Seaweeds in Producing Safe and Sustainable Bio-Packaging

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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.

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

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][6][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][10][11], aquaculture feed, biofuels, application in wastewater treatments, or to produce innovative and ecologic materials to replace plastic equivalents [12][13][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 to find new, eco-friendly material to develop biodegradable packaging [15][16][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 the 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][23][24][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][28][29][30][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][37][38][39][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.

2. 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 [42]. 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 [43], 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			

Seaweed	Extract/Compound in Biofilm Formulation	Biofilm Mechanical Characteristics	Reference
Sargassum siliquosum	Alginate	Adequate tensile strength, elongation at break, water vapor permeability and water solubility due to addition of $CaCl_2$	[44]
Sargassum natans, Laminaria japonica	Crude extracts	Enhanced physicochemical, mechanical, and thermal properties due to addition of cellulose nanocrystals	[45]
Phylum Rhodophyta			
Kappaphycus sp.	Crude extract	Reduction in brittleness, weak tensile strength	[46]
Kappaphycus alvarezii	κ-carrageenan	Evidenced good physical, mechanical and thermal strength of bioplastic films	[47]
Eucheuma cottonii	Semi-refined carrageenan	Biofilm with refined carrageenan showed higher tensile strength and thermal resistance compared with semi-refined carrageenan	[48]
	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	[49]
	Agar (alkali extraction)		
Gracilaria vermiculophylla	Agar	Transparency, clearness, flexibility, and mechanical strength enhanced by addition of glycerin	[50]
Eucheuma spinosum	Crude polysaccharides	The addition of glycerol as plasticizer enhances biofilm physical and mechanical properties	[51]

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 [52]. Alginate, agar, carrageenan, and cellulose from seaweeds have shown excellent film-forming properties, and are very easy to process [53].

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 [54].

Lim et al. [44] 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 research suggested that alginate from *Sargassum siliquosum* as a suitable candidate for the synthesis of bioplastic films [44].

Doh et al. [45] 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 [46] 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 [46]. 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. [47] 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 [48].

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 [49]. Therefore, agar extracted from *Gracilaria salicornia* is interesting for future possibilities in commercial applications of bioplastic films. A study by Sousa et al. [50] reported that biofilm obtained using agar from *Gracilaria vermiculophylla* showed transparency and clearness similar to the commercial counterpart [55]. 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 [56].

Bioplastics were synthesized recently by Darni et al. [51] 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 [57]. 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 [58].

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 [59]. 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 [60]. 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. [61], 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* [62][63]. In addition, crude polysaccharides extracted from red alga *G. ornota* showed antimicrobial activity against *Escherichia coli* [64]. The antimicrobial activity of pure carrageenan did not exhibit an effect against *S. aureus, Escherichia coli*, or *L. monocytogenes* [65][66]. Meanwhile, in the study of Kanatt et al. [67], 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 [67]. Lipid oxidation has a strong impact on food quality; it can lead to a decrease in shelf-life and nutritional value of food [68]. 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. [69] 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 [70]; 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.

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