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Effect of Extraction Method from Brown Seaweeds *Padina sp.* on Quality of Alginate for Edible Film Application: A Review

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Author's contribution

The sole author designed, analysed, interpreted and prepared the manuscript.

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Review Article

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ABSTRACT

Aims: This review aims to study the effect of the extraction method on the quality of the alginate extracted from *Padina sp.* and study its application for edible films.

Study Design: This research was conducted using a systematic literature review method.

Place and Duration of Study: Library sources are research articles that are available online and can be accessed through websites or search engines by entering keywords that are relevant to the topics.

Results: The results obtained are various information about the alginate of *Padina* sp seaweed extracted from several methods such as the acid pathway method, the potassium method without the addition of potassium hydroxide and the calcium method with the addition of potassium hydroxide, and the application of alginate for edible films. It is recommended that further research be carried out to obtain alginate results with more diverse extraction methods.

Keywords: Padina sp.; alginate; edible film; extraction.

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1. INTRODUCTION

Indonesia is a large country with an area consisting of one-third of the land and two-thirds of the sea traversed by the equator so Indonesia is rich in marine resources [1]. Indonesia increases its natural potential [2]. It can increase the nation's competitiveness by utilizing its existing resources in Indonesia in contributing to the world market (Sumardi and Fernandes, 2018). Indonesia has a water area of 5.8 million km2 and in Indonesian waters, there are as many as 27.2% of the world's marine fauna and flora species [3]. Of the 8,642 species, there are approximately 555 types of seaweed in the world in Indonesia. That way, Indonesia is a country that has a plasma source of the world's biodiversity in the form of seaweed, which is 6.42% [4].

Padina sp. is one of the Phaeophyta species or brown seaweed that grows in Indonesian waters. Padina sp. can be extracted and the seaweed extraction results produce alginate [5,6]. Alginate is a type of polysaccharide that makes up the cell wall components of brown algae (Phaeophyta). The components that make up alginate consist of guluronic and mannuronic acids with 1,4-Dmannuronic acid and -L-guluronic bonds [7].

Industrial activities are increasing rapidly and generating new innovations, especially in the food industry sector [8.9]. In the food alginate is used as a balance industry, regulator. thickener, and emulsifier. In foodstuffs, generally, food is very easy to decrease in quality and quality [10]. One way to slow down and prevent this can be done with proper packaging. Packaging materials derived from plastic are still often and are still widely used. But the composition of svnthetic materials from plastic causes environmental pollution [10]. With this problem, various kinds of food packaging made from organic materials have been developed or are renewable and have inexpensive economic value. One of these types of packaging is edible packaging.

Indonesia, although a developing country, has a large food industy sector [9]. In the business chain, profit must be considered. However, environmental responsibility must also be considered [11]. One of the developments is edible film products. The edible film is packaging made from organic materials (Umaraw and Verma, 2015). Edible films have biodegradable properties that can be broken down by microbes and have the function of preventing oxidation in a food product. Edible films are also used to replace plastic polymers because they are economical, renewable, and provide good physical characteristics [10].

Therefore, this review aims to determine the effect of the brown seaweed extraction method on the quality of alginate derived from *Padina sp.* and the application of alginate for the manufacture of edible films.

2. MATERIALS AND METHODS

2.1 Alginate

Indonesia is an archipelagic country with 17,504 islands, and as a maritime country, Indonesia has great potential to develop seaweed raw materials of high economic value that can be utilized in various product innovations. Several types of Indonesian seaweed that have been traded for a long time are *Gelidium sp., Turbinaria sp., Sargassum sp., Padinaria sp., Euchema sp., Hypena sp., and Gracilaria sp., Sodium alginate* is the most processed product of brown seaweed found.

The main component of the arginophyte cell wall, namely alginate, is composed of guluronate and mannuronic acids with 1,4 -D-mannuronic acid and -L-guluronic acid bonds [12,13] (Estervag et al., 2009). Alginate content of brown seaweed depends on the type of environmental conditions, harvest time, and extraction method used and is influenced by the plant part used and the plant part of the extracted brown seaweed [12,14]. Alginates are used to stabilize mixtures, dispersions, and emulsions due to their gelling properties and increased viscosity [15,16].

Because alginate is a linear molecule with a high molecular weight, it is very easy to absorb water. Therefore, alginate is very often used as a thickener. Alginic acid is commonly found in brown seaweed as calcium, magnesium, and sodium salts [17]. Alginate in the form of salt or sodium alginate can be produced from seaweed, one of which is Turbinaria sp. and Padinaria sp. The physical properties of sodium alginate are white to vellowish, almost odorless and tasteless, and in the form of flour or fiber. Alginate solutions that dissolve in water form a gel due to the presence of calcium ions and other polyvalent metal cations in acidic solutions. The chemical structure of alginate can be seen in the image below.

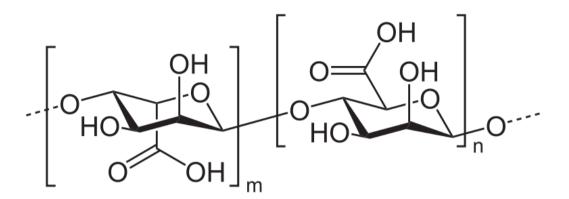


Fig. 1. Chemical structure of alginate Source: Wikipidia.org

Over the past ten years, many studies have investigated the use of alginates in food packaging design. Edible alginate films are inherently transparent, uniform, and highly soluble in water. The interaction of increasingly insoluble ions in water is induced by alginate cross-linking during film synthesis through ionic interactions and hydrogen bonding using divalent cations such as Ca2+. Therefore, coatings and films for food packaging applications can usually be made from solutions of calcium and sodium alginate which react rapidly, allowing intermolecular associations that generally involve GG blocks. As is often mentioned in the literature, alginates have excellent film-forming properties but need to be plasticized with glycerol or sorbitol to reduce brittleness after drying. Glycerol and sorbitol increased the flexibility of the alginate film, but only glycerol was able to increase the water vapor and oxygen permeability of the alginate film.

In one study, a longer shelf life in freshwater fish (Megalobrama amblycephala) was observed when using a calcium alginate coating containing tea polyphenols or ascorbic acid. In general, the addition of nano strengths (nano-strengthening agents) as whisker cellulose (cellulose whiskers) can improve the mechanical properties (tensile strength, modulus) and water barrier of alginate films, as in the case of food films based on pure alginate-acerola. Therefore, these alginate films can be used as edible coatings on various foods such as fresh vegetables and fruits, or simply to extend shelf life. This is evidenced by many studies showing that the addition of bioactive agents such as antioxidants and antibacterial agents to alginate in food wrappers is more beneficial than industrial processes that apply bioactive agents directly to food. Therefore, a wide variety of antibacterial agents, including

chemical and natural (essential oils, phenolic compounds), are efficiently incorporated into alginate films for food packaging development. For example, we can refer to recent studies combining antioxidants and antibacterial agents for food applications in alginate films and coatings such as trans-cinnamaldehyde; carvacrol, methyl cinnamate; pomegranate peel extract; thyme oil; acetic acid/lactic acid; nisin/EDTA; resveratrol; tea polyphenols; garlic oil; ascorbic acid; lycopene; clove essential oil; and tocopherol.

2.2 Edible Film

Food packaging is a packaging process that uses appropriate packaging materials to store and protect food in the hands of consumers and maintain its quality and safety. One of the commonly used packaging materials is plastic. In addition to containing very dangerous chemicals. the use of plastic also produces large amounts of waste that are difficult to decompose. Increased public awareness of health and environmental issues has led to an increase in demand for biodegradable packaging that can guarantee food safety, including packaging with edible coatings and films. The edible film is a thin layer made of hydrophilic colloid and fat or a mixture thereof which acts as a mass transfer inhibitor and can be used as a carrier for antibacterial compounds that can protect products from pathogens [18]. According to Santoso et al. (2007), the edible film can be defined as a packaging material in the form of a thin sheet (foil) that has been performed before being used to wrap food.

Edible film and coating are different things when viewed from the application process. The coating is applied directly to the surface of food

ingredients and printed, while the edible film is a thin laver that is first printed into sheets and then applied (Winarti et al., 2012). Pavlath and Orts [19] found that all types of materials used to coat or package various foods to extend the shelf life of edible products can be eaten with food, with or without further transfer called edible films. Examples of the use of edible films include the packaging of sweets, sausages, fruits, and dry soups [20]. Edible films act as the main package for wrapping food, as a barrier to mass transfer such as oxygen, carbon dioxide, and ethylene, and are involved in the respiration process. In addition, edible films can also be used as a carrier for food ingredients, including vitamins, minerals, antioxidants, antibacterial substances, preservatives, as well as ingredients to improve the taste and color of packaged products. The function and appearance of edible films depend on their mechanical properties. Mechanical properties are determined not only by the manufacturing process and application method but also by the composition of the material. The materials used to make edible films are relatively inexpensive. and biodegradable, and the manufacturing technique is simple. The principle behind the formation of edible films is that polymer chains interact to produce larger and more stable polymers [21].

Applying edible films and coatings is an easy way to increase the physical strength of foods, reduce particle agglomeration, and improve the visual and tactile properties of food surfaces [22]. They can also protect food from oxidation, water absorption/desorption, microbial growth, and other chemical reactions [23]. The most common functions of edible films and coatings are as a barrier to the movement of petroleum, gas, or vapor, and as a carrier for active ingredients such as antioxidants, antibacterial agents, colors, and flavors [24]. Edible films and coatings improve food quality by extending shelf life and increasing food safety.

2.3 Leaching Process

According to McCabe and Smith [25], solid-liquid extraction (Leaching) is the process of separating a solute contained in a solid by mixing the solid with a solvent (solvent) so that the solid and liquid mix, and then the solute can be separated from the solid because it dissolves. In the solvent, the solid-liquid extraction occurs in two phases, namely the overflow phase (extract) and the underflow phase (raffinate/dregs). The determination of the method used for extraction is based on the number of dissolved substances, the nature of the solid, the distribution in the solid, and the size of the particles. If the solute is evenly distributed throughout the solid, the material closest to the surface will dissolve first. Usually, the Leaching process is divided into three stages, namely:

- (1) First, is the change in the shape of the solute that is taken up in the solvent that seeps in.
- (2) Second, the process of liquid diffusion from the inside of the solid particles to the outside.
- (3) Third, the transfer of the solute from the solid to the solvent.

2.4 Methodology

This review uses a systematic literature review method. The systematic literature review is a term used to refer to a particular research methodology or research, the development of which is carried out to collect and evaluate research related to the focus of a particular topic. The systematic literature review is done by reviewing previous research. This review is carried out by identifying, analyzing, and synthesizing literature that is related or relevant to the topic to be discussed [26]. The purpose of a systematic literature review is to identify, evaluate, and synthesize research data that is in accordance with predetermined topics to answer questions from certain research or hypotheses [27].

3. RESULTS AND DISCUSSION

3.1 Classification, Morphology, and Habitat of *Padina sp.*

Padina sp. has a thin, soft, and smooth physique that is shaped like a wide fan. Talus from *Padina sp.* is divided into lobes or thin sheet segments [28]. The thallus of this seaweed is upright with a diameter of up to 15 cm. The color of the thallus is yellowish green to yellowish brown [29]. The following is the classification of *Padina sp.* [29].

Kingdom : ChromistaPhylum: OchrophytaClass: PhaeophyceaeOrder: DictyotalesFamily: DictyotaceaeGenus: Padina

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Fig. 2. Seaweed Padina sp.

Padina sp. can grow in clear waters to waters with a depth of 15-20 meters [30]. It grows in coastal waters where the substrate is sandy, slightly muddy, attached to rocks, attached to dead coral fragments, and sometimes attached to other macroalgae [29]. *Padina sp.* can live well at salinities in the range of 30-34%. And a good

pH for the growth of *Padina sp.* which ranged from 6.8-9.6 [31].

3.2 Content of Padina sp. Seaweed

The content of *Padina sp.* can be seen in Table 1.

3.3 Alginate

Alginate is a type of polysaccharide, its constituents consist of monomers of mannuronic acid (M units) and guluronic acid (G units). These monomers vary in the number and distribution of the alginate polymer chains. Species of algae, age, and body parts consisting of leaves and stems of algae affect the distribution, length, and unit ratio between M and G alginates. This also affects the chemical and physical properties of the GG block which plays a role in the formation of alginate gels [35]. The structure of alginate according to Lee and Mooney [36] can be seen in Fig. 3.

Table 1. Seaweed content of Padina sp.

Seaweed	Content	Reference
Padina sp.	Water content 12.09%, ash content 54.51%, fat content 0.23%, protein content 7.64%, carbohydrates 24.72%, and crude fiber 0.83%	Chowdhury et al., [32]
	9.79% water content, 21.80% ash content, 1.27% fat content, 18.03% protein content, 40.81% carbohydrates, and 8.30% crude fiber	Arguelles dan Sapin, [33]
	Water content 13.64%, ash content 38.02%, fat content 1.6%, protein content 12.33%, carbohydrates 48.06% and fiber content 20.02%	Khadijah <i>e</i> t al., [34]

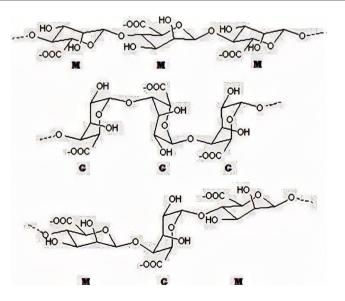


Fig. 3. Structure of alginate molecule in M and G. block types

The extraction process is a way to get alginate from brown seaweed itself [37]. Where there is a principle in extracting alginate according to Sinurat and Marliani [38], which goes through several stages including demineralization, neutralization, extraction, filtration, precipitation, and bleaching.

The results of the alginate extraction of *Padina sp.* of several methods can be seen in Table 2.

3.4 Water Content

On the water content parameter, the results ranged from 8.00%-19.68%. The water content of the alginate produced from the research of Viswanathan and Nallamuthu [43] does not meet the quality standard of alginate quality which refers to the Food Chemical Codex (FCC) which is <15%. In the research of Hamrun and Rachman [39]; Herlina et al., [42]; Hamrun et al., [5,6]; and Kautsari and Ahdiansyah (2018) the percentage of water content is not reported. The high and low water content is influenced by the process of soaking the alginate with a solution during the extraction process. The drying process of the alginate itself after the extraction process also affects the water content of the alginate [48].

3.5 Ash Content

In the ash content parameter, the results ranged from 17.34%-31.24%. In the research of Hamrun and Rachman [39]; Herlina et al., (2016); Hamrun et al., [5,6]; and Kautsari and Ahdiansvah [47] the resulting ash content is not reported. The value of ash content is influenced by the washing process after the alginic acid formation process until the pH is close to a neutral number. The addition of 1% HCl is also a factor in decreasing the value of the ash content [41]. The ash content of research conducted by Permatasari, et al. [40] is 17.34%. Meanwhile, in the research of Mushollaeni [46] and Septiani et al. [45], the resulting ash content of sodium alginate is 28.59% and 31.24%. The ash content in the three studies did not meet the quality standards of alginate quality based on the Food Chemical Codex (FCC), which ranged from 18%-27%. In each alginophyte according to Mushollaeni [46], the ash content produced is also influenced by the habitat of the seaweed growing. Padina sp, whose habitat is at the bottom of the water has more ash content. With the high ash content produced in the research of Mushollaeni [46] and Septiani et al. [45], it is suspected that the use of HCI has not been optimal.

Method	Parameter							
	Water content	Ash Level	Viscosity	yield	рН	Reference		
Sour	-	-	-	28.4%	-	Hamrun dan Rachman, [39]		
	9,07%	17,34%	75,20 cP	5,89%	7,35	Permatasari et al., [40]		
	9,00%	20,83%	10,42 cP	20,59%	6,57	Pasaribu et al., [41]		
	-	-	-	18,48%		Herlina et al., [42]		
	8,00 %	-	13,33 cP	-	-	Hamrun et al., [5,6]		
Calcium without	19,68%	23,01%	-	-	-	Viswanathan dan Nallamuthu, [43]		
КОН	13,1%	20%	57,6 cP	-	9,72	Rashedy et al., [44]		
	-	-	160 cP	14%	-	Hamrun et al., [5,6]		
	10,87%	31,24%	55,10 cP	17,82%	8,21	Septiani et al., [45]		
Calcium with KOH	12,7%	28,59%	-	16,93%	-	Mushollaeni W, [46]		
	-	-	-	6,65%	8	Kautsari dan Ahdiansyah, [47]		

Table 2. Extraction of *Padina* sp. alginate seaweed extract

3.6 Viscosity

In the viscosity parameter, the results ranged from 10.42 cP-75.20 cP. The results of research by Hamrun and Rachman [39]; Herlina et al., Viswanathan and Nallamuthu [42]: [43]: Mushollaeni [46]; and Kautsari and Ahdiansvah (2016) for the viscosity parameters are not shown. In other studies, the results of the viscosity parameter have met the quality standard of alginate quality based on the Food Chemical Codex (FCC) which is 10-5000 cP. The difference in viscosity value indicates the occurrence of degradation at the time of alginate extraction. The presence of degradation causes a decrease in the weight of the alginate molecule so that the viscosity of the alginate also decreases [5,6]. In addition, the precipitation process with ethanol solution according to Jian et al. [49]. The increase in the viscosity value also depends on the arrangement of the guluronic acid and mannuronic acid chains [50]. Viscosity value can also be influenced by the concentration of alginate, alginate gel strength, degree of polymerization, and molecular weight [35].

research of Hamrun and Rachman [39], amounting to 28.4%. The high or low yield of sodium alginate produced is caused by the type of species, the habitat where the seaweed grows, the climate, and the extraction method used [51]. The higher the yield obtained, the better the alginate extraction process used. The low yield obtained can be caused by the treatment of flouring, milling, and sieving [38].

3.8 pH

The resulting pH in the study ranged from 6.57-9.72. In the research of Hamrun and Rachman [39]; Herlina et al., [42]; Hamrun et al., [5,6]; Viswanathan and Nallamuthu [43]; and Mushollaeni [46] the results of the pH parameters were not reported. In the literature reported, the pH parameter is in accordance with the quality standard of alginate quality based on the Food Chemical Codex (FCC) which is 3.5-10. Alginate will be stable if it has a pH ranging from 5.5-10 at room temperature. If the pH is below 5.5, the alginate solution will form a gel. The pH value affects the difference in the solution used. If the pH is more than 10, the sodium alginate solution is unstable and precipitation occurs on sodium alginate if the pH is less than 3.5 [45].

3.7 Yield

The resulting yield is in the range of 5.89%-28.4%. The highest yield is in the

Component Film	Method	Plasticizer	Results	Application	Reference
Sodium alginate, essential oils	Casting	Glycerol	 The resulting edible film has a thickness of 0.109 mm, water vapor permeability of 0.006 g.h-1 .mm.m- 2.Kpa-1, elongation of 54.245%, and tensile strength of 2.136 Mpa. Edible films can improve the quality and shelf life of food, make food less perishable, and reduce microbial growth The resulting edible film has an antibacterial effect 	Films and coatings for food	Mahcene et al., [10]

Table 3. Application of alginate as edible film

against foodborne pathogenic bacteria - Edible films can

Component Film	Method	Plasticizer	Results	Application	Reference
			increase the melting point value and reduce thickness, moisture, and oxygen		
Alginate, Spirulina platensis powder	Casting	Glycerol	- The value of the thickness of the edible film is 0.2 mm, the value of the water vapor transmission rate of this edible film does not meet the standards, the elongation is 17.09%- 58.236% (not yet met the standard), the tensile strength value is 0.0883-4.01 (meet the standard), phenol content is 52.373%, antioxidant activity is 37.695% and solubility is 50.21%. - The resulting edible film can prevent food oxidation	food coating	Hayati et al., [55]
Sodium alginate, lemongrass oil	Casting	Glycerol	 Increase the shelf life The resulting edible film has a thickness of 0.047 mm, a solubility of 63.19%, a water vapor transmission rate of 0.1333 g/h.m2, a tensile strength value of 10.63 MPa, and an elongation at break of 36.78%. Edible film sodium alginate with citronella oil is effective against gram-positive bacteria such as <i>Bacillus subtilis</i> and <i>Staphylococcus aureus bacteria</i>. 	food coating	Othman et al., [56]
Sodium alginate, pectin, whey protein concentrate	Casting	Glycerol	- The resulting edible film is transparent - Edible film has a thickness of 41-50 m, water vapor permeability of 7.2- 9.6 (1010 g mm h-1 cm-1 Pa-1), the	food coating	Chakravartula et al., [57]

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Component Film	Method	Plasticizer	Results	Application	Reference
			tensile strength of 11- 62 Mpa, elasticity of Young's modulus of 288- 1467 MPa and elongation at 16%- 40%		
Sodium alginate and pectin	Casting	Glycerol	 The resulting edible film is homogeneous and transparent Has a high water absorption rate compared to pectin or composites The high content of pectin affects the internal structure of the film The resulting edible film has a thickness of 12.5-26.8 m, water vapor permeability of 0.84-1.73 10-10gm- 1s1Pa-1, tensile strength of 22.5-42.3 MPa, and elongation at break. (elongation) of 5.9-14.9% 	food coating	Galus dan Lenart, [58]
Alginate, whey protein isolate, soy protein isolate, gelatin	Hot Melt Extrusion	Glycerol	 The resulting edible film has an elongation of 88.3%, a tensile strength of 1.84 MPa, a thickness of 0.147mm The edible film produced by the addition of whey protein is more transparent than the addition of soy protein isolate, which is yellowish brown in color. 	Sausage coating	Harper et al., [59]
Sodium Alginate Powder	Casting	Sorbitol	- The resulting edible film has a tensile strength of 50.7 N/mm2, an elongation of 5.6%, - The resulting edible film is transparent.	food coating	Jost et al., [60]

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3.9 Edible Film

Alginate can be used to make films because alginate films have low permeability to oils, fats,

and oxygen. This low permeability can inhibit the occurrence of oxidation in fruits and vegetables [61,62]. This film also serves to extend the shelf life of fruits and vegetables and can delay the

ripening of fruits and vegetables. The film can also prevent spoilage caused by microbes by inhibiting microbes and slowing down respiration. In addition, alginate used as an edible film has good mechanical characteristics and can reduce oxygen transfer [52].

The edible film is a thin layer, where the layer is made of materials that can be consumed directly. Materials for making edible films are divided into 3, namely hydrocolloids (polysaccharides and proteins), lipids, and composites (a mixture of hydrocolloids and lipids). These three materials can be recycled naturally (biodegradable) [53]. The purpose of making edible films is that the edible films can later be consumed together with packaged food. Edible films are safe and healthy because thev do not cause harmful environmental impacts and can reduce waste from plastics that cannot be decomposed by soil microorganisms (non-biodegradable) [54].

There are four kinds of methods that can be used for the manufacture of edible films, including the solvent casting method, the hot melt extrusion method, the semisolid casting method, and the rolling solvent casting method. These methods have different characteristics and have their own advantages and disadvantages. The most common method and has been used for a long time is the solvent casting method because it is easy and considered the most practical [53]. The results of the application of alginate as an edible film can be seen in Table 3.

The differences in film components, manufacturing methods, and plasticizers produce different edible films in each library. Alginate application for an edible film produced edible film in accordance with the standards and good quality, namely the research of Mahcene, et al. [10], the film components used are sodium alginate and essential oils. The edible film was obtained in the form of a thickness of 0.109 mm, water vapor permeability of 0.006 g.h-1 .mm.m-2.Kpa-1, elongation of 54.245%, and tensile strength of 2.136 MPa. The resulting parameters are in accordance with the quality standards of edible film quality based on the reference of the Japanese Industrial Standard (JIS). In this study, the edible film produced can improve the quality and shelf life of food, make food not easily damaged, and reduce microbial growth. The resulting edible film also has an antibacterial effect against pathogenic bacteria derived from food [63,64]. Edible films can increase the melting point value and reduce thickness,

moisture, and oxygen [65-68]. In addition, good edible films were also produced in the research of Othman, et al. [56], which produced a film thickness of 0.047 mm, a solubility of 63.19%, a water vapor transmission rate of 0.1333 g/h.m2. a tensile strength value of 10.63 MPa and an elongation at break of 36.78%. The resulting edible film can effectively fight gram-positive bacteria, namely Bacillus subtilis bacteria and Staphylococcus aureus bacteria found in food, and increase the shelf life of food products. Alginate quality of Padina sp. was better obtained using the calcium channel extraction method without KOH immersion. Factors affecting the quality of alginate, such as extraction method, temperature, extraction time, alkali concentration, and pretreatment during extraction should also be considered and with p9nel components [69,70].

4. CONCLUSION

The conclusion obtained in this review is that the alginate quality of Padina sp. is better obtained by using the calcium pathway extraction method without KOH immersion. It is also necessary to pay attention to factors that affect the quality of alginate, such as extraction method, temperature, extraction time, alkali concentration, and pre-treatment at the time of extraction. In the table for the application of alginate for the manufacture of edible films, good quality edible films are obtained, namely with film components in the form of sodium alginate with essential oils and sodium alginate with lemongrass oil. The film components used are sodium alginate and essential oils. The edible film was obtained in the form of a thickness of 0.109 mm, water vapor permeability of 0.006 g.h-1 .mm.m-2.Kpa-1, elongation of 54.245%, and tensile strength of 2.136 MPa. The resulting parameters are in accordance with the quality standards of edible film quality based on the reference of the Japanese Industrial Standard (JIS).

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COMPETING INTERESTS

Author has declared that no competing interests exist.

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