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## Sustainable applications of carrageenan as a next-generation biopolymer in intelligent and active food packaging

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Carrageenan is extracted from seaweeds and is considered a natural polysaccharide that has emerged as one of the most promising biopolymers for sustainable food packaging applications. Due to the increasing demand for eco-friendly packaging over conventional plastics, carrageenan-based films serve as a versatile solution that aligns with environmental and consumer acceptability of their use. Carrageenan is recognized as a sustainable and eco-friendly bio-material for food packaging due to its excellent film-forming abilities, biocompatibility, low cost, and functional properties. It offers a sustainable solution for preserving and extending the shelf life of food products by providing a protective film that can incorporate active constituents, such as antimicrobial and antioxidant agents, to enhance food quality and safety. As a biodegradable and natural polymer, carrageenan-based packaging helps to address environmental concerns while effectively maintaining the freshness of various food items throughout storage and distribution. Active constituents may include antimicrobial agents, antioxidants, oxygen scavengers, ethylene absorbers, or other bioactive compounds that can perform one or more functions. These compounds could be immobilized onto the package, allowing for controlled release into the food matrix. This review categorizes various types of carrageenan-based packaging, including edible coatings, active packaging, and innovative packaging integrated with sensors, which address food packaging needs and evaluate the factors determining the suitability of these biomaterials. The effects of packaging on physical attributes, chemical stability, microbial growth, organoleptic quality, and shelf life of the food are discussed. The flexibility of carrageenan, integrated with innovative materials and efficient processing technologies for packaging, is also included. Furthermore, the potential toxicity, consumer acceptability, challenges, and opportunities of carrageenan packaging are explored, along with future perspectives and government encouragement for carrageenan cultivation, which provide future directions for research in this arena.

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### Sustainability spotlight

This review addresses the sustainability of carrageenan, a marine-derived biopolymer, for next-generation food packaging applications. Emphasizing its biodegradability, biocompatibility, and film-forming abilities, carrageenan supports SDG 12 (Responsible Consumption and Production) by providing an eco-friendly alternative to conventional plastics by reducing plastic waste. Integration with active compounds enhances antioxidant, antimicrobial, and mechanical properties, enabling active and intelligent packaging solutions that extend shelf life and monitor food quality which contributing to SDG 2 (Zero Hunger) and SDG 14 (Life Below Water) by minimizing food loss and protecting marine ecosystems. The advancements and challenges guide future sustainable packaging innovations of carrageenan. These advancements support circular economy principles, environmental stewardship, and sustainable food systems, advancing a greener future in food packaging and related sectors beyond food, including pharmaceuticals and cosmetics.

## 1. Introduction

Carrageenan is a polysaccharide abundant in nature, derived from red seaweed, commonly known as *Rhodophyta*. The term

carrageenan is derived from seaweed (name originating from the Sanskrit [karkhanda] and Irish Gaelic).<sup>1</sup> Carrageenan is a high-molecular-weight marine polysaccharide obtained mainly from *Eucheima denticulatum* and *Chondrus crispus*. Carrageenan is generally classified into three types, namely iota ( $\iota$ ), kappa ( $\kappa$ ), and lambda ( $\lambda$ ). Carrageenan is commonly used as a thickener and stabilizer in the food processing sector. The market demand for carrageenan is expected to grow due to its increasing need in the food and beverage industries and

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consumer preferences for natural and plant-based products. Carrageenan production contributes to the local economy and the livelihoods of the region's residents. It is estimated that the global carrageenan market will experience growth in the upcoming years due to its increased applications in the cosmetics, pharmaceuticals, and food industries. Additionally, several types of research are ongoing on the application of carrageenan in industries other than food, which may increase the demand for carrageenan production.<sup>2</sup>

Carrageenan is a versatile ingredient in the food manufacturing industry. It's prized for its ability to hold water, thicken, gel, and stabilize various products, and it is often found in meat, dairy, and flour-based foods.<sup>3</sup> In dairy, it imparts a creamy texture and can even serve as a fat substitute in low-fat options, such as cottage cheese and ice cream. It acts as a stabilizer, preventing separation and maintaining a consistent texture in milk-based beverages. When it comes to meat products, particularly processed items like sausages and deli meats, carrageenan enhances tenderness and juiciness. It is also employed in sauces to enhance their viscosity, contributing to a more desirable texture.<sup>4</sup> Food packaging is another field where carrageenan is commonly used. It is selected for developing films due to its low cost, film-forming properties and biodegradability.<sup>5</sup> It has been identified that kappa-carrageenan can form transparent films with good structural and mechanical properties when compared to iota and lambda carrageenan.<sup>6</sup>

However, these types of films have limited water vapor barrier properties and low water resistance capabilities, as they are inherently hydrophilic in nature.<sup>7</sup> These minor drawbacks are overcome by using carrageenan with other hydrocolloids to form films. For example, a film made from the combination of starch and  $\kappa$ -carrageenan exhibits increased water vapor permeability and mechanical strength.<sup>8</sup> Other methods, such as cross-linking, reinforcing with hydrophobic fillers (insoluble proteins, waxes, resins, etc.), the addition of nanoparticles, and blending natural and synthetic polymers, are used to enhance the properties of carrageenan-based films and overcome their shortcomings. The films or packaging materials used are designed to improve the shelf life of the product by preventing surface browning, dehydration, and oxidative rancidity.<sup>9</sup> Carrageenan is also widely employed in pharmaceutical formulations due to its diverse functional properties. It is used for stabilizing emulsions and suspensions, thereby ensuring the uniform distribution of active ingredients.<sup>10</sup>

Furthermore, carrageenan plays a significant role in controlled-release drug delivery systems. Its ability to form gels allows for a slowed release of active pharmaceutical ingredients, leading to prolonged therapeutic effects and improved patient adherence.<sup>11</sup> The bio adhesive properties of carrageenan enhance the retention of drug formulations at the target site, making it particularly useful for topical and mucosal drug delivery.<sup>12</sup> Carrageenan's excellent compatibility with various other excipients facilitates the development of complex drug delivery systems, accommodating multiple active ingredients without compromising the product's stability or effectiveness.<sup>13</sup>

Carrageenan has advantages over other petroleum-based films, including biodegradability, non-toxicity, and water

solubility. The aqueous carrageenan solution is used either as a film or directly coated on the food surface. The films are cast in plates and then molded into bags. The physical and chemical modification of carrageenan is performed to enhance the effectiveness of the film and overcome its inherent shortcomings.<sup>14</sup> Some recent studies suggest that carrageenan films can be developed to change color, indicating spoilage in products, and used to monitor the quality of food products. Additives like carbon dots can enhance the film's antioxidant and antibacterial properties, thereby helping to extend the shelf life of foods.<sup>15,16</sup> This review mainly focuses on discussing their types, extraction techniques of carrageenan, selection of carrageenan for films, physical and mechanical properties, such as mechanical, rheological, and thermal properties, potential challenges in packaging, and recent innovations and challenges in the industry to provide a practical recommendation and a glimpse into the future of sustainable packaging solutions.

## 2. Carrageenan source, types and extraction

Carrageenan is a sulfated polysaccharide composed of alternating units of  $\beta$ -D-galactopyranose and  $\alpha$ -D-galactopyranose or 3,6-anhydro- $\alpha$ -D-galactopyranose, linked by  $\alpha$ -1,3 and  $\beta$ -1,4-glycosidic bonds. It is extracted from algal species that are rich and abundant in seaweeds, such as *Euchema cottoni* (mainly kappa carrageenan) and *Euchema Spinosum* (mainly iota carrageenan). Generally, carrageenan is classified into three broad types: kappa, iota, and lambda grades. Kappa carrageenan requires heat to dissolve in water.<sup>10</sup> It can form complex and brittle gels with syneresis, and the water gel strength of this type of carrageenan increases with the blending of salts containing  $K^+$  potassium ions in the formulation. The kappa type has more synergistic effects when blended with locust bean gum, compared to other hydrocolloids. Iota carrageenan dissolves partially in cold water, and complete dissolution needs heating. It forms soft, elastic, freeze-thaw stable gels with no syneresis.<sup>17</sup> Water gel exhibits increased strength when combined with  $Ca^{2+}$  ions. Lambda carrageenan is completely cold-soluble in water. It forms a viscous solution and is non-gelling, and primarily used for stabilizing thickening systems and suspending particles. It is stable in neutral or alkaline media and insoluble in alcohol and other organic solvents.<sup>9</sup>

The carrageenan treatment process involves a matrix of variables, including alkali, temperature, raw weed, and time. These four variables will influence conversion levels, affecting gelling behavior, influencing viscosity, and impacting process yields. The process of semi-refined carrageenan (SRC) begins with the withdrawal of seaweed. It is transferred to a rotating drum to remove sand and dirt. It is then cooked with potassium chloride (KCl) and potassium hydroxide in water.<sup>5</sup> Then it has subjected for extensive washing (4 times) to ensure maximum quality of carrageenan has separated from seaweed using a hot extraction method. Also, it removed the unwanted plant fiber during this process. The dissolved carrageenan, which makes up about 2–7% of the wet seaweed plant, is separated from the



extraneous solids by filtration. The carrageenan is precipitated into a fibrous or gel-like material, dried, ground, and sampled for quality control testing.<sup>18</sup> The entire process is closely monitored to obtain optimum gelling viscosity characteristics. For the bleaching step, agents such as  $\text{Ca}(\text{OCl})_2$  and  $\text{NaOCl}$  are used as bleaching agents, followed by rinsing with water.<sup>9</sup>

It is used for steam drying and cabinet air drying. The particle size is reduced in the hammer mill, and the metal detector separates any iron fillings. Furthermore, it is ground into a fine powder, blended with other additives, and packed in drums or bags. Storage should be under ambient conditions, with the temperature and relative humidity not exceeding 35 °C and 60%, respectively.<sup>19</sup> Refined carrageenan (RC) varies depending on the gel-press process. In this process, seaweed is extracted using hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) at 95 °C, followed by two stages of filtration: coarse filtration and fine (polishing) filtration using a filtration aid. The filtrate is then subjected to gelation by the addition of salts at 32 °C, followed by potassium chloride (KCl) precipitation. The resulting gel is pressed to remove excess water, after which the material is passed through metal detection. Finally, the refined carrageenan is compressed into pellet or powder form for storage.<sup>20</sup> Both iota and kappa carrageenan solutions typically gel at room temperature, and require heating to revert to a liquid state for further processing.<sup>21</sup> Formulating, casting, and moulding a product at elevated temperatures, followed by cooling, results in a carrageenan-based binder. Therefore, the set and melting temperatures of carrageenan gels depend on the cation concentration.<sup>9</sup>

### 3. Criteria for selecting carrageenan for packaging films

The selection criteria for carrageenan for packaging films include several key factors such as biodegradability,<sup>22</sup> non-toxicity,<sup>10</sup> and water solubility.<sup>5</sup> Its mechanical and thermal properties are significant, as is its effectiveness as a barrier against external elements.<sup>23,24</sup> Additionally, antibacterial characteristics are crucial for improving food preservation and helping monitor freshness.<sup>25</sup> While carrageenan has some inherent limitations, these can often be overcome through various modifications.<sup>26</sup> Chicken patties demonstrate that essential oils, such as oregano at 0.02% and thyme at 0.03%, exhibit antimicrobial properties best suited for edible coating applications, reducing the level of surface microbes, including *Staphylococcus aureus*, *Listeria species*, and *Salmonella pullorum*. The release profile of curcumin differed among the films, with carrageenan showing the fastest release, alginate an intermediate release, and chitosan the slowest.<sup>27</sup> The sustained release of curcumin from the chitosan/montmorillonite nanocomposite is a result of powerful electrostatic interactions. The acid-insoluble ash of  $\kappa$ -carrageenan, semi-refined carrageenan and refined carrageenan can affect the edible film pH and tends to degrade from a neutral pH of 6.6 to an acidic pH of 4–5.5, leading to film damage. The regulations based on EU and JECFA standards of 2% acid insoluble matter apply to refined carrageenan, while the refined carrageenan range is 8–15% for good film performance.<sup>28</sup>

Generally, kappa carrageenan can be used for seaweed straw applications as a film barrier, with an optimum water gel strength of at least  $700 \text{ g cm}^{-2}$  and a potassium gel strength of at least  $1000 \text{ g cm}^{-2}$ , which can be tested at a concentration of 1.5% w/w in 0.2% KCl at 20 °C. The final edible kappa films, with a pH range of 7–8, were designed to prevent colour change and nutritional alteration, thereby reducing the risk of textural loss and enzymatic browning in apples transported from 4 °C to 37 °C storage conditions.<sup>29</sup> Buffer acids and a combination of acidulants can influence the pH and gel strength of the films.<sup>30</sup> Moreover, the buffer salts will tend to influence the gel strength of the film. The addition of citric acid for lemon and orange-based products is based on the solubility of water. Buffer salts, such as sodium, potassium, and calcium, are commonly used in conjunction with the bases of citrates and phosphate salts. Buffer salt should be mixed first before adding the acid. The buffer salt helps stabilize the pH level. Further heating of the solution after the addition of the acid would result in a decrease in the gel strength of the packaging material composition. This is because acids have a depolymerizing effect on carrageenan, and this effect is exacerbated by high temperatures.<sup>31</sup> The significant advantages of choosing the right buffer salts and acidulants can provide a highly elastic, pliable, resilient, and thermoreversible gel with quick-setting properties. Furthermore, it gives soft, transparent, and elastic gels.

Carrageenan exhibits thixotropic behavior, particularly at low concentrations of iota-carrageenan. This property enables iota gels to become fluidized when subjected to agitation or shear forces, making them more liquid-like; upon resting, the gels recover to an elastic state. Specifically, when shear rates reach 1000 rpm, the gel structure of dilute iota carrageenan breaks down, but it reforms once the shear is removed. This thixotropic characteristic is advantageous for suspending insoluble particles in flowable systems, such as antibiotic suspensions.<sup>32</sup> In contrast, kappa carrageenan water gels lack thixotropic properties; once disrupted, they require reheating and cooling to regain their gel structure.<sup>33</sup> Furthermore, freezing temperatures between  $-6 \text{ °C}$  and  $-3 \text{ °C}$  cause approximately 50% of the water content within packaging films to freeze, which is critical for storage stability. Understanding these rheological and thermal behaviors is essential for optimizing carrageenan applications in food packaging.<sup>34</sup>

### 4. Classification of packaging types derived from carrageenan

Carrageenan is used to create biodegradable, non-toxic, and water-soluble packaging for food, in the form of both films and coatings. These can be cast into films from aqueous solutions or applied directly onto food surfaces as “wraps”. To improve their performance in areas like mechanical strength, heat resistance, barrier protection, and antibacterial action, carrageenan-based packaging can be modified. These enhancements broaden their use in extending food shelf life and monitoring freshness.<sup>26</sup>

#### 4.1 Edible coatings and films

Carrageenan is used in edible coatings because it is biodegradable, renewable, and safe for consumption. It significantly



improves food quality by enhancing appearance and texture, while also reducing moisture loss, controlling microbial growth, and extending shelf life. This makes it ideal for a wide range of food applications.<sup>35</sup> Films made using a 3% iota carrageenan and 30% glycerol combination have been studied for their wetting properties. The results show that increasing the concentration of glycerol from 30% to 90% on a solid basis affects the gelation effect and water resistance capacity of the film. This is due to the “phates” group present in carrageenan, which can form macromolecular aggregates due to strong electrostatic interactions.<sup>36</sup> Generally, carrageenan films have been used to pack dry fruits like figs and roasted almonds. Walnut films consist of trehalose, orange essential oil, Tween 20 or Tween 80 series and 5% (w/v) of glycerol combination. This is due to the use of trehalose to prevent aroma and color loss in the product. At the same time, the essential oil can exhibit antibacterial or antimicrobial properties against Gram-positive bacteria, such as *Staphylococcus aureus*.<sup>37</sup> Incorporating raw spices into a semi-refined kappa carrageenan edible film effectively controls surface microbial growth on chicken breasts during storage. Specifically, this coating significantly reduces *L. monocytogenes* to undetectable levels after four days and achieves a six-log reduction in *Streptococcus aureus* after six days, demonstrating its strong antimicrobial capabilities.<sup>38</sup> Fresh-cut fruits, such as papaya, jackfruit, guava, watermelon, and muskmelon, can be coated with a combination of sodium CMC and refined carrageenan, which have been tested for color indication and edibility. It has different intensities of colour, ranging from dark blue to green. The CO<sub>2</sub>-releasing tablets were placed inside the package, increasing the respiration rate by approximately 10% and maintaining controlled atmospheric conditions. A seven-day shelf-life study of fresh-cut fruits revealed an initial color shift from blue to green, confirming continued edibility. Extended storage under refrigeration (10–15 days) resulted in a further color transition to varying intensities of yellow-green, indicating that the fruits remained edible beyond the initial seven-day period.<sup>39</sup>

#### 4.2 Active packaging films

Active packaging films are often incorporated with bioactive compounds such as essential oils to impart antibacterial properties.<sup>138</sup> Besides oregano oil (*Origanum vulgare*), other commonly used essential oils include thyme (*Thymus vulgaris*), clove (*Eugenia caryophyllata*), cinnamon (*Cinnamomum verum*), and lemongrass (*Cymbopogon citratus*) oils, all of which exhibit significant antimicrobial activity against foodborne pathogens.<sup>40,41</sup> Carrageenan can be incorporated into films as an active packaging material to form biodegradable films, thereby enhancing food safety and shelf life while minimizing the use of chemical preservatives.<sup>42</sup> Active packaging for shrimp utilized pure kappa carrageenan coated with zinc oxide and anthocyanin extracted from kohlrabi peels, which can enhance the antioxidant properties and improve UV protection by 85.2% compared with the control film. The films exhibit 99% antioxidant properties, as confirmed by the ABTS assay detection method. The results were compared with those of the DPPH method, which has 58%

antioxidant property and is more resistant to *Listeria monocytogenes*. After 12 hours of incubation, 8.1 log CFU mL<sup>-1</sup> were found in the microbial analysis.<sup>22</sup> Studies show that combining carrageenan and agar with tea essential oil and zinc oxide nanoparticles using a pickering emulsion process improves mechanical properties by approximately 30%. In addition, the presence of the oil emulsion increases hydrophobicity by about 10% and provides good antioxidant activity due to catechins and epicatechins in the tea extract.<sup>43</sup>

Carrageenan-polyvinyl alcohol (PVA) films enriched with calcium (Ca<sup>2+</sup>) and sodium (Na<sup>+</sup>) ions and incorporated with gallic acid exhibit higher water vapour permeability and reduced mechanical strength. The presence of gallic acid enhances antibacterial activity, showing greater resistance against *Escherichia coli* and *Staphylococcus aureus*, attributed to increased electrostatic interactions and a rise in surface hydrophobicity to approximately 15%. This enhanced hydrophobicity improves binding affinity to fruits and vegetables such as apples and plums, making the films suitable for active packaging applications.<sup>44</sup> Carrageenan films were used for banana packaging to reduce ethylene levels and respiration rates, thereby controlling the ripening process. The addition of a modified titanium nanotube can extend the shelf life by up to 12 days, retaining good organoleptic properties such as color and aroma, while also resulting in less weight loss and shrinkage.<sup>26</sup>

In fish packaging applications, Mandarin fish have a more volatile base nitrogen content, which can lead to an off-smell due to the presence of dimethyl and trimethyl amines in the fish. Carrageenan coating with anthocyanin and cinnamaldehyde may have the potential to improve the shelf life of fish by 13.3%. According to the ABTS method results, 92.77% antioxidant retention was detected, whereas the DPPH method showed 48% antioxidant property in the active films.<sup>45</sup> Carrageenan-sodium alginate films incorporated with *Allium sativum* for meat packaging applications exhibit enhanced antioxidant and antimicrobial properties against *Listeria monocytogenes*, reducing microbial counts to below 2.5 log CFU g<sup>-1</sup> after 48 h of incubation. Antioxidant activity, measured using the ABTS method, reached 71.4% compared to control films.<sup>45</sup>

#### 4.3 Biodegradable films

Carrageenan's biocompatibility, biodegradability, and excellent film-forming abilities make it a prime choice for creating environmentally friendly packaging films. These films offer a sustainable alternative for food packaging, boosting both safety and quality while aligning with consumer preferences for minimally processed, preservative-free products.<sup>24</sup> Carrageenan has been utilized as a preservative to prevent diffusion into food products, such as confectionery gummies and frozen cakes, which can inhibit the growth of microbes on the surface. Recent trends have shown that biopolymer polysaccharides can exhibit synergistic effects in biodegradability. For instance, a combination of arrowroot starch (2%) and iota carrageenan (0.5%) has demonstrated good crystalline properties and excellent biodegradation in both soil and seawater.<sup>46</sup> This leads to



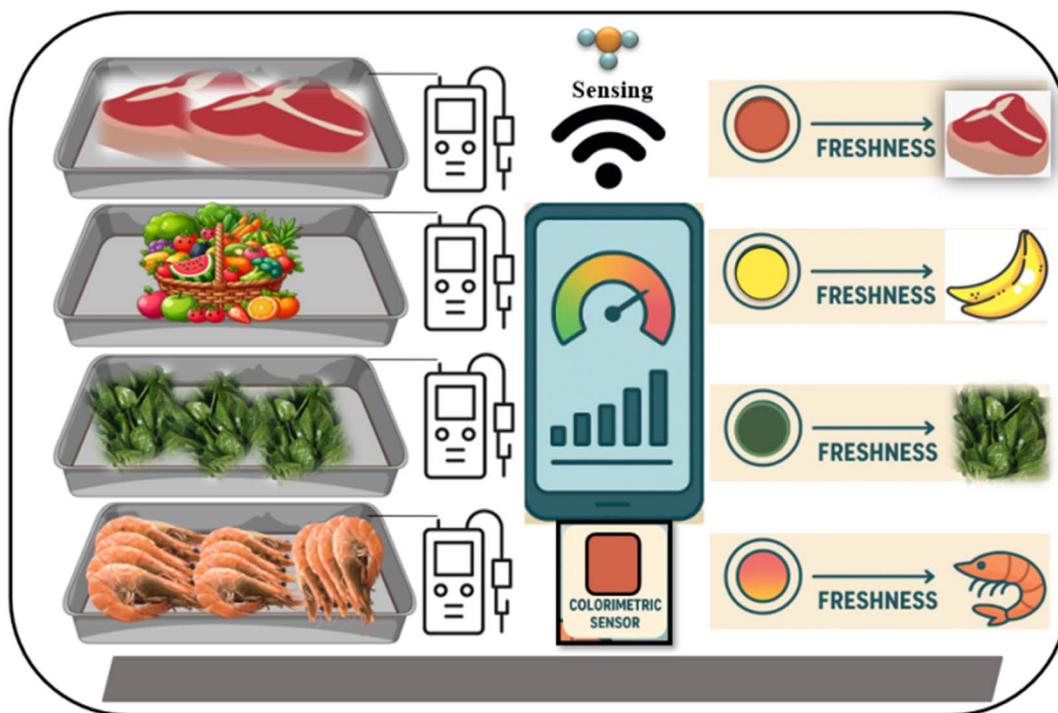


Fig. 1 Smart packaging integrated with sensors for different food applications.

a smooth surface with high transparency. The results show that arrowroot films exhibited biodegradation in 7 days in soil, whereas control films showed biodegradation after 30 days in soil. Moreover, the shelf life of cherries and tomatoes can be increased by up to 10 days using a composite biopolymer.<sup>47</sup>

One of the challenges in biodegradable films is the lack of mechanical strength. This problem can be addressed by introducing pectin (33%) and kappa carrageenan (67%) with nanoclay (1.5%) plasticized with 2% water, which exhibits positive behavior in terms of good barrier properties and enhances dispersion.<sup>48</sup> Under conditions like high temperature and humidity, films start to degrade or lose their texture. This can be prevented by the addition of cassava starch and  $\kappa$ -carrageenan-starch interaction, which, when produced by the solvent casting method, yields good transparent and opaque films that can be used to pack freshly baked goods, such as bread and croissants, making them suitable for this biodegradable application.<sup>49</sup>

#### 4.4 Smart packaging integrated with sensors

Smart packaging utilizes sensors to monitor the condition of packaged food in real-time, providing valuable information on parameters such as temperature, humidity, gas composition, pH, and freshness status. These sensors can be embedded in or attached to packaging materials and function as indicators, data carriers, or communication devices, such as RFID tags and near-field communication (NFC) systems.<sup>50</sup> Smart packaging sensors detect physiological and chemical changes caused by microbial growth or spoilage and can alert producers and consumers through colorimetric changes or electronic signals.<sup>51</sup> This technology enhances food safety, reduces waste, and

improves shelf-life management by facilitating monitoring throughout the supply chain.<sup>52</sup> This system produces excellent outputs that aim to extend shelf life and reduce food waste.<sup>53</sup> Integration with electrochemical sensors was used to detect the freshness of fruits and vegetables like bananas, carrots and shallots, which can extend shelf life during transportation. Gas sensors are highly sensitive and can be used to detect gases like ethylene during the banana and mango ripening process. They are also used to identify toxicity, for leak detection, and assess the ripeness of the product through color changes.<sup>54</sup> Advances in gas sensor technology, such as paper-based electric gas sensors (PEGS) have enabled their application in high-value products like spinach. These sensors can be linked to smartphones *via* near-field communication (NFC) and function as an in-line monitoring tool to assess the freshness of spinach.<sup>55</sup>

Furthermore, a combination of gelatin and kappa carrageenan at a (1 : 3) ratio with curcumin concentration ranging from 1–4% was able to produce halochromic carrageenan films, which are used to package chicken meat. This synergistic effect will enable the binding ability to encapsulate elements and carriers, which can be tightly bound to films, thereby paving the way for potential smart packaging applications.<sup>56</sup> For seafood-based products, blueberry anthocyanins were used with a 6% diatomite infusion in konjac and carrageenan films, which are used to monitor shrimp spoilage. This synergistic combination improves the tensile strength, surface hydrophobicity and elongation at break by 5 to 7% compared to standard carrageenan films. The colorimetric sensor indicated the colour change from bright pink to bluish violet which can be directly linked to the total volatile content, *i.e.*, amine content such as



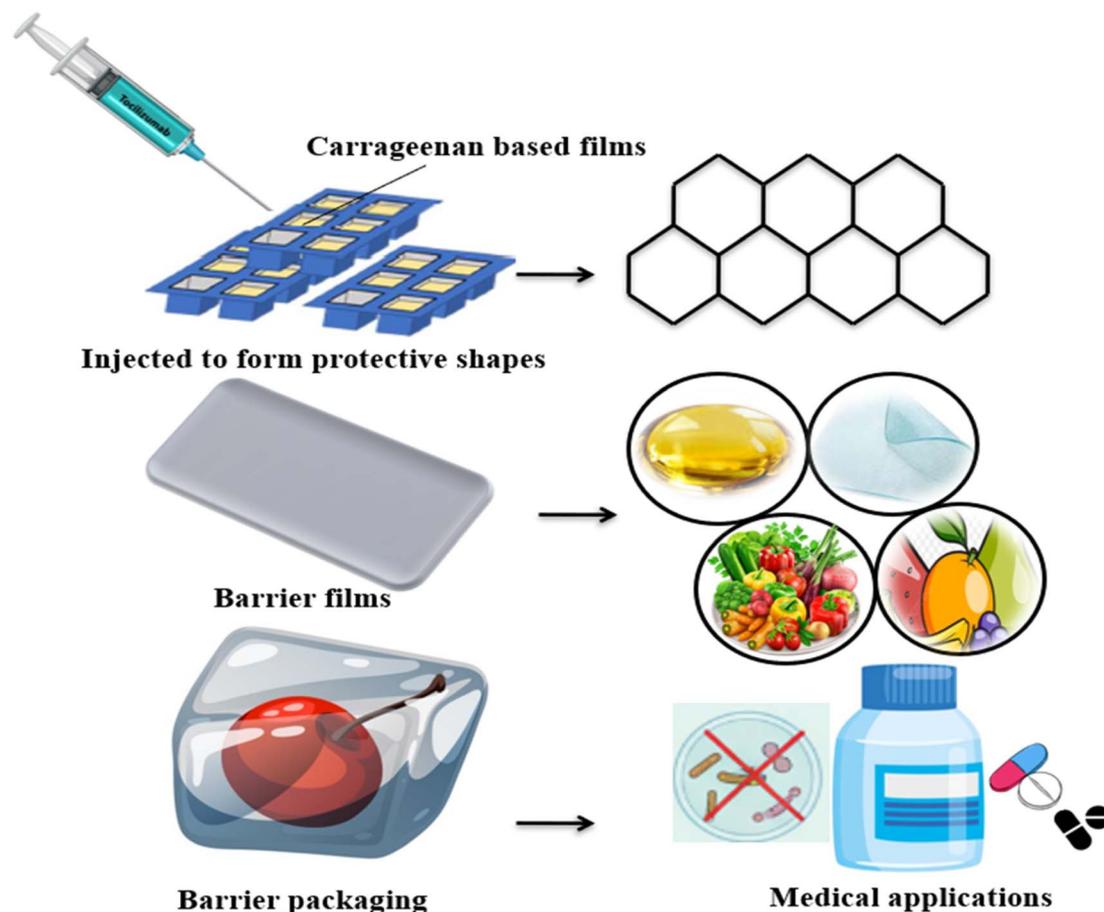


Fig. 2 Mechanism of film formation on injectable-type packaging.

dimethylamine, trimethyl amine and ammonia ( $\text{NH}_3$ ) during the storage of shrimp and the pH changes in the shrimp. Increasing the amine concentration enables sensors to detect food deterioration, which can accelerate the color change, off-odor, and release of volatile gases from the shrimp, and a color change was observed.<sup>57</sup> Blueberry anthocyanin solutions exhibit colour changes based on pH: bright red at pH 2, pink at pH 3–4, and a gradual reduction in intensity as the pH increases, with further reduction at neutral and alkaline pH levels of 7. Colour changes from reddish-brown to yellow indicate spoilage, which is detected by the colorimetric sensors. This could be a potential development opportunity for pH indicator intelligent packaging films using carrageenan for seafood applications. Fig. 1 illustrates the smart packaging integrated with sensors for different food product applications.

#### 4.5 Composite packaging

Carrageenan-based composite films are biopolymer materials that excel at forming films. These films can be further enhanced by incorporating functional compounds and nanofillers, which enhance their mechanical strength, barrier protection, and overall functionality. This makes them ideal for use in active and intelligent food packaging.<sup>58</sup> Silver-carrageenan nano-composite films were developed using argemone albiflora

extract, a biodegradable and eco-friendly material that exhibits enhanced mechanical and antimicrobial properties, making it more suitable for food packaging applications. Similarly, kappa carrageenan added with two antibacterial types, one with positively charged spermidine carbon dot (CD) films and the other with carrageenan polyvinyl alcohol (PVA-CP) nano-composite biopolymer with low migration and high transparency, could prevent microbial contamination and act as effective packaging in pork samples. The combination of carrageenan and gelatin exhibits good mechanical properties, whereas a combination of carrageenan with agar shows a reduction in water solubility and poor elongation at break.<sup>59</sup> Gelatin and kappa carrageenan infused with lime essential oil (LEO), which contains secondary metabolites such as limonene,  $\gamma$ -terpinene, and hexane groups like geranyl acetate, cyclohexene,  $\beta$ -pinene, and terpineol, can reduce the presence of food-borne pathogens, including *Staphylococcus aureus* and *Candida albicans*. The composite film exhibits reduced mechanical properties with increased film thickness (depth of less than 3 mm), making it most suitable for functional active packaging. A composite coating, made from a blend of carboxymethyl cellulose (0.3%) and carrageenan (0.2%) polysaccharide gums, with added citric and oxalic acids, was applied to ready-to-eat pears. This coating effectively minimized



oxidative damage, physiological decay, and browning.<sup>60</sup> The influence of newly developed composite edible coatings on strawberry quality was investigated during a 15-day storage test. The study revealed that varying amounts of hyaluronic acid in the coating directly impacted the preservation of key strawberry characteristics, including reduced weight loss, stabilized pH, total solids, and titratable acidity.<sup>61</sup>

#### 4.6 Injectable type packaging

Injectable packaging involves materials that can be readily injected into molds to form protective shapes, commonly used in the food and pharmaceutical sectors to ensure product freshness and safety. Carrageenan-based films are a safe option for this type of packaging in both food and medical applications. Their injectability can be further enhanced, and their mechanical strength improved, by incorporating materials such as green graphene oxide (GO), leading to more durable films.<sup>62</sup> Kappa carrageenan has been utilized in biomedical studies for stimuli-responsive hydrogels, which have demonstrated strong antibacterial properties against *Staphylococcus aureus* and *E. coli*, as well as in packaging films.<sup>63</sup> Fortifying the actives in the carrageenan film could be a potential way to reduce micro-nutrient deficiency. Carrageenan could be used to laminate the films over the surface of nutraceutical products, such as soft gel and oral strip films, thereby integrating the new product with high functional benefits.<sup>64</sup> Modified carrageenan can be achieved through the interaction of kappa carrageenan with dialdehyde, which has potential for active food packaging applications. Gelatin films (~3%) incorporated with modified  $\kappa$ -carrageenan *via* an injection process and incubated at 37 °C for 24 h exhibited improved stability and barrier properties compared to control films. It could be used as a potential packaging material for jamun fruit, berries, and tomatoes to preserve their antioxidant properties and anti-pigmentation effects.<sup>65</sup> Fig. 2 illustrates the mechanism of film formation on injectable type packaging on carrageenan-based films.

#### 4.7 Pouch and moulded packaging

Carrageenan is used to create versatile food packaging in the form of films that can be cast into pouches or molded into shapes. These films are produced from aqueous solutions and can be further modified to enhance their mechanical strength, thermal resistance, barrier properties, and antibacterial characteristics, making them highly effective for various food packaging applications.<sup>26</sup> Unrefined carrageenan, combined with protein sources using the thermo-moulding method, can be used to make tiny pouches and edible films that could be successful food packaging. In case the film contains 7% glycerol as a plasticizer, it can behave as a hydrophilic film, which can degrade the film under extreme environmental conditions. This can reduce the water vapour permeability of carrageenan due to the molecules being tightly held between the gel matrices, which hold the water together.<sup>66</sup> The processing method employed here was injection moulding and film blowing, which can produce bubble-forming and flexibility characteristics. Furthermore, the antioxidant properties of moulded films can

be enhanced through the addition of extracts and vitamin C during the packaging process. Table 1 presents the collaborative data on carrageenan-based films in various food applications.

#### 4.8 Intelligent packaging films

Carrageenan films combined with anthocyanins from natural sources such as blueberry and sweet potato peel extracts have been engineered to function as pH-sensitive colorimetric sensors, capable of indicating food freshness through visible color changes.<sup>67,68</sup> A film infused with blueberry anthocyanins and diatomite incorporated into konjac and carrageenan matrices was developed. This film exhibited improved tensile strength, surface hydrophobicity, and elongation at break by 5–7% compared to standard carrageenan films.<sup>67</sup> Intelligent packaging was made from carrageenan and jaboticaba peel extract though incorporation of the peel extract at two different concentrations like 0% (control), 50% and 100% w/w with plasticizer glycerol (concentration 60% w/w) in the film which decreased mechanical properties (from control to 100% concentration) like tensile strength ( $7.72 \pm 0.49$  to  $3.24 \pm 0.32$  MPa), and elongation at break ( $10.75 \pm 2.01\%$  to  $3.28 \pm 0.62\%$ ), but it showed improved thickness ( $0.039 \pm 0.0024$  to  $0.055 \pm 0.0018$  mm), Young's modulus ( $74.75$  to  $101.57$  MPa), and opacity ( $4.40 \pm 0.54$  to  $14.86 \pm 2.18$  abs at  $660 \text{ mm}^{-1}$ ) and reduced swelling index ( $95.08 \pm 1.12$  to  $92.40 \pm 1.00$ ) and water vapour permeability ( $1.89 \times 10^{-11}$  to  $1.34 \times 10^{-11} \text{ g m}^{-1} \text{ Pa}^{-1} \text{ s}^{-1}$ ).<sup>69</sup> As the anthocyanins present in the jaboticaba peel extract change colour (from pH 1 to 14) under varying buffer conditions, it has the potential to act as intelligent packaging in the film.<sup>70</sup>

## 5. Properties of carrageenan-based packaging materials

### 5.1 Mechanical properties

The mechanical properties or mechanical strength of a film are generally determined by its elongation at break and tensile strength. The inclusion of other components into the carrageenan matrix modifies the mechanical properties of the carrageenan-based films.<sup>5</sup> These modifications improve the film's resistance and durability during handling and transportation.<sup>26</sup> The film made by incorporating kappa carrageenan with grapefruit essential oil shows enhanced tensile strength. The tensile strength increases with the increasing concentration of grapefruit essential oil, from  $65.20 \pm 4.71$  MPa to  $98.21 \pm 6.35$  MPa.<sup>76</sup> In a film composed of semi-rigid components made with potassium hydroxide, the tensile strength ranges from 38.1 to 47.5 MPa. The material exhibits enhanced mechanical properties, enabling it to withstand applied stress without breaking in a film made with semi-rigid carrageenan indicating that the films are strong and can withstand stress.<sup>77</sup> Plasticizers such as sorbitol, glycerol and PEG-400 enhance the mechanical properties, including tensile strength and elongation at break.<sup>78</sup> A study of silica and silver nanoparticles incorporated into  $\kappa$ -carrageenan films identified that the composite has enhanced mechanical properties, with the tensile strength of the film ranging between 23.8 and 41.5 MPa.<sup>79</sup>



Table 1 Carrageenan films in different food applications and their effects on films

Food product	Film type	Dose level and size of the film	Effect on the films	Detection method	Effect on the film system	Mode of action	Mechanism	Reference
Chicken patties	Edible coating ( $\kappa$ -carrageenan + EO)	0.02% Oregano and 0.03% thyme	99.99% Reduction surface microbes	Microbial colony count	6-log reduction of <i>Listeria monocytogenes</i>	Antimicrobial action	Essential oils disrupt the cell membrane	27
Dry fruits (Figs)	Carrageenan film + orange EO	5% Glycerol and Tween 20 or Tween 80	30% Less aroma loss; antibacterial	GC-MS and microbial assays	Retained 95% aroma and color compared to the control	Antibacterial	Essential oil bioactives kill Gram+ bacteria	37
Shrimp	Active film ( $\kappa$ -carrageenan + ZnO + anthocyanin)	Not specified	Reduced microbial load 58% (DPPH) with 99% antioxidant (ABTS assay)	ABTS, DPPH, and microbial count	8.1 log CFU ml <sup>-1</sup> after 12 h incubation	Antimicrobial and antioxidant	ROS scavenging and cell membrane disruption	22
Chicken breasts	Semi-refined $\kappa$ -carrageenan + spices	Not specified	99.9% Reduction <i>L. monocytogenes</i>	Microbial assay	Undetectable <i>L. monocytogenes</i> after 4 days	Antimicrobial	Spices damage bacterial cell walls	38
Papaya and jackfruit	Coating (sodium CMC + refined carrageenan)	Not specified	10% Longer shelf life and color retention	Colorimetric test	Color change from blue to green sustained for 7 days	Preservation	pH-responsive anthocyanin	39
Banana	Kappa film with TiO <sub>2</sub> nanotubes	Not specified; <0.1 mm thickness	12 days shelf-life extension	Microbial assay	20% Less weight loss, 15% less shrinkage, weight loss, and organoleptic evaluation	Preservation	Reduces ethylene and respiration rate	26
Mandarin fish	Pigmented coatings (anthocyanin & cinnamaldehyde)	Not specified	13.3% Less volatile nitrogen with ABTS 92.77%; DPPH 48%	ABTS, DPPH	30% Increased antioxidant retention	Antioxidant	Radical scavenging	45
Meat packaging	Carrageenan+ alginate + <i>Allium sativum</i>	Not specified	Reduced microbial growth <2.5 log CFU g <sup>-1</sup> with ABTS 71.4% antioxidant	ABTS assay	2.5 log CFU g <sup>-1</sup> less <i>L. monocytogenes</i>	Antimicrobial	Garlic bioactives inhibit bacteria	71
Seaweed dodol	$\kappa$ -Carrageenan edible film	Not specified	Reduced bacteria by 40%	Microbial colony count	40% Lower microbial counts vs. control	Antimicrobial effect	Barrier to moisture and microbes	72
Ambon bananas	Edible coating + peppermint oil	1.5% Carrageenan, 0.5% glycerol, and 0.5% peppermint	5 days delayed ripening	pH and soluble solid measurement	8% Less weight loss at day 10	Preservation	Reduces metabolic activity	73





Table 1 (Contd.)

Food product	Film type	Dose level and size of the film	Effect on the films	Detection method	Effect on the film system	Mode of action	Mechanism	Reference
Apple slices	Edible film + 1.25% carrageenan + 10% palm oil	Film thickness 30 $\mu\text{m}$	Retained 91% vitamin C; 8% weight loss	Vitamin C content and weight loss	Less discoloration and better moisture retention	Preservation	Moisture barrier & antioxidant	74
Fresh-cut mangoes	Anti-browning dip (incl. carrageenan)	$5 \text{ g kg}^{-1}$	30–50% Reduction in browning with pH 5.8–5.9	Visual browning assessment	Significantly less enzymatic browning	Preservation	Inhibits polyphenol oxidase	75
Pork samples	$\kappa$ -Carrageenan + spermidine carbon dots	Film thickness not specified	30% Sustained antimicrobial effect	Microbial assays	Low microbial contamination during storage	Antimicrobial	Electrostatic cell membrane disruption	59
Confectionery gummies	Carrageenan film for preservative diffusion	Not specified	Preservative migration controlled	Microbial assays	Up to 70% reduction in microbial growth	Antimicrobial	Preservative slow release	46
Dry goods (breads)	Cassava starch + $\kappa$ -carrageenan film	Not specified	15% improvement in tensile strength	Mechanical testing	Increased flexibility and durability	Stability	Improved cross-linking	49
Strawberries	Composite coating (hyaluronic acid + carrageenan)	0.1–0.5% Coating thickness	Reduced 12% weight loss and preserved pH and acidity	pH, weight loss, and total acidity	Retained freshness for 15 days	Preservation	Maintains membrane integrity	61
Jamun fruit	Gelatin + modified $\kappa$ -carrageenan film	3% Gelatin, $\kappa$ -CGN modified dose with film thickness 25–30 $\mu\text{m}$	No toxicity and enhanced antioxidant retention	Stability and antioxidant assays	Improved antioxidant activity on packed fruit	Protective effect	Antioxidant encapsulation	65
Chicken meat	Carrageenan + lime EO composite film with essential oil contents 1–2%	Film thickness 30 $\mu\text{m}$	Reduced microbial count by $\sim 85\%$ after 7 days	Microbial colony count	Maintained safety and slowed spoilage	Antimicrobial	EO disrupts microbial membranes	76
Fresh-cut guava	Sodium CMC + refined carrageenan coating	Coating spray, weight $0.05 \text{ g cm}^{-2}$	Extended shelf life by 6 days	Food product	Form/dose level/Size	Dose concentration	Health effects	25
Grapefruit	Carrageenan + grapefruit EO film 0.5–1%	Film thickness 25–40 $\mu\text{m}$	Tensile strength increased by 50%; UV blocked	Mechanical and UV transmission tests	Higher mechanical and UV stability	Antimicrobial	Cross-linked polymer network	76

## 5.2 Rheological properties

The properties of films that relate to their flow behavior are expressed as rheological properties. The viscosity of carrageenan depends on the composition of other ingredients present in it. It varies depending on the temperature and shear rate. Carrageenan films typically exhibit viscoelastic properties, meaning they display both elastic and viscous properties. These properties make the films flexible and able to handle mechanical stress.<sup>24</sup> In films made from kappa carrageenan and hydroxypropyl methyl cellulose, sorbitol, glycerol and PEG-400 are used as plasticizers. These plasticizers reduce the intermolecular forces between the polymer chains, making the film more flexible and ductile. The addition of plasticizers reduces the film's viscosity, resulting in shear thinning behavior, *i.e.*, the higher the shear stress, the easier the flow of the film. They help to maintain the balance between the elasticity and viscosity of a film.<sup>78</sup> A combination of starch and carrageenan exhibits non-Newtonian, thixotropic behavior, where the viscosity decreases under constant shear and regains its original viscosity at rest, which helps maintain the stability of the films. The concentration of starch and carrageenan alters the viscosity; an increase in carrageenan concentration increases the viscosity.<sup>8</sup> The addition of montmorillonite and nanocrystals, such as cellulose, strengthens the films, making them resistant to deformation by improving their structural integrity.<sup>80</sup>

## 5.3 Barrier properties

The barrier properties of the film encompass several essential characteristics, including protection against light, water vapor permeability, and surface hydrophobicity. Carrageenan films typically exhibit poor water vapor permeability due to their hydrophilic nature, which can limit their application in food packaging.<sup>81</sup> To address this limitation, carrageenan is often crosslinked with other ingredients, such as nanoclays, proteins, or lipids, which can improve moisture barrier properties by increasing film density, strengthening inter-polymer interactions, and reducing water molecule diffusion.<sup>82</sup> The barrier efficiency is influenced by factors including film density, the balance of hydrophilic and hydrophobic components, and the integrity of the film's matrix structure.<sup>5</sup> These films possess barrier properties against gases such as oxygen and carbon dioxide, which help prevent food spoilage.<sup>26</sup> The water vapour transmission rate (WVTR) measures the amount of water vapour that can pass through the film. A lower WVTR is mandatory to maintain a product's quality and shelf life. An edible film made from a combination of carrageenan, pectin and curcumin exhibits a lower WVTR when the concentration of pectin and curcumin is increased.<sup>83</sup> The composite film of carrageenan, polyvinyl alcohol, and spermidine carbon dots has shown strong resistance to water vapor transmission. The combination of carrageenan with polyvinyl alcohol is responsible for these strong barrier properties. Additionally, the film exhibits low migration properties, making it difficult for components of the film to leach into the food product. This ensures the food safety of the packed products and prevents cross-contamination.<sup>59</sup> A study that included a composite film

of  $\kappa$ -carrageenan incorporated with guar gum and locust bean gum indicates an improvement in the barrier properties of the film. The incorporation of guar gum reduced the water vapour permeability of the films which helps to retain the inherent moisture of the product.<sup>84</sup>

## 5.4 Thermal properties

The potential and reaction of the films to higher temperature conditions are expressed as thermal properties, which are generally determined using a differential scanning calorimeter and thermogravimetric analysis. During the film formation process, the addition of various plasticizer substances, such as glycerol and sorbitol, can influence the interaction between the carrageenan film components, thereby altering the thermal properties of the film. Moreover, an increase in the concentration of glycerol decreases the melting point, which leads to good stability of the film even at temperatures as high as 200 °C. On the other hand, sorbitol does not decrease the melting point and is unstable at higher temperatures.<sup>5</sup> Films made with carrageenan have good thermal stability, *i.e.*, they can withstand up to specific temperatures without any degradation. The film does not release harmful components during high-temperature treatments, such as pasteurization or cooking. This ability of the film ensures the quality of the food packed. Thermal properties also influence other properties, such as barrier and mechanical properties. For example, if the structure of a film remains unchanged at higher temperatures, those films would also exhibit better mechanical strength and barrier properties at higher temperatures. It has been found that the addition of other substances to carrageenan films modifies their chemical structure, thereby improving their thermal properties.<sup>26</sup>

The thermal stability of films made with kappa carrageenan and grapefruit essential oil is analysed using derivative thermogravimetry (DGA), Differential Scanning Calorimetry (DSC), and thermogravimetric analysis (TGA), and the analysis reveals that the films made with grapefruit oil withstand higher temperature when compared to the normal/control film, that is the thermal stability of the film is enhanced.<sup>76</sup> Plasticisers such as sorbitol, glycerol and PEG-400 improve the films' thermal stability and help maintain the films' structural integrity at elevated temperatures.<sup>78</sup> The heat resistance of the  $\kappa$ -carrageenan film is increased up to 275 °C by adding hybridized cellulose nanocrystals and organically modified montmorillonite.<sup>80</sup>

## 5.5 Optical properties

The study on carrageenan-based composite films made with zinc oxide nanoparticles (ZnONPs) and carrageenan reports the films' surface color, transparency, and UV-blocking properties. Zinc salts, such as zinc chloride, zinc acetate, and zinc nitrate, are added to form zinc oxide nanoparticles. It is identified that incorporating ZnONPs does not affect the film's colour and transparency. However, the film exhibited different UV-blocking abilities depending on the type of ZnONPs incorporated. The composite film made with carrageenan and ZnONPs exhibited the highest UV-blocking ability.<sup>85</sup> The film containing titanium



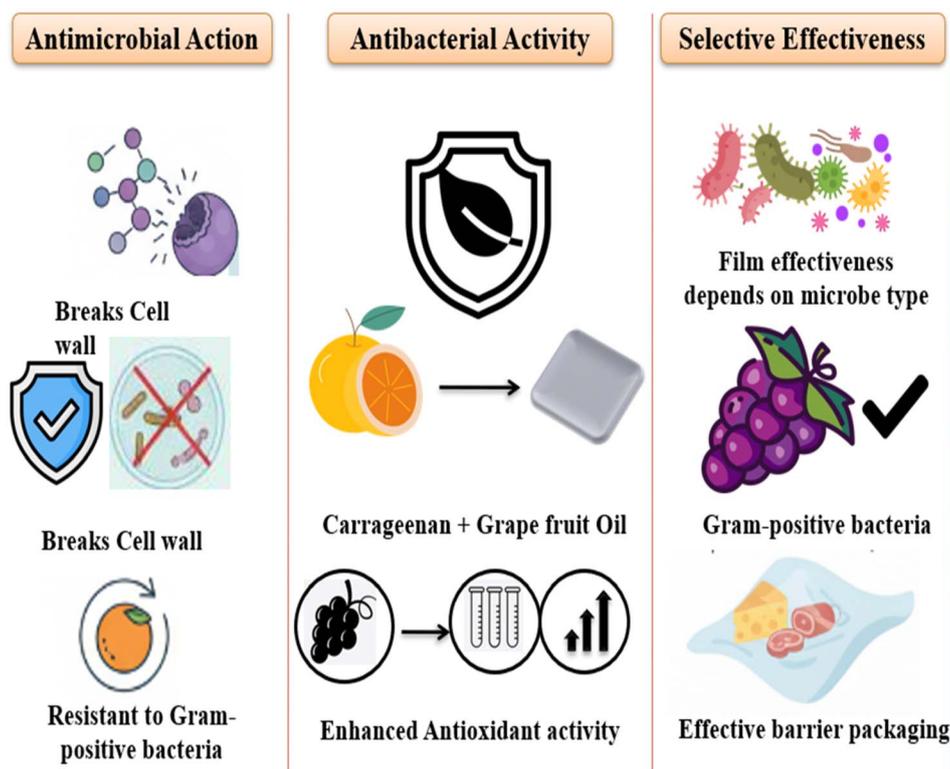


Fig. 3 Resistance of carrageenan films towards antimicrobial action.

oxide carbon dots and anthocyanins exhibited 100% UV protection. The film colour changes according to the pH, showing calorimetric responses in the range of pH 7–12. This property enables the film to serve as a visual indicator of food freshness. This ability of the film is analyzed using a shrimp freshness monitor; the color of the film changes from pink to yellow/brown, indicating the transition from fresh to spoiled shrimp, respectively. This property can be used to assess the quality of the product by retailers and consumers.<sup>86</sup> The incorporation of orange essential oil in the carrageenan matrix has decreased the transmission of UV and visible light; the transmission is found to be  $0.14 \pm 0.02\%$  at a wavelength of 356 nm. The transparency analysis also reveals that the film exhibits a certain degree of opacity, which can help protect the product from light exposure.<sup>87</sup>

### 5.6 Morphological characteristics

The morphology of carrageenan-based films significantly influences their mechanical, barrier, and optical properties, which are critical parameters for food packaging applications. Pure carrageenan films generally exhibit a smooth and homogeneous surface morphology, indicating a uniform polymer dispersion and strong intermolecular interactions within the film matrix.<sup>6</sup> However, the incorporation of nanoparticles such as zinc oxide (ZnO) and silver nanoparticles alters the surface topology, leading to roughened yet more compact and dense film structures.<sup>88</sup> Kappa-carrageenan films reinforced with silica and silver nanoparticles exhibited improved mechanical

properties due to enhanced film compactness, as corroborated by scanning electron microscopy (SEM), which showed a reduced porous network. Moreover, morphological studies have revealed that plasticizers such as glycerol or polyvinyl alcohol (PVA) affect the film microstructure by reducing brittleness and promoting flexibility.<sup>79</sup> The alteration of hydrogen bonding by plasticizers creates microdomains within the matrix, enhancing the elongation at break without compromising surface uniformity.<sup>78</sup>

Additionally, composite films blending carrageenan with starch or other polysaccharides exhibit phase-separated structures where synergistic molecular interactions result in films with balanced mechanical strength and biodegradability.<sup>8</sup> Advanced morphological assessments using atomic force microscopy (AFM) and SEM confirm that optimized solvent casting and drying conditions yield films with fewer cracks and homogeneous thickness distribution, critical for industrial scalability and consistency in packaging applications.<sup>89</sup> These structural attributes are vital for ensuring that films retain functional properties such as oxygen and water vapour barrier capacities while being sufficiently transparent and flexible for consumer acceptance.<sup>47</sup>

### 5.7 Antioxidant and antimicrobial properties

Carrageenan possesses inherent antibacterial properties, which are attributed to its ability to break down the cell walls of bacteria, thereby inhibiting their growth and reproduction. The antibacterial properties can be enhanced by using



Table 2 The overall property enhancement for carrageenan films

Properties enhanced	Combinations	Data	Benefits	Reference
Improved mechanical properties	Semi-refined carrageenan, cinnamaldehyde pickering (0.5%), and titanium oxide nanoparticles (3%)	Tensile strength – 21.86 MPa Elongation break – 34.2%	Maintains food quality at various temperatures and during transportation	92
	Carboxymethyl cellulose (CMC), hydroxyethyl cellulose (HEC), and $\kappa$ -carrageenan	Tensile modulus – 459% Tensile strength – 305% Fracture energy – 398%	Ability to withstand deformation under higher stress conditions	93
	Eugenol (0.4%), carrageenan, and cellulose nanofiber	Tensile strength – 38.08 $\pm$ 2.06 MPa Elongation break – 21.95 $\pm$ 9.02%	Enhanced overall durability and stability of the film	94
	$\kappa$ -carrageenan (3%), poly vinyl alcohol, gallic acid (6.25 & 10 w/v), and metallic ions	Tensile strength – 8.50 $\pm$ 0.61 MPa (kc3/GA6.25) and 10.28 $\pm$ 0.65 (kc3/GA10) Elongation at break – 2.36 $\pm$ 0.16% (kc3/GA6.25) and 1.19 $\pm$ 0.17% (kc3/GA10)	Stable films to maintain the structural integrity of the films	44
Enhanced thermal properties	Carrageenan, nano-clay, tetraethyl orthosilicate (TEOS), and aminopropyltriethoxysilane (APTES)	Tensile strength – 38.6 MPa (3% nano clay), 57 MPa (3% APTES), and 60 MPa (3% TEOS)	Improved mechanical properties without altering other properties	95
	Carrageenan, hydroxylpropyl methyl cellulose, and modified sago starch	Glass transition temperature, $T_g$ – 47.8 $^{\circ}$ C (max) Activation energy – 74.4 kJ mol $^{-1}$	Maintains the structural integrity of films up to certain temperatures	96
	Carrageenan and <i>chlorella vulgaris</i>	Activation energy – 42.16 kJ mol $^{-1}$	Requires higher energy to decompose the film	97
	Eugenol (0.4%), carrageenan, and cellulose nanofiber	Activation energy – 365.82 kJ mol $^{-1}$	Stable at higher temperatures	94
Rheological properties	Carrageenan and <i>chlorella vulgaris</i>	Glass transition temperature, $T_g$ – 65.3 $^{\circ}$ C (max) Activation energy – 64.7 kJ mol $^{-1}$	Used to develop capsules in the pharmaceutical industry	98
	Semi-refined carrageenan and potassium hydroxide	Elasticity – 50.24%	Can be utilized for multiple applications, as they are versatile	97
	Lambda carrageenan	Film thickness: 30.5–34.1 $\mu$ m	Can be molded into different shapes due to their flexibility	77
Barrier properties	Lambda carrageenan and sodium	Viscosity – 204 dL g $^{-1}$ Viscosity – 14.7 dL g $^{-1}$	The balanced viscosity of the film helps in more consumer acceptance and can be applied for various industrial uses	99
	Carrageenan and olive leaf extracts	Film thickness – 0048 $\pm$ 0.004 $\mu$ m	The desired thickness of films is developed by altering the viscosity	100
	$\kappa$ -carrageenan (2.5%), poly vinyl alcohol, gallic acid (6.25 & 10 w/v), and metallic ions	Water vapor permeability value – 5.36 $\pm$ 0.51 (kc2.5/GA6.25), and 3.76 $\pm$ 0 (kc2.5/GA10)	Essential to impart moisture barrier properties	44
	Carrageenan, nano-clay, tetraethyl orthosilicate (TEOS), and aminopropyltriethoxysilane (APTES)	UV transmission rate – 23% (TEOS), 18% (APTES), and 1% (nano clay)	Used for high UV protection	95
	Carrageenan and lignin	Water vapor permeability 1.14 $\times$ 10 $^{-9}$ g m $^{-2}$ pas.	Best for hydrophobic and UV protection	79



Table 2 (Contd.)

Properties enhanced	Combinations	Data	Benefits	Reference
Antioxidant	Carrageenan and layered double hydroxides	Contact angle – 78 °C, 100% UV protection	Good barrier properties against oxygen transmission	23
		Water vapor permeability – $1.98 \times 10^{-11} \text{ g m}^{-2} \text{ pas}$ Oxygen permeability – $1.05 \times 10^{-12} \text{ g m}^{-2} \text{ pas}$		
	Carrageenan, arrowroot starch (80%), and polyethylene glycol	Water vapor permeability – $85.12 \text{ g m}^{-2} \text{ 24 h}$	Improved barrier when compared with pure carrageenan films	101
	Eugenol (0.4%), carrageenan, and cellulose nanofiber	Radical scavenging efficiency – 58%	Helps to maintain the quality and safety of foods	94
	Carrageenan and olive leaf extracts	Radical scavenging efficiency – 89.5%	Prevent foods from oxidative spoilage	100
	Semi refined carrageenan, glycerol, and alpha tocopherols	DPPH assay inhibition – 88.59%	Can be utilized in active packaging	102

antimicrobials and modifying the film's composition. Films effective against specific bacteria can also be developed.<sup>26</sup> The antioxidant activity of the film made with kappa carrageenan and grapefruit oil is determined using ABTS (2,2'-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid)) and DPPH (2,2-diphenyl-1-picrylhydrazyl) assays. The results reveal an increase in antioxidant activity, which is attributed to the natural antioxidants present in grapefruit.<sup>76</sup> Composite films containing zinc oxide nanoparticles demonstrated potent antibacterial activity against specific pathogens, including *Escherichia coli* O157:H7 and *Listeria monocytogenes*. The film made with carrageenan (ZnO-NPsZC) (composite with zinc chloride) showed the highest antibacterial activity, proving that adding zinc oxide nanoparticles enhance the antibacterial activity of the carrageenan films.<sup>85</sup> A study investigating the antimicrobial properties of orange essential oil-incorporated carrageenan-based films showed that the films are significantly resistant to Gram-positive bacteria, especially *Staphylococcus aureus*. In the case of Gram-negative bacteria, the films exhibited only specific resistance to growth. Therefore, the effectiveness of the film may depend on the type of microorganism that grows in the food medium.<sup>87</sup> Fig. 3 illustrates the carrageenan films' resistance action towards antibacterial and antimicrobial activity.

### 5.8 Oxygen permeability

Oxygen permeability is a measure of the passing of oxygen through a particular material.<sup>138</sup> Materials with low oxygen permeability help maintain the freshness of food by reducing the oxidation process and improving the product's shelf life.<sup>90</sup> Carrageenan films have limited barrier properties. The addition of Layer Double Hydroxides (LDHs) to carrageenan reduces the oxygen permeability of the film by creating a more ordered and compact structure, which blocks the way for oxygen diffusion. These films exhibited lower oxygen permeability compared to pure/unmodified carrageenan films.<sup>91</sup> The effectiveness of the

film is determined by comparing the film with existing benchmarks. These properties make them suitable for industries other than food, such as pharmaceuticals and other industrial applications that require gas barriers.<sup>23</sup>

### 5.9 Gel strength and synergistic properties

The ability of carrageenan to form gels is due to the sulphur group present in it. When carrageenan is heated with water, it dissolves in the water and, upon cooling, forms a gel. This process is greatly influenced by temperature. The primary mechanism behind gel formation is ionic interactions, especially with cations such as potassium and calcium. These cations contribute to the formation of a three-dimensional network known as gels. During this process, water molecules are trapped within the gel network, which is essential for the film to maintain its integrity.<sup>26</sup> When carrageenan reacts with other polymers, such as pectin or gelatin, the film exhibits synergistic effects that enhance the gel-forming ability of carrageenan, thereby improving the mechanical properties, texture, and stability of the films.<sup>24</sup> The addition of hybridized cellulose nanocrystals and montmorillonite yields synergistic effects in films, resulting in strong mechanical properties, reduced water absorption, and improved water resistance compared to natural bio-nano composites with a single filler.<sup>80</sup> The overall collaborative table for property enhancement is illustrated in Table 2, and the gel formation mechanism is illustrated in Fig. 4.

## 6. Advancement and trends in carrageenan packaging

The advancement of carrageenan in packaging has gained attention in recent years due to its biodegradability, non-toxicity, and potential to replace synthetic polymers. Much



## GEL FORMATION MECHANISM

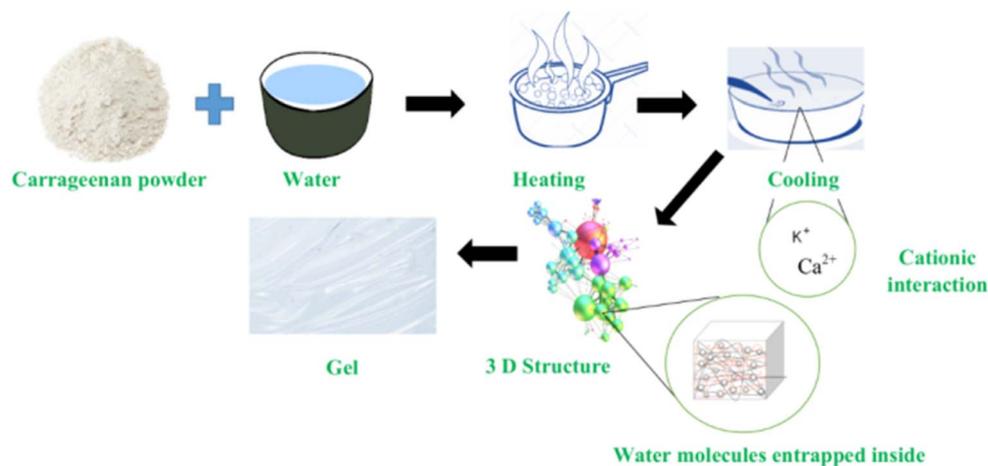


Fig. 4 Mechanism of gel formation in carrageenan.

research highlights the development of carrageenan-based films, which not only extend food shelf life but also incorporate active and intelligent features in packaging, such as biopolymer blending and enhancement of properties through various extraction techniques. These films have gained widespread acceptance among consumers.<sup>24</sup> Below are some trends and advancements in the packaging sector.

### 6.1 Modification of carrageenan

Carrageenan has wide applications due to its non-toxicity, transparency, biodegradability, renewability, barrier properties against gas and moisture, and film-forming ability. Due to certain limitations, such as a low water vapour barrier and poor mechanical strength and water resistance properties, the need for adding or blending with other materials arises.<sup>103</sup> In terms of hydrophobic modification of kappa ( $\kappa$ ) carrageenan with 1-octyl chloride (alkyl halide) by the Williamson etherification reaction, at three different degrees of substitution (1, 2, and 3), all these modifications showed no change in the rheological properties. They improved the amphiphilic behavior of  $\kappa$ -carrageenan. The highest degree of substitution exhibited better amphiphilic behavior and emulsifying power.<sup>104</sup> A chemical-free method of extraction for hybrid carrageenan *Sarcopeltisskottsbergii* produces the mixed kappa/iota carrageenan in the ratio 6:4. The results showed, at a varying temperature from 110–220 °C, that the variation in the compound extraction takes place (110 °C for film applications and 140 °C for cosmetic and food applications), and showed enough rheological properties for its intended commercial uses.<sup>105</sup> Prior enzymatic pretreatment (cellulose) on alkali extraction of carrageenan from *Kappaphycus striatum*, with KOH and NaOH, was done. KOH exhibited significantly improved viscosity values (272–360 cP) and a higher gelation temperature (42.78 °C). However, no such effect was observed in cellulose-

treated and NaOH-extracted  $\kappa$ -carrageenan. This was mainly due to the better alkalization of  $\kappa$ -carrageenan by KOH compared to NaOH.<sup>106</sup>

Another effect of carrageenan with 1 M NaOH and without alkali modifications, in terms of its physical, chemical, water vapor, and thermal conductivity behavior, was studied. The unmodified method of carrageenan extraction at 60 °C showed the highest Young's modulus (1433.3 MPa) and tensile strength (146.4 MPa) values, which were due to stronger hydrogen bonding.<sup>107</sup> However, the modified method of carrageenan exhibited poor Young's modulus and tensile strength values due to the conversion of hydroxyl and sulfate ester groups into 3,6-anhydrogalactose, which in turn resulted in the highest elongation at break (33.2%) value when extracted at 80 °C. Irrespective of the modifications, the contact angle (°) values of the film improved at higher extraction temperatures, specifically at 80 °C, over 60 °C. This is because, at higher temperatures, hydroxyl and sulfate ester groups were removed, thereby increasing the surface hydrophobicity of the film.<sup>108</sup> There exists an inverse relationship between the contact angle (a measure of hydrophobicity) and the water vapor transmission rate. The highest value was shown by the native film (683.91 g per m<sup>2</sup> per day), while the least by the modified film (583.09 g per m<sup>2</sup> per day), both extracted at 80 °C. A higher extraction temperature rendered all the films hydrophobic.<sup>107</sup> These results showed that by prioritizing any particular properties, we can go for the specific type of film and chemical modifications showed improvements in the film properties, but it could be improved with further specific temperature and other property-based modifications.<sup>107</sup>

### 6.2 Structure and blending of carrageenan

Before blending carrageenan with other materials, the main properties and applications of carrageenan, particularly in



relation to its chemical structure, need to be discussed. The industrial production of carrageenan from seaweeds mainly depends on the four major species *Kappaphycus alvarezii*, *Sarcotialia crispate*, *Chondrus crispus*, and *Eucheuma denticulatum*.<sup>109</sup> Of these major carrageenan red algae sources (with their three different types of structure, like kappa, iota, and lambda), *Kappaphycusalvarezii* is most commonly used for kappa carrageenan extraction.<sup>110</sup> In general, there are five different variants of carrageenan, such as kappa ( $\kappa$ ), lambda ( $\lambda$ ), mu ( $\mu$ ), epsilon ( $\epsilon$ ), and iota ( $\iota$ ), which vary in molecular weights from 100–1000 kDa.<sup>111</sup> Carrageenan configurations, such as kappa and iota, are double helices in structure and withstand

a relative water vapour pressure of 0.9, but are not stranded by the single helix of lambda carrageenan. The carrageenan-based packaging material retained its shape even when exposed to water vapour, but dissolved in water when used alone.<sup>112</sup> Among them, the most widely used were the  $\kappa$ , followed by the  $\iota$  and  $\lambda$ , and further details are listed in Table 3.

When carrageenan was blended with galactomannans, namely locust bean gum and guar gum, the resulting composite films exhibited enhanced functional properties. Notably, a lower water vapour permeability of  $2.35 \times 10^{-11} \text{ g mm s}^{-1} \text{ m}^{-2} \text{ Pa}^{-1}$  was achieved with 25% locust bean gum, along with improved thermal stability and a maximum light transmittance of 57.06%

Table 3 Structural arrangements, properties and applications of carrageenan

Carrageenan type	Structural arrangement	Properties	Applications in the food industry	Reference
Kappa ( $\kappa$ )	One ester sulfate (ES) and one 3,6-anhydrogalactose (3,6-AG) per disaccharide Ester sulfate 25–30%	Forms a strong, brittle, rigid gel, especially with $\text{K}^+$ ions  Hot water-soluble sodium salts dissolve in cold water	Stabilizing, gelling and thickening agent; film-forming for packaging and edible coatings; meat, dairy, and confectionery	30, 76 and 113
	3,6-Anhydrogalactose: 28–35%	Linear polysaccharide; helical structure Tensile strength film range $\sim 38\text{--}98 \text{ MPa}$ Water gel strength $\geq 700 \text{ g cm}^{-22}$		
Iota ( $\iota$ )	Two sulfates and one 3,6-AG per disaccharide Ester sulfate: 28–30%	Forms soft, elastic, freeze-thaw stable gels with $\text{Ca}^{2+}$ Cold water partially soluble (requires heating for complete dissolution)	Used in salad dressings, soy milk stabilizers, and pet food gels	34 and 111
	3,6-Anhydrogalactose: 25–30%	Helical structure Lower tensile strength; thixotropic behaviour is advantageous for suspensions		
Lambda ( $\lambda$ )	Three sulfate groups, absence of 3,6-AG Ester sulfate 32–39%	Non-helical structure  Cold water soluble; no gel formation	Thickener in tomato sauce, dairy products, and creamy textures	99 and 114
	3,6-Anhydrogalactose: 30%	Pseudo-plastic (shear thinning) rheology Viscosity $\sim 204 \text{ dL g}^{-1}$		
Mu ( $\mu$ ) & epsilon ( $\epsilon$ )	Less common variants with modified sulfate 3,6-AG content	Variable gelation and solubility properties depending on structural modifications	Emerging interest in specialized food packaging applications	111
Alkylated kappa carrageenan	Less common variants: amphiphilic behaviour enhanced by octyl chloride substitution	Improved emulsifying capacity without sacrificing rheology	Potential for active food packaging	104
Kappa + locust bean gum or guar gum	Less common variants with modified sulfate	Reduced water vapor permeability $\sim 2.35 \times 10^{-11} \text{ g mm s}^{-1} \text{ m}^{-2} \text{ Pa}^{-1}$ at 25% LG; improved thermal stability; oxygen barrier	Enhanced edible films for packaging	84
Kappa + plasticizers (glycerol, PEG, PVA)	Less common variants with modified sulfate	More flexible, pliable, thermoreversible gels reduce tensile strength but increase elongation at break ( $\sim 21.4\%$ ) via reduced intramolecular H-bonds	Film processing and texture tuning	88



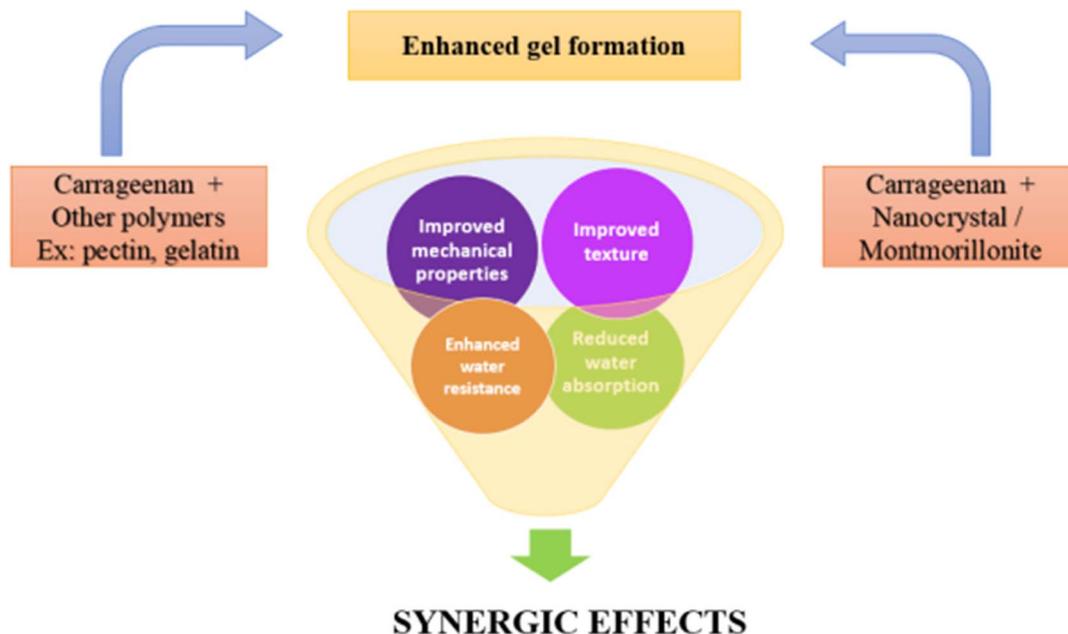


Fig. 5 Picture representation for synergistic effects of blended carrageenan films.

at 15% locust bean gum. Additionally, increasing galactomannan content led to reduced oxygen permeability and swelling ratio in the films.<sup>84</sup> On investigating carrageenan with different types of plasticizers, glycerol (G), polyvinyl alcohol (PVA), and polyethylene glycol (PG) and adding the additive palmitic acid (PA) at different concentrations, the overall results showed that both the addition of plasticizers and palmitic acid reduced the tensile strength. However, the plasticizers increased the elongation at break value, and palmitic acid increased the hydrophobicity of the film. The film, which showed the maximum value obtained for the tensile strength to be 5.725 MPa (92 : 3 w/w of C : PG), had an elongation at break value of 21.43% (81 : 15% w/w of C : PVA). It was reported that both these changes occurred due to a reduction in the intramolecular force and hydrogen bond strength, which caused an increase in the flexibility of the film by the reduction in the strain break force.<sup>88</sup> The pictorial representation is illustrated in Fig. 5.

### 6.3 Biodegradable packaging films

For the past two decades, researchers have focused on the biodegradability of the film, as it is not only eco-friendly and edible but also fertilizes the soil according to European standards for bioplastics. A film made from food-grade additives and biologically degradable polymers is called a biodegradable film.<sup>114</sup> In a study, different types of 2% (w/w) carrageenan (*Kappaphycus alvarezii*, kappa, refined, and semi-refined) were mixed with 0.9% (w/w) glycerol and with reinforced cellulose nanofibers at different concentrations (0.5, 10, and 15% v/w). The overall result showed that the film had improved tensile strength and a value of elongation at break greater than 10% (except for the *Kappaphycus alvarezii* film).<sup>94</sup> Tensile strength ranges from 10 to 100 MPa, and elongation at break outperforms at 10%. The incorporation of cellulose improved

elongation at break and tensile strength values but affected the physical properties. Different cellulose nanofibre (CNF) compositions at 15% maintains the same thickness of 0.060 mm, decreases the opacity from  $4.50 \pm 0.40$  to  $7.29 \pm 0.18$  and decreases the water solubility from  $60.00 \pm 4.60$  to  $43.92 \pm 0.39$ . Among these combinations, the best film that was suitable for the production of the active packaging was that with semi-refined carrageenan, glycerol, and 10% cellulose nanofibers, as it showed the highest mechanical properties (45.98 MPa), the highest temperature for decomposition and also the desired packaging material characteristics like lower water solubility (42.59%) and water vapour permeability.<sup>94</sup>

When a blended film was made from starch (arrowroot) and carrageenan, its degradability in terms of both seawater and a compost environment was examined. When blended with 0.5% carrageenan and 3.5% arrowroot starch, or a film with only 4% arrowroot starch, the film was biodegradable in 42 days in seawater (due to reduced thermal oxidation exposure) and within 7 days under soil conditions.<sup>115</sup> Investigation of a bioplastic cup made with carrageenan extracted from *Eucheuma cottonii* and with modified starch found that incorporating modified starch in high proportions increased water resistance but reduced biodegradability. The highest biodegradability (55.68%) was observed in films prepared using carrageenan alone. In terms of water resistance, the proportion with 3 g each of carrageenan and modified starch showed the best water resistance values of 48.86%, 71.33%, and 97.13% at low, room, and high temperatures, respectively.<sup>116</sup>

An investigation using the solution casting method on  $\kappa$ -carrageenan and nano-silica was conducted earlier for the application of films in food and pharmaceutical packaging. The best film was made from 2 g of  $\kappa$ -carrageenan (KC) dissolved in 200 mL of water (1% w/v), followed by the addition of sonicated 0.01 g of nanosilica (0.5% w/w of KC) and 0.8 g of glycerol (40%



w/w of KC). This optimized film demonstrated improvements in tensile strength (13.8%), tensile modulus (15%), and better crystallinity, as well as enhanced rheological properties (viscosity and storage modulus), and a reduced water vapor transmission rate (50%) compared to the control KC film.<sup>89</sup> The decrease in all properties mentioned above at increased concentrations is due to the higher concentration of nano-silica, as it formed agglomerates in the film at higher concentrations. However, at high concentrations of nano-silica (1.5% w/w of KC), due to agglomerates in the film, it improved the thermal properties, such as the melting temperature (16.8%) and enthalpy of melting (11%), compared to those of the control KC film. Silica's thermal insulating nature increased the 50% degradation temperature (11.5%) and 65% degradation temperature (32.9%).<sup>89</sup>

#### 6.4 Other advancements

Recent advances in carrageenan-based food packaging films focus on improving their properties and utility. Extraction methods are becoming more efficient through the use of combined techniques. Pure carrageenan films are now fortified with additional compounds, enhancing their physical and chemical properties. These improved films extend food shelf life by inhibiting microbial growth and reducing moisture loss. Research also explores their role in monitoring food freshness.<sup>5</sup> When kappa carrageenan was incorporated with the essential oil obtained from the *Cymbopogon winterianus*, it showed transparency (>90%), hydrophobicity, antibacterial activity against *Listeria monocytogenes* (LMG 16779) with a 31.67 mm Zone of Inhibition (ZOI) and a minimum inhibitory concentration (MIC) of 8  $\mu\text{L mL}^{-1}$  and antioxidant activity. The antioxidant and antimicrobial activity was attributed to the presence of more than 40% citronellal in the essential oil.<sup>117</sup> Optimizing the carrageenan-based biofilm with glycerol and montmorillonite concentration yielded the best film at 3% (v/v) and 20% (w/w), respectively. These films exhibited good values for tensile strength, elongation, and Young's modulus, which could be used in food applications as an alternative to commercial plastic films to a greater extent.<sup>118</sup> Kim<sup>119</sup> developed a uniform-sized silver nanoparticle of  $63.9 \pm 2.1$  nm, extracted from pine needles (3% w/w), which was incorporated into a carrageenan film (4% w/w) with glycerol (1.2 g). It exhibited properties such as perfect ultraviolet blocking, 87% antioxidant activity, and moderate antimicrobial activity, with 2 log CFU  $\text{mL}^{-1}$  and 3.5 log CFU  $\text{mL}^{-1}$  reductions in both bacteria, *E. coli* O157:H7 (Gram-negative) and *Salmonella aureus* (Gram-positive), respectively. The use of this film for food packaging was also suggested to extend the shelf life of packaged food products.

Another nanoparticle-based film developed using a combination of kappa carrageenan, silicon dioxide, hydroxyethyl cellulose, and silver nanoparticles, is suggested for use in packaging and biomedical applications due to its improved properties. This includes transparency, enhanced thermal properties (TGA T5% 115.7 and T10% value 150.4), mechanical properties, shear thinning effect, UV inhibition, and improved storage modulus.

This film also demonstrated a zone of inhibition (antimicrobial activity) against *Staphylococcus aureus* (34 mm), *Cronobacter sakazakii* (25.7 mm), *Salmonella typhi* (22.3 mm), *Bacillus subtilis* (19 mm), *Listeria monocytogenes* (19.7 mm), and *Bacillus cereus* (15.3 mm). The other films without incorporation of the silver nanoparticles showed no inhibition.<sup>120</sup>

A smart oxygen indicator tag was made from kappa carrageenan through electrospinning. The electrospinning solution at increasing temperature (25–45 °C) exhibited shear thinning behaviour (the power law suited best for that behaviour, but showed increased electrical conductivity from  $54.17 \mu\text{S cm}^{-1}$  to  $141.5 \mu\text{S cm}^{-1}$ ). The optimized condition for electrospinning was obtained with a voltage of 8 kV and a flow rate of  $0.4 \text{ mL h}^{-1}$  from a distance of 10 cm, and showed a non-woven type structure with 1–2 microns. Irrespective of the air conditions (ambient, *i.e.*, 21%  $\text{O}_2$  and reduced to 0.4%  $\text{O}_2$ ), the ultraviolet-activated tags showed 7 min as the photoactivation time, while the recovery time varied from 7.67–24.67 hours. This increased as the colour recovery of the film depended on the oxygen concentration. This tag remained stable for 2 months (60 days) when stored at 25 °C in the dark with a relative humidity of 65%.<sup>121</sup> This UV-activated electrospun oxygen tag suggests its applications in MAP for seal integrity, purity checks for MAP gases, and oxygen residue on food through visual color change.<sup>121</sup>

Another study on improving the physical, mechanical, and water vapor permeability of kappa carrageenan-based films was conducted by Shojaee *et al.*,<sup>6</sup> who found that incorporating *Zataria multiflora* Boiss essential oil and montmorillonite nanoclay at a 10% concentration improved the tensile strength from 26.29 MPa to 34.67 MPa. While *Zataria multiflora* Boiss essential oil (ZMBEO) incorporated at a 3% concentration reduced the water vapor permeability of the film by approximately 78%, the film made from  $\kappa$ -carrageenan, 5% montmorillonite nanoclay and 3% ZMBEO by the overlay method showed maximum antimicrobial activity with zone of inhibition (ZOI (mm)) values against *Salmonella aureus* ( $538.27 \pm 27.59$ ), *Bacillus cereus* ( $480.58 \pm 15.54$ ), *Escherichia coli* ( $359.12 \pm 12.11$ ), *Salmonella typhimurium* ( $243.35 \pm 10.34$ ) and *Pseudomonas aeruginosa* ( $168.40 \pm 15.19$ ). Only by reducing the concentration of ZMBEO to 1% in the aforementioned film composition, except for *P. aeruginosa*, did the film show some ZOI.<sup>122</sup>

A study was conducted using the most promising strain isolated from 25 different endophytic fungal strains for cellulase production. This strain, identified as *Amphiroa anceps*, was mass-cultivated and partially purified to extract carrageenan from red seaweed. Films were prepared with 1% (w/w) carrageenan, and combined with 0.75% (w/w) plasticizers like polyethylene glycol and glycerol in equal proportions along with 0.1 g of one of the antibiotics (tetracycline, amoxicillin, chloramphenicol, cephalixin, doxycycline, erythromycin, ofloxacin, ampicillin, or dicloxacillin) to form separate films. These films were evaluated against various food spoilage pathogens, including *Shigella* sp., *Vibrio parahaemolyticus*, *Escherichia coli*, *Vibrio cholerae*, *Salmonella* sp., and *Listeria* sp. Among them, the cephalixin-coated carrageenan film exhibited the largest zone of inhibition and thus shows potential for application in food preservation.<sup>123</sup>



## 7. Application of carrageenan in food packaging applications

Carrageenan-based films were utilized in food packaging due to their film-forming properties, allowing for transparent indicators on food products. Carrageenan film applications are widely applicable to all food products. However, the major utilized segments include meat and meat substitutes, confectionery, dairy products and their alternatives, and pharmaceutical segments. The rising demand for juicier, flavourful, and clean-label meat and meat alternatives is boosting the use of carrageenan, particularly kappa carrageenan, in processed meats. The increasing adoption of plant-based and flexitarian diets is driving the use of kappa carrageenan to enhance texture in meat analogues. Growth in ready-to-eat and easy meal solutions is fuelling carrageenan usage in the Asian meat industry.<sup>63</sup> In the confectionery market, the increasing demand for gummy candies and jellies among younger consumers is driving the use of carrageenan in the Asian confectionery market.<sup>3</sup> Health-conscious trends are boosting its inclusion in low-calorie and sugar-free snacks, catering to guilt-free indulgence. In fish and meat packaging, carrageenan composite films have demonstrated significant antimicrobial activity, effectively reducing pathogens such as *Escherichia coli* and *Staphylococcus aureus* by over 50%.<sup>124</sup> Moreover, the copper sulfide nanoparticle-carrageenan films showed a 52.6% and 69.8% reduction in the growth of these bacteria in beef, and carrageenan films infused with camellia oil reduced cryophilic bacterial growth on chicken.

Additionally, the antioxidant and antibacterial properties of carrageenan enable its use in dairy product packaging to inhibit microbial growth and lipid oxidation. Carrageenan composite films containing mulberry polyphenolic extracts changed color in response to milk spoilage indicators, such as pH and acidity, offering a freshness monitoring function. Aloe vera gel and carrageenan films inhibited microbial pathogens in ice cream, and carrageenan-black bean extract coatings reduced peroxide formation in cheese samples.<sup>26</sup>

Premium and artisanal confectionery brands are adopting carrageenan for quality and innovative texture. The dairy and alternative sector has been increasing its use of carrageenan packaging due to the growing demand for yoghurt and fermented dairy products in Asian countries, which is driving market growth.<sup>10</sup> Carrageenan has been used as a clean-label ingredient, driven by the inclusion of carrageenan in dairy products due to organic trends. The rising popularity of plant-based dairy alternatives, such as non-dairy creamers and cheeses, propels the adoption of carrageenan. Sustainability awareness encourages the use of carrageenan in eco-friendly and health-focused dairy substitutes. In the pharmaceutical segment, the Asian market is increasingly using carrageenan in liquid formulations for ease of consumption and administration. The rise of herbal and natural remedies is driving the inclusion of kappa-carrageenan in formulations, aligning with the demand for holistic health solutions.<sup>66</sup> Carrageenan is being adopted for its role in enhancing drug delivery systems and

improving efficacy and user-friendliness. Other applications for carrageenan include cosmetics, personal care, and other non-food packaging applications.<sup>63</sup>

## 8. Potential challenges with the packaging materials

The successful commercialization of carrageenan-based films requires overcoming various limitations. Although carrageenan is a biodegradable material, the extraction process for carrageenan has adverse environmental effects. There is a need for sustainable, green extraction techniques for carrageenan. Additives or plasticizers are used to improve the properties of carrageenan-based films. However, considering food safety, the additive must be non-toxic and must not compromise the film's biodegradability. The storage conditions and type of food can affect the ability of carrageenan films to extend the shelf life of food products.<sup>5</sup> The versatility of carrageenan as a packaging material is limited to certain foods. The antibacterial performance of films across different unit operations becomes challenging.<sup>24</sup> The processing of biopolymers is costly compared to traditional plastics, and they also have complex processes and technical difficulties when compared to plastic packaging. The lack of awareness among consumers regarding bio-polymer packaging is one of the biggest challenges for commercialization. Cultural concerns also play a crucial role in consumers' willingness to adopt these types of packaging films. The regulatory and legislative works also become complex in ensuring the standards of films.<sup>103</sup> There are several ongoing research studies in this field, and there is a need for further studies to overcome the above challenges.

## 9. Application and shelf-life extension in the product

A bilayer film was formed by two individual solvent casting methods that measures shrimp freshness by non-destructively monitoring it with visible colour change in the film. The most stable bilayer film was formed by incorporating anthocyanin (0.2%)-loaded liposomes (at three different concentrations: 0.2, 0.5, and 1%) and cholesterol (0.2%), along with carrageenan and agar (ALCA). An increase in lecithin proportion (0.2–1%) is directly proportional to the encapsulation efficiency of anthocyanins (36.06% to 46.99%), which increased the average particle size (1.37-fold), but at higher concentration of lecithin (0.2 to 0.5 and 1%) decreased the zeta potential (−49.01 to −19.96 mV) and slightly reduced the film's ammonia sensitivity. The ALCA film showed 55% lower values of exudation after 50 minutes than the film with non-encapsulated or free anthocyanin (ACA) film. Although encapsulation reduced the anthocyanin activity and water vapor permeability of the films, it improved the anthocyanin stability (maintained an  $\Delta E$  value less than 5) of the film from 4 days to 14 days at 25 °C.<sup>125</sup> With these findings, this ALCA film demonstrated its potential for use under highly humid packaging conditions.<sup>26</sup>



Seaweed dodol, when packed and stored at room temperature, showed a lower bacterial load ( $1.61 \times 10^6$  colonies per g) after 48 h in an edible  $\kappa$ -carrageenan film compared to baking paper ( $1.79 \times 10^6$  colonies per g). After two days of storage at room temperature, the microbial load remained within an acceptable range ( $1.61 \times 10^5$  colonies per g), and in terms of sensory aspects, it was satisfactory for up to 3 days. These preservations by carrageenan were due to the water vapor barrier properties of the  $\kappa$ -carrageenan film.<sup>72</sup> They have concluded that it could be an alternative to plastic films.<sup>72</sup> An edible coating made up of carrageenan (1.5% w/v) and glycerol (0.5% v/v), and carrageenan (1.5% w/v), glycerol (0.5% v/v) and peppermint oil (0.5% v/v) was used over the Ambon bananas (*Musa acuminata cavendish*). Carrageenan-coated bananas exhibited reduced total soluble solids and weight loss compared to uncoated bananas; however, the efficiency of the carrageenan coating decreased with the addition of peppermint oil. There is little difference in the water activity and pH of coated and uncoated bananas. Thus, carrageenan-based edible coating reduced the overall ripening process and reduced the discoloration in ambron bananas<sup>73</sup>

Another study was conducted on edible packaging films made from carrageenan and palm oil. The optimized film was made with a composition of 1.25% carrageenan and 10% palm oil (v/v) as a plasticizer.<sup>126</sup> This film showed good tensile strength ( $102.50 \text{ kgf mm}^{-2}$ ), elongation (7.04%), and oxygen

permeability ( $7.64 \times 10^{-19} \text{ cm}^3 \text{ cm/cm}^2 \text{ s cm Hg}$ ). The film covering apple slices over five days of storage showed better retention of colour, vitamin C (from 0.375 to 0.228%), and reduced weight loss (30.7%) of apple slices.<sup>126</sup> An anti-browning dip of pH 3.5–3.7 (citric acid  $8 \text{ g kg}^{-1}$ , *N*-acetyl-L-cysteine  $4 \text{ g kg}^{-1}$ , and calcium ascorbate  $20 \text{ g kg}^{-1}$ ) with or without any one of these coating like carrageenan ( $5 \text{ g kg}^{-1}$  pH 5.8–5.9), carboxyl methyl cellulose ( $10 \text{ g per kg CMC}$ ,  $5 \text{ g kg}^{-1}$  maltodextrin, pH 3.5–3.7) and chitosan ( $5 \text{ g kg}^{-1}$  with  $50 \text{ g kg}^{-1}$  of citric acid, pH 2.4) was studied in freshly cut mangoes. The best effect was produced by only anti-browning dipped fruits, followed by anti-browning dipped fruits coated with carboxymethyl cellulose, carrageenan and chitosan.<sup>75</sup>

Research was conducted with three individual films with kappa carrageenan (C), agar (A) and konjac glucomannan powder (KGP) and with two multi-component hydrogel films made, one with combinations of 1 g each of carrageenan, agar, konjac glucomannan powder and 0.9 g of glycerol 30% w/w as a ternary blended film (TBF), and another film with further incorporation of nano-clay 5% w/w (Cloisite®30B) in the ternary blended film (CTBF). CTBF showed the highest values for tensile strength (75.76 MPa) and water uptake ratio (548.61%), while the TBF showed higher water vapour adsorptive properties compared to those of all the other films. The incorporation of nano-clay not only improved the mechanical properties but also showed inhibition of *Listeria monocytogenes*. Both the TBF

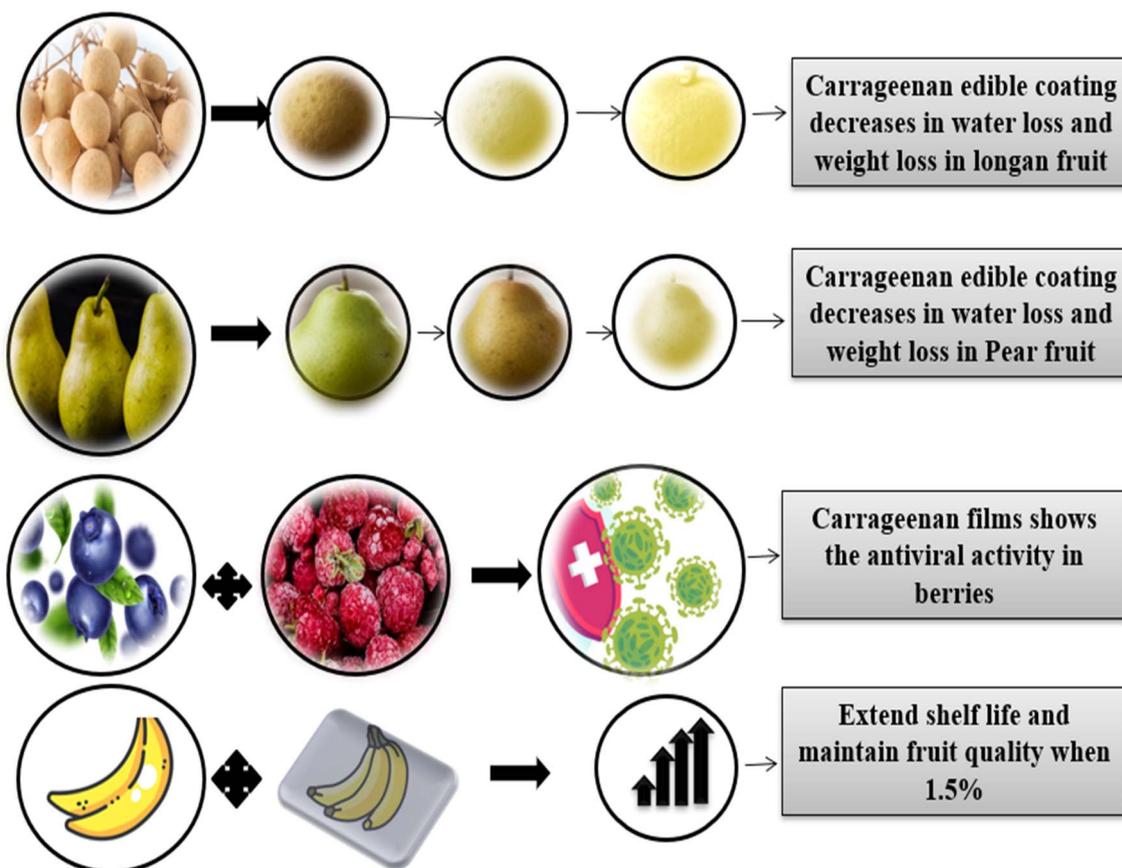


Fig. 6 The application of carrageenan films for the extension of shelf life.



and CTBF showed a reduced weight loss of 8.75% and 8.84%, respectively, in packed and stored spinach after 5 days at 55 °C.<sup>127</sup> Fig. 6 illustrates the application of carrageenan films for extension of shelf life.

## 10. Consumer acceptability of carrageenan films in packaging applications

An experiment conducted in Florida has reported innovations in the market, indicating that a total of 198 food products contain carrageenan. A simultaneous survey has also reported that carrageenan has been incorporated into various food materials, including different varieties of milkshakes, sauces, hamburgers, and different types of packaging films—the amount of carrageenan uptake in Florida ranges from 0 to 7.7 g per day. The statistical difference in intake per kilogram body weight was 40% superior in females compared to males. Additionally, the consumer's likelihood of developing new products incorporating carrageenan has increased.<sup>127</sup> Consumers are accepting transparent, differently coloured, and carrageenan-incorporated packaging films.<sup>49</sup> Different kinds of edible packaging films have also been developed, such as carrageenan, for wrapping chicken patties, which consumers accept due to their environmental protection and degradable qualities.<sup>27</sup> Consumers are more concerned about the disposal of wraps or packaging materials. They are most accepting of the market aesthetic point of view, particularly the different colours incorporated into packaging materials that are harmless from an environmental perspective.<sup>128</sup>

## 11. Toxicity, safety and legal compliance

Carrageenan has a very high molecular weight, which is not degraded or absorbed in the gastrointestinal tract of humans, indicating that it may have lethal effects on humans. The high proportion of carrageenan may lead to significant symptoms, such as gastroenteritis and runny stools. Although these effects are considerable, they do not indicate a highly toxic response or pose long-term health risks. Scientific studies have also concluded that carrageenan does not possess any genotoxic effects, indicating that there is no potential risk of DNA damage.<sup>129</sup> In some cases, it is reported that the mucin layer, which is present in the intestine, is decreased, leading to intestinal malfunction.<sup>130</sup>

Carrageenan, which is incorporated into food and beverages, is not digested in the GI tract. Researchers have reported that carrageenan has been transported through the epithelial cells of the GI tract in domestic guinea pigs. It has also been claimed that oral uptake of carrageenan may lead to an immunogenic response in the GI tract, resulting in ulceration and tissue damage. It is reported that there is no potential evidence of oncogenicity in the animals. It has been reported that consuming food-grade carrageenan has no oncogenic effects, effects on DNA replication, effects on the immune system, or

tumor-promoting effects.<sup>131</sup> The USFDA has declared carrageenan as “Generally Recognized as Safe” (GRAS), indicating that it is considered safe for both operational and functional purposes. Various experts from the WHO and FAO have reviewed and declared that the use of carrageenan in infant formula at a limit of 1000 mg L<sup>-1</sup> is not considered a risk.<sup>132</sup> It should be an animal component-free product, wherein all components can be recorded through a certificate of origin, segregated from non-animal sources, and produced using non-animal starting and processing materials, such as those free from TSE (transmissible spongiform encephalopathy) and BSE (bovine spongiform encephalopathy). Carrageenan conforms to the definition and specifications set forth by JECFA (FAO/WHO), FDA 21 CFR 172.620, and European Commission Regulation 231/2012/EC.<sup>133</sup> However, the manufacturer assures that the product complies with local regulations, particularly in the country where it is to be consumed. It should be free from non-GMO (non-genetically modified organisms), allergen-free, and certified by HALAL and KOSHER certificates, which should be obtained through audits. Carrageenan should be produced by a GMP/HACCP certified company that strictly complies with the specific criteria implemented by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) and Commission Directive 2009/10/EC regulations. It should be packed in multi-walled paper or kraft bags with a heat-sealed polyethylene liner. Carrageenan does not contain preservatives or any artificial additives, colour or ingredients.<sup>134</sup>

The degradation of kappa carrageenan in food packaging influences the toxicity levels, mainly through the breakdown of smaller oligosaccharides, which can exhibit various biological effects. Still, carrageenan was found to be safe for consumption when used as a packaging material for food products. The cytotoxic effects of degradation can induce cytotoxicity and apoptosis in human cells, particularly in intestinal and liver cells, potentially reducing the IC<sub>50</sub> values and resulting in reduced cell viability. Table 4 shows the toxicity of carrageenan in different categories of biodegradable films. The cytotoxicity associated with apoptosis, evidenced by chromatin condensation and nuclear fragmentation observed in treated cells, can lead to hormonal imbalance. In comparison, undegraded forms do not have any cytotoxicity effects related to degradation. The transition from a non-toxic one to a potentially harmful form underscores the importance of monitoring carrageenan degradation in food applications. The primary consideration should be associated with the degradation of carrageenan under storage and processing conditions, which would directly influence the toxicity levels in the foods. Despite the inflammatory properties of carrageenan, its use in food products should be carefully considered to mitigate health risks associated with the degradation of carrageenan.<sup>135</sup>

## 12. Future perspectives in carrageenan films for packaging

Carrageenan applications in packaging could be further improved by advancing existing methods (combined extraction,





Table 4 The toxicity of carrageenan films in biodegradable films

Type of film	Dose level	Lethal rate	Effect on the film	Toxicity manifestation	Mechanism	Reference
Food-grade carrageenan	Up to 1000 mg L <sup>-1</sup> (in infant formula)	No lethality	Film biocompatible; non-toxic	No genotoxic or carcinogenic effects	High molecular weight, not absorbed intact	132
Semi-refined kappa carrageenan	Typical use levels in edible films	No acute toxicity in animal studies	Effective as an antimicrobial coating, safe on food	No adverse immune or GI effects reported	Essential oils disrupt microbial membranes	27
Kappa carrageenan hydrogel	Injection in biomedical studies ( <i>in vitro</i> )	No lethality at therapeutic doses	Biocompatible hydrogel; antibacterial	No cytotoxicity in the tested cell lines	Binding to cell surfaces disrupts the bacterial wall	136
Degraded carrageenan products	High doses <i>in vitro</i>	Potential cytotoxicity at high doses	Induced apoptosis; cell viability reduction	Cytotoxic effects in intestinal and liver cells	Degraded oligosaccharides cause inflammation	135
Refined carrageenan (food grade)	Oral exposure in Guinea pigs	No tumorigenic or oncogenic effects	No immune system toxicity	Mild GI mucosal irritation at high doses	Not absorbed intact; the immunogenic response is minimal	131
Composite films (carrageenan + ZnO)	Film coating on meat surfaces	No acute lethality observed	Prolonged antimicrobial action	No permeation to systemic toxicity	ROS scavenging; bacterial membrane disruption	71
Kappa carrageenan-based film	Typical food packaging levels	No toxicity observed	Sustained antimicrobial/antioxidant activity	No intestinal mucosa damage	Physical barrier and bioactive incorporation	13
Semi-refined kappa carrageenan	1.5% w/w in 0.2% KCl solution	No acute toxicity observed	An effective edible coating reduces <i>L. monocytogenes</i> undetectably	No cytotoxicity; antimicrobial action	Essential oils disrupt cell membranes	38
Carrageenan + orange EO	5% EO, 5% glycerol	No toxicity detected	Retains 95% aroma and color in dry fruit packaging	Safe for food contact	Essential oil bioactive antimicrobial	37
Kappa carrageenan + zinc oxide + anthocyanin	Not specified	No lethality observed	Antioxidant 99%; UV protection 85.2%; reduces microbial load	No systemic toxicity detected	Reactive oxygen species scavenging	71
Carrageenan + spermidine carbon dots	Not specified	Non-toxic	Sustained antimicrobial effect; low microbial contamination	No toxicity; biocompatible	Electrostatic disruption of microbial cells	59
Kappa carrageenan + grapefruit EO	Concentration varies 0.5–1% EO	No toxicity	Tensile strength increased from 65.20 ± 4.71 to 98.21 ± 6.35 MPa	Non-toxic; improved film mechanical strength	Cross-linked polymer network	76
Gelatin + modified kappa carrageenan	3% gelatin; modified kappa CGN	No toxicity observed	Film stable with a good barrier and antioxidant retention	No toxicity; antioxidant effect on packed fruits	Antioxidant encapsulation	65

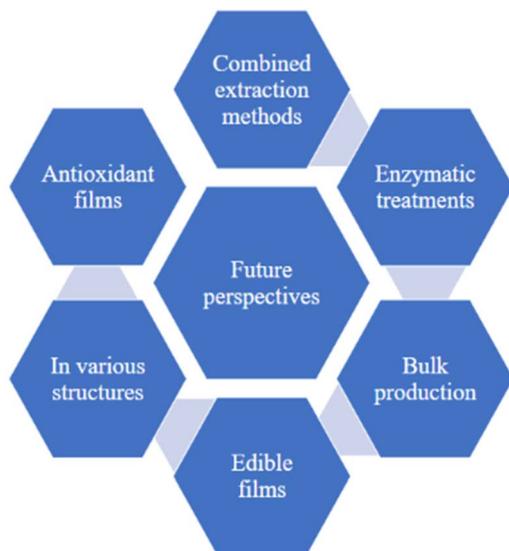


Fig. 7 Future perspectives of eco-friendly carrageenan-based packaging in commercialization.

pre-treatment before extraction, and other forms of carrageenan structural effects) and developing further functional packaging (edible, antioxidant, and active films), as discussed further below. There are possibilities for developing a multi-product extraction method, unlike the currently followed traditional method of single-product extraction. That is, along with carrageenan, some other application-related products could also be extracted.<sup>109</sup> The extraction of carrageenan from cellulose will open up new opportunities due to changes in its properties and potential for future film applications.<sup>109</sup> The composite film made with Guar gum or locust bean gum has shown better packaging properties and has the potential to be used in edible film applications in the future.<sup>84</sup> The bulk production of the developed biofilm made with *C. winterianus* essential oil and kappa carrageenan needs to be evaluated in the future, and the biodegradability of the film under various conditions should be assessed.<sup>117</sup> The developed kappa carrageenan and nano-bio-composite film with incorporated *Zataria multiflora* essential oil will be evaluated for different packaging materials.<sup>6</sup> Furthermore, the challenges will be overcome through cost-effective extraction, film interaction with a wide range of products, printability, storage stability, mimic or better properties than existing plastics, and year-round raw material availability, and other structural configurations of carrageenan improvements will be analysed, and the existing carrageenan film will be utilized in wider packaging applications which are given in Fig. 7.

### 13. Government encouragements of seaweed cultivation and processing

India is progressively establishing a comprehensive policy and governance ecosystem for seaweed cultivation by bringing it under the Department of Fisheries, while assigning export

facilitation to the Marine Products Export Development Authority (MPEDA). Dedicated committees for seed imports, biosecurity, and value chain coordination are strengthening regulatory oversight from cultivation to export. Supportive measures such as inclusion of seaweed under Priority Sector Lending, development of product and safety standards through the Food Safety and Standards Authority of India (FSSAI) and Central Drugs Standard Control Organisation (CDSCO), and improved market access through e-NAM and agriculture mandis are enhancing commercialization prospects. Ease of doing business is further promoted through geo-tagged cultivation databases, dynamic data portals, and simplified licensing frameworks. Investment incentives through Foreign Direct Investment (FDI) and Public-Private Partnerships (PPP) are being prioritized to develop processing units, cold-chain logistics, and supply-chain infrastructure in coastal and island regions. Parallely, skill development initiatives, including certificate and diploma programs are aimed at creating a trained workforce and sustainable coastal livelihoods. Research and innovation are being encouraged in high-value applications such as seaweed-based bioethanol, animal feed, pharmaceuticals, nutraceuticals, and biopolymers, alongside the development of frameworks to assess and monetize blue carbon credits from seaweed cultivation, thereby aligning economic growth with climate action goals.<sup>137</sup>

Social security and institutional strengthening form the backbone of inclusive seaweed sector growth. Comprehensive insurance coverage for crops, infrastructure, and farmers, along with inclusion under PMFBY, PM-KISAN, and Kisan Credit Card schemes, can significantly reduce financial risk. Mobilization of farmers through Self-Help Groups (SHGs), Farmer Producer Organizations (FPOs), and Joint Liability Groups (JLGs) will improve access to credit and markets, while cluster-level processing facilities, district-level marketing hubs, and Centres of Excellence for Seaweed will support seed supply, capacity building, processing, and R&D. Regions such as Lakshadweep, where *Gracilaria edulis* cultivation is gaining momentum, highlight the importance of species-specific and region-tailored strategies, as different seaweed species vary widely in yield, cultivation cycle, and market value.

### 14. Conclusions

Carrageenan, a natural polysaccharide extracted from red seaweed, has emerged as a highly promising biopolymer for sustainable food packaging applications due to its biodegradability, biocompatibility, film-forming ability, and cost-effectiveness. This review highlights its functional roles in edible coatings, active packaging, smart packaging integrated with sensors, and composite and injectable packaging forms. Despite carrageenan's hydrophilic nature, which limits its water vapour barrier properties, innovative approaches such as blending with other hydrocolloids, incorporating nanoparticles, and chemical modifications have significantly enhanced the mechanical, thermal, barrier, antioxidant, and antimicrobial properties of carrageenan-based films. These improved films effectively prolong food shelf life by reducing microbial growth,



controlling moisture and gas permeability, and maintaining the freshness of fresh produce. Moreover, monitoring can be achieved through sensors such as intelligent colorimetric sensors that are responsive to pH and volatile compounds. This potential extends beyond food packaging into pharmaceuticals, cosmetics, and biomedical applications, benefiting from its gel-forming and bio-adhesive properties, which support sustainable release systems and enhanced therapeutic efficacy. Safety evaluations consistently demonstrate that food-grade carrageenan is non-toxic, non-carcinogenic, and generally recognized as safe (GRAS) by regulatory authorities. However, attention must be paid to its degraded forms, which may exhibit cytotoxic effects under certain conditions. Challenges persist in commercialization related to sustainable extraction processes, ensuring the safety, improving mechanical strength under variable storage conditions, and addressing consumer awareness and regulatory frameworks. However, ongoing research on multi-product extraction, advanced composite films, and scalable fabrication techniques, coupled with government support for seaweed cultivation, promises to overcome these barriers. Overall, carrageenan represents a sustainable, multifunctional biopolymer with substantial potential to replace petroleum-based plastics, reduce environmental impact, and meet the rising demand for eco-friendly packaging solutions, thereby positioning it as a next-generation material for intelligent active food packaging with broad commercial and societal benefits.

## Author contributions

Devanampriyan Rajan: conceptualization; data curation; formal analysis; methodology; validation; writing – original draft; writing – review & editing. Kalaivani Sundaravadivelu, Sankari Rajan and Vigneshwaran V.: data curation; formal analysis; methodology; validation; writing – original draft. Chitra Devi Venkatachalam: supervision; validation; visualization; writing – review & editing. Rakesh Kumar Gupta: supervision; conceptualization; resources; writing – review & editing; validation; visualization; data curation.

## Conflicts of interest

The authors declare no conflict of interest.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

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