Review

Recent Developments in Edible Films and Coatings for Fruits and Vegetables

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Abstract: As a novel post-harvesting strategy, edible films and coatings for fruits and vegetables offer preservation measures to meet the growing needs of hunger and agricultural management. Albeit edible films and coatings would differ in their processing and physio-mechanical characteristics, but functionality is distinctly the same as they are designed to improve shelf-life, barrier, and nutritional properties of the food. With emerging concerns on sustainability, biomacromolecules have been widely considered for preparing edible films and coatings, which are Generally Recognized as Safe (GRAS) substances. Biopolymers, including polysaccharides, proteins, and lipids are the main sources of preparing edible films and coatings. These biomacromolecules make stable colloidal dispersions that deliver processing convenience with various formulation, blending, casting, coating, and film-forming methods. However, biopolymers based edible films and coating require improvements for their extended performance due to several structural and barrier limitations. Therefore, preparing blends and composites, incorporating target molecules to introduce different functionalities, and designing complex multilayers are among the many recent research approaches developed to overcome those limitations. Thereby ensuring enhanced food preservation and extended shelf-life, essential requirements of food waste management without or with minimal influence on the texture, flavor, and nutritional value of food and vegetables.

Keywords: biopolymers, Edible films and coatings, food packaging, polysaccharides, proteins

1. Introduction

Rapid population growth and the food supply chain crisis have exacerbated global hunger management. In addition, many geopolitical, socio-economical, and post-concurrent pandemic events have further aggregated the global food crisis. Global food waste

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generation in 2009 was 1.3 billion tons, and had reported that 32% of food produced for human consumption is wasted across the worldwide supply chain [1,2]. As per United Nations, Food and Agriculture Organization (FAO), approximately one-third (or 1.3 billion tons) of the global food production designated for human consumption is wasted annually [3]. The main challenge in modern agronomy is resolving the hunger crisis while delivering the adequate agricultural products and services. As a result, there is significant importance in developing novel food preservation and waste management strategies.

The magnitude of food waste can be equally expressed in terms of 3.3 gigatons (or 8% of the world's total) of CO_2 [4] and 250 km^3 of blue water consumption that would spread across 1.4 billion hectares (28% of the world's total) of agricultural landscape [4-7]. Global food wastes are responsible for nearly USD 2 trillion, including environmental costs (USD 700 billion) and social costs (USD 900 billion) [8], which compounded into approximately 10% of the gross domestic product (GDP) of the United States of America, USD 23 trillion [9]. Across the globe, developed and developing countries contribute USD 680 billion and less than USD 310 billion in food waste on average [10]. As a result, under sustainable development goals in 12.3, United Nations has declared to reduce per capita food waste by 50% by 2030 through enhancements in food security and environmental sustainability as a hunger management strategy amid projected exponential population growth [11]. Therefore, sustainable, innovative, high-performance packaging and coating solutions are required to accomplish the defined sustainable development goal.

Primary food commodities that generate food losses and wastes are classified into 10 subcategories according to FAO [12,13]: a) cereal and cereal products (wheat, maize and rice), b) roots and tubers (potatoes, sweet potatoes and cassava), c) oilseeds and pulses (from various sources such as peanuts, soybeans and olives), d) Fruits, e) Vegetables, f) meat, fish and seafood, g) dairy and dairy-related products, h) eggs and i) the products cannot be specified to the above. Among the above 10 subcategories, fruits and vegetables undergo the largest fraction of food losses and wastes in all regions from high to low-income countries [3,14]. On average, the total weight of vegetables (25%) and fruits (12%) contribute to total food waste [14]. Assessing wastage measures relevant to fruits and vegetables has many challenges. However, mechanisms of the waste generation that spread throughout the entire food chain can be indicated as follows [15]: 1) Primary production (in agricultural production and harvesting), 2) Secondary production (In postharvest handling and storage, processing, distribution, and retailing), 3) Consumption (in-household and out-of-home), Figure 1-a depicts the major wastes and losses in food consumption.

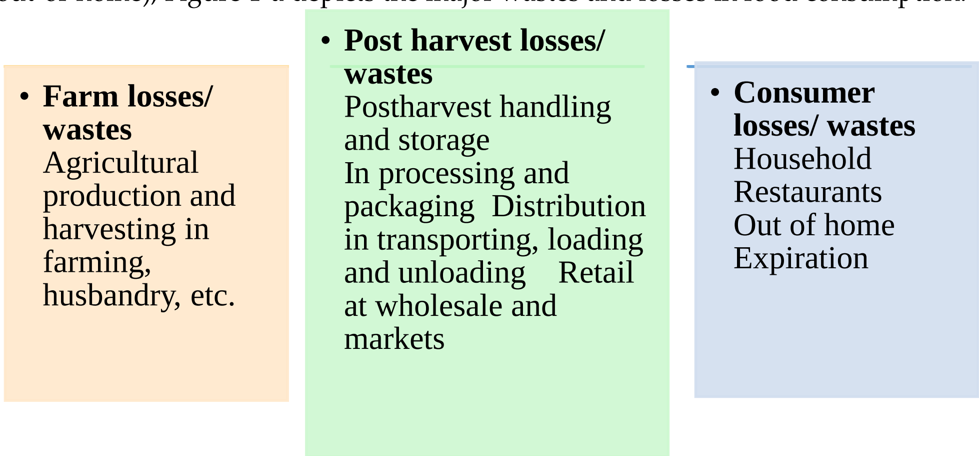

Figure 1-a. Main wastes and losses in food consumption [16]

Food that fails to meet the quality measures are considered food waste, and food losses lead to a decrease in food quantity or quality. Food waste and losses arise due to various reasons, including contamination, poor handling and storage, spoilage, microbial-

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fungal growth, and other factors [17,18]. Figure 1-b shows two major approaches to improve the food quality and mitigate food losses and waste include reducing preventa-ble food waste and the valorization of non-preventable waste.

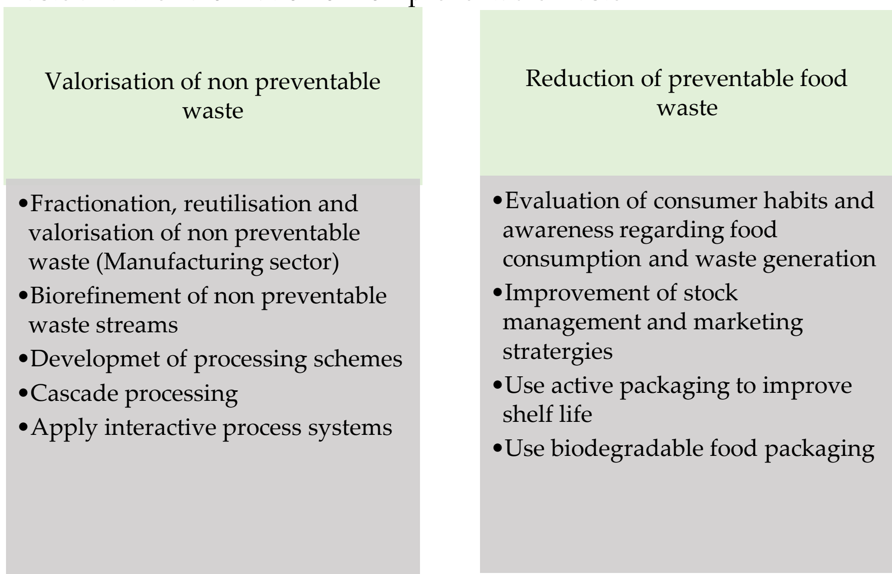

Figure 1-b. Main approaches to promoting food waste reduction and valorisation [16].

Edible films and coatings have recently been investigated as a commercially viable post-harvesting preservation strategy for fruits and vegetables. Interestingly, food can be consumed without removing the film or coating [19,20,21]. Edible films and coatings act as a thin protective barrier that extends shelf life in post-harvesting, processing, transportation, storage and consumption against dehydration, deterioration, spoilage and preserving appearances such as color, freshness, flavor and nutrients [22,23]. Furthermore, edible films and coatings do not devalue or alter the nutritional value of fruits and vegetables.

The applications of edible coatings and films are subjected to extreme safety guidelines. This involves good manufacturing practices and the use of food-safety materials monitored by the Food and Drug Administration (FDA) [24,25]. Another safety measure is to use Generally Recognized as Safe (GRAS) materials already approved by FDA [26]. However, not all GRAS substances are considered consumer safe, as there can be rare allergic reactions during consumption, including lactose intolerance from milk and Celiac disease from wheat gluten. Apart from FDA, the International standard organization (ISO) and European Unions (EU) are established regulatory organizations for safe applications of edible films and coatings [27].

Edible films and coatings for fruits and vegetables exhibit the potential to minimize preventable losses and waste, offering sustainable solutions to the global food crisis and hunger management. Therefore, this review explores the recent developments of edible films and coatings for fruits and vegetables. Furthermore, comprehensive coverage on characteristics and functionalities, processing, major types and their structural, chemical significance, different sources and their performances, health effects of edible films and coatings on health, and recent trends in edible films and coatings specific to fruits and vegetables have also been discussed.

2. Characteristics and functionalities of edible films and coatings

The definition of edible films and coatings generally refers to a thin barrier within the thickness range of 0.3 mm layer made from edible material that can be directly consumed or eaten [28]. Edible films and coatings are widely applied in contact with minimally processed fruits and vegetables. Even though the main functionality of edible films and coatings is similar, their processing techniques are used. For instance, edible films are pre-formed before contact with fruits and vegetables. In contrast, edible coatings are

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usually formed on the surfaces of fruits and vegetables, and layer formation occurs directly [29,30]. Both techniques make thin barriers from rigid matrices that can be further functionalized for extended applications.

When developing edible films and coatings, the following considerations are critical to assess [31]:

a) interactions with the food texture and surfaces,
b) ageing and prolonged performance with the shelf life of the food in contact,
c) changes in flavor, color, and texture of food due to the interactions with edible films and coatings,
d) response and sensitivity under storage / environmental conditions, and
e) processing conditions, including temperature, color and thickness.

2.1. Key functionalities of edible films and coatings

Edible films and coatings act as a protective barrier and provide a controlled atmosphere around fruits and vegetables. Figure 2 summarizes the key functionalities of edible films and edible coatings in fruit and vegetable preservation.

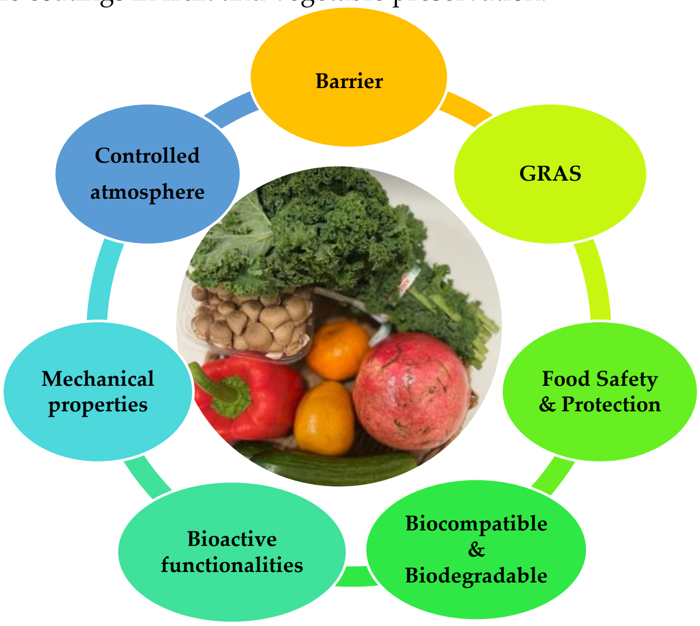

Figure 2. Major functionalities of edible films and coatings in fruits and vegetables preservation.

The major functionalities of edible films and edible coatings are as follows [31-33]:

- Protection from transport, handling, mechanical damages, and UV radiation
- Barrier properties:

a) Moisture barrier: minimize water vapor transmission to prevent dehydration.
b) Gas barrier: control oxygen and carbon dioxide levels pass through the protective layer
c) Volatile organic compounds (VOC) barrier: protection against organic vapors such as aromas and solvents and other additives and pigments.

- Prolong shelf-life
- Bioactivity: show antimicrobial and antifungal properties and acts as probiotics.
- Biodegradability

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- Structural integrity: melt above 40 °C without decomposition, water resistant, easily emulsifiable, non-sticky, or non-tacky, and deliver efficient drying.
- Maintain food quality: minimal influence on texture, flavor, or color
- Formulated from economical, relatively abundant, consumer safety GRAS materials

2.2. Common preparation methods of edible films and coatings

When processing edible films and coatings, raw material and the number of layers play a significant role. In addition, the nature of the edible film may govern overall film strength, solubility, surface activity, appearance, flavor, and texture in the mouth [24]. Edible films are primarily manufactured as sheets or thin wrap films and later used to cover food intact. Edible films and coatings are commonly prepared using melt extru-sion and solvent casting techniques.

2.2.1. Melt extrusion method

Main unit operations of melt extrusion include formulation, melt blending, extrusion, cooling, and storage. Sheet extrusion, blown film extrusion, and reaction extrusion are commercialized melt extrusion techniques applied to prepare edible films [34,35]. Generally, the extruder temperature and operating conditions are governed by the thermal and rheological properties of polymers. Interestingly, biopolymers with thermoplastic properties exhibit better performances in melt extrusion. However, process aids and plasticizers are added to enhance polymer melt flow during the extrusion [36]. Ultimate film quality and clarity are governed by the degree of crystallinity of the poly-meric matrix and the cooling rate. The polymeric matrix in the melt state undergoes recrystallization when cooling and may change its polymorphic state [37]. Higher crystallinity may deliver extended barrier performance over highly amorphous polymers. However, the amorphous fraction of polymers may significantly influence optical properties, controlling the clarity and transparency of films. The thickness of the films can be adjusted from rotor speed and extruder parameters. However, formulation errors may lead to phase separation, non-uniform distributions, and irregularity in thickness.

2.2.1. Solvent casting method

Solvent casting is another widely used mainstream film processing method [38]. Unlike melt extrusion, solvent casting can be complicated depending on the solvent system and intended application [39]. Hence, stable colloidal systems need to be designed based on the solubility parameters of the polymers and the pre-selected solvent system. Moreover, to obtain stable and uniform films, essential process aids such as emulsion stabilizers, either oil or wax-based surfactants, are added to facilitate mixing. In addi-tion, wetting and leveling agents are added to control uniform wetting and surface ten-sion and surface defects such as bubbles, pinholes, craters, and defoaming agents to avoid aeration of colloids, respectively [40,41]. Solvent removal and solidification are critical factors in the solvent casting method. The solidification process is vital for the quality of the film or coating and is facilitated by a precise drying system as drying transforms fluid into a solid-state transition. Factors including coating formulation, number of layers, wet film/coating thickness, viscosity, solution solids, solution temperature, coating accuracy, and coating substrate must be considered when designing coating or film in the solvent casting method.

Various drying systems are used in the solvent casting method depending on the requirements. Hot air convection, hot air impingement, steam, infrared, hot air flotation, and zoned drying are commonly employed drying systems. In addition, Rod, knife, and spin coating methods are used in the batch film and coating processes [42]. In recent

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approaches, multilayer films and coating have been tested to enhance performance [43]. Moreover, lamination, calendaring, slot extrusion coating methods, dip coating, and spraying have also been employed in edible films and coating processes [44].

3. Types of edible films and coatings and their structural and chemical significance

Most of the edible films and coatings are made from biomacromolecules, as they originate from natural sources with minimal toxicity and comply with GRAS. Also, biomacromolecules give extra processing convenience due to their biocompatibility and biodegradability. Matrices of biomacromolecules used for edible films and coatings can be segmented into three main categories considering their physicochemical properties, as in hydrocolloids (polysaccharides and proteins), lipid colloids (fatty acids, acylglycerol, waxes), and composites as illustrated in Figure 3.

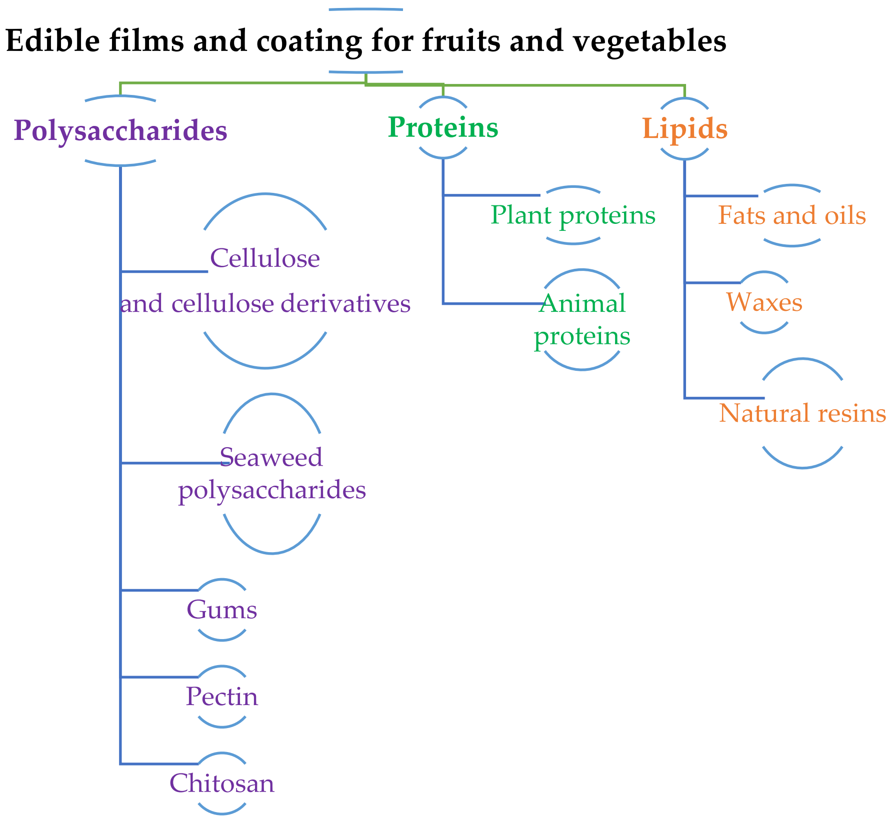

Figure 3. Sources of Edible films and edible coatings.

3.1. Hydrocolloids

Hydrocolloids are made of hydrophilic dispersion of colloids constituted of long-chain polymers, may be dispersed entirely or partially, or tend to swell in contact with water [45]. Dispersion of hydrocolloids results in changes in their physicochemical properties by forming stable gels or changing properties, including viscosity, thickening, emulsion, and stability [46]. Hydrophilic functional groups in biomacromolecules and hydrogen bonding play a crucial role in forming a stable hydrocolloid system [47]. Due to the presence of hydroxyl functional groups in their structure, polysaccharides are widely employed to prepare hydrocolloids. However, not all polysaccharides form hydrocolloids because of water insolubility, and the formation of hydrocolloids using water soluble sugars is limited by their low molecular weight [48]. Proteins are also classified as hydrocolloids in some contexts due to the structural hydrophilicity and intrinsic polydispersity of

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protein colloids [45,49]. Proteins sources, including gelatin, milk, egg, and isolates of proteins derived from vegetables, make stable hydrocolloids.

Based on origin, hydrocolloids can be divided into a) plant-based, b) animal-based, and c) modified hydrocolloids. Plant-based hydrocolloids are prominent in edible films and coatings due to their natural abundance and nontoxicity. Plant-based sources of hydrocolloids include pectin, starch, guar gum, locust bean gum, mannan, gum arabic, gum ghatti, tragacanth, agar, alginates, and carrageenan. Animal-based hydrocolloids are gelatin, collagen, whey, egg, and milk protein [50,51]. Among many animal-based hydrocolloids, gelatin is extensively used due to its relative abundance and easy extraction. Chitosan is another prominent animal-based hydrocolloid [52].

Hydrocolloids have also been modified to enhance stability, quality, safety, and nutritional values. For instance, hydrocolloids prepared from polysaccharides, including cellulose, pectin, and starch, have been modified to achieve extended performance [51]. Cellulose derivatives, including nano-fibrillated cellulose (NFC), nanocrystalline cellulose (CNC), carboxymethyl cellulose, methylcellulose (MC), hydroxypropyl methylcellulose (HPMC), and hydroxyethyl cellulose have been tested as hydrocolloids in the food industry [53]. Food hydrocolloiods have been prepared by modified starch with different granule sizes, amylose-to-amylopectin ratios, and functional properties such as gelling, thermal and textural [49]. Pectin extracted from fruits and vegetables is modified through esterification into low methyl ether pectin. High methyl ether pectins are industrially used as gelling and thickening agents [54,55].

Hydrocolloids of polysaccharides edible films (EF) and edible coatings (EC) for F&V are formulated from various high to low molecular weights and often exhibit thixotropic rheology. Gums of polysaccharides form micelles when dispersed in aqueous media through solvent-polymer hydrogen bonds [56]. As per the degree of hydrogen bonding and the intermolecular association between micelles and water, gums form a stable polymer-solvent system immobilized by water molecules [57].

Hydrocolloid systems of polysaccharides, hydrogels, and gums lead to highly viscous shear-thinning colloidal dispersions that are ideal for coating and film formation methods [58]. These hydrocolloids provide processing convenience and flexibility for continuous and batch production. However, formulation parameters such as concentration, drying conditions, and solidification methods (for water evaporation) are crucial to achieving quality EF&EC. During drying, EF&EC from polysaccharide molecules may rearrange into characteristic structural matrixes forming semicrystalline domains. The semi-crystallinity governs vital characteristics of EF&EC stability, solubility, durability, viscoelasticity, and physicomechanical, optical, and barrier performance [59].

3.2. Lipid Colloids

Colloids of lipid exhibit hydrophobic and emulsion properties. Lipid matrixes used for edible films and coatings are mostly fatty acids, glycerides, and waxes. Colloids from lipids are stabilized by physicochemically compatible surfactants [60]. Surface reactions and surface adsorption of lipid colloids are governed by the physical state of the hydrophobic matrix. Even though the lipid matrix is hydrophobic, it provides excellent affinity for binding other molecules, such as protein, to their surface [61].

In lipid colloids, the lipid-water interface provides a highly active reaction surface and moisture barrier performance. Common lipid colloids used in edible films and coatings include liposomes, micelles, nano emulsions, microemulsions, and solid lipid nanoparticles [62]. Furthermore, lipid colloids can be broadly categorized into [63] (i) solid-in-liquid

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dispersions, (ii) liquid-in-liquid dispersions, and (iii) dispersions of self-assembled molecules. For edible films and coating, lipid colloids are reliable due to their structural effectiveness in absorbing our digestive system, nontoxicity, favorable emulsion properties, physicochemical stability of lipid carriers, and surface reactivity [62,64]. Surface reactivity and surface binding of lipids are unique properties that have been extensively considered in designing edible films and coatings [65].

3.3. Composites

Composite films are heterogenous in nature by which a combination of biopolymers are used to achieve a distinct functional property from the each ingredient used [65]. When combining, hydrophobic and hydrophilic materials are blended to achieve targeted property [66]. As the biopolymers, carbohydrates, proteins and lipids are widely used in combinations [67]. Generally, fats are used to reduce water transmission, proteins provide mechanical stability and carbohydrates are used to control the exchange of gasses [22].

According to the number of biopolymers combined, composite edible films and edible coatings are categorized as binary and ternary [68]. Combinations of composites and be synthetic and natural polymers [65]. Binary films and coatings comprise of combinations such as protein-protein, carbohydrate-carbohydrate, carbohydrate-protein and lipid based binary films.

In order to meet packaging preservation that requires high performance, complex solutions such as multilayer, smart, and intelligent EF & EC have been designed. For such applications, natural polymer composites are used to achieve synchronized property enhancements and introduce functionalities. Composite applied for EC&EF are a combination of two distinct phases, continuous and discontinuous [68].

In conventional composites in EF&EC, the continuous phase consists of an edible biocompatible polymer, and the discontinuous phase consists of a filler or a modifier. Modern EF&EC can be formulated from multi-components, and there can be metaphases to deliver the complex functionalities required for EC & EF. All the elements in EC&EF must be covered by FDA and GRAS guidelines. Furthermore, composites for EC & EF can be designed for controlled release and active delivery of different molecules vital for F & V, such as antioxidants, vitamins, and antimicrobial compounds [68].

Bilayer composites of EC & EF use for F&V have been researched over the years to lessen the disadvantages of EC & EF made from single layers made from natural hydro and lipid colloids. Often context composites of EF & EC refer to a binary component system with two layers or more with distinct performances, made from lipid-hydrocolloids or different combinations of protein-protein and carbohydrate–carbohydrate, and protein - carbohydrate [69].

Composite films can be prepared in two different methods; layer form or emulsion of film-forming materials. Based on the number of polymers used, layered composites are classified as binary and ternary. Emulsion film forming method is considered more effective than layer form as layered method required more casting and drying processes and delamination can reduce the effectiveness over the time [66].

Bilayer films can be developed mainly from two different methods [70,71]; 1) Two steps method, continuous application over another matrix or 2) Single-step method- homogeneously dispersing with distinct emulsions or colloidal systems. However, bilayer films from the two-step method are described to be the best performing as the films from the

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dispersion method may tend to shrink method of application affects the characteristics of the films and coatings such as barrier properties [65].

4. Polysaccharide based edible films and coatings

Polysaccharides can be abundantly found in renewable sources such as plants, sea-weeds, algae, and microbial. However, the categorization of polysaccharides is complex. Based on their composition, polysaccharides can be divided into homoglycans with a single constituent in the main polymeric chain and heteroglycans with two or more constituents in the main polymeric chain, alternate or irregular sequence [72,73]. Physico-chemical properties of polysaccharides vary from the degree of polymerization, molecular weight, ring size, anomeric configuration, linkage type, and absence or branching [72,73]. Hence, solvent polysaccharide interactions of polysaccharides make them soluble, insoluble, and partially soluble. Insoluble polysaccharides are compact chains and tend to partial crystallization. Polysaccharides can act as sacrificing agents which can preserve moist nature of foods [65].

Polysaccharides can contain various linkage patterns such as C (1 → 3), C (1 → 4) and C (1 → 6) and the linkage type affects the solubility. Among the linkages C (1 → 6) linkage provides easier solubility [74]. The degree of branching and linearity of polysaccharides influence the gel forming ability and the stability of gels [75,76].

The solubility of polysaccharides governs by many factors. Hydrocolloids of poly-saccharides make stable hydrogen bonds with water because of their extremely hydrophilic nature [45]. Due to heterogeneity in natural polysaccharides in plant cell walls, certain pre and post-treatments and non-hazardous modifications may require for processing EF & EC [77,78]. Polysaccharidal solutions deliver green processing from EF & EC; such green formulation and processing routes can be established based on hydrocolloid chemistry in aqueous media [79]. This chemistry leads to reversible colloidal transformation from solution to a gel state, making most polysaccharide hydrocolloids make physical hydrogels [80].

Polysaccharides EF & EC are highly compatible with F & V as they are colourless and provide excellent barrier protection against oils and organic molecules [29,68,81]. However, polysaccharides EF&EC are less resistant to moisture and do not deliver sufficient water barrier, which is essential for F & V. Polysaccharides EF & EC are efficient in modifying the controlled environment in the preservation of F & V by reducing the respiration rate from moderate permeability to O_2 and CO_2 [82]. In recent research, active components such as antimicrobials, vitamins, antioxidants, bactericides, and preservatives are incorporated into polysaccharide matrixes. Active EF & EC from polysaccharides solutions can be designed through functionalization, crosslinking, and composites for advanced preservation, control release, bioactivity, protection, and water barrier properties [83]. The application of polysaccharides may still be economically reliable for F & V applications with a short shelf-life expectancy and have extremely thick natural protection peels as a low-cost preservation strategy. However, EF & EC may not interfere with the taste due to oxidation or rancidity and, when needed, dope other chemicals for extended performance.

4.1. Cellulose and cellulosic derivatives

Cellulose is the most abundant renewable material on earth and can be extracted from the cell wall of plants, algae, tunicates, and some bacteria [84]. The linear homo-polymer/homoglycan structure of cellulose is low density and has a degree of crystallinity varying between 40-70% having disordered, loosely packed amorphous regions that are susceptible to surface reactions [85,86]. In densely packed crystalline domains, intramolecular hydrogen bonding is prominent [87]. Cellulose anhydro glucose units of D-glycose are linked through β-1,4-glycosidic bonds [88]. The ultimate structure-property relationship of cellulosic edible films and coatings is governed by the degree of crystallinity, polymerization, and polymeric chain length [89]. Having significant importance, the degree of crystallinity of cellulosic fibers embodies toughness, strength, and fiber-fibril characteristics [90]. Furthermore, the degree of crystallinity and ratio between amorphous to crystalline domains directly impact physico-mechanical, optical, and barrier properties.

In the processing and formation of EF & EC, hydroxyl chemistry is responsible for hydrophilicity, chirality, chemical functionalization, insolubility in most aqueous sol-vents, infusibility, and solvent resistance of cellulose hydrocolloids [91]. Cellulose is insoluble in water, alkalines, and modifications may alter the solubility of hydrocolloids [92]. The low film stability and poor oxygen and carbon dioxide barrier are the key challenges with cellulosic hydrocolloids. Therefore, several research approaches have been executed to overcome the challenges of EF & EC from cellulosic and cellulose derivatives, such as modification, introducing functionalization, and composites. Cellulose extraction from high plants involves multistep chemical processing, drawing concerns for EF & EC under GRAS guidelines. Cellulose can be processed into hydrogels that are commercially used as micro cellulose or nanocellulose, which differ from the average fiber dimension of the hydrogel fibres. Nanocellulose from microbial sources is the purest form of cellulose. Hence, ideal for EF & EC. Crystalline nanocellulose, a product of chemical and enzymatic digestion of amorphous regions, has enhanced mechanical and barrier properties resulting from a high degree of crystallinity.

Processing convenience and formulation of cellulose-based edible films and coatings can be improved by modifying cellulose following esterification routes in the presence of chloroacetic acid or methyl chloride or propylene oxide to achieve carboxymethyl cellulose (CMC), methylcellulose (MC), hydroxypropyl cellulose (HPMC) or hydroxypropyl cellulose (HPC) respectively [29]. Edible films and coatings prepared from cellulose esters possess properties including odorless and tasteless, flexibility with moderate strength, optical features, repellent resistance to oil and fats, water solubility, moderate moisture and oxygen transmission, controlled release of bioactive, non-toxicity [93], compatibility with composites and laminates, and efficient membranes and separation [93,94]. Cellulose and cellulose derivatives provide strong adhesion between fibre and cellulosic interface, making them ideal for edible films and coating for fruits and vegetables [94].

Cellulose and its derivatives have been widely investigated as edible films and coatings. For instance, hydroxy propyl methyl cellulose (HPMC) was successfully tested as an edible coating for blueberry [95]. Sodium carboxymethyl cellulose (CMC) and hydroxyethyl cellulose (HEC) crosslinked with citric acid for probiotic entrapment in food, including fruits and vegetables [96]. Functionalized encapsulation is a promising, inexpensive, and environmentally friendly approach to improving the preservation properties of edible food and coatings. Plant based essential oils lemongrass (*Cymbopogon citratus*), rosemary pepper (*Lippia sidoides*), and basil (*Ocimum gratissimum*) encapsulated cellulose acetate, cellulose acetate propionate and cellulose acetate butyrate were investigated for fragrance and air freshening effects of edible films and coatings [97]. Essential oil incorporated cellulose esters enhance the physico-mechanical properties and plasticizing effects of edible films and coatings. Oils embedded in cellulose ester minimize moisture loss and improve the water and gas barrier [98,99]. For example, TEMPO oxidized cellulose esters were developed to enhance the biorefractory of edible films [100]. Cellulose esters were also substituted with acyl groups with C_2 to C_{18} to enhance the water and oxygen barrier properties and hydrophobicity [101]. The dispersed phase effect of steric acid was studied to improve the water barrier of CMC edible films. It has been concluded that the water vapor transmittance rate (WVTR) significantly dropped with loading more steric acid to CMC [102]. Microcrystalline cellulose (MCC) modified with lipid coating and unmodified

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composites of hydroxypropyl methylcellulose (HMC) was investigated for mechanical and barrier characteristics and reported up to 50% increase in mechanical properties and 40% to 50% improvement in water barrier with loading unmodified and modified MCC [102]. Composite biofilms of wheat gluten and cellulose acetate phthalate have also been tested for better permeability toward water and oxygen [103]. The preparation of polysaccharide and lipid bilayers is another approach to improve the barrier performance of edible films from cellulose and cellulose derivatives. Here polysaccharides facilitate the film forming, and lipid matrixes act as barriers against moisture transfers [104]. Edible films from MC, and HPMC, with saturated fatty acids with carbon chains ranging from C_{16} and C_{18}, have been designed with a thin lamination of beeswax at the cellulose-lipid interface to improve moisture barrier properties [105]. Bilayers of edible films from Corn Zein and MC have been reported for reducing WVTR [106]. In similar research, corn zein fatty acid was cast onto MC films, and the effect of corn zein-fatty acid concentration on mechanical properties and water vapor permeability of bilayer laminated edible films of cellulose ethers has been studied [107]. Hydrophobic ethyl cellulose (EC) and hydrophilic carboxymethyl chitosan hydrogel were used to design one-way bilayer films as humidity regulators to extend the browning of white button mushrooms [108].

Edible films of bacterial nanocellulose and konjac glucomannan composites were reported for good blending dispersion and film formation properties due to strong hydrogen bonding between two compatible matrixes [109]. Similar research has developed edible composites using sago starch and CMC nanoparticles to improve mechanical properties for applications in fruits and vegetables [110]. Another study has introduced CMC as a reinforcing filler to strengthen the mechanical properties of gelatin edible films [111]. In addition, antibacterial edible films and coatings cellulose esters were developed by preparing composites with chitosan [112] and silver nanoparticles [113]. Moreover, ginger and olive plant oils incorporated bacterial cellulose and CMC composites have been studied as antimicrobial edible coating for orange and tomato [114]. Cellulose based composites also show excellent.

Carrageenan exists in three forms, kappa carrageenan (κ-carrageenan), iota carrageenan (ι-carrageenan) and lambda carrageenan (λ-carrageenan) [115]. Edible films and coatings from carrageenan are known for excellent mechanical and barrier properties [116]. For example, edible films and coatings prepared from iota carrageenan exhibit excellent barrier properties for oxygen and preserving the deterioration and flavor of fruits and vegetables [117]. However, lambda carrageenan does not form stable gels and has limited use for preparing edible films and coatings [118]. The double-helical conformation of kappa carrageenan and linear structure form efficient three-dimensional (3-D) gels under standard cationic colloidal conditions. Therefore, widely used for edible films and coatings in food. Carrageenan based films and coatings also help minimize moisture loss, turgor, and oxidation [119]. Carrageenan based edible coatings show enhanced properties upon ultraviolet treatment on post-harvested longan fruits [120]. Carrageenan gum has been blended with different starch sources to achieve desired features. For instance, edible films developed from starch/carrageenan displayed improved mechanical and WVTR properties [121].

Edible films from rice starch and ι-carrageenan plasticized with stearic acids exhibited high physicomechanical and barrier properties with increasing the concentration of carrageenan and low loading of stearic acid [122]. Following a similar trend, edible composite films prepared from pearl millet starch and carrageenan gum using glycerol as a processing aid, increasing starch educed the water vapor permeability and mechanical properties [123]. Blends and composites have been used to improve the performances of carrageenan edible films and coatings. For instance, transparent, stable films and coatings prepared from carrageenan and rice starch hybrids displayed enhanced UV protection,

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oxygen barrier, and hydrophobic characteristics [124]. Surface properties of iota carrageenan edible films blended with glycerol plasticizer, glycerol monostearate surfactant, and fat showed improved surface properties [125]. Edible films prepared from κ-carrageenan, ι-carrageenan, and alginate blends have shown improved optical, barrier, and tensile performances [126]. I-carrageenan and sodium alginate blends exhibited good mechanical characteristics with emulsion stabilizers [127].

Agar is a heteroglycan with gelling and non-gelling fractions of agarose and agaropectin and is readily soluble in hot aqueous medium [128,129]. Gels from agar are known for making thermoreversible gels up to 55 °C – 60 °C having lower viscosity profiles which are ideal for edible films and coatings [129]. Agar-based edible films and coatings are generally stable and transparent with good mechanical properties [130]. However, edible films and coatings prepared from pure agar may draw concerns in industrial applications due to brittleness, low elasticity, less thermal stability, relatively medium gas barrier performance, and high-water vapor permeability [131]. Interestingly, edible films prepared from starch-agar-maltodextrin blends displayed improved barrier properties due to extensive hydrogen bonding and hydrophobic aggregations [132].

Blended films prepared using binary combinations of agar, cassava starch, and arabinoxylan have reported a decline in mechanical properties at higher loadings of cassava and arabinoxylan, wherein the water barrier significantly improved in agar-arabinoxylan films [133]. A similar study reported that edible agar films and coatings for fruits and vegetables required an optimum concentration of glycerol plasticizer to achieve good physical and mechanical properties [134,135]. Edible films and coatings of agar doped with essential oils exhibited improved mechanical and water barrier properties with antimicrobial and antioxidant activities [136]. Agar-based composites with nanoparticles and natural active ingredients have also been investigated for improving physiomechanical, thermal, and antioxidant characteristics suited for edible films and coatings [137].

4.2. Pectin

Pectin is one of the main constituents of plant cell walls with an anionic polysaccharide structure of β-1,4-linked *α*-D-galacturonic acid in which uronic and carboxyls can be fully or partially methyl esterified [138]. It is produced as a byproduct from the industrial processing of lignocellulosic biomass and used to develop active food packaging materials [67]. Edible films and coatings prepared from pectin show excellent mechanical and low water barrier characteristics [139]. For instance, pectin extracted from pineapple peels was tested as a natural plasticizer for biopolymer-based edible films and coatings [140]. Edible films and coatings prepared from pectins display improved O_2, CO_2, and ethylene barrier properties in the presence of hydrophobic additives [141]. The natural plasticizing effect of pectin has been further investigated using alginate/pectin blends for edible films and coatings [142]. It was reported that the WVTR of the films and coatings improved with increasing plasticizer and probiotic storage at different temperatures [142].

Pectin based edible films and coating were used as the carrier for oregano essential oils to improve antimicrobial properties against food related microorganisms [143]. Novel red color pectin extracted as a byproduct from *Hibiscus sabdariffa L.* showed excellent film-forming properties and was studied for strawberry preservation [144]. In a similar study, edible films and coatings prepared from pectin/whey protein blends cross-linked by transglutaminase were applied to preserve freshly cut fruits and vegetables [145]. This research demonstrated that fresh cut apples, potatoes, and carrots coated with pectin/whey protein blends were preserved for ten days [143]. Conversely, edible films and coatings developed from pectin/protein blends crosslinked by transglutaminase displayed a reduction in the hardness and chewiness of the fruits and vegetables they

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contained. Candelilla wax and pectin blends were also developed to improve hydrophobicity and barrier performance [146].

4.3. Chitin and Chitosan

Chitin is the second most available natural biopolymer on earth and is extracted from the exoskeleton of crustaceans, cell walls of fungi, and other species [147]. The repeating unit of chitin consists of poly (ß-(1-4)-2-acetamide D-glucose) [148]. Chitosan is derived from the deacetylation of chitin under alkali conditions, consisting of (ß-(1-4)-2-acetamido-D-glucose and (ß-(1-4)-2-acetamide-D-glucose units [149]. Chitin and chitosan are animal-based polysaccharides. The structural properties of chitosan depend on the degree of deacetylation and average molecular weight. Edible films and coatings of chitosan are transparent, tough, and flexible, with good oxygen barrier properties ideal for fruits and vegetable packaging. Chitosan also shows excellent antimicrobial properties against fungi, algae, and bacteria due to its polycationic nature [150,151]. Compared to other biopolymers, chitosan has antibacterial potential. Chitosan has been described as bacteriostatic rather than bactericidal [152].

Edible films and coatings developed from chitosan plasticized with 30% glycerol displayed enhanced biological and mechanical protection of strawberries from fungi at-tacks without altering the aroma, flavor, texture, and appearance of strawberries [153]. Moreover, edible films and coatings prepared from modified diethylaminoethylchitosan, were studied for maintaining freshness in perishable fruits [154]. Strawberries and bananas coated with modified diethylaminoethylchitosan exhibited antimicrobial protection against fungi that affect various fruits and extended the shelf life of selected fruits compared to pure chitosan [154].

In similar research, edible films and coatings developed from biguanide modified chitosan and alginate blend showed improved thermal and mechanical properties and antibacterial activity against gram (+) and gram (−) bacteria and reduced WVTR with increasing biguanide modified chitosan [155]. Edible films of multilayered emulsion composites of chitosan and beeswax crosslinked with tripolyphosphate exhibited reduced WVTR and adequate mechanical properties [156]. Sweet cherries coated with different ratios of chitosan effectively prevented moisture losses at 20 °C and improved shelf life by reducing microbial growth [157].

4.4. Gums

Gums are polysaccharides with a significant molecular weight and are soluble in aqueous systems making hydrocolloids through solvent-polymer hydrogen bonding [158]. In aqueous systems, polymer/gum molecules form micelles, leading to hydrocolloid formation with high viscosity [159]. These hydrocolloids can easily be cast into films and coatings, making them suitable for fruit and vegetable packaging. Gum polymers can be homoglycans or heteroglycans with linear or branched structures. Edible films and coatings prepared from gums show excellent mechanical, transparency, tear resistance, and plasticity. Moreover, edible films and coating of plant gums, including gum arabic, guar gum, xantham gum, and basil seeds gums, possess good barrier properties for oxygen, carbon dioxide, and moisture [160].

Gum arabic is extracted from gummy extrudes of Acacia species [161]. Gum arabic has a heteroglycan structure with a backbone composed of (1,3)-linked β-d-galactopyranosyl residues, with side chains comprising of 2–5 (1,3)-linked β-d-galactopyranosyl units attached to the primary chain by (1,6) linkages. The primary and side chains of gum arabic also contain other carbohydrate units, including l-arabinose, l-rhamnose, and glucuronic

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acid [162]. The composition and related physiochemical properties of gum arabic may vary from source to source. In most cases, gum arabic has been used as a component in blends. Edible films prepared from the blends of gum arabic and chitosan infused with cinnamon essential oil showed improved WVTR performance and low mechanical properties [163]. Edible coatings of gum arabic/starch for fruits formulated using glycerol and sorbitol as plasticizers exhibited effectiveness against moisture loss by 30%, preserving firmness, facilitating the respiration and delaying the ripening process [164]. Emulsion-based edible films and coatings of gum acacia showed antioxidant and antimicrobial activity and also contributed to improved inter-molecular interactions [165]. Grapefruit encapsulated edible films and coatings from emulsion-based seed proteins, and gum acacia displayed enhanced water vapor barrier, surface hydrophobicity, mechanical properties, and thermal stability [166].

Galactomannans are linear chains made from (1→4)-β-D-mannopyranosyl units with single side chains in a 3:1 ratio of (1→6)-α-D-galactopyranosyl [167]. Galactomannans form highly viscous water-binding colloidal systems that interact efficiently with the polymers [168]. Guar gum and tara gum are widely studied galactomannans for preparing edible films and coatings. However, tara gums have poor mechanical and barrier performance and require improvements using plasticizers, including glycerol [169].

Xanthan gum is an extracellular polysaccharide to prepare highly viscous colloids at low concentrations [170]. Generally, xanthan gum is used as an additive in edible films and coatings. Guar gum is hydrophilic, linear polymer with β (1 → 4) linkages of D-mannose and single-linked α (1 → 6)-D-galactose [171]. Guar gum is widely employed in preparing blends with other biopolymers such as starch [160,172-174]. Active packaging blends of guar gum and sago starch infused with carvacrol and citral showed improved mechanical properties and inhibition of Bacillus cereus and E. coli [175]. Monosaccharides have also been investigated as plasticizers for developing edible films from guar gum and pea starch blends [176]. Edible films of guar gum and pea starch incorporated with natural antimicrobial agents have demonstrated changes in mechanical and antimicrobial properties following a concentration dependent trend [177]. For instance, four different natural antioxidants, namely, epigallocatechin gallate, blueberry ash fruit extract, macadamia peel extract, and banana peel extract, were studied with edible films and coatings of guar gum and starch blends [178].

Basil seed gum is an acidic anionic gum, having a glucomannan (43%) structure with a glucose-to-mannose ratio of 10:2, and (1→4) linked xylan (24.29%) and a minor fraction of glucan (2.31%) [179]. Basil seed gum has been studied for preparing active edible films and coatings because of their mechanical, antioxidant and antimicrobial properties. It has been reported that the edible films of basil seed gum infused with oregano essential oils exhibited enhanced physico-mechanical properties [180].

Moreover, adding glycerol plasticizer into edible films and coatings prepared from basil seed gum hydrocolloids improved physical, mechanical, microstructural, and thermal characteristics [181]. In addition, edible films from nanoemulsions of *Zataria multiflora* essential oil incorporated with basil seeds gum displayed high mechanical properties and strong antimicrobial activity against gram-positive and gram-negative bacteria [182]. Nanoemulsion also delayed the release of volatile compounds [182].

Gellan gum is a bacterial exopolysaccharide produced from the aerobic fermentation of carbohydrate substrates in bacteria [183]. Gellan gum has a linear, anionic polymer structure with a degree of polymerization (DP) of about 50,000. The chemical structure of gellan gum comprises of repeating unit of β-d-glucose, l-rhamnose, and d-glucuronic acid and two acyl groups, acetate and glycerate, attached to the glucose residue adjacent to

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glucuronic acid [184]. De-esterification of gellan gum makes stronger films and also alters gel texture [159]. Polymer blends of gellan gum have been widely investigated for developing edible films and coating [185]. For example, edible films prepared from the blends of gellan gum and aloe vera gel showed improved mechanical properties [186]. Edible films and coating of gellan gum and Aloe vera blends have been designed for active food packaging [187]. Moreover, edible films of gellan gum integrated into proteins showed excellent mechanical properties and barrier performance [188].

4.5. Starch

Starch is a semicrystalline homopolymer with up to 20-40% crystallinity and consists of two major constituents: amylose and amylopectin [189]. Amylose is a liner polysaccharide structure made up of α-1,4 bonds, while amylopectin is a branched molecule in which the branch points consist of α-1,6 glyosidic bonds [190]. Co crystallization favors crystallization in amylose into single helixes [191]. Hydrocolloid properties of starch in edible films and coatings depend on crystallinity, amylopectin ratio, moisture content, molecular mass, degree of branching, and polymeric chain length of the source of origin [189,192]. Edible films and coatings prepared from starch shows mechanical, oil, and oxygen barrier properties [193]. Moreover, modified starch enhances physio-mechanical characteristics of films and coatings and processing processes [194]. However, due its high hydrophilicity, antioxidant, antimicrobial and other food additive agents are incorporated into the starch matrix.

Starch composites and blends have extensively been investigated for the active packaging of fruits and vegetables. For example, edible coatings were developed using a combination of cassava starch and glycerol, carnauba wax, and stearic acid as plasticizers for fresh cut fruits and vegetables [195]. Bioactive edible starch films and coatings were formulated using phenolic compounds [196]. Starch/chitosan blends were studied for antimicrobial properties against *lactobacillus* spp [153]. This study demonstrated reduced aerobic mesophilic and psychrophilic cell counts while maintaining pH and weight loss in refrigerated storage, extending product life from 6 days [153]. The effect of the amylose-to amylopectin ratio of different starch species, wheat, corn, and potato, on the physico-mechanical properties of edible films and coatings has been studied [197]. Edible films prepared from starch with higher amylose content displayed better mechanical resistance and barrier properties with higher moisture sensitivity due to their hydrophilic nature [197]. Among many starch sources, wheat starch has the least surface wettability because of low surface hydrophilicity at elevated temperatures [197].

Sunflower oil added corn starch edible films and coatings demonstrated improvements in mechanical and water barrier properties due to the low crystallinity of starch and microstructural changes with the loading of sunflower oils as a plasticizer [198]. The combined effect of plasticizers and surfactants on starch-based edible films and coatings was investigated using glycerol, Tween 20, and Spam 80 as plasticizers with soy lecithin as surfactants [199]. This study confirmed the synergistic contribution of the plasticizer and the surfactants in achieving high mechanical and water barrier properties. Further concluded that high loading of plasticizers led to a decline in mechanical properties and higher WVTR and surfactants contributed to improving mechanical properties in the absence of glycerol [199]. Transfer properties of glycerol loaded edible starch films were reported, confirming higher diffusivity/ transfer properties for 55% glycerol-loaded edible films over 33% glycerol loaded films [200]. Moreover, glycerol impacts the water diffusivity, oxygen permeability, and water vapor permeability performances in edible films and coatings. The same trend was observed with the edible film of native wheat prepared from varying concentrations of glycerol (0, 20, 30, 40, and 50,.wt%) [201]. The lowest WVTR was reported at 30% glycerol loading, but the degree of crystallinity was reduced. With

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increasing glycerol loading in the starch matrix, stress at break and Young's modulus decreased, and elongation increased [201]. Edible films of highly carboxymethylated starch (HCMS) plasticized using sorbitol, xylitol, mannitol, and glycerol showed reduced WVTR and decreased solubility with increasing plasticizer concentration [202-204]. Edible films and coatings have been developed from non-conventional arrowroot starch using the casting method prepared for plum packaging [205]. Results displayed an increase in WVP from 2.20 to 3.68 g mm/m2 day kPa, moisture content from 3.22% to 7.95% and a decrease in solubility in water from 22.45% to 13.89%, delivering extended post-harvesting preservation upto 35 days [205]. Furthermore, these edible films and coatings exhibited good film-forming ability with homogeneous, transparent and manageable appearances. The 2% coating indicated well adhesion, successfully minimizing moisture loss and controlling respiration rate [205]. A study on rheological properties of corn starch methylcellulose and glycerol edible films and coating exhibited high total viscoelastic recovery at a high shear rate, just like entangled polymer dispersions, due to the interactions of topological entanglements and dispersion stability [206].

5. Protein-based edible films and coatings

Edible films and coatings from protein sources may come from animal and plant origin [207]. Protein-based edible films and coatings are formulated with three main components; protein, plasticizer, and solvent. Most of the proteins in edible food pack-aging are insoluble-fibrous or globular-water soluble proteins [29]. Proteins make a cohesive film matrix with string interactions stabilized from uniformly distributed attractive forces of hydrogen, Van der Waals forces, covalent and disulfide bonding [208]. Protein-based edible films and coatings are biodegradable and compostable. The degradation process of protein-based edible films generates nitrogen in the soil, enhances soil nitrogen content, and functions as a fertilizer, delivering extra benefits than non-protein-based edible films and coatings [209]. Protein-based edible films act as bioactive peptides in digestion and deliver health benefits by antihypertensive and radical scavenging [210,211]. Characteristics of edible films and coatings from proteins are interlinked with composition, components, and processing technique [34,212]. The performances of protein-based edible films and coatings are associated with structural factors, including crystallinity, amino acid composition, additives, plasticizer content, hydrophobicity/hydrophilicity, nature of the surface charge, molecular size distribution, and three-dimensional shape [213,214].

5.1. Animal proteins – casein

Casein is the major dairy protein group in bovine or goat milk. There are four main subunits in casein: alpha s1-casein, alpha s2-casein, beta-casein, and kappa-casein that make up 38%, 10%, 36%, and 13% of casein composition [215]. The primary, secondary and tertiary structures of casein are less ordered and more flexible than typical globular proteins [216,217]. The structure of casein contains polar and hydrophobic domains with a degree of flexibility, giving limited proteolysis and enhanced functionality [218]. Each of the four protein fractions possesses unique properties that influence the film-forming ability of casein [219]. Edible films and coatings of casein in fruits and vegetables have been widely investigated due to the processing convenience of emulsion preparation in amphipathic dispersion systems [220]. The complex intermolecular binding of milk protein in edible films and coatings is well known for providing a good barrier to gas per meation and nutritional value [221]. Cohesive edible films and coatings can be designed from total milk proteins, wherein the continuous 3D film-coating network is governed by protein-protein interaction [220,222].

Edible films and coatings from casein and beeswax blends were developed from emulsions to reduce white blush and increase water vapor resistance in processed carrots by

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integrating stearic acid or acetylated monoglyceride [223]. Edible films prepared from casein and natamycin controlled efficiently mold growth and microbial activity [224]. Generally, the WVTR of caseinate edible films is affected by several factors, such as pH, calcium crosslinking, and lipid content [225]. A study assessing the permeability of casein and wax heat-sealed edible film reported that the WVTR reduced as wax content increased without a difference in oxygen permeability with loading waxes [226]. Extruded casein and wax blends for active edible films incorporated with potassium sorbate as the carrier exhibited good bacteriostatic properties, inhibiting E. coli growth for up to 20 days [227].

5.2. Animal proteins – whey protein

Whey protein is a byproduct of serum coagulation from casein extraction in cheese production [228]. Whey protein in edible films and coatings can be found as protein iso-late or whey protein concentrate, which forms an interactive 3D polymeric network upon drying [229]. Compared to polysaccharides and other proteins, whey protein can be used as an emulsifying, thickening agent, gelling agent, foaming, and water-binding agent resulting in films and coatings of whey protein inherit good mechanical, odorless, flexible, and transparent characteristics with moderate water vapor permeability and excellent oxygen gas barrier [230]. Film formation and gel characteristics of whey protein are governed by its secondary, tertiary, or quaternary globular structures and the presence of various combinations of cross-sulfur bonds, making them heat labile, dephosphorylated, and less calcium sensitive [231]. Whey protein coatings are proposed for microbial growth reduction, extending shelf life, and minimizing moisture loss and spoilage of fruits and vegetables without changing texture [232]. Furthermore, edible films and coatings from whey proteins can be used as an efficient carrier for antimicrobials, antioxidants, or other nutraceuticals [233].

The influence of protein to glycerol ratio in mechanical, optical, and moisture sorption properties of films and coatings prepared from whey protein isolates were analyzed, further justifying that increasing protein concentration leads to transparent films, higher moisture adsorption and enhanced tensile properties [234]. Edible films and coatings of whey proteins modified with almond and walnut oils have been reported for de-creased WVTR due to enhanced hydrophobicity, and the incorporation of almond oils provided plasticizing effect to the film [235]. Blends of whey protein and pullan plasticized using glycerol have been studied to determine optimum blending ratios for potential edible films and coatings applications [236]. The physical properties of edible films from whey proteins loaded with rapeseed oils have been studied for emulsion properties and reported significant improvements in tensile properties as the mechanical resistance decreased with lipids while the opacity caused to increase [237]. The blend of whey protein and pectin crosslinked with transglutaminase minimized microbial growth control and moisture loss with minimal impact on the texture properties, chewiness, and hardness [145].

5.3. Animal proteins – collagen

Proteins of animal origin are also considered for edible films and coatings due to excellent mechanical and oxygen and carbon dioxide barrier properties at low humidity conditions [238]. Collagen is extracted from the connective tissues of animals and is a major structural protein of the extracellular matrix [239]. The triple helical structure of collagen is known for determining its physio-chemical and colloidal properties [240]. A colloidal dispersion of collagen is desirable for making EF & EC via casting or extrusion and may require salt coagulation or covalent crosslinking for extended packaging requirements [241]. A recent study used acetic acid extraction technique for collagen, and EF & EC properties have

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proven acceptable after evaluating the film's mechanical, tensile strength, Young's modulus, elongation and WVR [242]. Chitosan/collagen edible films emulsified with cinnamon perilla essential oil displayed improved film properties and shelf-life, resulting from pickering nanoemulsification [243]. Coating food with chitosan films lowers the oxygen partial pressure in the package [244].

In another approach, hydrolyzed collagen and cocoa butter edible films plasticized from sucrose were proven for satisfactory optical properties, improvements in mechanical properties, and significant changes in WVTR [245]. Blends of collagen and alginates were used to prepare uniform films with no phase separation and showed good tensile, elongation at break, and WVTR properties [246]. Edible films and coatings developed from collagen and polysaccharide blends exhibited antimicrobial efficacy for staphylococcus aureus and Escherichia coli when 12 mg/mL cell-free supernatants were loaded into the films [247]. In similar research, hydrolyzed collagen and sodium alginate edible films and coatings EF & EC were developed as a low-cost alternative from byproducts with increasing thickness [248]. WVTR reduced up to 50% with slight inhomogeneity across the film [248]. Transglutaminase-crosslinked edible films and coatings from collagen were developed to improve the stability of the structure and packing of collagen fibers, improving mechanical, barrier, thermal stability, and morphology compared to the un-crosslinked [249]. Chitosan-lemon-essential oil nanoparticles incorporated collagen composites showed lower oxygen permeability, higher tensile strength (TS), and higher elongation at break [250270]. Furthermore, collagen-chitosan-lemon essential oil compo-sites preferentially inhibited lipid oxidation, microbial growth, and food deterioration [250].

5.4. Animal proteins – Gelatin

Gelatin is a secondary product derived from the hydrolysis of primary collagen proteins under mild heat or acidic or alkaline conditions into a partially denatured un-structured domain triple helical structural form [251]. Type A gelatins are derived from the acid processing of collagens, and type B gelatins are a product of alkaline or lime processing [252]. Blends of gelatin and starch have been widely tested for edible films and coatings [253]. For example, edible films and coatings prepared from gelatin/ starch blends plasticized using sorbitol and glycerol displayed improved transparency, mechanical properties, and satisfactory WVTR for crimson red grapes [254]. Furthermore, edible films and coating of crimson red grape act as an effective post-harvest treatment, maintaining fruit quality and preventing moisture loss without interfering with consumer taste [254]. Manioc starch and gelatin blends with different plasticizer combinations exhibited enhanced transparency, tensile performance, and WVTR [255]. However, as the sorbitol concentration increased, the stability of the edible film decreased due to broader glass transition and phase separation [256].

Antioxidant and antimicrobial edible films and coatings were also prepared from gelatin and chitosan blends by integrating plant ethanolic extracts. It was reported that increasing chitosan had increased the elasticity of the EF. According to the tolerance equivalent antioxidant test, these infused blends showed good antioxidant performance and excellent microbial growth inhibition against *Escherichia coli* and *Staphylococcus aureus* [257]. A similar study assessed the mechanical performance of gelatin and chitosan blends was indicated that chitosan loading significantly increased tensile strength and elastic modulus but reduced elongation at break, making films brittle ($p < 0.05$) [258]. Also, loading chitosan drastically improved the water barrier and water stability of the gelatin films ($p < 0.05$), and light barrier properties at 600nm against UV light [258]. In another approach, olive oil was incorporated into the gelatin matrix and delivered enhanced barrier properties and tensile performance with high stability against to UV light in emulsified films, making them potential for fruits and vegetable packaging [259]. Edible films from gelatin

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and casein blends crosslinked using transglutaminase enzymatic crosslinker exhibited elevated elongation ($P < 0.05$) compared to gelatin or casein alone, and crosslinks have improved water barrier as casein and gelatin (75:25) reported the lowest WVTR (5.06 ± 0.31 g mm/m2 d kPa) [260].

5.5. Plant proteins- Soy and wheat proteins

Soy and wheat are the two main plant-based proteins used for edible films and coating in food [261]. Soy protein isolates are hydrocolloids, and films and coatings can be processed following film-forming and casting methods. Films prepared from soy proteins are an excellent barrier against oxygen and moisture [262]. Moreover, the proper-ties, including appearance, tensile strength, and low WVTR of soy hydrocolloid films, can be enhanced with alkaline treatment [263]. Wheat glutens are also used for edible films and coatings. However, the application of wheat glutens in edible films and coatings is limited with increasing gluten-free food consumption. In general, plant proteins in edible films and coating are less than animal proteins due to food security concerns, limited availability and processing difficulties.

6. Lipid-based edible films and coatings

Edible films and coatings prepared from lipids are critical for minimizing moisture loss and as a hydrophobic barrier [104]. For Fruits and vegetables, moisture regulation preserves and extends shelf-life through a method similar to desiccation. In addition, lipids act as carriers for active molecules, keep the texture and flavors, and utilize as varnishes. Table 1 summarizes the different types of lipids used in edible films and coatings.

Table 1. Lipids in edible films and edible coatings

Lipids	Sources	Reference
Fats, oils, shortening and margarine	**a)** Various native fats and oil from animals, vegetables, and seeds Such as butter from dairy, lard, sunflower, mustered, olive, almond, peanut, coconut, palm, cocoa, etc **b)** Fractionated, concentrated, or reconstituted oils and fats ,and mono, di and tri-glycerides **c)** Hydrogenated or trans-esterified oils: Margarine and shortening	[264]
Waxes	**a)** Waxes from natural animals, insects and vegetables: Beeswax, carnauba wax, candelilla wax, genuine rice bran wax, and laurel wax **b)** Waxes from synthetic sources; paraffin, mineral, microcrystalline, oxidized-non-oxidized polyethylene wax	[265]
Natural resins	Resins from natural sources: Asafoetida, Benjoin, Chicle, Guarana, Myrrhe, oblibanum, Opponax, Sandaraque, etc	[266]
Essential oils and liquorices	Various essential oils from an extract from flowers, vegetables, animals and fruits; Citrus, rose, ginger, mint, etc	[267]

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Lipids are incorporated with hydrocolloids such as polysaccharides, proteins, cellulose, starches, and their derivatives in composites. Lipid blends and hydrocolloids can be mainly prepared from solvent blending and mixing. Other methods used for making composites are casting and film-forming methods with multilayer and bilayer films. In lipid/hydrocolloid composites, hydrocolloid fraction contributes to moisture absorbance, while lipids are responsible for moisture regulation. Additives are doped into lipids to introduce various functionalities to edible films and coatings. Functionalization of lipids helps improve the film-forming ability of colloidal emulsions and suspension phases and promotes adherence of films and coatings on the food surfaces. In addition, lipids act as emulsifiers, texturing agents, antioxidants, antimicrobial agents, enzymes, process aid, and gelling promoters. Plant based oils such as soybean, sunflower, high oleic linseed, rapeseed, and palm have been studied for edible films and coatings due to their moisture barrier properties, readily availability, less flavor impact and low cost [268].

6.1. Essential oil

Essential oils are volatile components obtained from plants and show distinct anti-microbial properties [269]. Furthermore, essential oils in edible films and coatings pre-vent chemical reactions, lipid oxidation, endogenous enzyme activity and microbial growth [136]. Natural essential oils also considered GRAS, help improve the mechanical, antimicrobial, antioxidant, and sensory properties of the in edible films and coatings in fruits and vegetables. For enhancing the shelf-life of fresh fruits and fruit cuts, essential oils with monoterpenes and sesquiterpenes are incorporated into films and coatings [270]. It was reported that plant essential oils are the main additive used as antimicrobial and antioxidant agents in edible films and coatings. Active components found in essential oils from various spices and herbs, specifically thymol, cinnamaldehyde, carvacrol, 1,8-cineole, and eugenol, give a greater extent of antimicrobial and antioxidant performance [271]. Essential oils make stable interactions between hydroxyl groups in biomacromolecules through ethers, aldehydes, and ketones pendant groups in their structure. Essential oils give hydrophobicity to edible films and coatings with improving water and moisture barrier properties [272,273]. Essential oils may improve tensile strength and elongation at the break due to plasticizing effect and capability to make crosslinks [274-276].

6.2. Waxes and resins

Waxes, referred to as esterified long-chain alcohols and fatty acids, belong to a diverse class of organic compounds that may exist as solid or semi-solid at ambient temperatures [277]. Based on the source of origin, waxes can be divided into three classes [267*];*

a) Animal waxes: bees wax, spermaceti wax, shellac wax, lanolin wax, Chinese insect wax.

b) Vegetable waxes: Carnauba wax, candelilla wax, bayberry wax, rosin wood wax, sugarcane wax, palm wax, esparto wax, cotton seed wax, oricury wax, rice bran wax, Japan wax, waxol.

c) Mineral and synthetic waxes: Montan wax, paraffin wax, ozocerite, synthetic wax, and microcrystalline wax.

According to the FDA, paraffin wax on raw fruits and vegetables is permitted [278]. Paraffin, carnauba, beeswax, candelilla and polyethylene waxes are widely used for food related applications alone or in combination [279]. Edible wax coatings are commonly applied for the post-harvest preservation of fruits and vegetables. Waxes provide good moisture and gas properties and preserve the surface appearance of fruits and vegetables. Edible waxes show high durability, stretching, and super-hydrophobicity [280]. However, it has been reported that waxes such as paraffin, carnauba, candelilla, and bee act as humidity barriers for edible films and coatings [281]. Wax-based micro-emulsions are widely used for fruits and vegetables coatings [282]. For instance, micro-emulsion coatings prepared from candelilla, beeswax, carnauba wax, polyethylene, and petroleum show distinct moisture barrier properties [283]. However, polyethylene and carnauba waxes were the glossiest and most brittle coatings [283].

Compared to ammonia-based emulsion wax coatings, edible coatings from morpholine wax performed a low barrier to oxygen and water vapor. Candelilla wax edible coating combined with biocontrol bacteria improved the quality and the shelf-life of strawberries and acted against *Rhizopus stolonifer* bacteria [284]. Carnauba wax incorporated edible coatings enhanced the shelf-life of mangos by decelerating the ripening process during storage [285]. Candelilla wax incorporated with bioactive compounds preserved the quality of tomatoes for a prolonged period [286]. Blends of waxes and fermented extract of tarbush improved the quality of apples by minimizing weight loss and ensuring firmness without altering the appearance and taste of apples [287].

Resins are another component of edible films and coatings. Terpene resin is obtained from the polymerization of terpene hydrocarbons derived from wood and is approved as a direct food additive [269]. Natural resins are added to the formulations of edible films and coatings to provide hydrophobicity, cohesiveness, and flexibility [266].

7. Health Effects

Edible films and coatings are primary packaging and according to Regulation No. 1935/2004 food contact materials should not transfer hazardous compounds to health into food [288]. Edible films and coatings were developed as an alternative for chemicals and synthetic materials which are harmful to human health [289]. Edible coatings can help to preserve antioxidants, phenolics and pigments of the food for a long time [289]. Probiotics can be added through microencapsulation technique [290]. Furthermore, edible coatings which are supplemented with bioactive compounds such as probiotics can give advantages on human health and if the food coating has a certain level of probiotics, the food can be considered as a functional food [289], [291]. Waxes and paraffins are considered as safe coatings for fruits and vegetables [291]. The cellulose monomers and starch- based biopolymer monomers are estimated as health safe and creates no health problems [292] However, nanocellulose which are used in edible coatings are non- toxic at a concentration of 0 – 50 μg/mL in human endothelial cells, and above the stated physiological changes [293]. Certain studies have shown that collagen showed discoloration, skin necrosis, granuloma formation, blindness, and foreign body reactions as clinical manifestations [293]. However, further detailed studies are required to assess the full effect of the edible films and coatings and human health to assure food safety.

8. Conclusion and Current Trends

Edible films and coatings are a pivotal part of food preservation amid current global hunger and supply chain crisis. Moreover, edible films and coatings prepared from renewable biomacromolecules have widely been investigated due to their relative abundance, cost-effectiveness, biocompatibility, nontoxicity and biodegradability. The colloidal chemistry of biomacromolecules, including polysaccharides, proteins, and lipids, governs their processing and film-forming ability, casting, lamination, and other properties. Hydrocolloids of biopolymers show excellent air and gas barrier properties, making them excellent candidates for developing edible films and coatings.

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Sustainable production approaches, materials from renewable sources, green syn-thesis, and eco and biotoxicity are crucial aspects in the modern packaging of edible films and coatings. Therefore, biomacromolecules, including polysaccharides, proteins and lipids, are extensively researched to develop future food packaging advancements. However, natural molecules have chemical and structural limitations in designing efficient films and coatings for fruits and vegetables. Hence, biopolymer blends, compo-sites, and multilayer approaches have recently emerged as novel techniques for developing edible films and coatings for fruits and vegetables. Furthermore, crosslinking, functionalization, and surface modifications using green routes have gained significant attention in enhancing the physical, chemical and biological properties of biopolymer-based edible films and coating. For instance, active edible films and coatings have been developed by doping active molecules such as antimicrobial, antifungal, enzymes, vita-mins, essential oils, and plasticizers. Moreover, active packaging, intelligent packing and nanotechnology-based approaches are the current trends in edible food packing applications.

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