CARRAGEENANS AS A MULTIFACETED BIOMATERIAL IN THE ERA OF BIOMEDICAL AND ENVIRONMENTAL INNOVATION

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ABSTRACT

Carrageenans are biomaterials of great interest due to their structural variability and physicochemical properties. This article provides a detailed review of the structural classification of carrageenans and its correlation with physical properties and specific applications, focusing on the various materials that can be obtained from them. Processing and modification methods are discussed to obtain gels, nanoparticles, films, and coatings, which possess unique properties that make them suitable for a wide range of applications. In the biomedical realm, carrageenans excel as scaffolds for tissue engineering, drug delivery systems, and wound healing materials, taking advantage of their biocompatibility and ability to form gels. In agriculture, they are used for the controlled release of fertilizers and pesticides, as well as biostimulants and soil improvers. In the food industry they act as thickening, stabilizing, and gelling agents in a wide range of products, from dairy to meat products, conferring texturising properties and improving stability. In addition, their role in environmental remediation is addressed, where in addition to their uses in agriculture to reduce the impact of different compounds, and water consumption, they are used for the removal of pollutants and water purification, thanks to their ability to form complexes with heavy metals and other pollutants.

This review highlights the potential of carrageenans to inspire future innovations in the design of materials and technologies, underlining the importance of exploring and fully exploiting their potential and that of their derivatives to address current and future challenges in various sectors.

Keywords: Carrageenans, biopolymers, applications, medical, environment, foods.

1. INTRODUCTION

The dynamic convergence between biotechnology and materials science has captured the attention of researchers and practitioners from various disciplines in the search for functional and sustainable biomaterials (1). Among these, carrageenans (CAR), a sulfated polysaccharide extracted from red algae has emerged as an exceptionally versatile component that goes beyond its conventional application in the food industry (2). Their ability to form gels, along with their biocompatible properties, have driven the exploration of CARs as an innovative biomaterial for various biomedical, pharmaceutical, biotechnological, agricultural, cosmetic, and environmental applications (3).

CARs are sulfated galactans that constitute one of the most abundant components in the extracellular matrix of red carragenophyte algae. These linear polysaccharides are formed by the repetitive arrangement of a disaccharide (carrage) pattern. These repeating units are formed from a β-D-galactose (G-unit) with 1,4 glycosidic bonding binding to a α-D-galactose (D unit) or a 3,6-anhydrous α-D-galactose (DA unit) (4). The presence or absence of the 3,6-anhydrous bridge in the α-D-galactose unit and the variability of the sulfation pattern and substitution of glycosyl, pyruvate and methoxyl groups in the polysaccharide give rise to different properties such as gelation, viscosity of the medium and selective absorption capacity of cations (5,6).

This particular chemical structure and the variability of CARs has meant that since their discovery they have been widely used in the food and cosmetics industry as one of the main texturizing agents (*i.e*. gelling, thickening or stabilizing), being necessary in the production of shampoo, beverages, dairy products and even toothpastes (7). However, due to its antioxidant, biodegradable properties, biocompatibility, and ability to form three-dimensional matrices, its use has been explored in recent years in different areas such as biomedicine, as a promising material in tissue regeneration or obtaining wound dressings, and tissue engineering (8–10), biotechnology and agriculture, for the controlled release of drugs, nutrients or biostimulants (11), and environmental remediation, due to its versatility in the removal of aquatic pollutants given the affinity for various compounds, expanding its use in water purification and mitigation of environmental pollution (6,9,12–14).

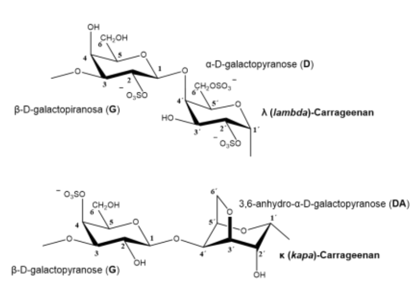
In this context, CARs stand as a central player in the 21st century biomaterials narrative, offering imaginative solutions to increasingly pressing biomedical and environmental challenges. By diving into this in-depth analysis, we will uncover the wealth of opportunities that CARs offer, opening the door to innovations that could transform the way we address crucial health and sustainability issues in the contemporary era. That is why throughout this exhaustive review, we will critically explore the properties and characteristics that make CARs a multifaceted biomaterial, examining pioneering studies and recent advances in the development of polymer-based biomaterials and highlighting their applications in different industries such as biomedical, food, agricultural, and environmental.

In addition, the current challenges and projections of this type of material are considered, identifying promising areas of research that could fully exploit the potential of this ever-evolving biomaterial.

2. Types of carrageenans and their structural characteristics

CARs are a group of linear sulphated galactans extracted from different species of Rhodophyta (red algae) such as Gigartina, Chondrus crispus, Eucheuma and Hypnea, and the type of CAR extracted varies depending on the reproductive phase of the plant (15–17). CARs are composed of alternating ring structures β-D-galactopyranose (G) linked 1-3 with α-D-galactopyranose (D) or 1-4 with 3,6-anhydrous α-D-galactopyranose (DA) (16,17). In 1953 a procedure for the fractionation of the CAR was suggested, initially obtaining two components that were called κ (kapa) and λ (lamda), from which new types of CAR were obtained that were designated with Greek letters without any specific system, and which were classified according to the presence of the 3,6-anhydrous bridge in the galactose residue and the position and number of sulfate groups.

The nomenclature for galactans based on their chemical structures was suggested in 1994 by Knutsen, who used letters that allowed the systematic description of the molecules, proposing two groups based on the basic repetitive backbone of the polymer, separating into carrageenans, as those polymers that are characterized by the alternation of G and D rings, and carragenoses, from the G and DA rings as shown in Figure 1 (18,19).



Greek name	Greek letter	Letter code
gamma	γ	G-D6S
delta	δ	G-D2S,6S
lambda	λ	G2S-D2S,6S
mu	μ	G4S-D6S
nu	ν	G4S-D2S,6S
xi	ξ	G2S-D2S
omicron	ο	G4S-D2S
psi	ψ	G6S-D6S
beta	β	G-DA
alpha	α	G-DA2S
theta	θ	G2S-DA2S
iota	ι	G4S-DA2S
kappa	κ	G4S-DA
omega	ω	G6S-DA

Figure 1. Nomenclature of carrageenans.

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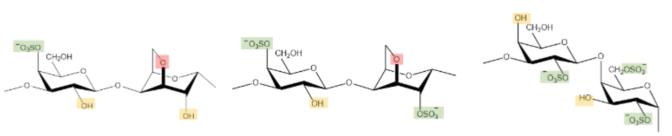
As a result of the fractionation procedures of galactans, the development of chemical, physical and enzymatic methods for their structural analysis made it possible to determine the structures of several CARs and to hypothesize the "masked repetitive structure", i.e., inherent in all galactans, which plays a determining role in their gel-forming properties (19). The exploration of these methods allowed us to identify much more complex structures of sulfated galactans, structural diversity and to find correlations between the structure of the polysaccharides and the taxonomy of the algae used for their extraction. Specifically, nuclear magnetic resonance imaging has been used to analyze the fragments and has been additionally coupled with other chromatographic methods such as gel permeation chromatography (GPC) or size exclusion chromatography (SEC) (17,20).

This is evidenced, for example, in research such as that carried out by Stortz and Cerezo, who strove to characterize around 42 repetitive units of different CARs, accounting for the chemical displacements of each of these types (20). This great variety of polysaccharides is traditionally divided into six basic forms: Iota (ι)-, Kappa (κ)-, Lambda (λ)-, Mu (μ)-, Nu (ν)-, and Theta (θ)-CAR, which are predominantly obtained by extraction from algae such as Kappaphycus alvarezii (Eucheuma cottonii), Eucheuma denticulatum (Eucheuma spinosum), Gigartinaceae, Petrocelidaceae, Phylophoraceae, and Chondrus, and even in some cases by transforming biological precursors into other carrageenans. as is the case with the use of μ and ν-CAR to obtain κ and ι-CAR (6,19–22).

In addition, it has been also identified that some gametophytic plants can produce a type of hybrid carrageenan (κ/ι), which consists of mixed polysaccharide chains containing κ and ι-CAR units that vary across a spectrum of purity from one to the other (5,22). Among those already mentioned, the most commercially important are κ, ι and λ, whose repeating unit has one, two, and three sulfate ester groups with calculated sulfate contents of 20%, 33%, and 41% (w/w) respectively (see Figure 2). It should be noted that in each of these types there are some variations since, in addition to galactose and sulfate, other carbohydrate residues, such as xylose, glucose, and uronic acids, may be present in CAR preparations, as well as some substituents, e.g., methyl ethers and pyruvate groups (22–24).



κ (Kapa)- Carrageenose (G4S-DA) ι (iota)- Carrageenose (G4S-DA2S) λ (lambda)-Carrageenan (G2S-D2S,6S)

Figure 2. More commercial carrageenan structures.

Different characteristics and properties of CARs such as solubility, viscosity, and gelling capacity have been evaluated, identifying that these are closely related to the amount and position of sulfonic groups, as well as factors such as the balance of cations in the aqueous solution (22). CARs are generally soluble in water, and insoluble in organic solvents, oils or fats, this characteristic depends essentially on the concentration of sulfate groups, the presence of anhydride bridges, type of associated cation, pH and temperature (25). For this reason, carrageenan-type CARs show a high hydrophilicity attributed to the presence of sulfate and hydroxyl functional groups, while the hydrophobicity of carrageenans originates from 3,6-anhydrogalactose bonds (26). On the other hand, the viscosity and gelling capacity in the presence of salts depends on the degree of formation and aggregation in the form of helices, and in the case of carrageenans, the sulfates in DA units are less favorable for interhelical aggregation than in the D units, which can be evidenced by the weak gelling capacity of α-CAR and the strong capacity of κ-CAR (27).

In terms of gel-forming capacity, each type of CAR has distinctive gelling properties, so the influence of the number and position of sulfate groups has been studied, as well as the effect of the counterion and its valence. In this regard, it has initially been found that the gel-forming capacity decreases with the increase of sulfate groups, so the gels obtained from κ-CAR are hard, strong and brittle, soft and weak ι-CARs, while λ-CAR has a low gelation capacity and only shows viscous behavior (25,28,29).

Regarding the effect of counterions and their valence, it has been found that some types of CAR exhibit a behavior of severe syneresis in the presence of some salts, so research shows that the influence of the valence of the contraion depends on the position and number of the sulfate group, being those CARs that alternate G-DA units. It has been found that despite the low gel-forming capacity of λ-CAR, in the presence of trivalent ions, it has been found that it is feasible to obtain them (27–29) (23).

The effect of temperature has been studied, finding that gel formation is essentially driven by phase separation that occurs when cooling a hot CAR solution in the presence of salt, causing polysaccharide chains to undergo a spiral-helix transition, where helices aggregate and form domains that permeate a three-dimensional lattice (30). This is observed in systems such as κ and ι-CAR, which have two forms during gel formation, the first is an unstructured random spiral, which occurs at elevated temperatures; and a structured double helix that usually takes place under cooling. Additionally, it has been found that the large thermal hysteresis between the setting and the fusion of the gel in the case of κ-CAR, suggests a superhelical gel structure, while the lack of this in ι-CAR gels indicates less helix aggregation or helical networks. This supports that the cross-links in the network are formed by double helix (5,31). It has been observed that there is a specific temperature where the random double helix coils are formed, which takes place during this cooling process, this is called the coil-helix transition temperature, and is an important parameter in the functional properties of CARs, so it allows us to think that temperature has effects on the conformational transition (24,27).

2.1. Biomaterials from carrageenans

CARs, due to their remarkable gel-forming capacity, the presence of a variable number of functional groups in their structure, and their inherent biocompatibility, have stood out as a promising candidate in the field of biomaterials, opening up a range of applications ranging from the food industry to tissue engineering and biomedicine (32). This marine biopolymer has become the focus of various breeding and modification strategies to develop functional materials, such as gels, micro and nanoparticles, films, and coatings, each adapted to specific needs through the application of chemical modification techniques (33,34).

These modifications not only improve the properties of CARs but also extend their range of applicability, demonstrating their versatility and potential in fields as diverse as agriculture and controlled drug release (35).

In this context, chemical modification emerges as a key strategy for the advancement of carrageenan-based biomaterials, offering the possibility of synthesizing derivatives with precise and improved characteristics. Techniques such as cross-linking, graphtization, and sulfation have emerged as fundamental methods in this process (36). Cross-linking, using agents such as metal ions or organic compounds, allows not only to improve the thermal and mechanical stability of gels but also to adjust their porosity and degradability, which are crucial for use in biomedical applications (37–39). On the other hand, graphtization facilitates the incorporation of lateral polymer chains, allowing the introduction of new functional groups that improve the interaction of the biomaterial with cells and biomolecules, and enabling the development of drug delivery systems sensitive to environmental stimuli such as changes in pH or temperature (40,41). Finally, the sulfation technique, which increases the negative charge density of carrageenan, significantly improves its solubility and ability to interact with proteins and other polymers (42). Thus, the sulfation technique, which increases the negative charge density of carrageenan, significantly improves its solubility and ability to interact with proteins and other polymers (42). This aspect is particularly relevant for the creation of more effective encapsulation matrices, allowing more detailed control over asset release and offering protection against unfavourable environmental conditions (43).

The implementation of these chemical modification techniques not only stands out for opening new paths in the functionalization of carrageenans, but also for allowing the customization of biomaterials for specific applications (44,45). The ability to design the structure and functionality of carrageenan derivatives at the molecular level represents a significant advance, offering ample potential for the development of innovative new materials adapted to the emerging needs of society and industry. (39,44,46–48)

2.1.1. Biomaterials in the form of gels

The production of carrageenan gels involves fundamental processes such as macromolecular gelation and ionic gelation. In macromolecular gelation, carrageenan chains, which are linear polymers, undergo a three-dimensional reorganization through covalent bonds or physical interactions, generating a gel with specific properties. This macromolecular gelling can be facilitated by gelling agents, such as metal salts, natural polymers or chemical agents, resulting in a cohesive three-dimensional structure (49). The resulting morphology of this gel exhibits remarkable physical and mechanical properties, including strength, elasticity, and water-retaining capacity (48,50). This method finds multifaceted applications, from the production of food, such as jellies and desserts, to its implementation in biomaterials for the controlled release of drugs.

At the same time, ionic gelation involves the formation of a three-dimensional gel, by interacting with metal ions such as calcium or sodium, to the polysaccharide dissolved in an aqueous solution, giving rise to a biomaterial with unique properties determined by the ionic bonds formed (51). The CAR, which is naturally anionic, forms ionic complexes with the metal ions present in the solution.

These complexes induce the formation of bonds between CAR chains, resulting in a three-dimensional gel structure (52). This resulting gelatinous lattice confers unique properties to the material, such as water-retaining capacity, structural stability and mechanical strength, usable in a variety of applications (53). In the biomedical field, gels formed from CAR by ionic gelation can serve as matrices for controlled drug release, scaffolds for tissue engineering (54), or coatings in medical devices (55,56). This versatility makes ionic gelling a valuable technique in the manufacture of biomaterials with specific and controlled properties (57).

The versatility of CARs expands towards the formation of hydrogels, a strategic modification to confer hydrophilic properties and improve the water absorption of biomaterials in the form of gels, obtaining three-dimensional structures capable of swelling while retaining their shape for extended periods of time (58,59). CAR hydrogel engineering involves a chemical or physical crosslinking phase, in which cross-linking agents, such as glutaraldehyde, and physical interactions such as hydrogen bonds or van der Waals forces play a fundamental role to give rise to a three-dimensional structure, essential to enhance the hydrophilic properties and water-holding capacity of the resulting hydrogel (60–62).

The fundamentals of this technique lie in the exact modulation of molecular interactions in the CAR polymer lattice. The proper choice of crosslinking agent, the type of modification applied to the biopolymer, and the reaction conditions are crucial to control the final properties of the hydrogel, such as absorption capacity, mechanical strength, and thermal stability (61,63). Understanding the mechanisms underlying the formation of the CAR gel during crosslinking is essential to optimize process efficiency and ensure the reproducibility of the desired properties in the resulting hydrogels (64). This integrated process, from the production of CAR gels to the engineering of hydrogels, demonstrates the versatile application of this polysaccharide in the development of polymeric materials with specific properties, offering a solid basis for various applications in medicine and industry (65,66).

2.1.2. Biomaterials in the form of micro and nanoparticles

CAR nanoparticle research and development has gained significant interest in fields as diverse as controlled drug release, biomedicine, and the food industry, given their ability to offer innovative and effective solutions in these areas (67,68). Unlike the gels described above, which are characterized by more extensive three-dimensional structures, nanoparticles of this polysaccharide have unique advantages such as their small size which, combined with a high specific surface area, allows for more precise manipulation of their properties, including the ability to modify their surface to suit specific functions, thus expanding their applicability in various areas (69).

This breakthrough translates into greater efficiency in applications where penetration and absorption at the cellular level are crucial, significantly improving interaction with active compounds. This not only enhances therapeutic efficacy in the field of health, but also optimizes functional properties in the food industry, offering new possibilities for product and process improvement (52,70). The ability to alter the surface of these nanoparticles paves the way for the design of specific nanocarriers, which can be targeted for precise drug delivery to specific tissues or to increase the stability and solubility of active ingredients in food (71).

Various techniques have been used for the synthesis of these nanoparticles, including the ionic gelation mentioned above, a technique that exploits the electrostatic interaction between CAR polysaccharide chains and metal ions, such as calcium (Ca^{2+}). When aqueous solutions of CAR are mixed with metal ions, the ions act as bridges between polymer chains, resulting in the instantaneous formation of a gel (72). This process is highly controllable and allows the formation of gel particles on the scale of nano to micrometers by adjusting the concentration of CAR and ions and mixing conditions. Therefore, it is particularly useful for encapsulating active compounds, protecting and releasing them in a controlled manner (73,74).

Emulsification and atomization are techniques that can follow ion gelation to produce even smaller particles. Emulsification involves finely dispersing a liquid phase (the CAR solution) into another with which it is immiscible, using emulsifying agents to stabilize the droplets formed (75). When the ionic solution is introduced, the droplets gel, forming nanoparticles (76). Atomization, on the other hand, uses mechanical forces to create fine droplets from a CAR solution that then solidify upon contact with an ionic solution or upon rapid drying (77). Both techniques allow precise control over the size and distribution of the particles formed (74).

Beyond ionic gelling, CAR nanoparticle production has benefited from top-down and bottom-up approaches, each with its own advantages. Mechanical milling is a top-down approach that involves the physical decomposition of solid materials into smaller particles using ball mills (78). This process can generate a wide range of particle sizes, including the nanometer range, by applying extreme mechanical forces (79). Critical variables include mill type, milling duration, ball size, and the mass-to-volume ratio of the balls to the material. Although effective in producing nanoparticles, this method can introduce impurities and structural defects due to the collision of the balls with the material (80,81).

While the Sol-Gel method is a bottom-up strategy that involves the transformation of monomers into a sun (a colloidal suspension of particles) that is then induced to form a gel (a solid three-dimensional lattice) through polymerization and crosslinking processes. This method allows exceptional control over the composition, morphology, and size of nanoparticles, facilitating the synthesis of materials with specific properties such as the encapsulation of active compounds, which allow controlled release in biomedical and food applications (80,82-83).

Each of these techniques offers unique advantages in the production of CAR nanoparticles, from precision in size and morphology to surface functionalization and encapsulation of active compounds. The choice of the appropriate method will depend on the specific objective of the application, considering factors such as the stability of the active compound, biocompatibility, and the efficacy of the release (52,84-85).

2.1.3. Biomaterials in the form of films and coatings

The use of CARs to develop films and coatings has captured interest from various areas, especially due to their potential to improve safety and extend the shelf life of food and pharmaceutical products (86-87). This fascination is due to the unique characteristics of CARs, which allow the formation of flexible, transparent structures with outstanding mechanical properties. These films can be modified to adjust their barrier properties, making them more effective against water vapor, gases, and lipid (88,89). The incorporation of nanoparticles or bioactive agents into these films introduces antimicrobial and antioxidant properties, resulting in active barriers that protect the food beyond the simple physical barrier, extending its freshness, and quality (87,90).

The process of obtaining these films and coatings has evolved with processing techniques, allowing the development of biomaterials with specific properties adapted to particular needs (91,92). A commonly employed technique is solution casting, which relies on the ability of CARs to form viscous solutions when dissolved in hot water. The addition of plasticizers such as glycerol modifies the structure of the gel formed as it cools, allowing the creation of flexible films that easily adapt to different surfaces. This method is ideal for applications that require consistent and accurate coverage (93,94).

Immersion coagulation is another relevant technique, which uses the property of CARs to gel in the presence of certain cations, such as potassium and calcium. This method is particularly useful for forming coatings around irregularly shaped objects or for encapsulation of substances, offering precise control over the texture and mechanical strength of the gel (95,96). In contrast, dip coating focuses on immersing the product in a CAR solution and then gelling it to form a thin protective layer, an especially valuable method for extending the shelf life of fruits and vegetables by forming a physical barrier that minimizes moisture loss (97–99).

Continuous innovation in the field of biomaterials has led to the exploration of advanced techniques such as electrospinning and 3D printing, which offer the ability to fabricate structures with high specific surface area, controlled porosity, and complex geometries. Electrospinning is a technique that allows the manufacture of films and coatings in the form of ultra-fine fibers, offering a high specific surface and porosity (96). The process involves applying a high voltage to a polymeric solution of CAR, generating a jet that, when the solvent evaporates, forms continuous fibers that are collected on a substrate (100,101). The rationale behind this technique lies in the ability to create nanometric structures that can mimic the extracellular matrix, being especially valuable in biomedical applications such as advanced bandages or scaffolds for tissue engineering (102).

3D printing has been also explored for the fabrication of different types of CAR structures using techniques such as fused deposition printing or stereolithography, it is possible to design and produce films and coatings with specific mechanical and controlled-release properties (103). The principle of this technique is based on layer-by-layer deposition of CARs, allowing the creation of three-dimensional structures with micrometer precision. This opens up new possibilities in the design of drug delivery systems and in the creation of active food packaging (104–106).

Finally, lamination is discussed as a method that involves assembling multiple layers of CAR films to improve mechanical strength and barrier against gases or vapors. This process can be combined with other biodegradable materials to obtain complementary properties (107,108). Lamination is based on the creation of multi-layer structures that can optimize the functionality of the coating or film, such as improving protection against moisture loss in food products or increasing resistance to oxygen penetration, crucial for preserving the quality of perishable foods (109,110).

These techniques expand the repertoire of methods available for the development of CAR films and coatings, highlighting the versatility of this polysaccharide as a biomaterial. A detailed understanding of the rationale for each technique is crucial for optimizing manufacturing processes and achieving the desired properties in the final product (91,111,112).

2.2 Application of biomaterials from carrageenans

2.2.1. Biomedical industry

In the biomedical field, the application of carrageenan as a biomaterial has stood out significantly in various areas, such as tissue engineering, regenerative medicine, and drug delivery (see Figure 3). Its ability to form three-dimensional arrays has boosted its utility, providing a structural and biochemical environment conducive to cell growth. The versatility of carrageenans is manifested in their ability to improve the efficacy of specific therapies by responding to external stimuli, such as temperature variations or the presence of ions, which expands the possibilities for applications in the biomedical field (8–11,21,113).

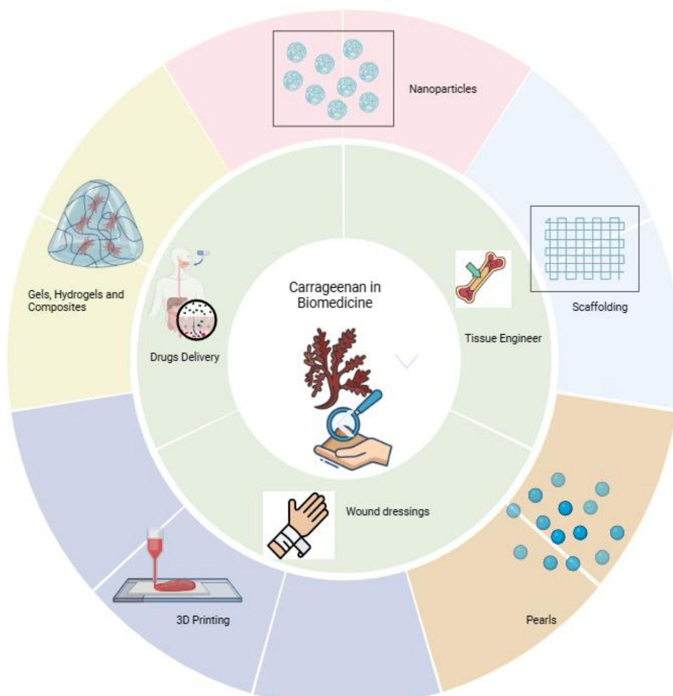


Figure 3. Biomaterials from carrageenan applied in biomedicine

One of the most promising applications of carrageenan in biomedicine is in tissue engineering and regeneration. Due to their gel-like structure, CARs can be used as scaffolds for cell growth and differentiation, promoting tissue formation in 3D structures. This is especially useful in the regeneration of soft tissues, such as skin and cartilage, where their ability to retain water and maintain a moist environment promotes cell healing and regeneration. A number of studies have reported the effectiveness and usefulness of CARs in this area, as they provide essential physical signals for cellular functions as well as directing bioactive molecules to target cells or damaged tissues (114,115).

For example, studies on 3D scaffolds of κ-CAR, chitosan and gelatin have been reported by Loukelis et al. in which mechanical and biological properties were evaluated in the engineering of bone tissues in which porosity percentages greater than 80% were achieved, in addition to showing in biological tests the significant proliferation of pre-osteoblasts in about 10 days. as well as the increase in calcium production, thus corroborating the influence of CARs in improving the biocompatibility and anticoagulant characteristics of the material (116). In addition, Kumari et al. reported the 3D bioprinting of CAR hydrogels for tissue engineered scaffolds loaded with bioactive molecules based on digital light processing for rapid fabrication of the complexes as an alternative for transplants, a material that has excellent printability, as well as swelling, biocompatibility, and biocompatibility properties. Rheology and suitability for printing cells lives. The presence of CAR is an excellent option in this material to improve the viscoelastic properties of the material that allow it to improve its shear behavior as well as its ability to recapitulate biomedical properties of soft human tissues (117). Similarly, bioactive porous scaffolds of CAR, combined with alginate and calcium silicate reported by Sathain et al., as well as the one mentioned above, show materials with better bioactive characteristics, but an increase in their mechanical properties of up to 50%, in deformation tests, as well as an increase in the capacity of bone induction due to the prescience of CARs, so they have been considered as promising candidates in the field of bioactive scaffolding. bone replacement (118).

The obtaining of injectable tissue repair hydrogels by combining CAR and green graphene, with better stability and mechanical properties that also recover their initial shape after eliminating applied stresses, as well as an increase in biocompatibility with fibroblasts that therefore translates into an increase in cell proliferation within the three-dimensional structure of the gel in times of up to 48 hours, as well as the combination of CAR and nanoresidonate for bone regeneration, which has exhibited exceptional characteristics in terms of its ability to swell and release maximum at neutral pH and temperatures of 37°C, are some of the innovative examples of the applications of biomaterials from CAR in tissue engineering (119,120).

In addition, carrageenan hydrogels have been investigated as controlled drug release systems. Its porosity and chemical composition allow the incorporation of various therapeutic agents, including drugs, proteins and living cells, which are released in a sustained manner at the desired site. This minimizes side effects and improves the efficacy of treatment, being especially relevant in the localized administration of treatments for cancer, inflammatory diseases, and chronic ailments.

Intelligent systems for drug transport use new polymeric materials with many advantages over conventional passive carriers for the treatment of bacterial diseases, cancer treatment, among others. Dozens of CAR-based smart biomaterials have recently been reported, an example of these being the use of antivirals or antibacterials as non-chemical drugs for the treatment of antiviral or antibacterial biomaterials. Together, they help prevent STDs, accelerating wound healing and reducing inflammation in infected wounds, thus being a multifunctional material with great applicability (21,121).

Other studies have also used CARs as a support matrix, however, they have varied the type of nanoparticles incorporated within the three-dimensional matrix such as chitosan and montmorillonite for cancer treatment, materials that have increased treatment efficiency by up to 96%, depending on the pH used in preclinical studies influenced by magnetic stimuli (122). The use of CAR nanospheres that respond to stimuli such as pH, temperature and magnetic fields is also reported, biomaterials that have significantly extended the half-life of drugs from 20 min to 4 h, thus increasing their release to 100% (123). Campos-Sánchez and his collaborators reported the use of λ-carrageenan as an immunosuppressive activator and anti-inflammatory of cantharidin in kidney leukocytes, the results showed the viability of the compound increased in peroxidase activity when incubated with CAR inhibiting respiratory burst and phagocytic activities, providing a detailed view of the improved properties of the material and its ability of the therapeutic compound to be transported by the material (124).

Another emerging field is the use of carrageenan in the manufacture of biomedical devices, such as bandages and wound dressings. These products take advantage of carrageenan's hemostatic and antimicrobial properties to speed up the healing process, reduce the risk of infections, and promote the formation of healthy tissue. In addition, their ability to form flexible, water-resistant films and coatings makes them ideal candidates for protecting wounds and burns during recovery, as the increasing incidence of skin wounds is a significant medical concern, prompting extensive research in the search for effective dressings. A study conducted by Narayanan et al. addressed this issue by evaluating the use of carrageenans as a support for isolated lytic coliphage, using bacteriophages as efficient antibacterial agents, in the creation of compatible heme hydrogel dressings for wounds. The results were remarkable, with latency periods of up to 10 min and a significant reduction in bacterial growth after two h. The presence of carrageenan positively influenced the compressive strength of the biomaterial, giving it greater elasticity. This approach presents itself as a promising and fascinating alternative for the treatment of wounds contaminated with bacteriophage agents (9). Similarly, the use of CAR as a bioactive membrane infused with reduced nanocomposites of graphene oxide and hydroxyapatite for wound reconstruction has shown improvements in wound healing processes in zebrafish models, thus corroborating the influence of CARs in improving membrane biocompatibility and strength, increasing the versatility of the material (125). Sathuvan et al. reported the development of a mechanical wound-detecting and wound-healing film from essential oil and CAR, which has an initial release with cellular adhesive capabilities that promotes wound healing *in vivo* (126). Carrageenan-based biomaterials are opening new frontiers in biomedicine, offering innovative solutions for tissue regeneration, controlled drug release, and advanced biomedical device development. Its sustainable nature, along with its unique properties, promises a future where regenerative medicine and personalized treatments become a more accessible reality for all.

2.2.2. Agricultural industry

In recent years, the agricultural industry has been concerned with meeting the increased demand for food and improving crop yields, while reducing the environmental impact of fertilizers, pesticides and the use of water resources for irrigation. Although chemical fertilization and pesticide use have increased yields per unit area of crops, traditional practices have proven to be insufficient because only a percentage of these substances are absorbed by crops, while others are lost through leachate, generating serious environmental consequences such as eutrophication and groundwater contamination (127–130). Moreover, considering that agriculture is one of the industries with the highest water consumption, where approximately 70% is used for irrigation, the use of materials with water absorption properties, which favor water retention in the soil, has been resorted to (131,132). Applications have been reported in the agricultural literature, including the use of hydrogels for the controlled release of fertilizers, pesticides, water retention and seed coating, as well as the application of carrageenans for crop protection and strengthening (see Figure 4) (132,133). Initially, coating with hydrogels or superabsorbent materials is an interesting alternative to reduce environmental impact given its slow rates of release of nutrients or active ingredients, compared to its uncoated analogues (127). This type of behavior is mainly attributed to the chemical or physical cross-linking mentioned in the previous sections, which produces a stable 3D matrix with interconnected superporous structures that create open channels that produce a capillary action, allowing the slow release of nutrients or pesticides by diffusion and providing a high water absorption capacity, acting both as a water reservoir and improving the physical properties of the soil (127,130).

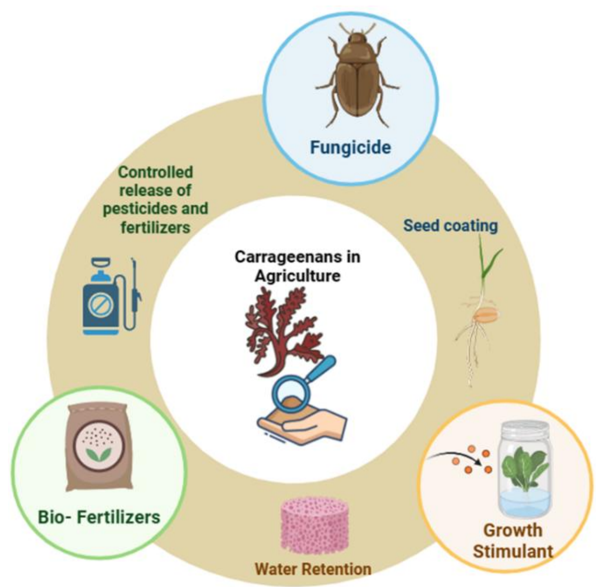


Figure 4. Biomaterials from carrageenan applied in agriculture.

Coatings of fertilizers and pesticides based on κ-CAR hydrogels or mixtures of κ-CAR with other natural polymers such as chitosan have been proposed for the production of different types of plants including tubers, bok choy, and pak choi (127,134,135). These investigations concluded that uncoated fertilizer granules released 12 to 18% less $N\text{-}NH_4^+$ and K^+ than coated fertilizer granules, and that they did not affect plant growth or tuber quality (127). Meanwhile, experiments at both laboratory and field level on pesticide release showed that after 25 to 30 days, the maximum amount of active ingredient was still in the soil, in addition to evidence of increased pesticide stability, protection from biotic and abiotic stress, and increased bioactivity against biotic stress compared to its uncoated analogues (134).

The effect of pH on the processes of swelling of materials and controlled release of different compounds has been studied, finding that carrageenan-based materials exhibit an inverse relationship with pH, evidencing a maximum swelling in acidic media (140,141). In the specific case of the release of NH_4^+, NO_3^-, and PO_4^{3-} ions, it was found that the release mechanism responds to a Korsmeyer-Peppas model, under a non-Fickian diffusion mechanism, in addition to the fact that there are no fractures in the polymer and a water absorption capacity of up to 300 times its weight in water is reached (130).

The use of hydrogels as water adsorbents in this industry to reduce the impact of irrigation, began in the 1980s, and has been highlighted in recent years, finding studies that report polyacrylamide-based materials with the capacity to store 95% of soil water, and even proposing superabsorbent polymeric materials, that enable the growth and development of plants in arid regions (132,136,137). That is why, recognizing the possibilities offered by this type of materials, biodegradable and low-toxicity alternatives have been sought, intensifying the development of research that proposes new environmentally friendly absorbent materials based on natural polymers such as cellulose, carrageenans and starch derivatives (138,139). Some studies report hydrogels composed of carrageenan/psyllium integrated in montmorillonite, used as a soil conditioner in the agricultural sector, a material that showed high degrees of swelling (2893%), and an increase in gel resistance, reaching (226 kPa), water retention capacity, and soil water percentage by higher than that 60% (131).

Other types of research focus on the use of purified marine algae polysaccharides and derived oligosaccharides as an important source of metabolites necessary to survive hostile, diverse, and competitive environments, in addition to their ability to trigger defense responses in plants and antimicrobial activity (135,142). For this reason, the use of seed coating, production of elicitors and carrageenan-based stimulants for crop protection and introducing the desired resistance in plants by activating or provoking their natural defense system, as well as promoting their growth has been studied (143–146). Among the research found aimed at the protection and promotion of plant growth, we find the use of λ or κ-CAR in the growth of banana and turnip plants (at the laboratory level), where it was concluded that vegetative growth, number of leaves, nutrient and pigment content, plant height, root length, pseudostem diameter, fresh weight, and soil fertility were significantly improved in those plants treated with λ or κ-CAR (142,146,147). The use of carrageenan was associated with structural improvements of the roots, which facilitated nutrient uptake, as well as increased chlorophyll, suggesting that this CAR increases the rate of photosynthesis, protein biosynthesis, and the biosynthesis of secondary metabolites that could eventually stimulate plant growth (146).

In the same vein, research studied the effects of elicitors made from k-CAR on the production of induced secondary metabolites, disease resistance capacity, and growth of chickpea and corn plants, finding that it can be used as a potent plant protectant, as well as as a growth-promoting agent, especially for chickpea plants (145,148). In addition, the fungicidal activity of κ-CAR in chili pepper plants was evaluated, where pathogenesis tests yielded a lower disease score for pretreated plants, indicating that this type of carrageenan extracts could be a potent natural fungicide and an inducer of disease resistance in plants (149).

Everything mentioned up to this point makes clear the versatility and diversity of applications that this type of materials can have in the agricultural industry, ranging from the preservation and nutrition of crops and soil taking advantage of properties of carrageenans, to the encapsulation and coating of different substances for their controlled release.

2.2.3. Food industry

The food industry has undergone a significant transformation in recent decades, driven largely by the continuous search for safe and sustainable alternatives to improve product quality. In this context, carrageenans have emerged as versatile biomaterials with revolutionary applications in this industry. These compounds have demonstrated not only their gelling and stabilizing properties, but also their ability to offer innovative solutions to specific challenges in food formulation, where their use for food encapsulation, edible sheet making, texturizing agents, and meat product formulation, etc. has been explored (see Figure 5) (150,151).

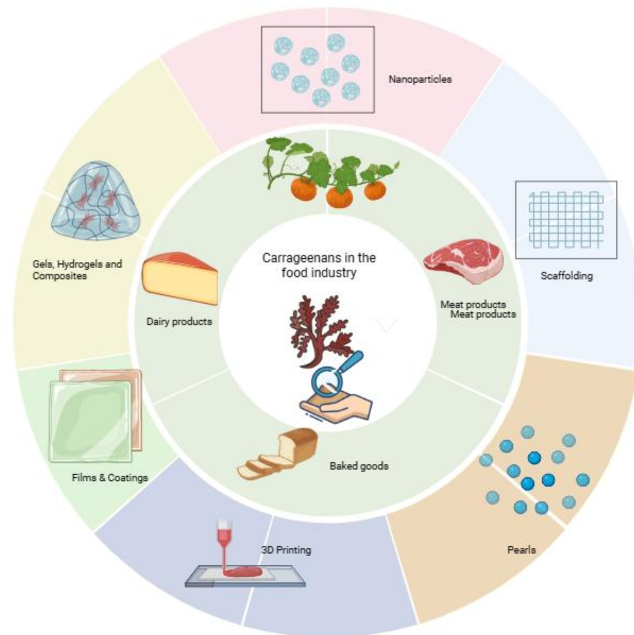


Figure 5. Biomaterials from carrageenan applied in the food industry.

Some of the publications that have been reported in recent years talk about the incorporation of CAR in the production of compound hydrogels, which represents an effective strategy to improve properties related to their field of application, because they improve the water retention capacity and stability of the materials obtained, as is the case of the study reported by Qiao et al., who explored the role of CAR in the fortification of gel structures that enhance the characteristics of a gel within an isolated soy protein system. This approach has resulted in robust gel structures applicable to the production of processed vegetarian and vegan foods, highlighting the significant improvement achieved thanks to the presence of CARs, demonstrating once again that the presence of CARs significantly improved gel-related properties.

Likewise, Dille et al., combined bean protein concentrated with CAR in these gelling processes, obtaining materials with improved rheological properties at small amplitudes in times of up to 19 hours, showing percentages of syneresis of 2% at higher concentrations of CAR. These advances open up new prospects for the formulation of foods with improved properties and extended shelf life (152,153).

The search for sustainable alternatives to plastic packaging has led to the development of edible films based on CAR and pectin in the presence of lemon balm as an alternative to plastic packaging that extends the shelf life of products as well as their quality and protects them against moisture, oxygen. Research, by Zioga et al. reveals that these biomaterials offer exceptional performance in protecting against moisture and preventing oxidative deterioration. These films, stronger and more rigid than those compared without CAR are presented as a promising alternative to extend the shelf life of food products, protecting them against moisture, oxygen, taste, aroma, and mechanical damage. This approach not only promotes sustainability, but also contributes to the quality and safety of packaged foods (154).

Another example where the applicability of CARs in the food industry is highlighted is as a stabilizer of sensitive food inks where parameters such as pH, temperature limit the application of dyes such as the natural blue pigment phycocyanin of Arthrospira platensis, Buecker et al., explored CAR complexation to improve pigment stability at pH 2.5 to 6.0 at room temperature and after thermal processes up to 90 °C, where they also attributes this improvement in stability due to the electrostatic interactions that were favored at pH > isoelectric point by the high amount of sulfate groups in the CARs.

Therefore, the results revealed that this complexation favored the stability of the pigment under environmental conditions and after thermal processes, highlighting the potential of carrageenans in the prevention of color degradation in food products. This approach not only contributes to reducing food waste, but also encourages the use of renewable materials in the food industry, aligning with growing demands for sustainability (155).

One of the most notable aspects is the role of carrageenans in improving the texture and stability of dairy products. Research has highlighted that the addition of carrageenans to products, such as yogurts, cheeses, and desserts, not only improves consistency and creaminess, but also contributes to the prevention of syneresis, a common phenomenon in dairy products that experience liquid separation. These results underscore the ability of carrageenans to enhance the consumer's sensory experience and extend the shelf life of dairy products. Thus, CARs have also been proposed as a thermal stabilizer in long-term gel structures and a texture modifier in semi-hard fresh cheese, where CARs act by affecting coagulation processes and modifying the cheese gel network by increasing the salt release process, achieving softer textured products and a lower storage modulus compared to the control product (156). Similarly, the use of CAR in the production of emulsion gels for pickering stabilization has been reported to be applied in the production of structured gels in the form of microgel films, which have presented better stabilization without forming aggregates and an improved gelling capacity influenced by the presence of CAR (157).

In the realm of processed meat products, carrageenans have proven to be effective binding agents, improving water retention and providing juicier and more palatable meat products. In addition, carrageenans' ability to form thermoreversible gels has enabled significant advances in the creation of meat products with improved textures, thus contributing to healthier and more attractive options for consumers. In addition, carrageenans' ability to form thermoreversible gels has led to advances in the creation of meat products with improved textures, thus contributing to the creation of healthier and more attractive options for consumers (151). An example of this is the study carried out by Gu et al., who reported the cultivation of meat with the potential to alleviate ethical, environmental and public health problems derived from the conventional breeding of domestic species, by 3D printing pork meat based on CAR and glucomannan, in the form of edible hybrid hydrogels with excellent biocompatibility and food safety as well as low cost, where the established spheroid model opens up possibilities for wide applications in cultured fat and represents a viable strategy for meat manufacturing (158).

In the context of baked goods, it is crucial to achieve desirable characteristics such as crumb structure, long shelf life, high volume, and resistance to aging, so in terms of bakery and pastry products, carrageenans have found application as dough structure and stability improvers. Research has shown how the inclusion of carrageenans in the formulation of breads and similar products not only improves texture and prolongs freshness, but also acts as a moisture-retaining agent, preventing desiccation and maintaining the sensory quality of the product for longer periods (159).

The contribution of carrageenan biomaterials to innovation in the food industry is undeniable. From improving texture and stability in dairy products to acting as binding agents in meat products and dough improvers in bakery products, the versatility of carrageenans opens up a range of possibilities for the formulation of more attractive, healthy and sustainable foods. As the research and practical application of these biomaterials continue to evolve, carrageenans are expected to play an even more prominent role in creating the next generation of innovative food products (160–162).

2.2.4. Environmental Remediation

In the field of environmental remediation, the versatility of carrageenans extends from what has already been mentioned in the section on agricultural applications, where the impact of fertilizers, pesticides and other chemical species that affect water resources is reduced, to the removal of aquatic pollutants given the ability to form gels in the presence of metal ions and the affinity for various compounds that make this biomaterial a valuable tool in the purification of the environment, water and mitigation of environmental pollution. Although the gelling properties of CARs make them biopolymers of interest, this has limitations in terms of mechanical properties and durability, so different strategies have been addressed through the incorporation of different compounds, materials and even the use of different techniques to obtain biomaterials that favor their use for the removal of pollutants of interest, ranging from dyes, through various organic pollutants known as of emerging concern and reaching metals.

Dyes in aqueous media are considered risky due to their toxicity and implications for the natural processes of aquatic fauna and flora, as well as the difficulty of removal, since most dyes are stable and resistant to light, temperature, chemicals, and bacteria, thus evading conventional processes (163–165). Among the materials proposed for the removal of cationic and/or anionic dyes, we find aerogels, absorbent membranes, multilayer materials, among others. Different researches have evaluated the use of these different materials, finding that, for example, hydrogels based on polyvinyl alcohol (PVA) with three types of CAR (separately), drastically increased the removal efficiency of methylene blue, going from 6.3% removal with the pure PVA membrane up to 98.8%, 97.0%, and 95.4% removal, incorporating κ, ι, and λ-CAR respectively. The results were associated with steric impediment due to the presence of sulfate groups in the repetitive units, and where it is highlighted that this type of mixtures favor the reuse of the membrane with a sustained elimination of 98% of the contaminant after five adsorption-desorption cycles (166). This was also observed in multilayer materials of ι-CAR, a natural polysaccharide similar to pectin, which showed a high absorption performance compared to other biosorbent polymeric systems, which is associated with the presence of a large number of sulfonic groups, hydroxyl and carboxyl, which enhance interactions with cation-type contaminants (167). Aerogels have also been explored with this objective, they found research in which a novel material based on grapefruit peel powder with polydopamine/polyethylenimine/κ-CAR is proposed, whose porous structure, compressible surface charge and adjustable in pH, is an alternative for the removal of cationic and anionic dyes, given the capacity to form π-π interactions and hydrogen bonds, and whose adjustable surface load allows selective absorption (168). This type of effects favoring molecular interactions π-π both within the material and with contaminants, was observed in research such as the one developed by Majooni et al. (2014) who proposed the encapsulation of carrageenan/graphene oxide hydrogels in 3D printed scaffolds, also affecting the mechanical properties of the gel increasing by 300%. It provided greater specific surface area and absorption capacity, increased cross-linking by increasing the temperature of ionic coagulation (169).

Pollutants of emerging concern are a wide range of chemical substances ranging from cosmetics and pesticides to pharmaceuticals such as antibiotics, whose presence in the environment is not recent, but whose harmful effects on aquatic ecosystems are, resulting in the generation of bacterial resistance in the case of the latter. For this reason, considering the harmful health effects of this type of substances in water, attention has been focused on developing different techniques for their removal (171,172). Among the materials designed for the removal of this type of contaminants are porous carbonaceous materials derived from CAR, double-net hydrogels, hydrogels in which nanoparticles are incorporated, among others.

Nogueira et al. (2018) obtained porous activated carbon materials from three different types of carrageenan (κ, ι, and λ) for the absorption of antibiotics, specifically ciprofloxacin, and reported that the use of this natural polymer significantly improves absorption properties, improving not only the speed (5 min), but also the yield given the high microporocity (about 1 $cm^3\ g^{-1}$) and specific surface area (above 2300 $m^2\ g^{-1}$) (173). κ-CAR/alginate hydrogels have also been used for the removal of ciprofloxacin, finding that this type of material has improved mechanical properties, anti-swelling and absorption capacity, in addition to the fact that the increase in the CAR ratio results in a higher viscosity and intermolecular interactions of the hydrogel, while alginate is attributed with the improvement of mechanical properties such as compressive strength and elasticity (174). Hydrogels with metal nanoparticles such as ZnO were used for the removal of norfloxacin, reaching 99.4% explaining the plausible absorption mechanism from electrostatic, π-π, and H bond interactions, with an elimination efficiency of 85% after seven cycles (175).

Finally, heavy metal pollution is a problem due to the fact that various industries, including mining, textiles, and agriculture, discharge considerable concentrations of metals, depositing and accumulating in food, soil, and living beings, and these are toxic even in low concentrations (170). One example for this purpose corresponds to tosyl-carrageenan/alginate beads have been designed for the removal of Pb^{+2} ions in aqueous solutions, where the anionic absorption sites provided by both carrageenan and algitane report an absorption capacity greater than 74 $mg\ g^{-1}$ (170).

In conclusion, although the use of carrageenan-based biomaterials for environmental remediation is relatively recent, the presence of functional groups that favor different types of interactions with pollutants of different species makes it a polymer of interest to continue exploring its use in this area.

CONCLUSIONS

In this paper, a detailed review of carrageenans was elaborated by exploring their structural classification, physicochemical properties, and their versatility and potential as biomaterials in a wide variety of applications. A number of materials derived from carrageenans were identified, such as gels, nanoparticles, films, and coatings, among others, that show unique and promising properties for various fields, including their use in the biomedical, agricultural, food, and environmental remediation fields.

Among the findings is that, in the biomedical field, the biocompatibility and ability to form gels make carrageenans ideal for applications in regenerative medicine and scaffolds in tissue engineering, where they can promote wound healing and facilitate the controlled release of drugs for specific treatments. The latter was also evidenced in the agricultural field, being used for the controlled release of fertilizers and pesticides, as well as water retention. Their ability to retain water and nutrients in the soil makes them valuable tools for improving crop quality and agricultural sustainability, which is why carrageenans have been employed as biostimulants and soil improvers, promoting plant growth and increasing production efficiency.

In the food industry, carrageenans play a central role thanks to their ability to improve the texture, stability and sensory quality of food, making them indispensable for the formulation of modern food products as thickeners, stabilizers and gelling agents in a wide variety of products, from dairy to even the production of meat products or their like. In terms of environmental remediation, the ability to form complexes with heavy metals and other pollutants makes them effective tools to mitigate negative environmental impacts and promote the health of aquatic ecosystems, developing materials that reduce the impact of different compounds on the environment supporting the controlled release of drug products, pesticides and fertilizers, such as the removal of pollutants of different types and water purification.

Finally, it can be concluded that these applications demonstrated the potential of carrageenans to inspire future innovations in the design of materials and technologies. However, much remains to be explored in order to fully exploit its potential in various sectors. Further research and development is required, to fully understand the capabilities of carrageenans and their derivatives, as well as to address current and future challenges in areas such as health, agriculture, and the environment. Recognizing carrageenans as a valuable source of biomaterials with a wide range of potential applications, versatility and unique properties, they are positioned as promising candidates to address important challenges in current and future society.

OUTLOCK

As research projections of this material, it is recognized that, although carrageenans are versatile natural polysaccharides, they face challenges in obtaining various biomaterials from them, which lie in the need to optimize the properties and functionalities of carrageenans to adapt them to specific applications. In addition, the variability in the quality and composition of carrageenans extracted from different algae species and processing methods adds complexity in the production of standardized biomaterials. Overcoming these challenges requires exploring multidisciplinary approaches that integrate biotechnology, polymer chemistry, and materials engineering to develop modified carrageenans with enhanced properties and even greater versatility in various industrial and biomedical applications.

CREDIT AUTHORSHIP CONTRIBUTION STATEMENT

Martina Zuñiga: Writing – review and editing, Writing – original draft, Research, review and editing. Diana Montoya: Writing – original draft, Research, review and editing. Bernabe Rivas Q.: Writing – review and editing, visualization, supervision, conceptualization.

CONFLICT OF INTEREST STATEMENT

The authors declare that they have no financial interests or personal relationships that could have influenced the work reported in this article.

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