



Review

Structure, Properties, and Recent Developments in Polysaccharide- and Aliphatic Polyester-Based Packaging—A Review

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Abstract: Food packaging plays an imperative role in the food processing sector by safeguarding foods from their point of harvesting until the moment of consumption. In recent years, biopolymers have attracted the attention of the scientific community as an alternative to conventional packaging materials. Among the available biopolymer sources, a lot of the focus has been on polysaccharides due to their superior barrier properties against gases, oils, and odors and their processing versatility. Moreover, there is also a growing interest in aliphatic polyester as a potential replacement for petrochemical-based synthetic plastics. Both polysaccharides and aliphatic polyesters have gained popularity in sustainable food packaging due to their unique characteristics, including their low cost, availability, biodegradability, gas and moisture barrier properties, film-forming capabilities, excellent heat resistance, and ability to be processed into films, trays, and coatings. This review highlights the structural features, properties, and recent advancements of several vital polysaccharides, namely, starch, chitosan, cellulose, alginate, pectin, carrageenan, and aliphatic polyesters, including polylactic acid (PLA) and polyhydroxybutyrate (PHB) for developing packaging materials, and their applications in the food industry. Conventional packaging and future perspectives of biopolymer-based food packaging are also comprehensively covered in this review.

Keywords: food packaging; biopolymers; polysaccharides; aliphatic polyesters; biopackaging



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1. Introduction

According to the World Health Organization (WHO), there are an estimated 600 million cases of foodborne diseases and 420,000 deaths yearly due to contaminated and unsafe food [1]. Therefore, it is crucial to intake safe and nutritious food to maintain a long and healthy life. Food can be contaminated at various stages throughout the production chain, from farm to consumption. Maintaining food safety is a top priority for public health and essential for attaining food security. Moreover, food safety and quality control systems are vital not only for protecting the health and well-being of consumers but also for supporting economic growth and boosting livelihoods by providing access to local and global markets.

Advanced food processing and packaging techniques are important to maintain a safe food supply worldwide. Food packaging is a key player in the food processing industry by safeguarding foods from their point of harvesting to the time of consumption. Food packaging protects its content from physical, biological, enzymatic, and biochemical damage. Beyond preservation, packaging also provides containment, utility, and communication [2]. The most used conventional food packaging materials include plastics, glass, metal, paper and paperboards, wood, and composites. These materials have long been the basis

of the packaging industry as they have various properties that meet food preservation requirements [3]. Even though conventional packaging materials fortify foods from different contaminants, other factors related to production costs and social and environmental aspects have raised many concerns over the past few decades.

Furthermore, single-use packaging materials prepared from conventional raw materials are the most widely employed in the food industry. They are predominantly disposed of right after use, causing numerous environmental concerns. For instance, over two-thirds of the total production of single-use packaging materials is used in the food sector alone. Due to changes in food production and consumption habits and the growing population, this number continues to rise [4]. Consequently, accumulating packaging waste in large quantities has made it extremely difficult to handle solid waste sustainably. Surprisingly, around 30–35% of municipal solid waste comes from global packaging waste, and food packaging accounts for approximately 60% of total solid municipal waste [5]. Hence, to address issues related to conventional food packaging, sustainable packaging materials with better thermal, mechanical, and barrier properties have significant importance in the food industry [6]. Interestingly, many investigations are underway to develop alternative food packaging materials due to increased consumer awareness about health, food quality, food safety, and environmental sustainability pertaining to food packaging [7].

In this context, one of the emerging solutions for safer and sustainable food packaging is using biopolymer-based packages that alleviate health and environmental concerns over conventional packaging [6]. Biopolymer-based packaging or biopackaging materials exhibit distinct features such as relative abundance, renewability, and biodegradability. Interestingly, the global market for biopackaging materials is expected to grow rapidly in the coming years. It is forecasted that the production capacity of bio-based packaging is set to upsurge from the 2019 figure of 2.11 million tons to around 2.43 million tons in 2024 [8]. Among many biopolymers, polysaccharides and aliphatic polyesters have been widely considered for developing biopackaging materials for the food industry. Biopolymers can either be directly extracted from natural biomass or chemically synthesized from biomass-derived monomers or microorganisms [9]. For example, natural polysaccharides such as starch, cellulose, chitin, alginate, pectin, and carrageenan and synthetic aliphatic polyesters such as PLA and PHB have been investigated for food packaging applications [10]. Although numerous review articles have been published on the use of biopolymers in food packaging applications, there is scarce information available explicitly emphasizing the structure, properties, and food packaging applications of polysaccharides and aliphatic polyesters [11–13].

This review first examines conventional food packaging materials, namely, paper, plastic, glass, and metal, to understand their strengths and weaknesses. Then, it sets the stage for a comparative study, discussing the advantages of biodegradable materials over conventional materials regarding functionality and environmental sustainability. After that, the current review focuses on the structural arrangement, properties, and recent advancements of polysaccharides and aliphatic polyesters for food packaging. The roles of polysaccharides, including starch, cellulose, chitosan, pectin, alginate, and carrageenan, and synthetic aliphatic polyesters, such as PLA and PHB, in biopackaging are elaborated upon. Recent trends and future outlooks of the biopackaging industry are also summarized in this review. Most importantly, we report the recent findings related to polysaccharide- and aliphatic polyester-based food packaging to disseminate knowledge on further advancing biopackaging materials and their potential as sustainable alternatives to conventional packaging.

2. Conventional Food Packaging

Food packaging is a complex system that protects food until it arrives at its destination in perfect condition through transportation, distribution, and storage. Various packaging materials have been introduced to provide the desirable functionality of a packaging system for different food items. The selection of proper packaging materials is essential

for maintaining product quality until usage, as well as fulfilling product requirements such as barrier properties and economic, environmental, and social factors [14]. Generally, conventional packaging can be divided into three categories based on their application: primary, secondary, and tertiary. Primary packaging comes directly into contact with the food product and provides a protective barrier, secondary packaging stores several primary packagers, and tertiary packaging is employed for bulk storage and transportation [3]. Paper and paperboard, plastics, glass, and metal are the primary packaging materials utilized for most commercial food products, and a combination of more than two packaging materials is used to provide the best protection for food products, as shown in Figure 1. The following section discusses the conventional packaging materials and their pros and cons.

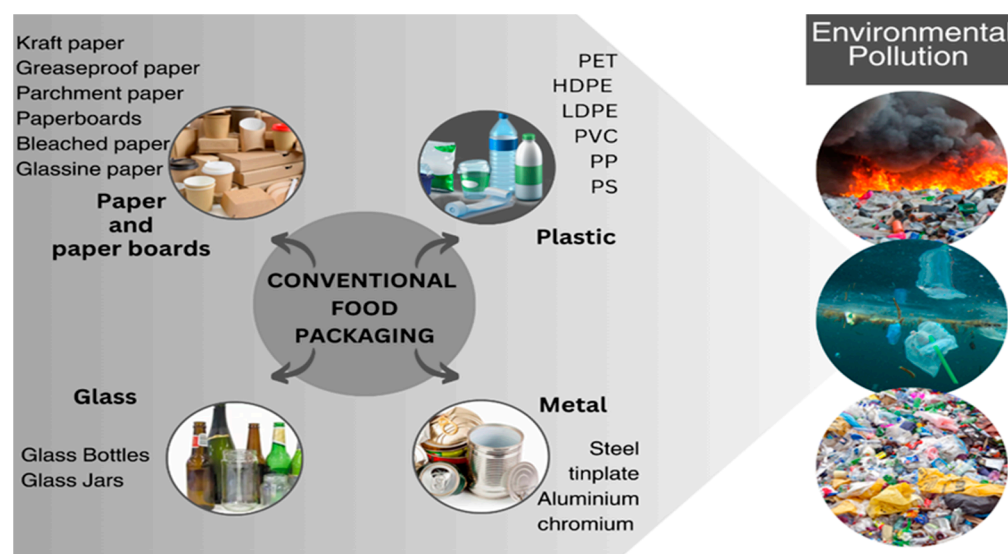


Figure 1. Main types of conventional food packaging materials.

2.1. Paper and Paperboard

Paper and paperboard represent over 30% of the worldwide packaging industry and are commonly applied in the food industry for product containment and preservation [15]. Paper and paperboard packages can be recognized as environmentally friendly due to their biodegradability, which promotes their usage in the food industry. Many paper and paperboard packages are available, from lightweight packages such as infusible tissues for tea bags to heavy tertiary packaging boxes. Paper and paperboard have been utilized for food packaging since the seventeenth century, and usage increased in the late nineteenth century [16]. Of total paper production, more than 92% is made from wood pulp and the remaining 8% is generated from agricultural by-products such as bagasse, straw, flax, and corn husks [17]. The quality of the paper depends on the type of raw materials used for paper development and their fiber and pulp properties [18]. Cellulose is responsible for providing a fiber-forming ability with long and straight fibers, hemicellulose in wood pulp is responsible for bond development during the paper-beating process, and lignin acts as a natural binding component for the paper development process [15]. After the pulping process, all unit operations, such as bleaching, beating, refining, and final treatments, are responsible for manufacturing paper with different qualities and appearances [19]. Table 1 summarizes the various types of paper packaging used in the food industry.

Table 1. Major types of paper used in food packaging.

Paper Type	Properties	Application	Ref.
Kraft paper	Type of coarse, high-strength, economical, porous, tear-resistant paper with a rough surface that can be coated or laminated	Beverage carriers, boxes, sacks, cartons, packages for flour, dried fruits, sugar	[15]
Greaseproof paper	Translucent, machine-finished, resistant to oils	Wraps cookies, confectionary, snack foods, highly oily foods	[20]
Parchment paper	Made from acid-treated pulp, not heat sealable, poor air and moisture barrier properties, high wet strength, greaseproof	Layer between pastry or meat slices, labels for fatty foods, cheese wrapping	[15]
Glassine paper	Glassy, smooth surface; transparent sheet; good grease and oil resistance; high density	Liner for baked goods, biscuits, cookies, cooking fats	[21]
Bleached paper	Soft and white, weaker compared to unbleached paper, expensive	Food labels, flour, sugar, fruits and vegetables	[22]
Paperboards	Thicker than paper, rigid, foldable; different types are available: whiteboard, liner board, food board, carton board, chipboard, corrugated board	Rigid boxes, beverage cartons, boxes for fruits and vegetables	[22,23]

Paper and paperboard packaging possesses several advantages, including low production cost, biodegradability, lightweightness, flexibility, printability, and recyclability [24]. However, they are also associated with environmental and health issues. For instance, chemicals added during pulping and coating to enhance the properties of final packages may migrate into foods, resulting in adverse health effects. Organic and inorganic dyes, mineral oil phthalates, and polyfluorinated substances are some of those migrants, and high mineral oil migration has been reported with recycled papers [15]. Other than that, many researchers have found that printing inks used for paper packages could cause cancer [25]. Moreover, the paper-making process requires a large volume of water and is energy-intensive, giving rise to vast amounts of wastewater and pollutants.

2.2. Plastic

Plastics are synthetic polymers with macromolecular structures obtained from repeating units of low molecular weight monomers [26]. During the last few years, plastics have become versatile and essential materials in the food industry. They are the second most used packaging material due to their flexibility, chemical resistance, low cost, lightweightness, and physical and optical properties. Plastics are commonly recognized as single-use packaging materials, contributing immensely to the environment and human health [27,28]. Global annual plastic production was estimated at 360 million tons in 2018 and it is projected that the worldwide production of plastics will reach 25 billion tons by 2050 [29]. Single-use plastic accounts for nearly 40% of overall plastic usage [30]. Moreover, it was estimated that China is the world's top producer of plastic materials, meeting around 29.4% of global demand, followed by Europe and North America, with 18.5% and 17.7% of the global market, respectively [31]. In the packaging sector, 50% of the plastics for the food packaging industry are obtained from fossil fuels, and these fossil fuel-derived plastics take many years to degrade [4]. Thermoplastics are popular in food packaging applications. A thermoplastic is any polymer that becomes pliable or rubber-like above a specific temperature, known as glass transition temperature (T_g), and solidifies below T_g after cooling. The transition from the rubber-like state to the glass state is a key characteristic of polymer behavior, providing significant changes in the physical properties, including elasticity and hardness. Table 2 summarizes different types of plastics and their applications in food packaging.

Table 2. Major types of plastics used in food packaging.

Plastic	Properties	Application	Ref.
Polyethylene terephthalate (PET)	Good barrier to gases and moisture; resistant to heat, mineral oils, solvents, and acids; transparent; tough	Beverage and mineral water bottles, jars, tubes, trays	[32,33]
High-density polyethylene (HDPE)	Good barrier to solvents and moisture, high tensile strength, opaque, high-temperature capability	Beverage and milk bottles, shopping bags, ice cream containers	[30,34]
Polyvinyl chloride (PVC)	High resistance to chemicals, high strength, good oil barrier properties, good heat sealability	Bottles, food wraps	[35]
Low-density polyethylene (LDPE)	Good heat sealing; resistant to acid, oils, and bases; rigid; flexible; transparent	Bakery, frozen, fresh produce, and meat packing; soft squeeze bottles	[36]
Polypropylene (PP)	Good water vapor barrier, resistant to gases and odors, high strength, puncture resistance	Containers for ice cream, margarine, yogurt, snack packs, and biscuit packs	[34]
Polystyrene (PS)	Brittle, rigid, poor barrier to moisture and gases, good insulation properties	Cutlery, food insulation boxes, meat trays, egg containers	[37,38]

From 1950 to 2018, 6.3 billion tons of plastics were produced; only 9–12% were recycled and incinerated and the remaining 79% were accumulated in the environment [39]. Due to their recalcitrant nature, plastic packages remain in the environment for decades, causing soil, water, and air pollution. The leaching of many hazardous chemicals in plastic packaging materials into the soil, underground, and into other water sources is an enormous consequence of landfilling after use [40]. Plastics also release significant amounts of greenhouse gases upon oxidation and incineration [41]. Different types of additives in plastics, such as bisphenol A, poly-fluorinated chemicals, phthalates, and brominated flame retardants, are toxic and potential carcinogens [30,42]. Also, synthetic packages are petroleum-based, contributing to the depletion of nonrenewable petroleum resources [4]. Furthermore, photodegradation causes these plastics to break into tiny fragments, eventually forming microplastics. Microplastics have long been known for their role in environmental implications and may enter the marine and human food chains, leading to health risks.

2.3. Glass

Glass is one of the oldest food packaging methods and dates back to 2500 BC. According to the American Society for Testing and Materials (ASTM), glass is an amorphous inorganic product of fusion that has been cooled to a rigid condition without crystallizing [43]. Glass is made of silica, which is naturally found in silica sand, with different additives by fusion at high temperatures [44]. Recycled glasses can be used as a substitute for virgin materials, reducing energy usage during glass production [45]. Glass bottles and jars are employed primarily in the food industry and generally contain 68–73% of silica, 10–13% of limestone, 12–15% of soda ash, and 1.5–2% of alumina [46].

Glass is applied in food packaging because of its excellent barrier properties, heat resistance, transparency, moldability, rigidity, and strength [47]. However, the main disadvantages of glass packages are their production costs, heavy weight, light permeability, and fragility. When comparing the interaction of food packaging material with packaged food, it was discovered that glass is the only packaging material that prevents toxic substances from being transferred from the package to the food and maintains the best quality of the food product [44]. However, the most notable setbacks of glass packaging include the high energy requirement and air pollution during glass production, which is three-fold higher than for plastic production [44].

2.4. Metal

Metals are widely employed in the food industry for various applications and, amongst the total usage packaging materials, 15% is accounted for by metal packaging [48]. Excellent barrier properties, good physical protection, recyclability, formability, heat resistance, and decorative ability enhance consumer preferences for metal packaging [49]. Steel, tinplate, aluminum, and chromium are metals used for packaging dairy products, fruits, vegetables, beverages, meat-based products, bakery, and confectionary products [48]. Steels provide a barrier to gases, odors, moisture, and light; coating improves heat sealability due to ductility [50]. Steel coated with tin is one of the most used materials in food packaging and is utilized to produce cans and sheets for bulk products [51]. Aluminum can be formed into different forms, such as cans, foils, and laminated films; they are lighter and weaker and can be alloyed and shaped easily compared to tinplate [22].

The main food safety issue of metal packaging is the migration of materials, including metallic compounds and their interaction with food. Bisphenol A, lead, chromium, aluminum, cadmium, mercury, and nickel coatings are some migrants from metal packaging to food [48]. Metal packaging causes minimal direct impact on the environment compared to plastic packaging other than landfilling. However, metal packaging contributes to drainage blockage and the pollution of aquatic sources and requires high energy during production; additionally, chronic metal and polyaromatic hydrocarbon co-exposure may lead to cancer [48]. Table 3 compares the advantages and disadvantages of mainstream packaging materials.

Table 3. Advantages and disadvantages of conventional packaging materials.

Packaging Material	Advantages	Disadvantages	References
Paper and paperboard	Low production cost, biodegradable, lightweight, flexible, printable, renewable, and recyclable	Combined with other packing materials, limited barrier properties, less durable, susceptible to damage, and unsustainable	[15,24]
Plastic	Versatile, lightweight, flexible, chemically resistant, low-cost, better physical properties, inert characteristics, easily processed, and recyclable	Non-biodegradable, causes environmental pollution, leaches many hazardous chemicals into foods, and dependent on fossil fuels	[22,41,44]
Glass	Durable, chemically inert, recyclable, transparent, good barrier properties, heat-resistant, and high-strength	Fragile, heavier than other materials, high production cost, and more energy consumption in production	[44,47]
Metal	Durable, excellent barrier properties, good physical protection, recyclable, good formability, good heat resistance, and versatility	High production cost, corrosive, and non-biodegradable	[48,50]

3. Biopolymers for Sustainable Food Packaging

Conventional petroleum-based packaging materials, primarily plastics, are currently associated with numerous concerns, including the accumulation of packaging waste, environmental pollution, climatic changes, and health effects. Therefore, packaging materials with enhanced environmental and sustainable attributes have received significant attraction for fulfilling food and environmental protection requirements [52]. According to the definition of sustainable food packaging, it must have numerous features, including safety for people and society throughout its entire lifecycle, cost-effectiveness, utilization of renewable energy sources and recycled materials, production process feasibility, and cleaner technologies [53].

Currently, biopolymers are considered sustainable and innovative packaging materials due to their relative abundance, renewability, nontoxicity, biodegradability, easy function-

alization, and environmental benignity [54]. They are polymers entirely biosynthesized by living organisms or chemically synthesized from natural sources [55] and can be easily degraded biologically [56]. Interestingly, these biopolymers have the capacity to substitute most conventional food packages [52,57].

These biopolymers contain different functional groups, such as hydroxyl, amide, amino, phosphate, and phenol, and there are three main types of natural biopolymers: polysaccharides, polypeptides, and polynucleotides [58]. Polysaccharides are made up of simple sugar units combined via glycoside bonds, amino acids are the monomers for polypeptides, and polynucleotides (DNA, RNA) are polymers of nucleotide monomers [59]. Moreover, biodegradable aliphatic polyesters, including polylactic acid (PLA) and polyhydroxyalkanoates (PHA), are chemically synthesized from bio-based monomers [56]. This review mainly focuses on the recent advances in polysaccharide- and synthetic aliphatic polyester-based biopackaging materials for the food industry. Figure 2 synthesizes the biopolymers of the three categories and their key features related to sustainable packaging discussed in this review.

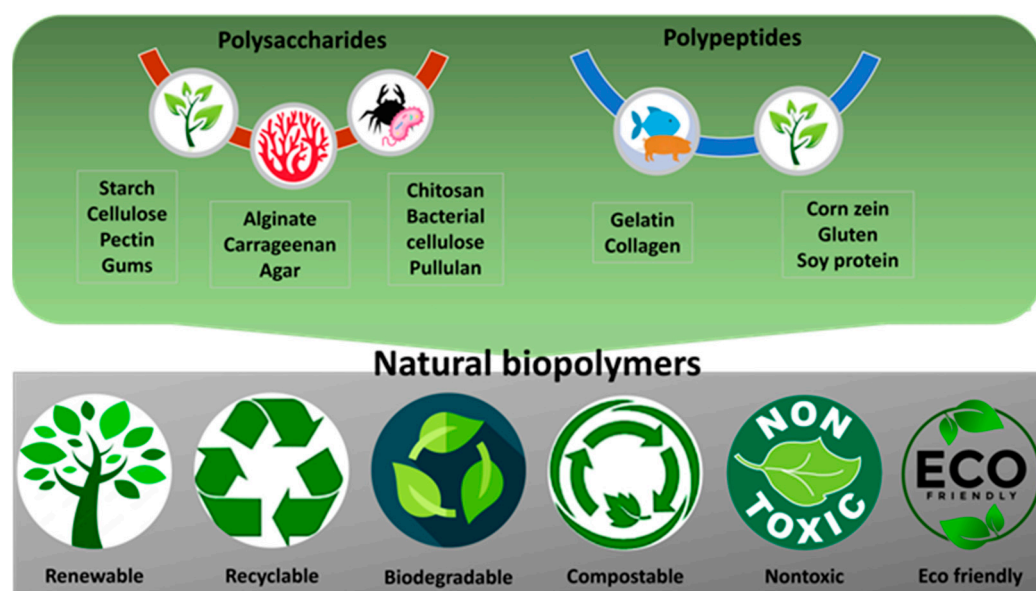


Figure 2. A summary of biopolymers and their key elements related to sustainable food packaging discussed in this review.

3.1. Polysaccharide-Based Biopackaging Materials and Their Applications in the Food Industry

Polysaccharides are the most abundant macromolecule in the biosphere and can be obtained from plants, animals, and microorganisms. Polysaccharides are a group of complex carbohydrates with different degrees of polymerization by α -1,4-, β -1,4-, or α -1,6-glycosidic bonds [7]. Wide varieties of polysaccharides have been used to develop biodegradable packaging materials, such as edible films and coatings since they have excellent barrier properties against gases, oils, and aromas and processing adaptability [60]. Edible films and coatings are thin layers of edible materials used to wrap or coat a product to give protective and functional benefits, which can be ingested together with the product. Many films formed using polysaccharide-based biopolymers show high tensile strength and percentages of elongation and are comparable to synthetic polymers [27]. The main disadvantage of polysaccharide-based films is their poor barrier properties against water vapor due to their hydrophilic nature [61]. Several polysaccharides, including starch, cellulose, chitosan, pectin, alginate, and carrageenan, have been explored for their potential to form films and coatings. The properties, including mechanical properties, solubility, barrier properties, and gelation, vary depending on the type of polysaccharide [62]. However, structural modifications can enhance these properties according to the final purpose, and different additives such as plasticizers, antioxidants, and antimicrobial agents can also

be incorporated with polysaccharides. Moreover, combining two or more biopolymers improves the properties of the blend films with excellent barrier properties [63]. The most widely explored polysaccharides in food packing include starch, cellulose, chitosan, pectin, alginate, and carrageenan.

3.1.1. Starch

Starch is a natural polysaccharide derived from plants and composed of amylose, a linear polymer with α -1,4-linked d-glucose monomer units, and amylopectin, a branched polymer with α -1,4-linked d-glucose monomer units and 1,6 linkages. The chemical structures of amylose and amylopectin units in starch are shown in Figure 3 [7]. Roots, tubers, and seeds are botanical sources of starch found in the form of granules. Starch is mainly isolated from wheat, corn, tapioca, cassava, potato, and rice [64]. Their granules are spherical, oval, or irregular in shape, with diameters ranging from 0.1 to 200 μm , and are insoluble in cold water [52]. Depending on the starch source, granule properties, and amylose and amylopectin content, the physiochemical and functional properties of starch can be varied. Most starches are semi-crystalline, with crystallinity between 15% and 45%. The crystalline regions contain short-branched chains of amylopectin and amorphous regions include amylose and branching points of amylopectin [65]. According to an X-ray scattering study of starch crystallites, there are two primary starches: type A and type B. Type A is mainly found in cereals, while type B is mainly in amylose-rich starches, including tubers and root crops. Moreover, mixed forms of type A and type B structures are called type C and are found in bean and pea starches [66].

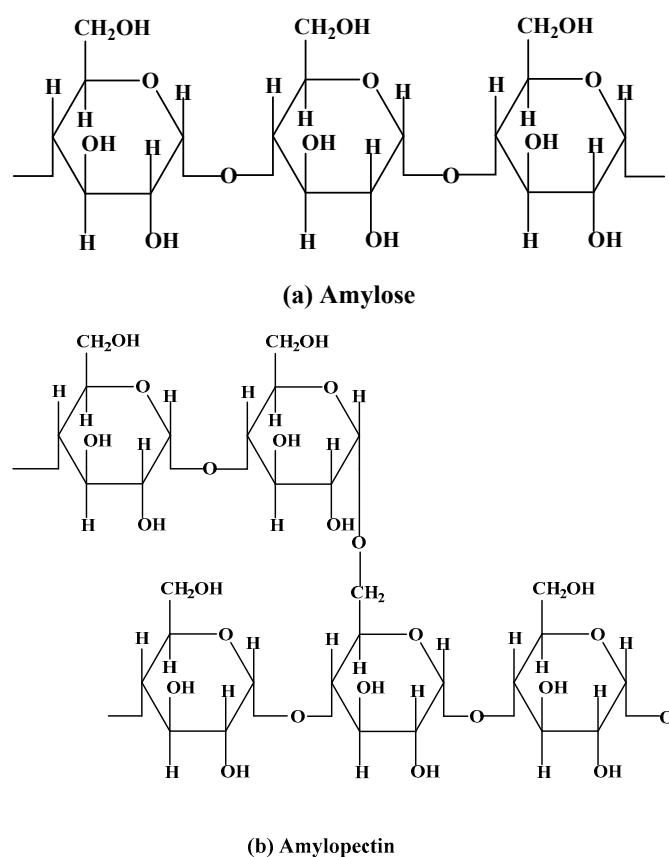


Figure 3. Chemical structures of (a) amylose and (b) amylopectin in starch.

Starch-based films exhibit low oxygen permeability but high hydrophilic properties. On the other hand, poor mechanical properties and retrogradation limit the use of starch-based films as food packaging material. However, incorporating plasticizers such as glycerol and sorbitol, grafting with hydrophobic polymers such as polylactic acid (PLA),

and blending with other polymers help overcome these drawbacks by improving chain mobility and flexibility [67,68]. Starch can be converted into thermoplastic starch by exposure to high temperatures and shear stress in the presence of water or plasticizers [69]. In the last few years, starch-based thermoplastics have been extensively used as bio-based compostable materials to develop bags, films, and food containers [7,70]. Two widely tested techniques for formulating starch-based films include solution casting or the wet method and extrusion or the dry method. Figure 4 depicts schematic representations of the wet and dry methods used for film preparation. The solution casting method is conducted by solubilizing starch in a solvent followed by drying. In contrast, the extrusion process is applied by plasticizing and heating starch above its T_g [71]. Regarding applications, different food products, including candies, bakery products, fruits and vegetables, dry products, and snacks, can be packed using starch-based films [72].

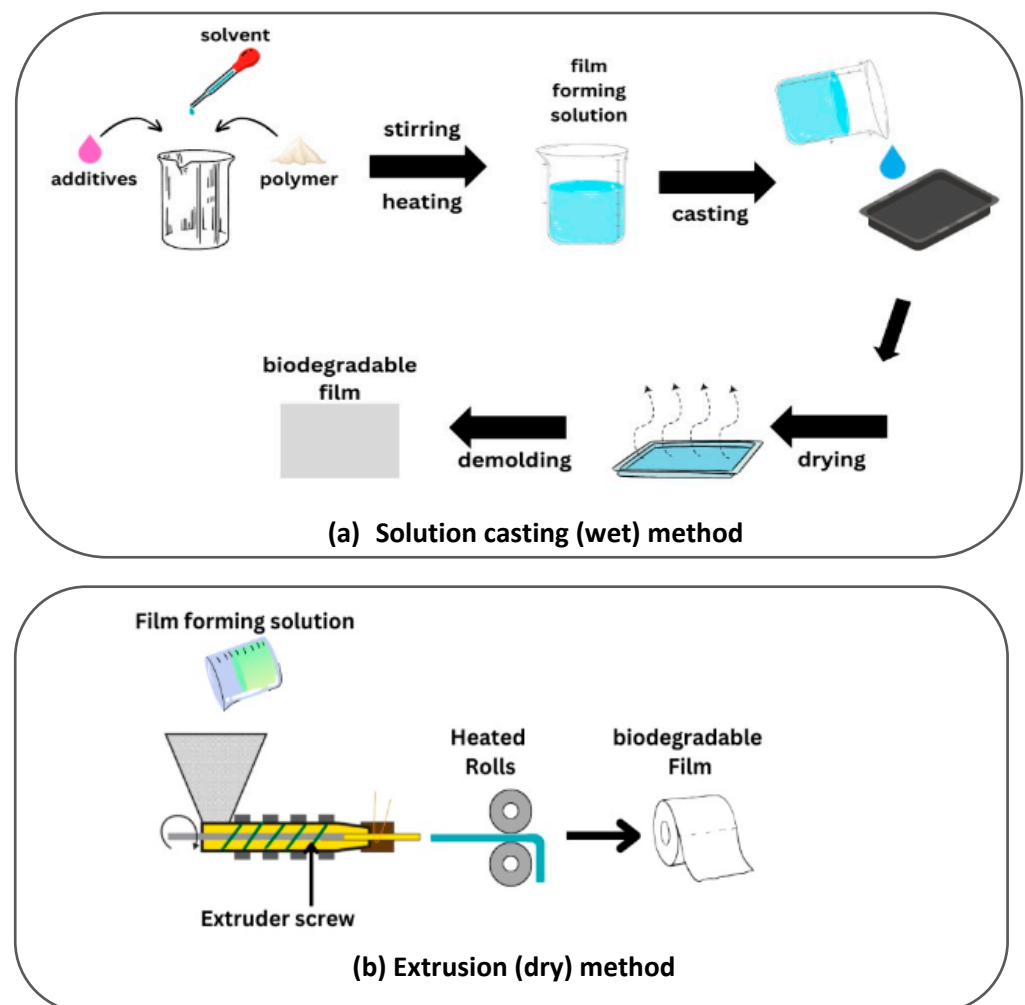


Figure 4. Schematic representations of solution casting (wet) and extrusion (dry) methods for preparing packaging films.

According to the literature, numerous studies have investigated the potential of starch-based materials for food packaging applications. For instance, Issa and coworkers studied the impact of sweet potato starch-based composite films with thyme essential oil and montmorillonite nano clay on fresh spinach leaves [73]. They showed that biodegradable packaging material effectively reduces microbial growth while enhancing the sensory quality of baby spinach leaves during cold storage [73]. Baek et al. developed an antioxidant film with cowpea starch and maqui berry extract to enhance the storage life of salmon by delaying lipid oxidation [74]. Recently, Chen et al. tested potato starch-based film incorpo-

rated with tea polyphenols to extend the shelf-life of fresh-cut fruits. Figure 5 shows the effect of potato starch-based film incorporated with different concentrations of tea polyphenols on the color of fresh-cut bananas. They demonstrated strong free radical scavenging activity and water vapor and oxygen barrier effects with high doses of tea polyphenols [75]. Garcia and coworkers suggested that the corn starch-based edible film with olive extract is an affordable and environmentally friendly food packaging material to prevent the oxidation of packed foods [76]. Behera et al. developed a novel biodegradable film using yam starch and bentonite, using it as an alternative to synthetic packaging materials [77]. Liu and coworkers enhanced the shelf-life of beef sauce by 4–6 days compared to a control by delaying rapid oxidation and microbial growth using an antimicrobial composite film prepared with cassava starch, konjac glucomannan, chitosan, and *Zanthoxylum armatum* essential oil [78]. Bangar and coworkers also prepared a pearl millet starch-based active food packaging material with cellulose nanocrystals and clove bud oil and investigated its potential for extending the shelf-life of red grapes [79]. Marichelvam et al. successfully synthesized a biodegradable film using starch extracted from the *Prosopis juliflora* plant and gelatin with similar properties to synthetic packaging materials [80]. The development of edible starch-based film using tef starch and agar with proper mechanical properties was conducted by Tafa and coworkers [81]. Ardjoum and coworkers evaluated the antimicrobial properties of a cornstarch-based film by incorporating *Thymus vulgaris* essential oils and ethanolic propolis extract against *E. coli* and *Listeria monocytogenes* [82].

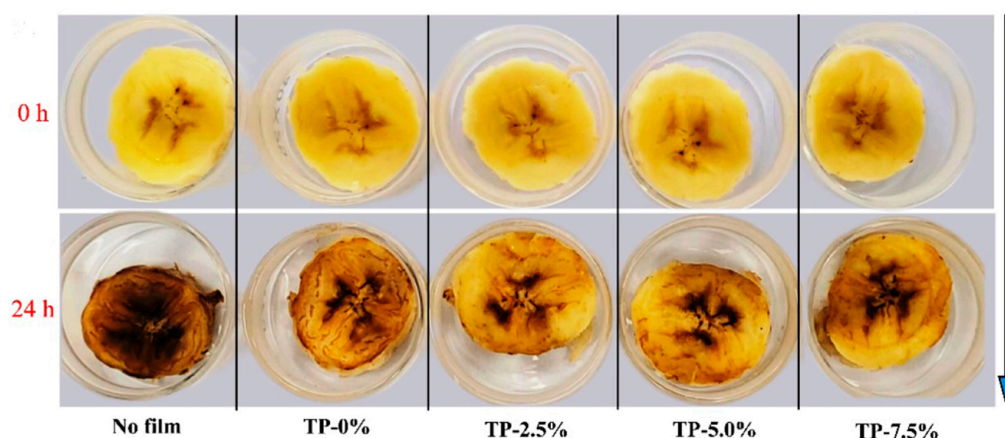


Figure 5. The effect of potato starch-based film incorporated with different concentrations of tea polyphenols on the color of fresh-cut bananas (reprinted with permission from [75]).

3.1.2. Cellulose

Cellulose is the most abundant biopolymer on the planet and the primary component in plant cell walls and natural fibers. Microorganisms, such as algae, fungi, bacteria, and tunicate family organisms, also biosynthesize cellulose [83]. Wood pulp and cotton fibers are the most used commercial sources of cellulose and, recently, plant-based waste products such as sugar cane bagasse, peel, husk, and shells are also considered for extracting cellulose [7]. Cellulose is a linear polymer composed of D-glucopyranosyl units linked by β -(1 \rightarrow 4) glycosidic bonds, which are covalently bonded between the equatorial -OH group of C4 and the C1 carbon atom through acetal functionalities [84]. This allows cellulose chains to be packed densely together, generating a strong inter-chain hydrogen bonding network. However, neat cellulose is not suitable for film development since it is highly crystalline, has a high molecular weight, and is insoluble in water due to strong intermolecular and intramolecular hydrogen bonding between chains [85]. Therefore, cellulose is converted into various derivatives, such as hydroxyethyl cellulose, methylcellulose, hydroxypropyl methylcellulose, and carboxymethyl cellulose, by breaking the polymer chains before being processed into bioplastic films to obtain unique chemical and physical properties [86]. Generally, the cellulose extracted from bacteria is much purer than

plant-based cellulose, with good strength, molding ability, and water-holding capacity [87]. Figure 6 represents the chemical structure of cellulose.

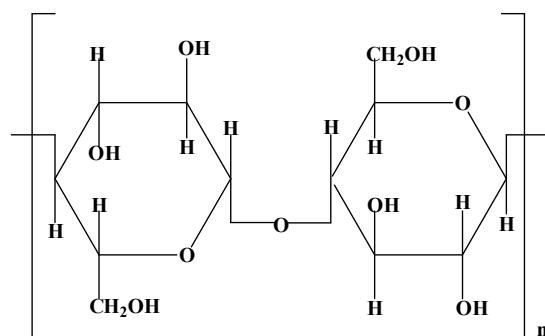


Figure 6. Chemical structure of cellulose.

The films prepared from cellulose derivatives display intriguing characteristics, including transparency, tastelessness, odorlessness, flexibility, robustness, and excellent barrier properties [88]. Hydroxypropyl methylcellulose is widely used to develop edible films with high water vapor barrier and mechanical properties [7]. Methylcellulose is the hydrophilic derivate of cellulose and it produces films with excellent gas and lipid barrier properties with less water vapor resistance [65]. Different methods, such as solution casting, layer-by-layer assembly, extrusion, coating, and electrospinning, are applied for fabricating cellulose-based packaging materials [89]. Moreover, studies have shown that blending cellulose with different additives such as biopolymers, plasticizers, and active agents could enhance the mechanical properties and storage ability [90].

Cellulose-based materials have broadly been tested for food packaging applications. Atta et al. prepared bacterial cellulose (BC)- and carboxymethylcellulose (CMC)-based bioactive food packaging material with olive oil and ginger oil as antimicrobial agents [88]. The authors revealed that the developed coating extended the shelf-life of oranges and tomatoes by inhibiting the growth of three bacterial strains and two fungal strains under different storage conditions for up to 9 weeks [88]. An epichlorohydrin-crosslinked hydroxyethyl cellulose functional composite film was prepared with polyvinyl alcohol and ϵ -polylysine as reinforcing agents by Zhang et al. The authors tested its antibacterial, barrier, and mechanical properties and ability to act as a packaging material for grapefruit. They reported that the shelf-life of packaged grapes could be extended for up to 6 days due to the excellent barrier properties and antimicrobial properties of the prepared film [89]. Yaradoddi and coworkers demonstrated the use of agricultural waste-derived carboxymethyl cellulose to develop cost-effective packaging materials with good mechanical and barrier properties [90]. Moreover, Romao et al. summarized previous research on cellulose-based films with antimicrobial and antioxidant properties for food applications [91]. Moradian and coworkers synthesized a bacterial cellulose-based active film with herbal extracts, including pomegranate peel, green tea, and rosemary, to pack button mushrooms [92]. The authors revealed that the fabricated film helped extend the shelf-life of mushrooms with its antioxidant and antimicrobial activity, as shown in Figure 7 [92]. Al-Moghazy et al. developed an active food packaging material with cellulose to enhance the storage life of cheese by delaying the growth of microorganisms [93]. Yordshahi and coworkers studied the impact of postbiotics-incorporated bacterial nanocellulose antimicrobial wrappers on ground meat [94]. They found that the developed films effectively extended the shelf-life of ground meat by reducing mesophilic and psychrophile counts [94]. The development of active food packaging materials using gallic acid-loaded hydroxypropyl methylcellulose and polyethylene oxide (PEO) nanofibers and their potential as a packaging material for walnuts was investigated by Aydogdu et al. [95]. Further, Liu and coworkers discussed numerous studies on developing biodegradable films prepared from cellulose and derivatives for the food packaging industry [96].



Figure 7. The physical appearance of fresh mushrooms packed with pure bacterial cellulose-based membranes (a) and active membranes containing pomegranate peel extract (b), green tea extract (c), and rosemary extract (d) after 5 days (1) and 15 days (2) of storage at 4 °C (reprinted with permission from [92]; copyright (2024) with permission from John Wiley and Sons).

3.1.3. Chitosan

Chitosan is the N-deacetylated derivative of chitin, with at least 50% free amine, the second most abundant natural polysaccharide on earth after cellulose. Chitin is made up of N-acetylglucosamine (β -1,4 linked 2-acetamido-D-glucose) monomers and can be found in the exoskeleton of crustaceans and cell walls of fungi and insects [97]. Chitin is also discarded as waste from the shrimp and crab processing industries. Chitosan polysaccharide is polymerized with two monomers, N-acetyl-D-glucosamine and D-glucosamine, randomly linked through β -(1-4) glycosidic bonds [98]. Chitosan can be deacetylated using concentrated alkali at elevated temperatures or by an enzymatic hydrolysis process in the presence of chitin deacetylase [99]. Figure 8 shows the structure of chitosan.

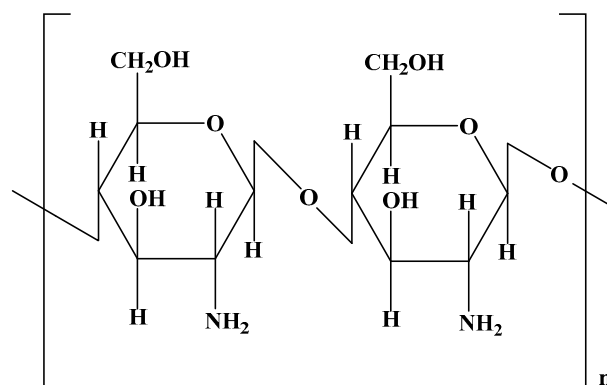


Figure 8. Chemical structure of chitosan.

Chitosan is insoluble in aqueous solutions due to its deprotonation at higher pH values (>6.5), thus limiting its usage in the food and pharmaceutical industries [100]. However, chitosan dissolves in weakly acidic solutions with a $\text{pH} < 6.5$ due to the protonation of the free glucosamine [59]. Chitosan also shows excellent antimicrobial properties against spoilage and pathogenic microorganisms such as fungi, bacteria, and viruses, making it versatile in different industries [97]. Generally, chitosan-based films exhibit good mechan-

ical properties, semi-permeability to gases such as carbon dioxide and oxygen, and low barrier properties [7,56].

The characteristics of chitosan-based films depend on numerous factors, including the degree of deacetylation, molecular weight, free amine regeneration mechanism, and solvent evaporation process [101]. The solution casting technique is widely practiced in preparing chitosan-based films. In the solution casting method, chitosan and other additives, such as plasticizers, are dissolved in a slightly acidic solvent, poured into a flat surface, and allowed to dry. The extrusion process is not applied to chitosan-based films because chitosan is not a thermoplastic and degrades before the melting point, hindering commercial usage [65]. Hence, numerous methods have been proposed to overcome these drawbacks. Blending chitosan with different thermoplastic polymers enhances the thermal properties and sealability of the film. On the other hand, incorporating different hydrocolloids and biopolymers increases the water vapor transmission rate and other mechanical properties [98]. Moreover, recent studies have also focused on incorporating natural antimicrobial and antioxidant agents and nanomaterials with chitosan-based films to improve shelf-life [102].

Biodegradable films and edible chitosan coatings have emerged as sustainable alternatives to compete with conventional non-biodegradable food packaging technologies. The development of chitosan-based films from edible cricket species and their application in the food industry was investigated by Malm and coworkers [103]. Zehra et al. prepared a chitosan-based film with thyme essential oil, zinc oxide (ZnO), polyethylene glycol (PEG), nanoclay (NC), and calcium chloride (CaCl₂) as additives [104]. Then, the film was tested for the preservation of collard greens under refrigerated conditions for 24 days. The authors reported that the chitosan-based film effectively extended the shelf-life of collard greens [104]. A chitosan-based edible coating incorporated with tomato plant extract enhanced the shelf-life of pork loin by reducing microbial growth, maintaining the sensory attributes of pork during 21 days of storage [105]. Wang and coworkers developed a packaging film for bread using chitosan blended with poly(ϵ -caprolactone) (PCL) and grapefruit seed extract (GFSE) by extrusion and compression molding techniques [106]. They reported improved food safety and bread shelf-life by inhibiting different foodborne pathogens [106]. Figure 9 shows the physical appearance of bread packed in the developed chitosan-based film over other packaging materials [106]. Xu et al. prepared a chitosan-based film with bacterial cellulose and curcumin that exhibited good mechanical properties and antioxidant activity, with the potential to be used for packaging foods with high fat contents [107]. Chitosan and whey protein hydrolysate composite films with improved physicochemical and mechanical properties were prepared by Al-Hilifi and coworkers [108]. Yao and coworkers successfully formulated chitosan- and polyvinyl alcohol-based active films incorporated with curcumin for food packaging applications [109]. Recently, De Carli developed a biodegradable active film based on chitosan from crayfish shells enriched with bioactive propolis extract. The prepared film exhibited good antioxidant, antimicrobial, and mechanical properties with the potential to package oxidation-sensitive food products [110]. Chitosan film with zinc oxide (ZnO) nanoparticles and sodium montmorillonite nanoclay was fabricated by Rodrigues et al. and demonstrated antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus*, with enhanced mechanical and physical properties [111]. Karkar et al. synthesized an active, edible, chitosan-based film enriched with *Nigella sativa* L. extract and investigated its potential to extend the shelf-life of grapes. They concluded that the developed film could prolong the shelf-life of grapes compared to the control, which was not covered [112]. Furthermore, applications of chitosan-based composites in the food industry were summarized by Kumar et al. [113].

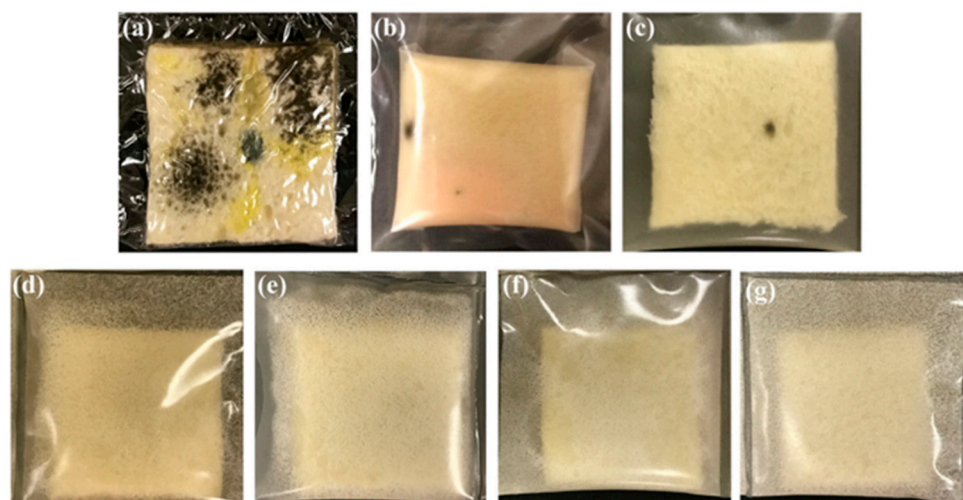


Figure 9. The physical appearance of bread packaged by (a) polyethylene (PE), (b) PCL/chitosan, (c) PCL/chitosan/GFSE 0.5 mL/g, (d) PCL/chitosan/GFSE 1.0 mL/g, (e) PCL/chitosan/GFSE 1.5 mL/g, (f) PCL/chitosan/GFSE 2.0 mL/g, and (g) PCL/chitosan/GFSE 2.5 mL/g films at 24 °C, 70% RH for 7 days (reprinted from [106]; copyright (2024) with permission from Elsevier).

3.1.4. Pectin

Pectin is a plant-based polysaccharide that makes up nearly two-thirds of the dry mass of the primary plant cell wall. This complex polysaccharide comprises 10–30% of plant primary walls and 2–5% of secondary walls [114]. It is a high-molecular-weight, heterogeneous, amorphous, white, colloidal carbohydrate found in fruits and vegetables, mainly apple pomace and citrus peel [115]. The pectin backbone comprises D-galacturonic acid units linked with α (1→4) linkages [60]. Figure 10 depicts the chemical structure of pectin.

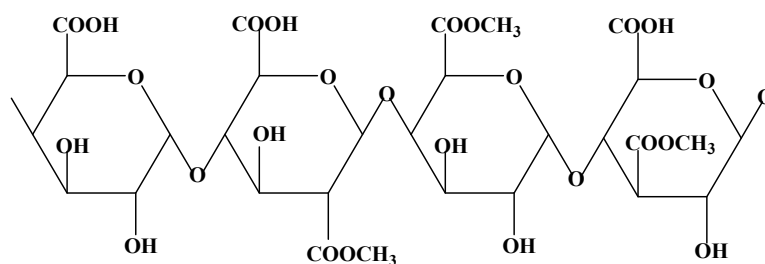


Figure 10. Chemical structure of pectin.

According to structural studies, pectin does not have a specific structure, comprising around 100–1000 saccharide units in a chain-like configuration. Therefore, it is a hetero-polysaccharide of three main domains, namely, homogalacturonan (HG), rhamnogalacturonan-I (RGI), and rhamnogalacturonan-II (RGII) [116]. Homogalacturonan is the major component of pectin polysaccharide, composed of (1-4)-linked α -D-galacturonic acid and methyl esters. Rhamnogalacturonan I consists of (1-4)-linked α -D-galacturonic acid and (1-2)-linked α -L-rhamnose. Rhamnogalacturonan II is a complex structure of (1-4)-linked α -D-galacturonic acid backbone with side chains and sugars [117]. The plant source and development stage govern the chemical composition, quantity, and structural properties of pectin.

In the food industry, pectin is mainly used as a thickening agent, colloidal stabilizer, gelling agent, and emulsifier, depending on its degree of esterification [116]. Besides these applications, pectin and its derivatives are used to develop biodegradable and edible food packaging, primarily for fresh fruits and vegetables. Depending on the degree of esterification, there are two types of pectin: high-methoxyl pectin or rapid-set pectin and low-methoxyl pectin or low-set pectin. High-methoxyl pectin is the best for producing

good films and coatings [118]. The research on edible films developed from pectin and its derivatives, such as pectate and amidated, exhibited good barrier properties against gas, oil, and aromas and excellent mechanical properties. Still, the major drawback is the lack of moisture barrier properties due to their hydrophilic nature [56]. The casting method and extrusion are effective approaches for producing pectin-based films [119]. The casting procedure entails spreading a prepared film-forming solution over a flat surface, followed by a drying process, while the extrusion process uses high pressure and temperature for film development [120,121]. However, the brittle nature, high water solubility, and poor moisture barrier qualities of pure pectin films limit the usage of pectin-based films. The brittle nature and poor elongations of these films can be overcome by incorporating plasticizers, which increases their flexibility [119]. Besides that, antimicrobial substances, emulsifiers, and other biopolymers have been incorporated to obtain packaging materials with better barrier properties and antimicrobial activity [121]. Pectin-based films are primarily used to pack fresh and minimally processed apples, berries, papaya, tomatoes, and carrots [122].

Pectin has gained attention as a promising biomaterial for manufacturing bio-based sustainable packaging films and coatings. For example, biodegradable films were prepared from pectin extracted from apple and pequi mesocarp and their mechanical, thermal, and barrier properties were tested for their application as food packaging materials [123,124]. Sadadekar and coworkers developed nano chitosan- and pectin-based food packaging materials incorporated with fennel essential oil and potato peel extracts to enhance antimicrobial and antioxidant properties [125]. The fabrication of biodegradable films from pomelo peel pectin, casein, and egg albumin was conducted by Sood and Saini [126]. The authors observed better mechanical, barrier, and thermal properties for their hybrid films compared to the pure films of casein and egg albumin [126]. In another study, an edible pectin-based biodegradable film enriched with mulberry leaf extract was tested for the shelf-life performance of capsicum fruit [127]. The authors confirmed that the edible pectin-based film is a promising material to extend the shelf-life of capsicum fruit for up to 12 days [127]. Jiang and coworkers prepared an active and intelligent film using white-fleshed pitaya peel pectin, betacyanins, and montmorillonite [128]. Their films exhibited good mechanical and antioxidant activity and colorimetric response to pH and ammonia, showing their ability to monitor the freshness of shrimp [128]. A pectin-based film with sodium alginate and castor oil exhibited good moisture, barrier, and mechanical properties and extended the shelf-life of capsicum and chili compared to uncoated samples [129]. Teleky et al. evaluated the physico-chemical and mechanical properties of pectin-based films enriched with phenolic extracts from apple pomace and stated their applicability in food packaging applications [130].

Han and Song prepared a mandarin peel pectin-based film with sage leaf extract [131]. The synthesized film could prolong the shelf-life of food, preventing the degradation of the nutritional value of packaged food [131]. Nisar and coworkers successfully devised a citrus pectin-based film incorporated with clove bud essential oil and investigated its mechanical, thermal, antibacterial, and antioxidant properties for food packaging applications [132]. Moreover, different studies on pectin-based films from agro-waste residues and their potential to be used in the food packaging industry were discussed by Mellinas and others [121].

3.1.5. Alginate

Alginates are natural biopolymers derived from the marine brown algae of the family phaeophyceae and can also be synthesized by bacteria, such as *Pseudomonas* and *Azotobacter* [133]. Alginates are known for their biocompatibility, biodegradability, low cost, low toxicity, ability to react with polyvalent metal cations, resistance to acidic media, and solubility at basic pH [101]. They are linear, unbranched, and water-soluble polysaccharides made up of polyuronic acid with three block structures, namely, poly- β -D-manopyranosyluronic acid (M) blocks, poly- α -L-gulopyranosyluronic acid (G) blocks, and M-G blocks linked by

(1–4) linkages, with varying characteristics and distributions throughout the chain [134]. Depending on the plant source and the stage of plant development, the chemical composition and order of M and G units may differ [135]. The physical properties of alginates are affected by the percentage of the three types of blocks, and the hardness of the three blocks diminishes in the sequence GG > MM > MG [136]. Figure 11 depicts the chemical structure of M and G monomer blocks, and the various combinations of M and G blocks can produce at least 200 distinct alginates [137].

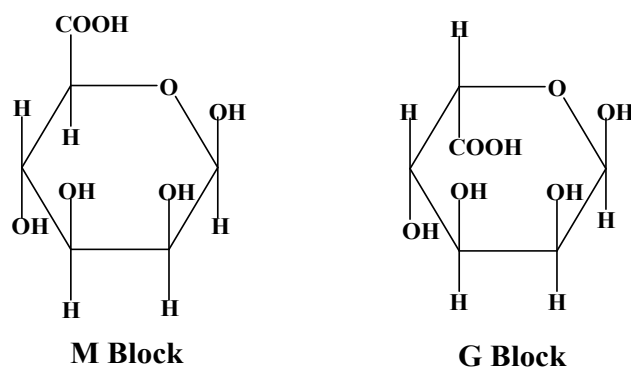


Figure 11. Chemical structure of M and G monomer blocks of alginate.

Alginates are commonly utilized in the food industry as a thickener, gelling agent, and colloidal stabilizer [138]. Also, alginates are used to develop food coatings by reacting with divalent cations, especially calcium (Ca^{2+}) ions, to generate water-insoluble polymers [139]. Alginate films can be fabricated via a two-step method: drying alginate solution and treatment with calcium salts to induce instantaneous crosslinking at the interface [84]. Alginates exhibit good film-forming characteristics; a uniform, transparent, and glossy appearance; flexibility; tastelessness; odorlessness; high tensile strength; tear resistance; low permeability to vapor and oxygen; and impermeability to fats and oils [137,139]. Furthermore, alginate-based films and coatings provide better barrier characteristics to bacteria and reduce the likelihood of microbial development in food products [140]. Alginate films were initially employed for fresh fruits and vegetables to reduce respiration rates since Ca crosslinking was more effective in attaching to a sliced fruit surface [139,141]. Apart from fruits and vegetables, alginate films are now applied to many other foods, including cheese, meat, and fish, resulting in the shelf-life extension of these products [137]. The properties of alginate films can be enhanced by incorporating different additives such as organic acids, essential oils, biopolymers, plant extract, and metallic nanoparticles [142].

Alginate-based edible coatings and films can efficiently maintain quality and lengthen the shelf-life of fruit, vegetables, meat, and cheese by controlling respiration and microbial growth and reducing dehydration [133]. Nair and coworkers studied the influence of alginate- and chitosan-based coatings enriched with pomegranate peel extract to extend the shelf-life of guava. The coating maintained the quality of guava at low temperatures for 20 days by improving visual and nutritional properties while delaying senescence [143]. Dulta et al. also prepared an alginate–chitosan-based film supplemented with nano zinc oxide (ZnO) and investigated its impact on orange quality for 20 days at 4 °C [144]. The film extended the shelf-life of coated oranges by more than 1.5 times compared to uncoated samples by delaying fruit senescence [144]. The impact of alginate-based edible coating with resveratrol on the shelf-life of rainbow trout fillet at refrigerated temperatures was examined by Bazargani-Gilani and revealed that the developed edible coating could extend the shelf-life of fish, with more health benefits [145]. Mahcene and coworkers summarized the potential of a sodium alginate-based film incorporated with essential oil to be utilized as a food packaging material to extend the shelf-life of foods [146]. An alginate-based bio-composite material prepared with pure reduced graphene oxide or mixed ZnO exhibited high antioxidant and antimicrobial properties against *E. coli* and *S. aureus*, potentially

extending food shelf-life [147]. Abdullah and coworkers prepared an alginate-based film incorporated with cinnamaldehyde for food packaging applications [148]. They stated high mechanical and antimicrobial properties against Gram-positive and Gram-negative bacteria, with a 98% reduction in microbial growth for their film [148]. Bata Gouda et al. evaluated the shelf-life of fresh-cut lotus root slices using sodium alginate mixed with an L-cysteine and citric acid coating [149]. The authors reported that their coating prevented microbial growth, browning, and membrane damage and extended the shelf-life of lotus root slices for 14 days at 4 °C [149]. Montone and coworkers synthesized alginate-based films charged with quercetin glycoside compounds and hydroxyapatite to coat fresh-cut papaya [150]. The results demonstrated the preservation of fruits by reducing microbial growth and respiration rate and preserving natural antioxidant compounds [150]. Packaging film based on alginate, carboxymethyl cellulose, and potato starch incorporating grapefruit seed extract was developed by Ramakrishnan et al. with excellent mechanical, antioxidant, and antimicrobial properties, which effectively extended the shelf-life of green chili by 25 days [151]. Feng et al. successfully developed a novel, cobalt-based, metal-organic framework-loaded, sodium-alginate-based packaging film with antimicrobial and ammonia-sensitive functions for shrimp freshness monitoring [152].

3.1.6. Carrageenan

Carrageenan is a marine-origin polysaccharide and can be isolated from red algae, most often from *Chondrus crispus*, *Kappaphycus alvarezii*, and *Eucheuma denticulatum* species [139]. Carrageenan is a linear hydrophilic polymer made up of sulfated or non-sulfated galactose and 3, 6-dehydrated galactose linked together by α -(1 \rightarrow 3) and β -(1 \rightarrow 4) glycosidic linkages [60]. There are three carrageenan types based on the position and quantity of sulfate groups on the disaccharide structures: kappa (κ), iota (ι), and lambda (λ) carrageenan [153]. In the food industry, kappa (κ) carrageenan is widely used, and the sulfate concentrations of the kappa (κ), iota (ι), and lambda (λ) carrageenan are 20, 33, and 41 percent (*w/w*), respectively [154]. The presence of sulfate ester groups in the structure influences the overall negative charge and water solubility of carrageenan [155]. Figure 12 shows the chemical structure of carrageenan.

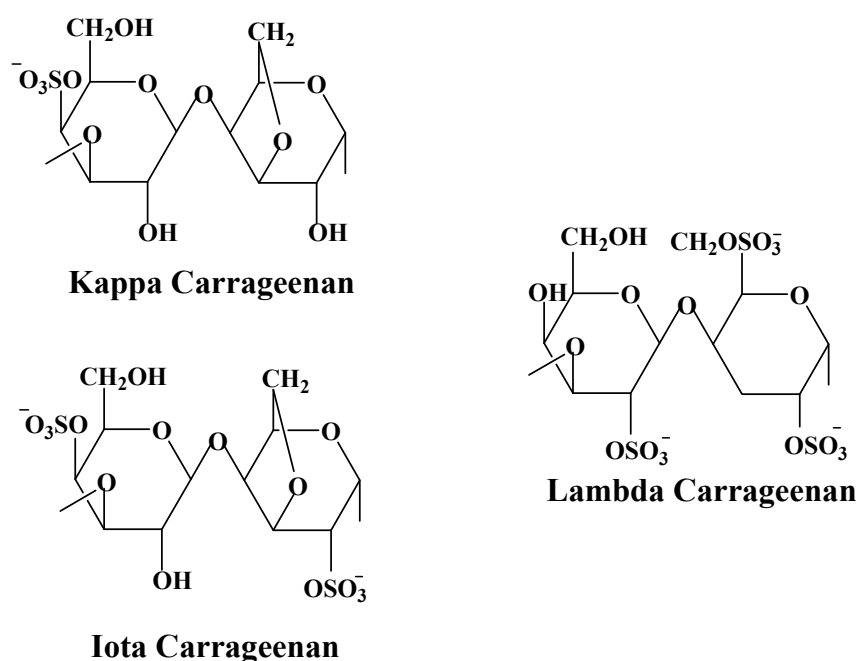


Figure 12. Chemical structure of carrageenan.

Carrageenan is commonly used in the food industry as a stabilizer, as a gelling and thickening agent in dairy products, in pet food, in infant food formulas, and for preparing

edible films [134]. Kappa (κ) carrageenan has been identified as the best type of carrageenan for edible food packages because kappa (κ) carrageenan produces robust, fragile, and firm gels, while iota (ι) carrageenan gels are weak, soft, and flexible [156]. Due to the higher gelling ability of carrageenan, random coils of polysaccharides change into double helices throughout the film-casting process by creating an extremely compact film with good mechanical and structural qualities [7]. Further, carrageenan-based coatings, edible films, and blends have been used in preserving freshly cut fruits due to the favorable properties of less moisture loss and gas exchange, preventing discoloration and maintaining the texture of the fruit [56]. Also, carrageenan films exhibit strong oxygen barrier properties and protect against lipid oxidation [157]. However, the water vapor permeability and high brittleness of carrageenan limit the usage of these polymer films in the food industry, and this can be overcome by blending them with natural or synthetic polymers [158]. Carrageenan film can be developed with antioxidant and antimicrobial properties by adding different essential oils and antimicrobial agents [159].

Recently, carrageenan-based food packaging films have received more attention due to their biodegradability, excellent biocompatibility, and availability. For example, Cheng et al. reviewed information on carrageenan extraction methods, methods of preparing biodegradable films, and their properties and applications for different food products to extend shelf-life [160]. Martiny and coworkers prepared carrageenan-based biodegradable films activated with olive leaf extract [161]. They reported that the developed film reduced aerobic mesophile growth and extended the shelf-life of lamb meat by acting as an active food packaging material [161]. Avila et al. investigated the mechanical, light barrier, antimicrobial, and antioxidant properties of carrageenan-based films incorporated with jaboticaba peel extract. They exhibited potential as active packaging materials for food applications [162]. Duan and coworkers fabricated a nanocomposite film from k-carrageenan, konjac glucomannan, and titanium dioxide nanoparticles and applied it to strawberry packing. They found that the shelf-life of strawberries could be extended for longer with the nanocomposite film compared to the conventional plastic package due to its excellent mechanical, thermal, barrier, and antimicrobial properties, as shown in Figure 13 [163]. Santos et al. evaluated the barrier, mechanical, and bioactive properties of developed k-carrageenan-based films incorporated with *Cymbopogon winterianus* essential oil as a novel food packaging material. The developed film presented high antioxidant activity and inhibited the growth of most foodborne pathogens [164]. Panatarani and coworkers protected minced chicken with semi-refined kappa carrageenan-based film mixed with cassava starch and ZnO and SiO₂ nanoparticles. The authors reported that the film exhibited improved water barrier and mechanical and optical properties while extending the shelf-life of minced chicken for up to 6 days [165]. Kim et al. synthesized a biodegradable carrageenan-based functional nanocomposite film with silver nanoparticles that was made using pine needle extract. The film exhibited intense antimicrobial activity, antioxidant activity, and UV protection, with the potential to be utilized for active food packaging applications [166]. Mahajan and colleagues developed a carrageenan-based edible film with *Aloe vera* that improved the microbiological and lipid oxidative stability of frozen dairy products during storage [167]. Color indicator film based on κ -carrageenan incorporating silver nanoparticles and red grape skin anthocyanin was developed by You et al. for fish freshness determination. The film displayed enhanced antioxidant activity, UV protection, antimicrobial activity against *E. coli* and *S. aureus*, and high mechanical strength, with a colorimetric response to pH and volatile ammonia variations [168].

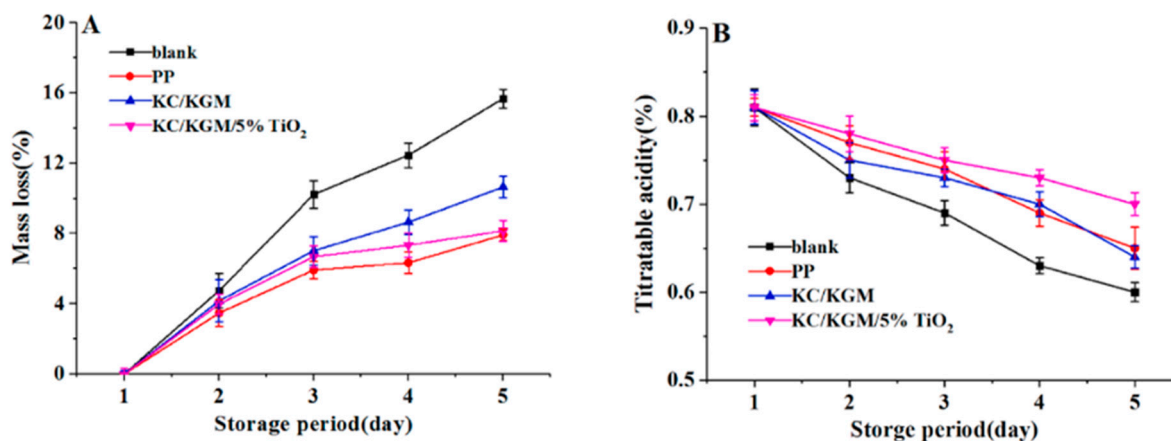


Figure 13. The effect of different packaging on the mass loss rate (A) and titratable acid (B) in strawberry: polypropylene (PP) film, k-carrageenan (KC)/konjac glucomannan (KGM) composite film, and KC/KGM/TiO₂ 5 wt% nano-composite film (reprinted from [163]; copyright (2024) with permission from Elsevier).

3.2. Aliphatic Polymer-Based Food Packaging

A major category of biodegradable polymers is aliphatic polyesters, with many exhibiting outstanding processability, biodegradability, and biocompatibility characteristics. Aliphatic polyesters are synthetic biopolymers obtained from the classical polymerization of renewable bio-derived monomers [62]. Polycondensation and ring-opening polymerization are the two main methods to synthesize aliphatic polyesters. Depending on the need and application, two different aliphatic polyesters are available: homopolymers and copolymers. Homopolymers are created when ester links combine the same monomer units, while copolymers are produced with two or more distinct types of blocks of monomer units [62].

The most researched aliphatic polyesters for a variety of applications are polylactic acid (PLA), polycaprolactone (PCL), polyglycolic acid (PGA) and polyhydroxybutyrate (PHB) and their copolymers [68]. Among these, PLA and PHB are mainly used to develop biodegradable packaging materials with high melting points between 160 and 180 °C [56]. The excellent heat resistance, gas barrier qualities, and ability to be processed into films, trays, and coatings of bio-based materials allow these aliphatic polyesters to be used as food packaging agents [169].

3.2.1. Polylactic Acid (PLA)

Polylactic acid (PLA) is one of the most widely used biodegradable polymers, with numerous applications in the commercial packaging industry. PLA is an aliphatic polyester made from lactic acid or lactide monomers [170]. A thermoplastic biopolymer called polylactic acid (PLA) is produced when bacteria ferment the carbohydrates in corn, sugarcane, or cassava [171]. The synthesis of lactic acid is the first stage in the multistep process that results in the synthesis of PLA, followed by the generation of the lactide monomer and the actual polymerization procedure [9]. Poly (L-lactide) (PLLA), poly (D-lactide) (PDLA), and poly (DL-lactide) (PDLLA) are the three stereochemical forms of PLA and, depending on the form, crystallinity, melting temperature, and tensile properties can differ. Figure 14 shows the chemical structure of PLA.

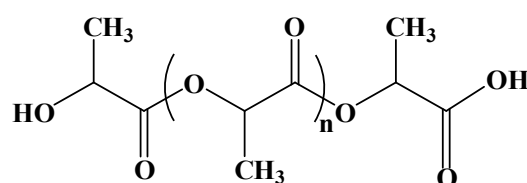


Figure 14. Chemical structure of PLA.

PLA is a good contender for several applications since it is an environmentally benign polymer with qualities like thermoplasticity and nontoxicity and equivalent mechanical properties to other traditional polymers like PP, PE, and polystyrene (PS) [172]. PLA has been authorized as a primary food packaging material because it is generally recognized as safe (GRAS) by the United States Food and Drug Administration (FDA). Different film-forming methods, such as extrusion, injection molding, thermoforming, and film blowing, can be employed to cast PLA-based sheets, films, and molded packages [4].

Features such as high recyclability, biocompatibility, low energy requirements in manufacturing, easy processability into various shapes, and good transparency and their desirable mechanical and barrier properties for oils and aromas have positively impacted the development of PLA-based packaging materials [173]. Studies have shown that PLA-based and polyethylene terephthalate (PET) bottles possess similar tensile strength and elastic modulus [174]. PLA bottle manufacturing utilizes 36% less energy and produces 44% less CO₂ than PET bottle manufacturing [4]. Also, PLA is superior to PP in terms of its tensile strength and O₂ and CO₂ barrier qualities. However, it has less toughness and impact strength than many non-biodegradable polymers [175].

Moreover, other drawbacks that limit PLA usage in the packaging industry include intrinsic brittleness, low-temperature resistance, and inadequate water vapor barrier qualities [176]. Depending on the application, the properties of PLA-based packages can be modified by blending with other biopolymers, crosslinking, or adding various natural fillers [62].

Different juices, milk, water, cheese, and yogurt are packaged using PLA-based packaging materials [10]. Food trays, films, bottles, sheets, and cups can be made of PLA, which is best for fresh products and those not needing to be protected from O₂ [177]. For example, Mohamad et al. developed a PLA-based film incorporating three active ingredients, namely, thymol, kesum, and curry, and investigated its impact on chicken meat [178]. They reported that the developed films extended the shelf-life of meat by 15 days while maintaining its sensory properties and microbial growth [178]. A PLA-based film activated with *Cinnamomum verum* essential oil was developed by Khanjari and coworkers and its impact on the sensory, microbial, and chemical properties of minced squab was assessed over 12 days at 4 °C. They revealed that the film reduced the growth of most bacteria, total volatile base nitrogen, and thiobarbituric acid reactive substances and maintained the sensory properties of minced squab [179]. Ardjoum et al. investigated the mechanical, thermal, and antimicrobial properties of PLA-based film enriched with *Thymus vulgaris* essential oil and an ethanolic extract of Mediterranean propolis and exhibited its potential as an active food packaging material [180]. PLA-, poly(butylene adipate-co-terephthalate)-, and starch-based films with salicylic acid were prepared and their potential as packaging for bananas was investigated by Ding et al. [181]. The authors reported that the shelf-life of bananas could be extended by 4–5 days due to the excellent barrier and mechanical properties of the films [181]. Hernandez-Garcia et al. evaluated the effect of a multilayer PLA film on the shelf-life of fresh pork meat compared to commercial, high-barrier, multilayer packaging films. Figure 15 depicts that the meat samples packaged in the PLA films maintained a reddish color and freshness at the end of storage compared to other samples [182]. Zhang and coworkers developed and characterized the properties of a PLA-based film enriched with cinnamaldehyde inclusions to preserve fruits [183]. The researchers found that the formed film exhibited enhanced barrier properties, tensile strength, morphology, crystallinity, and antibacterial activity against *Escherichia coli* and *Listeria monocytogenes* [183]. A film based on PLA, polybutylene-succinate-co-adipate, and thymol demonstrated antifungal properties against *Aspergillus* spp. and *Penicillium* spp. and prolonged the shelf-life of bread to 9 days [184]. Zhou and the research group reported that the developed sandwich-architected films based on PLA and pea starch were effective in extending the shelf-life of strawberries [185]. Wongthanaroj et al. examined the effect of nanocomposite films based on polylactic acid and cellulose nanocrystals on the browning reaction of cut avocados. The film exhibited a reduction in browning reactions and extended the shelf-life of avocados by 1.3 days at 23 °C and 5.4 days at 4 °C, with O₂ properties and antimicrobial

effects [186]. Further, more research on PLA-based intelligent food packaging materials with different natural antibacterial and antioxidant agents; their mechanical, physical, and antimicrobial properties; and the shelf-life extension of perishables was reviewed by Nasution and co-authors [187].

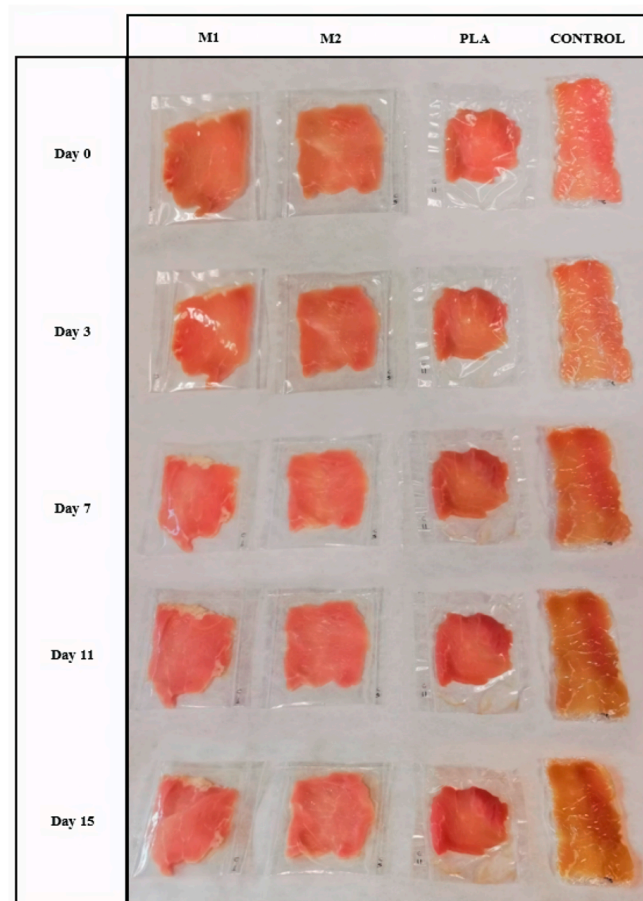


Figure 15. Visual aspect of pork meat fillets packaged in the M1 (PA6/EVOH32/PA6/LDPE) and M2 (EVOH48/EVA/EVOH48/EVA/coPP) multilayer films, PLA film (polylactide), and control (PVC cling film) during storage (reprinted with permission from [182]).

3.2.2. Polyhydroxybutyrate (PHB)

Polyhydroxybutyrate (PHB) is a well-known biodegradable poly- β -hydroxy alkanoate (PHA) synthesized by numerous bacteria as an internal carbon or energy reserve [177]. Nearly 75 different genera of bacteria can accumulate PHB, but *Ralstonia eutropha* has received the most attention because of its capacity to accumulate PHB in large quantities, and *Haloferax mediterranei*, *Bacillus megaterium*, and *Halomonas boliviensis* were also used for this purpose [188]. PHB is one of several biodegradable polymers that have reached commercial production and has a wide range of applications, from manufacturing packaging to more complex biomedical items [189]. This isotactic homopolyester has thermal and mechanical qualities similar to petrochemical polymers such as PP and PS and is biodegradable in various settings, including composting conditions and marine water. Due to their thermoplastic properties, homopolymers, polyhydroxy butyrate (PHB) and poly-3-hydroxybutyrate-co-3-hydroxy valerate (PHBV) copolymers, which are modified by copolymerization with hydroxy valerate, are used to develop packaging as alternatives to petroleum-based synthetic polymers [9]. Figure 16 shows the chemical structures of PHB and PHBV. Like other biopolymers, synthetic PHB also has biocompatibility, biodegradability, nontoxicity, thermoplasticity, and water barrier properties, making it widely available for packaging applications [177].

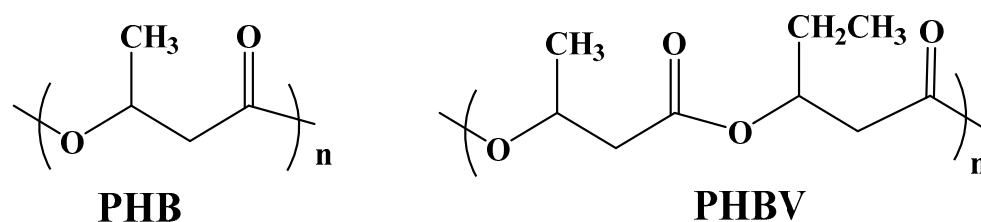


Figure 16. Chemical structures of PHB and PHBV.

As a pure homopolymer, PHB has a melting point of 180 °C, glass transition temperature of nearly 55 °C, and crystallinity above 50% [190]. Its high crystallinity and melting point makes it brittle and it has lower mechanical strength compared to petroleum-based plastics, restricting the usage of PHB for packaging applications [191]. The high production cost of PHB compared to petrochemical-based plastics due to the expenses of the substrate, fermentation, and downstream processes is another critical barrier to mass manufacturing and commercialization. Solution casting and extrusion methods are widely used to fabricate PHB-based packaging materials [54]. The drawbacks of PHB-based packaging materials can be overcome by using thermal treatments, developing copolymers, blending them with natural or synthetic polymers, and reinforcing them with natural fibers and inorganic fillers [192].

Recently, numerous studies have explored the potential of PHB-based films and coatings for food packaging applications. For example, Rech et al. developed a PHB-based film by incorporating nanosilica and clove essential oil as an active food packaging material for brown bread [193]. The prepared film extended the shelf-life of bread for up to 10 days compared to PE, with high antibacterial activity against *Escherichia coli*, *Aspergillus niger*, and *Staphylococcus aureus* [193]. Kumari et al. prepared PHB-based films enriched with different essential oils, including grapeseed, ginger, and bergamot oil, and characterized their mechanical, thermal, barrier, and antimicrobial properties [194]. The impact of methylcellulose- and chitosan-coated PLA-PHB film incorporated with olive leaf extract on preserving fresh pork burgers was examined by Fiorentini and coworkers [195]. The authors revealed that the shelf-life extension of the pork burgers was due to the reduction in lipid oxidation and the growth of *Enterobacteriaceae* [195]. Jiang et al. examined the mechanical and antioxidant properties of PLA-PHB-based film enriched with α -tocopherol to prolong the shelf-life of peach [196]. The results revealed that the film exhibited improved barrier, mechanical, and antioxidant properties and extended peach storage life by reducing malondialdehyde content and protecting cell wall structure [196]. Iglesias-Montes and coworkers successfully developed a film based on a PLA-PHB blend and chitin nanocomposite, suggesting its application in the food packaging industry [197]. Another study produced a PLA-PHB-based packaging film incorporating fennel oil and demonstrated its performance on the shelf-life of oysters [198]. The research confirmed that the developed film could prolong the shelf-life of oysters by 2–3 days due to its oxygen barrier properties and antioxidant and antibacterial ability [198]. Manikandan and colleagues developed PHB-based nanocomposites using graphene nanoparticles and examined the shelf-life extension of oxygen-sensitive foods [189]. Table 4 summarizes several recent studies on composite packaging materials, preparation methods, food types, mechanical and physical properties, and impacts on food quality.

Table 4. Applications of composite packaging materials in food packaging.

Packaging Material	Additives	Preparation Method	Food Sample	Properties of Packaging	Role as Food Packaging	Ref.
Potato starch-based film	Sodium Alginate Glycerol Essential oil	Casting method	Perishable food products	Water vapor transmission rate was 0.00254 g/m ² /h for the films.	Shelf-life extension and inhibition of the spoilage organisms <i>E. coli</i> and <i>B. cereus</i> .	[199]
Foxtail millet starch-based film	Clove leaf oil Sorbitol	Casting method	Cheese	Possessed ultraviolet light barrier properties, tensile strength (6.78–4.00 MPa), and elongation at break (66.26–99.48%) when the essential oil content was increased.	Reduced lipid oxidation and microbial growth compared to LDPE.	[200]
Corn, wheat, and rice starch-based coating	Chitosan	Coating method	Walnut	Thickness of starch films ranged between 0.19 ± 0.01 and 0.21 ± 0.02 mm. Water vapor permeability of films ranged from 20.63 ± 0.27 to 23.96 ± 0.25 g mm/m ² d kPa. Tensile strength ranged between 0.27 ± 0.04 and 0.89 ± 0.2 MPa.	Shelf-life extension due to reduced effects of oxygen, moisture, and temperature.	[201]
Chinese yam starch-based film	Sorbitol Glycerol Eugenol	Casting method	Pork	Plasticizer enhanced the mechanical strength and barrier to moisture and oxygen.	Due to its superior barrier and antibacterial qualities. increased the shelf-life of pork beyond 50%.	[202]
Cellulose nanofiber-based film	Zinc oxide nanorods Grapefruit seed extract	Casting method	-	Highly transparent, nanocomposite films with an enhanced vapor barrier (ranged from 0.46 ± 0.01 to 0.56 ± 0.02 × 10 ⁻⁹ g·m/m ² ·Pa·s) and UV blocking qualities.	Exhibited antimicrobial activity against food-borne pathogens and good antioxidant activity.	[203]
Chitosan-based coating	Glycerol	Coating method	Strawberry	-	Excellent antibacterial and antifungal activity for one week and maintained the appearance of strawberries.	[204]
Chitosan-based film	Apricot kernel essential oil	Casting method	Sliced bread	With addition of essential oil, water vapor transmission rate was decreased from 1394 ± 47 to 821 ± 31 g m ⁻² d ⁻¹ and tensile strength increased from 9.45 ± 0.53 to 19.36 ± 1.06 MPa.	Enhanced the shelf-life of bread, with antioxidant and antimicrobial activity against <i>E. coli</i> , <i>B. subtilis</i> , and fungal growth.	[205]

Table 4. Cont.

Packaging Material	Additives	Preparation Method	Food Sample	Properties of Packaging	Role as Food Packaging	Ref.
Chitosan-based films	Plant extracts obtained from oak, hop, and brown algae	Casting method	-	Blended films showed increasing moisture content (21.5–28.3%), total soluble matter (23.8–28.9%), and elongation at break (14.0–31.0%) for oak and algal extract-containing films but decreasing tensile strength (12.7 MPa–5.5 MPa) and Young's modulus (230.8 MPa–19.4 MPa)	-	[206]
Chitosan-based films	Pomegranate peel extract Glycerol	Casting method	Fruits and vegetables	Thickness (0.142–0.159 mm), tensile strength (32.45–35.23 MPa), opacity (0.039–0.061%), water barrier effect (1.32–1.60 g·mm/m ²), and gas barrier properties (93.81–103.45 meq/kg) of the films increased with increasing volume of pomegranate peel extract.	Extended storage life and improved quality.	[207]
Alginate-based films	Glycerol Aloe vera Frankincense oil	Casting method	Green capsicum	Mechanical properties and thermal stability were increased in the presence of aloe vera and frankincense oil. Water vapor permeability was decreased in the film containing aloe vera and oil from 21.53 ± 1.43 g mm/m ² day kPa for alginate to 8.18 ± 0.24 g mm/m ² day kPa.	Senescence retardation and resistance to the mass loss of green capsicums.	[208]
Alginate-based film	Glycerol Thymol	Two-stage cross-linking method	Fresh-cut apple	In comparison to sodium alginate films without thymol, thymol/sodium alginate composite films were shown to have poor water vapor permeability, water solubility, and swelling ratios but good tensile strength, elongation at break, and UV-vis light blocking capabilities.	Inhibited the growth of <i>Staphylococcus aureus</i> and <i>E. coli</i> and maintained apple weight, color, and appearance.	[209]
Alginate-based coating	Glycerol Thyme oil	Dipped method	Fresh-cut apple	-	Prevented bacteria growth, respiration, weight loss, and browning reaction while preserving firmness.	[210]

Table 4. Cont.

Packaging Material	Additives	Preparation Method	Food Sample	Properties of Packaging	Role as Food Packaging	Ref.
Alginate-based film	Kiwi peel extract Silver nanoparticles	Casting method	Cherry	Films exhibited high UV barrier qualities, water vapor resistance, and tensile strength.	Increased cherries' shelf-life by preventing moisture loss and protecting against microbial deterioration with strong antimicrobial and antioxidant properties.	[211]
Pectin-based film	Carvacrol Cinnamaldehyde	Casting method	Ham and bologna	Thickness of the films varied: apple films, from 0.128 to 0.135 mm; carrot films, from 0.041 to 0.049 mm; and hibiscus films, from 0.049 to 0.056 mm.	Improved microbial food safety by reducing the <i>L. monocytogenes</i> population with essential oil.	[212]
Pectin-based film	Glycerol Berry extract	Casting method	Salmon fillets	With the addition of berry extract, the thickness of the films was increased from 0.128 mm to 0.248 mm.	Improved shelf-life due to antioxidant and barrier properties.	[213]
Carrageenan-based film	Water extract of germinated fenugreek seeds Sorbitol	Casting method	Chicken breast	-	Improved the shelf-life of meat by controlling the growth of microorganisms on the surface of chicken breast.	[214]
Carrageenan-based film	ZnO nanoparticles Glycerol	Dipping method	Mango	Water vapor transmission rate of the film ranged from 65.88 ± 1.55 to $59.94 \pm 0.87 \text{ g m}^{-2} 24 \text{ h}^{-1}$, tensile strength ranged from 84.83 ± 4.67 to $121.53 \pm 6.57 \text{ MPa}$, and elongation ranged from 60.94 ± 6.03 to $65.91 \pm 2.49\%$ with the addition of ZnO.	Maintained firmness and delayed the discoloration and decay of mango.	[215]
PLA-PHB based films	Glycerol Cinnamaldehyde	Casting method	Salmon	PLA-PHB based film showed better tensile strength and excellent oxygen permeability rate compared to ethylene vinyl alcohol copolymer-based based film. Ethylene vinyl alcohol copolymer-based films had reduced water vapor transmission rates compared to PLA-PHB-based films.	Reduced the total bacterial count of the sample.	[216]
PLA-based film	Bergamot essential oils Nano-TiO ₂ Nano-Ag	Casting method	Mango	-	PLA nanocomposite films effectively extended the postharvest life and delayed the loss of mango firmness during the entire storage period.	[217]

Table 4. Cont.

Packaging Material	Additives	Preparation Method	Food Sample	Properties of Packaging	Role as Food Packaging	Ref.
PLA- and chitosan-caseinate-based film	Rosemary essential oil	Casting method	Fresh minced chicken breast	Elastic modulus of films ranged from 1133 ± 136 MPa for control sample to 2073 ± 89 MPa for chitosan- and oil-incorporated film. The tensile strength of the control film was 93 ± 9 MPa, whereas the value of the chitosan, caseinate, and essential oil-incorporated film was 160 ± 28 MPa.	Provided antioxidant effects and improved the shelf-life of fresh meat products.	[218]

4. Challenges and Future Perspectives of Biopackaging in the Food Industry

Consumer demand, advancements in industrial trends, environmental concerns, marketing methods, and customer lifestyles contribute to the growth of novel and inventive packaging strategies in the food sector [2]. Developing biodegradable packaging is an imperative step in the packaging industry toward reducing the environmental impact of conventional plastics and advancing sustainability objectives. The utilization of biodegradable and renewable materials is an excellent way to conserve the environment while adding economic value to neglected crops and industrial waste [219]. Despite the environmental friendliness of degradable packaging materials, several obstacles prevent their widespread application in food packaging. A major issue is the biodegradability via thermal, photo-induced, and chemical degradation. Major reasons for biodegradation are weak heat resistance, mechanical strength, and moisture and gas barrier properties. This reduces the shelf life of foods and renders materials less appropriate for several packaging applications [220]. Furthermore, certain biopolymers have low processability, demonstrating poor melt rheology and heat sensitivity, which poses difficulties for traditional polymer processing. Commercialization efforts are additionally impeded by insufficient legal requirements and a lack of regulations for biopackaging [221]. The efficacy of biopolymer packaging varies widely based on the product, and stability is highly affected by storage conditions like humidity and temperature. In order to overcome these constraints, several modifications have been introduced. For instance, the mechanical strength and barrier qualities of biodegradable materials can be enhanced by blending different polymers and using different modification techniques [222]. Biopolymers can be modified using various chemical, physical, and enzymatic methods to alter the physical and chemical properties of biopolymers for different applications [223]. Optimizing manufacturing procedures and exploring relatively abundant and low-cost biopolymers are important tactics for cutting costs in producing biodegradable packaging materials. Standardizing biodegradation testing methods and introducing new regulations and policies ensure consistent, reliable, and sustainable packaging solutions.

Currently, the improvement of primary biopackaging materials into active packaging materials incorporated with different active compounds such as antioxidant, antimicrobial, and anti-browning agents; colorants; flavors; vitamins; and enzymes is an exciting and rapidly evolving area of the food packaging industry to extend the shelf-life of foods and maintain their quality [224]. To gain optimal material properties for functional packaging films in particular applications, the blend of these various additives must be carefully examined [225]. Furthermore, O_2 scavengers, CO_2 releasers, ethylene scavengers, moisture absorbers, UV barriers, and antimicrobial packaging systems are common active packaging systems tested in the food industry [226]. In addition, intelligent food packaging has recently emerged as a novel technique to improve the functionality of biopackaging. Intelligent food packaging, also known as smart packaging, is a novel solution that utilizes

indicators and sensors incorporated in the packaging to monitor and report changes in food quality and safety. These indicators and sensors can detect various changes in the food system, including microbial activity and chemical and physical changes [227]. Some intelligent packages observe the real-time conditions of food and some provide a source for releasing bioactive agents during food spoilage [228].

Further, nanotechnology-integrated biopolymer-based food packaging can be developed by addressing the current issues with the mechanical, thermal, and barrier properties of biopolymers. The use of nanotechnology in biopackaging entails the production of bionanocomposites at the nanoscale or using nanoparticles to improve the properties of packaging material. Bionanocomposites are sophisticated materials composed of biodegradable polymers and nanoscale reinforcing elements with better mechanical, thermal, and barrier qualities than typical biodegradable polymers [229]. Future applications of bionanocomposites would rise with the development of smart and intelligent packaging as a new technology, incorporating novel programmable, artificial intelligence (AI)-based materials into bioplastics.

5. Conclusions

Economic, health, and environmental concerns associated with conventional packaging materials such as plastic, metal, paper, and glass have shifted consumers toward sustainable food packaging. One of the emerging solutions for sustainable food packaging is biopolymer-based packages that eliminate common waste and aid in minimizing the adverse effects of conventional packages on the environment. Biopackaging has been widely used in the food industry due to its degradability, renewability, nontoxicity, and edibility. The main biopolymers used to develop biodegradable food packaging are polysaccharides, which include starch, cellulose, chitosan, alginate, pectin, and carrageenan. Polysaccharides show excellent mechanical properties with superior barrier properties against O₂, CO₂, oil, and aromas while demonstrating poor moisture resistance. Moreover, synthetic aliphatic polyesters also exhibit desirable mechanical and barrier properties for oils and aromas. This review discussed the structure, properties, and recent developments in food packaging applications of polysaccharide-, including starch, cellulose, chitosan, alginate, pectin, and carrageenan, and aliphatic polyester-based polylactic (PLA) and polyhydroxy-butylate (PHB) biopackaging materials.

Although most of these biopolymers have poor mechanical and physical structures, numerous studies have demonstrated that polymer blends and composites have drastically strengthened structures and other properties. Also, biopackaging is vital in packaging perishable fruits; vegetables; meat, poultry, and fish products; cereals; bakery products; dairy products; and oil-fried products. Current trends in biopackaging include active packaging, intelligent packaging, edible coating and films, and bionanocomposites and blends. Nevertheless, despite these innovations, concerns related to shelf-life, finances, customer perception, and socioeconomics directly impact the replacement of conventional packaging, hindering biopolymers from being broadly commercialized in food packaging.

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