

### EDIBLE AND BIODEGRADABLE POLYMER FILMS:

CHALLENGES AND OPPORTUNITIES

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## STATUS SUMMARY

ackaging protects food from its environment. Quality and shelf life are reduced when food, through interaction with its environment, gains or loses moisture or aroma, takes up oxygen (leading to oxidative rancidity), or becomes contaminated with microorganisms. In multicomponent foods, quality and shelf life are also reduced when moisture, aromas, or lipids migrate from one food component to another. Food packaging also provides important information to the consumer and enables convenient dispensing of food. Food packaging, however, has become a central focus of waste reduction efforts. Packaging represents approximately 30wt% of municipal solid waste (MSW) but appears more significant because it occupies close to two-thirds of trash can volume due to its bulk (Hunt et al., 1990).

The public perceives plastic packaging, in particular, as using valuable non-renewable natural resources and as being neither biodegradable nor recyclable. Consumers are also concerned about the environmental effects of disposable food service items, such as containers, plates, cups, and utensils, as well as other plastic disposable items. Plastic packaging, however, makes up 13% of the 30wt% packaging share of MSW and, thus, represents only 4wt% of MSW (Rowatt, 1993). Plastic packaging is safe, strong, light-weight, and economical. Synthetic resin prices have recently been as low as \$0.50-0.75 per pound (Anonymous, 1995). Finished synthetic film prices, per pound, have recently ranged from \$1.00 for low density polyethylene (LDPE), \$2.00 for polystyrene (PS), \$3.00 for polyethylene terephthalate (PET), to \$5.00 for ethylene-vinyl alcohol copolymer (EVOH; Grimsley, 1995). Plastic also has helped to hold down packaging costs and waste. A recent study found that if all plastic packaging in Europe were replaced with alternative materials, the weight of packaging waste would increase by 400%, volume would increase by 250%, and packaging cost would increase 200% (Fleming, 1992).

Edible and biodegradable polymer films offer alternative packaging without the environmental costs. Interest and research activity in these films have been especially intense over the past ten years. Although edible films are not meant to totally replace synthetic packaging films, they do have the potential to reduce packaging and to limit moisture, aroma, and lipid migration between food components where traditional packaging cannot function. Biodegradable packaging, on the other hand, has been viewed by many as having the potential to totally replace synthetic, non-biodegradable packaging in some applications.

This Scientific Status Summary presents the challenges and opportunities for using edible and biodegradable polymer films. Non-polymeric materials such as lipids (e.g., waxes, acetylated monoglycerides, and fatty acids) and resins (e.g., shellac and wood rosin) are not addressed in this summary, except when used in combination with edible and biodegradable polymers to make composite films.

### **DEFINITIONS AND OPPORTUNITIES**

Edible Polymer Films. Generally, an edible film is defined as a thin layer of edible material formed on a food as a coating or placed (pre-formed) on or between food components. Its purpose is to inhibit migration of moisture, oxygen, carbon dioxide, aromas, and lipids, etc.; carry food ingredients (e.g., antioxidants, antimicrobials, flavor); and/or improve mechanical integrity or handling characteristics of the food. In some cases, stand-alone edible films with good mechanical properties can replace synthetic packaging films.

Effective edible films formed as coatings on foods by dipping, spraying, or panning could reduce packaging requirements and waste. An edible film coating, acting as an efficient moisture, oxygen, or aroma barrier, can reduce the amount of packaging. The barrier characteristics of an edible film coating may also allow conversion from a multilayer,

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multicomponent plastic package to a single component recyclable package.

Edible film coatings also may help maintain the quality of foods after packaging is opened by protecting against moisture change, oxygen uptake, and aroma loss. Edible films formed or placed between food components also can improve the quality of multicomponent foods.

Edible films with adequate mechanical properties could conceivably also serve as edible packaging for select foods (e.g., pouches for dried soup which would become part of the prepared soup). The sanitary condition of the edible package would need to be maintained during storage, transportation, and marketing. The end result would be source reduction and/ or improved recyclability of the remaining elements of the packaging system. Thermoforming, overwrapping, and shrinkwrapping of foods with previouslyformed edible films are also conceivable.

Biodegradable Polymer Films. The introduction of 'biodegradable" plastics that were combinations of natural (e.g., starch) and synthetic (e.g., LDPE) polymers created many skeptics. Although these materials disintegrated in composting operations, they did not completely biodegrade. A recent International Workshop on Biodegradability determined that, for materials to be called biodegradable, they must be degraded completely by microorganisms in a composting process to only natural compounds such as carbon dioxide, water, methane, and biomass (Anonymous, 1993a). Biodegradation includes two steps - depolymerization (chain cleavage) and mineralization to carbon dioxide, water, salts, etc. — and involves three key elements: appropriate microorganisms, well-tuned environment, and vulnerable polymer substrate (Kaplan et al., 1993). A warm, moist environment with acceptable range of pH, nutrients, and oxygen for the applicable microorganisms is conducive to efficient biodegradation processes.

Replacing conventional synthetic packaging with biodegradable polymers can reduce use of non-renewable resources and decrease waste through biological recycling to the biosystem. Use of biodegradable polymers that replace petroleum-based synthetic polymers may also slow the introduction of fossil-fuel-derived carbon dioxide into the atmosphere. Biodegradation or incineration of renewable biologically-derived polymers recycles carbon dioxide to the atmosphere, rather than increasing it. Consideration of biodegradable polymer films for packaging and other applications makes most sense when recovery of conventional synthetic plastic packaging for recycling or energy recovery is difficult. The effort to develop biodegradable polymers for food packaging has been especially urgent for the Navy, whose ships, along with non-military vessels, are now legally prohibited from dumping persistent waste at sea. Interest in biodegradable packaging for reduction of MSW is increasing as municipalities increase availability of curbside pickup for large-scale composting. Some municipalities are also providing equipment for small-scale domestic composting. In some cases, biodegradable polymer films could be handled by domestic garbage disposals and, subsequently, municipal waste treatment plants.

### SAFETY AND HEALTH ISSUES

Edible Polymer Films. Determination of the acceptability of materials for edible polymer films follows procedures identical to determining the appropriateness of such materials for food formulation (Heckman and Heller, 1986): (1) an edible polymer will be generally recognized as safe (GRAS) for use in edible films if the material has previously been determined GRAS and its use in an edible film is in accordance with current good manufacturing practices (food grade, prepared and handled as a food ingredient, and used in amounts no greater than necessary to perform its function) and within any limitations specified by the Food and Drug Administration (FDA); (2) if the edible polymer film material use is not currently GRAS but the manufacturer can demonstrate safety, the manufacturer may either file a GRAS Affirmation Petition to the FDA or proceed to market the material without FDA concurrence (self-determination); (3) the manufacturer may not need to establish that use of the edible polymer in

### DEFINITIONS

Elongation: Dimensionless measure of a film's ability to stretch. Specifically, the % change in length experienced by a material due to pulling stress before breakage.

Permeability: Also referred to as permeability coefficient. A measure of tendency to absorb, transfer, and desorb a permeant (e.g., water vapor or oxygen) from one side of a film to the other, when the two sides of a film material are exposed to different concentrations of permeant. Specifically, the amount of a given permeant that will transfer per unit time through a unit area of a given film material at unit driving force of permeant per unit thickness of film. Dimensions are: amount of permeant/time • area • driving force/thickness of film.

Tensile strength: Also known as ultimate tensile strength. A measure of a film's strength. Specifically, the maximum pulling stress (force/ cross-sectional area) that a material can sustain before breakage. Dimensions are: force/cross-sectional area.

Thermoplastic: Descriptive term applied to polymers that soften and eventually flow with increasing temperature, with no coincident chemical reactions. Upon cooling, these polymers return to their original state. This characteristic is generally possessed by linear polymers that are not crosslinked and that are not strongly hydrogen-bonded to adjacent polymer chains. Such tendency to soften and flow allows the extrusion of thermoplastic polymers to produce films and other structures.

edible films is GRAS if the material received pre-1958 FDA clearance and thus has "prior sanction"; (4) finally, if the material cannot be demonstrated to be GRAS or "prior-sanction," the manufacturer must submit a food additive petition to the FDA.

The materials that have received the greatest attention for edible film use are cellulose ethers, starch, hydroxypropylated starch, corn zein, wheat gluten, soy protein, and milk proteins. Food processors considering use of protein-based films must be aware that some consumers have wheat gluten intolerance (Celiac disease), milk protein allergies, or lactose intolerance. Use of such films as coatings on foods must be declared appropriately to the consumer, no matter how small the amount used. The nutritional quality of materials used for edible films may be affected, negatively or positively, by the temperature, pH, and/or solvents used in film preparation. Aside from these considerations, no intrinsic nutritional or health problems have been identified for edible films. In fact, edible films can be carriers of nutritional supplements, and protein-based films, depending on protein quality, can be an important nutritional enhancement of the food.

Attention to the microbial safety of edible films is guided by standard considerations of water activity, pH, temperature, oxygen supply, and time. Importantly, edible films are effective carriers of antimicrobials, which improve the microbial stability of film and food alike (Cuppett, 1994).

Biodegradable Polymer Films. FDA regulates all materials proposed for food packaging to ensure they are safe for food contact under conditions of intended use. There are several categories of biodegradable polymers acceptable for use as food packaging (Thorsheim, 1996): (1) if a biodegradable polymer developed for use in food packaging is found to be GRAS by the developer (self-determination) or affirmed as GRAS by the FDA, the polymer may be used in food packaging; (2) if a GRAS polymer is combined with a food-grade synthetic polymer to enhance biodegradable character, the developer should supply the FDA with information that addresses whether the proposed use of the product can be considered "good manufacturing practice." FDA requires information on the migration profile and identity of migrants from the biodegradable packaging to food during typical storage conditions and times; the agency also requests information on environmental aspects of the biodegradable package use; (3) if a biodegradable polymer developed for use in food packaging is not GRAS, it may be used only if a food additive petition and environmental assessment is approved.

Non-edible, polymeric materials available for biodegradable polymer films include certain cellulose-based products (e.g., cellophane), microbial polyesters (e.g., polyhydroxybutyrate/valerate copolymers produced by bacteria), biodegradable synthetic polymers (e.g., polylactic acid produced from fermentation lactic acid) and combinations of starch with biodegradable synthetic polymers (e.g., polyvinyl alcohol). Natural polymers or polymers derived from natural monomers offer the greatest opportunities, since their biodegradability and environmental compatibility are assured (Swift, 1993). Edible polymer film materials are also acceptable for biodegradable polymer films.

The challenge for the successful use of biodegradable polymer products is achieving controlled lifetime. Products must remain stable and function properly during storage and intended use, but then biodegrade efficiently later (Kaplan et al., 1993). This means avoiding environmental conditions conducive to biodegradation during product storage and use, and then optimizing environmental conditions for biodegradation at the intended time. Only by appropriately controlling water activity, pH, nutrients, temperature, oxygen levels, and time can package integrity and microbial stability be assured. Thus, biodegradable polymer films may be safely stored in dry environments and used with dry food products over a relatively long period of time, whereas acceptable time of storage in moist environments or time of use with moist foods would be limited.

### MATERIALS

Polysaccharide, Cellulose-based. Cellophane, a regenerated form of cellulose, was the first transparent, flexible packaging film. Since the introduction of synthetic thermoplastic packaging films in the 1950s, cellophane sales have dropped by 90% (Jenkins and Harrington, 1991). Biodegradable but not edible, cellophane makes a strong package due to its good tensile strength and elongation (Taylor, 1986). Cellophane film tears easily when notched, and this characteristic makes it an excellent material for easy-opening applications with tear tape. Other attributes include excellent printability and good machinability (Hanlon, 1992).

Like other natural polymers, cellophane is sensitive to moisture and is not a good moisture barrier. Because of their inherent hydrophilic nature, sensitivity to moisture is perhaps the greatest problem with all biological polymers. Cellophane is not thermoplastic and is, therefore, not heat-sealable. Because of its poor moisture barrier properties and non-sealability, cellophane is often coated (e.g., with nitrocellulosewax [NC-W] blend or polyvinylidene chloride [PVDC]) and is often additionally laminated (e.g., with metallized polyester). Coated cellophane has a water vapor permeability (WVP) as low as high density polyethylene (HDPE; Taylor, 1986). Cellophane is a good barrier to oxygen, fats, oils, and flavors at low relative humidity (RH; Hanlon, 1992; Salame, 1986), but its barrier properties are compromised at intermediate and high RH (Taylor, 1986). Coating with NC-W or PVDC reduces the influence

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of RH on these barrier properties. NC-W-coated cellophane biodegrades totally, whereas PVDC-coated cellophane degrades to small PVDC fragments (Whitehouse, 1990). Tables 1 and 2 summarize properties and applications of uncoated and NC-W-coated cellophane and other biodegradable films.

Unlike cellophane, *cellulose acetate* is a thermoplastic material. Films can be obtained by either extrusion or solvent casting. Cellulose acetate films are crystal clear and tough. Barrier properties against moisture and gases are not good, but cellulose acetate film is excellent for certain high-moisture products because it breathes and does not fog up (Hanlon, 1992). Cellulose acetate is also a good

barrier to greases and oils. Although chemical substitution of cellulose generally slows biodegradation, cellulose acetate exhibits mineralization (Kaplan et al., 1993). Thus, although cellulose acetate is not edible, it does appear to be biodegradable.

The usefulness of cellulose as a starting material for edible and biodegradable films can be extended by chemical modification to methyl cellulose (MC), hydroxypropylmethyl cellulose (HPMC), hydroxypropyl cellulose (HPC), and carboxymethyl cellulose (CMC). These *cellulose ether* films cast from aqueous or aqueous-ethanol solutions of MC, HPMC, HPC, and CMC tend to have moderate strength; are resistant to oils and fats; and are flexible, transparent,

Table 1 — Properties of Biodegrad	able and	Edible	Films
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MATERIAL	FILM PREPARATION	MOISTURE BARRIER <sup>a</sup>	OXYGEN BARRIER <sup>b</sup>	MECHANICAL PROPERTIES <sup>c</sup>	COST, \$/lb <sup>c</sup>
BIODEGRADABLE FIL	MS				
Cellophane	Aqueous suspension	Moderate	Good	Good	2.20 <sup>1</sup>
NC-W/Cellophane	NC-W coating	Good	Good	Good	2.40 <sup>f</sup>
Cellulose Acetate	Extrusion	Moderate	Poor	Moderate	1.60–2.10 <sup>g,h</sup>
Starch/PVOH	Extrusion	Poor <sup>e</sup>	Good <sup>e</sup>	Good	1.50-3.00 <sup>i,j</sup>
PHB/V	Extrusion	Good <sup>e</sup>	Good <sup>e</sup>	Moderate	3.00-6.00 <sup>l</sup>
PLA	Extrusion	Moderate <sup>e</sup>	Poor <sup>e</sup>	Good	1.00–5.00 <sup>i,j</sup>
EDIBLE FILMS		•			e Santa takan matakan kan santa mataki Mataki Mataki Mataki
MC	Aqueous-EtOH	Moderate	Moderate	Moderate	4.50-7.00
HPMC	Aqueous-EtOH	Moderate	Moderate	Moderate	4.75–7.00
HPMC:SA-PA/Wax	Aqueous-EtOH	Good	Moderate <sup>e</sup>	. NA	NA .
High Amylose Starch	Aqueous	Poor	Moderate	Moderate	0.60-0.70
Collagen	Aqueous suspension	Poor <sup>e</sup>	Good	Moderate	49.00–54.00 <sup>t</sup>
Gelatin	Aqueous	Poor <sup>e</sup>	Good <sup>e</sup>	NA	2.40-2.60
Zein	95% EtOH	Moderate	Moderate	Moderate	10.25-15.50
Gluten	Aqueous-EtOH	Moderate	Good	Moderate	0.80-0.90
SPI	Aqueous	Poor	Good	Moderate	1,30–1,70
Casein	Aqueous	Poor	Good <sup>e</sup>	NA	2.75–3.25
Casein-Beeswax	Emulsion	Moderate	Good <sup>e</sup>	NA	NA .
WPI	Aqueous	Poor	Good	Moderate	6.00-12.00
WPI-Beeswax	Emulsion	Moderate	Good	NA .	NA
Beeswax	Melt	Good	Poor	Poor	3.00-4.50
Shellac	EtOH	Moderate	Poor	Poor	3,50-6.00
est Conditions: ~38°C, 90	)/0%RH <sup>b</sup> Test Coi	nditions: ~25°C, 0-56	0%RH	CTest Conditions: ~25	
Poor = 10-100 g·mm/m <sup>2</sup> -0	<b>4</b> 1.0 <b>4</b>	100–1000 cm <sup>3</sup> μm/m		Moderate TS = 10-	
Moderate = 0.1-10 g·mm	· · · · · · · · · · · · · · · · · · ·	te = 10–100 cm <sup>3</sup> µm/		Moderate E = 10-5	
Good = 0.01–0.1 g·mm/m LDPE: 0.08 g·mm/m²·d·l		: 1–10 cm³ <i>µ</i> m/m²·d·k : 0.1 cm³ <i>µ</i> m/m²·d·kF		(LDPE: TS = 13 MF (OPP: TS = 165 M	

dFor Biodegradable Films: Compares to \$/lb resin (and finished film) costs for LDPE: \$0.50 (\$1.00); PS: \$0.55 (\$2.00); PET: \$0.75 (\$3.00); For Edible Films: Material cost range from suppliers

<sup>e</sup>Estimate.

fFinished film cost from supplier.

<sup>9</sup>Beach et al., 1994.

hFinished film cost from supplier is \$4.00/lb.

Beach and Price, 1993.

Ahmed et al., 1995.

E = Elongation

EtOH = Ethanol

EVOH = Ehylene vinyl alcohol copolymer

HPMC = Hydroxypropyl methyl cellulose

LDPE = Low density polyethylene

MC = Methyl cellulose

NC-W = Nitrocellulose-wax

OPP = Oriented polypropylene

PA = Palmitic acid

PET = Polyethylene terephthalate

PHB/V = Poly(3-hydroxybutyrate)-co-(3-

hydroxyvalerate)

PLA = Poly(lactic acid)

PS = Poylstyrene

PVOH = Poly(vinyl alcohol)

SA = Stearic acid

SPI = Soy protein isolate

TS = Tensile strength

WPI = Whey protein isolate

NA = Not applicable

Table 2 — Biodegradable Film Applications

MATERIAL	POSSIBLE APPLICA	TIONS <sup>a</sup>	
Cellophane	Decorative wraps, tapes		
NC-W/Cellophane	Baked goods, fresh proc	duce, processed meats, cheese and candy v	vraps
Cellulose Acetate	Baked goods and fresh	produce wraps, labels	· · · · · · · · · · · · · · · · · · ·
HPMC <sup>b</sup>	Dry food ingredient pour	ches	
Starch/PVOH	Compostable trash bags mulches, medical produc	s, disposable food-service items, paper coati cts	ings, loose-fill packaging, agricultural
Collagen <sup>b</sup>	Processed meat casings	and wraps	
PHB/V	Bottles, films, paper coa	itings	-
PLA	Compostable trash bags	, disposable food-service items, paper coati	ings, agricultural mulches, medical products
<sup>a</sup> Applications not l <sup>b</sup> Also edible	limited to those shown	NC-W = Nitrocellulose-wax HPMC = Hydroxyproyl methyl cellulose PVOH = Poly(vinyl alcohol)	PHB/V = Poly(3-hydroxybutyrate-co- (3-hydroxyvalerate) PLA = Poly(lactic acid)

odorless, tasteless, water-soluble, moderate barriers to moisture and oxygen (Anonymous, 1990; Hagenmaier and Shaw, 1990; Hanlon, 1992; Kester and Fennema, 1986; Nisperos-Carriedo, 1994; Table 1). MC is the least hydrophilic of the cellulose ethers, but MC film is still not a good moisture barrier. MC films, however, provide an excellent barrier against migration of fats and oils (Nelson and Fennema, 1991). HPC is the only edible and biodegradable cellulose-derived polymer that is thermoplastic and, therefore, capable of injection molding and extrusion.

A number of groups have investigated composite films composed of MC or HPMC and various kinds of solid lipids, such as beeswax and fatty acids (Debeaufort et al., 1993; Greener and Fennema, 1989; Kamper and Fennema, 1984a,b; Kester and Fennema, 1989a; Koelsch and Labuza, 1992; Martin-Polo et al., 1992; Park et al., 1994a). Many of these have water vapor permeabilities as low as LDPE. These composite films were all polymer-lipid bilayers formed either in one step from aqueous ethanolic solutions of cellulose ether with fatty acids, or in two steps by laminating pre-formed films with wax.

Pre-cast bilayer films, consisting of a layer of solid lipid (stearic acid [SA] and/or palmitic acid [PA] and/or beeswax) and a layer of HPMC and/or MC, substantially slowed the migration of water in several model food systems (Kamper and Fennema, 1985; Kester and Fennema, 1989b; Rico-Pena and Torres, 1990). Edible coatings, which include MC, HPMC, HPC, and CMC, have been applied to a variety of foods to provide a moisture, oxygen, or oil barrier and to improve batter adhesion (Table 3). MC and HPMC are also used in pharmaceutical tablet coating and to make edible pouches for dry food ingredients.

Cellulose can also be chemically modified to yet another ether, *ethyl cellulose* (EC), which is biodegradable but not edible. EC films can either be cast from non-aqueous solutions or extruded. Like the other cellulose ethers, EC films are poor moisture barriers, but they have been reported to be good oil and fat barriers (Hanlon, 1992).

Polysaccharide, Starch-based. Starches can be used to form edible or biodegradable films. Amylose (Rankin et al., 1958; Wolff et al., 1951), high amylose starch (Mark et al., 1966) and hydroxypropylated high amylose starch (Roth and Mehltretter, 1967) have been used to form selfsupporting films by casting from aqueous solution. These films appear to have moderate oxygen barrier properties but are poor moisture barriers, and their mechanical properties are generally inferior to synthetic polymer films. Starch exhibits thermoplastic behavior when a plasticizer such as water is added (Willettetal., 1994). Amylose and hydroxypropylated high amylose starch can also be extruded to form films (Jokay et al., 1967). The influence of moisture, however, on the stability of starch films limits their usefulness.

Amylose, hydroxypropylated starch, and dextrin films have been formed as edible coatings on foods to provide an oxygen or lipid barrier, and to improve appearance, texture and handling.

Starch composites, such as starch-polyethylene (starch-PE) mixtures, can be processed via extrusion, injection molding, or film blowing for the production of films or bottles (Röper and Koch, 1990). The starch component of these films can be metabolized by certain amylolytic bacteria. However, the PE component does not biodegrade; rather, it disintegrates. When packaging foods with such a starch-synthetic polymer composite, safety concerns arise related to premature biodegradation of the starch component and subsequent migration into the food of plasticizers, processing aids, stabilizers, and other additives associated with the synthetic polymer (Koelsch and Labuza, 1991).

Starch and polyvinyl alcohol (PVOH), a biodegradable synthetic polymer offering a broad range of properties, have been blended to yield thermoplastic materials with properties superior to starch alone.

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Some starch-PVOH blends appear to have potential for replacing LDPE films in applications where mechanical properties are critical for intended use and good moisture barrier properties are not necessary; other starch-PVOH blends are being explored for replacement of PS in disposable food-service items. Such single-use, quickly-used food applications avoid the safety concerns related to longer-term food packaging applications. Several companies are attempting to commercialize extrudable biodegradable blends containing high percentages of starch (Ahmed et al., 1995).

Other Polysaccharide Materials. *Alginate* films are formed by evaporation of an aqueous algi-

nate solution followed by ionic crosslinking with a calcium salt. They are impervious to oils and fats but are poor moisture barriers (Cottrell and Kovacs, 1980). Despite this, alginate gel coatings can significantly reduce moisture loss from foods by acting sacrificially. In other words, moisture is lost from the coating before the food significantly dehydrates. Alginate coatings are good oxygen barriers (Conca and Yang, 1993), can retard lipid oxidation in foods (Kester and Fennema, 1986), and can improve flavor, texture, and batter adhesion.

Coatings that include *carrageenan* as a major or sole component have been applied to a variety of foods to carry antimicrobials and to reduce moisture

Table 2 -	Edible	Polycacobarido	Coating	<b>Applications and Functions</b>	
Table 3 —	Ealble	Polysaccharige	Coating	Applications and runctions	,

MATERIAL AND APPLICATION	FUNCTION OF COATING A. Cellulose	REFERENCE
Methyl cellulose (MC)		, ne di Miliono de Territorio (1940). La <u>consta</u>
Pork and poultry pieces	Breading adhesion	Bauer et al., 1969
Glucose-oxidase-catalase enzyme preparation.	Inactivation protection during thermal processing	Scott, 1961
MC and hydroxypropyl methyl cellulose		
Potato products, onion rings	Oil barrier	Gold, 1969
Food pieces	Batter adhesion	Sanderson, 1981
MC and beeswax		
š	Moisture barrier	Greener and Fennema, 1989
Hydroxypropyl cellulose		•
	Moisture and oxygen barrier	Ganz, 1969
Candies	Moisture barrier	Krumel and Lindsay, 1976
Carboxymethyl cellulose		
Bananas	Oxygen and carbon dioxide barrier	Banks, 1984
Apples	Oxygen and carbon dioxide barrier	Banks, 1985; Drake et al., 1987; Santerre et al., 1989; Smith and Stow, 1984
Fresh fruits and vegetables	Oxygen and carbon dioxide barrier	Lowings and Cutts, 1982
Hard cheese, hard sausage	Mold suppression	Lück, 1968
Freshly-cut celery	Moisture barrier	Mason, 1969
Pears	Oxygen and carbon dioxide barrier	Meheriuk and Lau, 1988
Tomatoes	Oxygen and carbon dioxide barrier	Nisperos and Baldwin, 1988
Oranges	Oxygen and carbon dioxide barrier	Nisperos-Carriedo et al., 1990
Amylose	B. Starch	
Dried raisins	Clumping and sticking prevention	Moore and Robinson, 1968
Potato chips, french fried potatoes, specialty potato products	Oil barrier	Murray et al., 1971
Hydroxypropylated starch		
Confectionery product	Oil barrier	Brake and Fennema, 1993
	Oxygen barrier	
Jellied candies, caramels	Stickiness prevention	Jokay et al., 1967
Dextrins (starch hydrolysates)		,
	Moisture barrier	
Freshly sliced apples	Oxygen barrier	Murray and Luft, 1973
	T	able 3. Continued on page 67

loss, oxidation, or disintegration.

Low-methoxyl *pectin*, derived by controlled de-esterification, forms gels in the presence of calcium ions and can be used to develop edible films (Maclay and Owens, 1948; Schultz et al., 1948). The WVP of pectin films is quite high, the same order of magnitude as that for plain cellophane and other carbohydrate films. The WVP can be reduced signifi-

cantly by adding a wax coating onto the pectin film (Schultzet al., 1949). Although pectinate coatings are poor moisture barriers, they can retard water loss from food by acting as a sacrificing agent. Pectin coatings have been investigated for their ability to retard moisture loss and lipid migration and improve handling and appearance of foods.

Chitosan is produced commercially by de-acety-

Table 3 — continued from page 66. Edible Polysaccharide Coating Applications and Functions

MATERIAL AND APPLICATION	FUNCTION OF COATING	REFERENCE
The second of th	C. Seaweed Extracts	
	Texture improvement and moisture barrier	Allen et al., 1963a,b
Fish	Moisture barrier	Cottrell and Kovacs, 1980; Dziezak, 1991
Breaded foods, filled dough products	Moisture, lipid, oxygen barrier	Earle and McKee, 1985
the state of the s	Flavor, color, texture retention; breading adhesion	
	Batter adhesion	-
Ice cream	Drippings elimination	-
Lamb carcasses	Microbial growth reduction and chill rate increase	
Precooked ground pork patties	Oxygen barrier	
Beef pieces, steak	Moisture and oxygen barrier	
Carrageenan	Working and oxygon barrier	Timento of any 1070
	Moisture barrier	Bryan, 1972
	Mechanical disintegration protection, moisture barrier	=
	Shelf-life extension in cold storage	-
		-
	Surface microbial growth reduction	
	D. Pectinates Oil barrier	
Foods	Fungicide, vitamin, antioxidant carrier	Maclay and Owens, 1948
Cheese, dried sausages, ham, fish, frozen foods	Moisture barrier	Maclay and Owens, 1948; Schultz et al., 1948
Candied fruits	Stickiness reduction	Maclay and Owens, 1948; Swenson et al., 1953
Dates	Stickiness reduction and appearance improvement .	Swenson et al., 1953
Almonds		
	E. Chitin/Chitosan	
Apples, pears, peaches, plums	Oxygen and carbon dioxide barrier	Davies et al., 1989; Elson and Hayes, 1985
Fresh strawberries	Postharvest decay control	•
	Postharvest decay control	· · · · · · · · · · · · · · · · · · ·
		the state of the s
	F. Microbial Polysaccharides	
Pullulan  Peanuts, cashew nuts, instant noodles, fried confectioneries, dried sardines, dried meat products	Oxygen barrier	Yuen, 1974
•	Mechanical damage reduction, oxygen barrier	Yuen, 1974
Levan	. , ,	
·	Controlled release in the body	Kanlan et al. 1993

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lating chitin obtained from shellfish waste. It is biodegradable but has not yet been approved as a food ingredient in the U.S. Chitosan films that are clear, tough, and flexible and good oxygen barriers can be formed by casting from aqueous solution (Kaplan et al., 1993; Sandford, 1989). Chitosan-based coatings can protect foods from fungal decay and modify the atmospheres of fresh fruits.

Pullulan, levan, and elsinan are extracellular microbial polysaccharides that are edible and biodegradable. Pullulan films cast from aqueous solution are clear, odorless, and tasteless and good oxygen barriers (Conca and Yang, 1993; Yuen, 1974). Pullulan coatings have been used successfully as oxygen barriers to prolong food shelf life. Levan and elsinan also can be used as edible coating materials for foods and pharmaceuticals due to their low oxygen permeability (Kaplan et al., 1993).

**Protein-based.** *Collagen* is a fibrous, structural protein in animal tissue that can be converted into edible and biodegradable films. Because collagen is not thermoplastic, collagen film must be made by extruding a viscous colloidal acidic dispersion into a neutralizing bath followed by washing and drying.

Collagen film is not as strong and tough as cellophane but has reasonably good mechanical properties (Hood, 1987). Although no data on the WVP of collagen are available, its composition suggests that it is not a good moisture barrier. Collagen film is an excellent oxygen barrier at 0% RH, but oxygen permeability (OP) increases rapidly with increasing RH in a manner similar to cellophane (Lieberman and Gilbert, 1973).

Collagen is the most commercially-successful edible protein film. Collagen casings have largely replaced natural gut casings for sausages. Except for large sausages requiring thick casings, collagen casings are eaten with the sausage. Flat collagen films on smoked meats, such as hams, prevent the outer elastic netting from becoming imbedded in the meat during cooking. The collagen film is eaten with the meat product after removal of the netting. In addition to providing mechanical integrity to meat products, collagen film functions as an oxygen and moisture barrier (Baker et al., 1994).

Additional applications of collagen film have been explored. Collagen film overwrap on refrigerated and thawed beef round steak, packaged in high barrier bags or in PS foam trays with polyvinylchloride

Table 4 —	Edible !	Protein	Coating	<b>Applications</b>	and	Functions
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MATERIAL AND APPLICATION	FUNCTION OF COATING	REFERENCE
	A. Gelatin	
	Antimicrobial carrier	
Meat products	Mold prevention, oxygen barrier, handling abuse protection.	. Keil, 1961; Keil et al., 1960
Cut-up frozen turkey meat	Oxygen barrier and antioxidant carrier	. Klose et al., 1952
Smoked chicken meat	Moisture barrier	. Moorjani et al., 1978
Battered and breaded meats	Frying oil barrier	. Olson and Zoss, 1985
ogurt	Fruit separation	. Shifrin, 1968
Meat cuts	Oxygen and moisture barrier	. Whitman and Rosenthal, 1971
Zein	B. Corn Zein	
Eggs	Moisture and bacteria barrier; shell strength increase	. Meyer and Spencer, 1973
Rice	Vitamin adhesion	. Mickus, 1955
	Moisture and oxygen barrier	
Popcorn	Popping behavior	. Wu and Schwartzberg, 1992
Zein-acetylated monoglyceride		
Confectioneries	Oxygen, lipid, moisture barrier; antioxidant carrier; and stickiness prevention	. Alikonis, 1979; Cosler, 1957, 1959
Almonds, peanuts, pecans, walnuts	Oxygen, lipid, moisture barrier; antioxidant carrier	. Alikonis and Cosler, 1961; Cosler, 1958a,b,c
Intermediate-moisture food	Preservative carrier	. Torres et al., 1985a,b
Zein-vegetable oils		-
Nuts, confectioneries,	Oxygen and moisture barrier, antioxidant carrier	Andres, 1984
Zein/vegetable wax-oil laminate		
Dried fruit	Stickiness prevention; antioxidant carrier	Gunnerson and Bruno, 1990
	C. Wheat Gluten	Noznick and Rundus 1967
1400	· -	ible 4. Continued on page 6

(PVC) film, reduced exudation without significantly affecting color or lipid oxidation (Farouk et al., 1990). Another study found that collagen film performed as well as plastic film in maintaining the quality of frozen beef cubes (Conca, 1994; Rice, 1994). Collagen film, unlike synthetic polymer film, melts away as the meat thaws and cooks, eliminating need for plastic film waste handling.

Gelatin is obtained by hydrolytic cleavage of collagen chains. Edible coatings with gelatin reduce oxygen, moisture, and oil migration or can carry an antioxidant or antimicrobial (Table 4). Gelatin can also encapsulate low-moisture or oil-phase food ingredients and pharmaceuticals. Encapsulation protects against oxygen and light and defines ingredient amounts or drug dosages (Reineccius, 1994).

Other proteins that have been studied for film formation include *corn zein* (CZ), *wheat gluten* (WG), *soy protein isolate* (SPI), *whey protein isolate* (WPI), and *casein* (CS). Edible and biodegradable films from these proteins are generally obtained from ethanolic solution (CZ and WG) or aqueous solution (CZ latex, SPI, WPI, and CS). These films have mechanical properties similar to

MATERIAL AND

collagen film and inferior to cellophane (Brandenburg et al., 1993; Butler and Vergano, 1994; Gennadios et al., 1990; McHugh and Krochta, 1994a; Stuchell and Krochta, 1994). The poor mechanical properties of these protein films likely limit biodegradable packaging film applications, but paper coatings or edible food coatings are possibilities (Gennadios et al., 1994; Trezza and Vergano, 1994). These protein films have fairly large WVPs (Avena-Bustillos and Krochta, 1993; Gontard et al., 1992; McHugh et al., 1994; Park and Chinnan, 1995; Stuchell and Krochta. 1994). Their WVPs can be lowered by including wax or other lipid materials in the formulation (Avena-Bustillos and Krochta, 1993; Gontard et al., 1994; McHugh and Krochta, 1994b; McHugh and Krochta, 1994c). Such protein composite films, however, have not attained the low WVP of cellulose-ether-based composites. The OPs of films made from CZ, WG, SPI and WPI are quite low at 0-50% RH (Butler and Vergano, 1994; Gennadios et al., 1993; Li et al., 1993; McHugh and Krochta, 1994a). As with collagen film, RH has a large effect on OP for films from these materials. Little work has been done on the aroma-barrier properties of biodegradable and edible

Table 4 — contined from page 68. Edible Protein Coating Applications and Functions

APPLICATION	FUNCTION OF COATING	REFERENCE
Casein-acetylated monoglyceride	D. Casein	
ZucchiniApples and celery sticks Frozen fish Casein-stearic acid, beeswax, or acetylate	Moisture barrier	Avena-Bustillos et al., 1997
Peeled*carrots	Moisture retention	Avena-Bustillos et al., 1993, 1994b
Whey protein	E. Whey Protein	
Freeze-dried chicken dice	Mechanical disintegration protection	Alcantara et al., 1997
Peanuts	Oxygen barrier	Maté and Krochta, 1997; Maté et al., 199
Frozen salmon	Antioxidant carrier	Stuchell and Krochta, 1995
Whey protein, whey protein/acetylated mo		
Breakfast cereal, raisins, diced cheese, peas	s . Moisture barrier, stickiness prevention .	Chén, 1995
	F. Additional Materials	
Chicken parts	Breading adhesion	Suderman et al., 1981
Albumen, soy protein		
Raisins	Moisture barrier	Bolin, 1976; Watters and Brekke, 1961
Albumen, soy protein, wheat gluten		
Battered meats, etc	Batter adhesion	Baker et al., 1972
Casein, gelatin, soy protein, or zein/fatty a	icid amylose ester	
Dried fruits and vegetables	Moisture and oxygen barrier	Cole, 1969
Albumen, casein, gelatin, zein		
Nuts	Color carrier	Johnson, 1969
Albumen, casein, gelatin, soy protein-vege	etable oil	•
Polyod and fried goods shapelets	Oxygen and moisture barrier	D1 4007



films. The aroma permeability of WG film, however, was found to be only one-tenth that of LDPE (Debeaufort and Voilley, 1994).

Besides collagen and gelatin, corn zein is the only other protein that has been promoted commercially as an edible film or coating. The barrier, vitamin adhesion, and antimicrobial carrier properties of zein film coatings have been used on a variety of foods. Zein is also used on pharmaceuticals for coating capsules for protection, controlling release, and masking flavors and aromas (Gennadios et al., 1994). Zeincoated paper was judged equal to PE-laminated paper for quick-service restaurant packaging of fatty foods and was found to have good heat-sealing characteristics (Trezza and Vergano, 1994, 1995). Zein has also been explored as a plant-based replacement for animal-derived collagen in the manufacture of sausage casings (Turbak, 1972) and for the production of water-soluble pouches for dried foods (Georgevits,

Wheat gluten has been explored as a plant-based replacement for collagen in the manufacture of sausage casings (Mullen, 1971; Schilling and Burchill, 1972; Turbak, 1972) and as a means to improve the adherence of salt and flavorings to nuts and of batters to meats and other foods.

Soy protein has also been studied for the manufacture of sausage casings (Turbak, 1972) and in the production of water-soluble pouches (Georgevits, 1967). Soy protein in edible coating applications can improve batter adhesion and reduce moisture migration in raisins and dried peas.

Casein has been investigated for use in the production of water-soluble pouches (Georgevits, 1967). Caseinate coatings retained sorbic acid on the surface of a model food system and on the surfaces of intermediate moisture fruits and, when combined with lipid, protected fresh vegetables, dried fruits and vegetables, and frozen fish from moisture migration and/or oxidation.

Whey protein coatings effectively carried antioxidants for frozen fish, significantly reduced oxygen uptake and rancidity in roasted peanuts, and reduced disintegration of freeze-dried food. Whey protein and whey protein-acetylated monoglyceride coatings also reduced moisture migration into breakfast cereal and reduced stickiness of raisins.

Microbial Polyesters. Polyhydroxyalkanoates (microbial polyesters) can be produced by nutrientlimited fermentation of sugar feedstock. By manipulation of the growth medium, a random copolymer containing both hydroxyvalerate (HV) and hydroxybutyrate (HB) is obtained. The resulting copolymer poly(3-hydroxybutyrate)-co-(3-hydroxyvalerate) (PHB/V) is thermoplastic and fully biodegradable (Timmins et al., 1993). By changing the ratio of HV to HB, the resulting copolymer can be made to resemble either polypropylene (PP; low HV) or PE (high HV), with regard to flexibility, tensile

strength, and melting point (Kemmish, 1993). Polyhydroxybutyrate(PHB) is strong, stiff, and brittle, but HV content improves flexibility and toughness (Holmes, 1988). PHB/V has good chemical and moisture resistance (Kemmish, 1993) and is reported to possess good oxygen, moisture, and aroma barrier properties (Anonymous, 1993b). Good moisture resistance and barrier properties are consistent with the fact that PHB/V is relatively more hydrophobic than polysaccharides and proteins commonly used in biodegradable and edible films. Furthermore, while the oxygen barrier may not be as good as that for polysaccharides and proteins at low RH, it should not be as sensitive to increasing RH.

Uses being considered for PHB/V include beverage bottles, coated paperboard milk cartons (Hocking and Marchessault, 1994), and films (Ahmed et al., 1995).

Polylactic Acid. Polylactic acid (PLA) is a thermoplastic, biodegradable polymer based on lactic acid produced by fermentation of simple sugars. PLA-based materials have performed well in commercial medical applications, such as bioabsorbable sutures and implants. PLA can be easily hydrolyzed back to lactic acid, using only water, and then can be repolymerized (Ahmed et al., 1995). This may provide some advantages in recycling PLA. Recent advances have produced more economical PLA polymers of sufficient molecular weight to possess other useful properties. PLA has mechanical properties similar to PS (Sinclair, 1994). Modification of molecular weight and crystallinity results in properties that can also mimic PE, PP, or PVC (Gruber, 1994). Several U.S. and Japanese firms are developing PLA-based polymers (Ahmed et al., 1995). Product targets include food service containers and utensils, grocery bags, and compostable waste bags.

### CONCLUSION

Edible Polymer Films. The hydrophilic nature of edible polymers limits their ability to provide desired edible film functions. For all edible polymers, RH, which greatly influences properties, must be taken into account when considering applications. Use of edible polymer films and coatings as moisture barriers usually requires the formation of composite films that contain hydrophobic materials such as edible fatty acids and waxes. Cellulose ether-based composite films with moisture barrier properties comparable to LDPE involve formation of polymer-lipid bilayers from aqueous ethanolic solutions and/or lamination with wax. These films and coatings have greater integrity than lipid or wax structures alone. Elimination of ethanol and the second application (lamination) step, however, would increase the usefulness of this concept. Additional research is needed to develop such composite films and coatings from cellulose ethers and other edible polymers without need for non-aqueous solvents and/or multiple steps.

Because edible polymers can hydrogen bond effectively, they make good oxygen, aroma, and lipid barrier films at low-to-intermediate RH. Additional data on the effect of RH are needed to identify the range of practical use. Nonetheless, the barrier deteriorates with increase in RH. Therefore, potential applications include: (1) protective pouches or coatings for low-moisture food products vulnerable to exidation or aroma loss in conjunction with a simple. moisture-barrier packaging film bag (e.g., LDPE); (2) respiration-reducing coatings for fresh fruits and vegetables that are exposed to low RH during storage and transportation; and (3) lipid-barrier films or coatings separating lipid-rich ingredients from other components of heterogeneous foods. Edible polymers that are water-soluble and good emulsifiers may be favored in many food coating applications. Development of composite bilayer films will allow protection of edible film oxygen, aroma, and lipid barrier properties with a hydrophobic moisture barrier layer. Synthetic polymer films, e.g., EVOH copolymer, that are good oxygen and aroma barriers tend to be expensive. The additional cost incurred in coating or pouching foods with an edible film may be balanced by savings in eliminating such synthetic oxygen/ aroma barriers in multi-layer packages. Simplifying the package may also make it more recyclable, another advantage with economic consequences.

The mechanical properties of edible polymer films are generally inferior to synthetic films. The films, however, are adequately durable casings, wraps, or coatings on food products. They are also likely to adequately separate layers of heterogeneous foods or as small food pouches/bags.

Opportunities presented by the thermoplastic nature of certain edible polymers should be explored to enable production of edible films by extrusion. Potential materials include certain cellulose ethers, starch compounds, and certain proteins.

When considering possible edible film coating applications, attention must be given to the requirement that edible coating formulations must wet and spread on the food surface and upon drying form a film coating that has adequate adhesion, cohesion, and durability to function properly. These properties are influenced by both edible film formulation and methods of coating and drying. Lack of attention to these facts has probably resulted in inconsistent and unsatisfactory results in many studies. In addition, edible coatings must provide satisfactory appearance, aroma, flavor, and mouthfeel. Selective application to appropriate foods and good control of environmental conditions are necessary to ensure microbial stability. Addition of antimicrobials to edible films may widen application possibilities. Finally, any advantage of edible film coatings must be provided at an affordable cost. Unfortunately, little published research is available on food coating issues, sensory properties, microbial stability, or economics of edible films.

Biodegradable Polymer Films. For biodegradable packaging materials to compete with non-biodegradable synthetics, the critical mechanical, optical, and/or barrier properties for the intended application must be matched. This is especially difficult in the case of moisture barrier properties, because no biodegradable polymer approaches the hydrophobic character of synthetic polymers such as LDPE. Because of their inherent hydrophilic nature, biodegradable polymers are usually poor moisture barriers. They are, however, naturally good oxygen barriers at low RH, but oxygen permeabilities increase exponentially as RH increases.

Cellulose-based films have been used most extensively among biodegradable polymers. In spite of not being thermoplastic, cellophane has a significant presence commercially in food packaging, with use of coating and lamination to improve its properties. Coating with a nitrocellulose-wax (NC-W) blend provides a moisture barrier comparable to LDPE without sacrificing biodegradability, but at greater cost. Such coating allows short-term use in packaging of fresh bread, meat, and produce, and long-term packaging of low-moisture foods. Coating with the NC-W moisture barrier layer also helps retain oxygen barrier characteristics at medium to high RH.

Nonetheless, to develop economically viable biodegradable polymers for packaging, developers should use conventional synthetic polymer packaging conversion technology. For many uses, a combination of starch and PVOH can form thermoplastic blends that can be extruded to form films with mechanical properties similar to LDPE. Different starch-PVOH blends can be used to mold semi-rigid products currently produced from PS. PLA is a versatile thermoplastic material with properties that can also be modified to resemble LDPE or PS., PHB/V copolymer is a thermoplastic material that has adjustable properties dependent on the monomer ratio. PHB/V appears to have greater moisture resistance and lower WVP than the other biodegradable polymers. These properties could allow production of beverage bottles, coated milk cartons, and other containers useful for high-moisture products where starch-PVOH or PLA could not be used.

Several companies have commercialized starch-PVOH, PLA, and PHB/V polymers. None of the resulting products has obtained FDA approval for use in long-term food packaging (Beach and Ahmed, 1993a,b). PLA, however, has been self-determined GRAS by a major manufacturer (Thorsheim, 1996). Nonetheless, the key markets for biodegradable polymers for the nearer term include: nonfood packaging; personal and health care items; refuse (composting) and retail bags; loosefill packaging peanuts; medical gloves, gowns and masks; agricultural mulches and potting containers; and fast-food containers, cups, plates, and cutlery. Short-term use of these biode-

gradable polymer products in fast-food service applications avoids concern over premature biodegradation with longer-term packaging of high-water-activity foods. With all biodegradable polymers, the biodegradation characteristics must be taken into account when considering packaging applications, package storage conditions, and potential for insect infestation and microbial food safety problems.

Other issues related to biodegradable polymers must be considered. Easy, efficient sorting of biodegradable from non-biodegradable packaging would be essential to avoid contamination of recycling and biodegradation efforts (Evans and Sikdar, 1990). Secondly, designing a package to biodegrade precludes its being used as a resource for other products (Van Volkenburgh and White, 1993). Thirdly, biodegradation is slow and produces significant methane, with greenhouse-effect implications. In comparison, properly-designed incineration is fast and efficient and can capture important energy from natural (renewable/biodegradable) and synthetic polymers alike (Rowatt, 1993). Finally, biodegradable polymers are usually considerably more expensive than conventional synthetic polymers. Improvements in production practices, economies of scale, and increasing costs for fossil resources could all be necessary to produce a more favorable economic situation for biodegradable polymers.

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