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REVIEW ARTICLE

Biodegradable Films for Fruits and Vegetables Packaging Application: Preparation and Properties

H. P. S. Abdul Khalil[1] · Aparajita Banerjee[2] · Chaturbhuj K. Saurabh[1] · Y. Y. Tye[1] · A. B. Suriani[3] · A. Mohamed[3] · A. A. Karim[1] · Samsul Rizal[4] · M. T. Paridah[5]

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Abstract
Fresh-cut fruits and vegetables are highly perishable commodities. Packaging forms an important tool to maintain the shelf life of packed fresh-cut agricultural produce. Petroleum-based films are conventionally used for fresh fruits and vegetables. However, being nonbiodegradable and derivability from nonrenewable resources, these films lead towards serious ecological problems. To address this issue, various efforts have been focused on renewable and biodegradable films obtained from biopolymers. Widely studied biopolymers for film preparation are derived from biomass (gelatin, starch, cellulose, etc.), microbes (polyhydroxyalkanoates), and bio-derived monomers (polylactic acid). However, such films possess poor mechanical and barrier properties as compared to their commercial counterparts. Incorporation of various additives has been proposed to improve the film characteristics. In the present review, comprehensive information has been provided on different methodologies for film fabrication, properties, and applications of stand-alone bio-based films for packaging of fresh-cut produce. Furthermore, successful commercial implementation of such film is also summarized.

Keywords Biodegradable stand-alone films · Minimally processed food products · Shelf life · Packaging film properties

Introduction

The modern era has experienced a tremendous increase in the sale of minimally processed (MP) fresh-cut agricultural products. These products are ready-to-cook (RTC) or ready-to-eat (RTE) type which provides various advantages to consumers. However, suitable packaging is indispensable to increase the shelf life of packed products. Conventionally, nonbiodegradable plastics were the common choice for packaging materials owing to their easy processability, lower price, and high resistance to chemical and mechanical stress [1]. Petrochemical-based plastics such as polystyrene, polypropylene, and polyvinyl chloride have a good mechanical and barrier characteristics like tensile and tear strength, oxygen permeability, carbon dioxide permeability, and aroma transmission. Nevertheless, such polymers impose major environmental constraints due to their poor recyclability, derivability from nonrenewable resources, and non-biodegradability, thus leading to serious ecological problems [1]. Owing to such limitations, in the last few years, research has been focused on obtaining packaging materials from renewable resources and in this regard, biopolymers have gained increasing attention. Packaging biopolymers are highly safe due to the absence of harmful chemicals or toxins; furthermore, being biodegradable, they break down into harmless products that get absorbed into the soil [2]. Over the last two decades, the development and use of biopolymer-based packaging materials to extend the shelf life of fresh products has been receiving increased attention. However, researchers have been exploring the utilities of bio-based films for more than 100 years. A maize kernel protein

✉ H. P. S. Abdul Khalil
akhalilhps@gmail.com

1 School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

2 Department of Biotechnology & Microbiology, Tilak College of Science & Commerce, University of Mumbai, Mumbai 400032, India

3 Nanotechnology Research Centre, Faculty of Science and Mathematics, Universitii Pendidikan Sultan Idris, 35900 Tanjung Malim, Perak, Malaysia

4 Mechanical Engineering Department, Syiah Kuala University, Banda Ache 23111, Indonesia

5 Institute of Tropical Forestry & Forest Products, Universiti Putra Malaysia, 43400 Serdang, Malaysia

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film was developed by Osborne [3] in 1897. A patent has been filed in 1972 for the fabrication of wheat gluten-based film [4]. In the year 2000, films were prepared from apple puree for fresh-cut apples [5]. Thereafter, there have been various studies done for the development of biodegradable packaging for food applications. Table 1 presents the timeline of the development of biopolymer-based packaging for fresh-cut fruits and vegetables.

Biopolymers are broadly divided into three classes based on their source: polymers formed by a chemical process using renewable monomers as precursors such as polylactic acid, polymers such as polyhydroxyalkanoates synthesized from microorganisms, and polymers directly extracted from plant or animal biomass including polysaccharides and proteins (Fig. 1) [18]. For example, guar gum is a heteropolysaccharide having a mannose backbone which is obtained from the endosperm of an annual legume plant *Cyamopsis tetragonoloba*. In spite of the natural abundance of biopolymers, their usage in food packaging is limited due to their relatively poor mechanical and barrier properties as compared to conventional petrochemical-based packaging materials. Hence, various physical and chemical methods like thermal treatment, gamma irradiation, chemical modification, and incorporation of various additives such as nanoclay and plasticizer have been proposed in the past to overcome the limitation of biopolymer-based packaging [19].

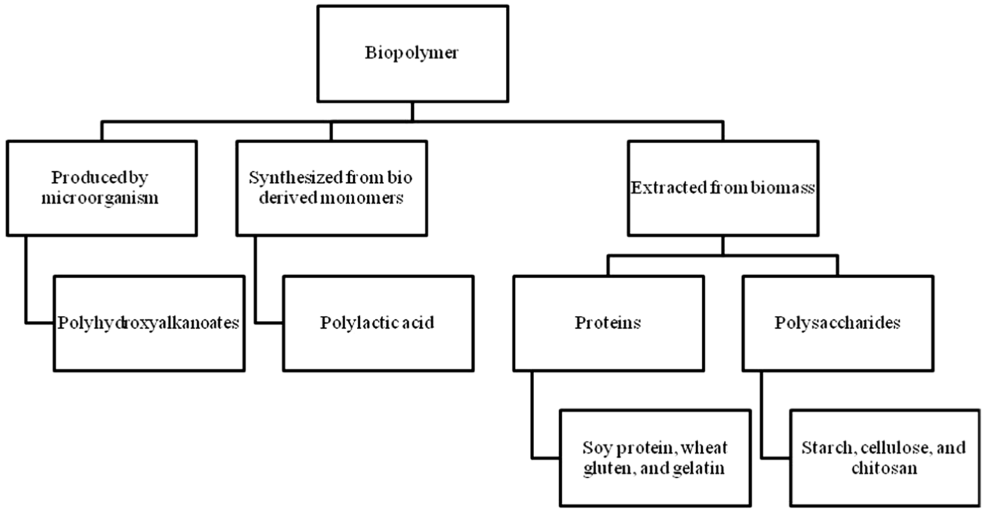
Protein networks can easily achieve strong intermolecular covalent bonds, close molecular packing, and reduced polymer mobility by means of cross-linking using thermal treatment. This was illustrated by Liu et al. [20] wherein peanut protein films made from thermally treated solution (70 °C for 30 min) were demonstrated to have improved tensile strength as compared to the untreated films. Functional properties of films can also be improved by chemical modification of the biomolecules. For instance, percentage elongation of the soy protein-based film increased significantly when mildly treated with alkali [21]. Physical treatment like the exposure of biopolymers to ionizing radiation can cause ordering of the polymeric chains which resulted in improved mechanical strength as observed by Saurabh et al. [1] for guar gum-based films when guar gum powder was exposed to a dose of 500 Gy. The elasticity of film is another vital property for packaging applications, and it can be improved by the addition of plasticizer like glycerol. Thus, various methods are available to enhance functional characteristics of the biodegradable film. Such modified or treated films can be suitably used as food packaging material for shelf life extension of food products which subsequently minimizes the usage of conventional plastics.

Edible coating and stand-alone film are two types of packaging generally used for consumable products. Edible coatings are an integral part of the food by forming an immediate coating on its surface and subsequently get consumed along with the coated product. However, the stand-alone packaging does not integrate with food surface and withstand independently. It is most commonly used as a food wrap and is not consumed along with the product. While edible coatings are well reviewed, little emphasis is given on biodegradable free-standing films. Consequently, there is a need for proper assessment on stand-alone biodegradable packaging for fresh-cut agricultural products, to sum up, the current trends and

Table 1 Chronological event of biopolymers' film usage in fresh-cut agricultural produce

Biopolymer	Properties/packaging applications	Year	References
Corn protein	Transparent films	1897	Osborne [3]
Zein	Packaging film	1941	Swallen [6]
Wheat gluten	Strong and flexible film have numerous uses particularly in the food	1972	Ann [4]
Corn, wheat, and soy proteins	Edible films	1988	Aydt et al. [7]
Corn and wheat protein	Enhancing shelf life of fresh foods	1991	Aydt et al. [8]
Chitosan, cellulose, and polycaprolactone	Modified atmosphere packaging of head lettuce, cut broccoli, whole broccoli, tomatoes, sweet corn, and blueberries	1997	Makino et al. [9]
Apple puree, fatty acids, fatty alcohols, beeswax, and vegetable oil	Fresh-cut apples	2000	McHugh et al. [5]
Zein films plasticized with oleic acid	Fresh broccoli	2001	Rakotonirainy et al. [10]
Polylactic acid	Green peppers	2007	Koide et al. [11]
Polylactide	Blueberries	2008	Almenar et al. [12]
Banana flour and chitosan	Asparagus, baby corn, and Chinese cabbage	2011	Pitak et al. [13]
Methylcellulose, polycaprolactone, and alginate	Broccoli	2013	Takala et al. [14]
Polylactic acid	Fresh-cut celery	2014	González-Buesa et al. [15]
Polylactic acid	Fresh-cut and cooked spinach	2015	Botondi et al. [16]
Polylactic acid	Fresh-cut melon	2016	Huijuan et al. [17]

Fig. 1 Different classes of biopolymer used for food packaging



future prospect. The main aim of this review is to effectively summarize different types of biopolymer-based packaging which have been studied to increase the shelf life of fresh-cut agricultural products. Special emphasis is given on the process of film formation, their properties, and subsequent application.

Importance of Packaging in Fresh-Cut Fruits and Vegetables

Fruits and vegetables are essential components of a healthy diet. The nutritional value of vegetables lies in their micronutrient content, fiber content, and bioactive phytochemicals. Micronutrients in vegetables generally comprised of vitamins and minerals which are required by humans in small quantities to orchestrate a range of physiological functions. Although these micronutrients are required in trace amounts, yet their deficiency causes a number of diseases in human. The USDA dietary guidelines recommend us to consume at least three to five servings of vegetables daily [22]. A daily intake of 400 g of fruit and vegetables is highly advised [23].

Owing to the convenience of minimum preparation time before consumption of minimally processed fruits and vegetables, they are gaining importance in food retail establishments [24]. Fresh agricultural produce has been processed into ready-to-eat (RTE) or ready-to-cook (RTC) products to increase their convenience without significantly altering their fresh-like characteristics [25]. The market for minimally processed fruits and vegetables is on the rise in Europe in last three decades. Furthermore, similarly in the USA, packaged salads and fresh-cut vegetables are among the fastest selling items in grocery stores [26]. In Asia, the fresh-cut produce market holds a major share of all products sold in the retail market [27]. The International Fresh-cut Produce Association (IFPA) defined fresh-cut produce as: "Trimmed, peeled, washed, and cut into 100% usable product that is subsequently bagged or pre-packaged to offer consumers high nutrition, convenience, and value while still maintaining freshness".

Mainly minimal processing operations ("mild technology") include peeling, cutting, washing, treatments with sanitizing agents, drying, etc. These processing operations alter the physical integrity of these products, making them more susceptible to spoilage than the original raw materials [28, 29]. Injury to fresh produce during processing operations renders them vulnerable to contamination via growth/survival of spoilage or pathogenic bacteria. About 80–90% of food pathogens are gram negative which predominantly include *Pseudomonas*, *Enterobacter*, or *Erwinia* species. Yeasts commonly include *Cryptococcus*, *Rhodotorula*, *Candida*, *Aureobasidium*, *Fusarium*, *Mucor*, *Phoma*, *Rhizopus*, and *Penicillium*. Pathogenic bacteria may include *Listeria monocytogenes*, *Clostridium botulinum*, *Aeromonas hydrophila*, *Escherichia coli*, *Salmonella*, *Yersinia enterocolitica*, and *Campylobacter jejuni* [30].

The industrial process accelerates the degradation of the minimally processed agricultural products and leads to biochemical changes such as an increase in respiration rate which speeds up the oxidation processes, degradation of cell membranes and enzymatic browning, and loss of tissue texture. Overall preservation of sensory, nutritional, and microbial qualities of fresh-cut produce is a major challenge for fresh-cut food industries. Minimally processed agricultural produce can be preserved by using techniques such as refrigeration, preservatives, mild heat treatments, microwave processing, diminution of water activity, ionizing radiation, high-pressure technology, high intensity pulsed electric field, pulsed light disinfectants, ozone technology, hurdle technology (an intelligent combination of different preservation techniques which secures microbial and sensory qualities of the product), etc. [31]. However, an adequate packaging is indispensable for the maintenance of the overall quality of processed products. While petroleum-based packaging has been used as packaging material for a considerable time, the modern era is experiencing an aversion towards its use owing to environmental concern. Furthermore, being in direct contact with the product, the conventional food packaging materials can be a source of contaminants by migration of chemicals from packaging into the food. Hence, more emphasis is given on the use of biopolymers-based food packaging. Much work has been done for the development of stand-alone food packaging from biomass which is further commercialized in food industries.

Biodegradable Polymers: Sources

Biodegradable films can be divided into three categories as per their components: hydrocolloids, lipids, and composites. Hydrocolloids are hydrophilic polymers of natural (including proteins and polysaccharides) or synthetic origin that generally forms a gel in water due to the presence of many hydroxyl groups; thus, they have an excellent film-forming ability. Lipids are broadly defined as hydrophobic or amphiphilic molecules which include waxes, acylglycerols, and fatty acids. Composites are made up of two or more constituent (hydrocolloids and/or lipids) to produce an amalgamation with improved characteristics than that of the individual components. Extensive research has been done on proteins and polysaccharides film owing to their suitable physicomechanical properties. Such films are able to maintain aerobic condition around packed products, thus extending their shelf life [32].

Polysaccharides

Polysaccharides are long chains of monosaccharide units linked together by glycosidic bonds. Owing to simple processing and wide availability, significant research has been done on carbohydrate-based biopolymer than proteins for the development of biodegradable packaging.

Starch

Starch (polymer of glucose) is a storage compound produced by most green plants consisting of linear amylose and branched amylopectin units. It is most commonly used for the production of films because it is cheap, plentiful, biodegradable, and easy to handle [33]. In fact, the first commercial "biodegradable" film was developed from a mixture of granular starch and synthetic polymers [34]. Thermoplastic starch can be produced by using an extruder employing both thermal and mechanical energy on starch. Thermoplastic starch is commercialized due to its excellent properties, and today, it dominates bio-based compostable material market [35]. Starch-based films have moderate gas barrier properties which can be improved by restructuring the native conformation of the starch and by adding synthetic substances. Various efforts for the development of starch-based films have been reported in the past [36, 37]. Starch when mixed with glycerol as plasticizer, the resultant films enhanced the shelf life of strawberries from 14 to 21 days [38]. Furthermore, in the above studies, glycerol content had a significant effect on mechanical properties of films. Puncture strength decreased from 14.6 to 12.1 N with the increase in glycerol content from 1.3 to 2 g/100 g of solution. Han et al. [39] studied the effect of beeswax as a plasticizer in pea starch-based edible films. Developed films were shown to have low water vapor permeability due to the hydrophobic component of beeswax. Water vapor permeability decreased from 7.78 to 6.56 g mm $m^{-2}h^{-1}kP^{a-1}$ with beeswax concentration from 0 to 40% (*w*/*w* of pea starch). Gamma irradiation has been widely used for the improvement of mechanical properties of starch-based edible films by radiation-induced cross-linking between polymeric chains. A similar effect of irradiation on enhancing the functional properties of the starch-based film was earlier observed by Kim et al. [40]. The effect of commonly used plasticizers like glycerol and polyethylene glycol, along with glutaraldehyde or calcium chloride as a cross-linking agent on mechanical properties and water vapor transmission rate of cassava starch was also studied [33]. Table 2 shows the studies conducted on the shelf life extension of fresh-cut fruits and vegetables using biopolymer including starch-based biodegradable packaging.

Chitosan

Chitosan is a linear polysaccharide commercially produced by alkali deacetylation of chitin. It is generally a copolymer comprised of D-glucosamine along with N-acetyl-D-glucosamine [47]. The unique cationic nature possessed by chitosan relative to other neutral or negatively charged polysaccharides enables it to impart strong and wide spectrum antimicrobial properties [48, 49]. Several models have been projected for antimicrobial activity of chitosan but the most acceptable is electrostatic interactions between the positively charged amino groups (NH_3+) at pH values lower than 6.3 (the pKa of chitosan) and the negatively charged surface of bacteria. This electrostatic interaction results in twofold interference: (i) by altering the permeability of the membrane wall consequently aggravating internal osmotic imbalances, and therefore, preventing microorganisms' propagation, and (ii) by peptidoglycans hydrolysis in the cell wall of microbes, leading to the seepage of intracellular electrolytes, for instance, potassium ions and other low molecular weight proteinaceous fractions including proteins, nucleic acids, glucose, and lactate dehydrogenase [50]. Corroborating this proposition, Fernandez-Saiz et al. (2008) recently demonstrated that the release of soluble protonated glucosamine fractions from chitosan was responsible for its antimicrobial activity [49].

Table 2 Research on biopolymer-based film on shelf life extension of fresh agricultural produce

Polymer	Claim	References
Master-Bi® bag (based on starch)	Quality of packed tomatoes in biodegradable packages was similar to the quality of tomatoes stored in low-density polyethylene bags for 3 weeks	Kantola et al. [41]
Yam starch and glycerol	Shelf life of fresh strawberries was 14 days and for samples packed in starch film it was 21 days	Mali et al. [38]
Banana starch and chitosan	Developed composite bags to protect asparagus, baby corn, and Chinese cabbage against *Staphylococcus aureus*	Pitak et al. [13]
Chitosan and Na-caseinate	Antimicrobial effect against fungi and yeast when developed film applied on carrot slices	Moreira et al. [42]
Chitosan, methyl cellulose, and vanillin	Reduction of four logs in microbial population in fresh-cut pineapple on day 6	Sangsuwan et al. [43]
Carboxymethyl cellulose containing potassium sorbate	Strong activity against *Aspergillus* species in fresh pistachios	Sayanjali et al. [44]
Methylcellulose and polycaprolactone/alginate films incorporated with antimicrobial agents	Films controlled *Salmonella typhimurium* growth for 12 days and *L. monocytogenes* and *E. coli* growth for 4 days on broccoli florets when stored at 4 °C	Takala et al. [14]
Polylactic acid with Allium extract	Developed film was found to be effective for ready-to-eat salads up to 5 days of storage at 4 °C	Llana-Ruiz-Cabello et al. [45]
Wheat gluten	Extend shelf life of strawberries by 12 days at 7–10 °C	Tanada-Palmu et al. [46]
Zein films plasticized with oleic acid	Extend shelf life of fresh broccoli florets by 6 days	Rakotonirainy et al. [10]
Apple puree with fatty acids, fatty alcohols, beeswax, and vegetable oil	Reduced moisture loss and browning in fresh-cut apples for 12 days at 5 °C	McHugh et al. [5]

Chitosan-based films and coatings have excellent mechanical properties and selective permeability of carbon dioxide and oxygen gasses. However, high water vapor permeability of chitosan films limited its applications [51, 52]. Chitosan along with other biopolymers are studied in the past to overcome poor barrier properties [49]. Maize starch/chitosan-based composites demonstrated improved elasticity and water vapor permeability as compared to film fabricated with either of the biopolymers [53]. Chitosan/starch film-forming suspensions and chitosan solution both demonstrated a pseudoplastic behavior. Water vapor permeability of starch film was 13.2 to 21.2×10^{-11}, for chitosan film it was 4.5×10^{-11} g $m^{-1}s^{-1}Pa^{-1}$, and for chitosan/starch film it was $3.8–4.5 \times 10^{-11}$ g $m^{-1}s^{-1}Pa^{-1}$. The observed improvement in barrier properties was due to the interactions between hydroxyl groups of starch and amino groups of chitosan. In another set of study, an edible film with innovative characteristics was obtained by combining the antimicrobial properties of chitosan with the property of self-sealing banana (variety Kluai Namwa) starch [13]. The presence of starch makes composite film water soluble and sealable, thus wrapping of food products is easier, while the presence of chitosan gives an antimicrobial property to film, thus extending the shelf life of packed products. Such composite bags could protect the asparagus, baby corn, and Chinese cabbage against *Staphylococcus aureus* spoilage and consequent food poisoning [13]. Chitosan-based composite coating formulations, as well as films, have been shown to prolong the shelf life of a wide variety of food products like banana, mango, and capsicum due to its antimicrobial property [54]. The conduct of biopolymers like chitosan-based biodegradable packaging films on shelf life extension of fresh-cut fruits and vegetables has been demonstrated in Table 2. Application of chitosan induces the production of plant defense enzymes such as chitinase which protect it from fungal and invertebrate attack. A composite formulation based on derivatized chitosan (Nutri-Save) is used for the shelf life extension of fresh produce [2]. Biodegradable films incorporated with an antimicrobial agent can be used in food packaging to reduce microbial loads, thus extending the shelf life of fruit and vegetables. However, being inherently antimicrobial, chitosan-based films prolonged the shelf life of packed products without any antimicrobial agents.

Cellulose

Cellulose consists of a linear chain of β (1 → 4) linked D-glucose units. A number of cellulose derivatives are used for the development of films with improved properties as compared to native cellulose films. Cellulose acetate films are most widely used in food packaging than other derivatives due to its improved gas and moisture barrier properties [55]. Extensive work has been done on chitosan-cellulose composite films. The cellulose and chitosan form homogeneous composite films due to similarities in their primary structures which resulted in excellent functional properties. Möller et al. (2004) reported that 1% of chitosan in chitosan-HPMC (hydroxypropyl methylcellulose) composite films were effective against *L. monocytogenes* [56]. Chitosan/methyl cellulose-based composite film demonstrated antimicrobial activity against *E. coli* on fresh-cut cantaloupe. The abovementioned film also effectively reduced the viable count of *Saccharomyces cerevisiae* yeast inoculated on cantaloupe and pineapple [43]. Furthermore, the water vapor transmission rate of native HPMC film was around 270 g m^{-2} day^{-1} atm^{-1} and by addition of chitosan along with cross-linking agent, it was reduced to 40% of its initial value. García et al. (1998) demonstrated the use of cellulosic films for wrapping and maintaining the qualities of strawberries [57]. Such films proved to be effective for enhancing the shelf life of packed samples by maintaining an optimum atmosphere inside packets. Although cellulose possesses excellent film properties, however, due to its high cost, it is not feasible to use them for the preparation of biodegradable films on a commercial scale. Table 2 shows the studied conduct on the shelf life extension of fresh-cut fruits and vegetables using cellulose-based films.

Alginate

Alginate is extracted from seaweed and it is mainly composed of a linear copolymer of D-mannuronic and L-guluronic acid monomers. It possesses good film-forming properties that make alginate predominantly valuable in food applications [58]. Divalent cations like Ca^{+2}, Mg^{+2}, Mn^{+2}, or Fe^{+2} are used as gelling agents in alginate film preparation. Calcium is more efficient in gelling of alginate as compared to other cations. Properties of alginate films can be further modified by incorporating starch, oligosaccharides, or simple sugar [58]. There are various reports on the incorporation of antioxidants in alginate films and its subsequent effect on the shelf life extension of fresh produce. Norajit et al. (2010) investigated the physical and antioxidant properties of alginate film incorporated with ginseng extracts for packaging applications [59]. Water vapor permeability of pure alginate film was $1.72 \pm 0.07 \times 10^{-7}$ g•m^{-2}•day•Pa and tensile strength was 22.2 ± 4 MPa; with the addition of ginseng extract, it changed to $2.28 \pm 0.13 \times 10^{-7}$ g•m^{-2}•day•Pa and 10.76 ± 1.77 MPa, respectively. The observed increase in permeability was insignificant, thus incorporation of ginseng extract did not affect the barrier property of alginate film. Active films were also developed by using methylcellulose, polycaprolactone, and alginate along with antimicrobial agents including acetic acid, rosemary extract, and Asian spice essential oil [14]. The obtained film effectively controlled *L. monocytogenes* and *E. coli* growth during the first 4 days and *Salmonella typhimurium* growth for 12 days on fresh broccoli stored at 4 °C.

Polylactic Acid

Polylactic acid or polylactide (PLA) is a thermoplastic aliphatic polyester derived from renewable material like corn, tapioca roots, and sugarcane. PLA is classified as GRAS (Generally Recognized As Safe) by the US Food and Drug Administration and authorized by the European Commission to be used in contact with food and for these reasons it is an excellent material for fabricating biodegradable packaging [60]. New food packaging systems were developed by using PLA containing *Allium* spp. extract for ready-to-eat salads [45]. The developed film was found to be effective up to 5 days for storage of salads at 4 °C. PLA-based stand-alone film also used for extending shelf life of various agricultural products including blueberries [12], celery [15], spinach [16], and melon [17].

Polycaprolactone

Polycaprolactone (PCL) is a type of biodegradable polyester, prepared by ring-opening polymerization of ε-caprolactone. A laminate was produced by gluing a chitosan-cellulose film to a PCL film by Makino et al. [9]. The obtained biodegradable laminate was found to be suitable for packaging and storage of cabbage, broccoli, tomatoes, sweet corn, and blueberries. Different types of PCL-based composites were prepared for the shelf life extension of fresh broccoli [14, 61].

Protein

Proteins are the structural and functional components of plant and animal cells made up of amino acids linked together by peptide bonds. Proteins from both plant and animal sources have been widely reported for films preparation. Commonly used plant proteins are soy protein, wheat gluten, corn zein, and whey protein whereas gelatin, collagen, and keratin are commonly studied animal proteins for fabrication of film. However, considerations regarding food allergy, wheat gluten intolerance (celiac disease), milk protein intolerance, and religious beliefs/banning must be taken into account when protein-based films are used for food packaging.

Wheat gluten-based films extended the shelf life of refrigerated strawberry to 12 days at 7–10 °C [46]. Authors also concluded that the gluten film was more promising for controlling decay than the gluten-based coatings. In a different study, zein films plasticized with oleic acid was proposed for extending the shelf life of broccoli [14]. Fresh broccoli florets preserved their original firmness and color after 6 days of storage at low temperature when packed in glass jars sealed with zein films. Besides, fresh-cut fruits and vegetables biopolymer-based film were also studied for the shelf life extension of seafood, poultry products, and bakery. Composite films were prepared from barley bran protein and gelatin containing grapefruit seed extract by Song et al. (2012) [62]. Incorporation of 0.5% of grape seed extract decreased the tensile strength of composite film from 38.17 ± 3.25 to 31.41 ± 0.08 MPa as compared to film with no seed extract.

However, after 15 days of storage, populations of *E. coli* O157:H7 and *L. monocytogenes* inoculated on salmon packed with the composite active film decreased compared to the control film without grapefruit seed extract. There are few reports available on protein-based stand-alone film on fresh agricultural produce. Future research can be focused on protein film for fresh-cut produce owing to their excellent mechanical and barrier properties.

Lipids

Lipid coatings and films are mainly used for their hydrophobic properties, representing a good barrier to moisture loss. Waxes, fatty acids, and acylglycerols are the commonly used lipids for film formation. However, owing to its lack of structural integrity, lipids form very weak films. The poor mechanical properties of such lipid films can be overcome when they mix with hydrophilic materials like polysaccharides to form emulsion film or bilayer films. The bilayer film is mainly composed of hydrocolloid film laminated with the lipid layer. However, simply adding hydrophobic materials in film formulation cannot improve the barrier properties of an edible film against moisture, unless a homogeneous lipid dispersion inside the hydrocolloid matrix is achieved [63]. Through a careful selection of the types of fatty acid based on its chain length and nature of derivatization as well as its concentration, a composite formulation with particular characteristics can be tailor-made for a specific application. A mixture composed of hydrocolloids, emulsifiers, and lipid molecules demonstrated shelf life extension of several fruits and vegetables [64]. McHugh and Senesi (2011) developed biodegradable films from apple puree with different amounts of fatty acids, fatty alcohols, beeswax, and vegetable oil for wrapping of sliced apples [5]. Such wraps significantly reduced moisture loss and browning in fresh-cut apples up to 12 days at 5 °C [65]. There are various successful examples of biopolymer-based packaging films for the shelf life extension of fresh-cut fruits and vegetables (Table 3).

Film Preparation Methods

For film formation, biopolymers are dissolved in appropriate film formation solution followed by casting over a suitable base. Selection of a base material is important in the film-forming process because good casting plates help in easy removal of films without any tears and wrinkles. Teflon and glass plates have been widely used as casting surface because dried films could be easily peeled off (stripped) from these plates [69]. Saurabh (2015) demonstrated that for guar gum films, a glass plate was suitable for casting when compared with Teflon plates as shown in Fig. 2 [67]. Guar gum films casted on glass plate were more transparent than the films casted on Teflon plates.

During fabrication of films, components including biopolymer along with other additives must be dispersed and dissolved in a suitable solvent especially water. Plasticizers, antimicrobial agents, and nanoparticles can be added in this process to impart enhanced elasticity, antimicrobial activity, and mechanical strength, respectively, to films. Regulating pH and/or heating the film-forming solutions may be done for some types of polymer especially protein to facilitate their dispersion and induction of cross-linking [68]. The formation of films generally involve inter- and intramolecular associations or cross-linking of polymer chains which resulted in the formation of semi-rigid 3D networks. The degree of cohesion depends on polymer structure, solvent, temperature, and additives like plasticizers, nanoparticles, cross-linking agents (glutaraldehyde), etc. Next step involves removal of solvent used for dissolution of the film-forming components. This is done by drying at a desired temperature and relative humidity to obtain free-standing films with suitable mechanical and barrier properties. Rate and temperature of drying have been demonstrated to influence the mechanical properties of films. Drying of films is quick when infrared rays are used; thus, they have an advantage over conventional oven drying [70]. An optimum moisture level of 5 to 8% is desirable in the dried film for its easy peeling from casting plates. Finally, conditioning of biopolymer-based film, i.e., storage at relative humidity, is necessary to bring all the samples at the required moisture content for mechanical and barrier properties analysis since bio-based films are sensitive to its moisture content [1]. The solvent casting method is one of the most frequently used methods for preparation of biopolymer-based biodegradable films [71]. General steps involved in the preparation of biodegradable stand-alone films are demonstrated in Fig. 3. However, the film fabrication process changes with the change in components of the film. Prior to the addition of a hydrophobic component like essential oil in the biopolymeric solution, it is usually mixed with an emulsifier like Tween-80 to facilitate the compatibility between hydrophobic component and matrix. The resultant solution is homogenized at high speed for production of a homogeneous and smooth film. This method led to the production of emulsion films; such film is usually weaker in mechanical strength as compared to native film without oil. Beeswax or other solidified components whose melting points are less than the boiling point of the solvent can be added to the film-forming solution. Further processing of the obtained solution like mixing, casting, and drying should be done above melting temperature of the solidified components. This method is only limited to incorporation of such material whose melting point is lower than the boiling point of the solvent. Another problem during film preparation is foam formation which is more in protein solution as compared to polysaccharide solution; thus, degassing

Table 3 Different types of biodegradable films for the packaging of fresh-cut agricultural produce

Biopolymers	Packed product image	Reference
Zein and polycaprolactone	Carrots 	Mensitieri *et al.* [65]
Polyester based biodegradable films	Lettuce 	Del Nobile *et al.* [66]
Guar gum, beeswax, grape pomace and nanoclay	Pomegranate 	Saurabh [67]
Oriented poly(lactide)	Mango 	Chonhenchob *et al.* [68]
Oriented poly(lactide)	Pineapple 	Chonhenchob *et al.* [68]

is an important step before casting of protein suspension for film formation. Removal of bubbles can be done by employing a vacuum pump. In preparation of nanoclay incorporated film, nanoclay is intercalated or exfoliated in a suitable solvent prior to its addition in biopolymeric suspension. If numbers of additives are more than two and they have cumulative effects on various properties of films, then various optimizing tools like Response Surface Methodology is used. The main principle of RSM is to relate film properties of regression equations that express interrelations between additive concentrations and film's characteristics. It was earlier used by various authors consecutively to understand the individual and interactive effects of additives' content on film properties [72]. Overall, it can be concluded that the solvent casting method with little modification is frequently used for the preparation of films including additive pretreatment like

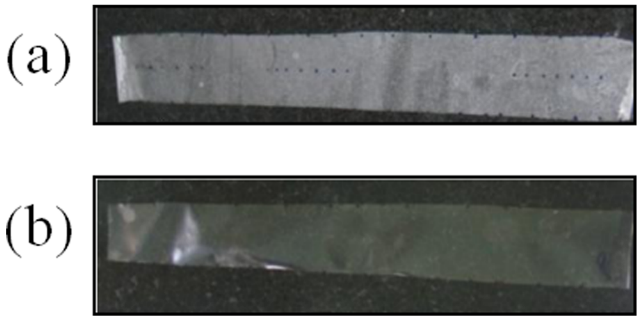


Fig. 2 Guar gum-based film casted on **a** Teflon plates and **b** glass plates [67]

intercalation of nanoclay, emulsification of hydrophobic components, degassing, etc.

Properties and Additives of Biodegradable Polymer-Based Films

Mechanical and Barrier Properties of Films

Cling wrap films are widely used for preserving a variety of fresh fruits and vegetables in supermarkets owing to its good clarity and thus are effective for display. Furthermore, cling film packaging preserves moisture and aroma of wrapped foods, thus keeping it fresh. Biodegradable polymer-derived films are generally poor in barrier properties, thus limits its applicability. Widespread usage of bio-based films depends on its mechanical and barrier characteristics when compared with commercially available packaging materials. Mechanical properties of any film determine its ability to protect food against physical damage. Mechanical properties of packaging

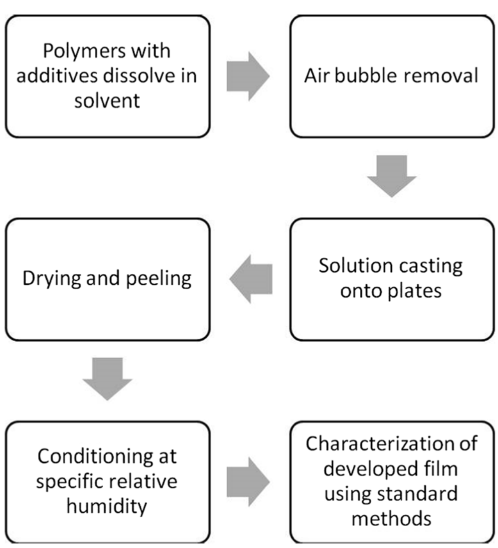


Fig. 3 Flow chart of a solvent casting method for biodegradable film preparation

materials include Young's modulus, tensile strength, puncture strength, elongation at break, etc. The most widely used standard to determine the mechanical characteristics of different types of biodegradable films is the American Society of Testing and Materials (ASTM) D882-02 standard test method for tensile properties of thin plastic sheeting. Each mechanical characteristic as mentioned above helps in evaluation of a film, thus assists in determining its potential application.

Headspace gaseous composition is one of the important aspects of storage of minimally processed agricultural produce. An elevated O_2 percentage inside the packaging usually leads to unfavorable reactions like oxidation and browning while a lower O_2 percentage may facilitate the growth of anaerobic bacteria. Thus, maintenance of an optimum ratio of O_2 and CO_2 percentage is vital in food packaging. An O_2 content of less than 2% will favor the growth of anaerobic bacteria like *Clostridium botulinum* and microaerophilic bacteria like *Listeria* spp. and lactic acid bacteria [28]. Barrier properties of a packaging film are the main factor in determining the headspace gas composition of a packed sample. Generally, polymers are moderately permeable to small molecules like O_2, CO_2, water, and organic vapors. The O_2 barrier is calculated by the oxygen permeability coefficients (OPC) which signify the quantity of oxygen that infuse in a definite time and pressure through per unit of area of a packaging material ($kg{\bullet}mm^{-2}s^{-1}Pa^{-1}$). Thus, packaging films with low OPC may extend the shelf life of a packed product by reducing the O_2 percentage of a packet to the point that inhibits oxidation. Generally, biodegradable film has an OPC value of one or more order of magnitude below the OPC of synthetic films such as polyethylene terephthalate (PET) and polystyrene for same applications. The CO_2 barrier is measured by the CO_2 permeability coefficients (CO_2PC) which signify the quantity of CO_2 that permeates during a definite time and pressure through per unit of area of packaging films ($kg{\bullet}mm^{-2}s^{-1}Pa^{-1}$). The ratio of CO_2 transmission rate to O_2 transmission rate for fresh-cut produce packaging films ranges from 2 to 5 with an average value of 3 [73]. The ASTM D3985-05 standard test method is widely used for O_2 transmission rate through plastic film and sheeting using a coulometric sensor. The ASTM F2476-05 test method is used for the determination of CO_2 transmission rate through barrier materials using an infrared detector.

Equilibrium moisture content is one of the important factors responsible for the physical and/or chemical deterioration and dehydration of packed fresh agricultural produce. Thus, the water vapor barrier characteristic of food packaging films is of great importance for maintaining or extending the shelf life of fruits and vegetables. The water vapor barrier is determined by the water vapor permeability coefficients (WVPC) which specify the quantity of water vapor that transmit in a definite time and pressure through per unit of area of packaging material ($kg\ mm^{-2}\ s^{-1}\ Pa^{-1}$). Water vapor permeability of biopolymer-based film is usually determined by standard

ASTM E 96-95. Saurabh (2015) [67] developed guar gum-based composite film having better mechanical and comparable barrier properties as compared to commercially available cling film and other biopolymer-based films (Table 4). Polyvinyl chloride (cling film) film had a tensile strength of 42 ± 7 MPa and water vapor transmission rate of 35 ± 5 g/m^2/d, however, and for guar gum-based hybrid film, it had a value of 122 ± 18 MPa and 39 ± 4 g/m^2/day, respectively.

Incorporation of Various Additives

An important function of food packaging is to maintain the microbiological quality of a packed sample. Suitable films also assist in maintaining sensory and nutritional parameters. Furthermore, packaging films can act as carriers for food additives, including antioxidants, colorants, flavoring agents, and antimicrobial compounds [75]. Various researchers demonstrated that addition of different types of antimicrobial agent in packaging films effectively control the microbiological growth on food and extend its shelf life. However, the antimicrobials used in edible film formulation must be classified as food-grade additives or GRAS by the relevant regulative authority. There are numerous groups of antimicrobials that have the potential to be used into edible films including organic acids like acetic, benzoic, lactic, propionic, sorbic, etc.; fatty acid esters like glyceryl monolaurate; and polypeptides like lysozyme, peroxidase, lactoferrin, nisin, etc. These compounds typically inhibit the propagation of food pathogens. Methylcellulose and chitosan-based composite films containing 2% of organic acid salts potassium sorbate or sodium benzoate were very active against *Rodotorula rubra* and *Penicillium notatum* [76]. Plants, herbs, and spices, as well as their extracted essential oils or concentrations, and compounds isolated from different extracts contain a wide range of compounds that are well known to inhibit the metabolic activity of bacteria, yeasts, and molds [77]. Essential oils of various spices and herbs like angelica, anise, cardamom, cinnamon, cloves, coriander, fennel, garlic, nutmeg, oregano, parsley, rosemary, sage, thymol, etc. have inhibitory effects towards a wide range of spoilage or pathogenic bacteria, molds, and yeasts. Various types of polysaccharides like starch, cellulose and its derivates, chitosan, alginate, and fruit puree-based films containing natural antimicrobial agents have been studied extensively for inhibiting the growth of food microbes. However, there are limited studies available in the literature for protein-based films incorporated with natural antimicrobial agents. Proteins which are mainly studied for active films are whey protein isolate, soy protein, soy protein isolate, egg albumen, wheat gluten, and sodium caseinate. Ayala-Zavala et al. (2013) developed an edible pectin-based film incorporated with cinnamon leaf essential oil with high antioxidant content that can effectively reduce the bacterial growth on fresh-cut peach [78]. From the broad variety of target microbes studied for food poisoning, human pathogens of the genus *Listeria*, *Escherichia*, and *Salmonella* have been extensively studied. For example, hydroxypropyl methylcellulose (HPMC) film incorporated with nisin demonstrated antimicrobial activity against *L. monocytogenes* ATCC 15313, *S. aureus* IP 58156, *Kocuria rhizophila* ATCC 9341, and *Aspergillus niger* [75]. In another set of study, Valencia-Chamorro et al. (2008) showed that the stand-alone film prepared from hydroxypropyl methylcellulose/lipid composite incorporated with organic acids, salts of parabens, and mineral salts had antimicrobial activity against postharvest pathogens of fresh citrus fruit, i.e., *Penicillium digitatum* and *Penicillium italicum* [79]. In a different study, sodium alginate films containing lactoperoxidase demonstrated significant antimicrobial activity against different types of pathogenic bacteria [80]. Besides affecting antimicrobial properties of films, antimicrobial agents also significantly affect its mechanical properties [81]. A hypothesis was proposed by Han (2005); according to the authors, if the molecular weight of the biopolymer is higher than the incorporating antimicrobial agents, then no considerable effect on strength of film will be observed since antimicrobial molecule cannot change the polymeric conformation [82]. However, tensile properties can be significantly altered if there is chemical/physical interaction between incorporating additives and the base polymer matrix. Antimicrobial agents like essential oils are hydrophobic in nature; thus, their addition in a biodegradable film may result in low water vapor permeability as compared to native films [83]. Furthermore, antimicrobial molecules can also improve the gas barrier properties of bio-based films. Surface morphology of antimicrobial films is roughed as compared to native films because additives disturb the homogeneity of films [81].

Nanotechnology and its application in food science have recently been studied by several researchers. Starch/clay nanocomposite films have been obtained by dispersing MMT

Table 4 Comparison between commercially available cling film and different types of biopolymer-based film

Polymers	Thickness (μm)	Tensile strength (MPa)	Young's Modulus (GPa)	WVTR (g/m^2/day)
Polyvinyl chloride (cling film) [67]	12	42 ± 7	0.12 ± 0.02	35 ± 5
Guar gum, beeswax, and nanoclay [67]	29	122 ± 18	99 ± 8	39 ± 4
Cassava starch and montmorillonite [74]	71	21–25	1.1–1.2	1082–2000
Chitosan and polylactic acid [60]	67	30–25	0.017–0.011	165 ± 7

nanoparticles via polymer melt processing techniques [37]. Such films incorporated with nanoclay were reported to have increased tensile strength. Fabricated nanocomposite films also had high water vapor barrier property due to the introduction of tortuous path. Tensile strength increased from 19 to 22 MPa and Young's modulus enhanced from 979 to 1135 MPa with nanoclay content from 0 to 4%, respectively. Some organically modified nanoclay also demonstrated antimicrobial activity [84]. The most commonly researched nanoparticle in active food packaging films is silver nanoparticles which are well known for its strong activity against a wide range of microorganisms [85], with high-temperature stability and low volatility [86]. Li et al. (2009) have reported the use of such nanocomposite films for retardation of decay in Chinese fruits [87]. In a study by Costa et al. (2012), the feasibility of calcium- and alginate-based films loaded with silver and montmorillonite (Ag-MMT) nanoparticles for shelf life enhancement of fresh-cut carrots were demonstrated [88]. However, migration of additives from films to food remains an important constraint in the use of such films. Although, additive migration from biodegradable nanocomposite film to vegetable products is considered to be minimal. There are very few reports available on the migration of additives from biopolymer-based films to food products. Thus, it is necessary to have more detail analysis and accurate information regarding migration rate during storage and processing of product which are likely to have an impact on human health.

Nutraceuticals can also be incorporated into edible film's formulation. Many studies in the past have been focused on the incorporation of minerals, vitamins, and fatty acids into edible film solution to improve the nutritional importance of some agricultural produce where micronutrients are not present or found in very low amount. Mei and Zhao (2003) showed that edible films prepared from milk protein can carry a high percentage of calcium, i.e., up to 10% (*w*/*v* of polymer) and vitamin E up to 0.2% (*w*/*v* of polymer) [89]. Addition of nutraceuticals can also result in improvement in film properties. Park and Zhao (2004) reported that the water vapor barrier property of chitosan films improved by increasing the concentration of mineral zinc lactate up to 20% (*w*/*v* of polymer) or vitamin E in the film matrix [90]. In the last few years, studies have been mainly focused on the addition of probiotics to obtain functional edible films. Tapia et al. (2007) developed the first edible films using gellan with a probiotic compound containing viable *bifidobacteria* for fresh-cut apple and papaya [91]. The authors further reported that the viable cell count of > 10^6 cfu/g *Bifidobacterium lactis* Bb-12 were maintained up to 10 days during storage at low temperature for both the fruits. This study demonstrated the feasibility of polysaccharide coatings to carry and support viable probiotics on fresh-cut agricultural produce.

Maintenance of sensory characteristics of packed food products is an important aspect in the evaluation of film applications. In order to slow down the flavor changes during food storage, the encapsulation of aromatic compounds has been investigated as a possible approach to minimize the effect of degradation like oxidation, browning, and lipid peroxidation. Marcuzzo et al. (2010) encapsulated 10 different types of aromatic compounds in carrageenan-based films including ethyl acetate, ethyl butyrate, ethyl isobutyrate, ethyl hexanoate, ethyl octanoate, 2-pentanone, 2-heptanone, 2-octanone, 2-nonanone, and 1-hexanol [92]. These edible films aim towards the gradual release of aroma compounds and thus sustain the sensory quality like aroma and taste during storage. Furthermore, Hambleton et al. (2009) demonstrated that the film matrices made up of polysaccharides like alginate can protect encapsulated aroma compounds like n-hexanal due to its low oxygen permeability [93]. Studies have demonstrated that polysaccharide matrices can encapsulate aroma compounds, thus preserve the organoleptic quality of food systems. However, protein-based films or blends are yet to be studied as protective matrices for aroma compounds. Nevertheless, in one study, Monedero et al. (2010) found that the incorporation of beeswax was essential for the improvement of the capacity of soy protein isolate-based films to retain n-hexanal [94]. Fresh-cut fruits and vegetables tend to lose their visual appeal owing to cut edge browning. Oxidation of phenolic compounds polyphenol oxidase (PPO) and peroxidase (POD) produces quinone compounds which on polymerization form brown deposits. Ponce et al. (2008) showed that chitosan-based films incorporated with olive and rosemary oleoresins when applied on butternut squash (*Cucurbita moschata Duch*) slices the action of polyphenol oxidase (PPO) and peroxidase (POD) slowed down within 5 days of storage period, thus lowering the browning of packed products [95]. The obtained result was due to the presence of antioxidants in olive and rosemary in the biodegradable film.

Commercially Available Biodegradable Films

Numbers of industries have dedicated their effort towards the development of commercially viable and sustainable food-related packaging applications using various biopolymers. A few examples of such companies are listed in Table 5 which reflects the diversity of bioplastics' applications in food packaging and their recognition at commercial scale to produce environmentally responsible packaging materials. Del Nobile et al. (2008) used biodegradable Novamont films for prolonging the shelf life of minimally processed lettuce by 5 days as compared to traditional package film (oriented polypropylene) which extends the shelf life by 4 days only at 4 °C [66]. Novamont (Novara, Italy) produce a wide range of biodegradable film including monolayer and multilayer co-extruded film based on a blend of biodegradable polyesters. Furthermore, Novamont commercializes MaterBi®, a type of

Table 5 Companies involved in production of bioplastics for food-related packaging applications

Company	Biopolymer	Application
Bioenvelop	Starch	Moisture barrier coating films for food
EverCorn, Inc. (subsidiary of Japan Corn Starch Co., Ltd.)	Starch	Films for food wrapping
FKur Kunststoffe GmbH	Cellulose/polylactic acid	Composite film for food packaging applications
Metabolix	Polyhydroxyalkanoates	Utilized as cast film
NatureWorks LLC	Polylactic acid	Food wrapping
NODAX	Polyhydroxyalkanoates	Good odor and oxygen barrier properties, customizable mechanical properties, thus highly suitable for food packaging
Fresh Del Monte Produce Inc	Polylactic acid	Packaging of fresh-cut fruits
ILIP	Polylactic acid	Packaging of fresh-cut to fresh whole produce

granular starch that can be processed as a thermoplastic material for manufacturing of films or bags. Lucera et al. (2010) studied the effect of different packagings like oriented polypropylene based (OPP) bag and biopolymeric co-extruded polyesters (COEX, Novara, Italy) on the shelf life of fresh-cut zucchini [96]. The authors reported that samples packaged in both biodegradable and nonbiodegradable films showed comparable microbial and sensory qualities till the end of 9 days of storage at 5 °C. Similar results were observed when PLA film (Tohcello, 25 μm, Japan) was used to pack green pepper and stored for 7 days at 10 °C [11]. In another study, the postharvest shelf life of blueberries was 9 days at 23 °C when packaged in PLA containers (VersaPack®, Wilkinson Industries Inc., Fort Calhoun, NE) or PET clamshell containers [12]. Nowadays, several starch-based films are commercialized; for instance, Bio-P-TM®, a product of Bioenvelope (Japan), and BIOPAR® (Biop Biopolymer Technologies AG, Germany) are potato starch-based material and completely biodegradable. PLA is a thermoplastic polymer derived from renewable resources like corn starch or sugar cane and second most commonly used material for manufacturing of biodegradable films. PLA can be produced either by ring-opening polymerization of lactide using various metal catalysts or by direct condensation of lactic acid monomers at less than 200 °C. Lactic acid chirality produces many types of PLA including poly-L-lactide (PLLA) and poly-D-lactide (PDLA). Mixing of PLLA and PDLA resulted in improvement in functional characteristics of resultant blends depending on the mixing ratio. Extrusion of PLA leads to a wide range of materials that can be used in film and sheet casting, 3d printing, injection molding, and spinning. It can also be used as decomposable packaging material, cups, bags, mulch, etc. A purified protease from *Amycolatopsis* PLA depolymerize can degrade PLA [97]. The higher crystallinity of PDLA decreases its biodegradability rate as compared to PLA. One major disadvantage of PLA is its low glass transition temperature which makes PLA-based plastic cups unsuitable for hot liquid and other fills. Its temperature stability can be maximized by using blends of PDLA and PLLA [98]. Another limitation of PLA is its inherited brittleness which can be overcome by using suitable plasticizer. Recent research trend paves the way for PLA to overcome its barrier for wide-scale commercial applications. Various studies have demonstrated that biodegradable film can be successfully used to replace widely used nonbiodegradable films for packaging of fresh-cut fruits and vegetables.

Conclusion

The environmental concern of petroleum-based packaging film has led to an amplified research in the area of biodegradable packaging films. Biopolymers are eco-friendly and renewable; thus, they are a potential source for bio-based plastics. Numerous studies have been conducted in the past for exploring the feasibility of biopolymer-based films to improve the shelf life of fresh-cut agricultural produce. Starch, cellulose, polylactic acid, polyhydroxyalkanoates, gelatin, and chitosan are widely used for this purpose. Antimicrobial agents like silver nanoparticles, essential oils, nisin, lysozymes, etc. further facilitate bio-based film to enhance the microbial safety and sensory qualities of fruits and vegetables. Apart from the selection of appropriate biopolymers and active components, film formation procedure like mixing and drying temperature, relative humidity of storage, casting plates, pretreatment of additives, etc. also significantly alter the characteristics of developed films. Developed biodegradable active films must have some tailored characteristics like water vapor transmission rate, oxygen barrier property, and carbon dioxide permeability in a specific range for commercial applications of food packagings. The current trend in the biopolymer-based film indicates that the cumulative effort is in the right direction which has led to the foundation of various industries commercially producing biodegradable packaging films. However, further research is needed in the field of bioplastics to exceed the petroleum-based counterparts in terms of wider applicability.

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