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An Overview of the Alternative Use of Seaweeds to Produce Safe and Sustainable Bio-Packaging

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Abstract: In modern times, seaweeds have become widely involved in several biotechnological applications due to the variety of their constituent bioactive compounds. The consumption of seaweeds dates to ancient times; however, only from the last few decades of research can we explain the mechanisms of action and the potential of seaweed-derived bioactive compounds, which has led to their involvement in food, cosmetic, pharmaceutical, and nutraceutical industries. Macroalgae-derived bioactive compounds are of great importance as their properties enable them to be ideal candidates for the production of sustainable "green" packaging. Diverse studies demonstrate that seaweed polysaccharides (e.g., alginates and carrageenans) not only provide health benefits, but also contribute to the production of biopolymeric film and biodegradable packaging. The dispersion of plastics and microplastics in the oceans provoke serious environmental issues that influence ecosystems and aquatic organisms. Thus, the sustainable use of seaweed-derived biopolymers is now crucial to replace plasticizers with biodegradable materials, and thus preserve the environment. The present review aims to provide an overview on the potential of seaweeds in the production of bioplastics which might be involved in food or pharmaceutical packaging.

Keywords: seaweed; bioactive compounds; bioplastic; sustainability; biodegradable packaging

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1. Introduction

Seaweeds, also called marine macroalgae, are multicellular photosynthetic organisms that can be found in all aquatic environments. They are divided into three main groups according to their color, which is produced by the presence of pigments. Brown seaweeds (phylum Ochrophyta, class Phaeophyceae) have an abundance of pigments that vary from yellow to dark brown [1], while red seaweeds (phylum Rhodophyta) contain high amounts of carotenoids, chlorophyll (*a* and *d*), phycoerythrin, phycocyanin and allophycocyanin [2,3]. Green seaweeds (phylum Chlorophyta) possess mainly chlorophyll, which has a key role in photosynthesis. Its greenish pigmentation conveys the typical green color in plants, algae, and cyanobacteria [4].

The variety of compounds present in seaweeds possess unique properties, which have positive effects on organisms; in fact, the use of seaweeds has been common since ancient times for alimentary and medicinal purposes, especially in Asian countries [5–7].

With the development of scientific research and new techniques, it has been possible to individuate compounds extracted from seaweeds and investigate their curative effects on human organisms, such as antioxidant, antimicrobial, antitumoral properties [8]. Moreover, seaweeds might be also employed in several industrial applications, such as the production of fertilizers [9–11], aquaculture feed, biofuels, application in wastewater treatments, or to produce innovative and ecologic materials to replace plastic equivalents [12–14], contributing towards a sustainable solution to protect the environment from the discharge of non-biodegradable plastic.

Nowadays, plastic industries are responsible for a very high amount of plastic waste, especially due to the production of packaging; thus, it is of high priority that we find new, eco-friendly material to develop biodegradable packaging [15–17]. As the global population increases, the demand for plastic products also increases, along with the level of plastic waste in the environment [18]. More than eight million tons of plastic waste is dispersed into oceans every year, causing global environmental pollution and corrupting the proper functioning of ecosystems [19]. The growing necessity to decrease the use of petroleum-based plastic products has led research to look for new sources of raw material with the same characteristics of plasticizers, but also being biodegradable and non-harmful to human health and the environment.

To attenuate the plastic waste, an ecologic alternative may be the development of "bioplastics"; this term refers to plastics derived from biological sources that are biodegradable and renewable. Bioplastics are easy to recycle, and their production requires less cost and energy relative to oil-based plastic [20]. Most of the food, cosmetic, and pharmaceutical products in the market are packaged in plastic, which takes more than 400 years to decompose, having a major impact on the environment. Therefore, the need for new sources of biodegradable film is a crucial point for the safeguard of our planet.

Starch and cellulose are the main polysaccharides of vegetal origin tested for packaging materials [21]. Starch occurs widely in nature as reserve polysaccharide in plants; it has been considered for the development of biodegradable films as starch is easily obtained from natural sources, and is renewable, abundant, low cost, and has the ability to form an odourless, colourless, and transparent biofilm [22–25]. The low oxygen permeability of the starch matrix makes it an interesting polysaccharide for food preservation [23,26], even though so far limited applications have been developed as starches possess poor water barrier properties, a low melting point, and lower mechanical strength compared with petroleum-based plastics. Starch can be found in several vegetal sources, such as peas, rice, corn, potatoes, and tapioca [27–31].

Cellulose is a low-cost, biodegradable and water-insoluble material that is promising for the development of biodegradable films [24,32]. It is the most abundant biomass resource on earth, and is found in plants, fruits, and vegetables [33]. Cellulose derivates, such as hydroxypropyl methylcellulose (HPMC) and methylcellulose (MC), have been used to form biofilm due to their mechanical resistance [34]. Moreover, the methyl group substitution to the cellulose backbone led to the solubility of HPMC and MC in cold water; thus, when heated, they form a gel, while in cold water they are soluble. Both HPMC and MC are potential compounds for the production of strong, clear, odourless, tasteless, oil-resistant, and water-soluble films [21].

Ashfaq et al. [35] recently developed a new biofilm using the whole fruit. The use of papaya has been promoted for its strong photoprotection activity, as the UV screening capacity of food packaging film is beneficial to prevent spoiled food. The UV transmittance of developed films was calculated; papaya film (a mixture of gelatin, papaya, corn starch, and glycerin) revealed the highest transmittance compared to film with soy protein addition, as was the case for the combination of glycerol and gelatine. Moreover, papaya film exhibited higher transparency and elasticity, and slower degradability, compared to film with soy proteins. In conclusion, papaya biofilm revealed effective UV protection properties and qualities that makes it comparable to commercial plastic films.

On the other hand, macroalgae have more potential as a source of bioplastics due to their higher biomass, fast reproduction, they are easily maintained in all environments, and are cost-effective [20]. In addition to their biodegradability property, seaweed-derived biofilms might exhibit antimicrobial activities, as seaweeds produce antimicrobial compounds, identified as phenols, fatty acids, carbohydrates, proteins, and minor compounds [36–40], which take part in mechanisms of antibiotic defence developed to survive the harsh environments seaweeds inhabit. The integration of the whole seaweed, or seaweed extracts, with antimicrobial activity in food packaging manufacture might increase the shelf-life of foods and prevent the development of foodborne pathogens. A recently

published study by Cabral et al. [41] listed the main antimicrobial compounds recently isolated from multiple seaweeds and their main antimicrobial properties. Most compounds exhibit broad-spectrum antibiotic activity, confirming seaweed has the potential to shield against microbes and foodborne pathogens.

This review aims to provide a better understanding of seaweeds as a potential resource used to produce biodegradable and antibacterial plastic. Seaweed-derived biological compounds, such as carrageenan, agar, and alginate, possess unique physical, optical, mechanical, thermal, antioxidant, and antibacterial properties, and the biodegradability of seaweed-derived bioactive compounds make them ideal candidates to produce bioplastic packaging for medicine and food.

2. Potential Algal Bioactive Compounds for Bioplastic Production

The accumulation of petroleum-based plastic on land and into oceans is of growing concern to society, as it can result in physical issues for organisms that ingest plastic or are entangled in fishing nets or plastic ropes, and also lead to the indirect consumption of microplastic previously ingested by animals subsequently consumed by humans [42]. The accumulation of microplastics can lead to an accumulation in the organism of toxic chemical compounds, incorporated into/onto plastics. According to Cole et al. [43], these chemical substances can accumulate in higher trophic levels, and thus into seafood, creating health issues for humans. Entanglement and ingestion of microplastic debris can be lethal or sub-lethal, causing reduced food particle capture and swallowing, impaired reproduction ability, loss of sensitivity or mobility, the inability to escape from predators, and/or decreased growth. Gall and Thompson [44] reported that sea turtles, marine mammals, and all types of sea birds are species most negatively affected by entanglement in, and ingestion of, plastic pollution. New materials based on biopolymers might provide a sustainable solution to replace synthetic plastic with edible and non-harmful bioplastic. Various studies have already proven that bioplastic can be produced from starch, crops, and microalgae [45–49]. The use of cultivable marine resources will avoid the use of land and resources destined for crops; it also potentially avoids deforestation, as seaweed cultivation is performed in outdoor or laboratorial conditions [50–52]. Moreover, the large-scale production of seaweeds reduces costs and increases compound availability.

Seaweed-derived biological compounds most suitable to produce biofilms or bioplastics are alginate, carrageenan, and agar [53–55].

2.1. Alginate

Alginate is a polysaccharide derived from alginic acid and its derivatives and salts [56,57]. Alginates are anionic linear polysaccharides found in high amounts in brown seaweeds, up to 40% of the dry weight, and have been reported as able to form edible films. They are comprised of polymers of alginic acid, with monomer units of β-D-mannuronic acid (M) and α-L-guluronic acid (G) joined by 1,4 linkages [58] (Figure 1).

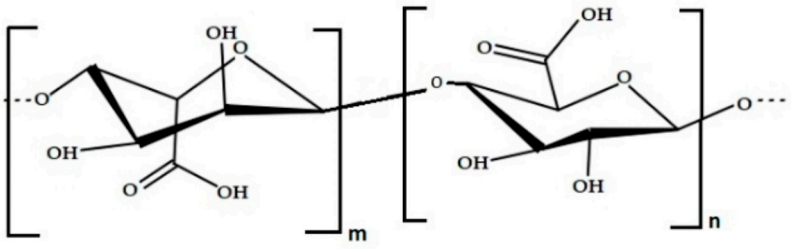

Figure 1. Chemical structure of alginic acid.

The physicochemical and mechanical properties of gels made from alginate differ based on the M/G ratio and the length of the structure; thus, a high content of guluronic acid results in stronger gelling properties, and a more elastic gel. On the other hand, low M/G ratios result in strong and brittle gels with good heat stability, but show syneresis after

freeze–thaw processing [56,57]. Due to its excellent stabilizing and thickening properties, alginate is commonly used in food products and medicine [56,59,60].

Alginates are highly hydrophilic; thus, it is important to combine the matrix with other elements to provide more resistance upon water contact. Moreover, the presence of ions influences the solubility of alginates, while their ability to form gels depends on type of bonds among cations [56,57].

The addition of calcium in the alginate matrix provides more stability and resistance to the membrane, which can be an interesting step for the development of biodegradable materials with antimicrobial properties and non-toxic packaging [59,60].

2.2. Carrageenan

Carrageenan, mainly found in red algae, is a linear polysaccharide formed from alternating sulfated or non-sulfated galactose units linked with α-1,3-glycosidic and β-1,4-galactose bonds [61] (Figure 2).

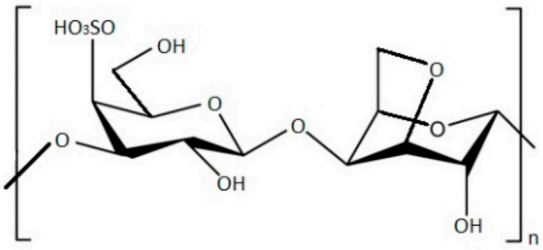

Figure 2. Chemical structure of κ-carrageenan.

Depending on the position at which the sulfate group is connected to the galactose unit, carrageenans can be divided into different types (κ-carrageenan, ι-carrageenan, λ-carrageenan, γ-carrageenan, ν-carrageenan, ξ-carrageenan, and μ-carrageenan). In nature, carrageenans are mostly hybrid; thus, their properties vary based on the bonded sulfate group [62].

Commercial carrageenans are widely used in ice creams, paints, water gels, and pharmaceuticals. They are normally divided into three structural types (κ-, ι-, and ʎ-forms) according to the number of sulfated groups connected to the galactose unit, where the number, chemical location, and arrangement of these groups defines the carrageenan's function and bioactivity power [63]. These different types of carrageenan are obtained from different species, e.g., κ-carrageenan is predominantly extracted from the species *Kappaphycus alvarezii*. κ-carrageenan forms gels that are hard, strong, and brittle; in contrast, ι-carrageenan is mainly produced by *Eucheuma denticulatum* (trade name "*spinosum*"), and it gives the soft and weak form to gels. Lastly, ʎ-carrageenan is obtained from different species of the genera *Gigartina* and *Chondrus* [64].

Due to their gelling, thickening, and stabilizing properties, carrageenans are widely applied in the food industry [57]. The Food and Drugs Administration (FDA) and the European Food Safety Agency (EFSA) have approved the commercial forms of ʎ-, κ-, and ι-carrageenans as food additives [65]. In the last few decades, the biological potential of carrageenan has been explored in medical field, with positive outcomes; in fact, it has been discovered that carrageenan possesses anticoagulant and antithrombotic activity [66], antivirus activity [67], antitumoral activity [68], and antioxidant properties [69].

All types of carrageenans are soluble in water, although their aqueous solubility is influenced by temperature, pH, ionic strength of the medium, and the presence of cations. The sulfate and hydroxyl groups determine their hydrophilic characteristic, while their hydrophobicity derives mostly from the 3,6-anhydro-α-D-galactopyranose units [57].

The hydrophobicity of carrageenan represents a disadvantage for the manufacturing of resistant packaging; however, bonding carrageenan with hydrophobic compounds to reinforce the matrix of the compound might be a solution for enhancing the proper-

ties of carrageenan, conferring their strength into eco-friendly, cost-effective packaging material [70].

2.3. Agar

The main structure of agar is chemically characterized by the repeating units of D-galactose and 3,6-anhydro-L-galactose, with a few variations, as well as by a low ester sulfate content. The structure of agar consists of two groups of polysaccharides: agarose, a neutral polysaccharide, and agaropectin, an oversimplified term for the charged polysaccharide [71–73].

Agarose is accountable for the gelling capacity of agar, which makes it very useful in skin care, herbal medicines, and pharmaceutical applications; it also has excellent film properties [74] (Figure 3).

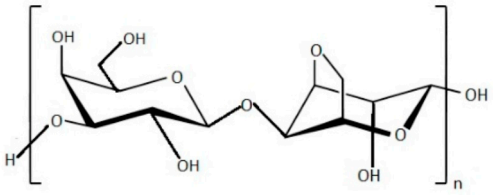

Figure 3. Chemical structure of agarose polymer.

Agar, as well as carrageenan, is widely involved in the commercial food processing industry due to their ability to act as stabilizers, emulsifiers, and thickening agents. Both are already used in gel-based food products, such as desserts, jams, jellies, and bakery products. Gels produced from agar are usually tense and lucid, but the addition of sugars increases their strength [75].

Agar possesses low hydroscopic property, which is an advantage in the production of packaging; moreover, agar films are biologically inert, and can easily interact with different bioactive substances and/or plasticizers to help the formation of an elastic and soft gels [76–78].

3. Physicochemical Characteristics of Seaweeds Phycocolloids

The hydrocolloidal and gelling properties of phycocolloids in seaweeds are dependent on their polymer structure, concentration in a solution, temperature, pH, and syneresis potential [79–81].

The physicochemical quality of phycocolloids is heavily influenced by the seaweed species, environmental conditions, extraction methods, and treatment processes [79,82]; thus, a deep understanding of these factors will allow us to discover the most efficient techniques to obtain high-quality phycocolloids.

In the form of salts, carrageenans possess high gel strength [83]. Both κ-carrageenan and ι-carrageenan gels are stable at room temperature. One exception is λ-carrageenan, which has typical nongelling property, and is the only cold-water soluble carrageenan in its native form. According to Robal et al. [84], κ-carrageenan enriched with cations enhances the gel formation and strength of phycocolloids. Paula et al. [85] studied the physical properties of glycerol-plasticized edible films made with κ-carrageenan, ι-carrageenan, and alginate; κ-carrageenan exhibited higher tensile strength and elasticity, higher moisture permeability, and lower opacity compared to ι-carrageenan, while alginate films revealed higher transparency [85].

Similar to carrageenan, alginate also forms rigid and stable gel matrix in the presence cations, especially Ca^{2+} [86]. Films obtained from sodium alginate with 1%–3% (*w/v*) calcium chloride solution exhibited an increase in tensile strength and elongation properties, and reduced opacity [87,88].

Agar films, compared to carrageenan and alginate films, have a lower tensile strength and water vapor permeability. On the other hand, they exhibit twice the elongation value

of κ-carrageenan film, and with better elasticity [89,90]. Due to their gelling properties and viscosity, seaweed phycocolloids are widely used as stabilizers and/or thickening and gelling agents in the manufacturing of food, pharmaceuticals, and cosmetics.

With new methodologies, it has been possible to investigate the behaviour of these seaweed-derived compounds when incorporated into biofilms, and thus to use them as potential candidates to develop biofilms and packaging [91–94]. Different methods of film formation had significant effects on the physical properties and microstructures of the film. An example is given by Li et al. [95], in which chitosan–alginate films were produced through different processes. Their results showed that biofilm prepared through layer-by-layer assembly combined with ferulic acid crosslinking has enhanced mechanical properties, opacity, and hydrophobicity compared to films prepared by direct mixing, crosslinking alone, and layer-by-layer assembly alone.

Seaweed-derived bioactive compounds possess biological properties that can assure the safety of the packaged product [96]. Due to their biocompatibility, non-cytotoxicity, and antimicrobial properties, biopolymers from seaweeds are excellent candidates to develop safe packaging for food and pharmaceuticals [97]. Bioplastics made from extracted, seaweed-derived biopolymers are reported to be more resistant to microwave radiation due to the photoprotection properties of seaweed compounds, and thus can conserve the quality of packaging for food or pharmaceuticals [98].

The extraction of carrageenan and agar typically involves hot water as a solvent, as they possess great solubility. Alginate, on the other hand, requires hot alkali as the major solvent, as alginic acid is composed of water-insoluble salts. Thus, through alkali extraction, alginate salts are converted into water-soluble alginate salts.

Despite the requirement of hot water for optimal extraction, agar and carrageenan are industrially extracted with alkali extraction, since hot water extraction weakens their rheological properties (e.g., gel strength); consequently, the quality of these phycocolloids is not optimal for bioplastic formulation. In the work of Khalil et al. [56] are reported the percentages of gel strength obtained with treated and untreated extraction of agar and carrageenan, and their results show that, with alkali extraction, the gel strength is optimized, but the yield of production decreases.

Common Extraction Processes for Commercial Seaweed Hydrocolloids

Before extraction, it is important to clean the seaweed to remove epiphytes, impurities, sand, debris, salts, and contaminants.

Agar and carrageenan are submitted to alkali pre-treatment to improve gelling properties through the conversion of unstable sulfate molecules into 3,6-anhydro-L-galactopyranose (3,6-AG) [56]. Alginate undergoes formaldehyde pre-treatment to eliminate the color pigments in the seaweed tissue and increase the alginate yield, and hydrochloric acid (HCl) pre-treatment to "clarify" the phenolic compounds and formaldehyde residue while promoting the conversion of insoluble alginate salts (calcium, magnesium, etc.,) into soluble salts [99–103].

Hot water extraction is performed for agar and carrageenan, and is followed by alkali extraction to obtain compounds with desirable properties and functionalities through the manipulation of various parameters, such as temperature, time, pH, solvent concentration, etc. For alginate, only alkali extraction is performed.

All three compounds are further neutralized by removing excess chemicals and solvents; subsequently, through precipitation and filtration, the residuals are eliminated, and the pure compound is obtained; in the last steps, drying and milling are performed to obtain dry and purified final products ready for commercial purpose (Figure 4) [56].

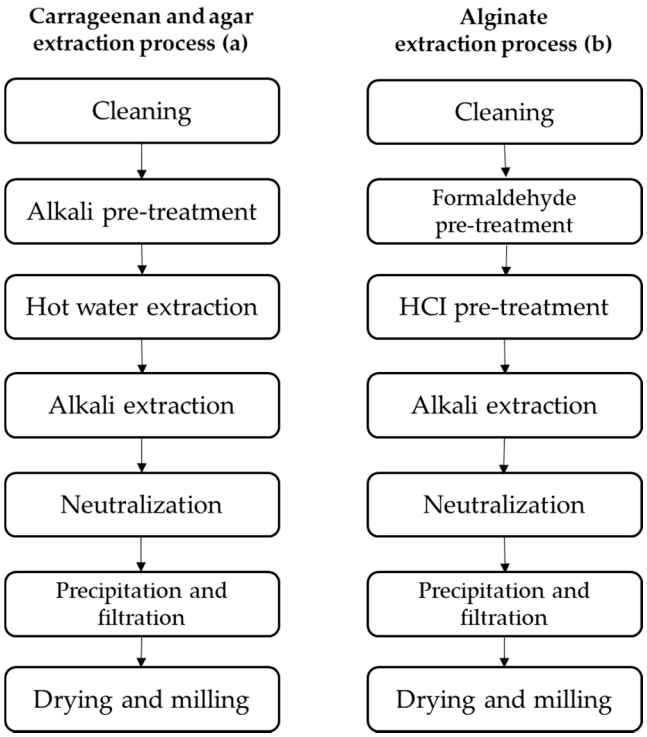

Figure 4. Common extraction process for commercial seaweed hydrocolloids: carrageenan and agar (**a**) and alginate (**b**).

Some of the limitations of common hydrocolloid extraction include the high consumption of time, energy, and water. Moreover, to obtain an optimum yield, a huge quantity of chemical solvents are used, some of which are health hazards; because of the lack of control throughout the whole production process and discharge, these chemicals can be a serious threat to human health and the environment [104,105]. Another downside of traditional extraction process is the expensive cost of ethanol used during precipitation of carrageenan to obtain refined carrageenan [104].

To overcome these disadvantages, new industrial processes of extraction that are cheaper and eco-friendly have been researched. The "green" extraction and processing methods are reported by Khalil et al. [56], which are as follows: microwave-assisted extraction (MAE), enzyme-assisted extraction (EAE), ultrasound-assisted extraction (UAE) [106], supercritical fluid extraction (SPE), pressurized solvent extraction (PSE), reactive extrusion and photobleaching processes. Some of these methods are already used to extract bioactive compounds from plants [107,108]. Nevertheless, all these techniques possess pros and cons in terms of cost, yield of production, and time consumption [56].

To prove the effectiveness of dry and purified phycocolloids for producing bioplastics, it is necessary to perform tests to evaluate the physical, optical, mechanical, thermal, antioxidant, and antibacterial properties, and biodegradability. Physical properties include thickness, solubility, water vapor permeability, water vapor transmission rate, and moisture content of the films. Optical properties include transparency, opacity, and light transmittance values. Mechanical properties include strength and elongation at break. Antioxidant properties can be measured via total phenolic content and the DPPH radical scavenging activity test. Antimicrobial properties are analyzed by inhibitory effects of the purified phycocolloids against bacteria, such as *Escherichia coli*, *Listeria monocytogenes*, and *Salmonella Typhimurium*, while biodegradability is usually tested via a soil burial test [109].

Seaweed bioplastics biodegrade in the soil over a short time period, and no plastics residuals are dispersed into the environment [110], which is useful in those applications related to human health, such as foods or drugs packaging [111].

If the quality of phycocolloids is suitable to produce biofilms, the matrix of the phycocolloids can be enriched with other polymers, hydrophobic components, and/or nanoparticles, forming a hybrid material [53,112] with stronger mechanical strength and water barrier properties, which are properties required for strong, seaweed-based packaging.

4. Promising Seaweeds to Produce Bioplastics

To define the quality of an optimal bioplastic, mechanical characteristics must be considered, such as tensile strength, elongation at break, thermal resistance, and water vapor permeability. Tensile strength is the maximum stress that a material can withstand while being stretched or pulled before breaking. Generally, the tensile strength and Young's modulus of plant fibers increases with increasing cellulose content of the fibers. Elongation at break is the ratio between changed length and initial length after breakage of the tested material. In a matrix with natural polymers, it expresses the ability to resist changes of shape without crack formation. Thermal resistance is a heat property, and represents the difference in temperature at which a material can resist [113]. Vapor permeability is a material's ability to allow a vapor (such as water vapor or, indeed, any gas) to pass through it. According to the ISO 11092:1993 [114], water vapor permeability is "a characteristic of a textile material or composite depending on water vapor resistance". The higher the value of the permeability of the material, the more rapidly water and vapor can pass through it.

Films and bioplastics developed from seaweed-derived biopolymers that follow these characteristics are considered potential new materials to produce bio-packaging. Some current investigations on seaweeds employed in bioplastic production are summarized in Table 1.

Table 1. Seaweeds investigated for the development of biofilms for packaging manufacture.

Seaweed	Extract/Compound in Biofilm Formulation	Biofilm Mechanical Characteristics	Reference
Phylum Ochrophyta, Class Phaeophyceae			
Sargassum siliquosum	Alginate	Adequate tensile strength, elongation at break, water vapor permeability and water solubility due to addition of $CaCl_2$	[59]
Sargassum natans, *Laminaria japonica*	Crude extracts	Enhanced physicochemical, mechanical, and thermal properties due to addition of cellulose nanocrystals	[115]
Phylum Rhodophyta			
Kappaphycus sp.	Crude extract	Reduction in brittleness, weak tensile strength	[116]
Kappaphycus alvarezii	κ-carrageenan	Evidenced good physical, mechanical and thermal strength of bioplastic films	[117]
Eucheuma cottonii	Semi-refined carrageenan	Biofilm with refined carrageenan showed higher tensile strength and thermal resistance compared with semi-refined carrageenan	[118]
	Refined carrageenan		
Gracilaria salicornia	Agar (photobleaching extraction)	Tensile strength and elongation at break higher for biofilm obtained with agar from photobleaching extraction Thermal resistance and biodegradability higher in alkali extraction agar film	[119]
	Agar (alkali extraction)		
Gracilaria vermiculophylla	Agar	Transparency, clearness, flexibility, and mechanical strength enhanced by addition of glycerin	[77]
Eucheuma spinosum	Crude polysaccharides	The addition of glycerol as plasticizer enhances biofilm physical and mechanical properties	[120]

Seaweeds belonging to the genera *Kappaphycus*, *Eucheuma*, *Gracilaria*, *Porphyra*, *Gelidium*, *Pterocladia* for red seaweeds, *Ulva*, *Codium*, *Enteromorpha* for green seaweeds, and *Macrocystis*, *Laminaria*, *Ascophyllum*, *Lessonia* for brown seaweeds, have been investigated for their high polysaccharide content, which might allow the production of plastic biofilms [121]. Alginate, agar, carrageenan, and cellulose from seaweeds have shown excellent film-forming properties, and are very easy to process [122].

Alginate is the compounds most frequently used for bioplastic production. It is extracted from brown seaweeds, usually from *Laminaria* sp. and *Ascophyllum nodosum*. It is commercially sold for its gel properties; however, these properties depend on sequence, composition, and the ratio of alginic acid monomers [87].

Lim et al. [59] investigated the alginate extract from the brown alga *Sargassum siliquosum* as a raw material for the synthesis of bioplastic film. During the treatment process, alginate was mixed with sago starch, sorbitol, and calcium chloride ($CaCl_2$). The physical properties of the biofilm were then analyzed. Their results indicate that the biofilm developed using a mixture of 2 g of alginate powder from *Sargassum siliquosum* and 15% *w/w* of sorbitol treated with 75% *w/w* of $CaCl_2$, appears to possess adequate properties (tensile strength, elongation at break, water vapor permeability, and water solubility). This study suggested that alginate from *Sargassum siliquosum* as a suitable candidate for the synthesis of bioplastic films [59].

Doh et al. [115] prepared biofilms using crude extracts and cellulose nanocrystals from *Laminaria japonica* and *Sargassum natans*. It has been noticed that the presence of cellulose nanocrystals enhanced the physicochemical, mechanical, and thermal properties of the biofilm, providing a positive vision towards the use of cellulose to produce bio-packaging.

Hanry and Surugau [116] investigated the biofilms obtained from pure κ-carrageenan and whole seaweed of *Kappaphycus* sp. The aim of this research was to compare the properties of both biofilms and determine whether the carrageenan extraction process could be avoided. Their results showed a reduction in brittleness for biofilms derived from the whole algae, but they were weaker due to the low presence of carrageenan and, consequently, weaker binding intermolecular forces. However, the biofilms produced were both potential candidates to replace petroleum-based plastic and non-degradable packaging [116]. For instance, the best use for this type of bioplastic is single-use packaging for powders, fast foods, candies, or to contain daily-use pharmaceuticals, such as integrators or pills, which do not require great mechanical properties and are easy to open. Another investigation performed by Sudhakar et al. [117] on κ-carrageenan from *Kappaphycus alvarezii*, evidenced good physical, mechanical, and thermal strength of bioplastic films, suggesting that further research would be beneficial to develop bioplastic film from *Kappaphycus* sp. in the market.

The physical and biological properties of carrageenans derived from the red seaweed *Eucheuma cottonii* have been investigated to evaluate these compounds as raw materials for bioplastic production. Semi-refined carrageenan flour was obtained from *Eucheuma cottonii*, while refined carrageenans flour was purchased. Bioplastic with extracted carrageenans showed higher antimicrobial activity, but less strength, than bioplastic produced with refined carrageenans. Furthermore, the thermal resistance was higher in bioplastic made from refined carrageenans. However, both bioplastics met the requirements to be used in the market [118].

Agar from *Gracilaria salicornia* was extracted to identify its physicochemical properties as raw material for bioplastic products. Two extraction methods for agar were tested, and different biofilms were developed to evaluate their properties. The results showed that tensile strength and percent elongation of biofilm obtained using agar from photobleaching extraction (PB) was higher than biofilm from agar obtained by alkali extraction (AE), while thermal stability was higher in the AE agar film. Moreover, the AE agar film was completely decomposed after 30 days in the soil burial test [119]. Therefore, agar extracted from *Gracilaria salicornia* is interesting for future possibilities in commercial applications of bioplastic films. A study by Sousa et al. [77] reported that biofilm obtained using agar from *Gracilaria vermiculophylla* showed transparency and clearness similar to the commercial

counterpart [123]. The addition of glycerin as a plasticizer gives flexibility and mechanical strength, making agar-based films suitable for packaging foods or coating pharmaceuticals. Agar-based film presents thermal resistance and antimicrobial activity, making it another potential candidate; therefore, more accurate studies would be required [124].

Bioplastics were synthesized recently by Darni et al. [120] using a matrix combined with a filler of sorghum stalk, polysaccharides from *Eucheuma spinosum*, and glycerol as plasticizer. The addition of filler and plasticizer in bioplastic synthesis enhances its physical and mechanical properties. The presence of these substances must be optimized to improve the bioplastic characteristics; however, *Eucheuma spinosum* might be a source for alternative to plastic packaging.

Among green seaweeds, ulvan extracts showed film-forming property and can be used as a filler or reinforcement in pharmaceutical and cosmetic applications [125]. Ulvans are unique polysaccharides with high viscosity and gelling properties, which might make them potential agents for biofilm production. Even though there are no studies at present on the production of bioplastic from ulvan, these extracts exhibit high thermal resistance and mechanical strength, all properties in line with the characteristics of optimal bioplastics [126].

Among all green algae polysaccharides, cellulose was found to be the most suitable for developing biodegradable plastic; its properties make cellulose capable of forming hydrocolloids in a suitable solvent system, and thus able to exhibit excellent mechanical performance [127]. Moreover, it is cheap, biodegradable, and renewable. For example, cellulose from *Cladophora* sp. is very robust and not susceptible to chemical reactions. Its robustness and excellent mechanical, thermal, and morphological properties make this material an interesting possibility as a bio-packaging material for foods or pharmaceuticals [128]. Therefore, green seaweed compounds should be further investigated as they show interesting properties that make them potential candidates for creating biodegradable plastic.

Active Biofilms

The incorporation of seaweeds into other polymers changes the mechanical, thermal, optical, and chemical properties of the materials. Carina et al. [129], in a recent published work, focused on the potential development of active packaging, which is identified as packaging that interacts with the product in a positive way, to improve the safety and shelf-life of the product and to maintain the original sensory properties. The inclusion of seaweed extracts into biofilm formulation can provide a defense for the product against bacteria, oxidation, and UV rays.

However, it is important to verify the biological effect of mixed and pure polysaccharides in biofilms. In some studies, crude extracts incorporated into biofilms exhibit antimicrobial activity. For example, mixed polysaccharides extracted from the brown seaweed *Nizamuddinia zanardinii* inhibited the growth of *E. coli* and *P. aeruginosa*, while crude fucoidans extract from the *Saragassum polycystum* exhibited inhibitory effects against *V. harveyi, S. aureus*, and *Escherichia coli* [130,131]. In addition, crude polysaccharides extracted from red alga *G. ornota* showed antimicrobial activity against *Escherichia coli* [132]. The antimicrobial activity of pure carrageenan did not exhibit an effect against *S. aureus, Escherichia coli*, or *L. monocytogenes* [133,134]. Meanwhile, in the study of Kanatt et al. [135], pure κ-carrageenan from *Kappaphycus alvarezii* was used to evaluate the antimicrobial activity of the Gram-positive bacteria *S. aureus* and *B. cereus,* and the Gram-negative bacteria *E. coli* and *P. fluorescens*. In vitro results show antimicrobial activity only against Gram-positive bacteria, and no growth inhibition of Gram-negative bacteria. The inclusion of κ-carrageenan in polyvinyl acetate (PVA) film showed an effective zone of inhibition against *S. aureus* and *B. cereus*, proving that the antimicrobial activity of the pure extract could be retained and affect the film, protecting the wrapped products. Aqueous seaweed extract from *Kappaphycus alvarezii* also showed an increased antioxidant activity after incorporation into PVA film, compared to pure PVA film [135]. Lipid oxidation has a strong impact

on food quality; it can lead to a decrease in shelf-life and nutritional value of food [136]. Thus, the formulation of packaging with antioxidant compounds leads to a decrease in the amount of lipid oxidation and protein degradation within the packaging, avoiding degradation of the product coated on the film. He et al. [137] investigated the antioxidant activity of the green seaweed *Ulva lactuca*, the red seaweeds *Gracilaria lemaneiformis* and *Sarcodia ceyclonensis*, and the brown alga *Durvillaea antarctica*. Their results showed that crude polysaccharide from the green seaweed showed the highest activity, followed by *Durvillaea antarctica* and *Sarcodia ceyclonensis*.

Moreover, the inclusion of seaweed components in biofilm can lead to a photoprotective effect on packaging; some species possess photoprotective compounds capable of absorbing UV rays. Methanol extracts of *Sargassum* sp. and *Eucheuma cottoni* showed photoprotective activity against UV radiation, probably due to the presence of flavonoids, phenols, and triterpenoids [138]; they can potentially be used as a raw material for sunscreen products or be included in biofilm formulation, enhancing the properties of active packaging.

Although the biological activities of pure seaweed compounds/extracts incorporated into biofilms should be further investigated, several studies have shown that most seaweed polysaccharides exhibit antioxidant, antimicrobial, and photoprotection activities, suggesting that their inclusion in biofilms can lead to the manufacture of safe and active packaging.

5. Future Perspective and Conclusions

The interest in seaweed hydrocolloids grows, and they have been considered potential candidates to produce biodegradable plastic. In support of this, the London start-up company Notpla has created a natural, edible, plastic-like material that can biodegrade within four to six weeks. The membrane is made from seaweed farmed in northern France; first, it is dried and ground down into powder, and then transformed into a thick, gloopy fluid, which dries to form a plastic-like substance. This natural plastic could lead to a decrease in plastic pollution all over the world, which generates 300 million tons of plastic waste each year. Another project involving sustainable plastic is the project Mak-Pak, launched by the collaboration between researchers of Alfred Wegener Institute, Hochschule Bremerhaven University, and the fast-food chain Nordsee. This project developed the production of a sustainable macroalgae-based packaging for fast food, which has been already tested by real consumers.

Nevertheless, packaging made from pure phycocolloids does not meet the criteria for bioplastic of commercial use, due to their poor mechanical and water barrier properties. The addition of biopolymers or plasticizers will improve the performance of the biomaterial. The antioxidant and antimicrobial properties of seaweed-derived bioactive compounds will also improve the maintenance and shelf-life of food and medicines.

The use of seaweed for biodegradable, safe, and hygienic packaging attracts the interest of the marine pharmacology and food industry, which may not only exploit the potential of seaweed-derived bioactive compounds for the development of novel and natural drugs, or healthy food products and supplements, but also provide a sustainable packaging that preserves them and does not alter their properties, in a sustainable and eco-friendly way.

Unfortunately, the traditional extraction of phycocolloids has some disadvantages, which can be solved by the adoption of more eco-friendly extraction methods, which are already proven; however, they still need continuous research to reduce implementation costs and to persuade industrial investment and further studies to develop these sustainable technologies.

The potential of seaweed-derived bioactive compounds is of great interest for the development of sustainable and eco-friendly bioplastic, which can be seen as a solution for replacing petroleum-based plastics and to avoid the large production of plastic waste.

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