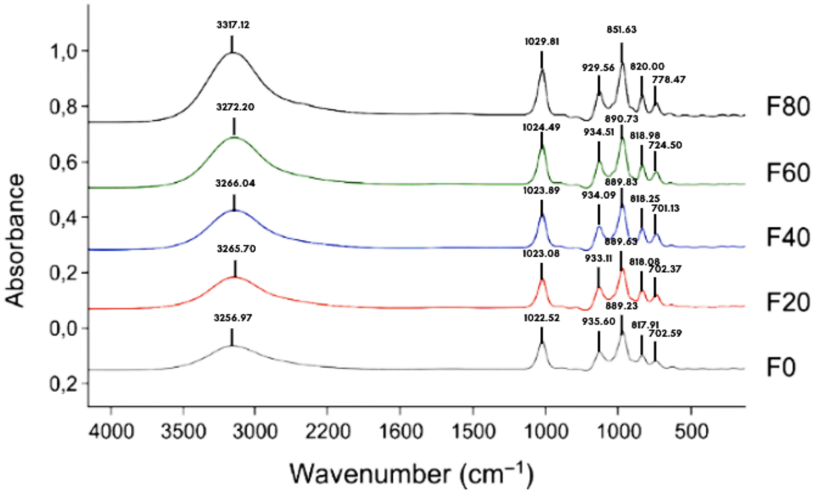

Figure 4. Overlay of FTIR spectra of bioplastics.

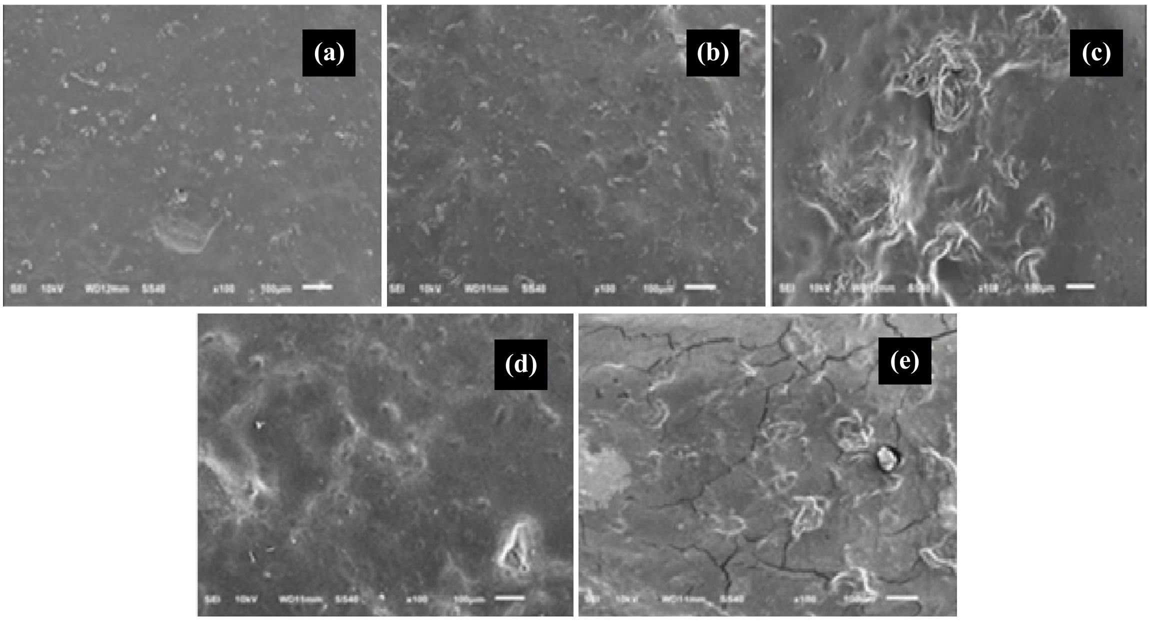

Figure 5. SEM Bioplastic Surface 100 times magnification. (a) Bioplastics without added waste, (b) added 20% waste, (c) additional waste 40%, (d) additional waste 60%, and (e) Bioplastic composite with 80% waste addition.

3.4 Bioplastic surface (SEM)

Figure 5(a) shows a Scanning Electron Micrograph (SEM) of the surface of carrageenan-alginate bioplastic without the addition of waste, while Figure 5(e) is a SEM image of carrageenan-alginate bioplastic with the addition of 80% waste (L80). The surface of bioplastics without added waste tends to be rough, while bioplastics with added waste look uniform and homogeneous. Alginate is more hydrophilic than carrageenan, so alginate will occupy the outside of the bioplastic film during the manufacturing process so that the surface of the bioplastic is slightly different from the inside of the bioplastic [42].

The SEM image shows that there is no phase separation which shows good compatibility between waste and carrageenan, alginate, and glycerol. This finding can be caused by first: the waste has the same hydrophilic character as the carrageenan-alginate matrix so that good adhesion occurs so that the bioplastic surface is more homogeneous, second: the carrageenan, agar, and waste melt and mix during heating and stirring so that the surface becomes homogeneous, and the cause third: carrageenan, alginate and waste are thought to melt together during the mixing process to form a stable mixture. Other research shows that carrageenan can form a homogeneous structure when mixed and heated [43].

Different results were shown with the addition of 80% waste, [Figure 5(e)] which showed the presence of cracks, which were thought to be caused by the bonds between each alginate, carrageenan, and waste molecules from being stronger than the composite which prevented deformation before cracking, or the waste prevented the formation of crosslink bonds between carrageenan polymer chains with alginate [8]. These cracks are evidence that the addition of 80% waste has an effect on matrix discontinuity at high waste levels. The occurrence of cracks was also reported in starch films [44] and starch-agar composites with the addition of *Eucheuma cottonii seaweed* [3].

4. Conclusions

The addition of *Kappaphycus alvarezii* seaweed extraction waste increases the biodegradability of carrageenan/alginate composite bioplastics but does not improve its mechanical properties. In general, the addition of 20% decarrageenan waste produces the best bioplastic because no cracks occur and hydrogen bonds between polymers are maintained. These results indicate that *Kappaphycus alvarezii* seaweed decarrageenan waste can be used as a filler in carrageenan/alginate composite bioplastics. Findings reveal that incorporating waste enhances flexibility and biodegradability but diminishes tensile strength and water resistance.

Acknowledgements

I would like to thank Dr. Ervia Yudiati for her invaluable guidance and support throughout this project. I am also grateful to my research assistant, Zalsabila Sulistyana for her technical expertise and administrative assistance. This work was supported by the Research Institute, Diponegoro University under the RPP scheme [Contract numbers 609-09/UN7.D2/PP/VIII/2023]

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