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Preface

Biodegradable polymers play an essential role in innovative food packaging while causing no harm to the environment. Unfortunately, conventional food packaging necessitates petrochemical-derived polymer as a packaging material, which endangers both the environment and human health. The significant limitations of conventional packaging materials are their non-biodegradability and the release of harmful gases such as dioxin and HCN during disposal. The application of biodegradable packaging material for food products is more prevalent in the European countries, and the USA is owing to its similarity in physicochemical properties with polypropylene-based plastic. However, despite its biodegradability and similarity in physicochemical properties with plastic, bioplastic's rapid usage is restricted due to its high production cost and complex production strategies. Certain biobased polymers are currently being tested for packaging applications in various food commodities. The mechanical and chemical properties of biobased plastic vary depending on its source. The moisture barrier properties, gas permeability, tensile strength, and heat stability of such plastic material determine its commercial acceptability in food packaging industries.

Based on the current scenario, the book *Novel Approaches for the Manufacture of Biodegradable Polymer-Based Food Packaging Materials* is a much-required effort in this series. The book is divided into 16 chapters that focus on novel approaches to the production of biodegradable polymers. Scope and importance of biodegradable polymers, animal, plant, and microbially derived biopolymers, nanocomposite biopolymers, polyhydroxyalkanoates, polylactide in food packaging, enzymes and metabolic engineering involved in biopolymer synthesis, downstream processing strategies, functionality test methods, application of biopolymers in packaging, standards for testing the biodegradability, and future prospects are presented and discussed in detail. The book proposes a new perspective with advancement and long-term solutions to existing gaps in the nanocomposite biopolymer production process and active packaging. This book will also provide a thorough understanding of the factors influencing the stability of biodegradable polymer films. Finally, the book predicted that microbially derived biodegradable polymers would play an essential role in food packaging in the near future. Hence, this book will serve as an asset for the researcher working in the relevant area, including researchers, scientists, teachers, and students.

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I am grateful to my co-editors, Prof. Dinesh Chandra Rai, Professor and Head, Department of Dairy Science and Food Technology, Institute of Agricultural Sciences, Banaras Hindu University, India, Prof. Kianoush Khosravi Darani, National Nutrition and Food Technology Research Institute of Iran and Ms. Veena Paul, Department of Dairy Science and Food Technology, Institute of Agricultural Sciences, Banaras Hindu University, India, for providing the immense support in compilation of this book. I am thankful to all my authors from India and Iran who contributed different chapters for this book as per their expertise. I am grateful to Springer Nature group for selecting our book proposal and permitting us to complete this task. I am thankful to external reviewers who approved the book proposal after thorough evaluation. I pay my sincere gratitude to our Director and Esteemed Dean for their constant encouragement. I am grateful to my Head of the Department Prof. Dinesh Chandra Rai, Department of Dairy Science and Food Technology, Institute of Agricultural Sciences, Banaras Hindu University, India, for his immense motivation and providing necessary facilities required for compilation of this book. I would like to pay my sincere thanks to my parents Shri Vishnu Dutt Tripathi and Mrs. Urmila Tripathi for their encouragement and support. I would like to pay my sincere thanks to my beloved wife Mrs. Priyanka Upadhyay and my kids Master Ojas and Baby Prajul for their kind support and cooperation. Lastly, I am thankful to almighty who blessed me to complete this task.

Contents

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Introduction: Scope and Importance of Biodegradable Polymers

1

Veena Paul, Abhishek Dutt Tripathi, Kamlesh Kumar Maurya, Pankaj, Dinesh, and Chandra Rai

Abstract

In today's world, the interest in biodegradable polymers has grown significantly. The demand for these polymers has skyrocketed, making them broadly utilized polymers with a range of several applications. Plant, animal, and microbially derived biopolymers have piqued the interest of researchers. Because of their inherent properties like biodegradability, biocompatibility, inexhaustibility, and economic availability, the need for biodegradable polymers has been enhanced. Recent trends in the food packaging industry have shown the employment of biodegradable polymers with improved characteristics. This chapter emphasizes biodegradable polymers' development, classification, application, challenges, and market opportunities. The application of these polymers in active and intelligent packaging are summarized. This chapter also highlights the application of biodegradable polymers as an assuring green technique for assuring the quality and safety of food.

Keywords

Biodegradable polymers · Food packaging · PHA · PLA · Bacterial cellulose · Biocomposites

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1.1 Introduction

Excessive use of plastics and their aggregation in the natural environment as a stubborn source of pollution have put life on earth in jeopardy (Geyer et al. 2017). About 30% of plastics are consumed as packages, and about 47% are biodegradable polymers. Remarkably, about 75% of biodegradable polymers are employed as packaging material in the food sector (European bio-plastics 2020). The crucial role of these polymers is to protect the product from harmful effects caused by microbiological contamination, chemical damage, and mechanical impact. The packaging material serves many benefits. It acts as a barrier against physical damage and environmental contamination, and it protects the food commodity from damage by any other external factors. It ensures quality and safe food with enhanced shelf-life (Cazón and Vázquez 2020). Food packaging is used in the manufacturing, preservation, storage, distribution, and preparation of food. Thus, to maintain the characteristics of food products, the choice of packaging material is essential (Ivonkovic et al. 2017).

According to current research studies, the European Union used 1,130,000 million food packaging materials in 2018. Consumption of packaging material has resulted in the generation of waste in a large amount. In 2013, data showed that the European Union generated 156.9 kg of waste from the packaging industry per inhabitant. It is likely to rise at a 4.2% annual rate in the coming years. The most common packages used in the food sector are polypropylene, high, low, and very-low-density polyethylene, polyvinyl chloride, polystyrene, and polyethylene terephthalate. These packages contribute to roughly 90% of total plastic production. These packages are economical with exceptional characteristics. However, they are nonbiodegradable and have numerous constraints. The waste management of synthetic polymers poses a severe environmental threat (Cazón and Vázquez 2020).

Presently, the need for biodegradable polymers has been extended worldwide as a potential packaging material. Nevertheless, biodegradable polymers in the food packaging sector are limited because of expensive production costs. This can be overcome by employing the production of naturally derived polymers. These are either plant-based, animal-based, or microorganism-based. Biodegradable polymers derived from natural resources can be used as novel packaging material in the food sector. Eco-friendly biodegradable packaging is expected to impact food product quality and market in the future significantly. Polysaccharides and proteins are the most diverse biopolymers used as films in the food packaging sector. These are naturally derived from renewable sources like plants, animals, or microorganisms. The commonly used polysaccharides in biodegradable packaging are alginate, carrageenan, chitosan, cellulose, and its derivatives, pullulan, and starch. Casein, gelatin, gluten, whey, soybean proteins, zein, and keratin are all proteins. Presently, biodegradable polymers of renewable origins cannot entirely substitute synthetic polymer from the food packaging market. However, they may partly serve as a significant stress factor. These polymers enable humidity control, exchanges of gases (internally and externally), compound migration, and undesirable aromas.

Furthermore, they can sustain and deliver antimicrobial or antioxidant agents into the food matrix (Cazón and Vázquez 2020).

This chapter aims to discuss the development and classification of biodegradable polymers for packaging in harmony with nature. Further, their application in the food industry and challenges and market opportunities are discussed.

1.2 Development of Biodegradable Polymers

Today, at the dawn of the twenty-first century, products derived from renewable sources are valued for environmental friendliness. In general, consumers worldwide are becoming more aware of biodegradable polymers. Due to the continuous buildup of synthetic polymers, there is a decline in arable lands, oil wells, and the gases released while burning waste has urged the fabrication of biodegradable polymers to be used as a packaging material (Ivonkovic et al. 2017).

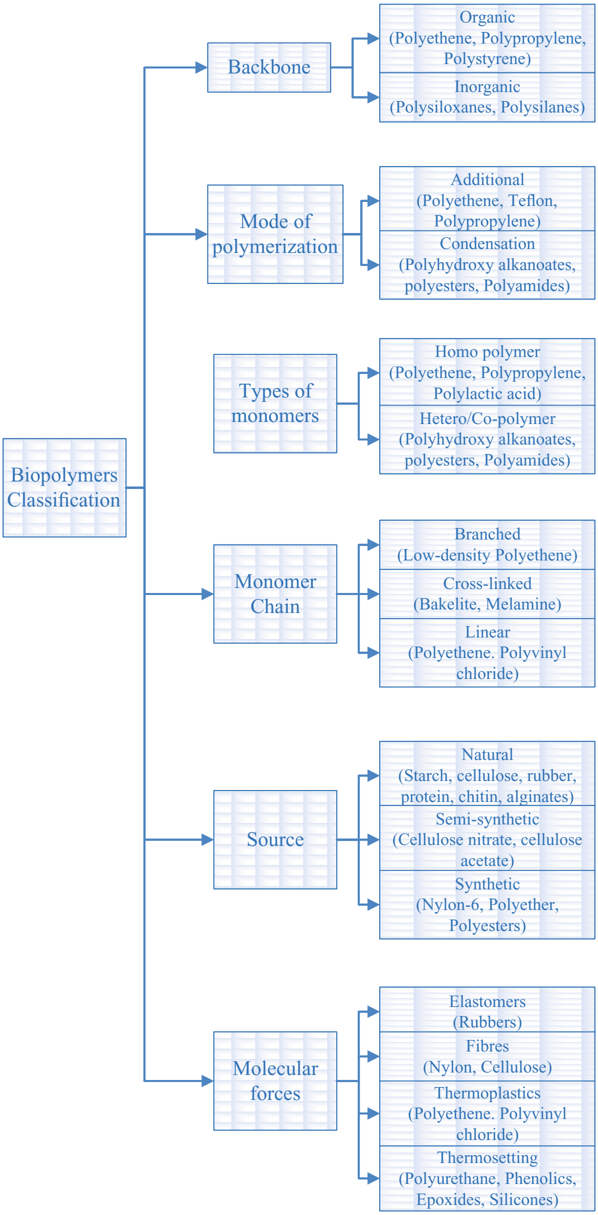
1.3 Classification of Biodegradable Polymers

Based on different criteria, biodegradable polymers are classified (Fig. 1.1). As described by European Bioplastics, biodegradable polymers are either biologically derived or biodegradable or both (Rai et al. 2021). They can be categorized as biodegradable and nonbiodegradable. Despite being derived from natural sources, not all biodegradable polymers are biodegradable. Their pathway and rate of degradation determine the biodegradability of polymers. Biodegradable polymers such as polyhydroxyalkanoate (PHA), starch, cellulose, polylactic acid (PLA), and their derivatives are used to make biodegradable polymers. Biodegradable biopolymers, like traditional fossil-based polymers, can be recycled. Instead, they can be degraded by microorganisms, thus, creating a zero-waste model (Rai et al. 2021).

Under natural or stimulated conditions, biodegradable polymers decompose from microbial enzymatic action into CO_2, H_2O, and inorganic compounds. However, comparing biodegradability is challenging because of the various standards, degradation processes, and circumstances used to evaluate polymer's degradation rate. The test duration is an essential parameter in these standard methods for determining biodegradation. In wastewater, the biodegradation tests are for not more than half of a year, but on the contrary, in marine water, the biodegradation tests require about 2 years (Sanchez-Salvador et al. 2021).

At present, out of the total plastic production (335 million tonnes), not more than 0.3% (0.91 million tonnes) are biodegradable plastics. PLA, polybutylene adipate terephthalate (PBAT), and starch blends are commonly fabricated biopolymers, accounting for 24%, 42%, and 17% of total production (European Bio-plastics 2020). Based on origin, biodegradable polymers can be categorized into four parts (Fig. 1.2).

Fig. 1.1 Different criteria for the classification of Biodegradable polymers



1.3.1 Agro-based Biopolymers

The agro-waste like sugarcane bagasse, banana peels, corn stalks, wheat straw, and jute can be valorized to produce biodegradable polymers (Fig. 1.2). Initially, the

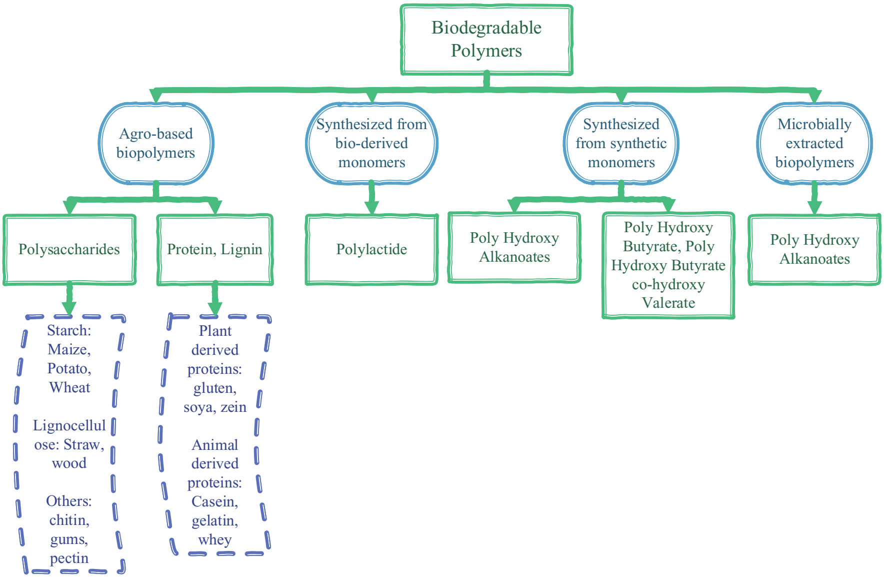


Fig. 1.2 Classification of biopolymers (origin-based)

substrate (agro-waste) conversion into functional biopolymer occurs via microbial breakdown of the substrate under favorable growth conditions (Rai et al. 2021).

Biopolymers derived from agro-waste consist of polysaccharides and proteins. Polysaccharides, also known as complex carbohydrates, are found in the essential structural parts of a plant or animal's exoskeleton, such as cellulose and chitin, generated by glycosidic linkages. On the other hand, protein macromolecules are fabricated of amino acids associated with peptide bonds. Each type of protein is distinguished by its amino acid composition (Sanchez-Salvador et al. 2021).

Starch, a natural ingredient that consists of amylopectin and amylose. The amylopectin ratio to amylose impacts the functionality and other properties of starch-based products. A high level of amylose increases film strength; nevertheless, amylopectin forms a film with low strength due to branched structure. The strength of polymers can be enhanced by employing plasticizers (Rai et al. 2021). Commercially starch is abundantly obtained from potatoes, rice, etc. (Amulya et al. 2021). Starch-based biopolymers are a suitable substitute for commercial packaging applications as they are obtained from renewable sources and have higher biodegradability and oxygen barrier property. Mater-Bi (MB™), a Novamont product, is comprised of starch and starch mixes (60%) and hydrophilic (40%) blends. About 26% of MB™ degrades under aerobic conditions within 3 months. This biodegradable polymer degrades completely in aerobic sludge in 100 days. MB™ produces methane and CO_2 under anaerobic conditions (Mohee et al. 2008). Table 1.1 shows the biodegradability rate of various biodegradable polymers.

Gómez and Michel Jr (2013) studied the blend of starch and PLA. They observed the conversion rate of starch-PLA bled under aerobic and anaerobic conditions.

Table 1.1 Biodegradability rate of various biopolymers

Types of biopolymers	It can be degraded in	Biodegradability rate		References
		(Days)	(%)	
Mater-Bi™	Marine water	236	68.9	Tosin et al. (2012)
Cellulose acetate	Municipal wastewater	12	44	Mostafa et al. (2018)
Polylactic acid	Municipal wastewater	28	39	Lambert and Wagner (2017)
Polyhydroxyalkanoate	Soil	60	35	Torres et al. (2019)
Polyhydroxybutyrate	Sea water	14	80	Rai et al. (2021)

Under aerobic conditions, the blend with 65% moisture composted to 51% in 4 months, whereas under the anaerobic situation, the same blend composted to 50% in 85 days (Gómez and Michel Jr 2013). These studies show that biopolymeric blends will likely replace synthetic polymers and can be a low-waste-generating alternative in the future.

Agro-waste, a cheaper substrate for industrial-scale production of polymers. Cellulose is obtained naturally in cotton linter, corn, wheat, paddy, and barley husk and stalk as a by-product (Rai et al. 2021). Due to its partial solubility in water, it cannot be used directly. However, it can be blended with plasticizers to suit various applications (Amulya et al. 2021). Cellulose has been used to make cellophane films widely used in packaging (Rai et al. 2021).

1.3.2 Bio-derived Monomer-Synthesized Biopolymers

Bio-derived monomers are derived from renewable sources. These monomers lessen carbon emissions. PLA, derived from lactic acid monomer from starch/sugar fermentation, is commercially used. It has improved mechanical properties and is broadly used for packaging with a short shelf life (Sanchez-Salvador et al. 2021). PLA is a viable choice in the packaging market (Amulya et al. 2021). PLA is created by polymerizing the enantiomers D-lactic Acid (PDLA) and L-lactic acid (PLLA). PLA can be made from both natural and synthetic processes. The natural renewable process is favored because it has less influence on the environment. The physical properties of PLA can be determined by the PDLA/PLLA ratio used during polymerization (Rai et al. 2021).

Because of its transparency, PLA has gained widespread acceptance in the packaging industry. Techniques like block polymerization/grafting and physical blending improve the physical properties of PLA. PLA biopolymers, both in their pure form and with additives such as starch, softwood, and sisal fibers, have been able to degrade (>50%) within 3 months when composted (Table 1.1). Due to their low carbon footprint and compatibility, such polymers are useful in the agricultural sector (Rai et al. 2021).

Poly(glycolic acid) (PGA) derived from sugar cane have a similar chemical structure as PLA, but their degradation rate is higher. Moreover, the copolymerization of PGA and PLA results in a linear poly(lactic-co-glycolic) acid (PLGA) polymer fabrication with improved physical and mechanical properties (Sanchez-Salvador et al. 2021). However, PGA has only been manufactured on a small scale and for medical use because of the expensive production process.

1.3.3 Biopolymers Synthesized from Synthetic Monomers

Polymers derived from synthetic monomers, like petroleum resources, have hydrolyzable groups such as esters, amides, urethanes, or biodegradable backbones such as polyesters and polyamides polyurethanes, polyanhydrides, or polycarbonates (Sanchez-Salvador et al. 2021). Table 1.2 illustrates the properties of commercially used biopolymers.

Poly(butylene adipate-co-terephthalate) (PBAT) is made from a synthetic monomer formed by polycondensing 1,4-butanediol with adipic and terephthalic acids. Because of its biodegradability, thermal, and mechanical properties, PBAT is regarded as one of the most appealing biodegradable polymers (Ferreira et al. 2019).

1.3.4 Microbially Extracted Biopolymers

Natural polyesters such as PLA, PHA, and PHB have been isolated by microbial degradation (Rai et al. 2021). Two types of biopolymers are derived from microorganisms. The first type consists of a class of natural biodegradable polyesters known as polyhydroxyalkanoates (PHAs). Microbially synthesized PHAs are highly crystalline and optically active compounds. PHAs have several applications due to improved physical and mechanical properties. At the same time, its application is restricted due to limited thermal property (Sanchez-Salvador et al. 2021). There is a majority of commercial PHAs. Poly-3-hydroxybutyrate (PHB) was the first commercial PHA discovered in 1925. Following PHB, poly(3-hydroxybutyrate-co-3-

Table 1.2 General properties of biodegradable polymers commercially used

Biodegradable polymers	Properties				
	Elongation at break (%)	Glass transition temperature (°C)	Melting temperature (°C)	Tensile strength (GPa)	Young's modulus (GPa)
PHB	5	–	177	43	–
PLA	<5	64	173–178	50	350–3500
PCL	–	−60	58–63	–	–
PBAT	>600	−30	110–120	32–36	20–60
PBS	230	−32	114	42	690
BC	–	–	–	200–300	2000

hydroxyvalerate) (PHBV) demonstrated reduced crystallinity with improved flexibility. Novel PHA polymers, such as poly(3-hydroxybutyrate-co-4-hydroxybutyrate) (P3HB4HB), poly(3-hydroxy octanoate) (PHO), and poly (3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx), have recently been developed using novel fermentation technology in association with metabolic engineering (Ke et al. 2017).

PHAs produced from bacterial fermentation have elastomeric properties (Mor et al. 2021). PHAs, as microbial-produced biopolymers, can be used in the packaging sector because of their high gas permeability and water barrier properties (Tripathi et al. 2021). Many microorganisms produce PHAs by utilizing lipids or sugars extracted from various agro-waste (Mor et al. 2021). After interfusing within this family, no less than 100 monomers are identified, resulting in materials with a wide range of properties. PHA polymer is classified into three types based on its properties and applications: pure PHA, PHB, and PHBV. Agricultural products obtained from oilseed crops and sugar crops aid PHA production. PHA typically accumulate intracellularly in granules accounting for approximately 90% of the cellular mass (Kabasci 2020). As a result, the production of pure PHA is restricted. PHA fabrication is typically 20–80% more expensive than conventional polymers because of increased production costs. The lack of transparency and low production volumes of PHA polymers are disadvantages. Inorganic nanofillers (like montmorillonite) may enhance the thermal stability of PHA (Khosravi-Darani and Bucci 2015). PHA exhibits good thermo-mechanical properties, similar to plastics; thus, it may replace PE and PP. PHAs are used as bio-additives. Recently, PHAs has been used as mulched films.

PHA microbial production using glucose generally produces PHB and PHV. PHB polymer is mainly biodegradable and biocompatible, ideal for various industrial, medical, and agricultural applications (Amulya et al. 2021). PHB differs from other biodegradable polymers based on their physical properties (water insolubility and hydrolytic degradation resistance), thermal stability, and biodegradability with no residue (Adeleye et al. 2020).

PHB is fabricated by bacteria like *Alcaligenes eutrophus*, *Cupriavidus necator*, and *Bacillus megaterium* (Yaradoddi et al. 2019). During the PHB extraction, initially, two molecules of acetyl-CoA condense and form acetoacetyl-CoA. Further, this molecule reduces and forms hydroxybutyryl-CoA. Then, this reduced molecule generates PHB. The produced PHB possess increased crystallinity and thermal properties than PHA. PHB-PLA blends possess improved properties than PLA polymers. A study on PHB-PLA blends was reported to have enhanced barrier properties. Nanoclay and PHB increase the thermo-mechanical properties (Helanto et al. 2019).

The bacterial cellulose (BC), produced through the microbial fermentation of *Achromobacter*, *Aerobacter*, *Agrobacterium*, *Alcaligen*, *Azotobacter*, *Escherichia*, *Komagataeibacter*, *Zoogloea*, *Pseudomonas*, *Rhizobium*, *Salmonella*, and *Sarcina*. BC is a gel-like, swollen, flexible membrane produced through biosynthesis. BC has gained interest as an excellent antibacterial agent because of its purity, exemplary nanofibrils network, and improved tensile strength and water holding capacity

(Sanchez-Salvador et al. 2021). During BC production, additional pretreatments are not necessary for removing lignin, pectin, and hemicellulose compared to other plant-based polymers requiring chemical pretreatments to expel the abovementioned substances. The BC is extracted through the bottom-up method. These polymers are biocompatible and possess high molecular weight, high water retaining capacity (>90%), and high crystallinity (80–90%) compared to plant-based polymers (Zinge and Kandasubramanian 2020).

Algal polymers have recently become popular due to their improved photosynthetic efficiency. The algal biomass is a sustainable source for biopolymer extraction. The algal polymers can be obtained as algal biomass and plasticizer blends. Chlorella has been primarily used in blends (Rai et al. 2021). *Spirulina* sp. has also been used to make blended polymers. The blending properties of *Spirulina*-based polymers differ from those of *Chlorella*-based polymers due to differences in their amino acid content (Cinar et al. 2020).

1.4 Biocomposites

Aside from the methods mentioned above, producing biocomposites using natural fibers has improved their physicochemical properties (Wu et al. 2020). Biocomposites are typically created by blending a natural ingredient with a biopolymer to fabricate a hybrid polymer that combines both individual component properties. Their applications can explain the increasing trend in the biopolymer market. Biocomposites are made up of a stiff and high-strength polymer matrix that serves as reinforcement. It protects the reinforcement from environmental degradation.

Synthetic polymers (like PE, PP, and polyethylene) are commonly used as matrix materials. Moreover, with the increased requirement of biopolymers, studies have been directed towards more renewable sources (Amulya et al. 2021). As a result, natural fibers and biological particles are widely used to reinforce composite preparation (Wu et al. 2020). The polymers with improved stiffness and thermal stability have been selected for reinforcement.

1.5 Biodegradable Polymer Application in the Food Sector

Biopolymers have been used as a food packaging material as cutleries, wrap, containers, and films in the food industry. Recently, polymers acquired from inexhaustible assets that can be reused and treated the soil have earned expanding consideration. The properties of biopolymers can be improved by employing polymer engineering to replace the synthetic packages used in the food industry. Polymers like PHBV have been used as a food wrap. The production cost of PHBV wrap is higher, but it degrades in active microbiological conditions in 5–6 weeks. Auras et al. (2006) created a polyethylene terephthalate and polystyrene blend for new food packaging applications.

1.6 Challenges and Market Opportunities

With the rise of ready-to-eat food, food packaging has become a significant portion of the packaging market, with global sales expected to reach USD 411.3 billion by 2025 (Grand View Research 2020). Despite the research and development approach for biopolymers, only a few biopolymers can match the market demand for food packaging material. Likewise, the market drifts show that the Asia-Pacific region is relied upon to have the quickest development because of the simple accessibility and minimal expense of crude material needed to produce bioplastics.

In 2020, 1.2 million tonnes of biopolymers were produced globally, out of which approximately 18.7% of biopolymers were PLA and starch blends. Packaging industries utilized almost 47% of the produced biopolymers. Novamont's Mater-Bi (MB™), Bio-on's Minerv-PHA™, and Mitsubishi Chemica's GS PLA® produce biopolymers for food packaging and other applications (Rai et al. 2021).

1.7 Conclusion and Future Prospective

All around the world, plastic reusing is quantitatively insufficient for ecological manageability. The waste valorization and biopolymer fabrication using agro-waste will be a step toward sustainable development to reduce carbon footprint.

Biopolymers with improved physical and thermomechanical properties are a sustainable and green approach to replace plastics. Bioplastics are ideal and are more liked over conventional petro inferred plastics than their eco-accommodating nature. The present significant concern is the contamination caused to nature by these nondegradable plastics. They have wide application in the clinical, horticultural, and packaging industry fields. Unlike manufactured plastics, they produce no profluent to the encompassing nature and are henceforth more secure to utilize. The fundamental issue in synthetic plastic is the absence of a waste administration framework that has been an incredible danger to humankind since a long time ago. These all perspectives lead to innovative work in the area of bioplastics.

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Plant-Derived Biopolymers in Food Packaging: Current Status and Market Potential

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Abstract

Plant-based biopolymers show many advantages as biofilms and coatings for food packaging purposes; however, the current commercial technologies mostly employ synthetic polymers and plastics to achieve proper mechanical and barrier properties, which have been proved to have a negative impact on packaging sustainability, lack of biocompatibility and biodegradability, and poor recyclability. Therefore, attempts to employ natural and plant-based biopolymers as packaging materials have increased considerably. In order to meet the requirements, plant-based biopolymers must be functionalized and/or incorporate into a matrix as an additive to improve the ultimate physical and chemical properties of the packaging matrix. Here, we reviewed essential information about biomaterials extracted from plant sources, such as starch, cellulose, gums, pectin, wheat gluten, soy protein, and zein, regarding the use of such biopolymers in the food packaging area.

Keywords

Plant-based biopolymers · Starch · Cellulose · Gums · Pectin · Wheat gluten · Soy protein · Zein

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2.1 Introduction

The food packaging industry has been associated with plastic materials due to their excellent barrier and low cost. However, there are ecological concerns on their detrimental effects such as emission of greenhouse gases, generation of high amounts of wastes deposited in landfills, and using up valuable nonrenewable petroleum products (Kumar et al. 2017). Biopolymeric materials could be a potential alternative to conventional packaging materials due to their excellent capabilities to reduce environmental problems, as well as inhibiting moisture loss, oxygen penetration, water absorption, solute transport, and also showing excellent biocompatibility and biodegradability (Aider 2010; Siracusa et al. 2008). Therefore, developing biopolymer films has been exploited increasingly over the last two decades. Based on various sources of extraction, biopolymers can be classified into plant- and animal-derived compounds. The increased demand for food quality and safety in the food industry has resulted in films formed by plant-based polymers (Kanmani and Rhim 2014). Plant-based biopolymers possess antioxidant and antimicrobial properties and usually result in modified physicochemical and mechanical characteristics of packaging material. Therefore, they can promote shelf life and durability of the packaged product. Plant-derived biopolymers can be extracted from different sources including seeds, leaves, and fruits (Mir et al. 2018). Due to the abundant plant species (approximately 300,000) and the growing reliance of human civilizations on plant extracts, the available plant-based polymers for food applications have rapidly increased throughout the centuries (Joppa et al. 2011; Alvarez 2014).

Plant polysaccharide-based polymers such as starch (Nogueira et al. 2018), cellulose (Zhao et al. 2019), gums (Cao and Bin Song 2020), pectin (Brito et al. 2019; Vaziri et al. 2019), and plant protein-based polymers such as wheat gluten (Sartori et al. 2018), soy protein (Zhao et al. 2016), and zein (Padua and Wang 2002) are promising plant-derived biopolymers for food packaging industry. Starch is one of the most favorable biopolymers due to its high availability and low cost and has been extensively used for food bio-packaging applications. Starch and its derivatives exhibit attractive properties such as moisture and air barrier, edibility, biodegradability, heat-sealing capacity, and safety (Shah et al. 2015). There are several kinds of sources for starch extraction like potato, corn, rice, cassava, and tapioca. It can be utilized as a thermoplastic matrix or a co-constituents with other commercial thermoplastics by destruction of its granular structure in the presence of chemical agents, heat, or pressure (Rouilly and Rigal 2002). Furthermore, starch has been used as a filler in small amounts (6–30%) to increase the biodegradability of the synthetic product (Bagheri 1999). Cellulose is the amplest natural polymer mainly found in wood. As cellulose is highly fibrous, crystalline, and insoluble in water, it cannot be used in its native form (Nechita and Iana-Roman 2020). Alternatively, cellulose derivatives such as cellulose acetate, ethylcellulose, methylcellulose, and hydroxyethyl cellulose are able to form edible films and are commercially available (Tang et al. 2012). Several researches resulted in higher biodegradability of synthetic plastics with cellulose fibers (Mwaikambo 2006). Gums are another group of polysaccharides, naturally produced by some trees, seeds, shrubs, and tubers, that can be used as film forming agents in food packaging industry (Nejatian et al. 2020). Pectin is an anionic heterogeneous branched polysaccharide that mainly composed of methoxy esterified α, d-1, 4-galacturonic acid units (Lochhead 2017). Pectin is able to form biodegradable edible films and also generate matrixes in the presence of calcium ions. As pectin is widely accessible from agro-waste materials and can easily be modified by demethylation, it is considerably attended in the food packaging industry (Krochta and De Mulder-Johnston 1997).

Wheat gluten is a complex water-insoluble protein obtained from wheat flour. The major components of wheat gluten are gliadins (soluble prolamins) and glutenins (glutelins) with incorporation of small amounts of starch, wheat oils, and insoluble hemicellulose (Pallos et al. 2006). Gluten-based films represent low oxygen permeability level and high carbon oxide permeability level, so they could be advantageously used for improving the shelf life of food products (Giacalone and Chiabrando 2013; Muratore et al. 2005). Soy proteins are mainly consisting of two globulin fractions, β-conglycinin (7S, ~35%) and glycinin (11S, ~52%). Among plant-based protein sources, soy proteins are the most widespread ones, and due to their low oxygen permeability, they are suitable for packaging applications (Rouilly and Rigal 2002). Zein films have lower water vapor permeability and higher oxygen permeability compared to wheat gluten. However, their tensile strength is comparative to that of wheat gluten matrixes (Tang et al. 2012). Corn zein represent unique solubility in alcohol and form film matrixes by involving four steps, including dissolving into ethanol or isopropanol solutions, heating, cooling, and casting on a petri dish (Tang et al. 2012). Many strategies such as utilization of cross-linkers and incorporation of lipids have been used to reinforce zein-based films. Plant-based biopolymers have gained interest due to their potential influence on the functional properties of biofilms.

In this chapter we cover essential information about plant-derived biopolymers as green sources of biopolymers for food packaging industry, notably their origin, structural, and physical, functional, mechanical, antioxidant, and antimicrobial properties. Furthermore, the present status and future market about the potential of plant-based biofilms and bio-coatings in food packaging industry will be discussed.

2.2 Plant-Based Biopolymers in Food Packaging Industry

Plastics, as common traditional packaging materials, have been applied in the food packaging industry from a long time ago. Although they may provide several desirable characteristics like transparency, stiffness, good mechanical performance, heat sealability, good barrier to carbon dioxide, oxygen, and aroma compounds, several ecological and waste disposal challenges have restricted their use in recent years. The process of recycling of these materials is also expensive and impractical, due to the presence of various contaminants and their nondegradability properties (Siracusa et al. 2008). According to the global ecological awareness, reducing conventional packaging materials, including plastics, metal, glass, paper, and paperboard, is needed. Accordingly, development of bio-based polymers, as a major alternative to produce bioactive and biodegradable films, has become a central focus of food packaging efforts. Plant-derived biopolymers, which include polysaccharides and proteins, are potentially applied to fabricate environmentally sustainable coatings and films to extend the shelf life of ingredients and food products. In addition, they can promote the value of these products by offering antibacterial, antioxidant, and discoloration properties (Brito et al. 2019). Besides, the substantial volume of food wastes produced mainly from fruits and vegetables during the manufacturing of beverages and other processed foods can be used to produce different compounds. These residues are mainly fruit layers and vegetable ends, which are often discarded during processing stages. However, valuable bioactive compounds and biopolymers can be extracted from these residues (Ayala-Zavala et al. 2010; Andrade et al. 2014). The scientific community has been focused on retrieving the bioactive molecule contents from food residues and developing new and sustainable strategies to minimize the waste, as well as to extend the shelf life of the packaged food products. Therefore, exploring valuable sources of biomaterials from plant residues and potential strategies to extract them is of paramount importance (Fai et al. 2016). Various polysaccharides and proteins derived from plant sources have been reported as low-cost and widely available biopolymers, which exhibit desirable biocompatibility, biodegradability, emulsifying capabilities, and film-forming capacities (Dehghani et al. 2018). It is also becoming increasingly important to utilized these biopolymers to contribute in synthetic packaging materials such as petrochemical-based plastics, namely, polyvinylchloride (PVC), polyethylene terephthalate (PET), polyamide (PA), polystyrene (PS), and polypropylene (PP), to improve the functional properties of the final product and to obtain biodegradable and eco-friendly compounds (Siracusa et al. 2008). Bioplastics with the highest proportion of renewable biopolymers and additives are gaining much attention in the field of food packaging. The performance of these biopolymer-based compounds strictly depends on their physical and functional properties during their interaction with the food for maintaining their quality and safety. In this regard, obvious understanding and recognition of physical, mechanical, and barrier properties and compatibility with the food is important for their potential incorporation. In particular, plant extracts present as one of the most promising natural biopolymers, as they not only modify physicochemical properties of packaging materials but also act as an antioxidant and antimicrobial agent to enhance their overall functions (Bifani et al. 2007). The plant-based biopolymers are extracted from various sources such as leaves, fruits, and seeds. The incorporation of grape fruit seed extract into carrageenan films (Kanmani and Rhim 2014), tea extract into chitosan films (Peng et al. 2013), beetroot and carrot extract into hydroxypropyl methylcellulose films (Akhtar et al. 2012), and raspberry extract into soy protein films (Wang et al. 2012) is one of the examples of plant-based film enrichment. The incorporation of plant-based materials to food packaging films has led to generous desirable properties. The properties of films are influenced by the individual compounds that made them, so the type and concentration of each compound affect the ultimate techno-functional properties. Basically, the physical and chemical interactions within these compounds determine the structural and functional characteristics of the film (Silva-Weiss et al. 2013). These characteristics include thickness, color, mechanical and tensile strength, solubility, water vapor and oxygen permeability, and antimicrobial and antioxidant properties. Thickness is a vital parameter in techno-functional modification of biodegradable films, which influences the shelf life of the packaged food. Several studies have exploited the positive impact of plant extracts incorporation into biofilms on their thickness. The addition of tea extracts has been purported to increase the thickness of film by the formation of strong interaction occurring in the composite film matrix (Peng et al. 2013). Blend of mango kernel extracts within edible films has been enquired, and the results showed an increased thickness in film compared to the control one (Maryam Adilah et al. 2018). Grape fruit seed extract has been utilized for thickness improvement of agarose-based films due to their high phenolic constituents that form linkages with hydrogen bonds (Lim et al. 2010). In another study, incorporation of grape fruit seed extract in carrageenan-based films resulted in thickness increase with increasing the concentration of extract (Kanmani and Rhim 2014). The presence of phenolic compounds in plant extracts can also modify the original color of films. This alteration strongly depends on the concentration and origin of extracts. Another study revealed that the incorporation of thyme extract in chitosan-starch biocomposite films increased more reddish color due to the formation of strong interactions between amine groups of chitosan molecules and hydroxyl groups of polyphenols (Talón et al. 2017). The effect of murta fruit extract and mango peels extract incorporation into methyl cellulose film and fish gelatin film were investigated and resulted in yellowish increasing (Maryam Adilah et al. 2018; López de Dicastillo et al. 2016). Transparency of films is another essential parameter for food packaging material in order to fulfill the consumer's willingness to observe the product through packaging. At the same time, they should provide an adequate barrier to light to protect the light and UV-sensitive compounds from degradation. Plant extracts offer protection against adverse effects of UV radiations by their intrinsic opacity properties, and thus the addition of plant extracts in packaging films results in less degradability rates. As an example, films of tuna gelatin containing the murta ecotypes leaves extracts revealed better light protection properties (Gómez-Guillén et al. 2007). Chitosan and polyvinyl alcohol films enriched with extracts of mint/pomegranate peel displayed lower transmittance and better barrier properties to UV light compared to the control ones. The incorporation of plant extracts reduced the transparency, whereas, in films with greater contents of polyvinyl alcohol, the opacity values further reduced and film was more transparent (Kanatt et al. 2012). Similar results were observed in carrageenan-grape fruit seed extract composite. The absorption at wavelength of 270–280 nm and transmittance at wavelength of 280 nm in the UV region and 660 nm in the visible light region was measured. Carrageenan film supplemented with grape fruit seed extract exhibited absorption peaks, while the control film without plant extract did not show any absorption peak. This could be attributed to the high content of polyphenolic compounds which are present in most natural extracts. Polyphenolic compounds involve in the light absorption at lower wavelengths. In contrast, the transmittance value of the control film was substantially higher than the composite film, and it decreased by the addition of plant extract content. Therefore, the carrageenan/fruit seed extract composite film can be applied as a potent biocomposite packaging material with excellent UV barrier and high transparency properties (Kanmani and Rhim 2014). Another study incorporated ginseng extract into alginate films to investigate the effect of plant extract treated films on the transparency and protection against UV light. At 280 nm, the blended film showed very low transmission in UV light compared to the control film (Norajit et al. 2010), which could delay in lipid oxidation caused by UV light exposure. Gingko leaf extract was blended with gelatin to form a plant-based biofilm. Similar results were obtained after spectroscopic scanning of the samples. Light transmission decreased by the addition of gingko leaf extract, mainly due to the presence of flavonoids and ginkgo lactone contents in the gingko leaf extract. In case of incorporating green tea extract, grape seed extract, and gingko leaf extract into gelatin film, an increase in opacity occurred, which showed higher light barrier properties (Wang et al. 2014).

Natural polymers consisting of plant extracts usually lack adequate mechanical and tensile strength. In order to prevent failure, mechanical behavior of the composite film should be precisely determined. The type and nature of plant-based polymers have significant influences on mechanical properties of the biocomposite films. Elongation at break and tensile strength are the two important properties that are related to film chemical composition, storage time, temperature, and film's resistance properties (Gurgel et al. 2011). Generally, plant extracts have been shown to improve the mechanical properties of the films due to the presence of polyphenols, which interact with natural polymer chains such as starch, protein, and lipid and act as a cross-linking agent. Table 2.1 reveals some of the recent studies on plant extracts incorporation into biopolymers and their effects on the resultant biofilm with regard to water vapor permeability (WVP), tensile strength (TS), thickness (T), and elongation at break (EB). The interactions between plant extracts and biopolymers modify the structure and techno-functional properties of biofilms such as color, thickness, solubility, water and oxygen permeability, mechanical properties, antioxidant, and antimicrobial properties.

The mechanical properties of soy protein isolate and fish gelatin films significantly increased when mango kernel extracts were added to the film (Adilah et al. 2018). The effect of fish gelatin incorporation on cross-linking degree was superior as compared to soy protein, because the linear structure of fish gelatin is more suitable to interact with phenolic compounds than the globular structure of soy protein. Besides, fish gelatin is capable to form a stronger network after solubility, simply by reassembling triple helix structures (Tulamandi et al. 2016). On the other hand, several studies revealed lower mechanical properties of plant-based films, especially those based on fruit residues (Il Park and Zhao 2006; Martelli et al. 2013). Accordingly, it is necessary to explore new reinforcing materials and potential strategies to extract and prepare these materials to feasibly overcome this obstacle.

Despite the wide application of plasticizers in commercial purposes, they are not able to fully adhere to other matrix components, thus resulting in poor integration. Bionanocomposites are a new category of materials that are considered as a

Table 2.1 Recent studies for biopolymers modifications based the incorporation of plant extracts for food packaging applications

Plant extract	Biopolymer	WVP	TS (MPa)	T (μm)	EB (%)	References
Mango kemel	Soy protein isolate	7.93–8.78	1.82–3.06	56–55	49.92–148.92	Maryam Adilah et al. (2018)
Thyme	Starch	6.9–8.3	7.3–9.8	–	29–30	Talón et al. (2017)
Murta leaf	Methyl cellulose	–	11.1–13.9	51–64.7	164–205	Hauser et al. (2016)
Murta fruit	Methyl cellulose	–	5.15–14.20	–	12–51.50	López de Dicastillo et al. (2016)
Cinnamon leaf oil	Bombacaceae gum	3.93–3.64	8.71–6.47	–	17.42–17.39	Cao and Bin Song (2020)
Fruit and vegetable residues	Pectin	–	2.9–1.8	12–24	16–30	Brito et al. (2019)
Grapefruit seed	Carrageenan	1.22–1.64	3.21–43.38	38.3–43.3	18.14–27.27	Kanmani and Rhim (2014)
Onion peel	Funoran	4.86–5.64	16.93–24.58	77–95	20.49–35.77	Ju and Bin Song (2020)
Betalains from red pitaya peel	Starch/polyvinyl alcohol	2.48–2.88	30.77–35.02	115–119	39.61–41.99	Qin et al. (2020)
Pomegranate peel	Zein	–	21–28	–	1.2–4.1	Cui et al. (2020)
Grapefruit seed	Gelatin	0.66–0.87	55.2–63.4	62.1–66.3	9.6–13.3	Riahi et al. (2021)
Olive leaf	Carrageenan	6.61–7.43	8.51–11.83	32–48	29.21–36.58	Martiny et al. (2020)
Caraway seed	Chitosan	11.9–13.5	20.54–32.21	10.5–11.8	13.33–17.91	Homayounpour et al. (2021)

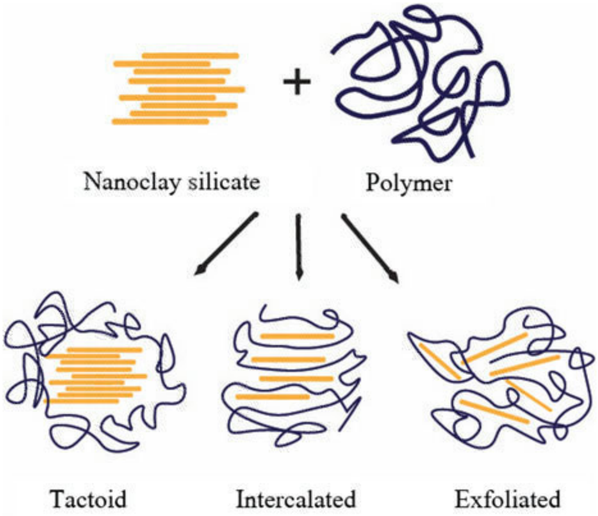

Fig. 2.1 Possible polymer clay structures

promising approach for improving mechanical, barrier, and thermal properties of the packaging films and coatings. Bionanocomposites are mainly consist of a biopolymer matrix and nanoparticles in three different dimensions, including isodimensional nanoparticles such as silica and metal nanoparticles, the ones with two dimension in nanometer range such as carbon nanotubes and cellulose nanowhiskers and the ones with one dimension in nanometer range such as layered crystals and layered silicate clays (Rhim and Kim 2014). Among these, layered silicate clays are widely employed as nanocomposite fillers due to their low cost, simple processability, and significant improvements. They consist of two coordinated tetrahedral silicone atoms that are fused to a octahedral sheet with a thickness of approximately 1 nm, and the lateral dimensions differ from tens of nanometers to several micrometers, depending on the source and the preparation method (Pavlidou and Papaspyrides 2008). For the formation of these bionanocomposites, layered silicate clays-polymer blends are commonly formed in three types, based on the type of clays and the processing conditions, consisting of immiscible tactoid, intercalated, and exfoliated, as shown in Fig. 2.1. In immiscible tactoid condition, clay particles are dispersed into the clay matrix, and the polymer cannot intercalate into the clay layers. Intercalation occurs when a polymer chain is inserted into the layers and a well-oriented multilayer morphology stacking is achieved. Exfoliation nanocomposite is a condition when the silicate layers are fully delaminated and dispersed and thus exhibit enormous improvements in performance properties, due to the higher interfacial area between polymer chains and layered clays (Rhim and Kim 2014).

Vegetable residues are another reinforcing materials that are known to add nutritional values to the biodegradable films, as well as enhancing flexibility and homogeneity of the film (Andrade et al. 2016). The addition of high methoxylated pectin with various granulometry fractions provided better mechanical properties by increasing film elongation and tensile strength at break and decreasing elasticity. It has been reported that the stiffness of the film depends strongly on the Young's modulus measurement. The larger the Young's modulus, the greater the stress required to undergo deformation, and thus the higher the strength and stiffness; and therefore, the greater the stress required to undergo deformation (Brito et al. 2019). The water sorption isotherms, which revealed thermodynamic equilibrium information between the water vapor and the film at the interface, proved to have lower accessible polar groups capable to provide water sorption sites. Water vapor permeability is one of the most important properties for food packaging purposes. Water vapor permeability depends on the chemical composition and morphology of the polymers and the environmental conditions, which influence solubility and diffusivity of water molecules within the matrix (Siripatrawan and Harte 2010). Murta extract-carboxymethyl cellulose biocomposite film resulted in lower water vapor permeability due to the leaf extracts incorporation (Bifani et al. 2007). The addition of green tea extracts had a significant positive effect on the water barrier properties of gelatin-based films. The abundant polyphenolic compounds, which are able to interact with polar groups of polypeptides found in gelatin, inhibit the formation of hydrophilic bonding of hydrogen groups with water (Siripatrawan and Harte 2010). Other researchers have noted the effect of plant-based extracts and biopolymers on barrier properties. The shelf life of the packaged food can be predicted based on the barrier properties of the biocomposite matrix (Wang et al. 2014). The solubility and moisture content of biodegradable films are important parameters affected by the nature of compounds. By the addition of hydrophilic polymers, more water molecules can bind to available hydroxyl groups, and thus, more water retains in the film. The amount of moisture content has been reported to be in the range of 16.43–35.75% in carrageenan-based films. Greater moisture values were observed with increasing carrageenan concentration. In another study, ginger extract-gelatin composite film resulted in higher moisture content as compared to gelatin film without ginger extract incorporation (Wang et al. 2014). It has been shown that plant extracts incorporation increases the solubility of films, mainly due to the presence of hydroxyl groups and their hydrophilic properties. The swelling behavior of the films is varied and strongly depends on the polymer structure and its thickness and concentration. The functional properties of food packages are enormously affected by the arrangement of various components in the biofilm composite, including plant-based extracts and polymers. The microstructure and morphology of the biofilm determine the homogeneity of the surface and porosity and thus could be responsible for water vapor permeability variations. It has been reported that the surface of gelatin-based films with plant extracts was less homogeneous and more brittle as compared to the films without any plant extract (Wang et al. 2014). This could be due to the denser cross-section microstructure of the composite film and the interaction of polyphenolic chains with hydrophobic molecules of gelatin. A similar result was observed in the soy protein-raspberry extract biofilm. The entanglement of soy protein side chains within plant extract molecules resulted in a more compact structure and subsequently affected the properties of the packaged product (Wang et al. 2012). The inclusion of