POLYMER BASED EDIBLE FILMS AND COATINGS

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Keyword: Natural polymers, edible films and coatings.

Abstract

Edible films have grown in popularity in recent years due to a number of advantages, including the ability to use edible packaging material instead of synthetic films. Edible films are environmentally friendly, but they do not pollute the atmosphere. As compared to non-environmentally friendly packaging materials, edible films have barrier properties, allowing packaging materials to be recycled more easily. Scientists have obtained and characterised new agricultural processing products, which are typically regarded as waste. The aim of this study is to present edible films and coatings in a concise manner by describing the related materials, their properties, and their applications.

1)Introduction

Edible polymers, which are primarily composed of polysaccharides, proteins, and lipids, are natural materials that can be consumed by both animals and humans without causing harm to their health (Kauhi et al., 2020). These polymers, which have been labelled as "generally recognized as safe" (GRAS) by the Food and Drug Administration, have become an alternative candidate that has sparked a lot of debate due to their beneficial properties. These polymers are increasingly being used in the functional food industry (food packaging and nutrition protection) as well as in biomedical fields (drug delivery, tissue engineering, and wound dressing) (Ali and Ahmed., 2018). Biodegradability, biocompatibility, and recyclability are all essential properties of edible polymers that make them a better option than non-biodegradable petroleum polymers. They can also help the environment by reducing pollution, improving material recyclability, producing eco-friendly goods, and ensuring sustainability (kabir et al., 2018). The emergence of new food product categories, such as healthy, easy, and environmentally sustainable high quality products, was one of the key reasons for finding healthy to eat polymers in the food industry. Furthermore, the modern consumer favours “green” consumerism, preferring less chemical additives, as well as assured protection, high nutritional value, overall quality, and extended postharvest shelf life. Various materials are used to enrobe (i.e., cover or coat) various foods in order to increase the shelf life of the product that can be consumed with the coated food. Generally, edible coatings and films are made from sustainable natural and biodegradable polymeric materials like polysaccharides, proteins, and lipids, or a mixture of these components (composite and conglomerate coating/film) (Dhall., 2013). Since the edible polymers are made entirely of renewable and safe-to-eat ingredients, they can degrade more quickly than other polymeric materials. Edible polymers have been widely used in biomedical research because of their flexibility, which allows them to meet a wide range of functional and design criteria for various tissue types. Furthermore, they can be subjected to a wide variety of physico-chemical modifications to meet particular tissue regeneration needs, making them ideal for biomedical applications. Material chemistry, shape structure, molecular weight, hydrophobicity/hydrophilicity, lubricating property, surface energy, degradation rate, water absorption, erosion mechanism, and solubility are all considerable factors in the selection of edible polymers for biomedical applications (Kauhi et al., 2020). On the other hand, edible polymers have received a lot of interest in cosmetics, non-renewable/renewable oil, and wastewater treatment, in addition to the food and biomedical industries. Edible polymers are gaining popularity in the cosmetics industry because of their ability to improve dispersion stability, texture, biodegradability, biocompatibility with skin and mucus, and sensory performance. Edible polymers have been used in energy applications as a source of renewable energy, electrolytes for energy storage systems, and generators. Natural polymers are also being researched for their ability to remove contaminants from water such as suspended solids, dyes, pesticides, toxicants, and heavy metals.

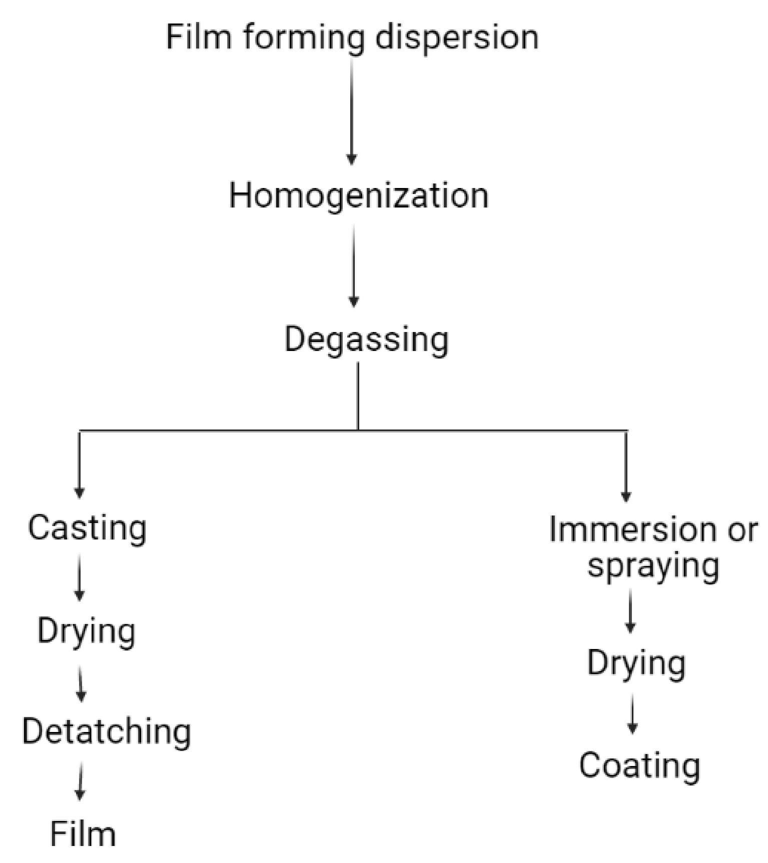


Figure: The processing of films and coatings is depicted schematically

2)CLASSIFICATION OF EDIBLE FILMS AND COATINGS

2.1. Hydrocolloids

Hydrocolloids are hydrophilic polymers with a lot of hydroxyl (-OH) groups in them. They are now commonly used as the base material for film-forming solutions to conduct and monitor food texture, taste, colour, and shelf-life (Nussinovitch and Nussinovitch., 2013). They are entirely or partially soluble in water and are primarily used to increase viscosity in the continuous process of the film forming solution with or without the application of external heat, i.e. as a gelling agent or thickener. They also have the ability to stabilise emulsions, preventing emulsion separation (if the coating or film contains oil-in-water or water-in-oil components) due to an increase in aqueous phase viscosity. The oil droplets' kinetic motion is decreased, which results in a slower rate of flocculation and coalescence in the film.

A.Polysaccharides

Polysaccharides are complex carbohydrate macromolecules composed of two or more monosaccharides connected by glycosidic linkages formed during condensation reactions (Neill et al., 2004). The form and linkage of monosaccharide units determine the physical properties of polysaccharides. They're insoluble in alcohol and nonpolar solvents, often white in colour, tasteless, and crystalline. Polysaccharides are also excellent energy savers, fuel storage molecules, and metabolic intermediaries. They are commercially assessable as stabilisers, thickeners, gelling and encapsulating agents, and crystallisation inhibitors in the food and non-food industries (Pjilips and Williams., 2009). Because of their versatile functionalities and biocompatibility, polysaccharides stand out among edible polymers. Chitosan, starch derivatives, cellulose, pectin, and exudate gums are used in polysaccharide-based edible films.

Examples of polysaccharides

(a) Starch and its derivatives

Starch, relatively inexpensive, abundant, odourless, tasteless, biologically absorbable, nontoxic, semipermeable to carbon dioxide, is colourless, and is resistant to oxygen (Shah et al., 2015). It has some physical properties similar to plastics, due to which, starch is being extensively researched in food industrial applications. Most starches are composed of anhydroglucose residues containing linear amylase and branched amylopectin derived from botanical sources such as corn, wheat, cassava, rice, potato, and yam, among others. Starch is made up of a linear glucose chain called amylose and a ramified glucose chain called amylopectin (Liu., 2005).

(b) Cellulose

Cellulose is the most abundant renewable plant resource with excellent film forming properties. It is made up of D-glucose units that are linked together by glycosidic bonds of β 1-4 linkage. The hydroxyl methyl groups of anhydroglucose residues found below and above the polymer backbone plane provide a solid packaging and highly crystalline structure for the polymer chain in its natural state of cellulose (Hassan et al., 2018). The water solubility of cellulose can be increased by treating it with alkali and then reacting it with chloroacetic acid, methyl chloride, and propylene oxide to produce carboxy methyl cellulose (CMC), methyl cellulose (MC), and hydroxypropyl cellulose (HPMC). MC, HPMC, HPC, and CMC films have good film forming characteristics, no odours or tastes, and are clear and solid. MC can also be used as a lipid migration barrier in bakery products (Badi et al., 2010). A number of other composite films made of MC or HP MC have been investigated. Water vapour permeability is equivalent to low density polythene in these films (LDPE).

(c) Chitoson and chitin

Chitin is a naturally occurring biopolymer that can be used in crustacean exoskeletons, fungal cell walls, and other biological materials. It's mostly poly (β- (1–4)-2-acetamido-D-glucose), which is structurally similar to cellulose but has an acetamide group in place of a secondary hydroxyl on the second carbon atom of the hexose repeat unit (Dhillon et al., 2013). Chitosan is a copolymer made up of β-(1–4)-2-acetamido-D-glucose and β-(1–4)-2- Amino-D-glucose units, with the latter accounting for more than 60% of the total. Chitosan's antimicrobial properties, in combination with its cationicity and film-forming properties, are defined in terms of degree of deacetylation and average molecular weight. Chitosan films have good mechanical properties and selective permeability to gases (CO2 and O2) (Remunan and Bodmeier., 1997). However, the fact that chitosan films are highly permeable to water vapour restricts their use, which is a significant disadvantage since an effective barrier to water vapour is needed. Strawberries, cucumbers, and bell peppers are among the fruits and vegetables used as antimicrobial coatings for chitosan films and as a gas barrier for apples, pears, peaches, and plums.

(d) Pectin

Pectin is a polysaccharide present in fruits and vegetables such as apple pomace and citrus peel (Dhanapal et al., 2012). Pectin is an anionic polysaccharide with a (14)-linked -d-galacturonic acid unit as its structural backbone. It's used in yoghurts, milk, ice cream, and jams as a stabiliser, thickening agent, and gelling agent. Food packaging could benefit from the use of pectin edible films and derivatives. The mechanical properties of pectin-based edible films reveal excellent mechanical properties, an excellent barrier to oil and scent, oxygen, and a high initial modulus, but they have poor moisture resistance, low elongations, and are brittle; the addition of plasticizer makes them more flexible. Crosslinked pectin films with polyvalent cations like calcium have good mechanical properties (Chaturvedi et al. 2019). Pectin films/gels are good at protecting low-moisture foods.

(e) Gums

The botanical origin, form, charge, and chemical structure of gums can all be used to classify them. Gums are mostly obtained from plants. Natural gums are widely used in the food and pharmaceutical industries. Tahir et al. (2019) concentrated on the gums that help in preservation of fruits and vegetables such as, Gum arabic, Xanthan gum, Guar gum, Gellan gum, Almond gum, Psyllium seed gum, Tragacanth gum, Locust bean gum, Flaxseed gum, Cashew gum, Karaya gum, Peach gum, Tara gum, Quince gum, Carrageenan gum. When a thin layer of edible coating solution (such as gum or gum infused with bioactive additives) is applied to fruits and vegetables, it acts as a barrier between the treated samples and their surroundings (Senturk et al., 2018). This barrier regulates gas and water vapour exchanges and slows the ripening process in fruits and vegetables, delaying many metabolic changes including ascorbic acid, polyphenols, anthocyanins, antioxidant activity, firmness, colour, and sensorial properties.

Table: polysaccharides according to their origin

0rigin of polysaccharide	Examples
Animal origin polysaccharides	Chitin and chitosan
Plant origin polysaccharides	Cellulose
	Starch
	Pectin
	Arabic gum
Marine origin polysaccharides	Alginate
	Agar
	Carrageenan
Microbial polysaccharides	Pullulan
	Gellan
	Xanthan gum

B.Protein

Protein is a heteropolymer made up of over a hundred monomers connected together by peptide bonds. A central carbon is bound to (a) a carboxyl group, (b) a hydrogen, (c) an amino group, and (d) R-group in each of these amino acids (Hanani et al. 2014). Proteins can be found in nature as fibrous or globular proteins; globular proteins are rolled over one another, while fibrous proteins are bound to one another in parallel. Protein films have stronger mechanical properties than polysaccharides because of their special structure. However, as opposed to synthetic polymers, protein films have a lower mechanical strength and have a higher water vapour permeability. Milk protein, Gelatin, collagen, corn zein, wheat gluten, soy protein is used as protein based film coatings.

(a) Milk protein

Whey protein and casein protein are two types of milk proteins (cow's milk contains around 33 g of protein per litre). Casein protein, which makes up 80% of milk protein, is made up of, and -casein components. Commercially available caseinates can be obtained by skimming milk at 20°C and pH 4.6 and are precipitated by alkali. For a wide range of pH, temperatures, and salt concentrations, casein-based films remain stable.

(b) Corn zein

Corn's key protein is zein, which is a polyamine. Since it is shiny, water-insoluble, bacteria-resistant, antioxidant, and forms adhesive film, it is a virtual hydrophobic and thermoplastic material.

(c) Collagen

Collagen edible films were previously used in meat products to retain moisture and give the product a uniform appearance.

(d) Gellatin

Collagen hydrolysis is the process used to make gelatin. Until processing meat products, the meat industry uses collagen films. When heated, collagen film acts as an edible skin that aids in the cooking of meat products (Hashim et al., 2015). Collagen, which is found primarily in animal skins, muscle, bones, and connective tissues, is pre-treated with an acid/alkaline solution before being heated to 40 degrees Celsius to produce gelatin. Pure and dry gelatin has a slight yellow colour and is translucent, tasteless, brittle, odourless, and glass-like solid. Gelatin is dissolved in hot water to make an edible film, and the dispersed solution is cast on a plate and dried in an oven. Gelatin-based edible films have improved film thickness and mechanical properties as protein content has increased, however water vapour has decreased.

(e) Soy protein

Soybeans are used to make soy protein. Soy protein is sold in a variety of ways, including soy flour (56% protein and 34% carbohydrate), soy isolates (90 percent protein, 2% carbohydrate), and soy concentrate (65 percent protein, 18 percent carbohydrate). Soy protein film is normally made from soy protein isolate. This is due to the nonprotein fraction in other soy protein types having a detrimental impact on film formability. Yuba films or baked film methods are commonly used to make soy protein edible films. Soy milk is boiled in a thin pot, and films are formed due to surface dehydration, which is then dried in the air. Spread soy protein isolates are baked on baking pans for 1 hour at 100 °C in the baked film form. In comparison to films produced by other plant proteins, soy protein films are smooth, versatile, and transparent (Denavi et al. 2009). In terms of gas barrier properties, soy protein films outperform lipid and polysaccharide films. As compared to low-density polyethylene, starch, and pectin, their oxygen permeability is at least 260 times lower when not exposed to moisture.

(f) Wheat gluten protein

Wheat gluten is a protein found in wheat flour that is made up of a variety of polypeptide molecules. Film forming properties can be found in wheat gluten due to its cohesiveness and elasticity. In 70 percent ethanol, gliadin is soluble, however, glutenin is not. Edible films can be made by drying an aqua solution of ethanol containing gluten. Hydrophobic bonds are essential in the formation of wheat gluten film structure, the breakdown of native disulfide bonds during heating, and the formation of new disulfide bonds during film drying, in addition to hydrogen. The addition of sorbitol reduces the barrier properties of film strength, elasticity, and water vapour barrier when gluten films are added to glycerin (plasticizer), but increases the stability of the film (Gennadios et al. 1993).

2.2. Lipids

Lipids are organic compounds found in nature, such as in animals, insects, and plants as well as herbs. The lipid functional groups' diversity is made up of phospholipids, phosphatides, mono-, di-, and tri-glycerides, terpenes, cerebrosides, fatty acids and fatty alcohol. The food industry has recently focused on the use of lipids in coatings and edible films for food preservation (Hassan et al., 2018). Lipids in coatings and edible films have a variety of benefits, including gloss, moisture loss reduction, cost reduction, and packaging complexity reduction.

(a) Waxes and paraffin

Waxes are made up of alcohol and/or esters of a long chain acid, they have a higher molecular weight. Waxes have a vegetal and animal basis and serve as protective coverings for tissues. These can be used in coatings or edible films to reduce moisture permeability and increase hydrophobicity. Aloe Vera gel and Candelilla wax was used by Akoh and Min, 2008 as edible coating on fresh fruits and declared that Candelilla coating, is a fruit preservation option that helped with weight loss, firmness, lightness, and appearance qualities as compared to fruits that were not coated. Beeswax, Brea gum, and glycerol were mixed however, Spotti et al. (2016) declared that Because of the decreased water vapour permeability, microstructure, and mechanical properties of beeswax, it was not useful in this film. Paraffin wax is made from fractions of crude petroleum distillate. Paraffin wax may be used on raw fruits, vegetables, and cheese in general. When waxes are applied to surfaces, they give them a lustrous sheen and also act as a moisture and gas barrier. When applied in thin layers, waxes are edible, but when applied in thick layers, they must be removed before food (certain cheese) is consumed.

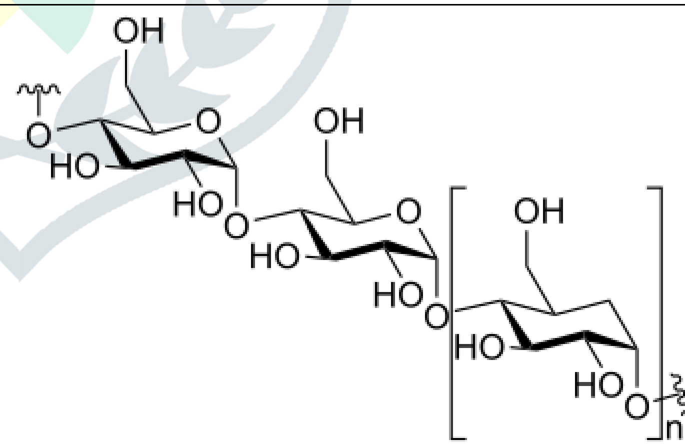
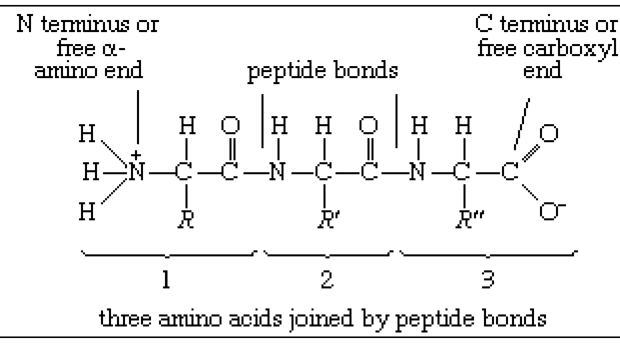
(b) Resins

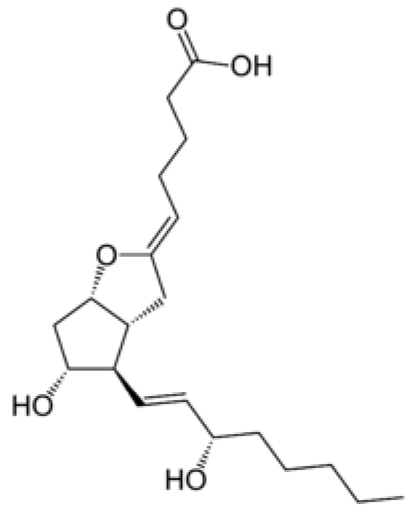
Resins are substances produced by plant cells in response to injury or infection in trees and shrubs, as well as by certain insects, such as *Laccifer lacca*, which produces shellac resin. Shellac is not authorised by GRAS, however it is used as a direct food additive in food coatings. It's widely used in the pharmaceutical industry, and it's thought to have only a few uses in the food industry.

2.3. Composites

To resolve the disadvantages of single-component edible films and coatings, composite coating is a term that combines the unique features of various individual coating materials in a desired combination. It is made either by layering different film forming materials, or in the form of emulsion, suspension, or dispersion in immiscible layers at a time, or by dissolving different materials in the same solvent and then applying the film or coating. Hagenmaier and Shaw, 1990 use lipid and hydroxypropyl methyl cellulose, Bertana et al., 2005 used combination of gelatin and fatty acid to ensure the coated products retain their quality and have a longer shelf life.

Table: Few examples and common structure of edible polymers

Edible polymers	**Examples**	**Common structure**
Polysaccharides	Starch, pectin, agar, alginate, cellulose derivatives, carrageenan, xanthan gum, guar gum, gum arabic, pullulan etc.	
Proteins	**Animal based**: gelatin, collagen, albumin, milk protein (casein and whey protein), etc. **Plant based**: zein, wheat gluten, soy, peanut, pea, nut proteins, etc.	

Lipids	waxes (beeswax, candelilla, carnauba waxes), phospholipids, fatty acids, triglycerides, glycolipids, etc	

3.Applications

a. Fruits, nuts, grains & vegetables

Respiration, transpiration during maturation and storage, or microbial development, especially moulds and rots, are the main causes of fruit and vegetable spoilage (Krochta et al., 1994). Waxes such as (Paraffin, Carnauba, Mineral Oils, Candellila, Beeswax, Polyethylene, Shellac) are commonly used for fruit coatings such as apple, strawberry, orange, peach, mango, date, guava, lychee, coconut banana, melon, celery, sweet corn, eggplant, radish, cabbage, tomato, cucumber, carrot, root crop, pumpkin, asparagus, pepper, and so on are widely used for fruit coatings. Waxes and oils, whether used alone or in combination with lipids, have a strong water barrier. However, a dense layer of the same causes anaerobic conditions. When corn zein films were applied to tomatoes, they resulted in delayed changes in colour, weight, and firmness. Antimicrobial agents may be added to edible films to help prevent spoilage of the product on which they are intended to be used (Campos et al., 2011).

b. **In dairy products and confectionaries.**

Milk and dairy products are a significant source of the nutrients needed for child growth and adult health. Despite the fact that cheese is the most diverse category of dairy products, the shelf life of the cheese is limited by microorganisms. Antimicrobial coatings added to the surface of the cheese are one method of limiting negative changes that can occur during handling and storage. A mixture of zein and ethanol, when used as a film-forming solution, produces a better result than normal confectionery glaze. Dyhr and Sorensen (1991) showed that normal conventional sugar coatings can be substituted by sorbitol-based hard coatings on chewable dragees. Edible films are now being used to make a wide range of food products, but the amount of data available in scientific and patent literature is limited.

c. **Meat and poultry**

Meat is characterised by its tissue structure, and it is often subjected to processes that encourage the growth of microorganisms. In recent years, antimicrobial and intelligent packaging have emerged as a food-safety stumbling block. Due to the vast and diverse range of processed meat and poultry products, various methods for managing food-borne pathogens and extending product shelf life are needed (Guo et al., 2014). Galus and Kadziska (2015) developed edible films and coatings to improve the mechanical properties, gas and moisture barriers, and sensory perceptions of meat products. Foaming, dipping, blowing, casting, brushing, person wrapping, or rolling have been used as methods of application (Embuscado and Huber., 2009).

4.Conclusion

For researchers, meeting current consumer expectations for sustainable natural, high-quality, and cleaner food packaging rather than non-biodegradable and nonrenewable petroleum-based polymers and plastics is a challenge. Due to their properties and good performance in food packaging, edible films and coatings made from edible ingredients are a major concern. Edible films are made from natural edible ingredients that are safe to eat by both animals and humans. Through a better understanding of the respiration process, several performances have been advanced that are effective in extending the shelf-life of fresh fruits. Polysaccharides, lipids, and proteins are the three types of normal, edible polymers. There is no single natural polymer that can provide all of the desirable edible film properties, such as moisture, lipid and oil barrier properties, volatiles, and gas barrier properties. flavourless, odourless, and tasteless; translucent or transparent; colourless; soluble in water; good ability to shape coatings and gelling properties; good mechanical and thermal properties; antimicrobial activity; lexible; flavourless, odourless, and tasteless. Recently, research in this area has increased, but some issues remain unresolved for practical application in food packaging, especially processing difficulties, as most recent research has focused on wet methods. Despite the challenges of translating laboratory research to commercial applications, edible films or coatings are safe alternatives to traditional plastic packaging in food packaging and biomedical systems. Hopefully, in a few years, they would be put to good use.

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