



# Seaweed derived alginate, agar, and carrageenan based edible coatings and films for the food industry: a review

Mayushi Malshika Jayakody<sup>1</sup> · Mihiri Priyanwadha Gunathilake Vanniarachchy<sup>1</sup> · Isuru Wijesekara<sup>1</sup>

Received: 27 July 2021 / Accepted: 20 December 2021

© The Author(s), under exclusive licence to Springer Science+Business Media, LLC, part of Springer Nature 2022

## Abstract

Accumulation of non-biodegradable plastics has adversely affected the environment. Hence, there is a need of promoting biodegradable polymer packages as substitutes for non-biodegradable plastic packages. Various studies have focused on utilisation of seaweed-derived polysaccharides in the development of coatings and films because of their renewability and sustainability for food packaging. Alginate, agar, and carrageenan are seaweed-derived polysaccharides that are widely used in the development of coatings and films due to their gelling ability. Alginates are mainly extracted from brown algae. Agar and Carrageenan are extracted from certain types of red algae. These developed coatings could be successfully utilized to extend the shelf life and maintain proper quality parameters of food during the shelf life. Films can be used to partially replace non-biodegradable polymer packages found in the market. Thus, the article reviews the basic information and applications of edible coatings and films from seaweed-derived polysaccharides in the food industry.

**Keywords** Alginate · Agar · Carrageenan · Edible films · Edible coatings · Packaging

## Introduction

At present, there is an increased interest in biodegradable materials for application in food packaging. This increased interest in biodegradable polymer packages is mainly due to the adverse environmental impact caused by the accumulation of large quantities of non-biodegradable plastic waste [1]. Recently, edible films and coatings have emerged as alternative eco-friendly packages to replace certain synthetic plastic-based packages in the food industry.

Edible coatings and films can be defined as primary packages which are developed from edible components. Generally, edible films can be introduced as self-standing structures in nature, while edible coatings adhere to the surface of the food component [2]. Films can be commonly used as

wrappers or separation layers; and can also form into casings, capsules, tiny pouches, and bags. Coatings involve the formation of films directly on the surface of the food. Therefore, coatings become part of the product and remain on the product itself through the shelf life and consumption [3]. Factors such as barrier properties, their capability for physical and mechanical protection, and capability to improve the food appearance are mainly considered when applying a coating of film in to a food product [4]. Other than that, the degradation rate of the film and release of harmful compounds into the food product by the applied coating or film should also be considered when utilizing an edible coating or film in the food industry.

Edible coatings and films have received an increased attention from consumers due to their advantages. The main advantage is that these edible coatings or films are regarded as safe to be consumed with food and they are eco-friendly. Edible films and coatings protect the food product from physical, chemical, and biological degradation. They prevent the transfer of moisture, gases such as oxygen, flavour, and oils between food and the surrounding environment and/or between different compartments in a heterogeneous food product [5]. They also extend the shelf life by protecting food products from surface microbial growth, light-induced chemical changes, and oxidation

✉ Mihiri Priyanwadha Gunathilake Vanniarachchy  
mihiripg@sjp.ac.lk

Mayushi Malshika Jayakody  
malshika@sci.sjp.ac.lk

Isuru Wijesekara  
isuruw@sci.sjp.ac.lk

<sup>1</sup> Department of Food Science and Technology, Faculty of Applied Sciences, University of Sri Jayewardenepura, Gangodawila, Nugegoda, Sri Lanka

of nutrients. At the same time, some of these coatings and films act as carriers of different kinds of active substances, like antioxidants, antimicrobials, colouring, preservatives, and flavours to improve the quality of food [6, 7]. Biopolymers such as lipids, proteins, and polysaccharides can be used for the development of biodegradable edible films and edible coatings [8]. Plasticizers such as glycerol and different types of additives can be included in to the formulation to modify and improve the physical properties and the functionality of edible films and edible coatings. Previous studies have revealed that seaweeds are rich sources of polysaccharides [9, 10]. Alginate, agar, and carrageenan are the three main polysaccharides found in seaweeds. These seaweed-based polysaccharides can be utilized in development of edible coatings and films for successfully incorporated in the food industry. Thus, the main intention of this review article is to provide information about applications of seaweed derived alginate, agar and carrageenan based edible coatings and films in the food industry.

## Properties of edible coatings and films

Edible films and coatings are regarded as thin layers of edible components. They generally act as primary packages and are consumed with the food itself [2, 11]. The main difference between an edible coating and an edible film is that an edible coating is generally applied in liquid form by dipping the product into the developed coating formulation while edible films are initially developed as sheets and later used as wrappers [12]. The development of coatings or films for the food industry must be economical. A good coating or a film will be transparent to opaque and must have the capability to tolerate slight pressure [13]. According to Dhall [13], properly applied coatings cover the product adequately. Such coatings should be water-resistant and reduce water vapour permeability. Another important point is that the applied coatings or films should not deplete oxygen or build up an excessive amount of carbon dioxide around the food

commodity since a minimum of 1–3% oxygen is required around a commodity to avoid a shift from aerobic respiration to anaerobic respiration. Additionally, the coating should have the ability to improve the appearance of the product, maintain the structural integrity, enhance mechanical properties, carry active agents such as antioxidants, antimicrobials, vitamins, etc. and retain volatile flavour compounds in the commodity. Another important thing is that coatings or films should show efficient drying performance and are also non-sticky. It should not impart an undesirable odour or flavour to the food commodity which will affect the actual quality of the commodity.

## Alginate

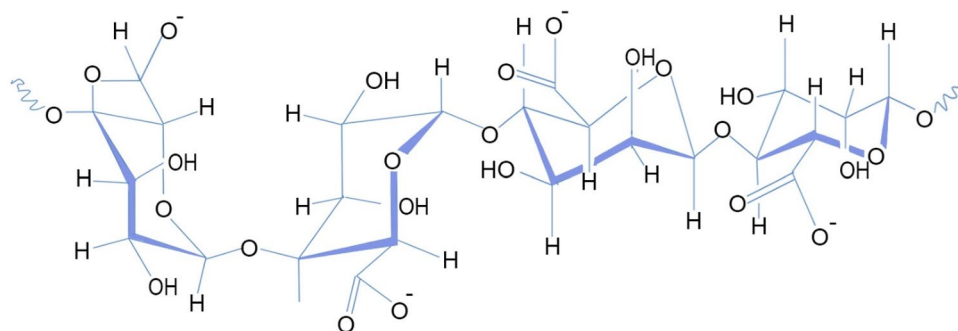
### Structure of alginate

Alginate is a glycuronan and an acidic linear polysaccharide consisting of residues of  $\beta$ -D-mannuronic acid (M block) and  $\alpha$ -L-guluronic acid (G block) which are arranged in a block-wise fashion along the polymer chain. Alginate can be homopolymers with polyguluronate or polymannuronate arranged in a block-wise manner or heteropolymers with a mixed sequence of these residues. Block structures arranged along the polymer chain are called G-blocks, M-blocks, and MG-blocks [14]. The chemical structure of alginate is shown in Fig. 1.

### Extraction methods of Alginates from seaweeds

Alginates are mainly extracted from brown algae varieties such as *Macrocystis*, *Laminaria*, *Ascophyllum*, *Ecklonia*, *Eisenia*, *Nereocystis*, and *Sargassum* [16]. Strong alkaline conditions are used to extract alginates from brown algae. Extraction of alginates can be done with or without adding formaldehyde. Sodium alginate extracted with a formaldehyde pre-treatment was observed lighter in colour than those that were extracted without pre-treating with formaldehyde. A higher extraction yield was obtained for the method

**Fig. 1** Chemical structure of alginate. (Source: [15])



without a formaldehyde pre-treatment step [17]. Alginate is commonly extracted by four main methods. Those methods are the hot extraction method, cold extraction method, high temperature (80 °C) alkaline extraction method, and room temperature alkaline extraction method.

### Cold extraction method

Cold extraction is usually carried out at a temperature around 27 °C. Initially, dried seaweed powder is soaked overnight in 1% CaCl<sub>2</sub> solution at 27 °C and washed with distilled water. After that, the seaweed sample is soaked in 5% HCl solution for 1 h and washed with distilled water. Then, stored in 3% Na<sub>2</sub>CO<sub>3</sub> overnight before separating the viscous mixture by centrifuging at 14,000×g. Finally, extracted sodium alginate was precipitated by adding an ethanol/water mixture (1:1, v/v) [18]. A previous study has reported the cold extraction yield of alginate in *Sargassum baccularia*, *Sargassum binderi*, *Sargassum siliquosum* and *Turbinaria conoides* as, 23.9%, 28.7%, 38.9% and 40.5% respectively [18].

### Hot extraction method

In the hot method, samples were treated similarly to the cold method, except that the storing time in CaCl<sub>2</sub> was 3 h at 50 °C [18]. The molecular weight and intrinsic viscosity of alginate vary according to the extraction method. Chee et al. [18] have observed that the intrinsic viscosity of alginate from *Sargassum baccularia* dropped drastically when it was extracted through the hot method compared to the cold method. Chee et al. [18] has observed higher molecular weights for alginate of *Sargassum baccularia*, *Sargassum binderi*, *Sargassum siliquosum*, and *Turbinaria conoides* extracted from the cold extraction method compared to the hot method. A previous study has reported the hot extraction yield of alginate in *Sargassum baccularia*, *Sargassum binderi*, *Sargassum siliquosum* and *Turbinaria conoides* as, 26.7%, 38.7%, 49.9% and 41.4% respectively [18]. The main advantage of hot extraction method over cold extraction method is the extraction yield. Hot method extracts more sodium alginate compared to the cold method [18].

### Room temperature alkaline extraction method

In the room temperature alkaline extraction method, 2% w/w Na<sub>2</sub>CO<sub>3</sub> is added to the dried seaweed powder and kept for 2 h. After that, it is filtered to obtain the filtrate. Then 0.2 N HCl was added to the filtrate and stirred well to form insoluble alginic acid. Then the mixture obtained was filtered and the resulting residue was dried to obtain

alginic acid. After that, an excess of 2% w/w sodium carbonate solution was added to alginic acid which forms soluble sodium alginate. Finally, an equal volume of ethanol was added to the solution to precipitate sodium alginate [19]. The alkaline extraction step and alcohol precipitation step can be repeated to maximize the extraction yield [20]. A previous study has reported the room temperature alkaline extraction yield of alginate from *Sargassum Subrepandum* as, 17.5% w/w [19].

### High temperature (80 °C) alkaline extraction method

In the high temperature (80 °C) alkaline extraction method, the sample was treated similarly to the room temperature alkaline extraction method except that the initial storing time of seaweed in 2% w/w sodium carbonate was done at 80 °C with constantly stirring it for 2 h [19]. The high temperature-alkaline extraction method has given higher alginate extraction yields for *Sargassum Subrepandum* compared to the room temperature-alkaline extraction method [19]. A previous study has reported the room temperature alkaline extraction yield of alginate from *Sargassum Subrepandum* as, 21% w/w [19]. As an advantage high temperature (80 °C) alkaline extraction method has given a higher extraction yield over room temperature alkaline extraction method [19].

### Properties of alginate

Alginate can be introduced as a polyuronide which is a natural ion exchanger. Solubility of alginate in a solvent is governed by pH, ionic strength, and the presence or absence of gelling ions in the solvent. Thus, to dissolve alginate in a solvent, it is necessary to make the pH of the solvent be above a certain critical value so that the carboxylic acid groups of alginate be deprotonated. At the same time, the solvent must be free of crosslinking ions. If not, gelation of alginate will take place [21]. To make alginate soluble in an organic media requires the formation of a tetrabutylammonium (TBA) salt. Previous studies have reported that a complete dissolution can be observed in alginate when TBA-alginate is in polar aprotic solvents containing tetrabutylammonium fluoride (TBAF) [22].

Among the properties of alginates, one of the most important properties of alginate is its ability to selective binding to multivalent cations. That feature is the basis of gel formation in alginate [23]. The increasing order of affinity of alginate for alkaline earth metals is, Ca<sup>2+</sup> < Sr<sup>2+</sup> < Ba<sup>2+</sup>. Generally, Mg<sup>2+</sup> ions and monovalent cations do not form gels with alginate. Divalent cations such as Pb<sup>2+</sup>, Cu<sup>2+</sup>, Cd<sup>2+</sup>, Co<sup>2+</sup>, Ni<sup>2+</sup>, Zn<sup>2+</sup>, and Mn<sup>2+</sup> can also form gels with alginate but their toxic nature limits their usage especially in the food industry. Gelling properties of alginate are

affected by certain factors such as proportion and length of the G block in the polymeric chain, the capacity to bind the number of ions, the type of gelling ions in the medium, and gelling conditions of the medium. When  $\text{Ca}^{2+}$  like divalent ions are introduced into the alginate solution, it forms divalent salt bridges by bounding  $\text{Ca}^{2+}$  ions between two chains. Hence, stimulates conformational changes in alginates such as alignment of the G-blocks and formation of an egg-box model [24]. It is stated that the presence of high content of G-Blocks in alginate will create rigid and dense gels, while the presence of high content of M-Blocks in alginate will build flexible, porous gels. Generally, alginate gels are not highly acid-resistant. They shrink at lower pH values [23].

### Application of alginate as a coating

Sodium alginate forms thick and stable gels with calcium ions in a solution by forming crosslinks between the carboxylate ions of alginate guluronate units and calcium ions [25]. Thus, this ability can be used in developing edible coatings and films in the food industry. Generally, alginate films are considered as poor moisture barriers. But the incorporation of  $\text{Ca}^{2+}$  ions enhances the moisture barrier property of alginate films [26]. In film formation, the charged state of alginate can be used. Alginate can be used only to increase the viscosity of a solution in the absence of divalent ions. However, the addition of divalent cations into the alginate solution initiates the gelation through an ion exchange process.

Fruits are highly perishable. The post-harvest quality maintained in harvested fruits generally affects the purchase decision of consumers. The research by Valero et al. [27] has studied the effect of the application of alginate-based edible coatings on plum quality during post-harvest storage. Plums (*Prunus salicina Lindl.*), for this study were plucked at the commercial ripening stage. 1% and 3% alginate coatings were used. The results of the study revealed that the alginate coating could be used to delay the post-harvest ripening process of plums. Even a 1% level retard the onset of ethylene climacteric peak. Both 1% and 3% treatments have shown effective in delaying weight loss, acidity loss, textural loss, and colour changes in plums. Olivas et al. [26] have studied the application of alginate coating on minimally processed “Gala apples” for preservation. Alginate films will be a good option to coat fresh apples, because these films become stronger when cross-linked with  $\text{Ca}^{2+}$  ions, and adhere to the apple surface via cross-linking (alginate-Ca-Pectin). Minimally processed freshly cut apples have a shorter shelf life than unprocessed apples since they are more prone to microbial spoilage, increased respiration rate, increased water loss, and increased ethylene production. According to the results of the study, it has revealed that alginate coatings can successfully maintain the quality of apple slices without causing anaerobic respiration. These

coatings reduce the water loss, textural loss, and browning of apple slices. It was observed a higher concentration of volatiles in coated apple slices than uncoated apple slices during storage.

Diaz-Mula et al. [28] have studied the application of alginate-based coatings to preserve quality and bioactive compounds present in sweet cherry fruit during storage. According to the results, a significant delay in colour change was reported in coated fruits than the uncoated control sample while no significant changes were reported in cherries treated with 3% and 5% coating formulations in colour during cold storage. Visual observations confirm the absence of and decay symptoms in any fruit either control or treated during the 16 days of cold temperature storage or after further storage life at 20 °C. Generally, weight loss increases during the storage period of a fruit. The weight loss increment for control, 1%, 3%, and 5% treated cherries after 16 days of cold storage were reported as  $6.81 \pm 0.08\%$ ,  $5.93 \pm 0.12\%$ ,  $4.88 \pm 0.15\%$ , and  $3.71 \pm 0.09\%$  respectively. Results have shown that there is a reduction in the weight loss percentage of coated cherries than the control and this weight loss percentage has decreased with the increment in the coating concentration. Results have also reported that the respiration rate was very low during cold storage. It is between 10 and 15  $\text{mg CO}_2 \text{ kg}^{-1} \text{ h}^{-1}$ . No difference in respiration rate was observed among control and coated cherries. Less titratable acidity reduction has been observed in coated fruits than in the control. According to the results, the initial titratable acidity (TA) of the control sample of cherries at harvest was  $0.91 \pm 0.01 \text{ g } 100 \text{ g}^{-1}$ . Reported titratable acidity of control, 1%, 3%, and 5% treated samples after the storage period were,  $0.57 \pm 0.01$ ,  $0.77 \pm 0.01$ ,  $0.81 \pm 0.02$  and  $0.85 \pm 0.02 \text{ g } 100 \text{ g}^{-1}$  respectively. It is also mentioned that the effect of coating on acidity retention could be a result of the lower respiration rate found in coated fruits. When considering the texture, reduction in fruit firmness has retarded in alginate-treated cherries while control fruits have exhibited a significantly higher reduction in firmness. Anthocyanin concentration at harvest was recorded as  $23.53 \pm 2.13 \text{ mg } 100 \text{ g}^{-1}$  and this level increased in control fruits until  $40.39 \pm 2.03 \text{ mg } 100 \text{ g}^{-1}$  at the end of the storage period, while these values were  $37.24 \pm 1.05$ ,  $32.04 \pm 1.27$ , and  $26.34 \pm 1.21 \text{ mg } 100 \text{ g}^{-1}$  for fruits coated with alginate at 1%, 3%, and 5% respectively. Increment in this anthocyanin content reflects the ripening of these cherries. Hence, the application of alginate coating has delayed this ripening process. Senturk et al. [29] have developed a uniform alginate-based edible coating for freshly cut cantaloupe pieces and strawberries. Generally, in the conventional coating process, a calcium solution is applied to the fruit after applying the alginate coating. This is to initiate the formation of crosslinking between alginate and calcium ions. In this study, a novel alginate coating aspect other than the conventional coating



process has been discussed. In the novel coating technique, an extra immersion step was introduced. Therefore, before applying the alginate coating on cantaloupe pieces, cantaloupe pieces were immersed in a calcium lactate solution which is a  $\text{Ca}^{2+}$  ion source. This novel alginate coating process has enabled to achieve a uniform coating layer on the fruit. Alginate coating solution used in the study was comprised of sodium alginate (1.25%, w/w), glycerol (2% w/w), surface-active agents such as, 1% span 80 (w/w) and 0.2% tween 80 (w/w) and Sunflower oil (0.2%, w/w). Stereomicroscope images have revealed that the novel coating process generates thicker and more homogeneous gel formation on the coated fruit. According to the results of weight loss analysis, the weight loss (%) of cantaloupe samples and strawberry samples has increased with the storage time. However, the weight loss (%) increment is less in coated cantaloupe pieces than in uncoated samples. Thus, an application of alginate-based coating has significantly decreased the increment of weight loss (%) of fresh-cut cantaloupes compared to the uncoated. On contrary, a significant increase in the water loss in coated strawberries than uncoated strawberries was reported as a negative effect. Maftoonazad et al. [30] have studied the Shelf-life extension of peaches by studying the effect on the respiration rate, firmness, acidity, pH, total soluble solids (TSS), and desiccation rate of peaches by applying sodium alginate and methylcellulose based edible coatings. According to the results, the respiration rate, moisture loss, and changes in quality parameters were observed to be much lower in coated peaches than in the control. The maximum acceptable shelf-life of the control sample at 15 °C was 15 days while the alginate-coated samples have maintained their acceptability up to 21 days and methylcellulose-coated samples have maintained their acceptability up to 24 days. According to the authors, the reduction in respiration rate and transpiration rate of peaches due to the application of the coating may be the reason responsible for maintaining the quality and increased shelf-life of peaches. Tabassum and Khan [31] have studied the quality parameters and shelf life of freshly cut papaya with an application of alginate-based edible coatings containing thyme and oregano essential oils in various concentrations as the lipid component of the alginate-based coatings. Results revealed that incorporation of thyme and oregano essential oils to alginate-based edible coating has the potential to improve weight loss, slow pH changes, delay consumption of organic acids and reduce the respiration rate of fresh-cut papaya. Coatings with essential oils have successfully maintained the microbial quality of fresh-cut papaya. Alginate based coatings can also applied on meat products. Alexander et al. [32] have applied an alginate edible coating and basil (*Ocimum* spp.) extracts on beef. Basil has used as an antioxidant in the coating formulation. The inclusion of basil extract in the alginate-based edible coating has increased the

antioxidant activity and has reduced the meat lipid oxidation more effectively than the coating without basil. The results have also reported that the coating could decrease the weight loss and could increase the tenderness of meat. They have concluded that the application of an edible coating containing natural compounds with antioxidant activity in animal meat products can improve their characteristics during shelf-life. Matiacevich et al. [33] have studied the application of an alginate, thyme oil and propionic acid for the preservation of fresh chicken breast. The sensorial analysis results have shown that there is no significant differences between coated and uncoated samples. Thus, there is no influence on buying decisions of consumers. The selected coating formulation developed using alginate (1% w/w) and propionic acid (0.5% w/w) was successful in increasing the shelf life of chicken breast by 33% with the lowest dehydration. Ruan et al. [34] have determined the effect of sodium alginate and carboxymethyl cellulose edible coating with epigallocatechin gallate on quality and shelf life of fresh pork stored at  $4 \pm 1$  °C for 7 days. Results revealed that the fresh pork coated with the coating had a significant inhibitory effect on its microbial growth, lipid oxidation and total volatile basic nitrogen. Thus, application of sodium alginate, carboxymethyl cellulose and an epigallocatechin gallate based coating can prevent the decay and significantly increase the shelf life of fresh pork. Summarised findings on some of the applications of alginate-based edible coatings on foods are given in Table 1.

### Application of alginate as a film

Pavlath et al. [35] have developed pliable biodegradable films from alginic acid with comparable physicochemical properties such as strength and insolubility in water, to those of synthetic non-biodegradable films used in food packs. This polymer is nontoxic. Hence, it could be used in food packaging. Alginic acid can form complexes by interacting with multivalent ions. The research states that the components can be mixed to cast as films. When the components are mixed, since the gel formation is instantaneous, it prevents casting. Uniform gelation in a film can be obtained by immersing a previously casted alginate-based film in an aqueous solution of multivalent cations. In this method, the concentration of the solution and the time period of exposure of the film to the multivalent cation solution could be used to control or influence the film properties. Multivalent ions of analytical reagents,  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ ,  $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ ,  $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ ,  $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ ,  $\text{ZnCl}_2$ , and  $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$  have been used in the study. Gel formation in films has been done by two techniques known as immersion and mixing. According to the results, air-dried sodium alginate films were not moisture resistant and readily dissolved in distilled water. Through immersing them in multivalent ions, their solubility has

**Table 1** Studies on application of alginate-based edible coatings to improve post-harvest quality of food

| Food  | Best coating Composition                | Composition        | Method of application             | Main findings   | References |
|-------|---|--------------------|-----------------------------------|---|------------|
| Plums | 3% (w/v)alginate and 20% (v/v) glycerol | Alginate, glycerol | Dipping the fruit twice for 1 min | <p>Application of an alginate coating has inhibited the onset of the ethylene climacteric peak</p> <p>3% alginate coating has highly inhibited the ethylene climacteric peak</p> <p>Application of an alginate coating has significantly decrease the weight loss in plums since edible coating act as an extra layer which coats the stomata leading to a decrease in transpiration rate of fruits</p> <p>3% coating was more effective than 1% coating in decreasing the weight loss</p> <p>Application of an alginate coating reduce firmness loss and acidity loss in plums and colour changes</p> <p>3% coating was more effective than 1% coating in reducing firmness loss, acidity loss and colour changes</p> <p>Increase the storage period of plums by 2 weeks</p> | [27]       |

Table 1 (continued)

| Food                              | Best coating Composition                                      | Composition  | Method of application   | Main findings  | References |
|-----------------------------------|---|--|---|--|------------|
| Minimally processed 'Gala' apples | Formulation 2 in terms of preventing water loss               | 3 coating formulations, Formulation 1 – Alg-Ca solution<br>Alginate, fructose, potassium sorbate<br>Formulation 2 – Alg-Ca-AMG<br>Alginate, fructose, potassium sorbate, Acetylated monoglycerid, glycerol alpha-monostearate and linoleic acid<br>Formulation 3 – Alg-Ca-MF<br>Alginate, fructose, potassium sorbate, Butter, glycerol alpha-monostearate and linoleic acid | Immersion of apple wedges in a CaCl <sub>2</sub> solution followed by coating solutions | All these coatings demonstrated delay in weight loss, firmness loss, and retarded onset of browning of apple wedges<br>Application of the coating will prevent water loss by producing a high relative humidity at the surface of slices and thereby reducing the gradient to the exterior<br>Among the 3 formulations, formulation 2 is the best coating to prevent water loss<br>Coatings containing saturated long chain fatty acids (e.g. AMG) exhibit better water vapour barrier properties than coatings contacting unsaturated short-chain fatty acids (e.g. butter)<br>Browning of slices were delayed since all these coatings act as barriers to oxygen which is necessary for browning<br>All these coatings prevent anaerobic respiration in apple slices<br>All these coatings allows retention of a higher concentration of flavour volatiles | [26]       |
| Sweet cherry fruits               | 3% (w/v) or 5% (w/v) alginate coating with 20% (v/v) glycerol | Alginate, glycerol,  | Dipping twice for 1 min   | Application of an alginate coating has successfully delayed the post-harvest ripening by reducing colour changes, acidity loss, firmness loss and respiration rate<br>Increase in alginate concentration has given promising results to quality parameters of cherries   | [28]       |

Table 1 (continued)

| Food                                | Best coating Composition   | Composition  | Method of application | Main findings  | References |
|-------------------------------------|--|--|-----------------------|--|------------|
| Fresh-cut Cantaloupe and Strawberry | -  | Sodium alginate (1.25% w/w), Glycerol (2% w/w), span 80 (1% w/w), tween 80 (0.2% w/w), Sunflower oil and (0.2%, w/w) crosslinking reaction with 2% calcium lactate   | Dipping               | Application of the proposed alginate coating has reduced water loss in fresh-cut cantaloupe pieces by plugging the stomata pores of cantaloupe pieces. Application of an alginate coating has significantly decreased the surface resistance of strawberry against water transfer. Application of an alginate based coating has promoted water loss in strawberries.   | [29]       |
| Peaches                             | Methyl cellulose coating was effective in extending the shelf life over alginate coating | 2 coating formulations<br>Formulation 1 - Methyl cellulose (3% w/v), ethyl alcohol and glycerol<br>Formulation 2 - Sodium alginate (2% w/v)<br>Crosslinking with 2% (w/w) CaCl <sub>2</sub> after applying the coating | Dipping               | Moisture loss in uncoated peaches was higher than in coated peaches.<br>Water vapour barrier properties of coating formulation 1 is higher than the coating formulation 2.<br>Alginate coating is better in reducing the respiration rate than methyl cellulose based coating.<br>Peaches coated with methyl cellulose exhibited the lowest decolorment in titratable acidity and it was greatest in the uncoated peaches.<br>Peaches coated with methyl cellulose exhibited the lowest textural degradation compared to control and alginate coated peaches.<br>Both coatings increased the shelf life of peaches. The maximum acceptable storage period for uncoated samples was 15 days at room temperature. It was extended by 40% for samples coated with sodium alginate and 60% for those coated with methyl cellulose. | [30]       |



Table 1 (continued)

| Food             | Best coating Composition   | Composition  | Method of application      | Main findings   | References |
|------------------|--|--|----------------------------|---|------------|
| Fresh-cut papaya | 1% Thyme essential oil based coating and 1% oregano essential oil based coating                              | sodium alginate (2%), Tween 80 (1% v/v), thyme or oregano essential oils (0.5%, 1.0%, 2.0% v/v)                                      | Dipping                    | Application of the alginate coating with essential oils has reduced the weight loss compared to application of the coating without essential oils<br>Delayed senescence of fresh cut papaya<br>Coatings containing essential oils have successfully maintained the microbiological quality of freshly cut papaya while alginate coating without essential oils has not maintained the microbial quality<br>Essential oils should be used at optimum concentrations to prevent masking of the original aroma of papaya | [31]       |
| Beef             | Coatings with basil extract  | Basil extract, sodium alginate and calcium chloride  | By submerging in solutions | Application of a coating has decreased the weight loss and increased the tenderness of beef during shelf life<br>The inclusion of basil extract in to the alginate-based edible coating has improved the acceptability of beef  | [32]       |
| Chicken          | Alginate (1% w/w), Sorbitol (1% w/w) and propionic acid (0.5% w/w) considering the shelf life of the product | Alginate (1% w/w), sorbitol (1% w/w), Essential Oil from thyme ( <i>Thymus vulgaris</i> L.) (0.5% w/w) and propionic acid (0.5% w/w) | Electric spray method      | Propionic acid and thyme essential oils were added as antimicrobial and antioxidant compounds in to the film<br>pH of meat surface, color and sensorial analysis has changed during the shelf life but has shown a similar behaviour between coatings<br>Coating containing alginate, thyme and propionic acid showed a significant reduction in weight loss compared to uncoated   | [33]       |

Table 1 (continued)

| Food | Best coating Composition   | Composition   | Method of application                | Main findings   | References |
|------|----------------------------|---|--------------------------------------|---|------------|
| Pork | SA-CMC-1.6EGCG formulation | Sodium alginate (SA), Carboxymethyl cellulose (CMC) at a volume ratio of 1:1 and Epigallocatechin gallate (EGCG) (0, 0.8, 1.2 and 1.6 g/100 mL) | Immersing in to the coating solution | Epigallocatechin gallate (EGCG) is the main component of tea polyphenols<br>Application of SA-CMC edible coating has significantly reduced the weight loss of pork samples during shelf life while the weight loss was further decreased with the increase of EGCG content<br>Microbial analysis results indicated that SA-CMC edible coating with EGCG has significantly inhibited the microbial growth and has extend the shelf life of pork<br>Results have also shown that the addition of EGCG in SA-CMC edible coating has a strong inhibitory effect on lipid oxidation of fresh pork during storage | [34]       |

decreased due to the formation of crosslinking between the carboxyl group of alginate and multivalent ions. The formation of cross-linking is a function of time and concentration. When alginate films were immersed in a multivalent ion solution, two competitive processes occur as dissolution of sodium alginate and diffusion of the multivalent ions to form cross-links. Thus, if the concentration of the solution is low, dissolution of the alginate film is prominent. Hence, an optimum concentration of the multivalent ions should be chosen. According to the results, treatment with magnesium salt has not given acceptable films at any concentration or treatment time. In zinc and iron, higher concentrations (10%) were needed to cause the insolubility of the film. They have reported that, depending on the applied ion, the tensile strength of the film will change [35]. The study done by Rhim [36] has developed an alginate-based film following a modified procedure to develop water-resistant alginate-based films with the aid of cross-linking alginate with Calcium ions. Control films in the study were without applying any  $\text{CaCl}_2$  treatment. Modified films were developed using two methods of  $\text{CaCl}_2$  treatments. The first method which is known as “mixing film” is by mixing different amounts of  $\text{CaCl}_2$  directly into the film-forming solution. The second method which is known as “immersion film” was developed by soaking the alginate films in different  $\text{CaCl}_2$  solutions. According to the results, the thickness of the film has changed according to the method. In the research, the moisture content of alginate films was observed to be different according to the preparation method. The moisture content of immersion films was significantly lower than the control films while the moisture content of mixing films was significantly higher than the control films. Thus, such a difference may be due to a change in hydrophilicity of films which intern affect the water sorption or permeation properties of films. The research has revealed that the thickness of the ‘mixed films’ was the thickest followed by the control and the ‘immersion films’. The reason behind less thickness of immersion films was solubilisation of alginate in  $\text{CaCl}_2$  solution. The results of the study revealed that the thickness of the immersion films can be increased by increasing the concentration of the  $\text{CaCl}_2$  solution. Research results also revealed that alginate films possess a higher tensile strength which is around  $33.6 \pm 3.1$  MPa. Application of  $\text{CaCl}_2$  treatment can further increase the tensile strength of these films while elongation at break has decreased by  $\text{CaCl}_2$  treatment. Hence, the highest elongation at break and the lowest tensile strength were observed in the control film. A higher tensile strength was observed in immersion films than mixing films while a higher elongation at break was observed in mixing films than immersion films. Results of the study have also revealed that the water vapour permeability of immersion films has decreased significantly while the change in water vapour permeability of mixed alginate films was not

significant.  $\text{CaCl}_2$  treatment has significantly affected the water solubility of films. Control alginate films without any treatment have completely dissolved in water. Mixing films has also disintegrated in water making it impossible to measure the water solubility of mixing and control films. The immersion films showed less water solubility. Hence, they have concluded that immersion films have improved properties than the direct addition of  $\text{Ca}^{2+}$  into the polymer blend. The study by Lim et al. [17] has developed a bio plastic film using the extract of *Sargassum siliquosum*. Statistical analysis of results of the study has shown that a bio-plastic film could be developed by using a mixture containing 2 g of extracted alginate powder and 15% w/w of sorbitol followed by the treatment with 75% w/w of  $\text{CaCl}_2$ . In the study, the authors have predicted an optimised film with a tensile strength of 33.90 MPa, elongation at break of 3.58%, water vapour permeability of  $2.63 \times 10^{-10}$  g  $\text{Pa}^{-1}$  s $^{-1}$  m $^{-1}$ , and water solubility of 33.73%. Alginate films developed incorporating cottonseed protein hydrolysates act as successful active packages for the preservation of fatty food susceptible to oxidation and microbial growth. These films have shown inhibitory effects against *Staphylococcus aureus*, *Colletotrichum gloeosporioides*, and *Rhizopus oligosporus*. A controlled and gradual diffusion of the compounds embedded in the film was observed when fatty foods were simulated. Thus, it shows that alginate films with cottonseed protein hydrolysates show promising effects as active packaging for the preservation of fatty foods [37]. Polymeric films incorporated with antimicrobial nanoparticles are also used in food packaging. Films developed with cellulose, sodium alginate, and copper oxide nanoparticles have been used to increase the shelf life of freshly cut pepper. Active packing film with cellulose nano whisker (0.5%) and Sodium alginate (3%) embedded with CuO Nanoparticles (5 mM) was successful in preventing the microbial contamination in freshly cut pepper up to 7 days [38]. The study done by Lourenco et al. [39] have utilized alginate-based edible films containing natural antioxidants from pineapple peel on beef steaks for microbial spoilage control, colour preservation, and barrier to lipid oxidation in beef steaks under the storage at 4 °C for five days. Results showed that control films without active compounds had no significant effect on decreasing the microbial load of aerobic mesophilic and *Pseudomonas spp.*, while the films containing encapsulated hydroalcoholic pineapple peel extract showed a significant inhibitory effect on microbial growth of meat at two days of storage. Alginate films containing peel encapsulated extract were effective for maintaining the colour hue and intensity of red beef meat samples. Thus, the pineapple peel antioxidants have the potential to retard lipid oxidation in meat samples. Summarised findings on alginate-based films that can be used as biodegradable packaging in the food industry are given in Table 2.

**Table 2** Studies on alginate-based films which can be used as biodegradable packaging in food industry

| Components of the film                    | Film preparation method | Plasticizer | Results   | Applications                             | References |
|---|-------------------------|-------------|---|--|------------|
| Alginate, glycerine and $\text{CaCl}_2$   | Casting                 | Glycerine   | <p>Alginate films without crosslinking with multivalent cations are transparent, pliable, not moisture resistant and readily dissolve in water</p> <p>Crosslinking of these films with multivalent cations decrease the solubility of immersion films in water</p> <p>Direct addition of multivalent ions in mixing films has created films with low strength and they dissolved in water</p> <p>Immersion of an alginate film in multivalent ion solution has resulted films with increased tensile strength and are resistant to dissolution in water</p>   | Food packaging                           | [35]       |
| Alginate powder, sago starch and sorbitol | Casting                 | Sorbitol    | <p>The sorbitol concentration in the film affect negatively on the tensile strength of the film and positively on percentage elongation at break of the film</p> <p>Water solubility of the film is influenced by <math>\text{CaCl}_2</math> treatment and sago starch concentration in the formulation. It is suggested that the effect of <math>\text{CaCl}_2</math> treatment on reduction of water solubility was highly significant when sago starch was absent from the formulation</p> <p>Best film formulation composition resulted a tensile strength of 33.90 MPa, an elongation at break of 3.58%, water vapour permeability of <math>2.63 \times 10^{-10} \text{ g Pa}^{-1} \text{ s}^{-1} \text{ m}^{-1}</math> and water solubility of 33.73%</p> | Alternative to conventional plastic bags | [17]       |

Table 2 (continued)

| Components of the film  | Film preparation method | Plasticizer | Results   | Applications  | References |
|---|-------------------------|-------------|---|---|------------|
| Alginate, glycerol, Cottonseed byproduct protein hydrolysate  | Casting                 | Glycerol    | Active packages could reduce lipid oxidation and microbial growth in packed foods by releasing the bioactive compounds during storage<br>Cotton by-product protein hydrolysate (PH) is the bioactive compound used in the study<br>Incorporation of PH has not affected on moisture content, water solubility and biodegradability of films<br>Increase in the PH concentration has increased the water vapour permeability of films<br>Increase in the concentration of Cottonseed byproduct protein hydrolysate has increased the barrier properties to visible light, film thickness and antimicrobial property<br>Good visible light barrier properties are important for food packaging since light can cause deterioration of lipid food<br>The total phenolic content and the antioxidant activity of these films has also increased with the increment in the hydrolysate concentration | Active packaging for the preservation of fatty foods        | [37]       |
| Sodium alginate (SA), copper (II) oxide nanoparticles (CuO NPs) and cellulose nano whiskers (CNW) (produced by acid hydrolysis of Microcrystalline cellulose) | Casting                 | Glycerol    | Polymeric films with antimicrobial nanoparticles can be developed for food packaging<br>The increment of the CuO NPs concentration and addition of the CNW significantly improved the antioxidant property of films<br>The active packing films comprised with CNW (0.5%)-SA (3%)-CuO NPs (5 mM) prevented the microbial contamination in fresh cut pepper up to 7 days   | Active food packaging for food industry, Freshly cut pepper | [38]       |
| Alginate, glycerol, encapsulated hydroalcoholic extract, pineapple peel juice microparticles, freeze-dried peel powder  | Casting                 | Glycerol    | Films with encapsulated hydroalcoholic peel extracts showed a good performance in the inhibition of initial aerobic mesophilic bacteria growth in meat samples, on their advanced multiplication phase  | Packaging for beef  | [39]       |

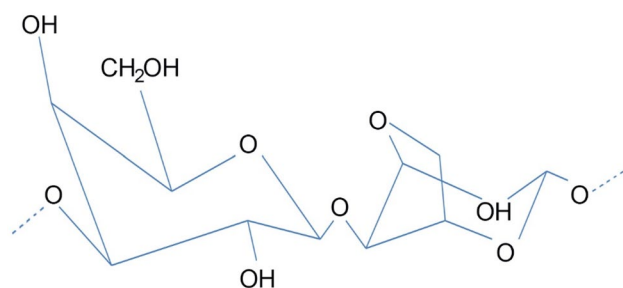


Fig. 2 Chemical structure of agar. (Source: [45])

## Agar

### Structure of agar

Carrageenan and agar can be introduced as the principal sulphated polysaccharides found in red seaweeds (Rhodophyta). The main difference to distinguish highly sulphated carrageenan from the less sulphated agar is the presence of D-galactose and anhydro-D-galactose in carrageenan and the presence of D-galactose, L-galactose, or anhydro-L-galactose in agar [40].

Agar can be introduced as, water-soluble and gel-forming polysaccharide which are the extracts of agarophyte members of Rhodophyta [41]. Agar are most commonly extracted from red seaweeds *Gelidium* and *Gracilaria* [42]. Agar is composed of agarose and agaropectin. These agarose and agaropectin differ in their physicochemical properties. Agarose can be introduced as a neutral polysaccharide and it is responsible for the gelling ability of agar. The basic repeating unit of agarose is the alternating 1,3-linked  $\beta$ -D-galactopyranose and 1,4-linked 3,6-anhydro- $\alpha$ -L-galactopyranose. Agaropectin is comprised of D-galactose, 3,6-anhydro-L-galactose. Agaropectin consists of several substituent groups such as sulphates, methyl esters, and pyruvates at different positions along the polysaccharide chain. Thus, agar is lightly sulphated [43, 44]. Agar can be extracted by alkaline treating the algal sample or without alkaline treating the algal sample. The chemical structure of agar is shown in Fig. 2.

### Extraction methods of Agar from seaweeds

#### Agar extraction without alkaline treatment

In non-alkaline treated extraction procedure, dried seaweed sample was boiled for 2 h with distilled water and filtered to separate agar from the algal residue [41]. Generally, the yield of agar extraction from *Gracilaria* species varies due to factors such as seasonal variation, environmental conditions,

extraction methods, and physiological factors [46]. It is stated that agar extraction at low temperature is responsible for lower extraction yields with higher sulphate concentrations [46]. Viscosity analysis results of agar extracted from *Gracilaria fisheri*, *Gracilaria edulis* and *Gracilaria* sp. revealed that the viscosity of the non-alkaline treated agar was higher than that of the alkaline treated agar [41]. The agar extraction yield of *Gracilaria fisheri*, *Gracilaria edulis* and *Gracilaria* sp. without an alkaline treatment are  $13.33 \pm 1.78$ ,  $10.90 \pm 0.92$  and  $39.42 \pm 0.71$  respectively [41].

#### Agar extraction with alkaline treatment

In the alkaline treated extraction procedure, the dried algal sample was incubated in 5% NaOH solution at  $80^{\circ}\text{C}$  for 2 h. Then washed to remove excess NaOH and neutralized in a 2%  $\text{H}_2\text{SO}_4$  solution for 1 h. Then, after complete eliminating of acids by washing in running tap water, agars can be extracted by boiling in water as described earlier. Finally extracted agar can be solidified and dried [41]. The agar extraction yield of *Gracilaria fisheri*, *Gracilaria edulis* and *Gracilaria* sp. with an alkaline treatment are  $39.55 \pm 7.59$ ,  $34.34 \pm 1.74$  and  $31.30 \pm 1.79$  respectively [41]. Generally, agar extracted from *Gelidium* has a low degree of substitution with low sulfate content, which will result in agars with high gel strength. On the other hand, agars from *Gracilaria/Gracilariopsis* have usually higher sulphate contents which will result in lower gelling capability [43]. Hence, the sulphate content of agar affects the gel strength. Praiboon et al. [41] have extracted agar from two Thai species of *Gracilaria* (*G. fisheri* and *G. edulis*) and one Japanese species (*Gracilaria* sp.). Results for the analysed samples have revealed that the gel strength of the alkaline treated agar is higher than the non-alkaline treated agar [41]. Since the gel strength of the extracted agar is high, it will be an advantage and will result in stronger edible films and coatings. Thus, it is better to select alkaline treated agar which developing edible films and coatings.

### Properties of agar

Generally, the physical properties and gelation of agar are related to the chemical structure of agar. Gelation is one of the most important properties of agar. Due to this property, agar can form reversible gels by cooling a hot aqueous solution [47]. Gelation of agar takes place when cooling the agar sol to a temperature below the gelation point of agar. During the gel state, agar molecule chains form a three-dimensional network by associating with each other. The basic structural units of this network are double helix [48]. Agar can be extracted by treating with NaOH or without NaOH treatment. It is reported that the gel strength of the crude extract



has increased with an increase in the concentration of NaOH treatment. Deformation of gels has decreased after application of the alkaline treatment. Decrement of deformation of gels after alkaline treatment is independent of the concentration and the incubation period in the alkaline treatment. But, gel strength is dependent on both these factors. Young's modulus of agar has also increased with the increase in NaOH concentration of the alkaline pre-treatment [49]. Agar obtained from the non-alkaline treatment has shown a higher sulphate content than agar obtained after alkaline treatment. According to a previous study, higher methoxyl content in agar was obtained from non-alkaline treated agar [49].

### Application of agar as coatings

Agar is a biopolymer with a simple extraction process. Agar is practically used as gelling, stabilizing, and encapsulating agents in pharmaceutical and biotechnological industries [50]. Additionally, previous research studies have utilized agar in development of edible coatings and films. The study "Biodegradable Agar extracted from *Gracilaria Vermiculophylla*: Film Properties and application to Edible Coating" done by Sausa et al. [50] has applied the coating developed by extracted agar on cherry and tomatoes. Results have shown that coatings made with extracted agar and glycerol have become effective in extending the shelf life of cherries and tomatoes. Agar as a polysaccharide coating agent resembles chitosan and gum Arabic for controlling postharvest diseases. Ziedan et al. [51] have developed an agar-agar-based edible coating for the management of postharvest diseases and to improve banana fruit quality. These post-harvest diseases include crown rot, neck rot, and finger rot which are mainly caused by *Colletotrichum musae* and *Fusarium moniliforme* [52]. Concentrations of 0.5, 1.0, and 2 g L<sup>-1</sup> of agar colloids were the coatings used in the study. Coating by dipping in a 2 g L<sup>-1</sup> colloid suspension for 10 min has given the most effective results while low concentrations such as 0.5 and 1.0 have become less effective since the less thickness of the coating and inability of the coating to completely cover the fruit surface. The application of agar-based coatings has shown effective in extending the storage shelf life of banana fruits by decreasing the weight loss percentage and fruit firmness loss [51]. The study by Cerqueira et al. [53] has applied coatings developed by polysaccharides from different sources for cheese. Chitosan, galactomannan from *Gleditsia triacanthos*, and agar from *Gracilaria birdiae* were the polysaccharides obtained for the study. Results reported an extensive mold growth on the surface of uncoated cheese when compared with the coated cheese. The study done by Moreno et al. [54] has developed three coating formulations with agar, alginate, and agar/alginate blend enriching with *Larrea nitida* (Ln) polyphenols. In the study, antioxidant and antiviral properties of *Zuccagnia punctata* Cav., *Larrea*

*divaricata* Cav., *Larrea cuneifolia* Cav., *Larrea nitida* Cav. (Ln) and *Tetraglochin andina* Ciald were studied. Ln extract was incorporated into the film formulation since, Ln extract has shown the most promising antimicrobial and antioxidant properties compared to other studied extracts, being a potential antiviral compound for the foodborne virus. These coatings have shown antiviral activity when applied to blueberries against murine norovirus (MNV). Thus, it is recorded that these coatings could be used as an alternative to reduce or eliminate foodborne viruses and protect the food against the oxidative processes. The coating formulation of agar and Ln extract was able to reduce the infectivity of MNV below the limit of detection after overnight incubation at 25 °C and after 4 days storage at 10 °C. A study done by Zhang et al. [55] have applied agar/sodium alginate and agar/sodium alginate and ginger essential oil based edible coatings on beef and the quality and shelf-life of fresh beef during refrigerated storage at 4 °C were studied. Microbial colony growth (total viable counts, psychrotrophs, *Escherichia coli*, *Staphylococcus aureus*, yeasts and molds), physico-chemical (thiobarbituric acid reactive substances, peroxide value), pH value, sensory evaluation, odor (electronic nose) and texture characteristics (springiness, chewiness and hardness) have analysed. Results revealed that the coating treatments significantly ( $p < 0.05$ ) retarded the oxidation of beef slices with thiobarbituric acid reactive substances and has reduced the microbial counts. Summarized findings on the application of agar-based edible coatings to improve the quality of food are given in Table 3.

### Application of agar as films

The agar/glycerol films (commercial and *Gracilaria* extracted) have shown good properties. One of the main functions of food packaging is to reduce the moisture content transferred between food and the surrounding. Agar is a hygroscopic biopolymer compared to other biopolymers such as "Sargaço",  $\kappa$ , $\iota$ -carrageenan, and commercial alginate. Therefore, agar-based films reduce moisture content migration from food and the surroundings than "Sargaço",  $\kappa$ , $\iota$ -carrageenan, and commercial alginate-based films. The oxygen permeability was identical for both commercial and *Gracilaria* extracted agar/glycerol films. The oxygen permeability of commercial and *Gracilaria* extracted agar/glycerol films were observed to be higher when compared to the synthetic polymers used in food packaging. Developed agar/glycerol films were transparent and optically clear [50]. Even though most studies utilize plasticizers in developing films, some studies report on the production of sustainable, cost-efficient, and successful food packaging films from unpurified agar extracts which contain impurities such as other polysaccharides like floridean starch and proteins which in turn exerts a plasticizing effect [56]. Wang et al.

**Table 3** Studies on application of agar-based edible coatings to improve the quality of food

| Food                             | Best coating Composition                                      | Composition  | Method of application | Main findings   | References |
|----------------------------------|---|--|-----------------------|---|------------|
| Cherry tomatoes                  | -   | Agar (1% w/w) and glycerol (15%)                                       | Dipping               | Application of the agar coating has decreased the weight loss of cherry tomatoes during storage when compared with the uncoated<br>Visual inspection of the coated and uncoated fruits revealed that the uncoated cherry tomatoes have lost their gloss whereas the coated cherry tomatoes have retained a light gloss up to the end of the shelf life  | [50]       |
| Banana ( <i>Musa acuminata</i> ) | Dipping in a 2 gL <sup>-1</sup> colloid suspension for 10 min | Agar colloid at concentration of 0.5, 1.0 and 2 gL <sup>-1</sup> (w/v) | Dipping               | All these coating formulations can suppress postharvest rot diseases of banana fruits for 10 days compared to control after infestation at 25 ± 2 °C<br>All these formulations can suppress disease incidence (%) and severity of finger rot, crown rot, neck rot and flower end rot for 15 days after infestation compared to control<br>Banana fruit coated with agar colloid at 2 g.L <sup>-1</sup> for 10 min had less weight loss (%), firmness loss (%) and soluble solid concentration (SSC) than uncoated bananas for shelf lives of 10 and 15 days<br>Scanning electronic microscopic observation of coated banana fruit surface with 2 g L <sup>-1</sup> of agar colloid for 10 min had significantly fewer cracks with a smooth surface and limited mycelia of pathogenic fungi compared to the uncoated | [51]       |

Table 3 (continued)

| Food   | Best coating Composition                                       | Composition   | Method of application | Main findings  | References |
|--------|--|---|-----------------------|--|------------|
| Cheese | Galactomannan (1.5% w/v), glycerol (2% w/v) and oil (0.5% w/v) | 3 coating formulations Chitosan, galactomannan or agar, glycerol, glycerol/sorbitol mixture and corn oil<br>Chitosan based coating contained lactic acid and Tween 80 exceptionally | Brushing the surface  | <p>Tween 80 was used in chitosan formulation to increase the wettability and to improve the compatibility between cheese surface and chitosan solution</p> <p>Galactomannan coating solution with addition of oil exhibited a good adhesion to the cheese surface</p> <p>Water vapour permeability of coatings change with the integration of sorbitol and with different concentrations of glycerol. Water vapour permeability decreases with the increase of sorbitol</p> <p>Increase in galactomannan or agar concentration in coatings resulted decrease in water vapour permeability due to stronger gel network</p> <p>Addition of oil resulted in decrease in water vapour permeability of agar and galactomannan coatings</p> <p>Agar coating displayed a decrease in the value of carbon dioxide permeability with the increase of agar and sorbitol concentrations</p> <p>Coated cheese with the best coating formulation had lower gas transfer rates, decrease in the relative weight loss and a less mold growth (visually) compared to the uncoated cheese</p> | [53]       |

Table 3 (continued)

| Food        | Best coating Composition   | Composition   | Method of application    | Main findings  | References |
|-------------|--|---|--------------------------|--|------------|
| Blueberries | Agar based formulation   | 1 g of agar, alginate or agar/ alginate mixed in a 1:1 ratio, 50 mg of <i>Larrea nitida</i> extract, 0.3 g of glycerol in 100 mL of distilled water | Dipping for 2 min        | <i>Larrea nitida</i> extract possess antimicrobial and antioxidant properties and exhibit antiviral properties against pathogens<br>These coatings can be used as antiviral edible coatings<br>Effective in eliminating murine norovirus (MNV) in artificially contaminated blueberries<br>Incorporation of the extract in to the coating formulation has reduced the transparency of the coating<br>Incorporation of this extract did not significantly modify the stretchability and stiffness of the coating films but water vapour barrier properties were significantly improved due the higher hydrophobic nature of the extract | [54]       |
| Beef        | Sodium alginate-agar and ginger essential oil containing coating | Sodium alginate (0.360 g), agar (1.440 g) and Tween 80 (0.1 g) for 100 mL of distilled water and ginger essential oil (5%v/v)                       | Immersed in the solution | Shelf life of uncoated, samples coated with agar/sodium alginate and agar/ sodium alginate containing ginger essential oil were approximately 7 days, 10 days and 16 days. Thus, the results have concluded that the shelf life extension of beef by coating with agar / sodium alginate and agar / sodium alginate containing ginger essential oil were 3-6 days and 9 days respectively compared to the uncoated sample  | [55]       |

[57] have developed and characterized an agar-based edible film reinforced with nano-bacterial cellulose which has a potential to be used as a packaging for food industry. Results have revealed that the addition of nano-bacterial cellulose at 10% level has significantly decreased the moisture content, water solubility and water vapor permeability of films and has increased the tensile strength. Phan et al. [5] have studied the functional properties of films developed using agar and starch varieties. As starch-based films, cassava starch-based, normal rice starch-based and waxy rice starch-based films have been studied. According to the results, transparent, clear, homogeneous and flexible film was obtained from an aqueous solution of 3% agar which was plasticized with glycerin. A similar appearance has been observed for the film made of cassava starch, except for its less flexibility than agar-based film. Films obtained from the normal rice starch and waxy rice starch were slightly opaque and brittle. The average thickness of the agar-based and waxy rice starch-based films were observed to be between 35 and 38  $\mu\text{m}$ , whereas that of cassava starch and normal rice starch-based films varied between 48 and 50  $\mu\text{m}$ . The authors suggested that these differences in thicknesses could be due to the differences in the viscosity and the matter concentration of the film-forming solutions. The authors have also taken Environmental scanning electron microscopic view of these films. According to the results, agar-based film micrograph has shown relatively smooth and continuous cross-section without pores or cracks, which confirms a dense and homogeneous structure while starch-based films have displayed an irregular and rough structure. On the other hand, agar-based film has displayed higher moisture sorption at the same RH conditions, which shows to have better moisture barrier properties than cassava starch-based film. Finally, they have revealed that agar-based and cassava starch-based films which were plasticized with glycerine, were transparent, clear, homogeneous, flexible, and easily handled. Thus, the results have suggested that agar-based film and cassava starch-based films, which show better functional properties, are promising systems to be used as food packaging or coating instead of normal rice starch-based and waxy rice starch-based films. Guerrero et al. [44] have studied the surface characterization of agar-based films developed by agar extracted from *Gelidium sesquipedale* (Rhodopyta). Agar-based films have been developed by incorporating different amounts (25 and 50%) of soy protein isolate (SPI) using a thermo-moulding method. Functional groups of soy protein isolates and agar were studied by Fourier transform infrared (FTIR) spectroscopy and the surface composition of agar-based films by X-ray photoelectron spectroscopy (XPS). Replacement of agar by SPI alters the surface of agar-based films, such as the orientation of the hydrophobic groups and hydrogen bonding interactions. These alterations were observed by measuring the contact angle. Changes in

orientation of the hydrophobic groups and hydrogen bonding interactions influence the functional properties of films. Thus, the study has concluded that SPI contents lower than 25% could be adequate to prepare films with good functional properties. The authors have also stated that the interactions between polysaccharides and proteins could promote the formation of a compact and reinforced three-dimensional network. Previous studies have also studied the impact of addition of essential oils, plant extracts, and nanoparticles in to edible films and coatings. In most of the time, some essential oils and nanoparticles cannot be implemented in food formula directly, therefore, as a solution for that application of such substances as a part of food packaging is becoming popular [58]. It is stated that the addition of nanoparticles can improve the mechanical and antimicrobial properties of food packaging [59]. Gold nanoparticles are becoming popular in nanopackaging due to their antibacterial characteristics as well as their inert and nontoxic nature [60]. Atef et al. [61] have characterized the physical, mechanical, and antibacterial properties of agar-cellulose bionanocomposite films incorporated with savory essential oil. Results revealed that the savory essential oil (SEO) incorporated films can be used as active packaging for foods in order to improve the shelf life and safety. Incorporation of savory essential oil has decreased tensile strength, young's modulus and water solubility and has increased the percent elongation at break of films. These films were more effective against gram positive bacteria than gram negative bacteria. De Lacey et al. [62] have developed agar-based films containing green tea extract and probiotic bacteria for extending the shelf life of fish. The results of the study has concluded that the films with green tea extract and probiotic films could extend the shelf-life of the coated fish fillets approximately for a week and it could be a way to incorporate beneficial probiotic bacteria to the fish. Da Rocha et al. [63] have studied the effects of agar-based films incorporated with fish protein hydrolysate or clove essential oil on flounder (*Paralichthys orbignyanus*) fillets shelf-life. The study has concluded that the clove essential oil can be used as a natural biopreservative to extend the flounder fillet shelf life. Summarized findings on agar-based films that can be used as biodegradable packaging in the food industry are given in Table 4.

## Carrageenan

### Structure of carrageenan

Carrageenan is found in certain red seaweeds. Carrageenan can be extracted from red algae varieties such as *Chondrus*, *Gigartina*, *Eucheuma*, and *Hypnea* [64]. They are galactose-rich polysaccharides. The composition of

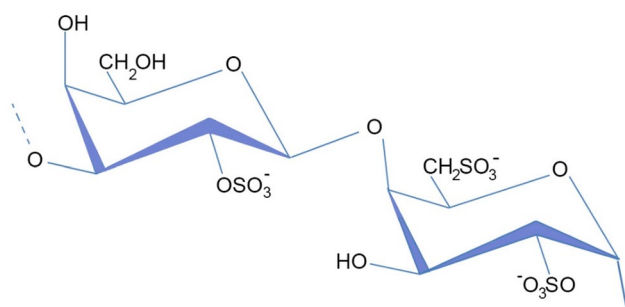
**Table 4** Studies on agar-based films which can be used as biodegradable packaging in food industry

| Components of the film                 | Film preparation method   | Plasticizer | Results  | Applications                  | References |
|--|---|-------------|--|-------------------------------|------------|
| Agar and glycerol                      | Spread the film forming solution over an acrylic plate, using an automatic knife film applicator and allowed to dry | Glycerol    | At same water activity levels, agar film are less hydrophilic when compared to commercial alginate and κ,-carrageenan film<br>Films with good moisture barrier properties can be developed using agar<br>The oxygen permeability of agar films are higher than synthetic polymer based films   | A packaging for food products | [50]       |
| Agar and glycerol                      | Casting   | Glycerol    | Agar based films with purified agar cannot be processed without incorporating a plasticizer due to excessive brittleness of agar<br>Glycerol contents lower than 30% in purified agar films has resulted very rigid and brittle films which are not optimum for food packaging<br>The unpurified extracts obtained from <i>Gelidium sesquipedale</i> seaweed were richer in other types of polysaccharides, such as floridean starch and other components, such as proteins. As opposed to the purified and commercial agars, the presence of these impurities in the extract induced a plasticization effect and the addition of glycerol was not required to obtain flexible films<br>Films with optimal mechanical performance, water barrier performance, improved resistance to high humidity conditions and added functionalities can be developed using unpurified agar based extracts without incorporating plasticizers | Food packaging                | [56]       |
| Agar based film with agar and glycerol | Casting   | Glycerol    | Heat sellable, transparent and a flexible films could be developed using agar<br>Environmental scanning electron microscopic observation of the developed agar film has revealed that the smooth, homogeneous and continuous cross sections without pores or cracks could be observed due to formation of strong and homogeneous gels before setting of the film   | Food packaging or coating     | [5]        |



**Table 4** (continued)

| Components of the film  | Film preparation method   | Plasticizer | Results  | Applications   | References |
|---|---|-------------|--|--|------------|
| Soy protein isolate, agar and glycerol                                  | Then the film forming solutions were freeze-dried and the powder was thermally compacted using a caver laboratory press to obtain films by thermo-molding process | Glycerol    | Gloss of the films increase with the increase of the agar content<br>Incorporation of agar content lower than 25% could be adequate to prepare films with good functional properties   | Soy protein and agar based films could be employed as short-term use or one-time use plastic products in food industry | [44]       |
| Agar, glycerol, glucose, green tea extract, probiotic bacteria solution | Casting   | Glycerol    | The green tea and/or probiotic film resulted a reduction in H <sub>2</sub> S-producing bacteria counts and total viable bacteria during the shelf life<br>Application of these films have also caused a decrease in total volatile basic nitrogen, trimethylamine nitrogen and pH<br>The total viable counts, H <sub>2</sub> S-producing microorganisms and total volatile basic nitrogen were maintained below the limits of acceptability during 15 days for the fillet covered with the green tea probiotic film, compared to the rest of the samples | Packaging for fish fillets   | [62]       |
| Agar, protein hydrolysate, clove essential oil and glycerol             | Casting   | Glycerol    | Application of the film was successful in maintaining the microbial, physical and mechanical properties of fillets for 15 days storing at 5 °C<br>Incorporation of protein hydrolysate has increased the water solubility, water vapour permeability, elongation at break and yellowness of films<br>Clove essential oil incorporated films were better in preserving the fish quality indices   | Packaging for fish fillets   | [63]       |



**Fig. 3** Chemical structure of carrageenan. (Source: [15])

carrageenan is comprised of water-soluble linear sulfated galactans, 3-linked  $\beta$ -D-galactopyranose, 4-linked  $\alpha$ -D-galactopyranose, or 4-linked 3,6-anhydrogalactose [65]. There are three major types of carrageenan with differences in chemical structures and properties. Those 3 types are, kappa carrageenan ( $\kappa$ -carrageenan), iota carrageenan ( $\iota$ -carrageenan) and lambda carrageenan ( $\lambda$ -carrageenan). Kappa carrageenan has 3-linked, 4-sulfated galactose and 4-linked 3,6-anhydrogalactose in its structure. Iota carrageenan has a structure analogous to kappa carrageenan, but iota carrageenan has an additional sulfate ester group on C-2 of the 3,6-anhydrogalactose residue. Lambda carrageenan is with 2-sulfated, 3-linked galactose unit, and a 2,6-disulfated 4-linked galactose unit [66]. The chemical structure of carrageenan is shown in Fig. 3.

### Extraction of carrageenan from seaweeds

During the extraction of native carrageenan, the seaweed samples are placed in bi-distilled water ( $50 \text{ mL g}^{-1}$ ), pH 7 at  $85^\circ \text{C}$  for 3 h. But, the industrial carrageenan extraction method follows an alkaline extraction process. There, seaweed samples were placed in a solution ( $150 \text{ mL g}^{-1}$ ) of NaOH (1 M) at  $80^\circ \text{C}$  for 3 h followed by neutralization to pH 8 with HCl (0.3 M). After the extraction, solutions were filtered hot, under suction, twice, through cloth and glass fibre filter. Then the filtered extract was evaporated in a vacuum to one-third of the initial volume. Finally, carrageenan was precipitated by adding the warm solution to twice its volume of ethanol (96%) [67]. Extraction of carrageenan from *Eucheuma cottonii* with and without 0.5% alkaline treatment has given a yield of 35.1 and 49.4% respectively [68]. Alkaline treated extraction of carrageenan from *Hypnea bryoides* has given a yield of 33% [69]. Mostly, carrageenan is precipitated by adding alcohol. 2-propanol is the commonly used. Kappa-type carrageenan can alternatively be precipitated as gelled fibers by spraying the solution into a cold 1–13% KCl [68].

### Properties of Carrageenan

Carrageenan is identified as a sulphated polygalactan. The ester-sulfate content of carrageenan is up to 15 to 40% and the average relative molecular mass can be above 100 kDa. Carrageenan is grouped into different types as  $\lambda$ ,  $\kappa$ ,  $\iota$ ,  $\epsilon$ , and  $\mu$  considering the solubility in potassium chloride. Factors that affect for properties of carrageenan type are the number and position of ester sulphate groups and 3,6-anhydrogalactose content present. Higher levels of ester sulphate content in carrageenan type reflect lower solubility and lower gel strength of carrageenan type [70]. Commercial carrageenan have shown that sulphate ester group content of  $\kappa$ -carrageenan,  $\iota$ -carrageenan, and  $\lambda$ -carrageenan can be 25–30%, 28–38%, and 32–39% respectively. However, differences in sulphate contents can occur due to changes in seaweed species and batch [71]. It was reported that  $\lambda$ -Carrageenan is readily soluble in cold or hot aqueous solutions while  $\kappa$  carrageenan is soluble in hot solutions.  $\kappa$ -carrageenan precipitates when an aqueous solution is treated with potassium ions [70]. Generally, carrageenan forms highly viscous aqueous solutions. The viscosity of the solution depends on factors such as temperature, concentration, presence or absence of other solutes, type of carrageenan, and molecular weight. Viscosity increases with the concentration and decreases with temperature. Depolymerisation of carrageenan occurs during acid catalysed hydrolysis and complete loss of carrageenan functionality occurs at elevated temperatures and low pH values [70]. When proper conditions were given, aqueous solutions of  $\kappa$ -carrageenan and  $\iota$ -carrageenan form thermo-reversible sol-gels. But this gelation does not take place in  $\lambda$ -carrageenan due to the presence of more electrolyte groups. Carrageenan is heavily used in the food industry as gels or viscous agents [72].

### Application of carrageenan as coatings

Previous studies have mentioned that the usage of carrageenan as a food additive dates back to the 15th century [73]. Application of Carrageenan-based edible coatings and films could be possible for foods such as fresh and frozen meat, ham or sausages casings, oily foods, dry solid foods, in soft capsules, and non-gelation capsules [74]. Banu et al. [75] have studied the effect of the application of edible coatings by the extracts of *Kappaphycus alvarezii* and *Sargassum tenerrimum* on quality characteristics and shelf life of tomato (*Lycopersicon esculentum mill*). The study focused on the factors such as weight loss, texture, ascorbic acid content, juice content, total acidity of the uncoated (control) and coated tomatoes during the storage period [75]. According to the results, coating by 3% *K. alvarezii* extract was more effective in maintaining the weight of the tomatoes. Texture losses can be seen prominently during the prolonged storage

of tomatoes. Thus the results of the study revealed that the texture of the coated tomatoes was firm and fresh than the control. It was also noticed that the increase in coating concentration has increased the firmness. Tomatoes coated with 3% *K. alvarezii* extract were effective to maintain the firmness of the fruit and increased the shelf life for 28 days. Tomatoes coated with 3% *K. alvarezii* extract were also effective in maintaining the highest juice content and lowest reduction in total acidity during the shelf life of 28 days. From the results, it was also observed that there was a drastic decrease in the ascorbic acid content of tomatoes during the shelf life. The research results have found that the effectiveness of the retention of ascorbic acid content during the storage life was considerably high when tomatoes were coated with 3% *K. alvarezii* extract. Similarly, the application of 3% of *S. tenerrimum* extract coating also showed a higher retention in ascorbic acid during the storage period than the control sample [75]. Research on the application of carrageenan as an alternative coating for papaya has been conducted by Hamzah et al. [76]. Papaya is a climacteric fruit. Hence, it continues to ripen throughout the post-harvest life. A lot of changes take place in fruits during ripening. Though these changes are important, an uncontrolled ripening process can lead to rapid degradation of the quality of papaya leading to a high post-harvest loss. Thus, it is important to control the ripening process during storage and distribution to supply the best quality papaya to consumers. At present, a variety of post-harvest treatments such as hot water treatment, modified or controlled atmosphere packaging, and gamma irradiation have been applied to maintain the post-harvest quality of papaya. Another technique that is gaining attention as a post-harvest treatment for fruit including papaya is the application of edible coatings. Hence, Hamzah et al. [76] have studied the application of carrageenan as an alternative coating for papaya. They have also optimized the carrageenan-based edible coating by response surface methodology. For the optimization, carrageenan concentration and glycerol concentration have been used as the two independent variables and the optimization study was carried out using a two-factor central composite design (CCD). Through optimization, they have predicted the levels of carrageenan concentration and glycerol concentration that would give maximum firmness and lowest colour components. According to the results, carrageenan concentration contributed more significantly to the fruit firmness and glycerol contribute more towards the changes in colour components. The best coating formulation was found as carrageenan and glycerol levels at 0.78% (w/v) and 0.85% (w/v) respectively. Application of carrageenan-based edible coating on papaya has resulted in higher firmness and lower colour component value ( $L^* a^* b^*$ ) indicating a delay in ripening and extension of the shelf-life of coated papaya. Ribeiro et al. [77] have studied the ability of starch, carrageenan, and chitosan-based

coatings to extend the shelf life of strawberries. According to the results of the study, strawberry has a low energy surface. Strawberry surface has a surface tension of 28.94 mN/m, and it has a polar and a dispersive component of 5.95 and 22.99 mN/m, respectively. The critical surface tension of the strawberry surface is 18.84 mN/m. Analysis results after the application of coating solutions revealed that oxygen permeability and opacity of carrageenan coating were significantly lower than that of starch coating. It has been confirmed that the addition of calcium chloride to coatings decreases the microbial growth rate of the fruit, the minimum rate of microbial growth was obtained with strawberries coated with chitosan and calcium chloride. The minimum loss of firmness was observed in strawberries applied with carrageenan coating and calcium chloride. The minimum weight loss was observed in strawberries coated with chitosan and carrageenan coatings, both with calcium chloride. The study done by Khojah [78] has developed a coating comprising of fish gelatin, k-Carrageenan, and extracts obtained from pomegranate peels for preserving the fish filets. Gelatin extracted from blue tilapia (*Oreochromis aureus*) was microencapsulated with k-Carrageenan loaded with different concentrations of 0.5%, 1.0%, 1.5%, and 2.0% pomegranate peel extract. Carrageenan : Gelatin weight ratio of 1:2 was maintained during the entire experiment by taking a carrageenan concentration of 0.75% w/v and a gelatin solution of 1.5% w/v. 1% of microcapsule powder was used for coating. Generally, within the storage microbial counts tend to increase in fish. Application of gelatin/carrageenan/pomegranate peel extract-based coating could effectively reduce the microbial counts of total aerobics, psychrotrophs, yeast and molds, and enterobacteriaceae groups throughout 30 days of refrigerated storage. The count decrease was dependent on the concentration of pomegranate peel extract in the coating formulation. The analysis of spoilage chemical parameters such as total volatile basic nitrogen, peroxide value, and thiobarbituric acid reactive substances in coated samples indicated that application of the coating based on the given components could hinder the increase of their values during storage. Finally, all the coated samples were judged as “acceptable” for consumption, up to the 30th day of storage by the sensory analysis. Tran et al. [79] have developed a composite film combining alginate, kappa carrageenan, glycerol, and gac pulp (*Momordica cochinchinensis*). Gac pulp is a rich source of pectin, essential oil, and carotenoids thus, this material could be a good source to be incorporated in composite films. Gac pulp, sodium alginate, kappa-carrageenan affect the thickness and tensile strength of films. They have found that sodium alginate and carrageenan significantly affect the physical and mechanical properties of the film, while Gac pulp significantly affects the colour parameters and physical properties. The suggested optimum formulae is 1.03% w/v, 0.65% w/v, 0.4% w/v and

**Table 5** Studies on application of carrageenan-based edible coatings to improve the quality of food

| Food  | Best coating composition                     | Composition   | Method of application | Main findings   | References |
|---|--|---|-----------------------|---|------------|
| Tomato ( <i>Lycopersicon esculentum</i> mill) | Coating with 3% <i>Kappaphycus alvarezii</i> | 1%, 2%, and 3% extracts of <i>Kappaphycus alvarezii</i> and <i>Sargassum tenerrimum</i>   | Coated                | Seaweed extracts can successfully improve the shelf life and maintain the quality of tomatoes<br><i>Kappaphycus alvarezii</i> and <i>Sargassum tenerrimum</i> extracts used in the study are comprised of phytochemical constituents such as alkaloids, phenolic compounds, flavonoids, terpenoids and tannin<br>Both these extracts possess antibacterial activity<br><i>K. alvarezii</i> extract exhibit a higher antibacterial effect compared to <i>S. tenerrimum</i> extract at equal concentration levels<br>Application of any of these coating gave better quality results for texture, total soluble solid content, total acidity, total acid content and juice on 28th day of storage compared to uncoated tomatoes   | [75]       |
| Papaya  | 0.78% (w/v) carrageenan with glycerol        | Carrageenan (0.2–0.8%, w/v), Glycerol (0–1%, w/v) and citric acid to adjust the pH to 5.6 | Dipping for 2 min     | Papaya is a climacteric fruit<br>With the ethylene climacteric peak, softening, aroma development, changes in peel and changes in pulp colour can be observed<br>Carrageenan based coatings possess good gas barrier properties and has the ability to retard the moisture loss by acting as sacrificing agents<br>Application of the best formulation determined through multiple response optimization analysis as a coating, has given a higher firmness and lower colour component values extending the shelf life by retarding the postharvest quality deterioration of papaya<br>Application of the coating reduce oxygen permeability which influence in reduction of respiration rate and thereby delay ripening, chlorophyll degradation, and hydrolysis activities of softening | [76]       |

Table 5 (continued)

| Food         | Best coating composition  | Composition   | Method of application | Main findings  | References |
|--------------|---|---|-----------------------|--|------------|
| Strawberry   | Carrageenan coating (Carrageenan 0.3% (w/v), Glycerol 0.75% (w/v), Tween 80 (between 0.01 and 0.1% (w/v)) with CaCl <sub>2</sub> was the best considering the external firmness of fruits | 3 coating formulations<br>Starch based (starch, citric acid, sorbitol)<br>Carrageenan based (Carrageenan, citric acid, glycerol, tween 80)<br>Chitosan based (chitosan, HCl, NaOH, Tween 80)<br>From the above formulations, 6 formulations were studied either by adding CaCl <sub>2</sub> or without adding CaCl <sub>2</sub> | Spraying              | The smallest loss of weight was obtained with chitosan and carrageenan coatings, both with calcium chloride<br>The soluble solids percentage differences between coated and uncoated fruit were statistically not significant<br>During the shelf life, carrageenan-coated fruit appeared to have a more glossy/shiny surface than others<br>The industrial application of the carrageenan coating with calcium on fresh strawberry fruit will improve the external firmness of the fresh strawberries   | [77]       |
| Fish fillets | Coating with 2% pomegranate peel extract  | Fish gelatin, k-carrageenan (and Extract of Pomegranate Peels(0.5%, 1.0%, 1.5% and 2.0% w/v)<br>pH was set to 3.6 using 2.5% (v/v) acetic solution  | Dipping               | The physicochemical attributes, gel network formation, thermal and mechanical properties of fish gelatin has enhanced by mixing k-carrageenan in coating formulation<br>Pomegranate peel extract contains elevated amounts of bioactive polyphenolic compounds and possess antimicrobial activity, anti-inflammatory, prebiotic activities, anti-cancer activity, and anti-diabetic activities<br>Incorporation of pomegranate peel extract at concentrations 1.5% and 2.0% has completely inhibited psychrotrophic bacteria, yeast, molds and enterobacteriaceae groups during the shelf life<br>Coating has acted as an oxygen barrier which hinder the microbial activities by preventing oxygen penetration into microbial cells<br>Antimicrobial property of the coating has influenced in preventing bacterial decomposition of protein and oxidative deamination of non-protein nitrogen in fish fillets<br>Oxygen barrier property and antioxidant property of the coating prevent fat oxidation in fish fillets<br>Application of the coating has resulted in prolongation of the refrigerated storage of coated fish fillets up to 30 days | [78]       |

Table 5 (continued)

| Food         | Best coating composition   | Composition   | Method of application               | Main findings  | References |
|--------------|--|---|-------------------------------------|--|------------|
| Chicken meat | Konjac glucomannan (0.5%), carrageenan (0.5%), camellia oil (3.5% w/v) and Tween 80 (10%, w/w of camellia oil) | Konjac glucomannan (0.5%), carrageenan (0.5%), Glycerol (30%), camellia oil (0%, 2.0%, 2.5%, 3.0% and 3.5% w/v) and Tween 80 (10%, w/w of camellia oil) | Immersed in the coating formulation | Best coating composition was successful in extending the shelf-life of chicken meat up to 10 days at refrigerated storage (4 °C) compared to control samples | [82]       |

0.85% w/v of sodium alginate, kappa-carrageenan, Gac pulp and glycerol respectively. It is stated that the films prepared under these optimum conditions are suitable to be used as a food coating. There are also previous studies which used carrageenan based coatings with ZnO nano particles to inhibit the growth of food pathogens like *E.coli* and *S. aureus* in mangoes in order to preserve the shelf life of mangoes. Application of carrageenan and ZnO nanoparticle based coating has resulted in reducing total acidity, maintain firmness and delay the discoloration and decay [80, 81]. Zhou et al. [82] have developed a konjac glucomannan/carrageenan based edible coating incorporating camellia oil to evaluate the effect of the application of the coating on quality and shelf life of chicken meat. The results revealed that the application of the coating formulation is successful in extending the shelf life of chicken meat by decreasing the weight loss, pH, thiobarbituric acid reactive substance, total volatile nitrogen and microbial counts when compared to uncoated samples. Summarized findings on the application of carrageenan-based edible coatings to improve the quality of food are given in Table 5.

### Application of carrageenan as films

Karbowiak et al. [74] have studied the wetting properties at the surface of iota-carrageenan-based edible films and the structural and functional properties of those films. The film composition was comprised of 3% of Carrageenan, 30% of anhydrous glycerol, fat, and Glycerol monostearate. Three different concentrations of fat as 30%, 60%, and 90% (w/w total solid basis) were studied. In conclusion, it is stated that charged polysaccharides such as carrageenan, which possesses a strong electrolyte character due to their sulphate groups can interact with other film components such as lipids and additives and the resulting macromolecular network after gelation affect the water barrier properties of the film. Tye et al. [71] have studied the preparation and characterization of native seaweed, alkaline modified, and unmodified carrageenan-based films. In the study, physical, mechanical, water vapour barrier properties and thermal properties of carrageenan-based films were studied. Further, variations of carrageenan after chemically modifying were determined using Fourier Transform Infrared Spectroscopy (FTIR) analysis. According to the results, unmodified carrageenan is suitable to produce biopolymer films with the best mechanical properties such as films with high tensile strength and Young's modulus while chemically modified Carrageenan-based films have exhibited better thermal stability and water vapour barrier property. Chemically modified carrageenan-based films are less hydrophilic as compared to the native seaweed and unmodified carrageenan films. FTIR analysis shows that alkali modification of the carrageenan has removed the hydroxyl and sulphate



ester groups in the carrageenan structure and has formed 3,6-anhydrogalactose, which will result in improving the flexibility of the film. Variation of extraction temperature will also cause significant changes in the properties of films. According to the results, an increase in extraction temperature has resulted in reducing the tensile strength and Young's modulus, but it has improved the elongation at break and hydrophobicity. Thus, it shows that the chemical modification of carrageenan can enhance all the properties of the film provided that an appropriate extraction temperature was selected. Nur et al. [83] have developed kappa-carrageenan-based films from *Eucheima cottoni* incorporated with various types of plant oils such as olive oil, corn oil, soybean oil, and sunflower oil. Films were developed by dissolving 1% (w/v) of kappa-carrageenan in distilled water and incorporating 50% of glycerol (w/w based on carrageenan powder weight), plant oil (1–3% v/v) and Tween 80 as an emulsifier in quantities proportional to the plant oil. Films developed by incorporating lipids were thicker than the films developed without incorporating lipids and the thickness increased with the increase in oil concentration. Pérez-Mateos et al. [84] stated that an increase in thickness of films with the increase in oil concentration may be due to conformational changes in the film matrix. According to previous research, carrageenan-based films have exhibited good gas barrier properties but, those films possess high water permeability due to their hydrophilic nature. Hence, the high water permeability of these films limit their application as food packaging in the food industry [85]. This incorporation of plant oils has improved the water vapour permeability of films by significantly reducing the moisture content and solubility of films in water. The observed decrease in moisture content of films incorporated with plant oils may be due to a decrease in the hydrophilic nature of carrageenan films since the hydroxyl groups present in carrageenan interact with the plant oils. Polysaccharide and oil interact and form non-covalent bonds between the hydroxyl group of carrageenan and plant oils, which in return would reduce the hydroxyl group availability for interaction between carrageenan and water molecules, which would consequently lead to a more water-resistant film [86]. The incorporation of plant oils will also affect the plasticizing effect of films. Among the developed films, the carrageenan-based film developed by incorporating olive oil was the best film with the best water barrier and mechanical properties [83]. Manuhara et al. [7] have developed a semi-refined kappa carrageenan-based edible film for nano-coating application on minimally processed food. The film solution was prepared by dissolving 1, 1.5, and 2 g of carrageenan in 100 mL of distilled water and incorporating sorbitol (1% v/v) as a plasticizer at 60 °C. When the carrageenan concentration was increased in the film formulation, film thickness and tensile strength have increased but elongation at break and water vapor transmission rate (WVTR)

has decreased. Thus, the recommended formulation was the formulation with 2% carrageenan. 2% carrageenan-based film demonstrated a thickness of 0.053mm, tensile strength of 21.14 MPa, Elongation at break (%) of 12.36%, and a water vapour transmission rate of 9.56 g/h.m<sup>2</sup>. The study done by Simona et al. [87] has developed films comprised of carrageenan, orange essential oil, trehalose, glycerol, and Tween 20 or Tween 80 in different proportions. If these films were used to pack dried fruits, trehalose in the film will assist in preserving the aroma and colour. Essential oils in the film will enhance the antioxidant property and will provide an antimicrobial effect to the food. It is stated that both these orange essential oil and trehalose possess UV protective properties. As the final outcome, the research showed that the combination of orange essential oil and trehalose in carrageenan-based films can develop films with good barrier protection against UV-VIS radiation. This property is essential to reduce lipid oxidation in foods. The barrier protection against UV-VIS radiation increases with the increase of the concentration of trehalose and orange essential oil. These films have also shown resistance to Gram-positive bacteria (*Staphylococcus aureus*). The study done by Farhan et al. [88] have used a semi refined kappa carrageenan based active edible film developed incorporating water extract of germinated fenugreek seeds on chicken breast. Results have revealed that the films have a good capability to control the microorganism growth on the surface of chicken breast samples. Martiny et al. [89] have studied the effect of carrageenan films with olive leaf extract for lamb meat preservation. Incorporation of olive oil extract has increased the thickness and water vapour permeability of the film and has decreased the tensile strength of the film. Incorporation of olive leaf extract has successfully decreased the growth rate of psychrophiles in packaged lamb meat. Summarized findings on carrageenan-based films which can be used as biodegradable packaging in the food industry are given in Table 6.

### Advantages of usage of seaweed-based coatings and films in the food industry

At present, the application of edible films as well as edible coatings have received more consideration as promising methods to enhance the shelf life and quality of various food products. Edible coatings and films provide various functions in foods such as physical protection, moisture, and gas inhibition, carriers of many active compounds such as antioxidants, probiotics, antimicrobial, and flavouring agents [90–93].

Alginate, agar, and carrageenan are seaweed-derived natural food ingredients that are generally regarded as safe materials to be used in food. Therefore, alginate, agar, and carrageenan can be used in formulations of edible films and

**Table 6** Studies on carrageenan-based films which can be used as biodegradable packaging in food industry

| Components of the film  | Film preparation method | Plasticizer | Results  | Applications   | References |
|---|-------------------------|-------------|--|--|------------|
| Carrageenan, glycerol, glycerol monostearate and fat  | Casting                 | Glycerol    | Surface properties of films differs with the nature of the used additives<br>Fat was used to improve the moisture barrier properties of the film<br>Contact angle can be used to determine the hydrophobicity of a film. Large contact angle represent a hydrophobic surface while a small contact angle represents a hydrophilic surface  | Fresh and frozen meat<br>Fish<br>Ham or sausage casings<br>Dry foods | [74]       |
| k-Carrageenan, glycerol, plant oils (corn oil, soybean oil, olive oil and sunflower oil) and tween 80 | Casting                 | Glycerol    | Hydrophilic nature of carrageenan which result films with high water permeability limit their application in food packaging. Oils are hydrophobic in nature. Therefore incorporation of oils in to film formulation enhance the moisture barrier property of carrageenan films<br>Incorporation of plant oils has increased the film thickness and values for elongation at break while the moisture content, solubility, opacity and tensile strength of films has decreased significantly. Plants oils will also contribute to the plasticizing effect of these films<br>Incorporation of olive oil gave the best film properties compared to the other oils | Food packaging   | [83]       |
| Carrageenan and Sorbitol  | Casting                 | Sorbitol    | When the carrageenan concentration increased, thickness and tensile strength of carrageenan based edible films increased while the elongation at break and water vapour transmission rate decreased  | Food packaging   | [7]        |
| Pectin, k-carrageenan and organically modified nano-clays   | Wet casting method      | -           | Inclusion of nanoclays has effected positively on enhancing the barrier properties to water vapour and CO <sub>2</sub> of a polymeric matrix composed by kappa-carrageenan and pectin<br>Water vapour permeability of films was reduced by 35% of its initial value at 10% nanoclay content under high RH conditions<br>Permeability for CO <sub>2</sub> was reduced by 50% for 1% nanoclay content  | Food packaging with enhanced barrier properties                      | [85]       |

Table 6 (continued)

| Components of the film  | Film preparation method | Plasticizer | Results  | Applications                          | References |
|---|-------------------------|-------------|--|---------------------------------------|------------|
| kappa-carrageenan, glycerol, <i>Satureja hortensis</i> essential oil (SEO) and Tween 80 | Casting                 | Glycerol    | <i>Satureja hortensis</i> essential oil (SEO) possesses antioxidant and antimicrobial characteristics<br>Films with SEO had lower moisture content compared to films without SEO<br>Films with SEO had lower tensile strength and water vapour permeability compared to films without SEO.<br>Water vapour permeability reduced with the increase of SEO concentration<br>Increase of essential oil concentration has also reduced the transparency of the films<br>Research revealed that Carrageenan films incorporated with SEO have a good potential to be used as an active biodegradable packaging material that controls food pathogens<br>SEO incorporated films have exhibited inhibitory effect on <i>Staphylococcus aureus</i> , <i>Bacillus cereus</i> , <i>Escherichia coli</i> and <i>Salmonella typhimurium</i> . | Films and coatings for food           | [86]       |
| Carrageenan, Trehalose, orange essential oil, glycerol and Tween 20 or Tween 80         | Casting                 | Glycerol    | Research revealed that the combination of trehalose with orange essential oil improved the barrier protection against UV-VIS radiation and the protection increases with the increment in trehalose and orange essential oil concentration. Experimentally produced packaging has also shown the greatest antimicrobial activity against <i>Staphylococcus aureus</i><br>The compatibility of trehalose in the film matrix was better in films with 0.3 g of carrageenan than with 0.5 g of carrageenan  | Fruits Vegetables<br>Edible packaging | [87]       |
| Semi refined carrageenan, sorbitol, water extract of germinated fenugreek seeds         | Casting                 | Sorbitol    | Application of the film resulted in extending the shelf life of chicken breast by improving the microbial quality  | Chicken breast                        | [88]       |
| Carrageenan, glycerol, olive leaf extract   | Casting                 | Glycerol    | Incorporation of olive leaf extract in to films has imparted an antimicrobial capacity during the storage of lamb meat   | Lamb meat                             | [89]       |

coatings in functional food without any safety risk [94]. Generally, food additives like, antimicrobials, probiotics, antioxidants, vitamins, and flavour compounds that are incorporated into foods are highly sensitive to external factors such as gases (oxygen), temperature, and light. Thus, the application of a coating developed using alginate, agar, or carrageenan would act as a shield and protect the food product against losing these valuable compounds due to harsh environmental conditions [94]. Alginate, agar, and carrageenan are highly hydrophilic, films out of these polysaccharides can act as barriers to oxygen, protecting the food against lipid oxidation. The quality loss of packed food is commonly caused by oxygen [94]. Oxygen is responsible for degradation processes in foods such as lipid oxidation, enzymatic browning, microbial degradation, etc. Lipid oxidation causes off-flavour, off-colour, and nutrient loss in foods. Therefore, the application of an edible coating or a film with good oxygen barrier properties could effectively reduce these degradation processes in foods. Additionally, the effect of oxygen on food could also be delayed with the incorporation of antioxidant agents, such as ascorbic, and citric acids into coating and film formulations of alginate, agar, or carrageenan [95]. Another issue in the food industry is the incompatibility of the solubility of food additives and food which result in phase separation and precipitates in the food. Usage of cinnamon leaf oil as a preservative in food causes phase separation is an example of such an issue. Encapsulation of such kinds of additives with edible coating matrices developed using seaweed-based polysaccharides is an important method that can be equipped to improve the solubility of insoluble additives. Moreover, one of the most attractive advantages of the use of alginate, agar, and carrageenan in edible coatings is the ability to control the release of encapsulated bioactive compounds by simple parameters such as humidity, temperature, changes in pH, and mechanical rupture of the matrix. This control of the release of the additive at a given point is crucial to improve quality and ensure food safety and functionality [96, 97]. Seaweed-based coatings are considered as less expensive packaging materials which can be used in the food industry. These coatings and films can improve the aesthetic appearance of a food product by minimizing the impact of physical damage, hiding scars, and improving surface shine. Other than that, the application of proper packaging in fruits and vegetables plays a main role in protecting the harvested products, minimizing respiration rate and ripening, reducing ethylene, eliminating microbial activities, and controlling water loss [98]. Seaweed-polysaccharide based coatings and films can act as moisture barriers when applied on the surface of fruits and vegetables which prevent weight loss during postharvest storage. Other than that, alginate, agar, and carrageenan coatings and films act as gas barriers for controlling gas exchange between the fresh produce and its surrounding

atmosphere, which would slow down respiration rate, deterioration, enzymatic oxidation, browning discoloration, and texture softening during storage. Application of alginate, agar, and carrageenan coatings or films will also restrict the exchange of volatile compounds between the fresh produce and its surrounding environment which prevents the loss of natural volatile flavor and color compounds from fresh produce and the acquisition of foreign odours [97].

### Limitations of usage of seaweed-based coatings and films in the food industry

There are certain limitations of using seaweed hydrocolloids in the development of edible coatings and films. One limitation is that it is difficult to optimize and control the production of agar, carrageenan, and alginate since the yield of those hydrocolloids are highly dependent on the origin, geographical location, and harvesting season of the seaweed source. Additionally, extraction variables such as temperature, extraction time, seaweed source, particle size, pH, and concentrations of solvents affect the hydrocolloid yield and their rheological properties [99]. The extraction process of these hydrocolloids with an optimum yield and quality requires large quantities of chemicals such as formaldehyde, potassium chloride, sodium hydroxide, potassium hydroxide, etc. Overflows of pollutants and effluents generated during extraction steps with these chemicals due to the lack of control throughout the whole production process could pose a serious threat to human health and the environment [99]. Costs involved in the extraction processes are another issue. As an example, precipitation of alginate and carrageenan requires large quantities of ethanol. Ethanol is a highly expensive chemical. Therefore, a high cost is involved in the extraction process of alginate and carrageenan [99]. Another negative point is that most of these films and coatings developed using alginate, agar, and carrageenan suffer due to their high sensitivity to moisture, and other stress factors such as, high pressure, ultrasound, microwave radiations, electric field, and other radiations. Therefore, commercial applications of these films and coatings have become limited [8].

### Conclusions

Alginate, agar, and carrageenan are polysaccharides that are extracted from marine algae. Under proper conditions, these extracts can form gels. Gel formation of alginate is mainly governed by its ability to selectively binding to multivalent cations, while the gel formation ability of agar and carrageenan are related to their chemical structure. These extracts can be successfully utilized for the development of edible coatings and edible films to be utilized

as packaging in the food industry. The principle behind the formation of films and coatings using these extracts is their gel-forming ability. These coatings and films can be applied to fruits, vegetables, dairy products, meat-based products, and some other dry food items. Application of seaweed-derived coating on fruits has shown positive results on post-harvest quality of fruits. When these coatings were applied on fruits, decrements in weight loss, decrements in post-harvest diseases due to microbial growth, decrements in textural losses, decrements in acidity losses, and colour changes have observed most of the time. Thus, the application of a coating on fruits can successfully delay the ripening process of climacteric fruits enabling extension of the post-harvest shelf life. When these coatings were applied on some other food items such as cheese and beef, it will reduce the microbial growth within the shelf life.

Films can be used to partially substitute non-biodegradable polymer-based packages utilized in the food industry. They can be commonly used as wrappers, or separation layers or can be formed into casings, capsules, and tiny pouches. By incorporating Ca<sup>2+</sup> divalent cations, alginate films with higher tensile strength could be obtained. No such ions were used in developing agar or carrageenan-based film. However, commercialization of these films and coatings made out of natural polymers is limited due to their high sensitivity to moisture, and other stress factors such as high pressure, ultrasound, microwave radiations, electric field, and other radiations. Therefore, it is suggested to conduct further research on improving the commercial quality of edible films and coatings to expand the commercial applications.

**Acknowledgements** This review article is based on the work supported by University Research Grants [Grant No. ASP/01/RE/SCI/2019/16], University of Sri Jayewardenepura, Gangodawila, Nugegoda, Sri Lanka.

**Author contributions** All authors contributed to the study's conception and design. The original draft of the manuscript was written by MMJ. Reviewing was done by MPGV and IW.

**Funding** This work was supported by the University Research Grants, University of Sri Jayewardenepura, Gangodawila, Nugegoda, Sri Lanka [Grant No. ASP/01/RE/SCI/2019/16].

**Availability of data and material** Not applicable.

**Code availability** Not applicable.

## Declarations

**Conflict of interest** The authors declare that there are no conflicts of interest regarding the publication of this paper.

**Ethics approval** Not applicable.

**Consent to participate** Not applicable.

**Consent for publication** Not applicable.

## References

1. S. Muizniece-Brasava, L. Dukalska, I. Kantike, In: The 6th Baltic Conference on Food Science and Technology, pp. 187–192 (2011)
2. B. Hassan, S.A. Chatha, A.I. Hussain, K.M. Zia, N. Akhtar, *Int. J. Biol. Macromol.* **109**, 1095–1107 (2018)
3. A. Gennadios, *Protein-based films and coatings* (CRC Press, Boca Raton, 2002)
4. M. Iñiguez-Moreno, J.A. Ragazzo-Sánchez, M. Calderón-Santoyo, *Polymers.* **13**, 3271 (2021)
5. T.D. Phan, F. Debeaufort, D. Luu, A. Voilley, *J. Agric. Food Chem.* **53**, 973–981 (2005)
6. J.H. Han, *Innov. Food Packag.* (2014). <https://doi.org/10.1016/B978-0-12-394601-0.00009-6>
7. G.J. Manuhara, D. Praseptiangga, D.R. Muhammad, B.H. Maimuni, A.I.P. Conf, Proc. (2016). <https://doi.org/10.1063/1.4941509>
8. E. Tavassoli-Kafrani, H. Shekarchizadeh, M. Masoudpour-Behabadi, *Carbohydr. Polym.* (2016) doi:<https://doi.org/10.1016/j.carbpol.2015.10.074>
9. M.M. Jayakody, M.P.G. Vanniarachchy, I. Wijesekara, *Journal of Tropical Forestry and Environment.* **9**, 93–100 (2019)
10. M.M. Jayakody, M.P.G. Vanniarachchy, W.L.I. Wijesekara, *Vidyodaya Journal of Science.* (2021) <https://doi.org/10.31357/vjs.v24i01.4963>
11. S. Galus, E.A. Arik Kibar, M. Gniewosz, K. Kraśniewska, *Coatings.* **10**, 674 (2020)
12. V. Falguera, J.P. Quintero, A. Jiménez, J.A. Muñoz, A. Ibarz, *Trends Food Sci. Technol.* **22**, 292–303 (2011)
13. R.K. Dhall, *Crit. Rev. Food Sci. Nutr.* **53**, 435–450 (2013)
14. M. Iwamoto, M. Kurachi, T. Nakashima, D. Kim, K. Yamaguchi, T. Oda, Y. Iwamoto, T. Muramatsu, *FEBS Lett.* (2005). <https://doi.org/10.1016/j.febslet.2005.07.007>
15. J. Venkatesan, B. Lowe, S. Anil, P. Manivasagan, A.A. Kheraif, K.H. Kang, S.K. Kim, *Starch-Stärke.* (2015). <https://doi.org/10.1002/star.201400127>
16. L. Pereira, S.F. Gheda, P.J. Ribeiro-Claro, *Int. J. Carbohydr. Chem.* (2013) <https://doi.org/10.1155/2013/537202>
17. J.Y. Lim, S.L. Hii, S.Y. Chee, C.L. Wong, *J. Appl. Phycol.* (2018) <https://doi.org/10.1007/s10811-018-1603-2>
18. S.Y. Chee, P.K. Wong, C.L. Wong, *J. Appl. Phycol.* (2011) <https://doi.org/10.1007/s10811-010-9533-7>
19. N.S. Basha, R. Rekha, A. Letensie, S. Mensura, *J. Sci. Res.* (2011) <https://doi.org/10.3329/jsr.v3i3.6770>
20. A. Mohammed, R. Bissoon, E. Bajnath, K. Mohammed, T. Lee, M. Bissram, N. John, N.K. Jalsa, K.Y. Lee, K. Ward, *Carbohydr. Polym.* (2018) <https://doi.org/10.1016/j.carbpol.2018.06.067>
21. S.N. Pawar, K.J. Edgar, *Biomaterials.* (2012) <https://doi.org/10.1016/j.biomaterials.2012.01.007>
22. S.N. Pawar, K.J. Edgar, *Biomacromolecules.* (2011). <https://doi.org/10.1021/bm201152a>
23. K.I. Draget, C. Taylor, *Food Hydrocoll* **25**, 251–256 (2011)
24. T.S. Parreidt, K. Müller, M. Schmid, *Foods.* (2018). <https://doi.org/10.3390/foods7100170>
25. F. Bi, S.J. Mahmood, M. Arman, N. Taj, S. Iqbal, *Phys Chem Liquids.* (2007) <https://doi.org/10.1080/00319100600745198>
26. G.I. Olivás, D.S. Mattinson, G.V. Barbosa-Cánovas, *Postharvest Biol. Technol.* (2007) <https://doi.org/10.1016/j.postharvbio.2006.11.018>



27. D. Valero, H.M. Díaz-Mula, P.J. Zapata, F. Guillén, D. Martínez-Romero, S. Castillo, M. Serrano, *Postharvest Biol. Technol.* (2013) <https://doi.org/10.1016/j.postharvbio.2012.10.011>
28. H.M. Díaz-Mula, M. Serrano, D. Valero, *Food Bioprocess. Technol.* (2012). <https://doi.org/10.1007/s11947-011-0599-2>
29. T. Senturk Parreidt, M. Lindner, I. Rothkopf, M. Schmid, K. Müller, *Foods*, **8**, 203 (2019)
30. N. Maftoonazad, H.S. Ramaswamy, M. Marcotte, *Int. J. Food Sci. Technol.* **43**, 951–957 (2008)
31. N. Tabassum, M.A. Khan, *Sci. Hortic.* (2020) <https://doi.org/10.1016/j.scienta.2019.108853>
32. S. Alexandre, A.C. Vital, C. Mottin, R.M. do Prado, M.G. Ornaghi, T.R. Ramos, A. Guerrero, E.J. Pilau, I.N. do Prado, *J. Food Sci. Technol.* **58**, 3835–3843 (2021)
33. S. Matiacevich, N. Acevedo, D. López, *J. Food Process.* **39**, 2792–2801 (2015)
34. C. Ruan, Y. Zhang, Y. Sun, X. Gao, G. Xiong, J. Liang, *Int. J. Biol. Macromol.* **141**, 178–184 (2019)
35. A.E. Pavlath, C. Gossett, W. Camirand, G.H. Robertson, *J. Food Sci.* **64**, 61–63 (1999)
36. J.W. Rhim, *LWT*. **37**, 323–30 (2004)
37. J.G. Oliveira Filho, J.M. Rodrigues, A.C.F. Valadares, A.B. de Almeida, T.M. de Lima, K.P. Takeuchi, C.C.F. Alves, H.A. de Figueiredo Sousa, E.R. da Silva, F.H. Dyszy, M.B. Egea, *Food Hydrocoll.* (2019) <https://doi.org/10.1016/j.foodhyd.2019.01.052>
38. K. Saravanakumar, A. Sathiyaseelan, A.V.A. Mariadoss, H. Xiaowen, M.H. Wang, *Int. J. Biol. Macromol.* **153**, 207–214 (2020)
39. S.C. Lourenço, M.J. Fraqueza, M.H. Fernandes, M. Moldão-Martins, V.D. Alves, *Antioxidants*, **9**, 667 (2020)
40. E. Gómez-Ordóñez, P. Rupérez, *Food Hydrocoll.* (2011) <https://doi.org/10.1016/j.foodhyd.2011.02.009>
41. J. Praiboon, A. Chirapart, Y. Akakabe, O. Bhumibhamon, T. Kajiwara, *Science Asia* (2006) doi:[https://doi.org/10.2306/scienceasia1513-1874.2006.32\(s1\).011](https://doi.org/10.2306/scienceasia1513-1874.2006.32(s1).011)
42. R.B. Said, F. Mensi, H. Majdoub, A.B. Said, B.B. Said, A. Bouraoui, *J. Appl. Phycol.* (2018) <https://doi.org/10.1007/s10811-018-1414-5>
43. C.M. Rocha, A.M. Sousa, J.K. Kim, J.M. Magalhães, C. Yarish, M. do P. Gonçalves, *Food Hydrocoll.* (2019) <https://doi.org/10.1016/j.foodhyd.2018.10.048>
44. P. Guerrero, T. Garrido, I. Leceta, K. de la Caba, *Eur. Polym. J.* (2013) <https://doi.org/10.1016/j.eurpolymj.2013.08.014>
45. E.A. El-Hefian, M.M. Nasef, A.H. Yahaya, E-J. Chem. (2012) <https://doi.org/10.1155/2012/781206>
46. B.W. Souza, M.A. Cerqueira, A.I. Bourbon, A.C. Pinheiro, J.T. Martins, J.A. Teixeira, M.A. Coimbra, A.A. Vicente, *Food Hydrocoll* **27**, 287–292 (2012)
47. A. Imeson, *Food Stabilisers, Thickeners and Gelling Agents* (Wiley-Blackwell, New York, 2010), pp. 31–47
48. K.C. Labropoulos, D.E. Niesz, S.C. Danforth, P.G. Kevrekidis, *Carbohydr. Polym.* (2002) [https://doi.org/10.1016/S0144-8617\(02\)00085-1](https://doi.org/10.1016/S0144-8617(02)00085-1)
49. A. Chirapart, Y. Katou, H. Ukeda, M. Sawamura, H. Kusunose, *Fish Sci.* (1995) <https://doi.org/10.2331/fishsci.61.450>
50. A.M. Sousa, A.M. Sereno, L. Hilliou, M.P. Gonçalves, *In Mater. Sci. Forum.* (2010) <https://doi.org/10.4028/www.scientific.net/MSF.636-637.739>
51. E.S.H. Ziedan, H.M. El Zahaby, H.F. Maswada, E.H. Zoeir, *J. Plant Prot. Res.* (2018) <https://doi.org/10.24425/122938>
52. H.A.E.R. Zoier, H.M.E. Zahaby, E.S.H. Ziedan, H.F. Maswada, *Plant Arch* **17**, 1463–1468 (2017)
53. M.A. Cerqueira, A.M. Lima, B.W. Souza, J.A. Teixeira, R.A. Moreira, A.A. Vicente, *J. Agric. Food Chem.* (2009) <https://doi.org/10.1021/jf802726d>
54. M.A. Moreno, H. Bojorges, I. Falcó, G. Sánchez, G. López-Carballo, A. López-Rubio, I.C. Zampini, M.I. Isla, M.J. Fabra, *Food Hydrocoll.* **102**, 105595 (2020)
55. B. Zhang, Y. Liu, H. Wang, W. Liu, K.L. Cheong, B. Teng, *Food Control.* 108216 (2021)
56. M. Martínez-Sanz, A. Martínez-Abad, A. López-Rubio, *Food Packag.* (2019) <https://doi.org/10.1016/j.fpsl.2019.100367>
57. X. Wang, C. Guo, W. Hao, N. Ullah, L. Chen, Z. Li, X. Feng, *Int. J. Biol. Macromol.* **118**, 722–730 (2018)
58. S. Paidari, N. Zamindar, R. Tahergorabi, M. Kargar, S. Ezzati, S.H. Musavi, *J. Food Meas. Charact.* (2021) <https://doi.org/10.1007/s11694-021-00979-7>
59. S. Paidari, R. Tahergorabi, E.S. Anari, A.M. Nafchi, N. Zamindar, *Foods*, **9**, 2114 (2021)
60. S. Paidari, S.A. Ibrahim, *Gold Bull* **54**, 31–36 (2021)
61. M. Atef, M. Rezaei, R. Behrooz, *Food Hydrocoll* **45**, 150–157 (2015)
62. A.L. De Lacey, M.E. López-Caballero, P. Montero, *LWT*. **55**, 559–564 (2014)
63. M. Da Rocha, A. Alemán, V.P. Romani, M.E. López-Caballero, M.C. Gómez-Guillén, P. Montero, C. Prentice, *Food Hydrocoll* **81**, 351–363 (2018)
64. G. Pierre, C. Delattre, C. Laroche, P. Michaud, *Polysaccharides*. (2014). [https://doi.org/10.1007/978-3-319-03751-6\\_69-1](https://doi.org/10.1007/978-3-319-03751-6_69-1)
65. G.A. Ramu, M. Shanmugam, R. Bhat, *Int. J. Biol. Macromol.* (2018) <https://doi.org/10.1016/j.ijbiomac.2018.02.089>
66. V.A. Cosenza, D.A. Navarro, E.N. Fissore, A.M. Rojas, C.A. Stortz, *Carbohydr. Polym.* **102**, 780–789 (2014)
67. L. Pereira, J.F. Mesquita, *J. Appl. Phycol.* **16**, 369–383 (2004)
68. K. Tarman, U. Sadi, J. Santoso, L. Hardjito, *Encyclopedia of Marine Biotechnology* (Wiley, New York, 2020), pp. 147–159
69. A.A. Al-Alawi, I.M. Al-Marhubi, M.S. Al-Belushi, B. Soussi, *Mar Biotechnol* **13**, 893–899 (2011)
70. J. Necas, L. Bartosikova, *Vet. Med.* **58**, 187–205 (2013)
71. Y.Y. Tye, A.K. HPS, C.Y. Kok, C.K. Saurabh, *IOP Conf. Ser. Mater. Sci. Eng.* (2018) <https://doi.org/10.1088/1757-899X/368/1/012020>
72. Y. Yuguchi, T.T. Thuy, H. Urakawa, K. Kajiwara, *Food hydrocoll* **16**, 515–522 (2002)
73. H.A. Khalil, C.K. Saurabh, Y.Y. Tye, T.K. Lai, A.M. Easa, E. Rosamah, M.R.N. Fazita, M.I. Syakir, A.S. Adnan, H.M. Fizree, N.A.S. Aprilia, *Renew. Sust. Energ. Rev.* **77**, 353–362 (2017)
74. T. Karbowiak, F. Debeaufort, D. Champion, A. Voilley, *J. Colloid Interface Sci.* (2006) <https://doi.org/10.1016/j.jcis.2005.07.030>
75. A.T. Banu, P.S. Ramani, A. Murugan, *Food Sci. Hum. Wellness.* (2020) <https://doi.org/10.1016/j.fshw.2020.03.002>
76. H.M. Hamzah, A. Osman, C.P. Tan, F.M. Ghazali, *Postharvest Biol. Technol.* (2013) <https://doi.org/10.1016/j.postharvbio.2012.08.012>
77. C. Ribeiro, A.A. Vicente, J.A. Teixeira, C. Miranda, *Postharvest Biol. Technol.* (2007) <https://doi.org/10.1016/j.postharvbio.2006.11.015>
78. S.M. Khojah, J. Aquat, *Food Prod. Technol.* **29**, 810–822 (2020)
79. T.T. Tran, P. Roach, M.H. Nguyen, P. Pristijono, Q.V. Vuong, *Food Hydrocoll* **99**, 105322 (2020)
80. S. Jafarzadeh, A.M. Nafchi, A. Salehabadi, N. Oladzaad-Abbasabadi, S.M. Jafari, *Adv. Coll. Interface. Sci.* (2021) <https://doi.org/10.1016/j.cis.2021.102405>
81. B. Meindrawan, N.E. Suyatma, A.A. Wardana, V.Y. Pamela, *Food Packag.* **18**, 140–146 (2018)
82. X. Zhou, X. Zong, M. Zhang, Q. Ge, J. Qi, J. Liang, X. Xu, G. Xiong, *Int. J. Biol. Macromol.* **183**, 331–339 (2021)
83. R. Nur Fatin Nazurah, Z.A. Nur Hanani, *Carbohydr. Polym.* **157**, 1479–1487 (2017)
84. M. Pérez-Mateos, P. Montero, M.C. Gómez-Guillén, *Food Hydrocoll* **23**, 53–61 (2009)

85. V.D. Alves, R. Castelló, A.R. Ferreira, N. Costa, I.M. Fonseca, I.M. Coelho, Proc. Food Sci (2011). <https://doi.org/10.1016/j.profoo.2011.09.038>
86. S. Shojaee-Aliabadi, H. Hosseini, M.A. Mohammadifar, A. Mohammadi, M. Ghasemlou, S.M. Ojagh, R. Khaksar, Int. J. Biol. Macromol. (2013) <https://doi.org/10.1016/j.ijbiomac.2012.08.026>
87. J. Simona, D. Dani, S. Petr, N. Marcela, T. Jakub, T. Bohuslava, Polymers. **13**, 332 (2021)
88. A. Farhan, N.M. Hani, Food Packag. **24**, 100476 (2020)
89. T.R. Martiny, V. Raghavan, C.C. Moraes, G.S. Rosa, G.L. Dotto, Foods. **9**, 1759 (2020)
90. S.M. Hashemi, D. Jafarpour, Food Sci. Nutr. (2020) <https://doi.org/10.1002/fsn3.1544>
91. S.M. Hashemi, D. Jafarpour, J. Food Sci. (2021) <https://doi.org/10.1111/1750-3841.15568>
92. S. Jafarzadeh, A. Salehabadi, A.M. Nafchi, N. Oladzadabbasabadi, S.M. Jafari, Trends Food Sci. Technol. (2021) <https://doi.org/10.1016/j.tifs.2021.07.021>
93. M. Sayadi, A. Mojaddar Langroodi, D. Jafarpour, J. Food Meas. Charact. (2021) <https://doi.org/10.1007/s11694-021-01096-1>
94. D. Zhang, M. Zhang, X. Gu, *Bioactive Seaweeds for Food Applications* (Academic Press, New York, 2018), pp. 153–175
95. J. Bonilla, L. Atarés, M. Vargas, A. Chiralt, J. Food Eng. **110**, 208–213 (2012)
96. J.F. Ayala-Zavala, H. Soto-Valdez, A. Gonzalez-Leon, E. Alvarez-Parrilla, O. Martin-Belloso, G.A. Gonzalez-Aguilar, J. Incl. Phenom. Macrocycl. Chem. (2008) <https://doi.org/10.1007/s10847-007-9385-1>
97. D. Lin, Y. Zhao, Compr. Rev. Food Sci. Food Saf. **6**, 60–75 (2007)
98. S. Jafarzadeh, A.M. Nafchi, A. Salehabadi, N. Oladzad-Abbasa-badi, S.M. Jafari, Adv. Colloid Interface Sci. (2021) <https://doi.org/10.1016/j.cis.2021.102405>
99. H.P. Khalil, T.K. Lai, Y.Y. Tye, S. Rizal, E.W. Chong, S.W. Yap, A.A. Hamzah, M.R. Fazita, M.T. Paridah, EXPRESS Polym. Lett. **12**, 296–317 (2018)

**Publisher's Note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.