Review

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Polysaccharide-Based Bioplastics as Green and Sustainable Packaging Materials

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Review

Polysaccharide-Based Bioplastics as Green and Sustainable Packaging Materials

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Abstract: Over the past few decades, synthetic petroleum-based packaging materials has increased, and the production of plastics has surpassed all other man-made materials due to their versatility. However, the excessive usage of synthetic packaging materials has led to severe environmental and health-related issues due to their non biodegradability and their accumulation in the environment. Therefore, bio-based packages are considered alternatives to substitute synthetic petroleum-based packaging material on the market. Furthermore, the choice of packing material in the food industry is a perplexing process as it depends on various factors, such as the type of food product, its sustainability, and environmental conditions. Interestingly, due to proven mechanical, gas and water vapour barrier properties, and biological activity, polysaccharide-based bioplastics show the potential to expand the trends in food packaging, including edible films or coatings and intelligent and active food packaging. This review discusses the structure, properties and recent developments in polysaccharide-based bioplastics as a green and sustainable food packaging materials.

Keywords: active packaging; alginates; bioplastics; carrageenan; cellulose; chitin; chitosan; gellan gum; intelligent packaging; polysaccharides; starch

1. Introduction

In recent years, synthetic polymer production has overwhelmingly grown worldwide, and a world without plastics seems unimaginable today. Although the first synthetic plastic, such as "Bakelite," appeared in the early 20th century, plastics' widespread use did not occur until World War II. The production of plastics has overtaken all other synthetic materials due to their versatility [1]. In

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2018, world plastics production totaled around 359 million metric tons, predicted to triple by 2050. This further exacerbates the inherent conflict between plastic's myriad uses and environmental impact [2]. Plastics have replaced metal and glass in massive volumes in modern-day packaging, and a significant fraction of global plastic production is utilized in packaging applications due to their flexibility, barrier properties, low cost, and ease in production [3–5].

Fossil fuel-based packaging waste contributes to a major part of municipal solid waste. The leaching of harmful chemicals, non-biodegradable nature, recycling issues, and the environmental impact caused by the excessive production and usage of synthetic plastics have led to a dire need to develop green materials [5,6]. Development and commercialization of bioplastics from renewable material can reduce the widespread dependence on fossil fuels, subsequently addressing environmental pollution. Moreover, synthesizing bioplastics from renewable sources is economical, sustainable, and environmentally benign [6]. Current packaging tendencies include interacting with the environment and self-preservation of food. Moreover, the packaging can improve food quality and shelf-life by integrating various functionalities in the packaging structure. Numerous studies are underway to incorporate nanotechnology into active packaging technologies to address the issues related to barrier properties of the packaging materials [6,7]. Furthermore, biopolymers have been widely investigated for preparing packaging materials [8].

Among many biopolymers, polysaccharide-based materials have recently gained attraction for preparing bioplastics owing to their renewability, relative abundance, nontoxicity, surface functionality, and biodegradability. Furthermore, bioplastics could help overcome the high dependence on fossil resources while reducing the carbon footprint compared to conventional plastic packaging materials. This review discusses structure and properties of polysaccharides derived from various sources, including plant, animal, and microorganisms and their food packaging applications, recent developments in polysaccharide-based bioplastics in food packaging and health impacts of using polysaccharides as bioplastic packaging.

2. Biopolymers

Biopolymers including polysaccharides, have been employed for preparing bioplastics due to their relative abundance, renewability, biodegradability, biocompatibility, non-toxicity, ease of handling, low-cost, and functional tunability [9,10]. Biopolymers can be categorized based on their fabrication methods and source of origin, and the classification of biopolymers is as follows [11,12].

1. Biopolymers extracted from biomass;
    a. Plant: starch (amylose/ amylopectin), cellulose, guar gum, pectin, protein including corn zein, gluten, and soy protein, lipids
    b. Animal: chitin/chitosan, protein including, whey protein, casein, collagen, and gelatin, lipids.
    c. Algae/seaweeds: alginate, agar, carrageenan, ulvan
2. Biopolymers produced by microorganisms: polysaccharides (dextran, gellan gum, pullulan, xanthan gum), proteins (polyamides from bacteria), polyhydroxyalkanoates (PHA), polyhydroxybutyrates (PHB).
3. Biopolymers chemically synthesized from bio-based materials: Polylactic acid (PLA)
4. Biopolymers chemically synthesized from petroleum-based materials: polycaprolactones (PCL) and polyesteramides (PEA).

Figure 1 depicts the classification of polysaccharides based on their origin.

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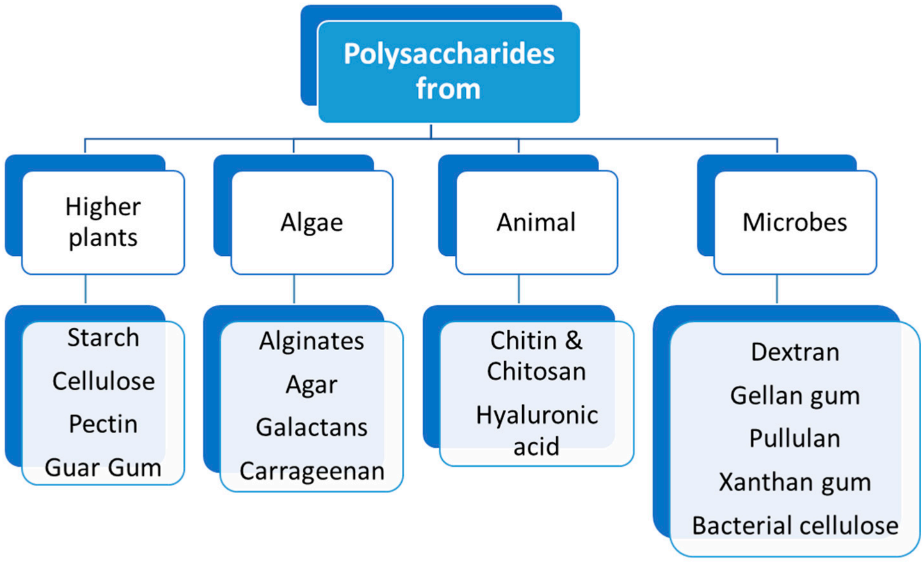

Figure 1. Classification of polysaccharides based on their origin.

3. Polysaccharide-based bioplastics

Polysaccharides are one of the most abundant natural biopolymers [10]. Therefore, polysaccharide-based bioplastics derived from biomass have been widely investigated over the last few years [13,14]. Polysaccharides, such as cellulose and starch derived from agricultural materials, chitin and chitosan derived from marine food processing wastes, pullulan from microorganisms, and many other polysaccharides from various natural sources have shown the ability to form bioplastics [15]. Moreover, bioplastics generated from the graft copolymerization of synthetic monomers or biomolecules have been studied for preparing bioplastics [16]. Certain additives, including plasticizers, antioxidants, and antimicrobial agents, have also been incorporated during bioplastic synthesis to enhance their functional properties [17–19].

Apart from biodegradability, the physicochemical and mechanical properties (flexibility, brittleness, and rigidity) of the biopolymers should be comparable to conventional plastics [11]. Utilization of polysaccharide is limited by their poor mechanical and barrier properties [9]. Generally, physicochemical, mechanical, and barrier properties of polysaccharides vary from their source of origin and other embodied constituent. Hence, developing packaging materials from those polysaccharides must be executed after carefully investigating their characteristics for intended applications.

4. Polysaccharides from higher plants

4.1. Starch

Starch is a naturally occurring polysaccharide found in abundance and exists as a heteropolymer in nature. Starch and its derivatives have been widely used to produce bioplastics [20]. Starch is comprised of two types of polymer chains: amylose and amylopectin. Amylose is the linear form with α-1,4-glycosidic linkage, whereas amylopectin possesses a branched structure with α-1,4- and α-1,6-glycosidic linkages [21]. Amylose (amorphous) and amylopectin (crystalline) are arranged in a semicrystalline structure in starch granule. Due to the semi-crystalline nature, starch granules are not soluble in water. Starch granules undergo gelatinization process during heating in the presence of excess water. This phenomenon occurs via hydration of amorphous region, loss of crystallinity and starch granule structure, unwinding of double helices amylopectin, and breakdown of hydrogen bonds between starch chains during the heating [21,22].

The ratio of amylose to amylopectin and branching degree of amylopectin determines the size and structure of starch granule as well as influences the physicochemical properties (crystalline

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structure, swelling capacity, gelatinization, paste properties, and retrogradation) of starch [21,22]. Direct applications of native starch is limited in industrial applications and native starch are modified using physical (hydrothermal methods, griding, and extrusion), chemical (etherification, esterification, crosslinking), and enzymatic hydrolysis methods [21].

Starch based polymers for packaging are mostly derived from maize, sugar cane, corn, and potato. However, corn is the most operative in the industry among those starch types [23]. Starch is employed in food and non-food applications, including cosmetics and pharmaceutical industries. Besides, it is used to obtain glucose, dextrin, ethyl alcohol, biofuel, and as stiffeners and binders [23].

High amylose content in starch contributes to high strength of film, whereas the presence of highly branched amylopectin (waxy starch) leads to films with poor mechanical properties. The Young's modulus and the tensile strength of the bioplastic films made of cassava increased with increasing amylose content due to the well-stacked polymer network [24]. The mechanical properties of starch-derived bioplastics can be improved by adding plasticizers, such as sorbitol and glycerol [25]. The type and content of the plasticizer significantly affects the water sorption and water vapour permeability (WVP) of starch-derived bioplastics films. For instance, the WVP increases with increasing plasticizer content [26].

Bioplastics developed from starch have many advantages, such as higher biodegradability, renewability, and good oxygen barrier properties, making them suitable alternatives for many commercial applications, including packaging [27]. However, starch possesses several limitations, including high water absorption and poor mechanical properties [23].

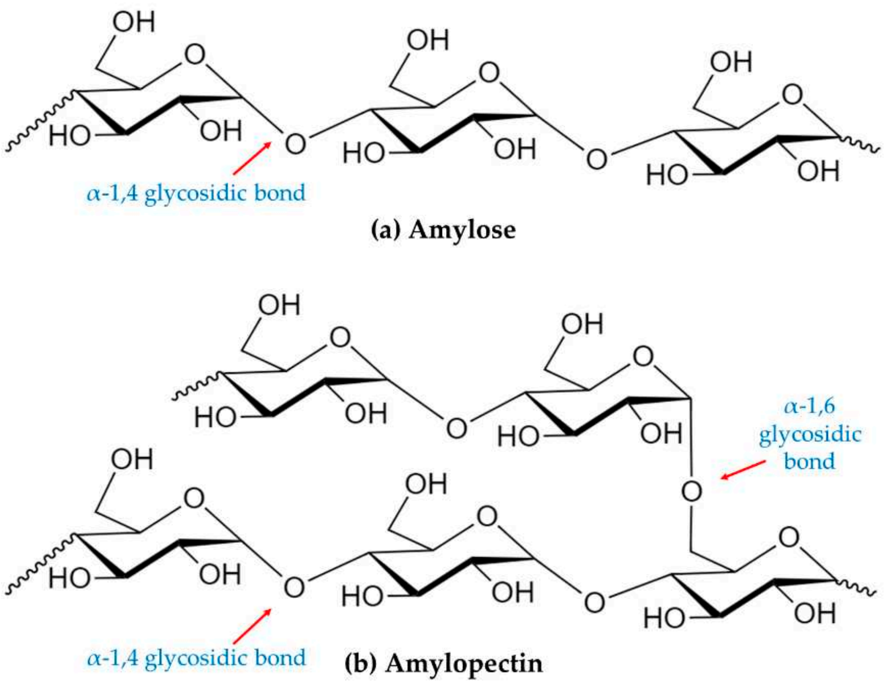

Figure 1. Structure of the amylose and amylopectin.

4.1.1. Thermoplastic starch

Thermoplastic starch (TPS) is TPS obtained by the spontaneous destruction of the starch crystalline structure in the presence of heat, mechanical shear, and plasticizers. This phenomenon is called gelatinization. Plasticizers used in TPS processing are water and glycerol, alcohol, polyols, sugars, urea, and acetamide [28–31].

The most commonly used techniques for bioplastic preparation are casting and thermo-molding. However, extrusion and injection molding are widely accepted processing techniques for generating TPS. Extrusion technology has been widely used to process polymers [32], including TPS, which enable molecular weight reduction under high pressure, temperature, and shear stress [28]. Then TPS can be mixed with other ingredients, such as polylactic acid (PLA) and polycaprolactone (PCL), to fabricate the bioplastic film. [28–30,33,34].

TPS is an amorphous material that can flow like synthetic polymers, thus can be suitable for conventional molding and extrusion technologies [28]. TPS has poor processability and mechanical properties. TPS has moisture sensitivity and low temperature resistance. Furthermore, mechanical properties of TPS vary with the storage time due to the loss of plasticizer. Thus TPS can be blended with various synthetic and natural polymers to improve their properties [35]. TPS blend were prepared using various polymers, such as polyethylene (PE) including low-density PE (LDPE)/ linear low-density PE (LLDPE) [36,37], high density PE (HDPE) [38], polypropylene (PP) [38,39], polystyrene (PS) [35,40], polyvinyl alcohol (PVA) [41], etc.

High compatibility, crystallinity, tensile strength, hardness, and stiffness, are important factors in food packaging. Furthermore, for a packaging material, elasticity or elongation is very important as a material with high elongation absorbs a large amount of energy before breaking. St-Pierre *et al*. [36] reported that TPS/PE (LDPE or LLDPE) blend has high elongation at break even without any interfacial modifier.

TPS using various plasticizers have been widely studied. Of which, glycerol has been widely studied as plasticizer for starch due to its high boiling point, availability, and low cost [35]. Schlemmer *et al*. [40] used a solvent casting technique to prepare TPS/PS blends with glycerol and buriti oil as plasticizers, while Mihai *et al*. [35] studied the fabrication of extruded foams from TPS/PS blends with glycerol as a plasticizer. Yoon *et al*. [41] reported the effect of functional groups (hydroxyl and carboxyl groups) of plasticizers on the properties of starch/PVA blends with different plasticizers, namely, glycerol, malic acid, tartaric acid, and succinic acid. Degree of swelling and solubility were found to be higher in film with malic acid and tartaric acid due to their higher hydroxyl and carboxyl groups, i.e., higher hydrophilicity. Another study reported the role of a complex plasticizer prepared from a mixture of urea and glycerol on the starch/PVA blends [42]. The result showed that the complex plasticizer formed stronger and more stable hydrogen bonds with water and starch/PVA molecules than the single plasticizer. Therefore, the starch blends with complex plasticizers displayed better mechanical characteristics. Another study compared the morphology of TPS/LDPE blend prepared by one-step extrusion with the reprocessed TPS/LDPE blend [43]. Results indicated that one-step processing of the used behave as typical thermoplastic immiscible blends and could achieve highly elongated morphological properties [43].

4.1.2. Modified starch

The industry already produces chemically modified starch derivatives by the surface modification of starch granules [44,45]. Modified starch is prepared by physically (gelatinization, extrusion, foaming, and impregnation), chemically (esterification and grafting), enzymatically, or biotechnically treating native starch to change its properties [44]. Modified starches are commonly used as emulsifiers and stabilizers, viscosifying agents, coatings, and thickeners [45,46]. A study reported that starch chemical modification produces a biodegradable material with appropriate mechanical strength, flexibility, and water barrier properties to be used as packaging material [45,47]. Moreover, hydrophobically modified starch through acetylation, esterification, and grafting of highly reactive hydrophobic functional groups improves the hydrophobicity of starch by replacing the hydroxyl groups in starch, which leads to an enhanced interfacial compatibility between starch and the hydrophobic polymer [48,49].

Modification improves the processability by reducing the gelatinization temperature or hot paste viscosity. The modification also improves film formation and emulsification [50]. Moreover, proteins in starch form a network and assist the modified starch in enhancing plasticity and elasticity [44–47].

In a study, polybutylene succinate (PBS) was blended with five types (A, B, C, D, E) of modified tapioca starch and tested for food packaging applications [46]. Five modified starch grades (A, B, C, D, E) showed various properties: moisture content of 11.1, 8.1, 7.2, 8.6, and 11.2%; bulk density of 0.63, 0.62, 0.59, 0.53, and 0.54 g/cm^3; gelatinization (T_g) temperature of 51, 45.2, 44.9, 60.3, and 69.4 °C;

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maximum Brabender viscosity of BU 1291, 228, 405, 75, and 0 717; viscosity of 5.5; 6.5; 6.3; 5.7; and 6.1 CP, respectively. According to the observed results, starch A and B blends has good elongation at break and blending capability makes them to be used as food wrap and food container materials, while starch D blend as grocery plastic bags because of its good tensile property [46].

4.2. Cellulose

Cellulose, the most abundant macromolecule on earth. Cellulose can be extracted from plants, especially from vascular plants, algae (*Cladophora* and *Valonia* spp.), bacteria (*Gluconacetobacter* and *Sarcina* spp.), fungi, some protozoa (*Dictyostelium amoebae*), and agricultural residues [51,52]. The plant cell wall is the major source of cellulose, and consists of cellulose, hemicellulose, and lignin, at the ratio of 4:3:3, which differs from the source. Besides the three components, natural lignocellulosic materials include a small amount of pectin, nitrogenous compounds, and ash [51,53]. Cellulose is a linear homopolymer composed of glucose units linked by β-(1–4)-glycosidic bonds. The hydroxyl groups in cellulose form strong intramolecular and intermolecular hydrogen bonds, which enables cellulose to form a stable three-dimensional crystalline structure. Cellulose is surrounded by hemicellulose and lignin [53–55]. Thus, cellulose extraction requires a pretreatment process to remove lignin, pectin and other non-cellulosic materials [52,55].

Cellulose and its derivatives have been tested for packaging applications [56]. Cellulose composites showed excellent mechanical properties, reinforcing capabilities, biodegradability, and availability. For instance, Carrillo *et al*. prepared the cellulose lyocell fibre/cellulose acetate butyrate composite [57]. They displayed increased tensile properties, dimensional stability, fibre and matrix compatibility, and biodegradability. Carboxymethyl cellulose (CMC) based films fabricated by incorporating bioactive Chinese chives root extract (CRE) showed higher oil resistance property in addition to the improved physical and barrier properties, antioxidant and antimicrobial activity (against *B. cereus*, *S. aureus*, *E. coli*, and *S. typhimurium*), which is desirable for packaging of oil products [58]. Peptidopolysaccharide developed using 2,3-dialdehyde cellulose and antimicrobial nisin peptide showed improved mechanical property, lower water holding capacity, and excellent antimicrobial activity against *S. aureus* and *E. coli*. This active film also showed extended shelf life of fresh pork meat stored at 4°C for 6 days [59]. In another study, antimicrobial packaging film was prepared using cellulose acetate butyrate/organically modified montmorillonite (OMMT) incorporated with carvacrol and cinnamaldehyde [60].

4.2.1. Nanocellulose

The potential of cellulose nanoparticles or nanocellulose has been used for the preparation of nanomaterial with various functions. Cellulose NPs have a strong tendency for self-association due to the omnipresence of surface hydroxyl groups. These inter-particle interactions can cause aggregation during the preparation of the nanocomposite [61].

Because of the excellent dispersion level of cellulose NPs in water, cellulose NPs can be mixed in both water-soluble polymer and aqueous polymer dispersion (latex). Then the solid nanocomposite film can be obtained by simple casting and water evaporation [61].

Nanocellulose is typically divided into groups based on their preparation techniques: cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs), and bacterial cellulose (BC) [62,63]. CNCs can be produced by acid hydrolysis, and hydrochloric and sulfuric acid is widely used. Other acids, including sulfuric, phosphoric, hydrobromic, and phosphotungstic acids are also used for CNCs preparation [62,64–66]. Other than the chemical methods, enzymatic methods also used to prepare CNCs [62]. CNFs are obtained by mechanical methods, including high-pressure homogenization, microfluidization, refining, or grinding [62]. However, the synthesis of CNFs often requires chemical pretreatment before mechanical disintegration processes.

The geometric dimensions and final properties of CNCs and CNFs directly depend on the cellulosic source, the preparation and processing conditions, and the possible post- or pretreatments. CNCs have a few nanometers diameter and lengths ranging from 10 to 500 nm, while CNFs have diameters of 3–50 nm and lengths of a few micrometers, and BC are in the range of 20–100 nm in diameter and micrometers lengths [67,68].

Nanocellulose and its composites, have been tested for packaging applications. WVP decreases when the cellulose fibres are disintegrated to the nanoscale level. The gas permeability of micro-fibrillated cellulose (MFC) is reduced in a dry atmosphere when decreasing the size of the cellulosic particles because of the dense structure with no porosity of the nanofilm. Improved gas barrier of at high humidity levels can be achieved by chemical modification of NPs or hybridization with other materials [69]. Chemical modification of the NPs improves the gas barrier properties of nanocellulose films at high relative humidity levels [61]. Further, nanocomposite films can serve as carriers for active substances such as antioxidants and antimicrobials. Thus, can improve food quality and extend food shelf-life [61,70].

Cellulose based nanocomposites widely studied for biodegradable and antimicrobial food packaging material [71–73]. Carboxymethyl cellulose (CMC)-based nanocomposite containing Ag, ZnO, and CuO NPs prepared using solution casting method, showed excellent mechanical, water vapour barrier, and antimicrobial properties [71]. Furthermore, guar gum/CMC-based film incorporated with halloysite-nanotubes (HNT) and litchi shell extract (LSE) showed improved elongation at break, UV barrier property, and antioxidant activity, and improved the oxidation stability in roasted peanut for 8 days. Thus, this guar gum/CMC/HNT/LSE film can be used as a packaging material for low water activity and oxygen sensitive food products [74].

5. Algal Polysaccharides

5.1. Alginates

Alginates are naturally occurring water-soluble polysaccharides derived from brown seaweeds (Phaeophyceae) and from bacterial sources. Even though there are many alginates sources, algae, including *Laminaria hyperborea*, *Laminaria digitata*, *Laminaria japonica*, *Ascophyllum nodosum*, *Macrocystis pyrifera*, and bacteria like *Azotobacter vinelandii*, and *Pseudomonas aeruginosa* are widely used species for industrial production [75].

Alginate is a linear biopolymer comprised of two distinct structural units linked by 1-4 glycosidic bonds, β-D-mannuronic acid (M) and α-L-guluronic acid (G) (Figure 3) [76]. The structural properties and composition of alginates may vary based on their source of origin, geography, and growth conditions [77] However, the M and G units content and their length of each blocks varies with different sources [78]. Alginates can be organized into two different segments; (a) homopolymeric G blocks [poly (G)] or homopolymeric M blocks [poly (M)], (b) heteropolymeric MG blocks randomly assigned into G and M sequence as alternating or short interchanging G and M units [76,79]. The composition and sequence of these M and G units in alginates chains govern the physicochemical properties and thermal processing [80].

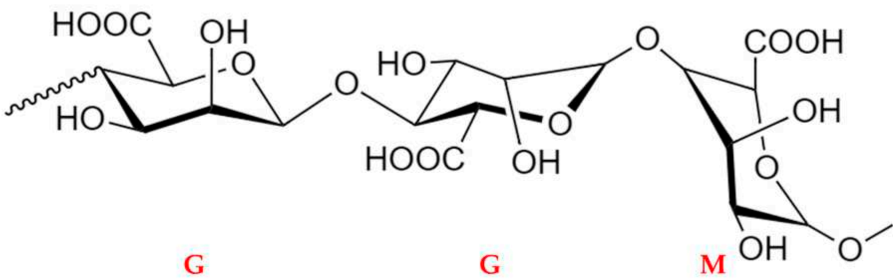

Figure 3. Structure of alginic acid.

The content or length of G and M blocks in alginates significantly affect its gelling properties. Alginates with less than 20-25% (molar fraction) of G-blocks is unable to produce gels, whereas G block in higher content produces brittle and strong gels, and alginates with high M blocks forms softer and more elastic gels [81].

The anionic nature of alginates (due to the presence of carboxyl group) makes stable hydrogels/gels with the presence of cations, including Na^+ and Ca^{2+} [80,82]. Alginates are extracted from their dried, grounded sources using alkaline treatments (e.g., sodium carbonate) followed by precipitation of sodium alginates salts using calcium chloride [80,83]. Alginates powders can be processed into alginates fibre through a wet spinning process [80].

Alginates can be molded into composites, hydrogels, and scaffold, and widely used in food, biomedical, cosmetics, textile, and other industrial applications due to its biocompatibility, low toxicity, low cost, high-stable gelling and thickening ability [80,84–86]. Alginates have been used in various application. including enzyme immobilization or carriers [87], drug delivery, wound dressing, tissue engineering scaffold, and cell culture [80,88–90]. Alginates fibre with cotton fibres can be used to fabricate woven or knitted eco-friendly flame-retardant textile fabrics [90]. Furthermore, alginates have been widely used in edible films formation (using solvent casting and extrusion) and coating (using dipping, spraying, and vacuum impregnation) for food packaging applications [91].

The addition of fillers, reduced graphene oxide (rGO) and mixer of ZnO-rGO into alginate matrix showed improved electrical conductivity and antimicrobial activity against *E. coli* e *S. aureus*, which makes the film suitable for extending the shelf life and food sterilization at low temperature (e.g., pulsed electric field and ohmic heating) [92]. Incorporation of sulphur NPs (S NPs) at 2% into calcium cross-linked alginates films improved the tensile, water vapor and UV barrier properties, hydrophobicity, and bactericidal activity against *Listeria monocytogenes* [93]. Alginates/S NPs composite film can be used as packaging material for frozen foods with high moisture content which are susceptible to contamination of *Listeria monocytogenes* (e.g., meat products) [93]. Various other active food packaging material have been developed by incorporating metal and metal oxide (TiO_2, ZnO) and essential oil or plant extract, such as cumin essential oil, Alovera, oregano essential oil, cottonseed protein hydrolysates, which have the proven potential application in packaging of fish, meat and fatty foods (Table 1) [94–97].

5.2. Carrageenan

Carrageenan (CG) is a hydrophilic linear polysaccharide extracted from red seaweeds (Rhodophyceae) [98]. CG is a sulfated polygalactan with 15-40% ester-sulfate content, forming α-1,3 and β-1,4-glycosidic linkages through alternate units of D-galactose and 3,6-anhydrous-galactose (3,6-AG) (Figure 4) [99,100]. CG solutions exhibit gel-forming and viscosifying characteristics. Based on the structural significance and solubility in potassium chloride, CG can be classified into different forms, such as λ, κ, ι, ε, and μ. All these structural confirmations contain sulfate groups between 22 to 35%. The aforementioned structures are not definitive of chemical structures but signify compositional differences (3,6-AG content) and degree of sulfation at specific locations [99,101,102]. Higher levels of ester sulfate resulted in lower solubility temperature and lower gel strength [99,102]. As the free acid is unstable, commercial grades exist as stable sodium, potassium, and calcium salts or as a mixture. The physical and rheological properties of the CG are influenced by cations and conformation of the sugar units in CG polymer chain [99,103].

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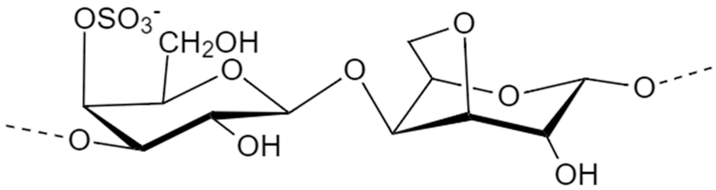
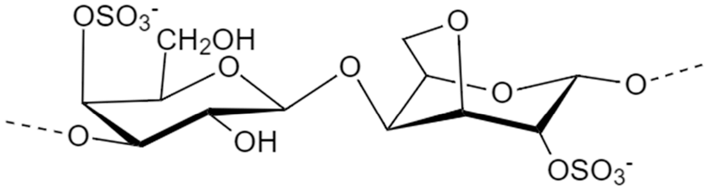
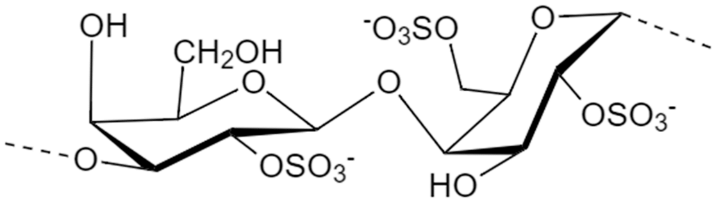


(a) κ-carrageenan



(b) ι-carrageenan



(c) λ-carrageenan

Figure 4. Chemical structure of carrageenan: (a) κ, (b) iota, (c) lambda.

CG forms thermoreversible gel via ion-induced coil-helix conformational transition upon cooling, i.e., randomly arranged CG coils (at higher temperatures, i.e., >50 °C) undergoes a conformational transition into ordered double-helical structures, which aggregate to form three-dimensional structure at low temperatures and/or in the presence of the cations [104–106]. Furthermore, the concentration and the nature of cations influence the gelling properties of CG, particularly on iota and kappa-CG. Kappa-CG in the presence of KCl forms stronger gels than those with NaCl, LiCl, $MgCl_2$, $CaCl_2$, and $SrCl_2$ [102].

CG has been used for many pharmaceuticals and biomedical applications due to its antiviral, anti-tumour, immunomodulatory, and anticoagulant, and antithrombotic activities. CG is frequently used in the food industry due to its gelling, thickening, and emulsifying properties [99,101]. CG blends, reinforcements, composites, and multilayering have been extensively researched to design complex and active packaging materials [100,107,108]. Nanocellulose reinforced CG exhibited substantial enhancement of mechanical and water vapour barrier properties [109]. The plasticized semi-refined kappa-CG was reported as potential edible food packaging material with improved barrier and mechanical properties [110].

CG has relatively reduced mechanical and water vapour barrier properties. To improve these properties, CG can be blended with natural polymers, like starch, cellulose, chitin, chitosan, and alginate [102]. Kappa-CG/alginate blend exhibited excellent film-forming ability due to the improved tensile and moisture barrier properties of kappa-CG and the improved elongation and transparency of alginate [111].

6. Polysaccharides from animal origin

6.1. Chitosan

Chitin (Poly [β-(1-4)-2-acetoamido-2-deoxy-D-glucopyranose]), a polymer of N-acetyl-D-glucosamine, is widely distributed in nature, especially in higher fungi, algae, protozoa, arthropods, nematodes, and molluscs. Its derivative, chitosan (poly[β-(1-4)-2-amino-2-deoxy-D-glucopyranose), a polymer of D-glucosamine, has reactive amino groups. Chitosan is a heteropolymer made of D-glucosamine and a small fraction of N-acetyl-D-glucosamine residues [112–114]. Chitosan adsorption ability is higher than chitin, which has relatively fewer amino groups [112]. Figure 5 shows the chemical structures of chitin and chitosan.

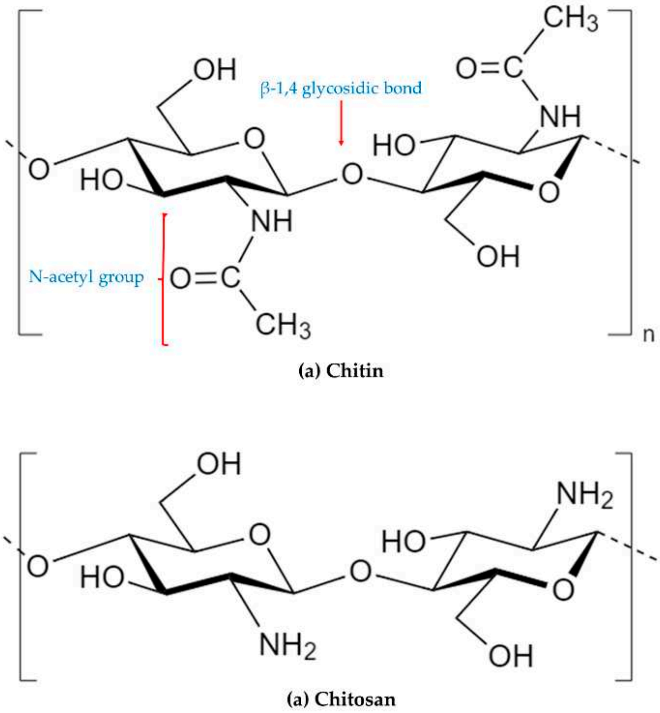


Figure 2. Chemical structure of (a) chitin and (b) chitosan.

The advantages of chitosan in food packaging applications include: biodegradability, biocompatibility, non-toxicity, bio-adhesive, edibility (coating for fruits and vegetables), and bacteria and fungi-static properties. Chitosan films have a selective permeability to gases, including O_2 and CO_2 and good mechanical properties [9,115,116]. Chitosan has a potential packaging polymer, particularly as an edible packaging or coating due to its antimicrobial activity against yeast, bacteria and fungi, thus can be used to extend the shelf life of foodstuffs [116,117]. Moreover, chitosan has good mechanical property incomparison to other natural polymers, such as starch and gluten [116]. However, high sensitivity to moisture conditions limits the application of chitosan in packaging.

It was also reported that the addition of curcumin (0.5%) increased the mechanical, hydrophobicity, and oxygen permeability of the chitosan/bacterial cellulose composite active film as well as showed excellent antioxidant activity. Furthermore, this composite film is a promising food packaging material as it exhibited excellent preservation of strawberries compared with ordinary PE film and significantly delayed the oxidation of edible oils [118].

In a study, chitosan-coated cassava starch films were prepared with 1-4 wt% chitosan solutions and coated with free starch films containing 2-6 wt% glycerol [119]. The results showed that the mechanical properties with chitosan coating concentration significantly increased the tensile stress at maximum load and tensile modulus and decreased elongation at break. Furthermore, concerning physical properties, a remarkable decrease in water uptake was observed due to the contribution of hydrophobicity of the chitosan coating layer. Generally, reducing wettability and WVP is preferable for packaging film applications. Studies have also shown a decrease in water susceptibility for chitosan-based membranes with beeswax and a reduction in WVP with oleic acid, neem-oil, cinnamon essential oil, among others [120]. Incorporation of ZnO NPs and neem essential oil improved the tensile strength, water vapour barrier properties, and antibacterial activity against *Escherichia coli* [121]. CS film incorporated with magnetic-silica nanocomposite (MNP/Si) and turmeric essential oil (TEO) showed antimicrobial activity against *Bacillus cereus* over 14 days of storage in packaged *Surimi* [122]. Active packaging film fabricated using CS/halloysite nanotubes (HNT)/*Citrus limetta* pomace extract (LPE) showed improved antioxidant activity, and can be used to improve the oxidative stability of lipids [123]. CS has fungistatic, bacteriostatic, antioxidant activity, and enzyme inhibition properties, which can be enhanced by incorporating NPs, essential oils, plant extracts, other biopolymers, antimicrobial and anti-browning agents. Due to these properties, chitosan based coatings can delay the postharvest decay and extend the shelf life of harvested fruits and vegetables without changing their nutritive and sensorial properties [124]. Various studies showed the application of chitosan composites in food packaging applications (Table 1).

6.2. Hyaluronic Acid

Hyaluronic acid (HA) (Figure 6) is a linear non-sulfated glycosaminoglycan, composed of repeating units of D-glucuronic acid and N-acetyl-D-glucosamine [125–127], linked through alternating β-1,4 and β-1,3 glycosidic bonds [128].

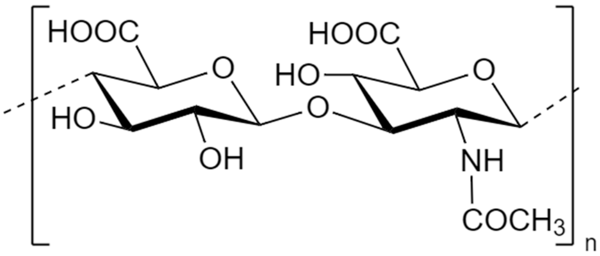


Figure 2. Chemical structures of hyaluronic acid.

HA is mainly found in the extracellular and pericellular matrix, but the occurrence in intracellular has also been reported [129]. HA can be separated from animal sources (such as rooster combs) and microbial sources, but in industrial context, HA is widely produced by microbial fermentation method in high purity and good yield. *Streptococcus* genera, including type C *Streptococcus* (*Streptococcus equisimilis*, *Streptococcus zooepidemicus*, *Streptococcus uberis*), type A *Streptococcus* (*Streptococcus pyogenes*, *Pasteurella multocida*), are widely used in the HA production [130–132]. Type C *Streptococcus* has mainly used in the HA production using microbial fermentation due to its less pathogenicity [132]. Resides of exotoxins and immunogens can found in HA produced from type C *Streptococcus*, which may limit its wider applications. To overcome this safety issue and the low HA synthesis efficiency, many studies focused on use of genetic engineered microorganisms, such as *Escherichia* coli, *Bacillus* spp., *Lactococcos lactis*, *Corynebacterium glutamicum*, *Agrobacterium* spp. [132–134].

HA has unique structural, rheological, physiological, and biological properties, such as higher water binding capacity and viscoelasticity as well as lack of immunogenicity and toxicity and received higher interest in cosmetic, biomedical, and food industrial applications [133–135].

HA is an excellent lubricant and shock absorber due to its unique higher water binding capacity and viscoelasticity [134]. HA is a highly hydrophilic polysaccharide and uses as a vital component in skincare products as a moisturizer [129]. In addition, biocompatibility, non-immunogenicity, unique viscoelasticity, make HA perfect for diverse biomedical applications, such as supplementation of joint fluid for arthritis, surgical aid in eye surgeries, facilitating wound healing, and drug delivery agent for various administration routes [135,136].

Surface modifications, crosslink formation, blends, and composites have been investigated to improve the mechanical and barrier functionalities of HA for advanced packaging applications [137–140]. For instance, recent research findings tested successful applications of HA films for diverse applications in edible films/ primary packaging [141,142] and active packaging [143,144]. Furthermore, studies have highlighted the effectiveness of using blends [145,146], composites [147], and crosslink formation [137,140] to improve desired properties of HA based packaging.

Coating of eggs with the composite of HA, 0.025% curcumin, and 0.025% cellulose nanofibre (CNF) extended the shelf life by 14 days, which has the potential application in egg preservation at 25 °C and 70% humidity [141]. The crosslinked HA/PVA bearing styrylpyridinium groups (PVA-SbQ) composites exhibited better thermal stability, mechanical, UV light barrier, and water vapor barrier properties and has the potential application as food packaging materials [130]. Antimicrobial edible food packaging was developed by encapsulated the angelica root (*Angelica sylvestris*) oil (ARO) into a Zein/HA/Gelatin based biofiber through emulsion electrospinning [148].–

7. Polysaccharides from microbial origin

7.1. Gellan Gum

Gellan gum (GG) (Figure 7) is a linear anionic extracellular natural polysaccharide produced by the aerobic fermentation of a gram-negative nonpathogenic bacterium, *Sphingomonas elodea* (formerly known as *Pseudomonas elodea*) [149–151]. The GG is a heteropolysaccharide consisting tetrasaccharides repeating units, 1,3-β-D-glucose, 1,4-β-D-glucuronic acid, 1,4-β-D-glucose, and 1,4-α-L-rhamnose, containing one carboxyl group. The 1,3-β-D-glucose unit contains two acyl substituents: L-glyceryl at C2 and acetyl at C6 position [151,152]. There can be non-polysaccharide constituents in GG composition, such as cell protein and ash; however, those can be removed by filtration or centrifugation [153].

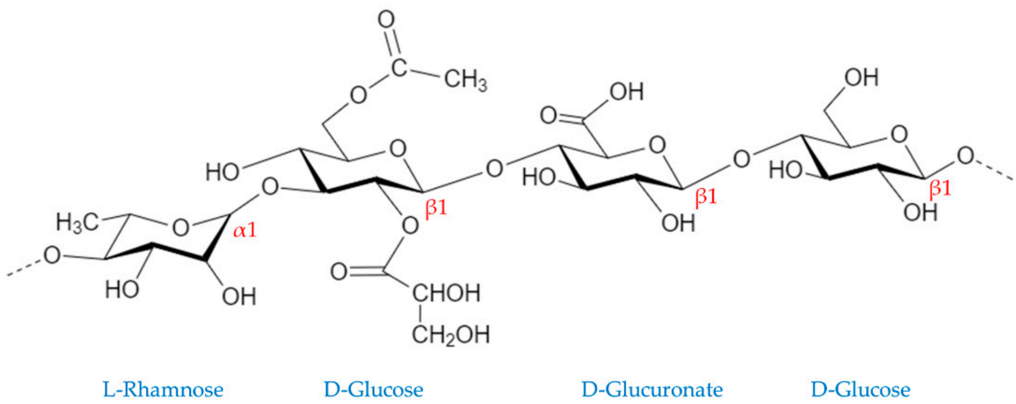


Figure 2. Chemical structures of gellan gum.

GG is a well-known food ingredient as a stabilizer, thickening agent and gelling agent and has wide applications in food packaging [120]. Furthermore, GG has been researched as a multifunctional additive for various pharmaceutical and biomedical applications as a source of regenerative medicine, stomatology, or gene transfer technology [154].

Commercial grade GG is a deacetylated form of native GG, which can be produced by alkaline treatment [150]. Commercially GG can be produces via controlled microbial fermentation process. The quality of commercial-grade GG may influence the process measures, as it is highly dependent on the metabolism and enzymic activity of the bacterium along with other vital parameters, such as process temperature, pH, stirring rate, oxygen transfer, and composition of the production medium [150].

Microbial exopolysaccharides, including GG, are water-soluble [153]. The type and the degree of acetylation or deacetylation presence in the polymeric backbone govern the mechanical and physicochemical properties and the functional differences of GG, such as hydration, gelation, stability, crystallinity, and textures [152,155]. Based on the degree of acetylation/deacetylation, GG exists in two forms: high and low acetylated GG. Gels from natural GG are soft and elastic, however, deacetylated GG makes brittle, firm, and optically clear gel after following various alkaline treatments [155].

GG solutions are known for their thermoreversible gel characteristics [156]. This sol-gel transition in thermoreversible GG gels is driven by the structural transition from double-helical at high temperatures to extended helical at low temperatures through the formation of junction zones while cooling down [155]. The gelation and gelling characteristics of GG solutions are strongly governed by their chemical nature, molar mass, and concentration of GG, the number of cations, and the valency of the cations present in solutions [155,157]. GG physicochemical structure favors the formation of gels at low concentrations. GG makes highly viscous solutions even at low concentrations, and deacetylated GG solutions show pseudoplastic rheology showing little thixotropy [158]. Hence, GG solutions provide processing convenience for many potential packaging applications using different water-based film-casting methods.

Successful attempts have been reported using GG in active and intelligent packaging to facilitate control release of active compound and monitoring food spoilage [159–161]. Versatile materials are incorporated into the GG matrix to make blends and composites and initiate crosslinking to achieve enhanced physicomechanical and barrier characteristics of GG films and coatings [162–164].

Antimicrobial food packaging developed from Konjac glucomannan (KG)/ GG/nisin which was reported antimicrobial activity against *Staphylococcus aureus* with increasing GG content [163]. In another study, coffee parchment waste (CP) was incorporated with GG to synthesis antimicrobial packaging, and showed Antifungal activity against against *Fusarium verticillioides*, *Fusarium* sp., and *Colletotrichum gloeosporioides* [159]. Packaging material developed from GG/2-hydroxyethyl cellulose (HEC)/lignin showed high ultraviolet (UV) protection: 100% protection against UVB (280 - 320 nm) and 90% against UVA (320 - 400 nm) [162].

Active and intelligent packaging film was developed using GG/heat-treated soy protein isolate (HSPI)/*Clitoria ternatea* (CT) extract, for controlling anthocyanins release and monitoring freshness in seafood which also exhibited antioxidant and antimicrobial activity against *B. cereus* [160]. In another study, GG/AgNPs bionanocomposite was fabricated as a safe meat spoilage indicator, which changes its color from yellow to colorless when exposed to H_2S, a volatile gas released from chicken breast and silver carp during storage [161].

7.2. Xanthan Gum

Xanthan gum (XG) (Figure 8) is an extracellular high molecular weight polysaccharide secreted by *Xanthomonas campestris* [165,166]. XG can be produced by aerobic fermentation process followed by heating to kill the bacteria, and precipitation of polymer using isopropyl alcohol [165,167]. XG has a broad spectrum of applications due to its excellent rheological and structural properties in food, biomedical, cosmetics, pharmaceutical, and textiles [165,166]. XG has excellent pseudoplasticity than many other thickeners [166]. XG improves the thickening, stabilization, gelation, and emulsification process. Therefore, XG is used in food and non-food (cleaners, coatings, polishes, and agricultural flowable) applications, to achieve desired product characteristics [165,168].

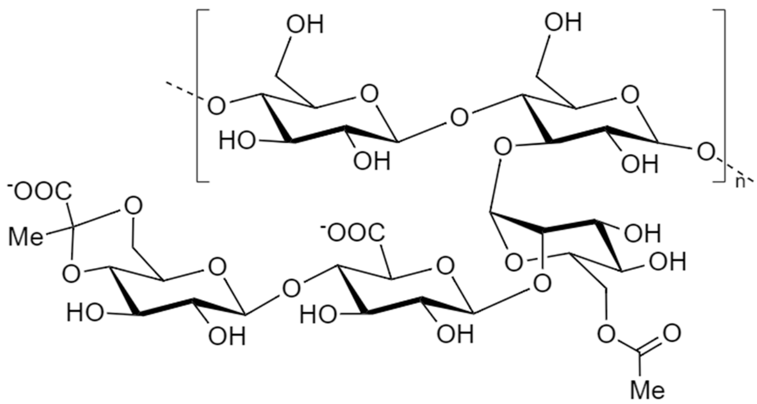


Figure 2. Chemical structures of Xanthan gum.

The chemical structure of the XG is an acidic heteropolysaccharide, and the pentasaccharide repeating units of XG with two glucose units, two mannose units, and one glucuronic [169]. XG has main chain consists of 1,4-linked β-D-glucose units, which is identical to cellulose, and the side chains consisting trisaccharide, β-D-mannose, 1,4-β-D-glucuronic acid, and 1,2-α-D-mannose, with internal mannose mostly O-acetylated, while the terminal mannose may be substituted by 4,6-linked pyruvic acid ketals [165,169]. The aforementioned structural uniqueness makes XG stable throughout a range of temperatures and pH [165].

XG is known for its good solubility in hot or cold water and makes highly viscous solutions even at low concentrations, exhibiting pseudoplastic rheological properties [170,171]. The unique chemical structure of XG has high surface activity, hence supporting different surface modifications that would enhance the chemistry of XG, such as compatibility, control release, water absorbency, and diffusivity for various applications [172–175].

Wide application of XG alone is somewhat limited, and XG is mostly blended with other polymers such as cellulose, gelatin, chitosan, agar, and clay [176–180]. Furthermore, high-performing composites of XG with organic and inorganic components have been successfully formulated for different packaging applications [172,180–183].

The properties, such as thickness, moisture content and WVP were reported to improve with the addition of XG (5%) in of gelatin-CMC film [176]. Lemon peel powder (LPP) incorporated with XG and TiO_2–Ag NPs (LPP/XG/TiO_2–Ag) showed improved mechanical property, antioxidant, and antimicrobial activity against *E. coli* and *Staphylococcus aureus* while decreasing the water solubility and WVP [184].

Raspberry pomace extracts (RPE) were incorporated into pectin/sodium alginate/XG composite film (PAX) proving the formulation of pH-sensitive colorimetric film used in monitoring the freshness of protein-rich food [185]. Fan et al., [186] also developed PAX edible composite film to preserve fresh cut vegetables or fruits. The XG/hydroxypropyl methylcellulose (HPMC)/tea polyphenols (TP) composite film (XHT) was synthesized for preserving fresh-cut bell peppers, which showed enhanced antioxidant activity, antibacterial activity *Staphylococcus aureus*, and retention of Vitamin C after 8 days of storage [187]. Addition of XG into chitosan reduced the WVP (10.41-10.68 $g^{-1}s^{-1}Pa^{-1}$) and exhibited antimicrobial activity against coagulase-positive *Staphylococcus*, *Salmonella* spp and coliforms and has the potential of preserving of refrigerated fish fillets [188]. Spray coating of XG solution mixed with citric acid (anti-browning agent) and glycerol (plasticizer) in fresh-cut lotus root resulted in decreasing enzymatic browning, inhibition of growth of *Bacillus subtilis* and improving the storage stability [189].

8. Packaging applications of polysaccharide-based bioplastics

Biolpolymers are widely used in the development biodegradable active, smart/intelligent food packaging material which serves as indicators, monitoring freshness of the food in real time by visual examination, maintaining the quality, reduce the loss [190–192]. Table 2 summarizes the recent developments of polysaccharide-based bioplastics in packaging applications. Table 2 also provides the packaging application and distinct properties of the bioplastics prepared from various polysaccharides.

Table 2. Properties and applications of polysaccharide-based bioplastics.

Polysaccharide type	Packaging application	Properties of the bioplastic	References
		Starch	
Starch/PBS	Food wrap and food container, grocery plastic bags	Very good elongation of break, outstanding bending capability (flexural modulus 378.69- 3188.48 MPa), good tensile properties (tensile strength [TS] 11.32- 18.13 MPa, Young's/tensile modulus [YM] 534.77- 2655.27 MPa)	[46]
Low density polyethylene/linear low density polyethylene/thermoplastic starch (LDPE/LLDPE/TPS)	Packaging applications	Adding starch at 15% yielded good mechanical properties (ultimate TS = 12.1 MPa, EB % = 250%), starch decreased the gloss%	[37]

Polypropylene (PP)/TPS	Biodegradable polymer	pseudo plastic in nature and exhibited shear-thinning behavior, EB is lower than PP, higher YM than PP.	[39]
Thermoplastic PVA/starch blend (TPPS)	Biodegradable polymer to replace starch polymers.	glycerol and urea as a complex plasticizer for TPPS increased TS (7.83 MPa) and EB (203%).	[42]
Cassava starch/glycerol/clay nanoparticles (NPs)	Biodegradable and cheaper food packaging	lower glycerol content presented better tensile and barrier properties, and clay NPs diminished the film permeability	[193]
Starch/clay (montmorillonite) NPs	Food contact material for vegetables	Increase of mechanical parameters (stress at peak = 6-22 MPa and YM = 450 -1135 MPa)	[194]
Carboxymethyl potato starch and citric acid (CA) (as a crosslinker and plasticizer)	Edible packaging	Highest tensile strength (160 kPa), Young modulus (650 kPa), and improved thermal stability (increased Tg 58 °C) was reported with CA at 30 wt%.	[195]
Cellulose			
Carboxymethyl cellulose (CMC)/ Chinese chives root extract (CRE)	Active packaging for sunflower oil	Higher oil resistance property, improved physical and barrier properties, antioxidant and antimicrobial activity against both Gram-positive (*B. cereus* and *S. aureus*) and Gram-negative (*E. coli* and *S. typhimurium*)	[58]
A 2,2,6,6-tetramehylpiperidine-1-oxy radical (TEMPO)-oxidized cellulose nanofibrils with free carboxyl groups (TOCN-COOH) prepared from the softwood celluloses	Biodegradable packaging	Flexible and highly transparent, higher YM (about 10 GPa) and lower elongation (about 5.1%) than those of the TOCN-COONa, lower oxygen permeability (0.049mL μmm^{-2} day^{-1} kPa^{-1}) than poly(ethylene terephthalate) films.	[196]
2,3-dialdehyde cellulose/nicin	Antimicrobial packaging for fresh pork meat at 4°C.	Improved mechanical property, lower water holding capacity, WVP, and oxygen permeability, Excellent antimicrobial activity against *S. aureus* and *E. coli*.	[59]
TEMPO-oxidized cellulose nanofibers (TOCN) prepared from the softwood and hardwood celluloses	High tech food and medicinal packaging material	Higher TS (about 200%) and YM (about 100%) than cellophane film. PLA film surface-coated with TOCN showed reduced oxygen permeability.	[197]
Hydroxyethyl cellulose, carboxymethyl chitosan and zinc oxide NPs	Composite film for food packaging	Exhibited lower water solubility and improved elasticity, thermal stability, UV shielding ability, antibacterial ability against Listeria monocytogenes and	[198]

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		Pseudomonas aeruginosa, improved crystallinity.	
Chitosan/bacterial cellulose composite with curcumin	Biodegradable food packaging for strawberry and edible oil.	Excellent barrier properties, hydrophobicity, mechanical, and antioxidant properties.	[118]
Cellulose acetate films with geranyl acetate (0.5% v/v and 1.0% v/v)	Food packaging	Antimicrobial activity against bacteria, Staphylococcus aureus and Escherichia coli and against fungi Aspergillus flavus.	[199]
	Alginate		
Gelatin/alginate film/1.5% oregano essential oil (OEO)	Antimicrobial food packaging for fish preservation	Increased antimicrobial effect on psychrotrophic bacteria, total viable count (TVC), and *Enterobacteriaceae*.	[95]
Alginate/Sulphur NPs	Antimicrobial film for frozen food with high moisture content (meat products)	S NPs at 2% improved the tensile strength by 12% and water vapor barrier by 41%, and UV barrier by 99%, hydrophobicity. Exhibited bactericidal activity against *Listeria monocytogenes*.	[93]
Alginate/Alvera/ZnO NPs	Antimicrobial edible coating for tomatoes	Improved mechanical, UV-shielding, and antimicrobial properties.	[94]
Alginate/cottonseed protein hydrolysates (CPHs)	Active food packaging for the preservation of fatty foods.	Increased the barrier properties to visible light, total phenolic content, antioxidant and antimicrobial (against *Staphylococcus aureus*, *Colletotrichum gloeosporioides* and *Rhizopus oligosporus*) activities. But increased the WVP without affecting moisture content, biodegradability, solubility or oil barrier property.	[96]
	Carrageenan		
PLA laminated on agar/κ-carrageenan/clay nanocomposite	Multilayer films for packaging of various types of food materials for keeping the quality and extending the shelf life.	Lamination with PLA layers (triple layer) improved WVP (5.0 × 10^{-11} g m/m^2 s Pa) and water resistance, decreased OTR (0.03 cm^3/m^2 day) in bionanocomposite film, thermal stability of the bionanocomposite also increase.	[200]
Alginate film prepared with $CaCl_2$ treatment using two methods: mixing films and immersion films.	biodegradable or edible films	Transparent film, increased TS and decreased EB. WVP of the immersion films decreased significantly, but did not decrease in mixing films.	[201]
Semi-refined kappa-carrageenan/ glycerol or sorbitol	Edible biodegradable packaging films	Addition of plasticizers at 30%, increased the TS, EB, moisture content, water solubility, WVP,	[110]

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		and reduced oxygen permeability. Increased transparency and their seal strength, reduced oil permeability.	
	Chitosan		
Chitosan/nano ZnO/ neem essential oil	Antibacterial food packaging	Addition of nano ZnO and neem essential oil improved TS, EB, and thickness, decreased the WVP, water solubility, and swelling property, and improved the antibacterial activity against *Escherichia coli*.	[121]
Chitosan-coated plasticized cassava starch films	Packaging film	chitosan coating increased the TS and YM, and decreased in EB, water uptake, wettability, and WVP	[119]
Chitosan/magnetic-silica nanocomposite/turmeric essential oil (CS/MNP/Si/TEO)	Antimicrobial packaging for Surimi	Antimicrobial activity against *Bacillus cereus* over 14 days of storage in packaged *Surimi*	[122]
Chitosan/halloysite nanotubes (HNT)/*Citrus limetta* pomace extract (LPE)	Active food packaging	Addition of LPE at 20% increased the crystallinity and antioxidant activity of CS film.	[123]
Chitosan/extract of propolis (PS)	Active food packaging for oxidation-sensitive food products	Improved thermal stability and mechanical property and reduced water solubility without affecting biodegradability (2×3 cm film buried in 5 cm depth in soil at 25°C for 15 days), exhibited antioxidant and antimicrobial activity against gram positive bacteria, *Arthrobacter* sp., *S. aureus*, and *S. hominis* and mould *M. rancensis*.	[202]
	Gellan gum		
Konjac glucomannan (KG)/gellan gum (GG)/nisin	Antimicrobial food packaging	Maximum TS = 17.5 MPa and lower moisture uptake value when adding 70% KG, antimicrobial activity against *Staphylococcus aureus* increased with GG content.	[163]
Gellan gum (GG)/ Heat-treated soy protein isolate (HSPI)/ *Clitoria ternatea* (CT) extract	Active and intelligent packaging films for controlling anthocyanins release and monitoring freshness in seafood	Showed colorimetric pH indicator properties, decreased TS and EB, improved antioxidant and antibacterial activity against *B. cereus*.	[160]

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Gellan gum/ silver NPs	Intelligent packaging for monitoring of meat spoilage	A colorimetric hydrogen sulfide (H_2S) sensor as ultra-strong binding ability of Ag with H_2S to form Ag_2S.	[161]
Gellan gum (GG)/ 2-hydroxyethyl cellulose (HEC)/ lignin (L) -	Food packaging with UV barrier property.	Incorporation of lignin improved the thermal and mechanical and hydrophobic properties, showed high ultraviolet (UV) protection: 100% protection against UVB (280 - 320 nm) and 90% against UVA (320 - 400 nm), showed antioxidant and non-cytotoxic activity.	[162]
Gellan gum/coffee parchment waste (CP)	Antimicrobial food packaging	Antifungal activity against against *Fusarium verticillioides*, *Fusarium* sp., and *Colletotrichum gloeosporioides*. Gallic, chlorogenic, p-coumaric, and sinapic acids along with caffeine were identified.	[159]
Xanthan gum			
Chitosan/Xanthangum	Packaging of refrigerated fish fillets	Reduced the WVP (10.41-10.68 $g^{-1}s^{-1}Pa^{-1}$), exhibited antimicrobial activity against *Staphylococcus*, *Salmonella* spp, and coliforms.	[188]
Low-molecular-weight xanthan gum	Foods to alleviate and resist the oxidative damage induced by reactive oxygen species (ROS)	Exhibited good free-radical scavenging activity and low cytotoxicity on Caco-2 cells injured by H_2O_2.	[167]
Gelatin/CMC film/Xanthan gum (XG)	Biodegradable food packaging	Addition of XG (5% w/w), improved thickness, moisture content, WVP, UV barrier properties.	[176]

9. Health and environmental effects of using biopolymers as food packaging

The petroleum-based materials possess serious health and environmental issues, such as greenhouse gas emission, environmental pollution, persistence in marine and terrestrial habitats. Though the biopolymers replace these petroleum-based materials, the true health and environmental impact of the biopolymers is need to be accessed [203–205].

To date, there are only a few studies conducted so far to assess the health effects and long term safety of polysaccharide-based packaging upon ingestion, absorption, metabolism, and excretion as well as the migration of additives/nano-fillers, crosslinking agents, stabilizers, etc., and the potential interaction between packaging material and food component [206,207].

Among primary, secondary, tertiary or quaternary packaging, primary packaging is considered the most important concern for health aspects as it is in direct contact with the food. Interaction between packaging material and food can occur through migration, permeation or sorption which impacts the sensory attributes of the food product and possibility of contamination with toxic materials that can affect consumers' health [208]. The bioplastic materials used for direct contact with food material (e.g., primary packaging materials), should comply with EU commission regulation No.10/2011 [209], and the novel interventions engineered nanomaterials covered by the EU

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Regulation 2015/2283 [210]. The potential migration, allergenic, and toxicological study for novel bio-based materials should be considered, despite only limited studies focused in this regard [209].

The long-term intake of food-grade carrageenans as an additive, may increase the incidence of intestinal inflammation or promote inflammatory recurrence in patients with colitis health issue [104].

Certain studies have reported that nanocellulose derived from bacteria and algae exerts physiological changes above non-toxic level (0–50 μg/mL) in endothelial cells, and induces fibrous bronchiolitis, pulmonary granulomas, inflammation, alveolitis [205].

In some instances, interaction between packaging material and food product can be beneficial, such as packaging material containing antimicrobial and antioxidant. However toxicological effects of eventual migration of active compounds/nano-fillers into food needs are needed to avoid health hazards and ensure food safety. Nanoparticle in active/intelligent and food contact material can only be used if explicitly authorized or named in Annex I of Regulation (EU) No 10/2011 [203].

The effect of biopolymers mainly depends on the type of feedstock (first generation, second generation, third generation) used for the biopolymer production and their health and environmental effects are listed in Figure 2 [205]. The greatest part of bioplastics currently available on the market are obtained from biomasses of the first generation due to the high yield of production, but extensive use of these source impact on food chain and food insecurity. Thus, many studies focus on development of biopolymers from second biopolymer materials, such as food wastes [204,205].

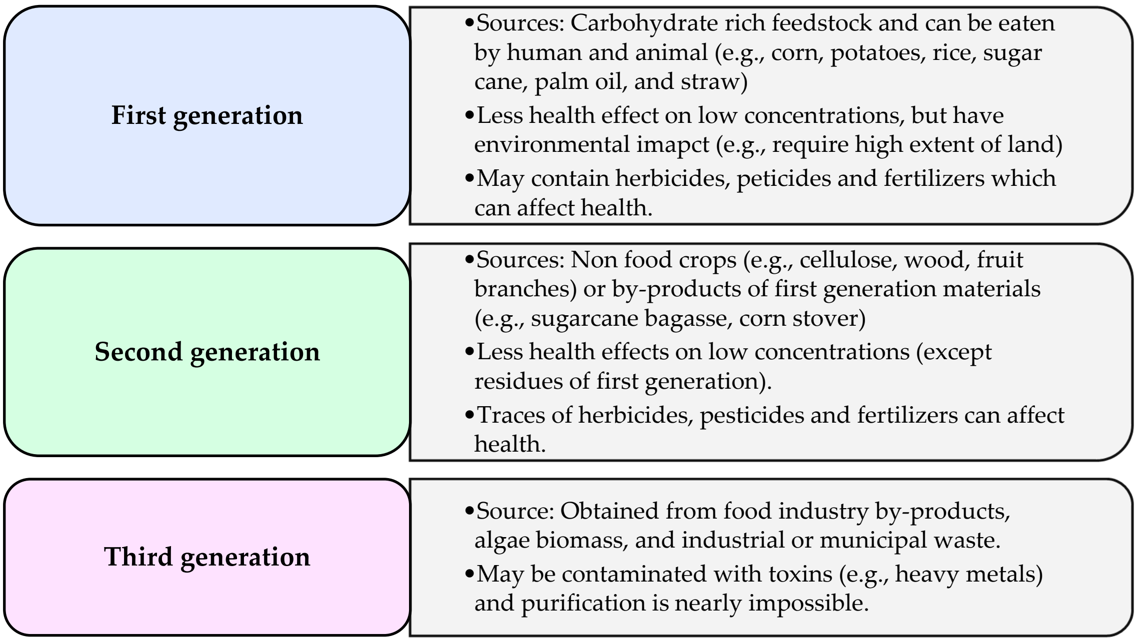

Figure 2. Sources and possible health risks of different raw materials used for biopolymer production.

Other than the toxicity, land-use, greenhouse emissions, and societal impacts must be accessed to study the sustainability of bioplastics [206].

Biodegradable and compostable biopolymers are a environmental friendly alternative materials and help to reduce CO_2 emissions and environmental impacts [206,211]. If a food packaging material is claimed to be biodegradable, the material should be tested for its extent of biodegradability or composability. Various international and European standards have been developed for testing of biodegradability of packaging materials; ASTM D6868, ASTM G21, ISO 14851:1999, and ISO 14853 [212].

10. Conclusions

Among numerous biopolymers, polysaccharides have been widely considered green, sustainable, nontoxic, renewable and environmentally benign materials for a diverse range of applications. This review focused on the structure and properties of polysaccharide-based bioplastics and their packaging applications. Due to the over usage of petroleum-based synthetic packaging materials and their related environmental issues, there is a significant interest in producing biopolymer-based packaging materials. However, critical setbacks of polysaccharide-based materials include sensitivity to moisture and mechanical strength, requiring modification or the addition of one more component in the system, which could increase the cost of the final product.

Interestingly, polysaccharide-based materials exhibit excellent gas barrier properties and biological activity, making them promising materials to expand the future of edible films and intelligent and active food packaging. Therefore, it is foreseen that the future growth of the development and application of polysaccharide multifunctional materials in the food packaging sector.

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