The Characteristics of Bioplastic Made from Sodium Alginate and Kappa Carrageenan

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Abstract

Synthetic plastic is commonly used daily, but its non-biodegradable nature poses a significant environmental threat. Therefore, it is urgent to explore sustainable alternatives such as bioplastics. Alginate, a polysaccharide, is a promising raw material for bioplastics. However, its poor elasticity characteristic necessitates the addition of other polysaccharides, such as carrageenan, to improve bioplastics' mechanical properties. This research aims to determine the characteristics (physical and mechanical properties) of bioplastics made from a mixture of alginate and kappa-carrageenan. The laboratory experimental methods were applied with a completely randomized design. The treatments are mixtures of alginate and k-carrageenan in the following ratios: 10:0 ($A_{10}K_0$), 8:2 (A_8K_2), 6:4 (A_6K_4), 4:6 (A_4K_6), 2:8 (A_2K_8), 0:10 (A_0K_{10}). The characteristics of bioplastic films measured were thickness, water resistance, tensile strength, elongation, and appearance. The optimal bioplastic formulation (4:6 ratio of k-carrageenan to alginate) exhibited a film thickness of 138.3 µm, 16.6% biodegradability, tensile strength of 22.965 N/mm², and 2.73% elongation. The scanning electron microscope showed that the surface structure of bioplastic A_4K_6 made from the ratios of k-carrageenan and alginate of 4:6 had fewer structure cracks and more surface smoothness.

Keywords: *k-carrageenan; alginate; characteristic*

INTRODUCTION

The presence of plastic waste in the sea has an impact that threatens the sustainability of the marine environment, endangering marine biota, ecosystem, health and economic decline in the fisheries sector, tourism, etc. Plastic waste will be fragmented into microplastics that pollute water, land, air, and groundwater environments not only visually but also ecologically for plants, animals, and humans (Amelia *et al*.,2021). One form of anticipation and management of plastic waste is using bioplastics which need to be produced in large quantities and is used widely around the world. Bioplastic is a plastic made from sustainable and renewable raw materials (Lim *et al*., 2021), such as beeswax (having lipids), gelatin, casein, gluten, soy and whey proteins (having proteins), starch, alginate, agar, carrageenan, pectin, cellulose and its derivatives, and chitosan (having polysaccharides) (Bartolo *et al*., 2021).

Hydrocolloid polysaccharides have the advantage of being able to form strong, easily biodegradable and environmental-friendly bioplastics (Khotimah *et al*., 2022). Alginate and carrageenan polysaccharides can be obtained from seaweed, including *Sargassum* sp. and *Kappaphycus* sp. The species of *K. alvarezii* is a marine product currently being developed into a prime fishery product (Prima & Andriyono, 2021). Carrageenan is a linear polysaccharide composed of galactose and 3.6-angidrogalactose units. Carrageenan can be produced by red algae (Rhodophyta). The type of carrageenan produced by *K. alvarezii* is Kappa carrageenan (k-carrageenan). Kappa carrageenan comprises (1,3)-D-galactose-4-sulfate and (1,4)-3,6-anhydro-D-galactose. Carrageenan also contains D-galactose-6-sulfate ester and 3,6-anhydro-D galactose-2-sulfate ester. The presence of a 6-sulfate group can reduce the gelation power of carrageenan, but the addition of alkali can cause the transelimination of the 6-sulfate group, which produces 3,6-

Diterima/Received : 02-01-2025, Disetujui/Accepted : 14-02-2025
DOI: https://doi.org/10.14710/jkt.v28i1.25887

anhydro-D galactose, thus the degree of molecular uniformity increases and its gelation power also increases (Winarno, 1990). In the manufacture of bioplastics, carrageenan gel can form a thin layer (Budiman, 2022), which forms a polymer like conventional plastic but is composed of natural materials, such as starch, starch, chitosan, lignin, rubber, gelatin, protein, pectin, wax, and fatty acids.

Alginate is a natural polysaccharide commonly found in the cell walls of all brown algae species. This natural polysaccharide is used for skin regeneration materials, accelerating wound healing etc. According to Stender *et al.,* (2019), chemically, alginate is a salt of alginic acid consisting of monomers (1-4)-β-D-mannuronic acid (M unit) and a-L-guluronic acid (G unit) which vary in number and distribution along the polymer chain. Alginate is another polymer to produce bioplastics. In general, alginate has a viscosity of 1% by weight in its solution between 10-5000 cP; pH 3.5; water content 5-20%; and particle size 10-200 standard mesh (Hamrun *et al.,* 2018). Alginate has three main properties, namely the ability to dissolve in water, sodium alginate dissolves in water at low ionic strength, resulting in a solution with high viscosity (Xie *et al*.,2024), increase the viscosity of the solution and the ability to form gels alginate can form stable gels in the presence of calcium ions or other divalent cations. (Eslami *et al*., 2023), and the ability to form films These films and coatings have strong potential for use in the production of active food packaging, assisting in food preservation by stabilizing and controlling the release of food preservatives such as antimicrobials and antioxidants (Zabihi.,2024)

While sodium alginate and kappa-carrageenan have been individually studied for bioplastic production, their optimal blend ratios to enhance mechanical and physical properties remain underexplored, such as tensile strength and elongation, thickness, opacity, and water resistance. This can be overcome by the addition of carrageenan in bioplastics that can provide an increase in viscosity because it is polyelectrolyte. Polyelectrolyte is a natural hydrophilic polymer with a linear chain of galactan sulfate that can form a strong polymer matrix and increase the tensile strength between molecules (Rusianto *et al*., 2020). This can cover the weakness of alginate, namely for a low level of resistance to water, due to the large content of hydroxyl groups (- OH) in its molecules (Khotimah *et al.,* 2022). This study aims to determine the optimal composition of sodium alginate and kappa-carrageenan blends for bioplastic production and evaluate their physical, mechanical, and biodegradability properties.

MATERIALS AND METHODS

The materials of the research are k-carrageenan and sodium alginate. Carrageenan powder has a pure white color while sodium alginate is in the form of a fine powder with a brownish white color. Both were purchased on commercial sites (shoppe), and there is no product certification. The research was conducted using the laboratory experimental method (Kerlinger, 1973) with a Completely Randomized Design with various treatments, carrageenan and alginate formulations as in Table 1. Based on previous journal references, the ratios were chosen that the 4:6 and 6:4 ratios had the best test results. Bioplastic characterization (thickness, water resistance, tensile strength and elongation, biodegradation, FTIR and SEM) was analyzed and compared among the treatments.

Table 1. The K-Carrageenan and alginate compositions in the bioplastics producing process

No	Formulation	Comparison	
		Carrageenan	Alginate
1	KK (K-Carrageenan)	10	0
2	A (Alginate)	0	10
3	KKA 2 (K-Carrageenan and Alginate ratio 2:8)	2	8
4	KKA 4 (K-Carrageenan and Alginate ratio 4:6)	4	6
5	KKA 6 (K-Carrageenan and Alginate ratio 6:4)	6	4
6	KKA 8 (K-Carrageenan and Alginate ratio 8:2)	8	2

Thickness test was conducted to determine the physical properties of bioplastics produced and then to compare among composition formulation. The thickness of the bioplastic was measured using a caliper with an accuracy of 0.01 mm at 5 different points (Yupa *et al*., 2021). Water resistance tests on bioplastics are used to measure its strength when it is used as a commercial product and to obtain the best bioplastic composition in physical tests. The water resistance test was carried out based on the method of Farhan & Hani (2017), as follows. Bioplastic was cut into 20 x 20 mm sizes, then soaked in a 50 ml of distilled water in a beaker glass at room temperature for 24 hours. After that, the bioplastic was removed, dried with tissue paper/filter paper, and then weighed. This step was repeated until a constant weight was obtained. Calculation formula as follows (Farhan & Hani (2017):

$$\text{Water resistance} = ((W_1-W_2)/W_1) \times 100\% \quad \ldots\ldots \quad (1)$$

Where, W_1 is the initial sample weight (g) and W_2 is the final sample weight (g)

Tensile strength and elongation tests on bioplastics are used to determine the best physical properties of bioplastic composition. Tensile strength and elongation were measured using the ASTM D882-18 method using a Universal Testing Machine. The bioplastic was cut into 2.5 x 15 cm sizes and pulled at a speed of 200 mm/min with a distance between clamps of 12.5 cm. Tensile strength was determined by dividing the tensile load (F) by the cross-sectional area of the sample (A). Elongation was measured by dividing the length of the sample after the tensile test (L_1) and the initial length (L) using the ASTM D882-18 formula.

$$\text{Tensile Strength } (N/mm^2) = F / A \quad \ldots\ldots \quad (2)$$

Elongation is calculated by dividing the increase in length when the film is torn (L_1) and the initial length of the film before being pulled (L) using the formula:

$$\text{Elongation} = (L_1/L) \times 100\% \quad \ldots\ldots \quad (3)$$

Biodegradation test on bioplastics was carried out using the Soil Burial Test method (Anggraini *et al*., 2013) as follows. Bioplastics were cut into 20 x 50 mm sizes, then put in a desiccator for 24 hours, then weighed (W_1). The samples were then burried in the soil for one week, then the samples were taken and reweighed (Selpiana *et al*., 2015). Biodegradation of the samples was calculated using following formula.

$$\text{Biodegradability } (\%) = (W_1 - W_2)/W_1 \times 100\ \%. \quad \ldots\ldots \quad (4)$$

Where, W_1 is the weight of sample before buried; W2 is the weight of sample after burried

The functional compounds groups in bioplastics were tested using Thermo Nicolet IS 10 FTIR with Deuterated Tri Glycine Sulfate (DTGS) detector. The test process begins by connecting the power cable connected to the power source properly, the computer is turned on by pressing the power button on the PC, click 2 Omnic desktop icons, then select experiment set up and find the menu settings in the Acquisition Parameter which contains a wave number of 4000-650cm-1, resolution 8, number of scans 32 with final transmittance format. The sample test is carried out by reading the air background first by clicking Col Bkg, placing the sample until it closes the Diamond ATR, then pressing the sample using the press knob, reading the sample by clicking Col Smp, waiting a few moments until the sample scanning is complete, then processing the data Where in this stage is baseline correction (yes), PDF, Overlay.

RESULTS AND DISCUSSION

Bioplastics made from only k-carrageenan or sodium alginate tend to be brittle and easily torn.

Bioplastics made of k-carrageenan and Alginate: The mixture with a higher ratio of carrageenan produces a stiffer, smoother, and slippery film, while those with more alginate produce a slightly rougher film. Bioplastics made from only k-carrageenan or sodium alginate tend to be brittle and easily torn. Bioplastics produced with several carrageenan and alginate formulations are presented in Table 2.

Table 2. Appearance of bioplastics produced with several k-carageenan dan alginate formulation

No	Treatment	Ratio KK and A	Bioplastic appearance	Note
1	KK	10:0		Clear, wrinkled at the bottom, stiff and smooth
2	A	0:10		Slightly yellowish and more transparent in the center, stiff at the edges of the sample
3	KKA 2:8	2:8		Yellowish, unwrinkled, and rough
4	KKA 4:6	4:6		Yellowish white, slightly wrinkled at the edges, slightly slippery
5	KKA 6:4	6:4		Slightly yellowish and more transparent at the bottom, wrinkled at the bottom, slightly stiff and slippery
6	KKA 8:2	8:2		Slightly yellowish and more transparent on the bottom, with brown spots, rough on the top, and relatively stiff on the bottom

The thickness of the bioplastic in this experiment was in the range of 56.8-139.5 µm. The highest bioplastic thickness value is produced by K-Carrageenan alginate with the ratio of 2:8 (139.5 µm) and the lowest made of alginate (56.8 µm) (Figure 1). All values of bioplastic thickness met the quality standards of the Japanese Industrial Standard (JIS), which are 250 µm - 25 mm.

In the water resistance test, all bioplastic produced was dissolved in the water, Except for the treatment of KA 4:6 (K-Carrageenan Alginate 4:6), it was seen to have been partially solved. Single biopolymer materials such as alginate and carrageenan need to be combined to obtain stronger and more water-resistant hydrogels, a smaller alginate composition affects water resistance due to the nature of alginate itself, which quickly absorbs water. Water resistance increases with the addition of carrageenan because carrageenan can bind water. There are several factors affect the solubility of the bioplastic, including the thickness of the bioplastic, the presence or absence of plasticizers, in which the more plasticizers, the greater the solubility, especially if the plasticizer is hydrophilic, in addition the increasing concentration of carrageenan cause higher percentage of water resistance of bioplastic (Inayah and Kusumayanti., 2022).

The highest tensile strength of bioplastic produced in this experiment has all alginate composition (Treatment A), i.e., 45.069 N/mm2, and the lowest in the carrageenan and alginate composition of 8:2 (Figure 2). The bioplastic that was produced from a higher alginate composition than k-carrageenan showed a higher tensile strength. This is in line with Pascalau *et al.* (2012), who state that alginate has a higher tensile strength compared to carrageenan, and carrageenan itself is the component that lowers the tensile strength properties. This is caused by the increase in hydrogen bonds formed in it, which will cause the chemical bonds to become tighter and stronger, so that it requires higher energy to make bioplastic stretch (Coniwati *et al*., 2014). of carrageenan alone or carrageenan that is greater than alginate makes bioplastics that are stiff and brittle.

There is the highest elongation value found in the composition of alginate bioplastic of 11.344. In the tensile strength and elongation test, the K-Carrageenan composition could not be tested because the sample was damaged during preparation because it was very dry and brittle. In comparison, other samples showed that increasing carrageenan concentrations made the elongation

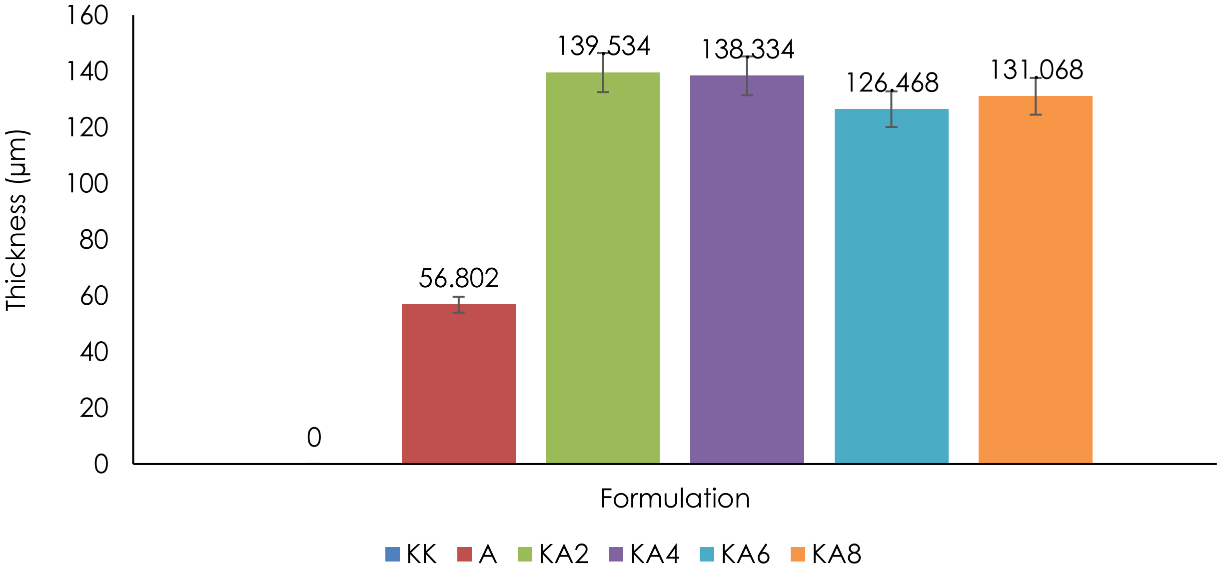


Figure 1. The thickness of bioplastic (µm) made of several k-carrageenan and alginate formulation
Note : KK = K-Carrageenan; A = alginate; KA 2:8 = K-Carrageenan Alginate 2:8; KA 4:6 = K-Carrageenan Alginate 4:6; KA 6:4 = K-Carrageenan Alginate 6:4; KA 8:2 = K-Carrageenan Alginate 8:2

smaller as seen in Figure 3. This can be caused by the nature of carrageenan itself which is stiff and brittle. In addition, the use of higher alginate concentrations makes elongation with better values. Alginate is the most beneficial component for elongation and tensile strength (Pascalau *et al*, 2012). Increasing the concentration of carrageenan increases dissolved solids in bioplastics, thus forming strong polymer chains and reducing molecular movement which produces stiff bioplastics. The addition of plasticizers can increase elongation and make bioplastics more flexible, but reduce tensile strength (Khalil *et al*., 2017), while in this test no plasticizers were used at all.

The biodegradation test on bioplastics of this experiment showed that the highest value was found in K-Carrageenan (KK) samples, i.e., 49.6%, and the lowest in alginate (A) and carrageenan alginate 8:2 (KA 8:2) samples, i.e., 15.5% (Figure 4). carrageenan itself is the most perishable component. This was caused by the fragile nature of carrageenan itself, making it easier to degrade by soil. The hydrophilic nature of carrageenan makes the sample take longer to degrade, while alginate has elastic properties so it takes longer to degrade in the environment. Test materials with higher tensile strength value make the biodegradation process slower (Astuti *et al.*, 2019).

Table 3. Wave Number of FTIR of bioplastics made of several k-carrageenan and alginate formulation

Absorption Peak	Wave Number					
	KK	A	KKA 2:8	KKA 4:6	KKA 6:4	KKA 8:2
-OH	3281.18	3247.08	3262.92	3302.10	3256.76	3295.28
Alkane (CH)	2991.91	2919.04	2918.30	2923.94	2920.77	2918.14
C=C	1643.13	1591.92	1594.39	1596.16	1596.18	1602.30
Organic sulfates	1411.03	1407.13	1407.77	1408.39	1407.61	1410.13
COO-	1015.20	1025.27	1023.45	1018.87	1019.31	1015.40

Note: KK = K-Carrageenan; A = alginate; KA 2:8 = K-Carrageenan Alginate 2:8; KA 4:6 = K-Carrageenan Alginate 4:6; KA 6:4 = K-Carrageenan Alginate 6:4; KA 8:2 = K-Carrageenan Alginate 8:2

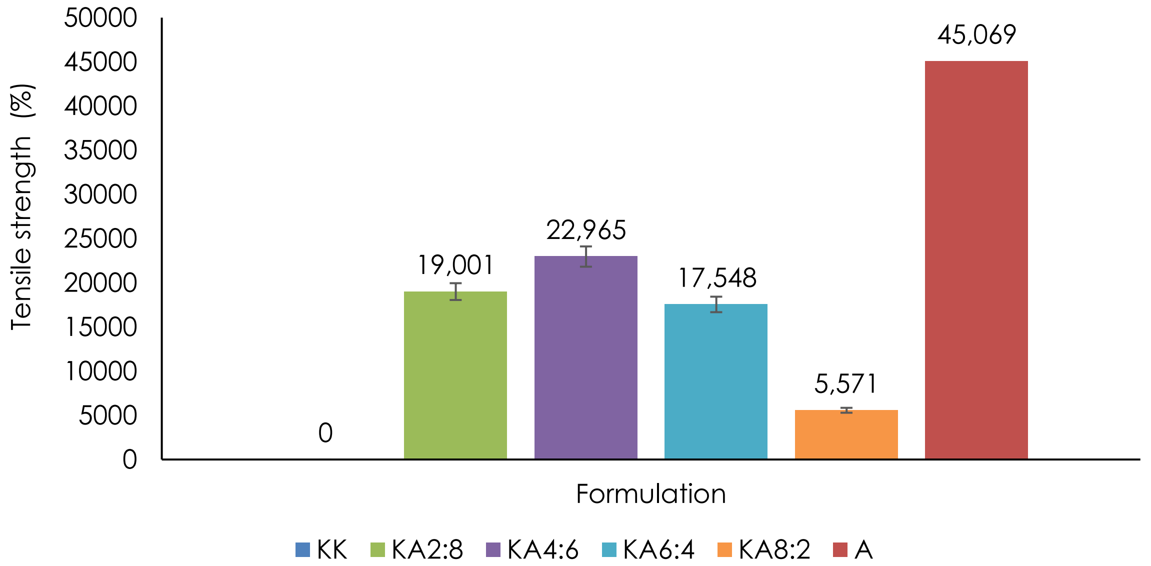


Figure 2. Tensile Strength Test (N/mm^2) made of several k-carrageenan and alginate formulation
Note : KK = K-Carrageenan; A = alginate; KA 2:8 = K-Carrageenan Alginate 2:8; KA 4:6 = K-Carrageenan Alginate 4:6; KA 6:4 = K-Carrageenan Alginate 6:4; KA 8:2 = K-Carrageenan Alginate 8:2.

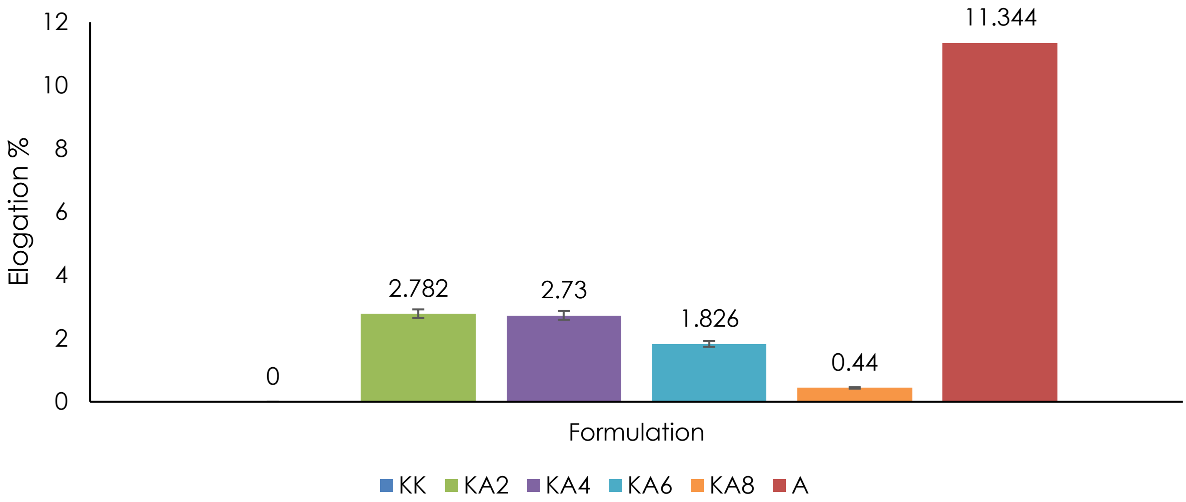


Figure 3. Elongation Test (%) on bioplastics made of several k-carrageenan and alginate formulation Note: KK = K-Carrageenan; A = alginate; KA 2:8 = K-Carrageenan Alginate 2:8; KA 4:6 = K-Carrageenan Alginate 4:6; KA 6:4 = K-Carrageenan Alginate 6:4; KA 8:2 = K-Carrageenan Alginate 8:2

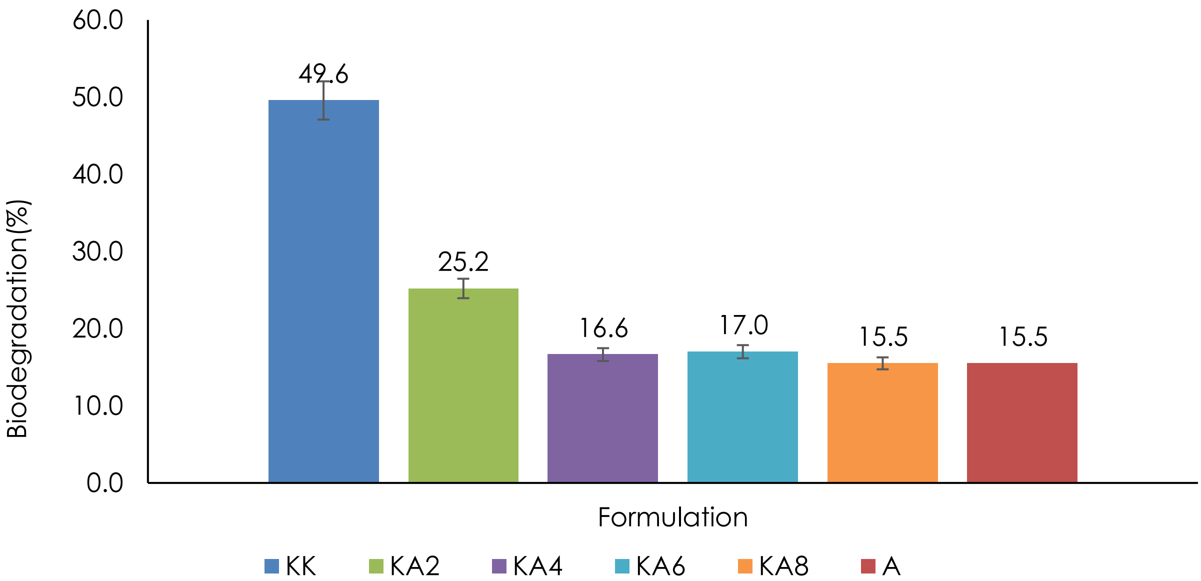
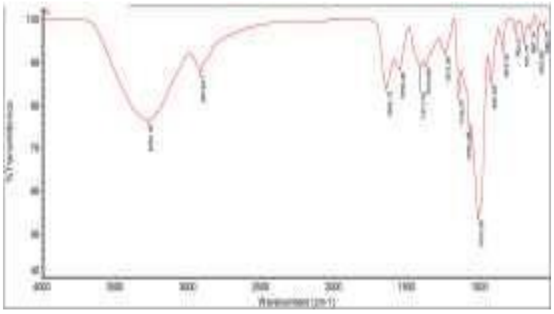
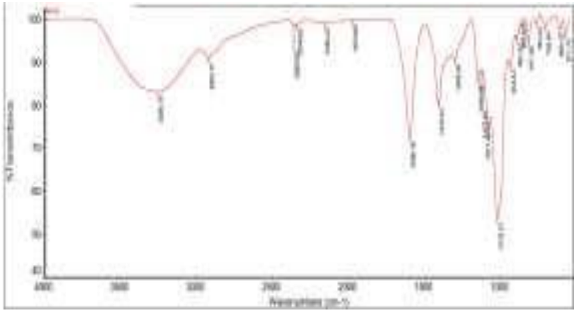


Figure 4. Biodegradation test (%) on bioplastics made of several k-carrageenan and alginate formulation Note: KK = K-Carrageenan; A = alginate; KA 2:8 = K-Carrageenan Alginate 2:8; KA 4:6 = K-Carrageenan Alginate 4:6; KA 6:4 = K-Carrageenan Alginate 6:4; KA 8:2 = K-Carrageenan Alginate 8:2.

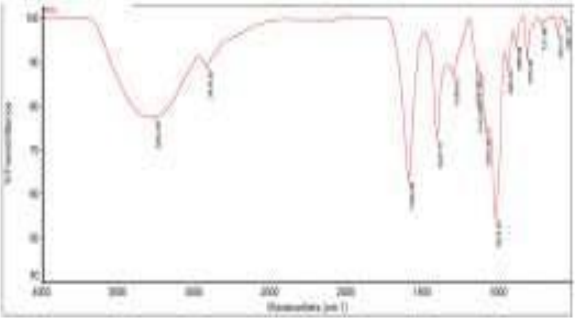
The results of FTIR analysis are shown in Table 3, in which the functional groups and their measured frequencies are referred to by Coates (2000). The absorption peak of wave numbers 3302-3247 cm^{-1} is ammonium ion usually the first absorption is intense and broad which the second absorption has weak to moderate intensity and narrow, both are often found in double band structures and can be used to characterize individual compounds. Wave numbers 2991-2918cm^{-1} are alkane CH, wave numbers 1643-1591cm^{-1} are C = CC aromatic ring stretch, wave numbers 1411-1407cm^{-1} are organic sulfates which are asymmetric/symmetric XO_2 stretch (NO_2 and SO_2) and wave numbers 1025-1015 cm^{-1} are cyclohexan ring vibration.



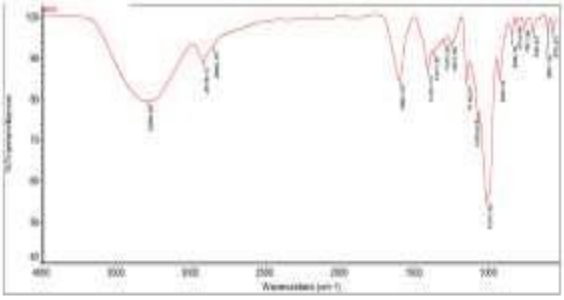
Kappa Carrageenan



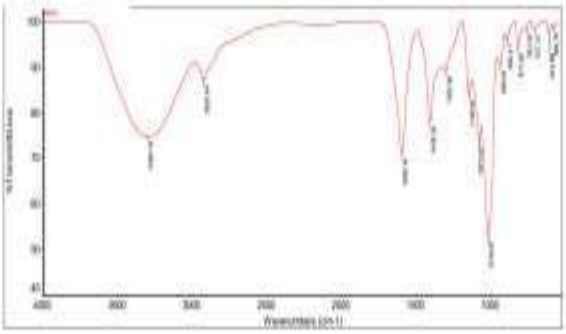
Kappa Carrageenan and Sodium Alginate 6:4



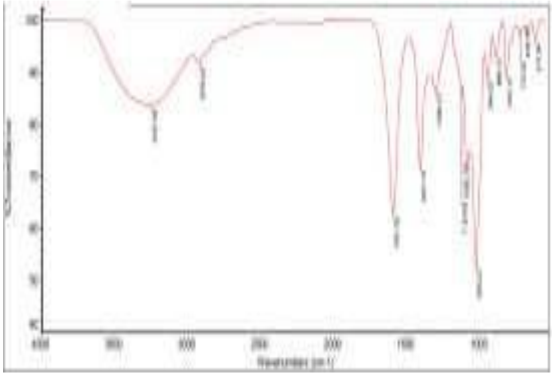
Kappa Carrageenan and Sodium Alginate 2:8



Kappa Carrageenan and Sodium Alginate 8:2



Kappa Carrageenan and Sodium Alginate 4:6



Sodium Alginate

Figure 5. FTIR results (cm^{-1}) produced from various bioplastic formulations

CONCLUSION

Bioplastics made of several k-carrageenan and alginate formulation produce thin, brittle, and stiff sheets. The best bioplastic sample is a mixture of K-Carrageenan and alginate of 4:6 that has a thickness value of 138.3 µm, partly dissolved in the water resistance, tensile strength of 22.965 N/mm^2, elongation of 2.73%, biodegradation of 16.6%. The SEM result shows the surface of the bioplastic is best in the composition of k-carrageenan and alginate 6:4 bioplastic. Further research is needed the addition of plasticizers to commercial samples. SEM test should be conducted for several test samples, not only the carrageenan-alginate 4:6 and 6:4 samples. Further research can be conducted by adding microbial test, water vapor transmission rate, solids transmission and opacity to test the feasibility of biodegradation standard.

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