Review Article

Edible Polymers: Challenges and Opportunities

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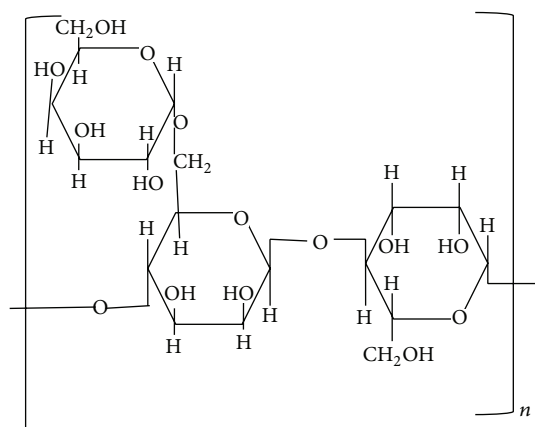Correspondence should be addressed to Subhas C. Shit; cipetahmd@gmail.com

Edible polymers have established substantial deliberation in modern eons because of their benefits comprising use as edible materials over synthetic polymers. This could contribute to the reduction of environmental contamination. Edible polymers can practically diminish the complexity and thus improve the recyclability of materials, compared to the more traditional non-environmentally friendly materials and may be able to substitute such synthetic polymers. A synthetic hydrogel polymer unlocked a new possibility for development of films, coatings, extrudable pellets, and synthetic nanopolymers, particularly designed for medical, agricultural, and industrial fields. Edible polymers offer many advantages for delivering drugs and tissue engineering. Edible polymer technology helps food industries to make their products more attractive and safe to use. Novel edible materials have been derived from many natural sources that have conventionally been regarded as discarded materials. The objective of this review is to provide a comprehensive introduction to edible polymers by providing descriptions in terms of their origin, properties, and potential uses.

1. Introduction

The increasing mandate for high quality, ready-to-eat food products with a long shelf life contributes to the expansion of new processing technologies which ensure that the product's natural properties and appearance were not dramatically transformed. Food wrapping contributes to a lot of wastes in our homes and it is something that most of us just accept. Edible polymers were the polymeric material which can be easily consumed by human beings or lower animals in whole or part via the oral cavity and given harmless effect to the health. Edible polymers may be applied directly on the surface as additional protection to preserve product quality and stability. The requirements imposed on edible polymers were determined by the specific properties of the product and changes in these properties during production and storage [1]. There were several reasons for investigating edible polymers. One of them was the introduction of new food product categories, such as safe, convenient, and high quality products. They protect food against the loss of nutrients. In practice, edible coatings which control the rate of transport of the product's molecular components from the inside to the outside of the packaging may slow down adverse reactions

which were responsible for undesirable changes in food products [1]. The efficiency of edible polymer was determined by their mass transport properties [2]. Technologist develops and formulates implantable drug delivery systems made of bioresorbable polymers that release precise quantities of therapeutic agents gradually over time [3]. Polymer drug conjugates were products of nanomedicine enrolled in cancer diagnosis and treatment [4].
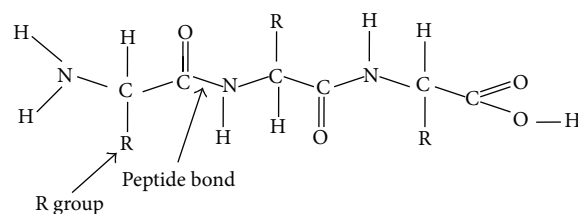
Natural polymers can be an alternative source for packaging development due to their precise taste and biodegradability. Edible polymers have appeared as a substitute for synthetic plastic for food applications and have received significant attention in recent years because of their advantages over synthetic polymer. The main advantage of edible polymer over traditional synthetics was that they can be consumed with the products. There is no package to dispose and even if the films are not consumed, they could still contribute to the reduction of environmental effluence. The edible polymers were produced exclusively from renewable, edible ingredients and therefore were anticipated to degrade more readily than other polymeric materials [5]. Edible polymer can expand the organoleptic properties of packaged foods, provided that they contain various components like
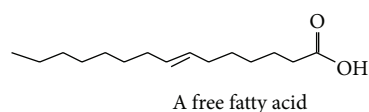
Scheme 1: Common structure of hydrocolloids.

Scheme 2: Common structure of polypeptide.

A free fatty acid

A triglyceride

A phospholipid

Scheme 3: Common structure of lipids.

flavorings, colorings, and sweeteners. Their use based on natural polymers and food grade additives has been continually increasing in the food industry and medical industries [6]. The edible polymer can be produced with a variety of natural products such as polysaccharides, proteins, and lipids, with the addition of plasticizers and surfactants. The functionality and performance of edible polymer mainly depend on their barrier, mechanical, and color properties, which in turn depend on film composition and its formation process. Food products were usually coated by dipping or spraying, forming a thin film on the food surface that acts as a semipermeable membrane, which in turn control the moisture loss or/and suppress the gas transfer [5]. The edible polymer also functions as carriers for antimicrobial and antioxidant agents. Production of edible polymer causes less waste and pollution; however, their permeability and mechanical properties were generally poorer than synthetic polymer.
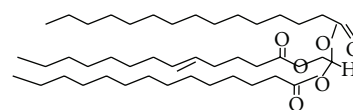
At present, around 150 million tons of plastic were produced yearly all over the world, and production and consumption continue to upturn [7]. Most of these plastics were crude oil based, and any increase in their production results in an increase of oil use and this causes serious environmental contamination, due to wasted and undegraded polymer. Edible and biodegradable polymers must be considered as a substitute to more traditional recycling procedures and this has stimulated researchers to synthesize new polymers that can be returned to the biological cycle after practice. Therefore, the use of agricultural polymers that were easily biodegradable would not only solve these problems, but also provide a potential new use for excess farm products. In the food packaging sector, starch-based material has received great attention due to its biodegradability, edible, commonly available, abundant, low cost, nonallergic, easy to use and thermoprocessable [8]. Components used for the preparation of edible polymer can be classified into four categories: hydrocolloids, polypeptides, lipids, and composites. Hydrocolloid films have good barrier properties of oxygen, carbon dioxide, and lipids (Scheme 3) but not to water vapor (Scheme 1). Most hydrocolloid polymers also possess superb mechanical properties (Tables 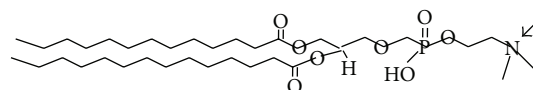1 and 2), which were quite useful for fragile food products. Among them, protein-based edible polymers were the most attractive ones. These polymers have impressive gas barrier properties compared with those prepared from lipids and polysaccharides. When being not moist, the O_2 permeability of soy protein-based film was 500, 260, 540, and 670 times lower than that of low density polyethylene, methylcellulose, starch, and pectin, respectively [9].

The mechanical properties of some protein-based edible polymers like rapeseed protein blended with gelatin (tensile strength—53.45 MPa) were better than polysaccharide and fat-based films [13]. Protein-based edible polymer can form bonds at different positions and offer high potential for forming numerous linkages (Scheme 2). Low value fish, such as lizard fish, were usually rejected from surimi manufacturing because of poor surimi gel strength. Hence, they have hitherto has mostly been used as animal feed and/or sold for low prices because of the lack of techniques for using them as foodstuffs. In order to obtain effective utilization of low value fish, including lizard fish, there was a need for more information on polymer produced from low value fish meat, their mechanical properties, and their applications. Nowadays there was a proecological tendency of dropping the use of synthetic materials. One of the problem-solving strategies was to replace the plastic packaging with new, biodegradable materials. Edible polymers seem to be a good alternative to the plastic foil. They were defined as material which can be consumed and provides a good barrier to moisture, oxygen, and solute movements for the food [14]. Because edible polymers were considered a packaging as well as a food component, they should fulfill a number of necessities, such as high barrier and mechanical efficiencies

Table 1: Tensile strength and elongation at break properties of hydrocolloid polymers [10, 11].

Polymer	Tensile strength (TS) MPa	Elongation at break (E %)
Cellulose derivatives	44–65	10–50
Collagen	1–70	10–70
Chitosan	10–100	20–80
Gelatin	25–140	7–22
Starch	35–46	1.7–3.4
Soy protein	3.7–4.5	152–160
Lentil protein	4-5	58–70
Whey protein	2.5–3.0	15–18
Peanut protein	3-4	147–150
Mung Bean	5.70–6.51	32–40
Low density polyethylene	16–18	>1000
Oriented polypropylene	50–60	73–100
Polyethylene terephthalate	81–85	19–25
Polyvinylidene chloride	65–75	18–23

and biochemical, physicochemical, and microbial stabilities; they should be nontoxic, nonpolluting, and of low cost [15].

2. Categories of Edible Polymer

The edible polymers can be comprised in any of the four categories:

(1) hydrocolloids, (2) polypeptides, (3) lipids, (4) synthetic and composite edible polymers.

2.1. Hydrocolloids. Hydrocolloids are often called hydrophilic polymers, of vegetable, animal, microbial, or synthetic origin, that generally contain many hydroxyl groups (polysaccharides) and may be polyelectrolytes. Polysaccharides were commercially available for use in food and nonfood industries as stabilizers, thickening and gelling agents, crystallization inhibitors, and encapsulating agents [16]. Many hydrocolloids were polyelectrolytes, for example, starch alginate, carrageenan, carboxymethylcellulose, gum arabic, chitosan, pectin, and xanthan gum.

Starch consists of two kinds of molecules, amylose (normally 20–30%) and amylopectin (normally 70–80%), which was primarily derived from cereal grains and tubers like corn (maize), wheat, potato, tapioca, and rice. Amylose was accountable for the film forming capacity of starch [17]. High amylose starch films have been made that were flexible, oxygen impermeable, oil resistant, heat sealable, and water soluble. Polymer of high-amylose corn starch or potato starch was more stable during aging [18]. Starch-based edible polymer exhibits physical characteristics similar to plastic polymer in which they were odorless, tasteless, colorless, nontoxic, biologically absorbable, semipermeable to carbon dioxide, and resistant to passage of oxygen. Since the water activity was critical for microbial, chemical, and enzymatic activities, edible starch-based films can retard microbial growth by lowering the water activity within the package.

The ability of polysaccharide-based (starch, carrageenan, and chitosan) coatings to extend the shelf life of strawberry fruit (*Fragaria ananassa*) was studied by Ribeiro et al., mainly for industrial applications [19]. Films, made with oxidized starch, showed improved tensile properties and fracture elongation percentage, with respect to those of the native starch [20]. *Kaempferia rotunda* and *Curcuma xanthorrhiza* are essential oils enriched in cassava starch-based edible coating which could extend patin fillet's shelf life and could be used as alternative fish preservation [21]. Alginates were derived from seaweeds and possess good film-forming properties that make them particularly useful in food applications. Alginate has a potential to form biopolymer film or coating component because of its unique colloidal properties, which include thickening, stabilizing, suspending, film forming, gel production, and emulsion stabilizing [22]. Alginate was used in various pharmaceutical preparations such as gaviscon, bisodol, and asilone. Gaviscon is a nonprescription medicine, which is taken by mouth to treat heartburn and gastroesophageal reflux disease. It was also observed that raft-forming chewable tablets prepared using an optimum amount of sodium alginate, calcium carbonate, and sodium bicarbonate could be an efficient dosage form in the treatment of gastroesophageal reflux disease [23]. 5-fluorouracil encapsulated with alginate beads was for the treatment of breast cancer [24]. Alginate based phenobarbitone tablets can be easily prepared for the treatment of chronic seizures in terms of controlled release of drug to obtain the effect over long time period [25]. Alginate was used extensively as an impression-making material in dentistry, prosthetics, life casting, and occasionally for creating positives for small-scale casting. It was also used in the food industry, for thickening soups and jellies. Edible polymer prepared from Alginates forms strong films and exhibits poor water resistance because of their hydrophilic nature [26].

Carrageenans were water-soluble polymers with a linear chain of partially sulfated galactans. These sulfated polysaccharides were extracted from the cell walls of various red seaweeds. Carrageenan film formation includes a gelation mechanism during moderate drying, leading to a three-dimensional network formed by polysaccharide-double helices and to a solid film after solvent evaporation [27]. It is used in desserts, ice cream, cream, milkshakes, salad dressings, sweetened condensed milks, and sauces, which is gel to increase viscosity. It is also used in beer, toothpaste, fruit gushers, soy milk, and so forth [28]. Recently, carrageenan films were also found to be less opaque than those made of starch [29].

Only four cellulose derivative forms are used for edible coatings or films: hydroxypropyl cellulose (HPC), hydroxypropyl methylcellulose (HPMC), carboxymethylcellulose (CMC), or methylcellulose (MC). Cellulose derivatives exhibit thermogelation; therefore when suspensions are heated they form a gel, whereas they return to original consistency when cooled [30]. The films casting from aqueous solutions of MC, HPMC, HPC, and CMC tend to have moderate strength, are resistant to oils and fats, and are flexible, transparent, flavorless, colorless, tasteless, water-soluble, and moderate barriers to oxygen. MC is more

TABLE 2: Water vapor permeability (WVP) properties of different polymers [12].

Film formulation	WVP ($g\,m^{-1}\,s^{-1}\,Pa^{-1}$)
Corn zein	5.35×10^{-10}
Corn zein plasticized with glycerol	8.90×10^{-10}
Fish skin gelatin	2.59×10^{-10}
Whey protein plasticized with sorbitol	7.17×10^{-10}
Wheat gluten plasticized with glycerol	7.00×10^{-10}
Gelatin (obtained from pigskin) plasticized with sorbitol	1.6×10^{-10}
Amylose	3.8×10^{-10}
Corn starch plasticized with glycerol	2.57×10^{-10}
Corn starch plasticized with sorbitol	1.75×10^{-10}
Amylomaize starch plasticized with sorbitol	1.21×10^{-10}
Hydroxypropyl methylcellulose withplasticizer and oil	1.90×10^{-10}
Amylomaize starch with sorbitol andsunflower oil	9.7×10^{-11}
Methylcellulose	8.70×10^{-11}
Methylcellulose 3%	$8.4\text{–}12.1 \times 10^{-11}$
Chitosan 2% (unknown source)	$3.66\text{–}4.80 \times 10^{-11}$
Chitosan 3%	$6.19\text{–}15.27 \times 10^{-11}$
Cellophane	8.4×10^{-11}
PVDC	2.22×10^{-13}
LPDE (low density polyethylene)	9.14×10^{-13}
HDPE (high density polyethylene)	2.31×10^{-13}

resistant to water and it is the lowest hydrophilic cellulose derivatives. However, cellulose derivative films are poor water vapor barriers because of the inherent hydrophilic nature of polysaccharides and they possess poor mechanical properties. A way to improve the moisture barrier would be the incorporation of hydrophobic compounds, such as fatty acids, into the cellulose ether matrix to develop a composite film [31]. As another example, Galus et al. [32] developed bilayers by adding a mixture of lipids (paraffin oil, paraffin wax, or a mixture of hydrogenated palm oil and triolein) onto a methylcellulose layer. Edible coatings made of CMC, MC, HPC, and HPMC have been applied to some fruits and vegetables for providing barriers to oxygen, oil, or moisture transfer.

Microcrystalline cellulose and powdered cellulose were used as inactive fillers in drug tablets and as thickeners and stabilizers in processed foods [33]. Cellulose powder was, for example, used in Kraft's Parmesan Cheese to prevent caking inside the tube. Methylcellulose (MC) and hydroxypropyl methylcellulose (HPMC) were used in coating formulations to reduce oil uptake in deep-fat frying potato strips and dough discs. MC coatings were more effective in reducing oil uptake than HPMC ones [34].

One method of enhancing the moisture barrier would be by incorporation of hydrophobic compounds such as fatty acids into the cellulose ether matrix to develop a composite film [5], for example, hydroxypropyl methylcellulose (HPMC). It was found that the moisture barrier was improved by stearic acid incorporation into the film forming solution; however, there were difficulties in preparing a homogenous composite film with both hydrophobic and hydrophilic compounds. For these reasons, homogeneous packaging film was prepared from HMPC by chemical

modification allowing an increase in hydrophobicity. Crosslinking of HPMC can be considered a useful approach to prepare nonwater soluble cellulose derivatives with interesting moisture barrier properties, which could be used as biodegradable packaging materials [35].

Pectin was a group of plant-derived polysaccharides that appear to work well with low moisture foods but was poor moisture barriers. Pectin was a heterogeneous grouping of acidic structural polysaccharides, found in fruit and vegetables, and mainly prepared from citrus peel and apple pomace. The conventional application was given the jellylike consistency to jams or marmalades and jelly beans, which would otherwise be sweet juices. In medicine, pectin increases the viscosity and volume of stool so that it is used against constipation and diarrhea [36].

The agar was hydrophilic colloids that have the ability to form reversible gels simply by cooling a hot aqueous solution. Agar exhibits features that make it convenient for cutting meats. It forms strong gels characterized by melting points far above the initial gelation temperature [37]. Agar has been used comprehensively as a gelling agent in the food industry. However, despite its biodegradability and its enormous gelling power, agar has not been used widely due to poor aging. Both photodegradation and fluctuations in ambient temperature and humidity alter agar crystalline, leading to the formation of microfractures and polymer embrittlement [38]. The effect of agar on the structure and the functional properties of emulsified edible films have been recently studied by Phan et al. [39].

Chitosan was one of the auspicious natural polymers with characteristics such as biodegradability, chemical inertness, biocompatibility, high mechanical strength, good filmforming properties, and low cost [40, 41]. Additionally, chitosan was a nontoxic cellulose-like polyelectrolyte polymer

hydrogel that was suitable for the production of artificial muscles, as this material undergoes a large volume change in response to changes in pH, temperature, or solvent composition [42]. Modern studies have suggested that chitin may play a role in a probable pathway in human allergic disease [43]. Chitosan was the second most abundant natural and nontoxic polymer in nature after cellulose. Some desirable properties of chitosan were that it forms films without the addition of additives and exhibits good oxygen and carbon dioxide permeability, as well as excellent mechanical properties and antimicrobial activity against bacteria, yeasts, and molds [44, 45]. Chitosan products were highly viscous, resembling natural gums. Chitosan can form translucent films to enhance the quality and extend the storage life of food products [46]. Pure chitosan films were generally cohesive and compact and the film surface has a smooth contour without pores or cracks. Chitosan was used in a wide range of applications such as wastewater treatment [47], separation membranes [48], food packaging drug delivery systems [49], and biosensors [50]. However, in sensor applications, the poor electrical conductivity of hydrogen results in a poor response time and a high operating voltage limits its applicability in devices. Hence, composites have been attempted by incorporating a rigid conducting polymer (such as PANI) into a flexible matrix (such as chitosan) to combine the good possibility of the matrix and the electrical conductivity of the conductive polymer [51]. The addition of tannic acid as a cross-linking agent of chitosan matrix has improved the mechanical properties and those related to the water affinity such as solubility, permeability, and contact angle [52].

Gums in edible-polymer preparation were used for their texturizing capabilities. In edible-forming preparations, guar gum was used as a water binder, stabilizer, and viscosity builder. Gum arabic, owing to its solubility in hot or cold water, was the least viscous one of the hydrocolloid gums. Xanthan gum was readily dispersed in water; hence high consistency was obtained rapidly in both hot and cold systems. A blend of guar gum, gum Arabic, and xanthan gum provided uniform coatings with good cling and improved adhesion in wet batters. The mesquite gum forms films with excellent water vapor (Tables 1 and 2) barrier properties when small amounts of lipids were added in their formulation [52].

2.2. Polypeptides. Polypeptide- (protein-) based edible polymer can be used for the individual packaging of small portions of food, particularly products that were not currently individually packaged for practical reasons, such as beans, nuts, and cashew nuts. Additionally, polypeptide-based edible polymer can function as carriers for antimicrobial and antioxidant agents. Additional possible application of polypeptide-based edible polymer could be their use of multilayer food packaging materials together with nonedible polymer. In this case, the protein-based edible polymer would be the internal layers in direct contact with food materials. Through functions relating to mechanical and barrier properties, protein-based edible polymer may be able to substitute synthetic polymer films.

Polypeptides were anticipated to be good oxygen barriers at low relative humidity. However, the fact that proteins were not totally hydrophobic and contain predominantly hydrophilic amino acid residues limits their moisture-barrier properties. Collagens were the major structural proteins of connective tissue, such as bone, hide, tendons cartilage, and ligaments. The main application of collagen films was as barrier membrane [53]. Films with a thickness of 0.01–0.5 mm are formed by air-drying a casted collagen preparation similar to ophthalmological shields and made of biodegradable materials. The drugs can be loaded into collagen membranes by hydrogen bonding, covalent bonding, or simple entrapment. They can be sterilized and become made flexible. This was used in ophthalmology in delivering drugs to yes and also used over wounds to make healing rapid and uniform [54]. Collagen polymer offers several advantages; it was biocompatible and nontoxic to most tissues; it has well-documented structural, physical, chemical, and immunological properties; it can be processed into a variety of forms, and it is readily isolated and purified in large quantities [55, 56]. Gelatin was unique in forming a thermoreversible substance with a melting point close to body temperature, which was mainly significant in edible and pharmaceutical applications. Fundamentally, gelatin was obtained by controlling hydrolysis from the fibrous insoluble protein, collagen, which was widely found in nature as the major constituent of skin, bones, and connective tissue. The characteristic features of gelatin were the high content of the amino acids glycine, proline, and hydroxyproline. Gelatin also has a mixture of single and double unfolded chains of a hydrophilic character. At approximately 40°C, gelatin aqueous solutions were in the sol state and form physical, thermoreversible gels on cooling. Gelatin was used to encapsulate low moisture or oil phase food constituents and pharmaceuticals [6]. Such encapsulation provides fortification against oxygen and light, as well as defining the amount of ingredient or drug dosage. In addition, gelatin films have been formed as coatings on meats to reduce oxygen, moisture, and transport of oil [57]. Addition, gelatin was competent to form clear and strong films and was used for microencapsulation and capsule coatings in food and pharmaceutical manufacturing. Microencapsulation is the process of surrounding or enveloping one substance within another substance on a very small scale, yielding capsules ranging from less than one micron to several hundred microns in size. Substances may be microencapsulated with the intention that the core material is confined within capsule walls for a specific period of time. Alternatively, core materials may be encapsulated so that the core material will be released either gradually through the capsule walls, known as controlled release or diffusion, or when external conditions trigger the capsule walls to rupture, melt, or dissolve [58]. Short chain gelatin peptides are effective in the treatment of stomach ulcers.

Zein was a prolamin protein and therefore dissolves in 70–80% ethanol [59]. Zein was a relatively hydrophobic and thermoplastic material [60]. Zein coating has also shown an ability to reduce moisture and loss of firmness and delay color alteration in fresh fruit like apples [61]. Furthermore, zein may similarly take part in the coating of conventional

packaging plastics. Zein's properties make it valuable in processed foods and pharmaceuticals, in competition with insect shellac. It was now used as a crust for candy, nuts, fruit, pills, and other encapsulated foods and drugs [62]. Wheat gluten was a water insoluble protein of wheat flour which was comprised of a mixture of polypeptide molecules and considered to be globular proteins. The cohesiveness and elasticity of the gluten give reliability to wheat dough and expedites film formation. The poor resistance of wheat gluten films to water vapor (Tables 1 and 2) was due to the hydrophilic nature of the protein and the substantial amount of hydrophilic plasticizer added to impart acceptable film flexibility. The properties of wheat gluten films can be upgraded by using a cross-linking agent such as glutaraldehyde or heat curing. Wheat gluten films can be abundantly biodegraded after 36 days in aerobic fermentation and within 50 days in farmland soil without liberating toxic products. It can be used in a wide selection of applications including milling, bakery products, meats, pasta, and breads [63]. Soy protein from soybeans has been extensively used as a food ingredient in nearly every food product available to the consumer, since it contains high nutrition and excellent functional properties. Since soy proteins were abundant, inexpensive, biodegradable, and nutritional, they show the potential to be developed as edible and biodegradable films [64, 65]. Milk proteins can be classified into two types: casein and whey proteins. The acid casein can be converted to functional soluble caseinates by neutralization through the addition of alkali. Edible protein films based on various caseinates can be obtained by solubilization in water followed by casting and drying. Laminated films that comprised casein did protect dried fruit and vegetables from moisture absorption and oxidation [12]. Caseinate-lipid emulsion coatings were fruitful in plummeting moisture loss from skinned carrots and zucchini [66].

Mung beans were of interest as a potential component of biopolymeric films because of their high protein content [67]. Bourtoom [68] synthesized and investigated the films from mung bean protein. It was found that the mechanical properties (tensile strength and elongation at break) of mung bean protein films had superior mechanical properties and water vapor barrier properties (Tables 1 and 2). These were better than other protein sources such as casein, soy protein isolate, wheat gluten, peanut proteins, and water-soluble fish protein films. However, the mung bean protein films still showed substantially lower mechanical and water vapor barrier properties (Tables 1 and 2), compared with some synthetic polymers (high density polyethylene, polyvinyl chloride, cellulose acetate, and polyester).

2.3. Lipids. Lipid compounds exploited as edible polymer consist of acetylated monoglycerides, natural wax, and surfactants. The most operative lipid substances were paraffin wax and beeswax. The primary utility of a lipid was to block passage of moisture due to their relative low polarity. In contrast, the hydrophobic characteristic of lipid forms thicker and more brittle films. Accordingly, they must be associated with film-forming agents such as proteins or cellulose derivatives. Generally, water vapor permeability decreases when the concentration of hydrophobicity phase rises. Lipid-based films were often maintained on a polymer structure matrix, usually a polysaccharide, to provide mechanical strength.

Paraffin wax was a plagiarist from the distillate portion of crude petroleum and consists of a mixture of solid hydrocarbon resulting from ethylene catalytic polymerization. Paraffin wax was permitted for use on raw fruit and vegetable and cheese. Carnauba wax was an exudate from palm tree leaves. Beeswax was produced from honey bees. Candelilla was obtained from candelilla plant. Mineral oil consists of a mixture of liquid paraffin and naphthenic hydrocarbon. Various types of wax were used as barrier films to gas and moisture (skin on fresh fruits) and to improve the surface appearance of various foods (e.g., the sheen on sweet). If applied as a thick layer, they must be removed before consumption (certain cheese); when used in thin layers, they were considered edible. Various types of wax (notably paraffin, carnauba, candellila, and beeswax) were the most efficient edible compounds providing a humidity barrier [69]. Acetylation of glycerol monosterate by its reaction with acetic anhydride yields 1-stearodiacetin. This acetylated monoglyceride displays the exclusive characteristic of solidifying from the molten state into a flexible, wax-like solid. Most lipids in the solid state can be stretched to only about 102% of their original length before rupturing. Acetylated glycerol monostearate, however, can be stretched up to 800% of its original length; water vapor permeability of this film was much less than that of polysaccharide film with the exception of methylcellulose or ethylcellulose. Acetylated monoglyceride coatings have been used on poultry and meat cuts to retard moisture loss during storage [6].

Shellac resins were a secretion by the insect *Laccifer lacca* and were composed of a complex mixture of aliphatic alicyclic hydroxyl acid polymers. Shellac was not generally recognized as safe substance; it was only permitted as an indirect food additive in food coatings and adhesives. It was mostly used in coatings for the pharmaceutical industry and only a few studies have been reported on foods [70]. Rosin and its derivatives were widely used in coating for citrus and other fruits. These coatings were designed primarily to impart high gloss at the time of inspection by the buyer, usually after application of the coating. When coatings were applied to fruit, they form an additional barrier through which gases must pass. Because coatings differ in gas permeance and ability to block openings in the skin, they have different effects on gas exchange. Citrus fruits with shellac- and wood resin-based coatings generally have lower internal O_2, higher internal CO_2, and higher ethanol content than fruits with wax coatings. High ethanol content, in turn, is an indication of off flavor. Shellac- and wood resin-based coatings also tend to increase the prevalence of postharvest pitting [6].

2.4. Synthetic and Composite Edible Polymer. Edible polymers may be diverse in nature, consisting of a blend of polysaccharides, protein, and/or lipids. This approach empowers one to utilize the distinct functional characteristics of each class

of film former. The combination between polymers to form films could be from proteins and carbohydrates, proteins and lipids, carbohydrates and lipids, or synthetic polymers and natural polymers. The main objective of producing composite polymer was to improve the permeability or mechanical properties as dictated by the need of a specific application. These heterogeneous films were applied either in the form of an emulsion, suspension, or dispersion of the nonmiscible constituents, or in successive layers, or in the form of a solution in a common solvent. The method of application affects the barrier properties of the films obtained. Kamper and Fennema [71] introduced the emulsion films from methylcellulose and fatty acids to improve water vapor barrier of cellulose films. Recently, many scientists have extensively discovered the development of composite polymer based on the work of Kamper and Fennema [71]. Examples of these studies were using lipid and hydroxypropyl methylcellulose, methylcellulose and lipid, methylcellulose and fatty acid, corn zein, methylcellulose and fatty acid, whey isolate and lipids, casein and lipids, gelatin and soluble starch, hydroxypropyl starch and gelatin, corn zein and corn starch [72], gelatin and fatty acid, soy protein isolates and gelatin [73], and soy protein isolate and polylactic acid [74].

Polyvinyl acetate was a nontoxic commercially important polymer prepared through emulsion polymerization which has been investigated as a coating film containing fungicides for protection of diverse foods and as a coating for pharmaceutical products. Biopolymers such as polysaccharides and proteins or even biocompatible synthetic polymers such as polymethyl methacrylate (PMMA) could be better alternatives for combinations with antimicrobial polymers aiming at antimicrobial chemotherapy in vivo. The use of antimicrobial macromolecular agents, however, was not restricted to in vivo applications; they may also be used ex vivo for water disinfection, food packing and preservation, and many antifouling applications [75]. For more than 50 years, techniques such as compression spray and dip coating and encapsulation have been used in the pharmaceutical industry to incorporate bioactive agents with polymers. Such polymers have largely included cellulose derivatives, polyethylene glycol (PEG), and poly (N-vinylpyrrolidone). Peppas and coworkers have initiated the use of pH-responsive complexation hydrogels of poly (methacrylic acid) grafted with PEG, referred to as P (MAA-g-EG), for oral protein delivery. Micelle-forming polymers, such as block copolymers of poly (ethylene oxide) and poly (propylene oxide), or Pluronics, have been thoroughly studied in drug delivery. Polyplexes formed by cooperative electrostatic interactions between polyethyleneimine (PEI) and DNA were widely studied for gene delivery [76].

An anionic copolymer based on methacrylic acid and methyl methacrylates was used for the coating of tablets and pills which resistance to gastric juice and improve the keeping property protection against moisture, light, and air coatings stable under tropical conditions [77]. Polymer spheres with a sugar coating on the outside and plastic coating on the inside have been made by European scientists. This gives them dual functionality to target and deliver drugs. Helmut Schlaad, from the Max Planck Institute of Colloids and Interfaces, Potsdam, and colleagues from Germany and Switzerland made the spheres by dissolving glycosylated polybutadiene-poly (ethylene oxide) block copolymers in water. When dissolved, the copolymers spontaneously formed hollow colloids called vesicles with a glucose coating on the outside and a poly (ethylene oxide) coating on the inside. The polymer vesicles could be used as living cell mimics or drug delivery vessels. They could be used to target drugs and biomolecules to injured or cancerous tissues [78].

3. Special Applications of Edible Polymers

Heightened consumer demand for enhanced keeping quality and freshness of foods has given rise to the concept of active packaging—a type of packaging that alters conditions surrounding the food to maintain product quality and freshness, improve sensory properties, or enhance product safety and shelf life. To meet the growing demand of recyclable or natural packaging materials and consumer demands for safer and better quality foods, new and novel food-grade packaging materials or technologies have been and continue to be developed. Examples of these packaging materials include biobased polymers, bioplastic or edible polymer packaging products made from raw materials originating from agricultural or marine sources [79]. The application of nanocomposites promises to expand the use of edible and biodegradable polymers [80]. It will help to reduce the packaging waste associated with processed foods and will support the preservation of fresh foods, extending their shelf life. Polymer composites were mixtures of polymers with inorganic or organic additives having certain geometries (fibers, flakes, spheres, and particulates). The use of nanoscale fillers was leading to the development of polymer nanocomposites and represents a radical alternative to these conventional polymer composites [81]. Pharmaceutical and food products were often coated with edible shellac, which must be of a sufficiently reduced viscosity. The application of polyvinyl acetate (PVA) was assessed as a potential protective edible coating in round-type tomato fruit on the green stage of maturity. The edible polymeric coating did not significantly affect brightness, weight loss, and RR; however, environmental conditions at which the fruits were maintained had significant effects. Despite the statistically nonsignificant effects, results showed that the PVA coating was associated with slight protection of the fruit compared to uncoated fruits [82]. Gelatin capsules [83] were small shells made from gelatin that were used to enclose various medications and supplements. Capsules can make it easier to take certain drugs and supplements, especially those in a powder or liquid form. The gelatin was almost odorless and tasteless, and it typically does not cause digestive problems for most people. Gelatin also usually was not interacting with other drugs. Gelatins from alternatives to mammalian species were gaining prominence, especially gelatins from marine fish species. Because of their good film-forming abilities, fish gelatins may be a good alternative to synthetic plastics for making films preserve foodstuffs [84].

Edible shellac coatings preserve fruits and vegetables during their long trek from the farm to your local produce stand.

To improve these coatings, Agricultural Research Service (ARS) scientists developed a new coating made of food-grade polyvinyl acetate that is cheaper to use and more effective than shellac at preventing postharvest fruit decay without discoloring the fruit. ARS has applied for a patent for this breakthrough [85]. It was cheaper to use and more effective than shellac at preventing postharvest fruit decay without discoloring the fruit. The coating can be applied to fruits and vegetables by dipping, spraying, or brushing on. Polyvinyl acetate (PVA) was a synthetic polymer that was used as an ingredient in chewing gum [86]. In addition to the polyvinyl acetate, the coating for fresh produce can include plasticizers, surfactants that aid coverage, gloss-enhancing additives, and other ingredients for specific uses, such as coating candy and baked goods. The new coating has several advantages over shellac, which slows fruit respiration and keeps the fruit firm [85]. Shellac tends to whiten or "blush" when it was wide open to moisture. This often occurs when apples were moved from cold storage to a humid environment. Another problem was that citrus fruits and some apple varieties develop "off" flavors when coated with shellac. And the fact that shellac coatings are primarily composed of insect exudates has made them objectionable to some consumers.

Several examples of snacks of orally soluble edible polymer, including their compositions and manufacturing techniques, were disclosed [87]. The snacks may include one or more layers of film that was orally soluble and fragments quickly upon placement in a human mouth without leaving substantial residue that can be felt by the human tongue or which needs to be swallowed or ejected from the mouth. An edible snack unit of tasty chocolate-flavored orally soluble polymer is comprised of a base medium formed in the shape of an ingestible snack, said base medium being formed at least in part from an orally soluble polymer, and said snack being orally soluble such that it disintegrates relatively slowly when placed on the roof of a human mouth and licked [88]. These film-forming edible polymers can include pullulan, hydroxypropyl methylcellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, polyvinyl pyrrolidone, carboxymethyl cellulose, polyvinyl alcohol, sodium alginate, polyethylene glycol, xanthan gum, tragacanth gum, guar gum, acacia gum, gum arabic, polyacrylic acid, methyl methacrylate copolymer, carboxyvinyl polymer, amylose, high amylose starch, hydroxypropylated high amylose starch, dextrin, pectin, chitin, chitosan, levan, elsinan, collagen, gelatin, zein, gluten, soy protein isolate, whey protein isolate, casein, polysaccharides, natural gums, polypeptides, polyacrylates, starch, gum karaya, gelatin, mixtures thereof, and others. Chewing gum consists of a gum base, sugar, corn syrup, softeners, and flavorings. The gum base was the insoluble part left in the mouth while chewing. The gum base was made of resins from trees, latexes, or the milky juices from plants, and man-made polymers. Thomas Adams desired to use the chicle as a rubber substitute, but instead, rubber was used to make chewing gum—unvulcanized styrene-butadiene synthetic rubber (SBR), others utilize polyvinyl acetate polymer (PVA) [89]. Modern chewing gum was made with a synthetic rubber, polyisobutylene, which not only makes it elastic, but also contributes to the obstinate, sticky

quality. US 3761286 [90] reveals a chewing gum composition comprising chewing gum and a water insoluble hydrophilic polymer of a hydroxyalkyl acrylate or methacrylate having controllably, releasable adsorbed there in a flavor, wherein the hydrophilic polymer can be poly-(2-hydroxy ethyl methacrylate). Gebreselassie et al. [91] invented swellable gum compositions which increase in volume upon chewing. The gum compositions include a super absorbent hydrophilic polymer in combination with a gum base or as a component of a gum base.

Researchers around the globe were trying to find ways of improving therapeutic efficacy of drugs by modifying the formulation technique, polymeric systems, and so forth. The use of novel edible polymers offers benefits. Care should be taken to properly select polymers while designing a delivery system. The eventual goal was to introduce cost effective, biocompatible, multifunctional, and less toxic polymers so that the delivery systems pass through the various phases of clinical trials and benefit the society. It was believed that the advances in polymer sciences will transfigure the design, development, and performance of polymer based drug delivery systems. PCL and PLA were chemically bonded onto starch and can be used directly as thermoplastics or compatibilizer [92]. Starch-g-PVA behaves good properties of both components such as processability, hydrophilicity, biodegradability, and gelation ability. Starch was a natural polymer which has widespread application ranging from a simple filler or binder to a more functional ingredient in the formulation of capsules, coatings, subcutaneous implants, and tablets [93]. It was easy to purify and soluble in water, which makes it convenient for delivery by injection and thus was considered as an ideal candidate for nanoparticle preparation. Protein based nanoparticles have the advantage of greater stability during storage and were easy to scale up as compared to other delivery systems [94]. It was a versatile protein carrier which was used in drug targeting for achieving better pharmacokinetic profile of peptide or protein based medicines.

Hyaluronic acid (HA), a natural polyanionic polysaccharide, was distributed widely in the extracellular matrix and the joint liquid of mammalians and approved for injections by the Food and Drug Administration (FDA) [95]. It was nontoxic, biocompatible mucoadhesive polysaccharide having negative charge and was biodegradable. It was mainly distributed in the connective tissue, eyes, intestine, and lungs. Guar gum was used as a binder, disintegrated in tablet formulations. It also acts as a stabilizer, emulsifier, thickening, and suspending agent in liquid formulations [96]. It has been widely used for colonic drug delivery applications. The swelling ability of guar gum was used in the delay of drug release from the dosage forms. Its utility as a carrier for colon specific drug delivery was based on its degradation by colonic bacteria. Chitin and chitosan derivatives were also used in the treatment of industrial effluents because of their affinity to metal ions. N-carboxymethyl chitosan has been used widely in pharmaceutical areas for achieving controlled release of drugs, orthopedic devices, and connective tissue. Pectin hydrogels can be used as a binder in tablet formulations and have been used in controlled-release matrix tablet

formulations [97]. Using an extruder/spheronizer, spherical pellets containing calcium pectate were prepared. These were then coated with pectin solution resulting in the formation of insoluble calcium pectinate gel around the pellets. The use of pectin to develop other oral controlled release drug delivery systems has been reported by many authors. Polysaccharides, for instance, contain hydroxyl groups that allow for direct reaction to drugs with carboxylic acid functions, thereby producing ester linkages that are biodegradable and thus facilitate the release of the drug in the body.

Finally, another relevant application of drug delivery systems was vaccines. Vaccines were classified into two groups, namely, proteins and nucleic acids. In both cases, they require polymeric carriers because they are susceptible to degradation by peptidases or nucleases. The DNA vaccine was a newly developed system. The DNA in this vaccine system encodes a protein antigen of interest that induces activation of the immune system. This DNA can be encapsulated into polymeric carriers, thereby protecting it from degradation [98]. It is then released into the phagosome, thus allowing it to reach the cell nucleus and express the foreign protein. Polymeric carriers of antineoplastic drugs can passively accumulate in cancerous tissues because of differences in the biochemical and physiological features of healthy and malignant tissues, while they can actively accumulate in the same tissues because they have been conjugated to targeting moieties. Scientists in the US have designed an ingestible electronic device that was composed entirely of edible materials and produces its own electric current [99]. Here the team designed and fabricated an ingestible current source consisting of flexible polymer electrodes and a sodium ion electrochemical cell. Having flexible polymer electrodes means they can be folded into an edible capsule, while the sodium ion cell serves as an on-board energy supply. The idea was for the patient to consume a pill that encapsulates the device. Polyesters based on PLA, PGA, and their copolymers PLGA and poly (ε-caprolactone) (PCL) have been extensively employed because of their biocompatibility and biodegradability. Polyesters have been used for the encapsulation of many types of therapeutic agents like cancer, bacterial, and parasitic infections [100].

4. Future Trends

A noble group of edible polymers was under enlargement, with the goal of allowing for the incorporation and/or controlled release of active compounds using nanotechnological solutions such as nanoencapsulation and multifaceted systems. Nowadays, nanotechnologies were being used to enhance the nutritional features of food by means of nanoscale additives and nutrients and nanosized delivery systems for bioactive polymeric compounds. Nanocomposites perception represents a motivating route for creating new and innovative materials, also in area of edible polymers. Materials with a large variety of properties have been realized, and even more due to be realized. Micro- and nanoencapsulation of active compounds with edible polymer coatings may help to control their release under specific conditions,

thus protecting them from moisture, heat, or other extreme conditions and enhancing their stability and viability. Coating foods with nanolaminates involves either dipping them into a series of solutions containing substances that would be adsorbed to a food's surface or spraying substances onto the food surface. These nanolaminate coatings could be elaborated entirely from food-grade ingredients and could include various functional agents such as antimicrobials, antibrowning agents, antioxidants, enzymes, flavorings, and colorant. In fact, the layer-by-layer electrodeposition technique [101] could be used to coat highly hydrophilic food systems such as fresh-cut fruits and vegetables including further vitamins and antimicrobial agents. The nanocomposite materials obtained by mixing natural, edible polymers, and sheets of crystalline solid layered offer a great variety of property profile. They were even able to compete, both in price and in performance, with synthetic polymeric materials. Consumer demands were driving research and development for alternatives to petroleum-based packaging materials including those with recyclable or edible properties, as well as those materials made from renewable/sustainable agricultural products. Edible films, gels, or coatings were considered biopolymers with numerous desirable properties and may be made from a variety of materials, including polysaccharides, lipids, and proteins, alone or in combination with other components. Edible biopolymers also have been developed from other sources and applied to foods, including fungal exopolysaccharides (pullan) or fermentation byproducts (polylactic acid).

5. Conclusions

The practice of edible polymers on many food products continues to develop. The potential paybacks of edible polymers as carriers of antimicrobial agents, flavors, antioxidants, coloring agents, vitamins, probiotics, and nutraceutical excuse continued research in this field of active packaging. A new trend in the storage of foods and in the food industry, nowadays, was the use of recyclable, renewable agricultural products for the production of packages, edible films, and coatings. Edible polymer technology plays an energetic role in drug discovery and was improving outcomes for patients today and addressing unmet therapeutic needs in the future. Vaccine drug delivery systems were now being proven to be patient friendly as they avoid the need to administer booster doses and provide a long term therapy in small doses. Edible polymer vaccines on the other hand open an attractive avenue for the oral delivery of vaccines. The development of new technologies to improve the delivery properties of edible polymer was a major issue for future research. At that moment, most studies on food applications have been conducted at a laboratory scale. However, further research should be focused on a commercial scale with the purpose of providing more accurate information that can be used to commercialize fresh cut products coated with edible polymers. Food industries were looking for edible polymers that could be used on a broad spectrum of foods and add value to their products while increasing their shelf life.

When the active ingredients (antimicrobials, antioxidants, and nutrients) were added to edible polymer, mechanical, sensory, and even functional properties can be dramatically affected. Scientists have researched new production methods to make coatings edible, safe, and fully functional. According to a broad range of research findings, edible polymer may be applied to prolong the shelf life of food products, control material exchange, and improve the products sensory properties, nutritive value, and attractiveness. Edible biopolymers were competitive as both commodity polymers and specialty applications where a specific functionality was valued. In a scenario where petroleum-based polymers were replaced by biopolymers, edible biopolymers from food crops would primarily be used in food applications, whereas other biopolymers could well cover the demand for commodity polymers. A continued development of functionality and processing was needed for a commercial breakthrough.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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