A Review on the difference of physical and mechanical properties of bioplastic from seaweed hydrocolloids with various plasticizers

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Abstract. Seaweed hydrocolloids such as alginate and carrageenan extracted from brown and red algae have the potential to be developed as bioplastic. Bioplastic is a polymer from natural materials that are biodegradable. Alginate and carrageenan-based bioplastics are brittle and inelastic. Plasticizers such as glycerol, sorbitol and polyethylene glycol can be added to formulate plastic to improve its properties. The aim of the research was to review the physico-chemical properties of the bioplastic made from seaweed hydrocolloids with various additions of plasticizers. The data used in this study were taken from several accredited national journals and international publications. The data were presented quantitatively and synthesized narratively to answer the hypothesis of the study. The results showed that the addition of plasticizer in bioplastic preparation affected the physico-chemical properties of bioplastics. The thickness and elongation at break of bioplastic increased after the addition of plasticizers. However, the tensile strength of the bioplastic has decreased after added with plasticizer.

Keywords: alginate; carrageenan; hydrocolloids; plasticizers

1. Introduction

Plastic waste pollution has become an environmental issue that has been widely discussed in the last decade. Plastic waste is a chemical that is difficult to decompose and takes more than 100 years to decompose [1]. Plastic waste that is difficult to decompose can cause damage to environmental ecosystems both on land and in waters. Even the impact of this environmental problem is contained in the 17 goals of the Sustainable Development Goals (SDGs) at points 14 and 15 regarding underwater life and terrestrial life. Data from the Ministry of Environment and Forestry states that the total waste heap in Indonesia reached 68 million tons in 2019, with 9.52 million tons of plastic waste [2]. Some of the garbage will be carried into the sea which will later cause problems for the marine ecosystem. Indonesia is also the second largest producer of plastic waste to the sea in the world after China with a total plastic waste of 1.29 million tons per year [3]. The main problem is the large amount of plastic waste produced by countries in the world because the need for plastic materials is still needed. Since 1930-1940 plastic has been mass-produced and became a necessity in various human daily needs at that time. However, the problems caused by plastic can disrupt the stability and ecosystem of an environment [4]. Bioplastics can be a solution to reduce the amount of plastic pollution that is difficult to decompose. The use of natural materials such as seaweed can be a biomaterial in the manufacture of bioplastics [4].

Seaweed is an aquatic resource that has the potential to be developed into alternative biomaterials. Indonesia's total wet seaweed production in 2017 reached 10,456 million tons, or 40% of the world's
total seaweed production. The most widely grown and cultivated species in Indonesia is *Kappaphycus alvarezii* [5]. This large amount of production needs to be developed to get a more valuable and useful product. Hydrocolloid in seaweed is a high value derivative. Hydrocolloid which has thickening properties and forms a gel in solution [6]. One of the hydrocolloids derived from seaweed is carrageenan and alginate.

Indonesia's carrageenan production was recorded by the Ministry of Marine and Fisheries reaching 13116 tons in 2017, with the utilization of approximately 50% [5]. On the other hand, the national demand for alginate still has to be met by importing it. The total market for Indonesian alginate imports from 2013-2016 was 1284.51 tons, 1268.62 tons and 1199.76 tons, respectively [7]. The data above identifies that Indonesia has quite abundant carrageenan and alginate products, and can be developed into a product.

Carrageenan is a polysaccharide extracted from seaweed of the Rhodophyceae family or red algae such as Eucheuma spinosum and *K. alvarezii*. Carrageenan is soluble in water, where for this type of kappa-carrageenan it dissolves in water at a temperature of 70°C [8]. Carrageenan will form a gel at low temperatures and will return to liquid at high temperatures. Carrageenan is a linear anionic polysaccharide consisting of galactose and anhydrogalactose. The benefits of carrageenan in the food and non-food fields are quite diverse, one of the non-food fields that can utilize carrageenan is in the manufacture of biomaterials. These biomaterials such as bioplastics or edible coatings are environmentally friendly [9]. The hydrocolloid nature of carrageenan can be a good film because it provides good mechanical properties for oxygen and carbon dioxide barriers so that it can counteract oxidation activity [10].

Alginate is one of the polysaccharides in the form of hydrocolloids derived from brown seaweed. Alginites found in brown algae are found in their cell walls [11]. Alginites found in brown seaweed in the industry can be used as thickeners, emulsifiers, stabilizers, suspension materials, and as film formers [12]. The nature of alginate is viscous and easily soluble in water, making alginate has the potential to be developed as a bioplastic making material. Brown seaweed *Sargassum* sp. which is one of the producers of alginate has thermoplastic properties that are easy to be printed into packaging films or bioplastics. The nature of thermoplastics is that they can change shape if given a temperature treatment, which will become a film sheet when cooled and will become a liquid when heated [13].

Bioplastics are plastics made from natural materials such as starch and are easily biodegradable or soluble in water. So far, bioplastics have been developed using raw materials from cassava. However, cassava is a food ingredient and Indonesia still imports cassava, so other alternative raw materials are needed. An alternative that can be used as raw material for making bioplastics is seaweed. The hydrocolloid content of seaweed which is capable of producing carrageenan and producing a film layer can be utilized for the manufacture of bioplastics [14]. However bioplastics derived from seaweed hydrocolloids have weaknesses, including being brittle and stiff [15]. The solution given to overcome the weakness of the brittle and rigid nature of plastic is to add material in the form of a plasticizer [16].

Plasticizer is a material that can be added to the raw material for making bioplastics, which can affect the elasticity and flexibility of bioplastics [17]. Glycerol is a plasticizer that can affect the characteristics of bioplastics [18]. Glycerol is the main component of fats and oils which are the simplest compounds [19]. In addition to glycerol, sorbitol is one of the ingredients that can be used as a plasticizer. In general, sorbitol is a natural sugar alcohol that can be used in various applications, such as sweeteners and emulsion stabilizers [20]. Sorbitol can also be used as a plasticizer because it can increase the flexibility and tensile strength of bioplastics. The nature of sorbitol is also non-toxic, so it remains an environmentally friendly product if it is used as an additional material for making bioplastics [21]. Determining the proper treatment or concentration ratio is also one of the problems that are often encountered during experimental research. In addition, the right type of plasticizer used as a plasticizer is a particular choice and can be a consideration regarding what type of plasticizer will be used. The type of plasticizer and the concentration used in the addition of making plastic will
directly affect the physical and mechanical properties obtained. Therefore, this study will show the results of the physical and mechanical properties of bioplastics from various plasticizers used.

The physical and mechanical properties of bioplastics can be used as an analysis to determine the quality of plastics. The physical and mechanical properties include thickness, tensile strength, and elongation or elongation [22]. Furthermore, the type of main constituent of bioplastics and additives such as plasticizers will directly affect the physical and mechanical properties [23]. Therefore, this study wanted to determine the effect of the types of seaweed hydrocolloids and plasticizers on the physical and mechanical properties of bioplastics.

2. Method

2.1. Time and place
The research was conducted from April to August 2020. The research location was in Bogor City, West Java, Indonesia.

2.2. Materials and tools
The material used in this research is a collection of published literature studies. Data and information are obtained from the internet, books, standards, national and international journals. The tool used to find secondary data is a laptop. Data analysis was carried out using Microsoft Excel 2013 and IBM SPSS version 25.

2.3 Research procedures
The research procedure used in this study is the Systematic Review according to [24] with the following steps:
1. Identification research questions (meta-analytical research questions)
2. Develop a meta-analysis research protocol
3. Setting the location of the research results database as the search area
4. Selection of relevant research results
5. Choose quality research results
6. Extraction of data from individual studies
7. Synthesis of research results using the meta-analysis method
8. Presentation of research results in the meta-analysis research report

The problem formulation questions used to collect the information needed in analyzing the results of different studies are as follows:
1. What are the physical and mechanical properties of bioplastic hydrocolloids with various plasticizer from various research sources?
2. What kinds of type hydrocolloids are used in the research?
3. What kinds of type plasticizer are used in the research?

The research protocol carried out was a literature review by collecting various reputable libraries. The data and information in this study were obtained through secondary data contained in national and international journals via the internet whose sources of information can be accounted for. The selected library sources must have the following criteria:
1. Library sources are scientific writings such as accredited national or international journals or books that have an ISBN number (International Standard Book Number)
2. The source of the literature focuses on research on the manufacture of bioplastics from seaweed hydrocolloids and plasticizers
3. The literature sources used focus on the discussion of thickness, tensile strength, and bioplastic elongation

Literature review begins with downloading literature on the manufacture of seaweed hydrocolloid bioplastics that will be used as data. Downloads of literature obtained regarding the manufacture of bioplastics based on seaweed hydrocolloids are obtained from sites, such as Google Scholar and Sciedirect. Additional information needed is obtained from data from government institutions,
including the Ministry of Maritime Affairs and Fisheries Indonesia and the Ministry of Environment Indonesia and Forestry as well as the Japanese Industrial Standard. The initial literature sources collected amounted to 30 libraries. The source is then selected based on the parameter data to be analyzed, namely thickness, tensile strength and elongation of the bioplastic. The data to be analyzed are the types of materials and plasticizers, namely alginate and carrageenan as well as glycerol, sorbitol and polyethylene glycol (PEG). The concentration of plasticizer used in this study and also used as a condition for the data used is a plasticizer with a control concentration (0%) and the addition of a plasticizer concentration of 20% of the control. These data are used to draw conclusions about the effect of adding plasticizer concentration to the bioplastic parameters to be analyzed.

The method of presenting the data used in this research is done quantitatively. The data set that will be used is selected according to the criteria mentioned above. The data were then analyzed in terms of the similarity of the manufacturing process and the concentration of the materials used. The data that has been selected is then analyzed quantitatively by using the Analysis of Diversity Prints (ANOVA one way). The data analyzed was the percentage of each change in the physical and mechanical properties of bioplastics after the addition of each plasticizer was 20% of the control treatment. The results obtained are then analyzed in a narrative manner by testing the correctness of the results from other researches. The hypothesis that wants to be proven in this study is "the application of plasticizer to seaweed hydrocolloid bioplastics can affect changes in thickness, tensile strength, elongation of the resulting bioplastic".

2.4 Data analysis
This research perform data analysis using Analysis of variance (ANOVA). Data were analyzed by Duncan's further test using Statistical Package for Social Science (SPSS) software.

3. Results and discussion
3.1. Physical and mechanical properties bioplastic with plasticizers
Bioplastics are polymers derived from natural materials and are easily degraded naturally [14]. Natural polymers that can be used as raw materials for the manufacture of bioplastics are polysaccharides, proteins, and lipids [25]. Raw materials for making bioplastics must have biodegradable properties, come from renewable materials and can be printed into film sheets after being dried or allowed to stand [26]. The manufacture of bioplastics has the same principle regardless of the difference in the raw materials used. According to [27] the process of making bioplastic begins with measuring or weighing the formulation of the material to be used. Furthermore, the formulation is dissolving, mixing, stirring, and heating the ingredients in one place. The next process is printing / dissolving the bioplastic solution and drying it until the plastic film is dry and forms a sheet. An important parameter that must be met by bioplastics in order to become good plastics is that the tensile strength of the plastic must be above 0.39 MPa [28] This are because the resistance and strength of the plastic produced is known through the parameters of tensile strength.

Hydrocolloid is one of the polymers that can be produced naturally. Hydrocolloid is a polymer component that has the ability to bind and absorb water and is soluble in water. This ability makes hydrocolloids able to thicken or form a gel in solution [6]. Seaweed is one of the plants that can produce hydrocolloids. One type of hydrocolloid derived from seaweed is alginate and carrageenan [29]. Alginate and carrageenan hydrocolloids can be used as a source of raw materials for making bioplastics, this is because hydrocolloids can form gels that can form film sheets when dried [30].

Carrageenan is a polysaccharide consisting of sulfated polyglycan chains. One of the characteristics of carrageenan that affect the formation of films or bioplastics is solubility and gelation. The process of dissolving a good carrageenan will produce bioplastics that are evenly distributed on the film sheet. While the gelation process will produce cross-linked polymer chains that form three dimensions, where this bond will immobilize water so that it will form a strong and rigid structure [36].
The development of carrageenan-based bioplastics is still in its early stages, when compared to plastics made from other biopolymers. Therefore, the only method of manufacture reported is by pouring or casting the solvent with carrageenan and then drying it to evaporate the water to leave the bioplastic solids. Carrageenan bioplastics containing calcium salts produce clear, elastic and soft gels, while other carrageenans will produce strong, rigid, and brittle films [37,4]. Similarly, the bioplastic properties or films produced from carrageenan exhibit a compact film [38] and high gelling ability [39] which enhance the mechanical properties and water barrier properties [40].

Alginate and carrageenan can be used as raw materials for making bioplastics. Alginate as a raw material for bioplastics is easily soluble, brittle, and has poor mechanical properties [41]. The poor mechanical properties in question are easy to break and inelastic or stiff [15]. Similar to alginates, carrageenans have water-soluble properties, low intermolecular distances, and low polymer chain mobility [16]. The physical and mechanical properties of alginate and carrageenan bioplastics need to be added with a plasticizer to improve these physical and mechanical properties. The addition of a plasticizer functions mechanically in reducing the intermolecular interactions of the polymer and is able to increase the mobility of the polymer, so that the bioplastic becomes more elastic [43]. Table 1 presents qualitative results from various literature on the effect of plasticizer application on the physical and mechanical properties of bioplastics. The provision of plasticizers presented in table 1 does not detail the types of plasticizers used, where the data presented are only to provide information that plasticizers can affect changes in thickness, tensile strength, and elongation of bioplastics. For quantitative and more detailed data results will be discussed in the next sub chapter.

The application of plasticizers can affect changes in the thickness of the bioplastic, as shown in table 1. The results obtained from [31,16] reported that the application of plasticizers will increase the thickness of the bioplastic. The increase in thickness occurs because the plasticizer added to the bioplastic material has an increase in the composition of the material, so the thickness will also increase. The addition and high concentration of materials used in the manufacture of bioplastics will increase the density and thickness of bioplastics [23]. Mixing materials for making bioplastics by the research of [31,16] used the same method, namely by dispersing with distilled water and then stirred and heated. Data from the research of [34] stated that the application of plasticizers increased the thickness of bioplastics. The plasticizer used in this study used two different types, namely polyvinyl alcohol and glycerol. The addition of these two types of plasticizers will increase the resulting bioplastic solids. Similar to previous studies, this study also uses aquadest to mix all types of materials used for the manufacture of bioplastics. In addition to the addition and amount of plasticizer concentration, external factors such as the shape of the mold used will affect the thickness of the bioplastic produced [44].

\[ \text{Table 1. Physical and mechanical properties of bioplastics with or without plasticizer.} \]

| Bioplastic          | Plasticizer added | Thickness (µm) | Tensile Strength (MPa) | Elongation at break (%) | Source |
|---------------------|-------------------|----------------|------------------------|------------------------|--------|
| Ca-alginate + Carrageenan | No                | 18             | 39.4                   | 13.0                   | [31]   |
|                     | Yes               | 21             | 29.7                   | 24.9                   |        |
| Kappa-carrageenan   | No                | 31.53          | 69.69                  | 3.1                    | [16]   |
|                     | Yes               | 36.70          | 58.27                  | 4.7                    |        |
| Kappa-carrageenan   | No                | -              | 24.85                  | 4.2                    | [32]   |
|                     | Yes               | -              | 2.28                   | 18.24                  |        |
| Alginate            | No                | -              | 59.9                   | 10.3                   | [33]   |
|                     | Yes               | -              | 15.6                   | 29.1                   |        |
| Sodium-alginate     | No                | 41.2           | -                      | -                      | [34]   |
|                     | Yes               | 55.2           | -                      | -                      |        |
| Semi refined        | No                | -              | 46.63                  | 6.54                   | [35]   |
| carrageenan         | Yes               | -              | 58.20                  | 14.5                   |        |
The tensile strength properties of bioplastics also change when plasticizers are added, as presented in table 1. Data from [31] shows a decrease in tensile strength after adding plasticizers. The results of research by [16,32,33] showed a decrease in the tensile strength produced after being given plasticizer treatment. The results from the four data sources both showed a decrease in the tensile strength of bioplastics after the addition of plasticizers. The decrease in the tensile strength of bioplastics after the addition of plasticizers is due to the fact that plasticizers can reduce intermolecular bonds. The decrease in intermolecular bonds occurs because the plasticizer can increase the bonding space, so that it can facilitate the migration of water vapor which results in the bond and the tensile strength of the plastic getting weaker [45]. Data obtained from [35] showed an increase in the tensile strength of semi-refined carrageenan (SRC) bioplastic after adding a plasticizer. The increase in tensile strength in this study shows the difference in results from the four previous journals which got the results of a decrease in tensile strength after adding a plasticizer. Increased tensile strength of bioplastics in the study of [35] can be caused by the addition of glycerol plasticizer using only 0.9% of the total solution material for making bioplastics. According to [37], the addition of small amounts of glycerol to SRC will increase the tensile strength of bioplastics. This can be attributed to the strong interaction between the glycerol polymer and the SRC polymer in hydrogen bonds. However, the amount of glycerol that is too high will still reduce the tensile strength of bioplastics, because glycerol can facilitate the migration of water vapor molecules resulting in the distance between bonds [43].

Other mechanical properties of bioplastics are presented in table 1, namely elongation at break or elongation at break. Elongation at break is the extension value of a material distance until it is broken or broken [23]. Research from [31,16,32,33,34,35] showed an increase in elongation after adding a plasticizer. This result is also in accordance with the statement of [43], that the application of plasticizers can increase the elongation of bioplastics. Plasticizer molecules will reduce intermolecular interactions and increase polymer mobility and will increase the percent elongation of the resulting bioplastic. This is because the increased mobility of the polymer will facilitate the migration of water vapor molecules [43]. The results obtained by [47] show that the application of sorbitol and glycerol as plasticizers to bioplastics has a saturation point limit, where the concentration of more than 60% is the saturation point and will not increase elongation and decrease elasticity. This is because the addition of a high concentration of plasticizer will reduce the cohesive forces between molecules, so that the resulting plastic polymer will become softer and inelastic [48].

3.2 Bioplastic thickness
The thickness of seaweed hydrocolloid bioplastic can be influenced by the type of hydrocolloid, plasticizer, and the concentration of the material used [11]. In addition, the area of the bioplastic mold used can also affect the thickness of the bioplastic [49]. According to the Japanese Industrial Standard 1975, the maximum thickness limit of bioplastics is below 250 m. Bioplastic thickness can be measured using a digital micrometer [16] and a manual screw micrometer [50].

The results of the previous bioplastic thickness data were designed completely randomly. Each data from seaweed hydrocolloid bioplastic with each plasticizer is the respective treatment used. The data in the table 2 is presented quantitatively by means of fingerprint analysis (one way ANOVA), namely by comparing the percentage change in thickness with each difference in plasticizer. The results of the average thickness change were obtained after getting the percentage change from the control treatment to the addition of 20% plasticizer treatment for each data source. These results were then used as replicates for each of the same plasticizers, then compared with the ANOVA test. The results were obtained from the ANOVA test. The results presented in table 2 are used as conclusions from the tested hypotheses.

The results shown in table 2 show changes in the thickness of carrageenan bioplastics treated with different plasticizers. Data were obtained from various literature sources listed in the table, then analyzed by one-way ANOVA to obtain the average change and the effect of each treatment. Some of the plasticizers listed include polyethylene glycol (PEG), glycerol and sorbitol. Carrageenan bioplastics with the addition of 20% PEG experienced an average increase of 13.1%. Meanwhile, the
The largest average thickness increase was found in the treatment of bioplastics added with sorbitol and 20% glycerol with 15.53% and 18.05% respectively. Although numerically there are differences in changes in the thickness of bioplastics from each treatment, the script letters contained in the data show that the differences in plasticizers have no significant effect on the resulting thickness changes (\(p>0.05\)). Theoretically, increasing the concentration and the number of types of materials used in the manufacture of bioplastics will further increase the solids contained, resulting in increased thickness [23].

The addition of 20% PEG as a plasticizer in the study of [16,51] in table 2 show an increase in thickness. According to [16] the increase in thickness that occurs when plasticizer is added is due to an increase in the volume of materials used in the manufacture of bioplastics. According to [45], PEG which is used as a plasticizer has hygroscopic properties or absorbs moisture. This causes the plastic polymer to contain more water, causing an increase in thickness.

The addition of glycerol plasticizer to carrageenan bioplastics listed in table 2 gives a change in thickness of 18.0.5%. Glycerol which is polar will dissolve with carrageenan which is also polar [36]. Glycerol has the property of absorbing higher water content, so it will cause an increase in thickness [16]. In line with the statement of the previous sentence, the more water that is bound, the more thickness will be produced [52]. The addition of sorbitol to carrageenan plastic will spread throughout the polymer matrix, so that the thickness of the plastic will increase [16,53].

Based on theory, the thickness of the bioplastic is influenced by the constituent polymer. The concentration of the constituent polymers will affect the total solids and polymers that make up the matrix of bioplastics [13]. The area of the mold and the type of additive used also affect the thickness of the bioplastic [44]. Plasticizer added to bioplastic can increase the viscosity of the solution so that the dissolved solids will increase. The increased solids of the bioplastic solution cause the film-forming polymer to increase, so that the thickness of the bioplastic will increase [45]. According to the results of research by [54], the type and concentration of plasticizers also significantly affect the thickness of the bioplastic. The thickness values obtained in this study ranged from 170 \(\mu\)m to 260 \(\mu\)m.

### 3.3 Bioplastic tensile strength

The mechanical properties of bioplastics in the form of tensile strength are an analysis that can determine the quality of a bioplastic. Tensile strength is the force required for plastic to stretch and is usually measured in Mega Pascal (MPa) units [11]. Plastic tensile strength can be measured using the CMT-10 Computer Control Electronic Universal Testing Machine [31]. The Japanese Industrial

| Bioplastic Plasticizer | Thickness \(\mu\)m (control treatment) | Thickness \(\mu\)m (after added 20% Plasticizer) | Δ Thickness (%) | Average Thickness change (%) |
|------------------------|--------------------------------------|-----------------------------------------------|-----------------|-----------------------------|
| Carrageenan PEG        | 31.53\(^1\)                          | 34.17\(^1\)                                   | 8.4             | 13.1±6.65a                  |
| Carrageenan PEG        | 97.6\(^2\)                           | 115\(^2\)                                     | 17.8            |                             |
| Carrageenan Sorbitol   | 31.53\(^1\)                          | 36.5\(^1\)                                   | 15.8            | 15.53±0.39a                 |
| Carrageenan Sorbitol   | 101\(^2\)                            | 116.4\(^2\)                                  | 15.25           |                             |
| Carrageenan Glycerol   | 98.2\(^2\)                           | 115.8\(^2\)                                  | 17.9            | 18.05±0.21a                 |
| Carrageenan Glycerol   | 31.53\(^1\)                          | 37.27\(^1\)                                  | 18.2            |                             |

\(^1\)[16] \(^2\)[51]

- sign in the table indicates a decrease changes

a, b, ab Different script letters signify a significant difference (\(p<0.05\)) for each treatment

\(\Delta\) Thickness Japanese Industrial Standard 250 \(\mu\)m>
Standard 1975 stipulates that the minimum standard of tensile strength of bioplastics is above 0.39 MPa. The tensile strength of bioplastics can be determined based on the type of constituent material [5]. Bioplastics can be made from seaweed hydrocolloids, such as alginate and carrageenan. Plasticizers can be added to the building blocks of bioplastics to make the plastic produced more elastic and less rigid [18]. The type and concentration of plasticizer treatment will result in changes in the tensile strength of bioplastics [16]. This is in accordance with the data presented in table 3, where the literature states that the application of plasticizers can change the tensile strength of bioplastics.

The results of the data The tensile strength of bioplastics was previously carried out in a completely randomized design. Each data from seaweed hydrocolloid bioplastic with each plasticizer is the respective treatment used. The data in the table is presented quantitatively by means of fingerprint analysis (one way ANOVA), namely by comparing the percentage change in tensile strength with each difference in plasticizer. The average results of changes in tensile strength were obtained after getting the percentage difference from the control treatment to the treatment of adding 20% plasticizer to each data source. These results were then used as replicates for each of the same plasticizers, then compared with the ANOVA test. The results obtained from the ANOVA test. The results presented in table 3 are used as conclusions from the hypotheses of this study.

Table 3. Average changes in tensile strength of bioplastics.

| Bioplastics | Plasticizer | Tensile Strength (MPa) (control treatment) | Tensile Strength (MPa) (after added plasticizer 20%) | Δ Tensile Strength | Average Tensile Strength Change (%) |
|-------------|-------------|--------------------------------------------|-----------------------------------------------|-------------------|-------------------------------------|
| Carrageenan | PEG         | 69.69<sup>1</sup>                          | 46.3<sup>1</sup>                               | -33.56            | -34.1±0.76b                         |
| Carrageenan | PEG         | 26.35<sup>2</sup>                          | 17.96<sup>2</sup>                              | -34.64            |                                    |
| Carrageenan | Glycerol    | 24.22<sup>2</sup>                          | 12.8<sup>2</sup>                               | -47.15            | -38.76±11.87b                        |
| Carrageenan | Glycerol    | 69.69<sup>1</sup>                          | 48.53<sup>1</sup>                              | -30.36            |                                    |
| Alginate    | Glycerol    | 38.5<sup>3</sup>                           | 8<sup>3</sup>                                  | -72.22            | -69.77±3.47a                         |
| Alginate    | Glycerol    | 52<sup>4</sup>                             | 17<sup>4</sup>                                 | -67.31            |                                    |
| Alginate    | Sorbitol    | 52<sup>4</sup>                             | 31<sup>4</sup>                                 | -40.4             |                                    |
| Alginate    | Sorbitol    | 59.9<sup>5</sup>                           | 23.2<sup>5</sup>                               | -61.27            | 50.84±14.76ab                        |

0.39 MPa is the tensile strength Japanese Industrial Standard. <sup>1</sup>[16] <sup>2</sup>[51] <sup>3</sup>[42] <sup>4</sup>[15] <sup>5</sup>[33]

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a, b, ab Different script letters signify a significant difference (p<0.05) for each treatment.

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Table 3 shows a change in the decrease in tensile strength after the plasticizer is applied. The results shown in table 3 show that the average change in each treatment produces a different script letter, which shows that different plasticizer treatments have a significant effect on changes in tensile strength (p<0.05). From all sources of carrageenan and alginate bioplastics, the addition of plasticizer treatment decreased more than 30% of the tensile strength of the control treatment or without plasticizer. However, the tensile strength value of bioplastics after a decrease is still at the standard for tensile strength of plastics set by the Japanese Industrial Standard, which is more than 0.39 MPa. The addition of 20% PEG to carrageenan bioplastics decreased an average of 34.1% of the tensile strength produced without PEG. [32] also reported that giving PEG resulted in lower tensile strength of kappa
Carrageenan films than without PEG administration. This is due to the hygroscopic nature of PEG [45], so the water vapor will provide space for the polymer which causes the intermolecular bonds to become weak [56].

Carrageenan and alginate bioplastics shown in Table 3 show a decrease in tensile strength values, where the tensile strength of carrageenan bioplastics after being given an addition of 20% glycerol concentration decreased by an average of 38.76%. While alginate bioplastics after being given the addition of glycerol by 20% decreased the tensile strength by 69.77%. Research from [47] showed a decrease in the value of the tensile strength of bioplastics after being given the addition of sorbitol and glycerol plasticizers. The decrease occurs because the plasticizer can reduce the tensile force between polymers when water evaporation occurs, so that the resistance of mechanical treatment and the tensile strength of bioplastics decreases [45]. This result is also supported by data from research by [16], as shown in Figures and 3 the addition of plasticizer concentration will be directly proportional to the tensile strength of bioplastics and will increase the elongation of bioplastics. Giving glycerol as a plasticizer can weaken the intramolecular and intermolecular bonds of bioplastics. This is because the polymer undergoes bond re-formation disorder and the molecular distance increases, so that the molecular movement in the polymer increases. This results in increased flexibility while the tensile strength will decrease [57].

The addition of sorbitol can reduce the tensile strength of the resulting bioplastic. The results in Table 3 show a decrease in the tensile strength of alginate bioplastics by 50.84% after being given the addition of sorbitol by 20%. These results are supported by the research of [51] that sorbitol as a plasticizer is able to reduce the tensile strength of bioplastics. However, when viewed from the type of each bioplastic hydrocolloid, the treatment with sorbitol resulted in the least reduction in tensile strength. This is because the ability of sorbitol to absorb water is lower than glycerol and PEG, so the ability to block hydrogen bonds between polymers is also lower [58].

### Table 4. Average change in elongation at break of bioplastics.

| Bioplastics | Plasticizer | Elongation at break % (control treatment) | Elongation at break % (after added plasticizer 20%) | Δ Elongation at break | Average Elongation Change (%) |
|-------------|-------------|------------------------------------------|-------------------------------------------------|----------------------|---------------------------------|
| Carrageenan | Glycerol    | 28.88<sup>2</sup>                         | 49.22<sup>2</sup>                              | 70.43                | 92.22±30.81<sup>a</sup>        |
|             | Glycerol    | 1.77<sup>6</sup>                          | 3.8<sup>6</sup>                                | 114                  |                                 |
|             | Sorbitol    | 3.1<sup>1</sup>                           | 5<sup>1</sup>                                  | 61.3                 | 74.84±19.15<sup>a</sup>        |
|             | Sorbitol    | 7.92<sup>2</sup>                          | 14.92<sup>2</sup>                             | 88.38                |                                 |
| Alginate    | Glycerol    | 12<sup>3</sup>                            | 21<sup>3</sup>                                 | 75                   | 60.48±20.54<sup>a</sup>        |
|             | Glycerol    | 37<sup>4</sup>                            | 54<sup>4</sup>                                | 45.95                |                                 |
|             | Sorbitol    | 11<sup>4</sup>                            | 16<sup>4</sup>                                | 45.45                | 49.91±6.31<sup>a</sup>        |
|             | Sorbitol    | 10.3<sup>5</sup>                          | 15.9<sup>5</sup>                              | 54.37                |                                 |

<sup>1</sup>[16]  
<sup>2</sup>[51]  
<sup>3</sup>[42]  
<sup>4</sup>[15]  
<sup>5</sup>[33]  
<sup>6</sup>[37]

- sign in the table indicates a decreasing change;  
 a, b, ab Different script letters signify a significant difference (p<0.05) for each treatment
3.4 Bioplastic elongation at break

Elongation at break or elongation at break is the ratio between the change in length and the initial length after damage to the specimen. This property determines the ability of a natural plastic to resist deformation without cracking or breaking. Elongation at break can be determined by tensile testing [22]. The elongation at break standard set by the Japanese Industrial Standard 1975 is 10-50%. Elongation at break is calculated based on the percent elongation of the plastic.

Alginate and carrageenan bioplastics have rigid properties, so the plastic will be brittle and easy to crack [59]. Plasticizer which is added to the material for making bioplastics will increase the elasticity so that the plastic produced will not be stiff and brittle. However, the addition of too much plasticizer will reduce the tensile strength of the bioplastic itself [16]. Therefore the addition of plasticizer must be in the appropriate concentration so that it will get the percent elongation at break and maximum tensile strength.

The results of the data from the elongation of bioplastics were previously designed in a completely randomized manner. Each data from seaweed hydrocolloid bioplastic with each plasticizer is the respective treatment used. The data in the table is presented quantitatively by means of fingerprint analysis (one way ANOVA), namely by comparing the percentage change in tensile strength with each difference in plasticizer. The results of the average change in elongation were obtained after obtaining the percentage difference from the control treatment to the treatment of adding 20% plasticizer to each data source. These results were then used as replicates for each of the same plasticizers, then compared.
with the ANOVA test. The results were obtained from the ANOVA test. The results presented in table 4 are used as conclusions from the hypotheses of this study.

Table 4 presents the results from several literature sources related to changes in bioplastic elongation after being given a plasticizer with a certain concentration. The same script letter in the data obtained shows that the change in elongation from each source is not significantly different (p>0.05), although numerically it shows different values. Glycerol carrageenan bioplastics experienced an average elongation increase of 92.22%, while glycerol alginate bioplastics experienced an average elongation increase of 60.8%. According to the results of research by [16], the increase in elongation due to the application of plasticizers was followed by a decrease in the tensile strength of the bioplastic. This can also be seen in figures 1 and figure 2, where there is a decrease in the tensile strength value and an increase in the elongation value of bioplastics when the concentration of plasticizer increases. According to [45] an increase in glycerol will also increase the mobility between molecular chains which results in a decrease in the OH group on glycerol which will form intermolecular bonds. The increase in glycerol will also reduce the cohesive bonds between polymers, so that the plastic will be more elastic.

Table 4 on alginate carrageenan bioplastics with sorbitol plasticizer also experienced an increase in the average elongation by 74.84% and 49.91%, respectively. Just like other plasticizers, sorbitol which is used as a plasticizer for carrageenan bioplastics can increase the elongation of bioplastics [51]. According to [60] the melting point of sorbitol crystals is 95°C while that of glycerol is 17.8°C. Crystallization of sorbitol on the film causes the tensile strength value to increase, but on the other hand decreases the flexibility or elongation value. This is because sorbitol crystallizes at room temperature.

Moreover, PEG used as a plasticizer for carrageenan bioplastics with the addition of 20% PEG from the control resulted in an increase in elongation of 16.13% [16]. [51] reported that giving PEG can increase the elongation value of carrageenan plastics. The hygroscopic nature of PEG causes the water vapor contained in the polymer to reduce intermolecular bonds so that it will increase molecular mobility [45]. However, excess moisture will cause the polymer to become saturated and break easily. Thus the provision of PEG as a plasticizer has certain limits so as not to cause the polymer to break easily [32]. This is due to the nature of PEG which absorbs water and can inhibit water evaporation, so that bioplastics will break easily [61].

Research by [16] in figure 1 and figure 2 above show the results of changes in the tensile strength and elongation of kappa carrageenan bioplastics with various plasticizers. These data show a relationship between tensile strength and elongation after being given plasticizers (glycerol, sorbitol, PEG), where the increase in plasticizer concentration is directly proportional to the increased in elongation and decreased in the tensile strength of bioplastics. This is also supported in the research of [51], that the addition of glycerol from 20% to 40% on hydroxypropyl methylcellulose carrageenan films reduced the tensile strength of bioplastics from 24.22 MPa to 12.80 MPa. In addition, the addition of glycerol from 20% to 40% can also increase the elongation of bioplastics from 28.88% to 49.22%. This is because plasticizers can reduce intermolecular bonds. The decrease in intermolecular bonds occurs because the plasticizer can increase the bond space, to facilitate the migration of water vapor which results in a weaker bond which results in a decrease in the tensile strength of the plastic and the plastic becomes softer and more elastic [45].

4. Conclusion
Seaweed hydrocolloid bioplastics such as alginate and carrageenan bioplastics have the weakness of rigid and inelastic mechanical properties. The addition of plasticizers is done to improve the physical and mechanical properties of bioplastics. This literature review research was conducted to conclude the hypothesis to be proven. The results show that the application of plasticizers can change the physical and mechanical properties of bioplastics that previously did not use plasticizers. Differences in plasticizers affect the tensile strength of the resulting bioplastics, while thickness and elongation are not affected by differences in plasticizers.
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