

Review Article

Edible films and coatings: characteristics and properties

Bourtoom, T.

*Department of Material Product Technology, Prince of Songkla University,
Hat Yai, Songkhla, 90112, Thailand*

Abstract: Edible films and coatings have received considerable attention in recent years because of their advantages including use as edible packaging materials over synthetic films. This could contribute to the reduction of environmental pollution. By functioning as barriers, such edible films and coatings can feasibly reduce the complexity and thus improve the recyclability of packaging materials, compared to the more traditional non-environmental friendly packaging materials, and may be able to substitute such synthetic polymer films. New materials have been developed and characterized by scientists, many from abundant natural sources that have traditionally been regarded as waste materials. The objective of this review is to provide a comprehensive introduction to edible coatings and films by providing descriptions of suitable materials, reviewing their properties and describing methods of their applications and potential uses.

Keywords: Edible film, coatings, characteristics, hydrocolloids, lipids, composites

INTRODUCTION

Edible films are defined as a thin layer of material which can be consumed and provides a barrier to moisture, oxygen and solute movement for the food. The material can be a complete food coating or can be disposed as a continuous layer between food components (Guilbert, 1986). Edible films can be formed as food coatings and free-standing films, and have the potential to be used with food as gas aroma barrier (Kester and Fennema, 1986). However, technical information is still needed to develop films for food application (Donhowe and Fennema, 1993). Edible films and coatings have received considerable attention in recent years because of their advantages over synthetic films. The main advantage of edible films over traditional synthetics is that they can be consumed with the packaged products. There is no package to dispose even if the films are not consumed they could still contribute to the reduction of environmental pollution. The films are produced exclusively from renewable, edible ingredients and therefore are anticipated to degrade more readily than polymeric materials. The films can enhance the organoleptic properties of packaged foods provided they contain various components (flavorings, colorings, sweeteners). The films can be used for individual packaging of small portions of food, particularly products that currently are not individually packaged for practical reasons such as pears, beans, nuts and strawberries.

The films can be applied inside heterogeneous foods at the interfaces between different layers of components. They can be tailored to prevent deteriorative inter-component moisture and solute migration in foods such as pizzas, pies and candies. The films can function as carriers for antimicrobial and antioxidant agents. In a similar application they can also be used on the surface of food to control the diffusion rate of preservative substances from the surface to the interior of the food. Another possible application for edible films could be their use in multilayer food packaging materials together with non edible films. In this case, the edible films would be the internal layers in direct contact with food materials. Production of edible films causes less waste and pollution, however, their permeability and mechanical properties are generally poorer than synthetic films (Kester and Fennema, 1986). Extensive research is needed on the development of new materials, methods of films formation, methods to improve film properties and the potential applications.

Classification of edible films and coatings

Edible films can be produced from materials with film forming ability. During manufacturing, film materials must be dispersed and dissolved in a solvent such as water, alcohol or mixture of water and alcohol or a mixture of other solvents. Plasticizers, antimicrobial agents, colors or flavors can be added in this process. Adjusting the pH

and/or heating the solutions may be done for the specific polymer to facilitate dispersion. Film solution is then casted and dried at a desired temperature and relative humidity to obtain free-standing films. In food applications, film solutions could be applied to food by several methods such as dipping, spraying, brushing and panning followed by drying. Components used for the preparation of edible films can be classified into three categories: hydrocolloids (such as proteins, polysaccharides, and alginate), lipids (such as fatty acids, acylglycerol, waxes) and composites (Donhowe and Fennema, 1993).

Polysaccharides

Polysaccharides used for edible films or coatings include cellulose, starch derivatives, pectin derivatives, seaweed extracts, exudate gums, microbial fermentation gums and chitosan (Krochta and Mulder-Johnson, 1997). Polysaccharides are generally very hydrophilic resulting in poor water vapor and gas barrier properties. Although coatings by polysaccharide polymers may not provide a good water vapor barrier, these coatings can act as sacrificing agents retarding moisture loss from food products (Kester and Fennema, 1986).

a. Cellulose and derivatives

Cellulose is composed of repeating D-glucose units linked through β -1, 4 glycosidic bonds (Figure 1A). In its native state, the hydroxymethyl groups of anhydroglucose residues are alternatively located above and below the plane of the polymer backbone. This results in very tight packing of polymer chains and a highly crystalline structure that resists salvation in aqueous media. Water solubility can be increased by treating cellulose with alkali to swell the structure, followed by reaction with chloroacetic acid, methyl chloride or propylene oxide to yield carboxymethyl cellulose (CMC), methyl cellulose (MC), hydroxypropyl cellulose (HPMC) or hydroxypropyl cellulose (HPC) (Figure 1B-1E). MC, HPMC, HPC and CMC film possess good film-forming characteristic; films are generally odorless and tasteless, flexible and are of moderate strength, transparent, resistance to oil and fats, water-soluble, moderate to moisture and oxygen transmission (Krochta and Mulder-Johnson, 1997). MC is the most resistant to water and it is the lowest hydrophilic cellulose derivatives (Kester and Fennema, 1986); however, the water vapor permeability of cellulose ether film is still relatively high. MC and HPMC have the ability to form thermally induced gelatinous coating; they

have been used to retard oil absorption in deep frying food products (Kester and Fennema, 1986; Balasubramanian *et al.*, 1997). MC could be applied as coating on confectionery products as barrier to lipid migration (Nelson and Fennema, 1991). A number of researchers have investigated composite films composed of MC or HPMC and various kinds of solids, such as beeswax and fatty acids (Kamper and Fennema, 1984; Kester and Fennema, 1986; Greener and Fennema, 1989a; Koelsch and Labuza, 1992; Debeaufort *et al.*, 1993; Park *et al.*, 1994a). Many of these have water vapor permeability comparable to low density polyethylene (LDPE). These composite films were all polymer-lipid bilayer formed from aqueous ethanolic solutions of cellulose ether fatty acids.

b. Chitin and chitosan

Chitin is the second most abundant naturally occurring biopolymer (after cellulose) and is found in the exoskeleton of crustaceans, in fungal cell walls and other biological materials (Andrade and Xu, 1997). It is mainly poly (β -(1-4)-2-acetamide-D-glucose), which is structurally identical to cellulose except that secondary hydroxyl on the second carbon atom of the hexose repeat unit is replaced by an acetamide group (Figure 2 A). Chitosan is derived from chitin by deacetylation in the presence of alkali. Therefore, chitosan is a copolymer consisting of (β -(1-4)-2-acetamido-D-glucose and (β -(1-4)-2-acetamide-D-glucose units with the latter usually exceeding 80% (Figure 2B). Chitosans are described in terms of the degree of deacetylation and average molecular weight and their importance resides in their antimicrobial properties in conjunction with their cationicity and their-forming properties (Muzzarelli, 1996). Chitosan can form semi-permeable coatings, which can modify the internal atmosphere, thereby delaying ripening and decreasing transpiration rates in fruits and vegetables. Films from aqueous chitosan are clear, tough, flexible and good oxygen barriers (Sandford, 1989; Kalplan *et al.*, 1993). Carbon dioxide permeability could be improved by methylation of polymers. Butler *et al.* (1996) observed that films from chitosan were rather stable and their mechanical and barrier properties changed only slightly during storage. Chitosan coatings are usually used on fruit and vegetable products such as strawberries, cucumbers, bell peppers as antimicrobial coating (El Ghaouth *et al.*, 1991a, 1991b), and on apples, pears, peaches and plums as gas barrier (Elson and Hayes, 1985; Davies *et al.*, 1989).

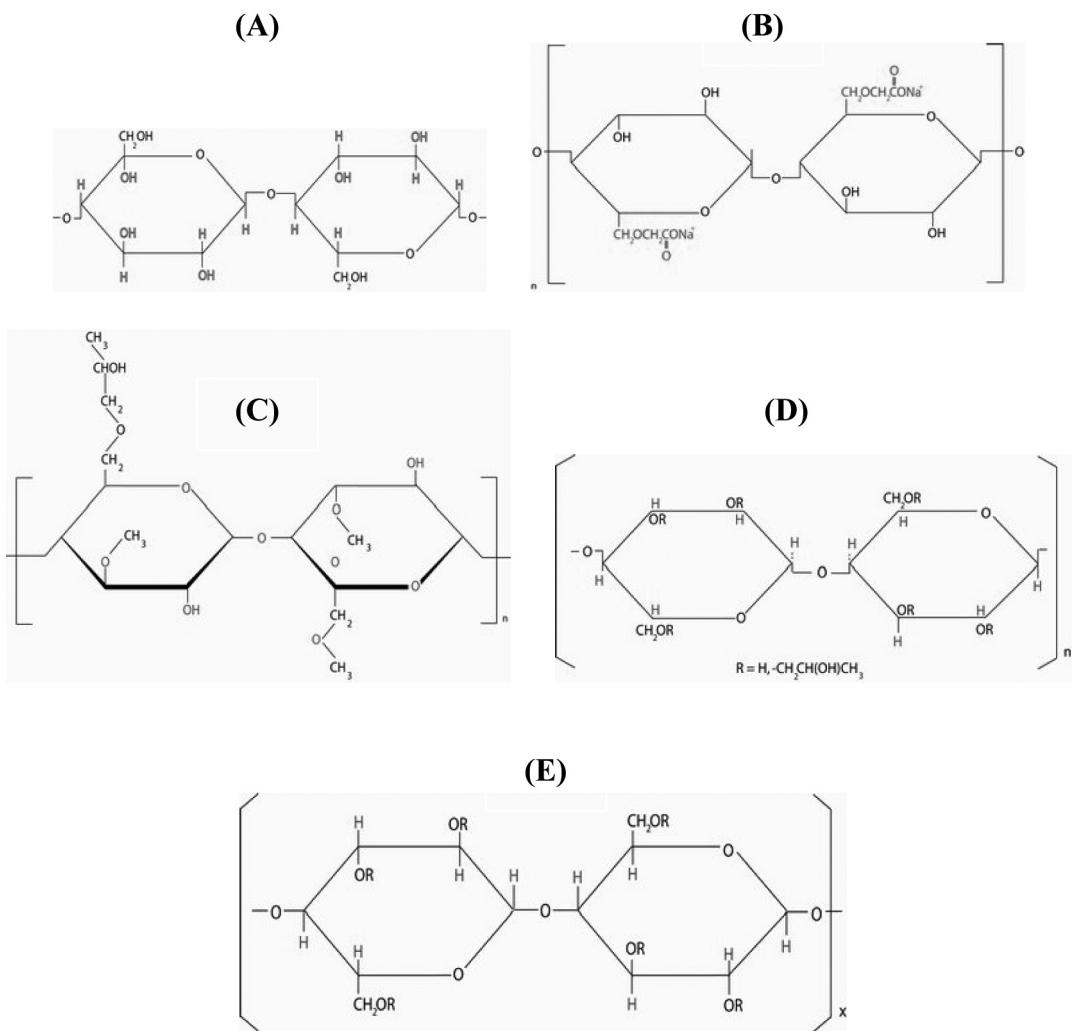


Figure 1: Structure of (A) cellulose, (B) carboxymethyl cellulose, (C) methyl cellulose, (D) hydroxypropyl cellulose and (E)hydroxypropyl methyl cellulose. (Source: Murray *et al.*, 2000)

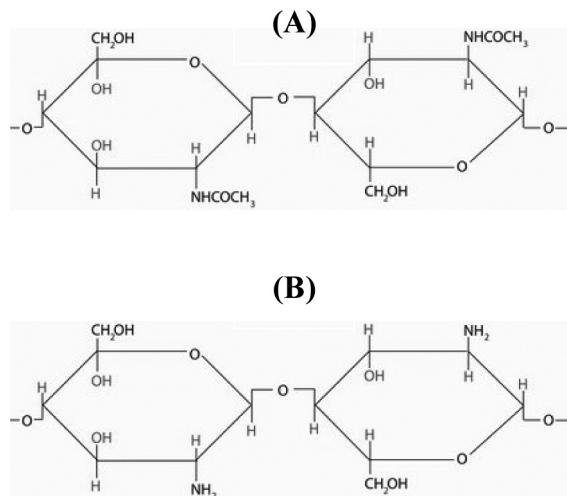


Figure 2: Structure of (A) chitin and (B) chitosan (Source: Muzzarelli, 1996)

c. Starch

Starch is a polymeric carbohydrate composed of anhydroglucose units. This is not a uniform material and most starches contain two types of glucose polymers: a linear chain molecule termed amylose and a branched polymer of glucose termed amylopectin (Rodriguez *et al.*, 2006). Starches are often used in industrial foods. They have been used to produce biodegradable films to partially or entirely replace plastic polymers because of its low cost and renew ability, and it has good mechanical properties (Xu *et al.*, 2005). High amylose starch such as corn starch is a good source for films formation, free-standing films can be produced from aqueous solution of gelatinized amylose and drying. Normal corn starch consists of approximately 25% amylose and 75% amylopectin. Mutant varieties of corn are produced which contain starch with up to 85% amylose (Whistler and Daniel, 1985). Mark *et al.* (1966) reported that films produced from high amylose corn starch (71% amylose) had no detectable oxygen permeability at RH levels less than 100%. This was true for both unplasticized and plasticized (16% glycerol) films. This result is surprising in light of the fact that addition of plasticizers and absorption of water molecules by hydrophilic polymers increase polymer chain mobility and generally lead to increased gas permeability (Banker *et al.*, 2000). Partial etherification of high-amylose starch with propylene oxide, to yield the hydroxypropylated derivative, improves water solubility.

Lipid films

Lipid compounds utilized as protective coating consist of acetylated monoglycerides, natural wax, and surfactants. The most effective lipid substances are paraffin wax and beeswax. The primary function of a lipid coating is to block transport of moisture due to their relative low polarity. In contrast, the hydrophobic characteristic of lipid forms thicker and more brittle films. Consequently, they must be associated with film forming agents such as proteins or cellulose derivatives (Debeaufort *et al.*, 1993). Generally, water vapor permeability decrease when the concentration of hydrophobicity phase increases. Lipid-based films are often supported on a polymer structure matrix, usually a polysaccharide, to provide mechanical strength.

a. Waxes and paraffin

Paraffin wax is derived from distillate fraction of crude petroleum and consists of a mixture of solid hydrocarbon resulting from ethylene catalytic polymerization. Paraffin wax is permitted for use on raw fruit and vegetable and cheese. Carnauba wax is an exudate from palm tree leaves (*Copernica cerifera*) (Figure 4A). Beewax (white wax) is produced from honeybees (Figure 4B). Candelilla is obtained from candelilla plant (Figure 4C). Mineral oil consists of a mixture of liquid paraffin and naphthenic hydrocarbon (Hernandez, 1994). Waxes are used as barrier films to gas and moisture (skin on fresh fruits) and to improve the surface appearance of various foods (e.g., the sheen on

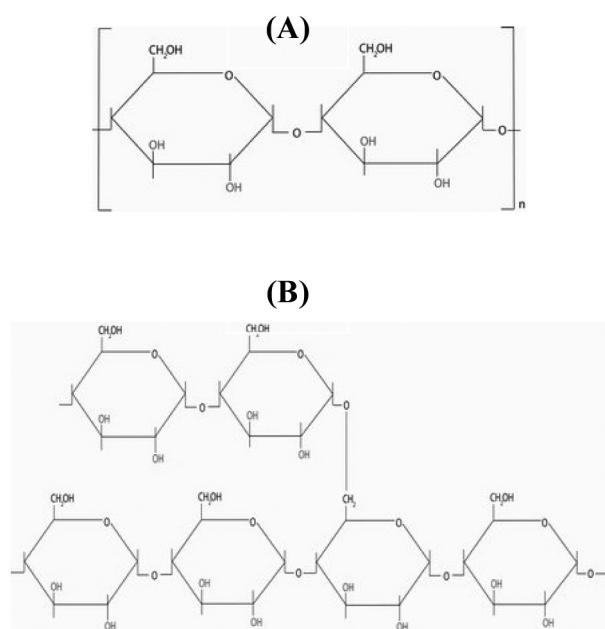


Figure 3: Structure of (A) amylose and (B) amylopectin (Source: Whistler and Daniel, 1985)

sweet). If applied as a thick layer, they must be removed before consumption (certain cheese); when used in thin layers, they are considered edible. Waxes (notably paraffin, carnauba, candellila and bee wax) are the most efficient edible compounds providing a humidity barrier.

b. Acetoglyceride

Acetylation of glycerol monostearate by its reaction with acetic anhydride yields 1-stearodiacetin. This acetylated monoglyceride displays the unique characteristic of solidifying from the molten state into a flexible, wax-like solid (Feuge *et al.*, 1953). Most lipids in the solid state can be stretched to only about 102% of their original length before fracturing. Acetylated glycerol monostearate, however, can be stretched up to 800% of its original length (Jackson and Lutton, 1952), water vapor permeability of this film is much less than that of polysaccharide films with the exception of methyl cellulose or ethyl cellulose (Kester and Fennema, 1986). Acetylated monoglyceride coatings have been used on poultry and meat cuts to retard moisture loss during storage (Kester and Fennema, 1986).

c. Shellac resins

Shellac resins are a secretion by the insect *Laccifer lacca* and is composed of a complex mixture of aliphatic alicyclic hydroxyl acid polymers (Figure 4D). This resin is soluble in alcohols and in alkaline solutions. Shellac is not a GRAS substance; it is only permitted as an indirect food additive in food coatings and adhesives. It is mostly used in coatings for the pharmaceutical industry and only few studies have been reported on foods (Hernandez, 1994). Rosins which are obtained from the oleoresins of the pine tree are residues left after distillation of volatiles from the crude resin. Resin and its derivatives are widely used in coating for citrus and other fruits (Hagenmaier and Baker, 1993). These coatings were designed primarily to impart high-gloss at the time of inspection by the buyer, usually after application of the coating. When coatings are applied to fruit, they form an additional barrier through which gases must pass. Because coatings differ in gas permeance and ability to block openings in the peel, they have different effects on gas exchange (Hagenmaier and Baker, 1993). Citrus fruits with shellac and wood resin-based coatings generally have lower internal O₂, higher internal

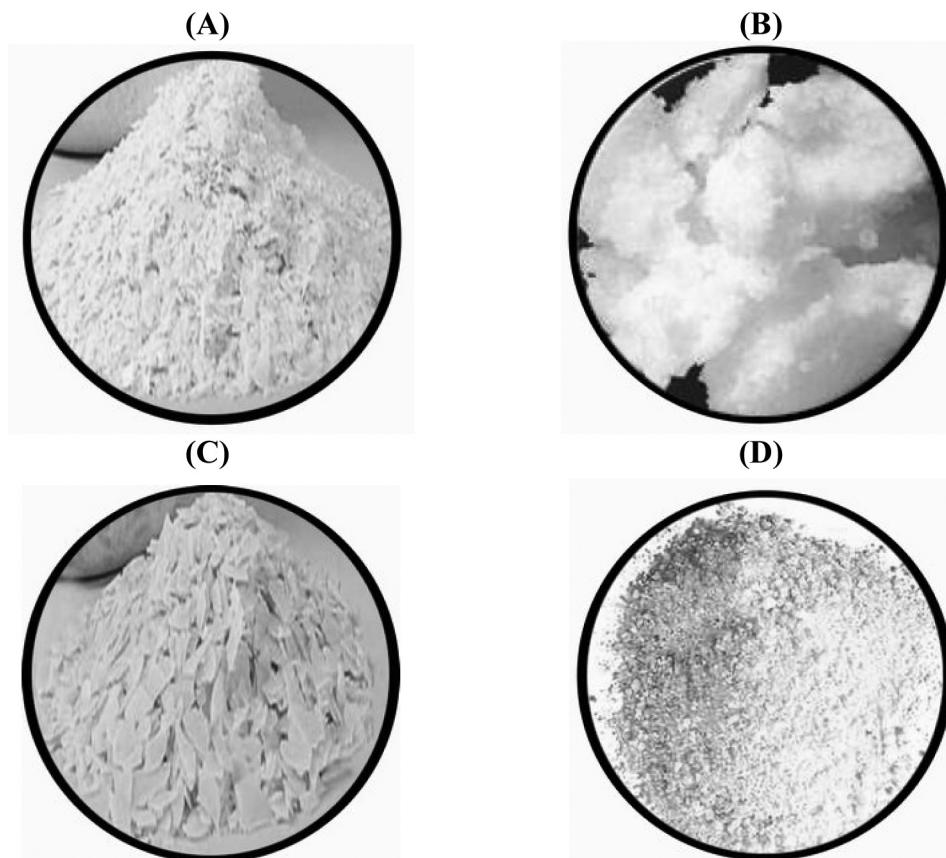


Figure 4: Illustrative pictures of (A) carnauba wax, (B) beewax, (C) carnauba wax and (D) shellac resins

CO_2 and higher ethanol content than fruits with wax coatings (Hagenmaier and Baker, 1993). High ethanol content, in turn, is an indication of off-flavor (Ahmad and Khan, 1987; Cohen *et al.*, 1990; Ke and Kader, 1990). Shellac- and wood resin-based coatings also tend to increase prevalence of post harvest pitting (Petracek *et al.*, 1997, 1998).

Protein films

In their native states, proteins generally exist as either fibrous proteins, which are water insoluble and serve as the main structural materials of animal tissues, or globular proteins, which are soluble in water or aqueous solutions of acids, bases or salts and function widely in living systems (Scope, 1994). Fibrous proteins are fully extended and associated closely with each other in parallel structures, generally through hydrogen bonding, to form fibers. The globular proteins fold into complicated spherical structures held together by a combination of hydrogen, ionic, hydrophobic and covalent (disulfide) bonds (Scope, 1994). The chemical and physical properties of these proteins depend on the relative amounts of the component amino acid residues and their placement along the protein polymer chain. Several globular proteins, including wheat gluten, corn zein, soy protein, and whey protein, have been investigated for their film properties. Protein films are generally formed from solutions or dispersions of the protein as the solvent/carrier evaporates. The solvent/carrier is generally limited to water, ethanol or ethanol-water mixtures (Kester and Fennema, 1986). Generally, proteins must be denatured by heat, acid, base, and/or solvent in order to form the more extended structures that are required for film formation. Once extended, protein chains can associate through hydrogen, ionic, hydrophobic and covalent bonding. The chain-to-chain interaction that produces cohesive films is affected by the degree of chain extension and the nature and sequence of amino acid residues. Uniform distribution of polar, hydrophobic, and/or thiol groups along the polymer chain increase the likelihood of the respective interactions. Increased polymer chain-to-chain interactions result in films that are stronger but less flexible and less permeable to gases, vapors and liquids (Kester and Fennema, 1986). Polymers containing groups that can associate through hydrogen or ionic bonding result in films that are excellent oxygen barriers but are susceptible to moisture (Salame, 1986). Thus, protein films are expected to be good oxygen barriers at low relative humidities. Various types of protein have been used as edible films. These include gelatin, casein, whey protein, corn zein, wheat gluten, soy protein, mung

bean protein, and peanut protein (Gennadious *et al.*, 1993; Bourtoom, 2008).

a. Gelatin films

Gelatin is obtained by controlled hydrolysis from the fibrous insoluble protein, collagen, which is widely found in nature as the major constituent of skin, bones and connective tissue (Figure 5A). Gelatin is composed of a unique sequence of amino acids. The characteristic features of gelatin are high content of the amino acids glycine, proline and hydroxyproline. Gelatin also has a mixture of single and double unfolded chains of hydrophilic character (Ross, 1987). At approximately 40 °C, gelatin aqueous solutions are in the sol state and form physical, thermoreversible gels on cooling. During gelation, the chains undergo a conformational disorder-order transition and tend to recover the collagen triple-helix structure (Ross-Murphy, 1992). Gelatin films could be formed from 20-30% gelatin, 10-30% plasticizer (glycerin or sorbitol) and 40-70% water followed by drying the gelatin gel (Guilbert, 1986). Gelatin is used to encapsulate low moisture or oil phase food ingredients and pharmaceuticals. Such encapsulation provides protection against oxygen and light, as well as defining ingredient amount or drug dosage (Gennadious *et al.*, 1994). In addition, gelatin films have been formed as coatings on meats to reduce oxygen, moisture and oil transport (Gennadious *et al.*, 1994).

b. Corn zein films

Zein is the most important protein in corn (Figure 5B). It is a prolamin protein and therefore dissolves in 70–80% ethanol (Dickey and Parris, 2001, 2002; Landry, 1997). Zein is a relatively hydrophobic and thermoplastic material. The hydrophobic nature of zein is related to its high content of non-polar amino acids (Shukla and Cheryan, 2001). Zein has excellent film forming properties and can be used for fabrication of biodegradable films. The zein bio-film is formed through the development of hydrophobic, hydrogen and limited disulfide bonds between zein chains (Guilbert, 1986). Edible films can be formed by drying aqueous ethanol solution of zein (Gennadious and Weller, 1990). Formation of films is believed to involve development of hydrophobic, hydrogen and limited disulfide bonds between zein chains in the film matrix (Gennadious *et al.*, 1994). The resulting films are brittle and therefore require plasticizer addition for increasing flexibility (Park, 1991). Zein films are relatively good water vapor barriers compared to other edible films (Guilbert, 1986). Water vapor barrier properties can be improved by adding fatty acids or by using a cross-linking reagent. But when

cross-linking agents are used, the edibility of those films is of concern (Alikonis, 1979). Zein coating have also shown an ability to reduce moisture and firmness loss and delay color change (reduce oxygen and carbon dioxide transmission) in fresh tomatoes (Park *et al.*, 1994b).

c. Wheat gluten films

Wheat gluten is a general term for water-insoluble proteins of wheat flour which is composed of a mixture of polypeptide molecules, considered to be globular proteins (Figure 5C). Cohesiveness and elasticity of gluten give integrity to wheat dough and facilitate film formation. Wheat gluten contains the prolamine and glutelin fractions of wheat flour proteins, typically referred to as gliadin and glutenin, respectively. While gliadin is soluble in 70% ethanol, glutenin is not (Gennadios and Weller, 1990). Although insoluble in natural water, wheat gluten dissolves in aqueous solutions of high or low pH at low ionic strength (Krull and Inglett, 1971). Edible films can be formed by drying aqueous ethanol solution of wheat gluten (Gennadios and Weller, 1990). Cleavage of native disulfide bonds during heating of film-forming solutions and then formation of new disulfide bonds during film drying are believed to be important to the formation of wheat gluten films structure, along with hydrogen and hydrophobic bonds (Gennadios and Weller, 1990). Addition of plasticizer such as glycerin in gluten films is necessary to improve film flexibility (Gennadios *et al.*, 1994). However, increasing film flexibility by increasing sorbitol content reduces film strength, elasticity and water vapor barrier properties (Gontard *et al.*, 1992). A review on gluten film was done by Gennadios and Weller (1990). Gennadios and Weller (1992) confirmed the effect of wheat gluten purity on film appearance and mechanical properties, i.e., a greater purity gluten results in a stronger and clearer films. Herald *et al.* (1995) investigated the effect of plasticizer size of wheat gluten; films prepared from spray-dried wheat gluten were stronger than films from flash-dried which had larger sized particles. When used as a coating on grade A-quality shell eggs, the egg quality was maintained for 30 days. Tensile strength of gluten films can be improved by using a cross-linking agent such as glutaraldehyde, or heat curing at 80°C (Gennadios and Weller, 1992; Koelsch *et al.*, 1992). Pochat-Bohatier *et al.* (2005) determined the influence of relative humidity on carbon dioxide sorption in wheat gluten films. The results showed that the permeability was based on the rise of CO₂ solubility and diffusivity with the increase in RH. The increase in the water content of wheat gluten improves the affinity between carbon dioxide and

the protein matrix, leading to outstanding sorption values for high RH.

d. Soy protein films

The protein content of soybeans (38-44%) is much higher than the protein content of cereal grain (8-15%). Most of the protein in soybeans is insoluble in water but soluble in dilute neutral salt solutions (Figure 5D). Thus, soy protein belongs to the globulin classification (Kinsella, 1979). Soy protein is globular in nature and is further classified into 2S, 7S, 11S and 15S fraction according to relative sedimentation rates (Gennadios *et al.*, 1994). The principal components are the 7S (conglycinin) and 11S (glycinin) fractions, both of which have a quaternary (subunit) structure (Kinsella *et al.*, 1985). Soy protein is high in asparagine and glutamine residues. Both conglycinin and glycinin are tightly folded proteins. While the extent of disulfide cross-linking of conglycinin is limited due to only two to three cysteine groups per molecule, glycinin contains 20 intramolecular disulfide bonds (Kinsella, 1979). Alkali and heating both cause dissociation and subsequent unfolding of glycinin due to disulfide bond cleavage (Kinsella, 1979). Edible films based on soy protein can be produced in either of two ways: surface film formation on heated soymilk or film formation from solutions of soy protein isolate (SPI) (Gennadios and Weller, 1992). Soymilk is produced by grinding soybeans with water followed by separation of milk from extracted soybeans. To form films from both soymilk and SPI, (a) heating of film solutions to disrupt the protein structure, cleave native disulfide bonds and expose sulfhydryl groups and hydrophobic groups, and then (b) formation of new disulfide, hydrophobic and hydrogen bonds during film drying are believed to be important to the formation of soy protein film structure (Gennadios *et al.*, 1994; Subirade *et al.*, 1998). The use of soy protein in the formation of films or coatings on food products has been investigated (Baker *et al.*, 1994; Gennadios *et al.*, 1994; Stuchell and Krochta, 1994; Kunte *et al.*, 1997; Rhim *et al.*, 2000, 2002)

Composite films

Edible films and coatings may be heterogeneous in nature, consisting of a blend of polysaccharides, protein, and/or lipids. This approach enables one to utilize the distinct functional characteristics of each class of film former (Kester and Fennema, 1986). The combination between polymers to form films could be from proteins and carbohydrates, proteins and lipids, carbohydrates and lipids or synthetic polymers and natural polymers. The main objective of producing composite films is to

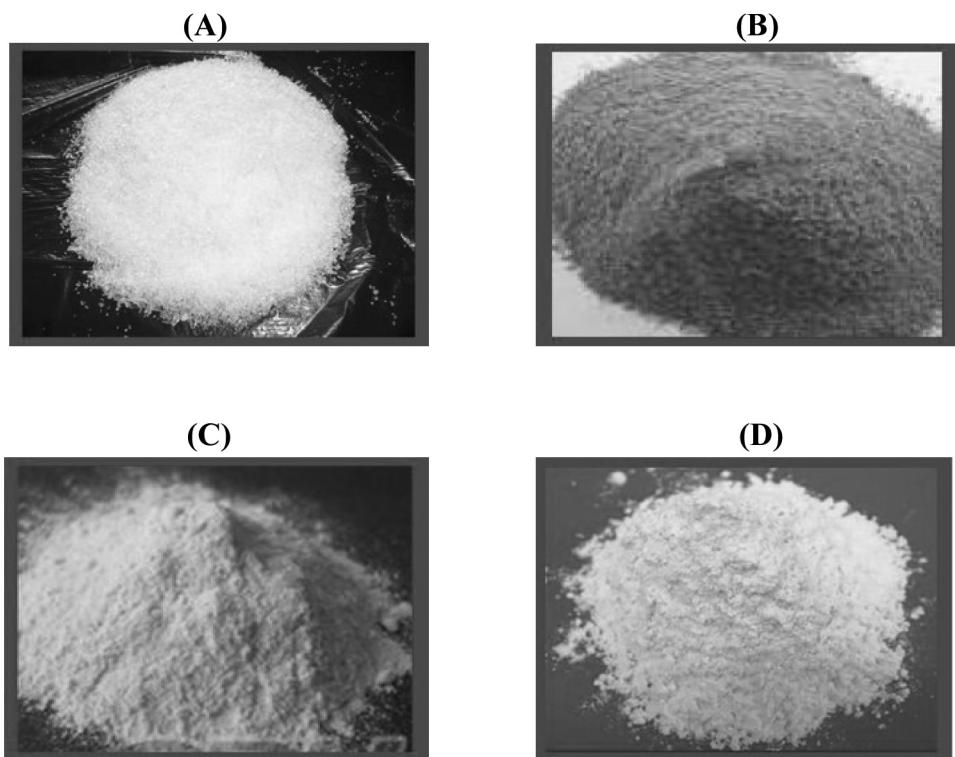


Figure 5: Illustrative pictures of (A) gelatin, (B) corn zein, (C) wheat gluten and (D) soy protein isolated

improve the permeability or mechanical properties as dictated by the need of a specific application. These heterogeneous films are applied either in the form of an emulsion, suspension, or dispersion of the non-miscible constituents, or in successive layers (multilayer coating or films), or in the form of a solution in a common solvent. The method of application affects the barrier properties of the films obtained (Guilbert, 1986). Kamper and Fennema (1984) introduced the emulsion films from methyl cellulose and fatty acids to improve water vapor barrier of cellulose films. Recently, many researchers have extensively explored the development of composite films based on the work of Kamper and Fennema (1984). Examples of these studies are using lipid and hydroxypropyl methyl cellulose (Hagenmaier and Shaw, 1990), methyl cellulose (MC) and lipid (Greener and Fennema, 1989b), MC and fatty acid (Sapru and Labuza, 1994), corn zein, MC and fatty acid (Park *et al.*, 1996), whey isolate and lipids (McHugh and Krochta, 1994), casein and lipids (Avena-Bistillos, 1993), gelatin and soluble starch (Arvanitoyannis *et al.*, 1997), hydroxypropyl starch and gelatin (Arvanitoyannis *et al.*, 1998), corn zein and corn starch (Ryu *et al.*, 2002), gelatin and fatty acid (Bertan *et al.*, 2005), soy protein isolate and gelatin (Cao *et al.*, 2007), soy protein isolate and polylactic acid (Rhim *et al.*, 2007)

Final remarks

The concept of edible films and coatings represents a stimulating route for creating new packaging materials. This is because edible films and coatings are available with a wide range of properties that can help to alleviate many problems encountered with foods. Edible films can be produced from materials with film forming ability. Components used for the preparation of edible films can be classified into three categories: hydrocolloids, lipids and composites. Hydrocolloid films possess good barrier properties to oxygen, carbon dioxide, and lipids but not to water vapor. Most hydrocolloid films also possess superb mechanical properties, which are quite useful for fragile food products. However, potential functions and applications of the films and coatings warrant increased considerations. Extensive research is still needed on the methods of films formation and methods to improve film properties and the potential applications.

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