

This report is a result of the combined effort of the project partners of the Food Biopack Project funded by the EU Directorate 12.



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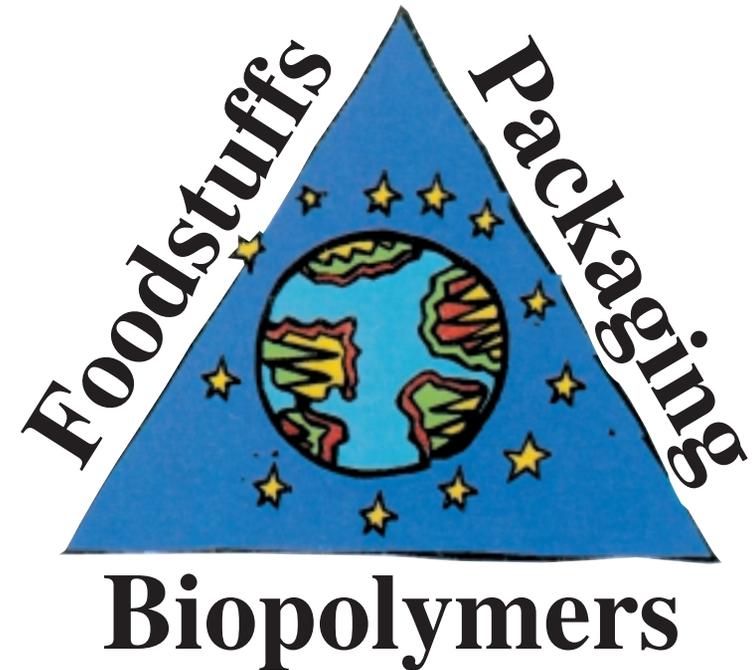
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Biobased Packaging Materials for the Food Industry



Biobased Packaging Materials *for the Food Industry*

STATUS AND PERSPECTIVES

Edited by Claus J Weber

– A European Concerted Action –

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Preface

At the turn of the last century most non-fuel industrial products; dyes, inks, paint, medicines, chemicals, clothing, synthetic fibres and plastics were made from biobased resources. By the 1970s petroleum-derived materials, had to a large extent, replaced those materials derived from natural resources. Recent developments are raising the prospects that naturally derived resources again will be a major contributor to the production of industrial products. Currently, scientists and engineers successfully perform developments and technologies that will bring down costs and optimize performance of biobased products. At the same time environmental concerns are intensifying the interest in agricultural and forestry resources as alternative feedstocks. A sustained growth of this industry will depend on the development of new markets and costs and performance competitive biobased products. A potential new market for these materials is food packaging, a highly competitive area with great demands for performance and cost.

The aim of this EU-concerted action project, "Production and application of biobased packaging materials for the food industry", is to evaluate the potential of biobased materials as food packaging. The mission of the report is to present the state of the art of biobased food packaging, and furthermore to outline the future scenarios and developments. In order to cover the whole area, project partners represent the whole production chain, from producers of biobased resins to converters, and food packaging users together with food scientists and polymer chemists.

The report consists of eight chapters and an executive summary, which altogether aim at covering all aspects of biobased food packaging materials. Chapter 1 gives a general introduction to the background of the project as well as to the interest in biobased food packaging. The biobased polymers, materials and packaging are presented in Chapter 2 together with an introduction to their properties. Chapter 3 focuses on the potential food applications of biobased materials and furthermore outlines the specific packaging demands of a range of food products. The emphasis in Chapter 4 is on legislative demands for food contact packaging materials and further, if any, specific considerations to

Acknowledgements

A definition of biobased food packaging materials

be made when dealing with biobased food packaging. Compostability, legislative demands and the process of documentation in relation to compostable packaging are described in Chapter 5. Chapter 6 deals with the environmental impacts of using biobased materials. The market of biobased materials, and moreover the future of the same, are the objectives of Chapter 7, and finally in Chapter 8, a joined conclusion of the potential of biobased packaging for the food industry is outlined.

To produce a state-of-the-art report of biobased food packaging turned out to be quite a challenge, taken the rapid pace of developments seen in this area into consideration. The presented publication does only report the information being part of the public domain and information on industrial R&D developments are not included. The state-of-the-art is very likely already to have moved on when these lines are being read. However, the report may also be read as a general introduction to the challenge of using biobased materials for food packaging.

This report is a result of the EU concerted action project: Production and application of biobased packaging materials for the food industry (Food Biopack), funded by DG12 under the contract PL98 4046.

"Biobased food packaging materials are materials derived from renewable sources. These materials can be used for food applications"

Abbreviations

Al	Aluminium
APET	Amorphous Poly(ethylene terephthalate)
BRED	Biomass for Green House Gases emission RE-Duction, a European project
CEN	The European Committee of Standardization
ECN	Energy Research Foundation
EPS	Expandable Polystyrene
EVA	Ethyl Vinyl Acetate
EVOH	Ethyl Vinyl Alcohol
FDA	Food and Drug Administration (USA)
GHG	GreenHouse Gasses
GWP	Global Warming Potential
HDPE	High Density Polyethylene
LCA	Life Cycle Analyses
LDPE	Low Density Polyethylene
LFP	Loose-Fill-Packaging
LLDPE	Linear Low Density Polyethylene
MAP	Modified Atmosphere Packaging
MDPE	Medium Density Polyethylene
OPP	Oriented Polypropylene
PA	Polyamide
PC	Polycarbonate
PE	Polyethylene
PET	PolyEthylene Terephthalate
PETG	Copolymer of PET and cyclohexane-dimethanol
PHAs	Poly(hydroxyalkanoates)
PHB	Poly Hydroxy Butyrate
PHB/V	Poly Hydroxy Butyrate/Valerate
PLA	Polylactic acid
PP	Polypropylene
PS	Polystyrene
PVC	Poly Vinyl Chloride
PVdC	Poly Vinylidene Chloride
RCF	Regenerated Cellulose film
RH	Relative Humidity
SCF	Scientific Committee on Food
SiOx	Silicium Oxide
Tg	Glass Temperature
TiO2	Titanium Oxide
Tm	Melting Temperature
UHT	Ultra High Temperature
WOF	Warmed-Over Flavour

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1. Introduction

The issue of sustainability has been high on the EU agenda for a number of years, encouraging academia and industry to develop sustainable alternatives thus aiming to preserve resources for future generations. At the same time, these sustainable alternatives address other key EU issues such as the use of surplus stocks in Europe and the production of higher added value agricultural products thereby promoting economic development in the European agricultural sector. The successful promotion and use of biological, renewable materials for the production of packaging materials will satisfy a number of the key EU objectives. To date, packaging materials have been, to a large extent, based on non-renewable materials. The only widely used renewable packaging materials are paper and board which are based on cellulose, the most abundant renewable polymer world-wide. However, major efforts are under way to identify alternative non-food uses of agricultural crops and the production of packaging materials, based on polymers from agricultural sources, could become a major use of such crops (Coombs and Hall, 2000; Mangan, C 1998). Indeed, such alternative biobased packaging materials have attracted considerable research and development interest for a significant length of time (Coombs and Hall, 2000; Mangan, C 1998) and in recent years the materials are reaching the market (see Chapter 7). The biological basis of the starting materials provides the material engineer with a unique opportunity to incorporate a very appealing functionality into the material, that of compostability. This property enables these new materials to degrade upon completion of useful life. Compostability has, so far, been the main focus for applications of biobased packaging materials which is the logical consequence for the vast amount of packaging materials used and the waste associated with it. Municipal plastic waste is difficult to deal with as it consists of a number of fractions of waste and several plastic types and it contains plastic types with a high degree of contamination from foodstuffs resulting in labour and energy intensive recycling. To date, prevention or enhanced recovery of materials has been used to extend the lifetime of the available non-renewable materials. Recovery methodology includes recycling, reuse, energy recovery, composting and biomethanisation. Re-use and re-cycling of food packaging materials is problematic, as they often comprise mixtures of

layers of different plastics to achieve optimal barrier properties of the material. Furthermore, caution must be exercised when re-using food contact materials, as there might be an unwanted build-up of contaminants from food components migrated into the packaging materials after several re-uses. Organic recovery by composting or biomethanisation offers an alternative waste disposal route, in which both left-over foodstuffs and the food packaging are disposed of in the same process. The bottleneck in using organic recovery is the development of biobased compostable packaging with the required properties for protection of food during storage and furthermore, a waste infrastructure for these compostable packages along with labelling to identify the compostable packaging must also be developed. So far, the potential compostability of these materials has been the central point of interest for commercialization although composting in many countries is not the common way of disposal. However, as the performance of the biobased materials progressively is being improved, new and more advanced applications, such as food packaging, are now becoming within reach.

The materials used for food packaging today consist of a variety of petroleum-derived plastic polymers, metals, glass, paper and board, or combinations thereof. These materials and polymers are used in various combinations to prepare materials with unique properties which efficiently ensure safety and quality of food products from processing and manufacturing through handling and storage and, finally, to consumer use. Notably, these materials fulfil a very important task as absence of packaging or insufficient packaging would result in fast deterioration of quality and safety giving way to massive commercial losses of valuable foodstuffs. Individual food products have specific optimum requirements for storage that the packaging materials must be able to provide. When contemplating the concept of food packaging, the entire dynamic interaction between food, packaging material and ambient atmosphere has to be considered. Hence, engineering of new biobased food packaging materials is a tremendous challenge both to academia and industry.

The biobased materials are interesting from a sustainable point of view. The question is whether they meet the standards of the materials used today or whether they even add value. This report summarizes the state-of-the-art of biobased food packa-

ging materials and provides scenarios for future use of biobased packaging materials in the food industry. The report is a result of the concerted action project "Production and application of biobased packaging materials for the food industry" sponsored by the EU Commission.

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2. Properties of biobased packaging materials

2.1. Introduction

Designing and manufacturing of packaging materials is a multi-step process and involves careful and numerous considerations to successfully engineer the final package with all the required properties. The properties to be considered in relation to food distribution are manifold and may include gas and water vapour permeability, mechanical properties, sealing capability, thermoforming properties, resistance (towards water, grease, acid, UV light, etc.), machinability (on the packaging line), transparency, anti fogging capacity, printability, availability and, of course, costs. Moreover, a consideration of the "cradle to grave" cycle of the packaging material is also required, hence, the process of disposal of the package at the end of its useful life must also be taken into consideration.

The aim of this report is to evaluate the potential of biobased packaging materials for the food industry, and the most important properties in relation to food applications can be narrowed down to four intrinsic properties of the material: mechanical, thermal, gas barrier and water vapour properties, and the focus of this chapter will be on these four properties.

Compostability, which is a very appealing property when the packaging meet its end of useful life, will also be described. For a detailed discussion of biodegradability/compostability and waste handling, please refer to Chapter 5 and issues of availability and costs are discussed in Chapter 7. Packaging of food and interaction between foods and packaging materials will be dealt with in Chapters 3 and 4, respectively.

The most common biobased polymers and potential biobased packaging materials are presented, followed by a discussion of their food packaging properties, and finally, procedures for processing biobased materials into food product packaging will be discussed.

2.2. Food biobased materials – a definition

As previously described, we have chosen a definition of biobased

food packaging materials based on their origin and use, leading to the following definition:

“Biobased food packaging materials are materials derived from renewable sources. These materials can be used for food applications”.

In addition, packaging materials recognized as biodegradable according to the standards outlined by the EU Standardization Committee are also included in the project. This amendment was included not to exclude materials which currently, of practical and economical reasons, are based on non-renewable resources, but at a later stage these materials may be produced based on renewable resources.

2.3. Origin and description of biobased polymers

Biobased polymers may be divided into three main categories based on their origin and production:

Category 1 Polymers directly extracted/removed from biomass. Examples are polysaccharides such as starch and cellulose and proteins like casein and gluten.

Category 2 Polymers produced by classical chemical synthesis using renewable biobased monomers. A good example is polylactic acid, a biopolyester polymerised from lactic acid monomers. The monomers themselves may be produced via fermentation of carbohydrate feedstock.

Category 3 Polymers produced by microorganisms or genetically modified bacteria. To date, this group of biobased polymers consists mainly of the polyhydroxyalkanoates, but developments with bacterial cellulose are in progress.

The three categories are presented in schematic form in Figure 2.1.

Updated and detailed description of the polymers presented in Figure 2.1 may be found in numerous excellent review papers and books published recently (Petersen et al., 1999; Chandra and Rustgi, 1998; Witt; et al., 1997; Guilbert et al., 1996; Krochta and Mulder-Johnston, 1996) and it is not the purpose of this report to repeat the work done so well by the previous authors. In general, compared to conventional plastics derived from mine-

ral oil, biobased polymers have more diverse chemistry and architecture of the side chains giving the material scientist unique possibilities to tailor the properties of the final package. The most common biobased polymers, materials and packaging will be presented in the following.

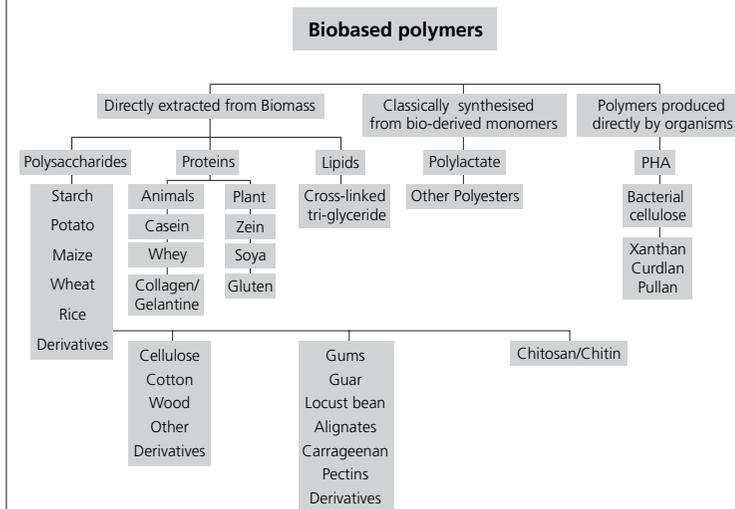


Figure 2.1 Schematic presentation of biobased polymers based on their origin and method of production.

2.3.1. Category 1: Polymers directly extracted from biomass

The natural Category 1 polymers, most commonly available, are extracted from marine and agricultural animals and plants. Examples are polysaccharides such as cellulose, starch, and chitin and proteins such as casein, whey, collagen and soy. All these polymers are, by nature, hydrophilic and somewhat crystalline – factors causing processing and performance problems, especially in relation to packaging of moist products. On the other hand, these polymers make materials with excellent gas barriers.

Polysaccharides

To date, the principal polysaccharides of interest for material production have been cellulose, starch, gums, and chitosan. Likely, the more complex polysaccharides produced by fungi and bacteria (Category 3 biobased polymers) such as xanthan, curdlan, pullan and hyaluronic acid, will receive more interest in the future.

Starch and derivatives

Starch, the storage polysaccharide of cereals, legumes and tubers, is a renewable and widely available raw material suitable for a variety of industrial uses. As a packaging material, starch alone does not form films with adequate mechanical properties (high percentage elongation, tensile and flexural strength) unless it is first treated by either plastization, blending with other materials, genetic or chemical modification or combinations of the above approaches. Corn is the primary source of starch, although considerable amounts of starch are produced from potato, wheat and rice starch in Europe, the Orient and the United States.

Starch is economically competitive with petroleum and has been used in several methods for preparing compostable plastics. However, a challenge to the development of starch materials is the brittle nature of blends with high concentrations of starch.

Overcoming the brittleness of starch while achieving full biodegradability in blends can be accomplished by the addition of biodegradable plasticizers. Common plasticizers for hydrophilic polymers, such as starch, are glycerol and other low-molecular-weight-polyhydroxy-compounds, polyethers and urea. Plasticizers lower the water activity thereby limiting microbial growth.

When starch is treated in an extruder by application of both thermal and mechanical energy, it is converted to a thermoplastic material. In the production of thermoplastic starches, plasticizers are expected to reduce the intermolecular hydrogen bonds effectively and to provide stability to product properties. Because of the hydrophilicity of the starch the performance of materials extruded with starch changes during and after processing as water contents changes. To overcome this challenge, many different starch derivatives have been synthesized; recently, site-selective modifications have been reported. Blending with more hydrophobic polymers produce formulations that are suitable for injection moulding and blowing films. Compatibility is an issue when these types of blends and laminates are used, and compatibilizers and other additives are used as processing aids.

Starch-based thermoplastic materials have been commercialized

during the last few years and are to day dominating the market of biobased, compostable materials (see chapter 7).

Cellulose and derivatives

Cellulose is the most abundantly occurring natural polymer on earth and is an almost linear polymer of anhydroglucose. Because of its regular structure and array of hydroxyl groups, it tends to form strongly hydrogen bonded crystalline microfibrils and fibres and is most familiar in the form of paper or cardboard in the packaging context. Waxed or polyethylene coated paper is used in some areas of primary food packaging, however the bulk of paper is used for secondary packaging. Cellulose is a cheap raw material, but difficult to use because of its hydrophilic nature, insolubility and crystalline structure. To make cellulose or cellophane film, cellulose is dissolved in an aggressive, toxic mixture of sodium hydroxide and carbon disulphide ("Xanthation") and then recast into sulphuric acid. The cellophane produced is very hydrophilic and, therefore, moisture sensitive, but it has good mechanical properties. It is, however, not thermoplastic owing to the fact that the theoretical melt temperature is above the degradation temperature, and therefore cannot be heat-sealed. Cellophane is often coated with nitrocellulose wax or PVdC (Poly Vinylidene Chloride) to improve barrier properties and in such form it is used for packaging of baked goods, processed meat, cheese and candies. However, there is considerable potential for the development of an improved cellulose film product or an improved production method as the existing product is problematic in both respects.

A number of cellulose derivatives are produced commercially, most commonly carboxy-methyl cellulose, methyl cellulose, ethyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose and cellulose acetate. Of these derivatives only cellulose acetate (CA) is widely used in food packaging (baked goods and fresh produce). CA possesses relatively low gas and moisture barrier properties and has to be plasticized for film production. Many cellulose derivatives possess excellent film-forming properties, but they are simply too expensive for bulk use. This is a direct consequence of the crystalline structure of cellulose making the initial steps of derivatization difficult and costly. Research is required to develop efficient processing technologies for the production of cellulose derivatives if this situation is to change.

Chitin/Chitosan

Chitin is a naturally occurring macromolecule present in the exoskeleton of invertebrates and represents the second most abundant polysaccharide resource after cellulose (Kittur et al., 1998). Chitin is chemically composed of repeating units of 1,4-linked 2-deoxy-2-acetoamido- α -D-glucose, and chitosan refers to a family of partially N-acetylated 2-deoxy-2-amino- α -glucan polymers derived from chitin. In general, chitosan has numerous uses: flocculant, clarifier, thickener, gas-selective membrane, plant disease resistance promoter, wound healing promoting agent and antimicrobial agent (Brine et al., 1991). Chitosan also readily forms films and, in general, produces materials with very high gas barrier, and it has been widely used for the production of edible coating (Krochta and Mulder-Johnston, 1997). Furthermore, chitosan may very likely be used as coatings for other biobased polymers lacking gas barrier properties. However, as with other polysaccharide-based polymers, care must be taken for moist conditions. The cationic properties of chitosan offer good opportunities to take advantage of electron interactions with numerous compounds during processing and incorporating specific properties into the material. The cationic property may further be used for incorporation and/or slow release of active components, adding to the possibilities for the manufacturer to tailor the properties (Hoagland and Parris, 1996). Another interesting property of chitosan and chitin in relation to food packaging are their antimicrobial properties (Dawson et al., 1998) and their ability to absorb heavy metal ions (Chandra and Rustgi, 1998). The former could be valuable in relation to the microbial shelf-life and safety of the food product and the latter could be used to diminish oxidation processes in the food catalyzed by free metals. So far, the major interest for chitosan as a packaging material has been in edible coatings. However, Makino and Hirata (1997) have shown that a biodegradable laminate consisting of chitosan-cellulose and polycaprolactone can be used in modified atmosphere packaging of fresh produce.

Proteins

Proteins can be divided into proteins from plant origin (e.g. gluten, soy, pea and potato) and proteins from animal origin (e.g. casein, whey, collagen, keratin). A protein is considered to be a random copolymer of amino acids and the side chains are highly suitable for chemical modification which is helpful to the mate-

rial engineer when tailoring the required properties of the packaging material.

For food packaging, edible coatings made of proteins are widely described in the literature (see Chapter 3), but thermoplastic processable polymers may also be made out of proteins (de Graaf and Kolster, 1998). Due to their excellent gas barrier properties, materials based on proteins are highly suitable for packaging purposes. However, like starch plastics mechanical and gas properties are influenced by the relative humidity due to their hydrophilic nature.

The major drawback of all protein-based plastics, apart from keratin, is their sensitivity towards relative humidity. Blending or lamination with other biobased materials may overcome this challenge with lower sensitivity towards humidity (see Section 2.5). So far, research in this field has been limited. Another way to modify protein properties is by chemical modification and, as seen in Figure 2.2, proteins contain a wide variety of chemical moieties which may help tailoring protein properties towards specific applications.

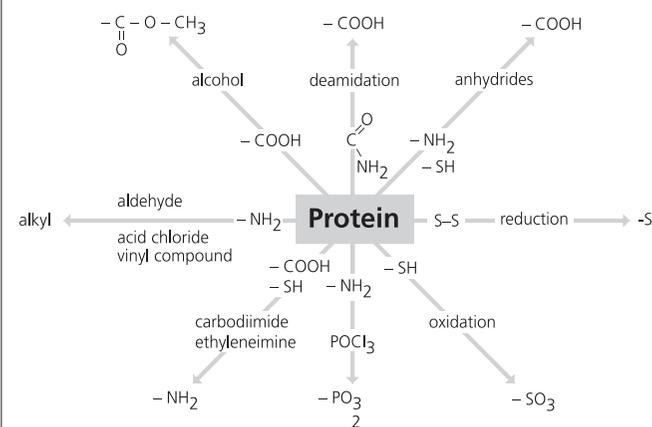


Figure 2.2 The numerous and diverse side chains of proteins offers the polymer scientist limitless opportunities to specifically tailor the properties of the final polymeric material by using chemical modification.

Casein

Casein is a milk-derived protein. It is easily processable due to its random coil structure. Upon processing with suitable plasticizers at temperatures of 80-100°C, materials can be made with mechanical performance varying from stiff and brittle to flexible and tough performance. Casein melts are highly stretchable making them suitable for film blowing. In general, casein films have an opaque appearance. Casein materials do not dissolve directly in water, but they show approx. 50% weight gain after 24 hours of immersion. The main drawback of casein is its relatively high price. Casein was used as a thermoset plastic for buttons in the 1940's and 50's. It is still used today for bottle labelling because of its excellent adhesive properties.

Gluten

Gluten is the main storage protein in wheat and corn. Wheat is an important cereal crop because of its ability to form a visco-elastic dough. Mechanical treatment of gluten leads to disulfide bridge formation formed by the amino acid cysteine which is relative abundant in gluten. The disulphide bridges are responsible for the creation of a strong, visco-elastic and voluminous dough. Processing is, therefore, more difficult than in the case of casein as the disulphide crosslinks of the gluten proteins have to be reduced with a proper reducing agent. Processing temperatures are, depending on the plasticizer contents, in the range of 70-100°C. Mechanical properties may vary in the same range as those for caseins. Gluten plastics exhibit high gloss (polypropylene like) and show good resistance to water under certain conditions. They do not dissolve in water, but they do absorb water during immersion. Due to its abundance and low price, research on the use of gluten in edible films, adhesives, or for thermoplastic applications is currently being carried out.

Soy protein

Soy proteins are commercially available as soy flour, soy concentrate and soy isolate, all differing in protein content. Soy protein consists of two major protein fractions referred to as the 7S (conglycinin, 35%) and 11S (glycinin, 52%) fraction. Both 7S and 11S contain cysteine residues leading to disulphide bridge formation and processing is, therefore, similar to gluten with similar mechanical properties. The best results are obtained with soy isolate (approx.90% protein) (Fossen and Mulder, 1998).

This behaviour in water is similar to that of gluten plastics.

Some patents from the beginning of the 1900 describe the use of soy protein as adhesives or plastics. Even the ancient Chinese used soy protein for non-food applications such as oil for lubrication. The most successful applications of soy proteins were the use in adhesives, inks and paper coatings.

Keratin

Keratin is by far the cheapest protein. It can be extracted from waste streams such as hair, nails and feathers. Due to its structure and a high content of cysteine groups, keratin is also the most difficult protein to process. After processing, a fully biodegradable, water-insoluble-plastic is obtained. However, mechanical properties are still poor compared to the proteins mentioned above.

The main drawback of all protein plastics, apart from keratin, is their sensitivity to relative humidity. Either blending or lamination can circumvent this problem. Research in this field has been limited until now.

Collagen

Collagen is a fibrous, structural protein in animal tissue, particularly skin, bones and tendons, with a common repeating unit: glycine, proline and hydroxyproline. Collagen is a flexible polymer. However, because of its complex helical and fibrous structure collagen is very insoluble and difficult to process. Collagen is the basic raw material for the production of gelatine, a common food additive with potential for film and foam production. Gelatine is produced via either partial acid or alkaline hydrolysis of collagen. Such treatments disrupt the tight, helical structure of collagen and produce water-soluble fragments that may form stiff gels, films, or light foams. Gelatine is a very processable material, but it is extremely moisture sensitive. Therefore, for prolonged use in packaging, research is needed for the chemical modification of gelatine to improve moisture sensitivity.

Whey

Whey proteins are by-products from the cheese production and are particularly rich in β -lactoglobulin. They have a relatively high nutritional value, are available in large amounts world-wide and

have been extensively investigated as edible coatings and films. This would seem to form the basis for a logical utilization strategy for this protein in packaging. Whey proteins are readily processable and have some potential as exterior films, if, as with gelatine, suitable modification strategies can be developed to reduce moisture sensitivity.

Zein

Zein comprises a group of alcohol soluble proteins (prolamines) found in corn endosperm. Commercial zein is a by-product of the corn wet-milling industry. Today, zein is mostly used in formulations of speciality food and pharmaceutical coatings. However, the potential supply of zein, estimated at 375,000 tons p.a. calls for expanded markets and drives research and development of novel value-added applications (Shukla, 1992). Film-forming properties of zein have been recognized for decades and are the basis for most commercial utilization of zein (Padua et al., 2000; Andres, 1984). Films may be formed by casting, drawing or extrusion techniques (Ha, 1999; Lai and Padua, 1997; Reiners et al., 1973). The films are brittle and needs plasticizers to make them flexible. Zein-based films show a great potential for uses in edible coatings and biobased packaging (Padua et al., 2000).

2.3.2. Category 2: Polymers produced from classical chemical synthesis from biobased monomers

Using classical chemical synthesis for the production of polymers gives a wide spectrum of possible "bio-polyesters". To date, polylactic acid is the Category 2 polymer with the highest potential for a commercial major scale production of renewable packaging materials. However, a wide range of other biopolyesters can be made. In theory, all the conventional packaging materials derived from mineral oil today can in the future be produced from renewable monomers gained by e.g. fermentation. Today, this approach is not economically feasible due to the cost of the production of the monomers. However, it is an obstacle that the PLA producers seem to have overcome with success (see Chapter 7).

Polylactic acid (PLA)

Lactic acid, the monomer of polylactic acid (PLA), may easily be produced by fermentation of carbohydrate feedstock. The carbohydrate feedstock may be agricultural products such as maize,

wheat or alternatively may consist of waste products from agriculture or the food industry, such as molasses, whey, green juice, etc. (Garde et al., 2000; Södergård, 2000). Recent results point out that a cost-effective production of PLA can be based on the use of green juice, a waste product from the production of animal feeds (Garde et al., 2000).

PLA is a polyester with a high potential for packaging applications. The properties of the PLA material are highly related to the ratio between the two mesoforms (L or D) of the lactic acid monomer. Using 100% L-PLA results in a material with a very high melting point and high crystallinity. If a mixture of D- and L-PLA is used instead of just the L-isomer, an amorphous polymer is obtained with a Tg of 60°C, which will be too low for some packaging purposes (Sinclair, 1996). A 90/10% D/L copolymer gives a material which can be polymerized in the melt, oriented above its Tg and is easy processable showing very high potential of meeting the requirements of a food packaging. The temperature of processing is between 60 and 125°C depending on the ratio of D- to L-lactic acid in the polymer (see Figure 2.5). Furthermore, PLA may be plasticized with its monomer or, alternatively, oligomeric lactic acid and the presence of plasticizers lowers the Tg. As outlined above, PLA offers numerous opportunities to tailor the properties of the finished material or package. PLA may be formed into blown films, injected molded objects and coatings all together explaining why PLA is the first novel biobased material produced on a major scale (see Chapter 7).

Biobased monomers

A wide variety of monomers, or chemical building blocks may be obtained from biobased feed stocks. These may be prepared using chemical and biotechnological routes, or a combination of both.

Since long, Castor oil has been recognized as an interesting starting material for making polyurethanes. Due to their water resistance some castor oil based polyurethane materials have found application in the electronics industry (Oertel, 1985) and coating market (Kase et al., 1987). Some seed crops and flax also contain fatty acids and oils where the major components of the recovered oil are β -linolenic acid, linoleic acid and oleic acid. This highly unsaturated material was of interest for application in

coatings and paints and in other potential applications utilizing an air drying process (Buisman, 1999). Other oils from marine and agricultural origin have been used in numerous applications including paints and other waterproof coatings (Carraher et al., 1981).

Oleochemicals, such as the unsaturated fatty acids oleic and ricinoleic acid, are derived from feedstocks such as coconut and castor beans and have long been recognised as useful chemical precursors in preparing polymeric materials. For example, oleic acid may be chemically transformed to azelaic (di)acid which has been used in polyamide synthesis. Other chemical transformations of oleochemicals result in the preparation of multifunctional alcohols, amines and esters. Some of these materials are prepared commercially by Cognis and Akzo Nobel amongst others, and are used for a variety of applications such as lubricants, surfactants and polycondensated monomers.

Carbohydrate sources such as woody material, molasses and maize give rise to a rich array of chemical and biotechnological transformations leading to a wide spectrum of potentially interesting chemicals. A well-established process which converts woody biomass to chemicals is the production of furfural. Furfural can be transformed to furfuryl alcohol which can be reacted to form a furan resin. As well as furfuryl alcohol synthesis a wide range of useful furan chemicals may be prepared although some are still in the development phase (Schiweck, 1991). Another example of the utilisation of woody materials is the preparation of levulinic acid from waste paper. Levulinic acid is a useful precursor for the synthesis of various lactones, furans and other functional building blocks. Plans to build a commercial plant for levulinic acid productions are being explored (Fitzpatrick, 1998).

Fermentation of carbohydrate materials using selected microorganisms has led to efficient pathways to the formation of multifunctional acids such as succinic acid. Diols, such as 1,3-propanediol, have also been prepared directly via fermentation. Pathways to the highly interesting monomers adipic acid and 1,4-butanediol, combine biotechnology and chemical transformation. In the case of adipic acid, glucose is transformed using microbes to muconic acid which is then chemically hydrogenated to adipic acid.

Terpene chemicals, isolated from pine trees for example, and transformed to other materials have resulted in the availability of a number of terpene based products such as terpineol, which are used as a fragrance ingredient (Gabelman, 1991). As well as this, other chemicals such as dipentene have been isolated and used to prepare resin materials. Due to the multifunctional nature of some basic terpene chemicals a wide range of derivatives are possible.

Protein engineering is a field of growing interest for the production of synthetic analogues to nature's polymers (O'Brien et al., 1998). Other developments include the possible production of biodegradable polymers currently derived from petroleum sources from biobased feedstock. An example of these developments is the work on Bionolle from renewable feedstock (Showa Denko HighPolymer, Japan). At present, biobased monomers may not be directly commercially attractive, however, biobased monomers derived by biotechnological pathways present promising alternatives to petrochemical polymer routes.

2.3.3. Category 3: Polymers produced directly by natural or genetically modified organisms

Poly(hydroxyalkanoates) (PHAs)

Poly(hydroxyalkanoates) (PHAs), of which poly(hydroxybutyrate) (PHB) is the most common, are accumulated by a large number of bacteria as energy and carbon reserves. Due to their biodegradability and biocompatibility these biopolyesters may easily find industrial applications. A general overview of the physical and material properties of PHAs, along with accomplished applications and new developments in this field, can be found in a recent review (Walle et al., (in press)).

The properties of PHAs are dependent on their monomer composition, and it is, therefore, of great interest that recent research has revealed that, in addition to PHB, a large variety of PHAs can be synthesized by microbial fermentation. The monomer composition of PHAs depends on the nature of the carbon source and microorganisms used. PHB is a typical highly crystalline thermoplastic whereas the medium chain length PHAs are elastomers with low melting points and a relatively lower degree of crystallinity. A very interesting property of PHAs with respect to food packaging applications is their low water vapour permeability which is close to that of LDPE.

PHB resembles isotactic polypropylene (iPP) in relation to melting temperature (175-180°C) and mechanical behaviour. PHBs T_g is around 9°C and the elongation to break of the ultimate PHB (3-8%), which is markedly lower than that of iPP (400%). An unfavourable ageing process is a major drawback for the commercial use of the PHB homopolymer. It has been reported in the literature that annealing can dramatically improve the mechanical properties of PHB by changing its lamellar morphology while subsequent ageing is prevented to a large extent. Incorporation of 3HV or 4HB co-monomers produces remarkable changes in the mechanical properties: the stiffness and tensile strength decrease while the toughness increases with increasing fraction of the respective co-monomer. Medium chain length PHAs, unlike PHB or its copolymers, behave as elastomers with crystals acting as physical crosslinks and, therefore, can be regarded as a class of its own with respect to mechanical properties. Elongation to break up to 250-350% has been reported and a Young's modulus up to 17 MPa. These materials have a much lower melting point and T_g than their PHB counterparts.

Applications that have been developed from PHB and related materials (e.g. Biopol) can be found in very different areas and cover packaging, hygienic, agricultural, and biomedical products. Recent application developments based on medium chain length PHAs range from high solid alkyd-like paints to pressure sensitive adhesives, biodegradable cheese coatings and biodegradable rubbers. Technically, the prospects for PHAs are very promising. When the price of these materials can be further reduced, application of biopolyesters will also become economically attractive.

Bacterial cellulose

To date, bacterial cellulose is rather unexploited, but it represents a polymeric material with major potential (Iguchi et al., 2000). Bacterial strains of *Acetobacter xylinum* and *A. pasteurianus* are able to produce an almost pure form of cellulose (homo-beta-1,4-glucan). Its chemical and physical structure is identical to the cellulose formed in plants (Brown, 1996). Plant cellulose, however, has to undergo a harsh chemical treatment to remove lignin, hemicellulose and pectins. This treatment severely impairs the material characteristics of plant cellulose: the degree of polymerisation decreases almost ten-fold and the form of crystallization changes.

Bacterial cellulose is processed under ambient conditions and the degree of polymerization is 15000, 15 times longer than cellulose from woodpulp. Bacterial cellulose is highly crystalline. In bacterial cellulose, 70% is in the form of cellulose I and the rest is amorphous. This composition results in outstanding material properties: a modulus as high as 15-30 GPa was determined across the plane of the film.

Production costs of bacterial cellulose are high due to the low efficiency of the bacterial process; approximately 10% of the glucose used in the process are incorporated in the cellulose. The high price of bacterial cellulose of approximately 20 Euro/kg hampers its applicability in low-added-value bulk products. Several high-added-value specialty applications have been developed. The material has been used as an artificial skin, as a food-grade non-digestible fiber, as an acoustic membrane, and as a separation membrane (Van Damme et al., 1996).

2.4. Material properties

2.4.1. Gas barrier properties

Many foods require specific atmospheric conditions to sustain their freshness and overall quality during storage. Hence, increasing amounts of our foods are being packed in protective atmosphere with a specific mixture of gases ensuring optimum quality and safety of the food product in question. To ensure a constant gas composition inside the package, the packaging material needs to have certain gas barriers. In most packaging applications the gas mixture inside the package consists of carbon dioxide, oxygen and nitrogen or combinations hereof. The objective of this section is to describe the gas barriers of biobased materials using mineral oil based polymer materials as benchmarks.

Literature provides a vast amount of information on the barrier properties of biobased materials. However, comparisons between different biobased materials are complicated and sometimes not possible due to the use of different types of equipment and dissimilar conditions for the measurements.

In Figure 2.3, different biobased materials are compared to conventional mineral-oil-based polymer materials. The figure is based on information from literature and on measurements of commercially available materials performed by ATO (Wageningen, NL).

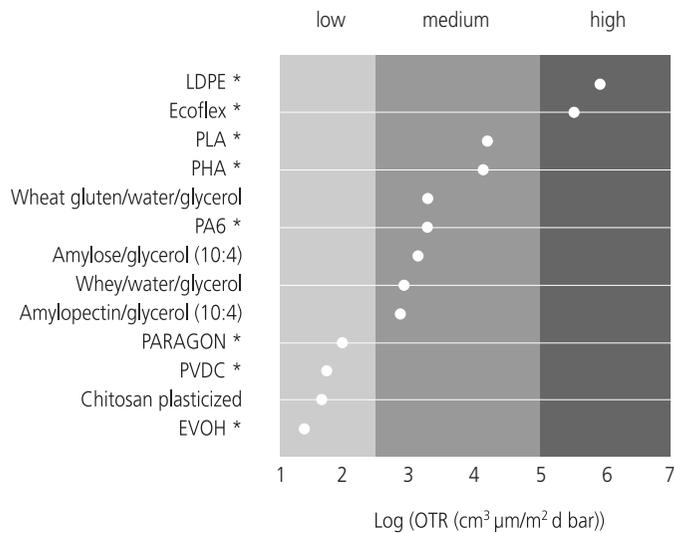


Figure 2.3 Comparison of oxygen permeability of biobased materials compared to conventional mineral-oil-based materials. Permeability of materials marked with * was measured by ATO, Wageningen, NL (23°C, 50% RH), information on other materials is based on literature (Rindlav-Westling et al., 1998; Butler et al., 1996).

As seen in the Figure 2.3, biobased materials mimic quite well the oxygen permeabilities of a wide range of the conventional mineral-oil-based materials and it is possible to choose from a range of barriers among the presented biobased materials. It is noteworthy that developments are still being made.

The conventional approach to produce high-barrier films for packaging of food in protective atmosphere is to use multi-layers of different films to obtain the required properties. A laminate that is often used in food packaging consists of an layer of EVOH or PA6 combined with LDPE combining the gas barrier properties of PA6 or EVOH with the water vapour barrier, the mechanical strength and the excellent sealing properties of the LDPE. A similar multi-layer approach for biobased materials may likewise be used to produce materials with the required properties. As seen in Figure 2.3 starch-based materials could provide cheap al-

ternatives to presently available gas barrier materials like EVOH and PA6 and an equivalent biobased laminate would be an outer-layer of plasticized chitosan, a protein or starch-derived film combined with PLA or PHA (see Section 2.5). Notably, the gas barrier properties of PA6 and EVOH are sensitive towards moisture and the LDPE creates a very effective water vapour barrier ensuring that the moisture from the foodstuff does not interfere with the properties of PA6 or EVOH. In the same fashion, PLA and PHA will protect the moisture-sensitive-gas-barrier made of polysaccharide and protein. Some interesting developments have made it possible to improve water vapour and gas properties of biobased materials many-fold by using plasma deposition of glass-like SiO_x coatings on biobased materials or the production of nano-composites out of a natural polymer and modified clay (Fischer et al., 2000; Johannson, 2000).

In general, the oxygen permeability and the permeability of other gases of a specific material are closely interrelated and, as a rule of the thumb, mineral oil based polymers have a fixed ratio between the oxygen and carbon dioxide permeabilities. This relation is also observed for biobased materials. However, for some biobased materials, e.g. PLA and starch, the permeability of carbon dioxide compared to oxygen is much higher than for conventional plastics (Petersen and Nielsen, 2000).

Gas barriers and humidity

As many of these biobased materials are hydrophilic, their gas barrier properties are very much dependent on the humidity conditions for the measurements and the gas permeability of hydrophilic biobased materials may increase manifold when humidity increases. Notably, this is a phenomenon also seen with conventional polymers. The gas permeability of high gas barrier materials, such as nylon and ethylvinyl alcohol, is likewise affected by increasing humidity. Gas barriers based on PLA and PHA is not expected to be dependent on humidity.

2.4.2. Water vapour transmittance

A major challenge for the material manufacturer is the by nature hydrophilic behaviour of many biobased polymers as a lot of food applications demand materials that are resistant to moist conditions. However, when comparing the water vapour transmittance of various biobased materials to materials based on mi-

neral oil (see Figure 2.4), it becomes clear that it is possible to produce biobased materials with water vapour transmittance rates comparable to the ones provided by some conventional plastics. However, if a high water vapour barrier material is required, very few biobased materials apply. Notably, developments are currently focusing on this problem and future biobased materials must also be able to mimic the water vapour barriers of the conventional materials known today.

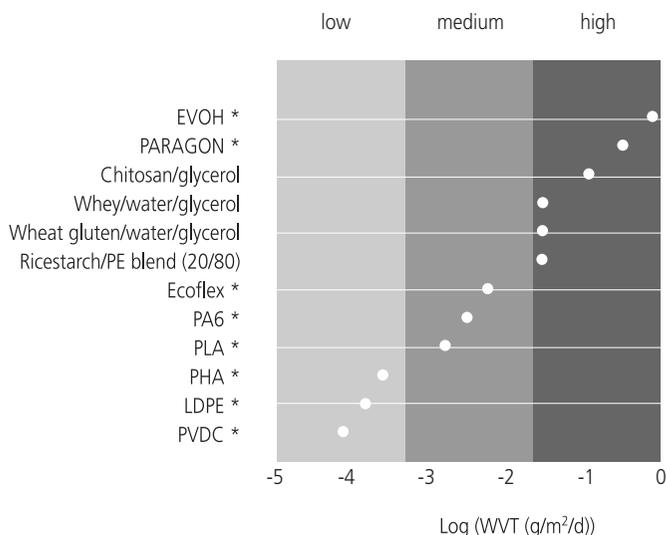


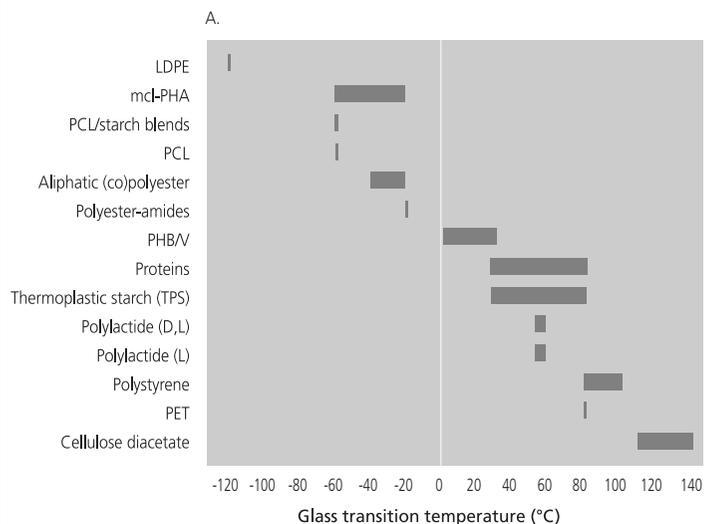
Figure 2.4 Water vapour transmittance of biobased materials compared to conventional packaging materials based on mineral oil. Water vapour transmittance of materials marked with * was measured by ATO (Wageningen, NL) at 23°C, 50% RH. Transmittance of other materials are based on literature and measured at same conditions (Rindlav-Westling et al., 1998; Butler et al., 1996).

2.4.3. Thermal and mechanical properties

Next to the barrier properties of the final packaging, the thermal and mechanical properties of the materials are both important for processing and also during the use of the products derived from these materials. Most biobased polymer materials perform in a similar fashion to conventional polymers. This indicates that

both polystyrene-like polymers (relatively stiff materials with intermediate service temperatures), polyethylene-like polymers (relatively flexible polymers with intermediate service temperatures) and PET-like materials (relatively stiff materials with higher service temperatures) can be found among the available biobased polymers.

The mechanical properties in terms of modulus and stiffness are not very different compared to conventional polymers. In figure 2.5 a comparison of the thermal properties of biobased polymers with existing polymers is made. The modulus of biobased materials ranges from 2500-3000 MPa and lower for stiff polymers like thermoplastic starches to 50 MPa and lower for rubbery materials like medium chain polyhydroxyalkanoates. Furthermore, the modulus of most biobased and petroleum-derived polymers can be tailored to meet the required mechanical properties by means of plasticizing, blending with other polymers or fillers, crosslinking or by the addition of fibres. A polymer like bacterial cellulose could for instance be used in materials which requires special mechanical properties. In theory, biobased materials can be made having similar strength to the ones we use today (Iguchi et al., 2000).



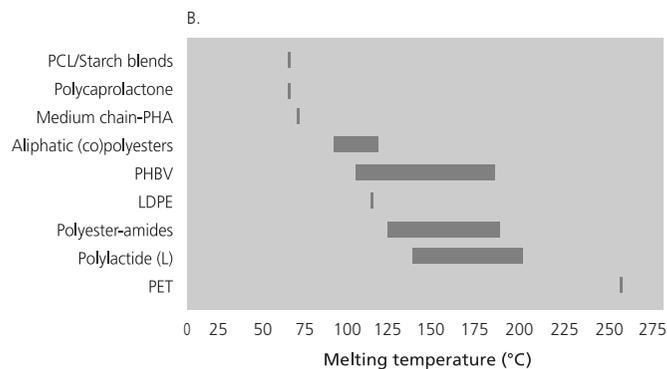


Figure 2.5 Comparison of the thermal properties of biobased polymers with conventional polymers. (All data is from company information).

2.4.4. Compostability

The issues of biodegradability and compostability are addressed in Chapter 5, but a comparison of the compostability of the materials is also provided in this chapter. Figure 2.6 compares the compostability of various biobased materials. Notably, the “composting time” depicted in the figure represents the approximate period of time required for an acceptable level of disintegration of the material to occur. This means that the original material should not be recognizable anymore in the final compost (fraction < 10 mm) nor in the overflow (fraction > 10 mm). The composting time does not reflect the time required for the biodegradation of the materials to be fully completed. The process could subsequently be completed during the use of the compost. The level of technology applied in the composting process highly affects the composting time needed for complete disintegration. Hence, it takes much longer to obtain a mature compost using low technology composting (e.g. passive windrow composting) than using high technology as in an intensively controlled tunnel composting process.

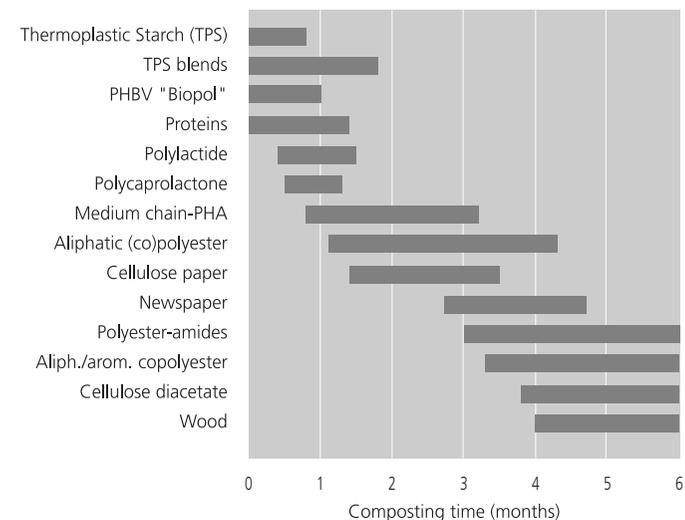


Figure 2.6. Indication of the time required for composting of various biobased and synthetic polymeric materials. Measurements of composting times were performed at ATO. The durations presented in this figure are based on an intermediate level of technology as observed in actively aerated and mechanically turned hall composting.

The durations presented in figure 2.6 are based on an intermediate level of technology as observed in actively aerated and mechanically turned hall composting. Furthermore, the composting time needed for complete disintegration is also affected by the particle size of the material. For example, wood is rapidly composted in the form of sawdust and small chips. A wooden log, however, takes more than one year to be completely disintegrated. The durations presented in this figure are based on dimensions regularly used for packaging applications.

The compostability of the materials are highly dependent on the other properties of the materials, e.g. the first step of the composting is often a hydrolysis or wetting of the material. The rate of this step is very much related to the water vapour transmittance and the water resistance of the material. Hence, the composting rate of a material will be dependent on its other properties.

2.5. Manufacturing of biobased food packaging

Engineering of a biobased package or packaging material requires knowledge of the processing and material properties of the polymers. If the properties of the native biopolymer are not identical to the required one, or if the polymer by nature is not thermoplastic, a certain modification of the polymer must take place. For very specific requirements (very low gas permeability or high water resistance) it is unlikely that one polymer will be able to provide all required properties even after modifications. Hence, it is necessary to use multiple materials in a composite, a laminate or co-extruded material.

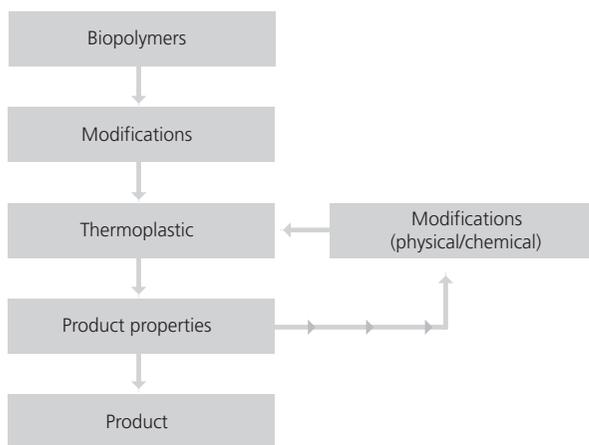


Figure 2.7. *Designing and manufacturing of biobased packages and packaging materials require a multistep approach.*

In this section the main categories of food packaging will be discussed. For these categories the main material requirements will be discussed and compared with the development of the materials from biobased polymers. Commercial and near commercial developments in this area will be mentioned.

2.5.1. Possible products produced of biobased materials

The fundamental repeating chemical units of the biobased materials described so far are identical to those of a significant body of the conventional plastics. Thus, in the broadest sense, polysaccharides possessing repeating acetal functionality can be

regarded as the naturally occurring analogues of the synthetic polyacetals; proteins (repeating peptide functionality) can be compared to the synthetic polyamides while polylactic acid is merely an example of the diverse group of polyesters. Clearly, however, the gross physical and chemical properties of native biobased materials and their synthetic counterparts are quite different and this is a feature of additional chemical functionality inherent in biobased materials. It should be expected that following requisite processing and product development of biobased materials resulting properties should equal or better those of the conventional alternatives. However, such processing and product development is not always trivial and is unlikely to be cost effective in all cases.

It is not surprising, therefore, that the current applications of biobased materials seek not to emulate the properties of conventional plastics, but to capitalize on inherent biodegradability and on other unique properties of these polymers. Biobased plastic applications are currently targeted towards single-use, disposable, short-life packaging materials, service ware items, disposable non-wovens and coatings for paper and paperboard applications. However, the possible products made from biobased resources covers a broader range, and some of the potential products and applications are summarized in Table 2.1. In general, the same shapes and types of food packaging can be made from synthetic and biobased resources. The question is whether the same performance can be achieved by using the biobased materials as with the synthetic ones.

Table 2.1 *The major processing routes to potential biobased products.*

Processing route	Product examples
(Co-)Extruded film	Packaging film
Cast film	Packaging film
Thermoformed sheets	Trays, cups
Blown films	Packaging film
Injection (blow-)moulding	Salad pots, cutlery, drinking beakers, cups, plates, drinks bottles, trays
Fibres and non-wovens	Agricultural products, diapers, feminine hygiene products, certain medical plastics, clothing
Extrusion coating	Laminated paper or films

2.5.2. Blown (barrier) films

Blown films comprise one of the first product categories to be developed based on mineral oil derived biodegradable polyesters. They have successfully been applied as garbage bags and related applications. Film blowing grades of renewable polymers have been developed based on PLA. Blown films based on these biopolyesters exhibit excellent transparency and cellophane-like mechanical properties. The sealability depends on the degree of crystallinity and good printability can also be achieved. The possibilities of film blowing PHB/V materials are at this time limited due to their slow crystallization and low melt strength.

In many food packaging applications, a water vapour barrier as well as gas barriers are required. No single biobased polymer can fulfil both these demands. In this case, the use of co-extrusion can lead to laminates which meet the objectives. Paragon (Avebe, NL) materials which are based on thermoplastic starch can be film blown in a co-extrusion set-up with polymers like PLA and PHB/V as coating materials, resulting in a barrier coating which, for example, proved to be successful in the packaging of cheese (Tuil et al., 2000). The use of Paragon tie-layers provides the adhesion between the coating and the base layer. In this way, starch-based materials could provide cheap alternatives to

presently available gas barrier materials like EVOH and PA6 (see Figure 2.3).

The properties (mechanical strength, gas and water vapour properties) of blown films can be improved by coating of a glass-like ultra thin layer of SiO_x or by producing nano-composites. Addition of nano-particles during processing of the film produces composites with improved water and gas barrier properties (Fischer et al., 2000) and ongoing developments at TNO industry (NL) aims at producing hydrophobic starches based on these composites. A similar approach is to use a glass-like ultra-thin coating of SiO_x improving the barriers of the material immensely (Johansson, 2000 and 1997).

2.5.3. Thermoformed containers

A next class of products is thermoformed containers for food packaging. In order to be able to thermoform a polymer it should be possible to process this material from the melt (extrusion) into sheets and consequently thermoforming these sheets just above the T_g or T_m of the material. Thermoformed products can be found based on PLA and PHB/V. Again, it is possible to produce thermoformed articles from laminates based on Paragon as well as other thermoplastically processable biopolymers.

2.5.4. Foamed products

Starch-based foams for loose fill applications (Novamont, (I), National Starch (USA) a.o.) have been commercially introduced with success some years ago and the market for these products is still growing. Foamed products like trays and clamshells based on starch for food packaging have not yet been introduced commercially. Products based on a molding technique from a slurry phase (Earthshell (USA), APACK (D)) are close to market introduction. These products are produced from starch base slurries with inorganic and agrofiber based fillers. Other proposed techniques include loose-fill molding (Novamont (I), Biotec (D)), foam extrusion (Biotec (D)), and extrusion transfer molding (Standard Starch (USA)) and expandable bead moulding (Tuil et al., (In press)). Foamed products based totally on PLA are still in a developmental phase.

In order to be able to use these starch-foamed products in food contact applications coatings should be applied on the starch-

based foams. Adhesion between the foam and the coating is of importance. Paraffin and other oligomer based coatings are proposed next to PLA and PHB/V based coatings. Protein and medium chain length PHA based coatings (ATO, 2000) are close to market introduction.

2.5.5. Coated paper

It is expected that paper will stay an important biobased packaging material. Paper and board materials have excellent mechanical properties, however, the gas permeabilities are too high for many food applications. The hydrophilic nature of the paper-based materials is a major challenge of these materials when packaging moist foods. To date, the paper-based materials have been coated with a thin layer of synthetic plastic which has provided the materials with the required gas property and water resistance. Alternatively, biobased materials might be used as coating materials thus paving the way for a 100% biobased packaging material. Paper-based materials coated with PE are readily repulpable as the hydrophobic PE is easily removed in the pulping process. Hence, paper-based materials coated with biobased, hydrophobic polymeric materials are, likewise, going to be repulpable.

2.6. Additional developments

To be able to produce a 100% biobased packaging development of biobased additives is needed. Additives used in the production of packaging are plasticizers, UV-stabilisers, adhesives, inks and paints, natural pigments and colorants. So far, few developments have been made in this field and it is suggested to direct research to this area.

2.7. Conclusions and perspectives

Developments of polymeric materials based on biological resources are being made with an ever-increasing rate making it almost impossible to produce a paper on the state-of-the-art of this area. The information presented here may very likely be outdated when these lines are being read and novel products, polymers and optimized performance of these are an expected scenario.

Biological derived polymers may be used for the production of all types of packaging (trays, cups, bottles, films, etc.) using the same equipment and technology used for conventional materials. However, these materials have to be well performing in order to be able to compete with the highly developed and sophisticated materials used today. Comparing the properties of biobased polymeric materials with the conventional synthetic petroleum-derived polymers shows a major potential of these polymers for the production of well-performing food packaging. However, when using proteins or polysaccharides in the materials their sensitivity towards relative humidity must be overcome. The biobased materials have an inherent potential of being compostable which may help the commercialization of these materials. Similar to the synthetic materials used today it will be necessary to use several polymeric materials in multi-layers or composites tailoring the properties of the packaging to meet the demands of specific foodstuffs. In general, the more diverse side chains and functional groups of biobased polymers, compared to conventional plastics derived from mineral oil, gives the resin and material manufacturer unique possibilities to tailor the properties of the finished package. This advantage should be used further to produce materials with even better properties than the ones we know today.

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3. Food biopackaging

3.1. Introduction

Food packages serve a number of important functions, including containment and protection of food, maintaining the sensory quality and safety of food, conferring convenience to food and communicating information about food to consumers (Robertson, 1993). This chapter focuses on biobased packaging for food and discusses critical packaging issues. The role that biobased packaging materials can play in protecting the sensory quality and safety of several groups of food products is discussed. In addition to sensory and safety aspects relating to food, it is recognized that other issues also require careful consideration in the development and selection of biobased food packages. These aspects, which are discussed superficially in the chapter, include logistical, marketing, legislative, environmental and financial constraints to the production of the biobased materials.

3.2. Food packaging definitions

Most commonly used food packages clearly fall into primary, secondary or tertiary packaging categories. For a variety of food products, however, conventional packaging does not provide optimal conditions for product storage (Petersen et al., 1999) and a number of approaches are used to design packages for specific products. Such product-specific packaging includes applying edible films and coatings, active packaging, modified atmosphere packaging (MAP), and using combinations of packaging materials.

3.2.1. Primary, secondary and tertiary packaging

Primary packaging materials are those which are in direct contact with foods. Their functions are to contain, protect and facilitate distribution and storage of foods while satisfying consumer needs with respect to convenience and safety (Brown, 1992). The properties of the primary packaging materials should be tailored according to the requirements set by the packaged foods. Primary packaging is packaging where the material and food may be separated from each other. Thus, edible coatings do not fall into the primary packaging category. However, edible films may perform similar functions to primary packaging.

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Secondary packaging is often used for physical protection of the product. It may be a box surrounding a food packaged in a flexible plastic bag. It could also be a corrugated box containing a number of primary packages in order to ease handling during storage and distribution, improve stackability, or protect the primary packages from mechanical damage during storage and distribution. Secondary packaging may also provide crucial information on lot number, production dates, etc. aimed at distributors and retailers. Furthermore, secondary packaging may be used for marketing purposes, e.g. a box that may be unfolded into retail display cabinets in the supermarket.

Tertiary packaging incorporates the secondary packages in a final transportation package system. Again, the purpose is to facilitate storage and handling and to protect the packaged product against mechanical damage, weather conditions, etc.. Examples of tertiary packaging are boxes, pallets and stretch foils.

3.2.2. *Edible coatings and films*

Edible coatings and films comprise a unique category of packaging materials differing from other biobased packaging materials and from conventional packaging by being edible. Films and coatings differ in their mode of formation and application to foods. Edible coatings are applied and formed directly on the food product either by addition of a liquid film-forming solution or molten compounds. They may be applied with a paintbrush, by spraying, dipping or fluidising (Cuq et al., 1995). Edible coatings form an integral part of the food product, and hence should not impact on the sensory characteristics of the food (Guilbert et al., 1997). Edible films, on the other hand, are freestanding structures, formed and later applied to foods. They are formed by casting and drying film-forming solutions on a levelled surface, drying a film-forming solution on a drum drier, or using traditional plastic processing techniques, such as extrusion. Edible films and coatings may provide barriers towards moisture, oxygen (O₂), carbon dioxide (CO₂), aromas, lipids, etc., carry food ingredients (e.g. antimicrobials, antioxidants, and flavour components), and/or improve the mechanical integrity or handling of the food product. Edible films and coatings may be used to separate different components in multi-component foods thereby improving the quality of the product (Krochta and De Mulder-Johnston, 1997). They may be used to reduce the amount of pri-

mary synthetic packaging material used in a product or allow conversion from a multi-layer, multi-component packaging material to a single component material. Edible coatings may also help maintain food quality by preventing moisture and aroma uptake/loss, etc. after opening of the primary packaging.

3.2.3. *Active packaging*

Packaging is termed active when it performs a role other than providing an inert barrier to external conditions. Active packaging solutions could involve the inclusion of an oxygen scavenger or an antimicrobial agent if microbial growth is the quality-limiting variable (Rooney, 1995).

3.2.4. *Modified atmosphere packaging*

Modified Atmosphere Packaging (MAP) is defined as the enclosure of food products in a high gas barrier film in which the gaseous environment has been changed or modified to control respiration rates, reduce microbiological growth, or retard enzymatic spoilage with the intent of extending shelf-life (Smith et al., 1995). For example, red meats are packaged in atmospheres in which the oxygen and carbon dioxide contents are elevated, relative to air, to maintain product colour, yet inhibit microbial growth.

3.2.5. *Combination materials*

Combining packaging materials in, for example, laminates or co-extrudates may improve barrier characteristics significantly. One example is combining cardboard and plastics in gable top beverage packages. Cardboard provides stability and light protection while the plastics contribute to an optimal packaging solution by providing a water vapour barrier.

3.3. **Food packaging requirements**

The packaging requirements of foods are complex. Unlike inert packaged commodities, foods are often dynamic systems with limited shelf-life and very specific packaging needs. In addition, since foods are consumed to sustain life, the need to guarantee safety is a critical dimension of their packaging requirements. While the issue of food quality and safety is first and foremost in the mind of the food scientist, a range of other issues surrounding the development of any food package must be considered before a particular packaging system becomes a reality (see Table 3.1).

Table 3.1. Food packaging requirements.

Area	Overall	Specific
Food Quality	Maintain or enhance sensory properties	Maintain taste Maintain smell Maintain colour Maintain texture
	Maintain the necessary microbiological standards	Should not support the growth of unwanted micro-organisms If necessary, can be pasteurized or sterilized
Manufacturing	Offer simple, economic processes for package formation	Sheet, film, containers, pouches Adequate mechanical properties
	Give compatibility in product filling	Dimensional stability Good runability on filling lines Closeability Compatibility with existing machinery
Logistical	Facilitate distribution	Conform to industry requirements (e.g. size, palletisation) Carry the required codes (bar code, product and sell-by)
Marketing	Enhance point of sale appeal	Good graphics Aesthetically pleasing Culture-specific consumer preferences Deliver the required functionality (e.g. openability, dust-free)

Environment	Not endanger human safety	Safe food contact interactions Avoid physical harm
	Use resources responsibly	Have a positive LCA
Legislation	Facilitate waste management	Should be recoverable. Ought to be recyclable, burnable or compostable
	National laws	Meet labelling, hygiene, migration conditions
Financial	Cost effectiveness	Acceptable price per food package Price of concomitant machinery

Biobased packaging materials must meet the criteria that apply to conventional packaging materials associated with foods. These relate to barrier properties (water, gases, light, aroma), optical properties (e.g. transparency), strength, welding and moulding properties, marking and printing properties, migration/scalping requirements, chemical and temperature resistance properties, disposal requirements, antistatic properties as well as issues such as the user-friendly nature of the material and whether the material is price-competitive. Biobased packaging materials must also comply with food and packaging legislation, and interactions between the food and packaging material must not compromise food quality or safety. In addition, intrinsic characteristics of biobased packaging materials, for example whether or not they are biodegradable or edible, can place constraints on their use for foods.

3.3.1. Replacing conventional food packaging materials with biobased materials – a challenge

One of the challenges facing the food packaging industry in producing biobased packaging is to match the durability of the packaging with product shelf-life. The biobased material must remain stable maintaining mechanical and/or barrier properties and functioning properly during storage of the food. Ideally, the material should biodegrade efficiently on disposal. Thus, environ-

mental conditions conducive to biodegradation must be avoided during storage of the food product whereas optimal conditions for biodegradation must exist after discarding. This situation presents an interesting challenge for the design and use of bio-based packaging materials since many of the factors that influence biodegradation (water activity, presence of microorganisms, temperature, composition of bio-material, etc.) also affect the rate of deterioration of the packaged food. In the case of edible films they may be required to operate as localized packages providing barriers to moisture or gases while the food is stored, yet they must become part of the food at the point of consumption.

Like conventional packaging, biobased packaging may need to supply consumers with mandatory product information as well as optional information such as cooking directions, recipes, etc.. These additional requirements provide further challenges for bio-based packaging applications. For example, new technology may be required to provide labelling information on biodegradable packages. Biodegradable and/or edible adhesives, labels or inks and solvents should be considered.

3.3.2. Biobased packaging – food quality demands

Defining the requirements of packaging in terms of maintaining food quality depends on how food quality is defined. The factors that contribute to consumer perception of food quality include sensory attributes (e.g. appearance, flavour, texture), nutritional content, safety concerns (e.g. microorganisms, residues), ethical issues (e.g. humane production methods), and the price of the food. In so far as biobased packaging materials impact on these attributes, they impact on food quality.

Deterioration in the sensory attributes, nutritional content and safety of foods is caused principally by physical and chemical changes in the food during storage and by microbial spoilage. Biobased packaging, like conventional packaging, should minimize these deteriorative changes in food products. Chemical changes in foods, leading to deterioration in quality, include browning reactions (non-enzymatic and enzymatic), hydrolysis of lipids and proteins, lipid and protein oxidation and glycolytic changes (see Table 3.2). To control these chemical reactions bio-based packaging materials must have the capacity to control one or more of the following: the gaseous atmosphere around foods

(oxygen, carbon dioxide, nitrogen), water activity, light, and temperature.

Table 3.2. *Packaging measures to prevent deteriorative changes in foods.*

Deteriorative change	Preventative properties of packaging
Chemical	
Rancidity (oxidation)	Oxygen barrier
Browning reactions	Light barrier
Fat degradation (lipolysis)	Moisture barrier
Protein degradation (proteolysis)	
Microbiological	
Growth of microorganisms	Oxygen barrier No/low oxygen atmospheres Oxygen absorbers Carbon dioxide emitters Moisture barrier Migration of antimicrobial agents from package
Physical	
Textural change (softening, hardening)	Moisture barrier Control of chemical, micro-biological changes
Crushing, bruising of product	Robust packaging Package stability

Deterioration in food quality due to microbial growth will be affected by the ability of the biobased packaging materials to control factors such as water activity, pH and nutrient migration. In addition to minimizing deteriorative changes originating from indigenous substrates within or on the surface of food products, packaging materials may be required to protect foods from exogenous influences. Food products may need to be protected from microorganisms originating from other sources or from bruising or crushing as a result of poor handling (Petersen et al., 1999).

Physical changes associated with deterioration in food quality include softening, toughening, loss of water holding capacity,

emulsion breakdown, swelling/shrinkage, and crushing/breakage (Petersen et al., 1999). Physical changes resulting from water uptake may be prevented by controlling moisture migration into foods or between different food components. Since chemical and physical changes do not occur independently of each other, controlling chemical reactions and microbial deterioration with biobased packaging materials may also contribute to the physical stability.

3.4. State-of-the-art in biopackaging of foods

The use of biobased packaging materials for food depends on availability, quantities, prices and properties of the materials. To date, considerable resources have been allocated to research, development and pilot scale studies, but usage of biobased packaging materials in the food industry is relatively limited. Technical packaging considerations as well as marketing aspects are important criteria when selecting a given packaging material or technique. These criteria are illustrated by numerous feasibility studies carried out for small and large food companies encompassing both technical and market-oriented aspects. However, the studies are confidential and are consequently not known to everyone.

“State-of-the-art” applications of biobased primary, secondary, and tertiary packaging as well as edible films and coatings are listed in Table 3.3. The Table clearly indicates that the literature on biobased primary, secondary and tertiary packaging is rather limited. This may be due to the short time frame that the materials have been available for testing and also due to the fact that customer-specific tests results are not available to the public.

Many food applications of coating materials have been investigated and tested over the years. The majority of these investigations have been undertaken in academic environments; a fact that is reflected in the huge volume of published scientific articles and reviews on edible films and coatings compared to the limited number of patents issued from industry. Producers of the coating materials or manufacturers of food products may not yet consider them to be of commercial interest because: (i) the coating materials, themselves, make up a minor part of the food product (typically 0.001-0.01%) and (ii) the application of the edible coating or film requires an extension of the processing

line with either a spraying, dipping or a pan coating unit. Not all factories have room for the extension or the cost for additional coating equipment is simply too high compared to the benefit obtained with the coating. Thus, while many applications of edible coatings/films have been investigated and identified and found to be very interesting from an academic point of view, there have undoubtedly been some successful commercial applications when it comes to large-scale use and implementation in the food industry the adoption of the listed applications is rather limited.

3.5. Potential food applications

Research and Development activities in the area of food biopackaging have intensified over the last decade. However, the lack of food biobased packaging materials on the market is evident and it appears that scientific studies on these materials are still very much in their infancy. Food manufacturers and packaging producers are currently testing biobased packaging materials for foods, but because of the confidential nature of the work it is difficult to get information on the findings. Thus, it is difficult to present the state of play in the market at present or to predict what will happen in the near future.

In this section, potential biobased packaging materials for particular food products are suggested bearing in mind product-specific requirements and that biobased packaging materials should, at least, meet the same food packaging requirements as conventional packaging materials. Different food categories are discussed in terms of deteriorative reactions that limit their shelf-life. Some of the materials suggested are not directly applicable presently and further optimization will be needed. However, the examples give an indication of the potential for biobased food packaging in the future. To present an overview of the potential food applications discussed in this section examples are summarized in Table 3.4.

3.5.1. Fresh meat products

Two factors are critical in the packaging of red meats: colour and microbiology (Robertson, 1993). In order to preserve the red colour of fresh meat, attributed to oxymyoglobin, a high oxygen level over the product surface is required. This level can be obtai-

Table 3.3 “State of the art” food application of biobased packaging materials and edible films/coatings.

Product example	Critical functions of packaging	Value added function	Examples of materials	References
MEAT PRODUCTS				
BIOBASED PACKAGING				
Beef and chicken	Absorb moisture		Trays of fresh pulp Mixture of wood pulp and starch	Pactiv, Omni-Pac, Germany Apack, Germany
Ground beef	Oxygen barrier Water vapour barrier		Starch-polyethylene films containing corn starch (0-28%), low- or high-molecular weight oxidised polyethylene and pro-oxidant	Kim and Pometto III (1994)
EDIBLE COATING				
Fresh meat	Moisture barrier	Antioxidants	Alginate, carrageenan, cellulose,	Lazarus et al. (1976)
Cured meat	Oxygen barrier	Antimicrobial agents	gelatin and soy protein	Suderman et al. (1981)
Casings	Carbon dioxide barrier			Wanstedt et al. (1981)
Cooked meat	Frying oil barrier			Keil (1961)
Beef	Adhesion			Moorjani et al. (1978)
	Mechanical Protection			Olson and Zoss (1985)
	Inhibit microbial growth			Kester and Fennema (1986)
				Siragusa and Dickson (1992)
				Ma-Edmunds et al. (1995)
				Chinnan et al. (1995)
				Stuchell and Krochta (1995)
				Balasubramaniam et al. (1997)
				Shaw et al. (1980)
				Robin et al. (1992)
				Feeney et al. (1992)
				Polansky (1993)
Product example				
SEAFOOD				
EDIBLE COATING				
Fish	Oxygen barrier Moisture barrier	Antioxidant (time-dependent migration)	Whey protein and acetylated monoglycerides	Stuchell and Krochta (1995)
Frozen fish	Moisture barrier Oxygen barrier Mechanical protection Batter adhesion	Antioxidants Antimicrobial agents Batter adhesion	Caseins, whey proteins, lipids, alginate, carrageenan	Bauer et al. (1969) Fischer and Wong (1972) Cottrell and Kovacs (1980) Guiseley et al. (1980) Torres et al. (1985) Hirasa (1991) Stuchell and Krochta (1995) Mu et al. (1996)
Frozen crustacean (shrimp)	Moisture barrier Oxygen barrier Mechanical protection Batter adhesion	Antioxidants Antimicrobial agents Batter adhesion	Caseins, whey proteins, lipids, alginate, carrageenan	Bauer et al. (1969) Fischer and Wong (1972) Cottrell and Kovacs (1980) Guiseley et al. (1980) Torres et al. (1985) Hirasa (1991) Stuchell and Krochta (1995) Mu et al. (1996)
READY MEALS				
BIOBASED PACKAGING				
French fries	Containment	Convenient food	Trays for french fries and chips made from coated pulp and starch	Used in Belgium
Hamburger pockets, sandwiches	Containment		Powdered starch, foams, shells	Used by McDonalds
EDIBLE COATING				
Pizza base/sauce	Moisture barrier	Thickening agent and moisture	Alginate, whey protein	Kamper and Fennema (1985)

Product example	Critical functions of packaging	Value added function	Examples of materials	References
DAIRY PRODUCTS				
BIOBASED PACKAGING				
Yoghurt	Mechanical protection Moisture barrier Carbon dioxide barrier	migration control	Cardboard + unspecified biobased plastic PLA	Non-scientific literature Has been used by Danone
Butter/margarine	Moisture barrier Light barrier Grease barrier	Anti fogging Snug down	10% PLA + 90% co-polyester 10% PLA + 90% co-polyamide 10% PLA + 90% starch 10% PLA + 90% PCL	
Cheese			Starch laminate	Van Tuil (2000)
Soft cheese	Moisture barrier Gas barrier Containment		Nitrocellulose lacquered cellophanes	UCB-films, Belgium
BEVERAGES				
BIOBASED PACKAGING				
	Moisture barrier		Paper cups coated with biobased degradable plastics, PLA, starch based, coated board	Sobol (1996)
FRUITS AND VEGETABLES				
BIOBASED PACKAGING				
Soft fruits	Containment		Nets of biobased biodegradable starch based plastics, pulp trays (from recycled paper), cellophane, corrugatedboard trays and transport boxes Nitrocellulose lacquered cellophanes	On the marked in Germany Bastioli (2000) UCB-films, Belgium
Product example				
Critical functions of packaging				
Value added function				
Examples of materials				
References				
Fruits (e.g. berries)	Containment		Pulp containers	Anon. (1989) Pactic, Omni-Pac, Germany
Fresh products (shredded lettuce and cabbage, head lettuce, cut broccoli, whole broccoli, tomatoes, sweet corn and blueberries)	Moisture barrier Oxygen barrier Carbon dioxide barrier Mechanical protection		Laminate of chitosan (14.5% by weight)-cellulose (48.3%) and polycaprolactone (glycerol (36.2%) and protein (1.0%))	Makino and Hirata (1997)
Cut vegetables		Anti fogging	Starch laminate	Van Tuil (2000)
EDIBLE COATING	Oxygen barrier Carbon dioxide barrier		Chitosan, carnauba wax, zein	Baldwin (1994) Hagenmaier and Baker (1994)
Mushrooms	Oxygen barrier Moisture barrier		Alginate	Nussinowitch and Kampf (1993)
Apples	Moisture barrier Oxygen barrier	Improve appearance (gloss) Delay browning and microbial growth (carrier of antioxidants and preservatives)	Prolong (sucrose-fatty acid ester) Nutri-Save (polysaccharide) NatureSeal™ 120 (cellulose based) Sodium caseinate-acetylated monoglyceride Calcium caseinate-acetylated monoglyceride	Lau and Meheriuk (1994) Baldwin et al. (1996) Park and Jo (1996) Avena-Bustillos et al. (1997)
Carrots	Moisture barrier Gas barrier	Retention of flavour Reduce white surface discoloration Retard senescence	NatureSeal™ 1000 (cellulose based) Carboxymethyl cellulose based Caseinate /beeswax Caseinate/stearic acid	Howard and Dewi (1995) Sargent et al. (1994) Avena-Bustillos et al. (1994a) Avena-Bustillos et al. (1993)
Avocados	Oxygen barrier Carbon dioxide barrier	Delay the onset of ripening	NatureSEAL™ (polysaccharide based film)	Bender et al. (1993)
Bell peppers/cucumbers	Moisture barrier	Chitosan: antifungal activity	Whhey protein isolate Sodium caseinate Sodium caseinate/beeswax	Lerdthanangkul and Krochta (1996) El Ghaouth et al. (1991)

Product example	Critical functions of packaging	Value added function	Examples of materials	References
Tomatoes	Moisture barrier Gas barrier	Uniform colour development Suppression of ripening	Chitosan Corn zein Durkex 500 (non-lauric vegetable oil) TAL Pro-Long (sucrose ester of fatty acids and sodium salts of carboxy methyl cellulose) Starch, cellophane, PHB coated paperboard tray overwrapped with starch bag, PLA coated paper-board over wrapped with starch bag Semperfresh™ FLO (sucrose esters)	Park et al. (1994) Nisperos and Baldwin (1988)
Loquat fruits	Moisture barrier			Helén (2000)
Beans	Moisture barrier		Cellulose based with polyethylene glyco, stearic acid, palmitic acid and lauric incorporated	Ayranci and Tunç (1997)
Strawberries	Moisture barrier	Improved sensory quality Retard senescence	Cellulose based with polyethylene glycol, stearic acid, palmitic acid and lauric incorporated Starch based	Ayranci and Tunç (1997) García et al. (1998)
Celery sticks	Moisture barrier		Caseinate-acetylated monoglyceride film	Avena-Bustillos et al. (1997)
Zucchini	Moisture barrier		Semperfresh™ (sucrose esters of fatty acids, mono- and diglycerides and sodium salts of carboxymethylcellulose) Calcium caseinate-acetylated monoglyceride	Avena-Bustillos et al. (1994b)
Satsume mandarins	Moisture barrier Oxygen barrier		Semperfresh™ Jonfresh™ (carnauba wax/shellac)	Bayindirli et al. (1995) D'Aquino et al. (1996)
Product example	Critical functions of packaging	Value added function	Examples of materials	References
Bananas	Carbon dioxide barrier Oxygen barrier Carbon dioxide barrier	Retard ripening	Prolong (sucrose esters and sodium carboxymethyl cellulose)	Banks (1985)
Pears	Moisture barrier Oxygen barrier Carbon dioxide barrier		Corn zein Semperfresh™	Park and Jo (1996)
Cabbage	Oxygen barrier	Reduction of browning by reducing oxygen permeability and increasing ethanol production	Sucrose fatty esters	Sakana et al. (1990)
Cut fruit	Oxygen barrier Moisture barrier	Antioxidant (prevent browning)		
SNACKS				
BIOBASED PACKAGING				
Potato chips	Light barrier Moisture barrier Oxygen barrier		Methylcellulose laminated with corn zein and stearic-palmitic acid	Park et al. (1996)
Confectionery	Moisture barrier		Cellophane	UCB-films, Belgium
Ice cream	Moisture barrier	Control of water migration	Acetylated monoglycerides and chocolate	Anon. (2000)
EDIBLE COATING				
Roasted peanuts	Oxygen barrier		Whey protein isolate Hydroxypropyl cellulose Zein	Maté et al. (1996) Ramos et al. (1996)
Nuts	Oxygen barrier Moisture barrier	Antioxidant (time-dependent migration)	Fats and waxes	Stuchell and Krochta (1995)
Pasty/topping Dessert topping/biscuit base	Moisture barrier	Thickening agent and control of water migration		Kamper and Fennema (1985) Silva et al. (1981)

Product example	Critical functions of packaging	Value added function	Examples of materials	References
DRY PRODUCTS				
BIOBASED PACKAGING				
Bread	Moisture barrier	Anti fogging	Starch laminate Starch Paper bags coated with biobased plastics, Window of PLA or starch (standard technology) Starch, PLA, coated cellophane Starch laminate Starch	Van Tuil (2000) Bastioil (2000) Holton et al. (1994) Helén (2000) Van Tuil (2000) On the market in Italy
Cruesli Dry pasta				
EDIBLE COATING				
Croutons/salad	Moisture barrier	Thickening and control of water migration	Fats and acetylated monoglycerides	Kamper and Fennema (1985) Silva et al. (1981) Seaborne and Ebberg (1989) Fennema et al. (1987)
Biscuit crumbyoghurt	Moisture barrier	Thickening and control of water migration	Fats and acetylated monoglycerides	Kamper and Fennema (1985) Silva et al. (1981) Seaborne and Ebberg (1989) Fennema et al. (1987)
OTHER				
Carrier bag			Starch	On the market in Finland and Italy
Garbage bag		Biodegradable	Wheat and maize starch + PCL	On the market in Denmark

Table 3.4 Potential food product applications of biobased packaging and edible films/coatings

Product example	Critical functions of packaging	Value added function	Examples of materials	References
FRESH MEAT PRODUCTS				
BIOBASED PACKAGING				
Fresh meat	High moisture absorption, high gas permeability (oxygen), high moisture barrier	Absorption of meat drip	Starch based drip pad Protein film Trays made from fresh pulp, starch, PLA and/or PHB/V Top lids produced from PLA, cellulose acetate or cellophane	
EDIBLE COATING				
		Reduction of oxidation (WOF), antioxidant release and moisture barrier	Different edible coatings combined with antioxidants	
READY MEALS				
BIOBASED PACKAGING				
	High moisture and gas barrier	Compostability for institutional foods	PHB film PHB tray Paperboard coated with PHB	
EDIBLE COATING				
Pizza base/sauce	High moisture barrier and heat resistance	Reduction of water migration and softening of the base	Edible film/coating of e.g. algininate or pectin between base and sauce	Kamper and Fennema (1985) Silva et al. (1981) Seaborne and Ebberg (1989) Fennema et al. (1987)
	High gas barrier (oxygen and carbon dioxide) and heat resistance	Reduction of oxidation (WOF), antioxidant release and moisture barrier	Different edible coatings combined with antioxidants	
DAIRY PRODUCTS				
BIOBASED PACKAGING				
Milk	High moisture, light	Moisture and oxygen barrier	PLA bottles	

Product example	Critical functions of packaging	Value added function	Examples of materials	References
Hard cheese	(and gas) barrier		PHBV bottles Paperboard cartons coated with PLA and/or PHBV	
	High moisture barrier, light and gas barrier (oxygen and carbon dioxide)	Higher carbon dioxide permeability compared to oxygen permeability	PLA and other biobased/biodegradable material with high carbon dioxide permeability compared to oxygen permeability	Södergård (2000)
Yoghurt, feta cheese, sour cream, quark, cottage cheese, processed cheese	High mechanical strength, high gas barrier (oxygen, carbon dioxide and light)	Mechanical strength and moisture barrier	Cardboard coated with a mixture of biobased/biodegradable materials PLA	
EDIBLE COATING		Moisture barrier	mcl-PHA latex	Van der Walle et al. (2000)
BEVERAGES				
BIOBASED PACKAGING	Acid resistant, inert to migration of flavour compounds (scalping), high moisture, (gas), (light), and aroma barrier, inert towards microorganisms	Resistance to scalping Moisture and oxygen barrier	PLA bottles/cups PHBV bottles/cups PLA coated with PHBV Paperboard coated with PHBV, starch or PLA	Haugaard and Festersen (2000)
FRUITS AND VEGETABLES				
BIOBASED PACKAGING	High mechanical strength, flexible, not opaque, balanced gas and moisture barrier, protection against crushing/bruising		Biobased stock in carriers Perforated PLA films Starch based trays Perforated cellohane films Perforated cellulose acetate films Gluten-based films	
Mushrooms			PLA films	Haugaard and Festersen (2000)
EDIBLE COATING		Moisture barrier, prevention of microbial growth and oxidation	Wheat gluten, pectin, beeswax	
SNACKS				
BIOBASED PACKAGING	High moisture, oxygen, and light barrier, protection against crushing		Whey protein isolate Hydroxypropyl cellulose Zein Paperboard coated with e.g. PHBV, PLA or starch	Maté et al. (1996) Ramos et al. (1996)
FROZEN PRODUCTS				
BIOBASED PACKAGING	High moisture, oxygen, and light barrier	Prevention of lipid oxidation Moisture and oxygen barrier Light barrier	Biobased materials incl. pigment	
EDIBLE COATING		Moisture, oxygen, and light barrier Prevention of moisture loss	Materials based on e.g. PLA, PHBV, or modified starch Cardboard coated with PHBV Zein Different edible coatings	Padua et al. (2000)
DRY PRODUCTS				
BIOBASED PACKAGING	High moisture and oxygen barrier	Moisture, oxygen, and light barrier	Cardboard and paper board coated with biobased materials (e.g. PHBV or PLA)	

ned by using oxygen permeable films in the packaging process. On the other hand, oxygen also supports the growth of bacteria, and discoloration, attributed to the brown pigment metmyoglobin, occurs rather quickly. This surface discoloration is even more pronounced in ground meats where the exposed surface area is hugely increased. As a result of its high water activity, unprotected chilled meat will lose weight by evaporation and its appearance will deteriorate (Robertson, 1993). Thus, low water vapour permeability is important in packaging of fresh meat. In cured meat products the pigment nitrosylmyoglobin oxidizes rapidly in the presence of light and oxygen. The onset of oxidative rancidity is also accelerated in the presence of light and oxygen. Thus, low permeabilities to oxygen and light are required of packaging materials for cured meat products. Raw poultry support microbial growth due to its high pH (5.7-6.7). Hence, packaging in modified atmospheres with a high level of CO₂ or vacuum extends shelf-life considerably. The myoglobin content of poultry is much lower than in beef and in other meats resulting in a relatively high colour stability of the product (Taylor, 1996).

Fresh meats are typically packed in oxygen permeable packs, vacuum packs or modified atmosphere packs. Where residual oxygen must be maintained at a very low level, vacuum packaging minimizes the colour and flavour defects associated with oxidation of muscle myoglobin and lipids, respectively. Modified atmosphere packaging (MAP), with 70-80% O₂ to maintain oxymyoglobin and 20-30% CO₂ to inhibit microbial growth is commonly used to package fresh red meats. "White" meats, such as poultry meats, are often packed in a mixture of CO₂ and N₂. However, some authors point out that more than 25% CO₂ may cause discoloration and off-flavour formation in poultry (Bartkowski et al., 1982). The "snug-down" effect obtained at high CO₂, when the CO₂ is dissolved in the water phase, is undesirable for several products giving the package a vacuum packaged look. It is possible to prevent the "snug down" effect by using N₂ in the gas-mixture (Parry, 1993). Active packaging, involving the use of oxygen-absorbing sachets, has been found to be useful in reducing photo-oxidation in cured meat products (Andersen and Rasmussen, 1992).

Packaging methods

Conventional packaging materials

Permeable films (PVC, PE-based, polyolefin based), PS, expanded PS, PETG, PA or PET or PVC/PVC or PVdC coating/LDPE or EVA or ionomer (Robertson, 1993), Saran (copolymer of PVdC and PVC).

Potential biobased materials

Starch is hygroscopic in nature and starch-based absorbent pads are expected to provide a potential alternative to the conventional absorbent pads for meat exudation. To avoid drying out it is important to design the pads for the specific product. Many biobased packaging materials have a relatively high oxygen permeability which would make them suitable for packaging fresh meats packed in air. A suggestion for biobased packaging of such meat products is a combination of a film based on plasticized proteins which has a high oxygen permeability, e.g. wheat gluten, whey (see Chapter 2), and a tray based on starch, pulp, PLA and/or PHB/V. Lids may be produced from PLA, cellulose acetate or cellophane. Furthermore, coating of meat might be useful for the reduction of oxidation (see Table 3.3).

3.5.2. Ready meals

The major challenge encountered with the shelf-life of ready meals arises from their heterogeneity (Labuza, 1982). The shelf-life of ready meals in chill-storage is largely determined by the extent of oxidative changes and the growth of microorganisms. To reduce deteriorative reactions in ready meals it is recommended to use packaging materials with low oxygen permeability and water vapour permeability. In cooked meats, oxidative changes occur rapidly and lead to the formation of the characteristic off-flavour described as "warmed over flavour" (WOF) (Stapelheldt et al., 1993). MAP with nitrogen to replace oxygen and carbon dioxide to inhibit microorganisms is often applied. Exclusion of oxygen is also important in pre-cooked frozen foods where lipid oxidation is a major contributor to deterioration (Labuza, 1982).

Packaging methods

Packaging methods include vacuum packaging, modified atmosphere packaging (e.g. 30% CO₂/70% O₂), and packaging in atmospheric air (Stapelheldt et al., 1993).

PE or laminate foil with low oxygen permeability is used (Stapel-feldt et al., 1993). Cardboard is often used around the primary packaging to protect the packaging from mechanical injuries during transportation and handling.

Vacuum packaging and MAP require packaging materials with low permeability towards gases. Available biobased packaging materials do not possess these properties and further optimization of the materials is required in order to make them useful for ready meals. A flexible film wrapped around a tray, both based on PHB, may be an option because of the relatively low oxygen (O₂) and water vapour permeabilities of this material (Hänggi, 1995; Krochta and De Mulder-Johnston, 1997). Paperboard coated with PHB might be a potential biobased packaging material for ready meals in the future. Unfortunately, coating of paper or board with PHB or other biobased materials is more difficult than coating with PE. This problem is caused by the lower adhesion of the biobased materials to paper or board. Furthermore, the sealing window for biodegradable plastics is more narrow than for PE with the result that, in addition to the higher material cost, the requirements of the packaging line are higher. These problems need to be solved before paperboard coated with biobased materials will offer an alternative to conventional packaging for food applications. PHB is presently not flexible enough for forming films or foils. PHB also tends to become brittle and to lose water vapour barrier properties (Hänggi, 1995). Thus, use of PHB in films or foils requires further process optimization before being considered for ready meals. Since ready meals often require re-heating, heat resistance, to allow heating of the food directly in the pack, is an additional need. The requirement for partitioning between food constituents in ready meals would appear to present a range of potential applications for edible films and coatings. For example, an edible film/coating, composed of alginate or pectin between the base and the sauce component of pizza, could reduce water migration between the sauce and base or edible coatings containing antioxidant components could reduce WOF in cooked meats. Since most biobased packaging materials are compostable, distribution of ready meals in compostable trays might be appealing in closed systems such as hospitals and residential homes for elderly people, because of the possibility of composting both food and trays directly after use.

Conventional packaging materials

Potential biobased packaging materials

Packaging methods

Conventional packaging materials

3.5.3. Dairy products

Milk, cream, fermented milk products, and processed cheese require low oxygen permeability packaging to avoid oxidation and growth of undesirable microorganisms. In addition, light initiates the oxidation of fats in dairy products and leads to discoloration, off-flavour formation and nutrient loss, even at temperatures found in refrigerated display cabinets. The oxidative reactions initiated by light may continue even if the products are subsequently protected from light. Dairy products should be protected from water evaporation, absorption of odours from the surroundings and high storage temperature to maximize shelf-life.

Different packaging technologies apply to different products. Thus, cold filling is used for milk, cream and fermented products, aseptic packaging is used for UHT milk, hot filling is used for butter and yoghurts, MAP packaging is used for milk powder, MAP packaging and hot filling is used for cheese. Several researchers have recommended fresh cheeses (e.g. cream cheese, decorated cream cheese, soft cheese, cottage cheese) to be packaged in modified atmospheres with N₂ and/or CO₂ replacing the O₂ in the package (Mannheim and Soffer, 1996; Fedio et al., 1994; Moir et al., 1993). However, spoilage caused by yeast and especially bacteria may still occur even at very low O₂ and elevated CO₂ levels (Westall and Filtenborg, 1998). Semi-soft and hard cheeses (whole, sliced, or shredded) have a relatively high respiration rate requiring a packaging material somewhat permeable to CO₂ to avoid blowing of the packaging. Meanwhile, O₂ must be kept out to avoid fungal spoilage and oxidation of the cheese. Mould ripened cheeses, such as white cheeses (Brie/Camembert) and blue-veined cheeses (Danablu/Roquefort), contain active fungal cultures. As a consequence, the O₂ content should not be too low as this may cause anaerobic respiration and production of off-flavours. Instead, these products require a balanced oxygen and carbon dioxide atmosphere tailored to each product to prolong shelf-life (Haasum and Nielsen, 1998; Nielsen and Haasum, 1997).

The packaging materials commonly used include: glass, PE-coated paperboard, plastic containers (HDPE) for milk; plastic containers, PE-coated paperboard cartons/with or without aluminium for UHT milk; plastic tubs (PS or PP)/aluminium foil heat

sealed to the rim of the container, PE-coated paperboard, glass bottle with foil cap, blow-moulded PE containers sealed with a close fitting plastic cap for cream; aluminium foil/greaseproof paper or vegetable parchment, paper, parchment, plastic tubs (PS or PVC) with a tight-fitting lid of the same material for butter; PA/PE, APET, PET or PVC/PVC or PVdC coating/LDPE or EVA or ionomer, PS, PP for cheese; air tight packages, metal cans, aluminium foil/plastic laminates with paper for milk powder.

PLA or PHB/V bottles or paperboard cartons coated with PLA or PHB/V could be used as packaging materials for milk because of their high moisture and oxygen barrier properties compared to the conventional HDPE bottles and PE-laminates. Exclusion of light from the bottles may be obtained by adding e.g. pigments to the polymer blend. Since cheeses respire, packaging materials with relatively high carbon dioxide permeability are required in order to avoid inflation of the packages. Compared to packaging materials conventionally used, biobased materials have relatively higher carbon dioxide permeability (O_2 : CO_2 permeability ratio of 1:7-14 for biobased materials and 1:4-5 for conventional materials has been found in a Danish project on biobased materials for foods (Biologically Based Packaging Materials for Foods; The Directorate for Food, Fisheries and Agro Business). Thus, packaging of cheese in biobased packaging, e.g. PLA, could be feasible. Cardboard coated with a mixture of biobased/biodegradable materials to obtain the proper mechanical and barrier properties is suitable for yoghurt, feta cheese, sour cream, fromage frais, cottage cheese or processed cheese. However, the lower adhesion of biobased materials remains an issue. Application of medium-chain-length PHA latex as cheese coating for prevention of moisture loss due to a low water vapour permeability is reported to be an alternative to conventional used cheese coatings (Van der Walle et al., 2000).

3.5.4. Beverages

Factors limiting the shelf-life of beverages include microbial growth, migration/scalping, oxidation of flavour components, nutrients and pigments, non-enzymatic browning, and, in the case of carbonated beverages, loss of carbonation. Thus, requirements of the packaging materials for beverages include low gas transmission and light permeabilities and resistance towards scalping (migration from food product to package). Packaging

Potential biobased packaging materials

Packaging methods

Conventional packaging materials

Potential biobased packaging materials

materials with high water vapour barrier properties are required to prevent penetration of the beverage through the package. For packaging of acidic beverages the material must be resistant to acids (Petersen et al., 1999).

Packaging methods for packaging of beverages include aseptic packaging with or without nitrogen injection (Sizer et al., 1988), hot (McLellan et al., 1987) and cold filling.

The packaging materials commonly used include: glass, HDPE, PP, PC, PET, PVC, PE/paper/PE/Al/PE, PE/paper/PE/Al/special coating (gable top packaging types) for water; glass, metal, HDPE, PE/paper/PE/EVOH/PE, PE/paper/PE/SiOx/PE, PE/paper/PE/Al (gable top packaging types) for juice; glass, metal, PET for carbonated soft drinks; glass, metal, PET for beer.

A Danish project on packaging fresh unpasteurized orange juice suggests that PLA and PHB bottles or cups could be used for packaging beverages (Haugaard and Festersen, 2000). The results showed PLA-cups to have relatively low water vapour permeability and high resistance to scalping compared to PE. Since PHB has a much lower oxygen transmission rate than PLA (Krochta and De Mulder-Johnston, 1997) and PHB has high water resistance (Hänggi, 1995), coating of PLA with PHB is expected to give a useful biobased packaging materials for beverages. Packaging materials based on 100% PHB are also expected to be useful for beverages. Paperboard coated with PHB, PLA or modified starch in order to improve the moisture and oxygen barrier properties of the paperboard could also be of potential use in packaging beverages. While the barrier properties of the suggested biobased packaging materials may match the beverage requirements, the mechanical strength is at present insufficient for the production of gable top type packaging materials.

3.5.5. Fruits and vegetables

Fruits and vegetables continue to respire, transpire and produce the ripening hormone ethylene even after harvesting with the result that concentrations of carbon dioxide, oxygen, water and ethylene change over time inside storage packs. Changes in gas composition may have a positive influence on the colour and flavour of the products, but they may also induce negative effects on texture, colour, shelf-life and nutritional quality (Lee et al.,

1995). Short-term preservation by reducing respiration and transpiration rates can be obtained by controlling factors such as temperature, relative humidity, gas composition (ethylene, oxygen and carbon dioxide), light, and by applying food additives and treatments such as waxing and irradiation. Physical damage (e.g. surface injuries, impact bruising) may stimulate respiration and ethylene production and accelerate the onset of senescence. The choice of proper packaging material is complex because it depends on the specific respiration and transpiration rates of the different products and the conditions in the supply chain. If the chosen packaging material is impermeable to CO₂, O₂ and H₂O, an anaerobic environment inside the packaging will develop and lead to microbial fermentation and product deterioration. If the packaging material is too permeable to water vapour, the products will dry out and the atmosphere in the packaging will contribute to a reduced storage life. The ideal packaging material has a permeability that takes the respiration processes of the products into account so that the atmospheric balance (CO₂/O₂ ratio) inside the packaging is optimal (Yam and Lee, 1995; Day, 1993). The packaging material should retain desirable odours, prevent odour pick-up, provide protection from light and give sufficient protection against mechanical damage.

Reduction of the O₂ content to less than 10% by using a passive or active modified atmosphere in the packaging (e.g. rigid tray wrapped in or sealed with plastic films) provides a tool for controlling the respiration rate and slowing down senescence although an adequate O₂ concentration must be available to maintain aerobic respiration. Packaging with bags, incomplete sealing or perforation of packages, individual shrink wraps, or bulk display where the consumers pick the product themselves, are used for fruits and vegetables.

Among the packaging materials used for fruits and vegetables are: monolayer PVC, perforated thin LDPE, LDPE/MDPE with EVA, kraft paper, LDPE, HDPE, white pigmented PVC or PP, expanded (foamed) PS, LLDPE, shrinkable film, regular net stocking or expanded (foamed) plastic netting, PET, moulded paper pulp with a thermoformed plastic liner, sleeve packs.

A potential application is net stocking carriers made of biobased materials for fruit and vegetables. Even though the gluten-based

Packaging methods

Packaging methods

Conventional packaging materials

Conventional packaging materials

Potential biobased packaging materials

Potential biobased materials

film has not yet been approved for food contact it may be a potential biobased packaging material for products such as mushrooms since it will provide beneficial MAP conditions and has a low price compared to conventional packaging materials. The properties of PLA-based materials may also be well-matched with the requirement of mushrooms for relatively high water vapour permeability and relatively low gas permeability (Haugaard and Festersen, 2000). Other possibilities for fruits and vegetables in general include perforated PLA, cellulose acetate and cellophane films wrapped around starch-based trays. In preventing microbial growth, oxidation or loss of moisture edible coatings from, e.g. wheat gluten, pectin, and beeswax could be used.

3.5.6. Snacks

The most common modes of deterioration of snack foods are loss of crispiness and development of fat rancidity. Thus, low water vapour and oxygen permeabilities are of the utmost importance. Mechanical strength is required of packages for snack foods and the exclusion of light has also been suggested (Quast and Karel, 1972).

Most snack foods are packaged by form fill sealing (Matz, 1993). For some snack products the air is removed and packages are flushed with nitrogen gas to protect against moisture absorption and retard the development of rancidity (Labuza, 1982).

Fried, extruded, and puffed snack foods are typically packaged in multi-layer structures. Packaging materials are usually pigmented, metallized, or placed inside paperboard cartons (Robertson, 1993). Spiral-wound, paperboard cans lined with aluminium foil or a barrier polymer are used for e.g. chips and nuts. In addition, metal cans are used for fried nuts; the container usually being gas flushed with nitrogen.

As shown in Table 3.3, biobased packaging materials based on whey protein isolate, hydroxypropyl cellulose, and zein have already been investigated for roasted peanuts (Maté et al., 1996; Ramos et al., 1996). As a result of the requirements for low permeability towards oxygen and water, paperboard coated with PHB, PLA, or modified starch for example, could potentially be used as biobased packaging materials for snacks traditionally packed in paper cans or cartons. In order to minimize penetra-

tion of light, the biobased materials could be combined with titanium oxide or other pigments.

3.5.7. Frozen products

The common modes of deterioration in frozen foods are pigment and vitamin degradation and oxidation of lipids (Petersen et al., 1999). Thus, requirements of packaging materials for frozen products include a high moisture barrier property to reduce moisture loss and freezer burn and oxygen and light barrier properties for protection against oxidation (Bak et al., 1999; Robertson, 1993; Christophersen et al., 1992). The packaging material should be resistant to tearing and puncturing (Labuza, 1982). For common polymeric films, satisfactory water vapour transmission rates are obtained at freezer temperatures below -20°C (Robertson, 1993). However, at the low temperature mechanical properties may be affected making the polymeric materials more brittle and sensitive to mechanical forces.

Most frozen products are packaged in air, with exception of fatty fish which is vacuum packaged or packaged in nitrogen (Labuza, 1982).

The majority of frozen fruits and vegetables are packed in polymeric films the major component being LDPE. Some films contain white pigments to prevent light penetration. Other conventional materials include waxed carton-board wrapped in a moisture-proof regenerated cellulose film and folding cartons with a hot melt coating of PVC/PVdC copolymer (Robertson, 1993). Films and wraps used for meat and seafood include cellophane, aluminium foil, PVdC, PE, and PS trays surrounded by films and wraps, and coated paper and cartons (Labuza, 1982).

Biobased packaging materials can be used for frozen products if their permeabilities under low temperature conditions can be reduced to levels comparable to those of conventional packaging materials. Packaging materials based on corn zein (Padua et al., 2000), PLA, PHB/V or for example modified starch might then be of potential use for frozen products. As an example, cardboard coated with PHB/V in which the cardboard gives low light transmittance and PHB/V gives medium transmittance of gas and low water vapour transmittance could be applied to frozen food products. PHB/V is not as brittle when folded as PLA, and it has a

higher adhesion to paper. Edible coatings could also find application in the prevention of water loss from frozen products.

3.5.8. Dry products

The most critical factors for dry products in relation to packaging are moisture uptake leading to loss of crispiness and oxidation of fats resulting in development of rancidity. Other modes of deterioration include oxidation of vitamins, breakage of products, loss of aroma, discoloration, mould growth, staling, and fat bloom depending on the product. Thus, the most important requirements for the packaging materials include high moisture, oxygen, and light barrier properties and high mechanical strength.

Most dry products are packaged under atmospheric conditions. Commercialization of MAP for bakery products is widespread in Europe whereas it is more seldom seen in the rest of the world.

Packaging materials for dry products include: underground pits or containers, piles of bagged grains and storage bins of different sizes, shapes and construction types for grains; bags, bulk bins, multi-walled Kraft paper bags, sometimes with an LDPE liner for flour; paperboard carton with a plastic window (cellulose acetate), OPP or coated LDPE films for dried pasta; LDPE bags in which the end is twisted and sealed with a strip of adhesive tape or perforated LDPE bags for bakery products; regenerated cellulose films coated with LDPE or PVC/PVdC copolymer and often with a layer of glassine in direct contact with the product if it contains fat, for biscuits; cookies and crackers, aluminium foil/LDPE sometimes containing a layer of paper, either between the foil and the LDPE or on the outside of the foil, PVC/PVdC copolymer/LDPE, molded PVC trays wrapped in aluminium foil or placed inside paperboard boxes or metal or glass containers for chocolate.

There are many opportunities for using biobased materials for dry products especially because the materials have relatively high water vapour barriers. Board and paper coated with biobased materials, e.g. PLA or PHB, are expected to be very useful for dried foods. Board and paper confer mechanical strength thus protecting products from breakage.

Packaging methods

Conventional packaging materials

Packaging methods

Conventional packaging materials

Potential biobased packaging materials

Potential biobased packaging materials

3.6. Conclusions and perspectives

Packaging of foods is a challenging task because food materials are complex and diverse. Using biobased materials to package foods brings additional challenges since biobased materials themselves possess diverse characteristics. To date, published information of the development of biobased packaging solutions for foods has primarily been the focus of academics as indicated by the state-of-the-art findings presented in this study. Food packagers have not used biobased materials for a number of reasons among them being a lack of knowledge about the materials themselves and their compatibility with existing packaging technology, an inability to recoup the additional cost of using biobased materials in large scale product packaging, and a reluctance to face the legislative hurdles that need to be overcome to permit the use of biobased materials. However, food manufacturers in collaboration with producers of biobased packaging materials are now testing biobased packaging for specific products. Availability of the raw materials for production of biobased materials, including PLA, at more favourable costs, will increase in the near future and food products packaged in biobased materials are likely to be introduced into the market place in the coming years.

Potential applications of biobased materials for specific food products have been identified using the product as a starting point. Product categories with the potential to utilize biobased materials include meat and dairy products, ready meals, beverages, snacks, dry products, frozen products and fruits and vegetables. In the short term, biobased materials will most likely find application in foods requiring short term chill storage, such as fruits and vegetables since biobased materials present opportunities for producing films with variable carbon dioxide/oxygen and moisture permeabilities. However, to succeed, biobased packaging of foods must be in compliance with the quality and safety requirements of the food product and meet legal standards and should preferably enhance the value of the product to justify any extra material cost. In this context, shelf-life testing is vital along with testing of durability and migration and verification of consumer acceptance of the packages. Close dialogue between food scientists and the manufacturers of biobased packaging materials is imperative if biobased materials are to make a significant impact on the food packaging sector.

3.7. References

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4. Safety and food contact legislation

4.1. Introduction

Packaging serves as a major defence against external hazards and normally gives a high level of protection. However, undesirable interactions between food and packaging materials can give rise to potential problems which effectively can be dealt with by careful design and construction of packages. The most well-known and undesirable interaction is migration of packaging components to the food and the food contact material legislation has been developed to deal with this problem. Other undesirable interactions are usually less likely to occur. These include microbiological contamination of packages, penetration of microorganisms, insects and rodents through packages and the collapse of packages under humid conditions. Microbiological contamination is dealt with by means of good manufacturing practice guidelines.

Both conventional and biobased materials are treated in exactly the same way in the European food contact material legislation and good manufacturing practice guidelines. However, due to differences in origin and properties between conventional and biobased materials some undesirable interactions are more relevant for one than for the other.

All current applications of biobased materials as food contact materials comply with European legislation. This fact clearly proves that biobased materials are as safe as conventional materials.

In this chapter the European legislation for food contact materials is clarified and relevant undesirable interactions for biobased packages are assessed by literature study. Finally, conclusions are drawn and recommendations are made.

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4.2. Biobased materials and legislation on food contact materials

Migration is an undesirable interaction between food and packaging that is actually caused by materials and articles coming into contact with food. Chemical contamination of food has become the driving force to prepare food legislation in the industrialized countries. Some biobased materials are "old" and well-defined, like paper and regenerated cellulose, and legislation on a harmonized EU or national level exists. But "new" materials have also been developed and the producer is responsible as to ensuring the safety and suitability for food contact. The safety of food contact materials is evaluated by considering the identity, toxicological properties and quantities of substances that migrate from the material into food during conditions of intended use. Biobased materials are treated in exactly the same way as conventional materials in this respect. Since "edible coatings" are by definition meant to be consumed, they are regarded as part of the food product and must fulfil the requirements in the legislation on foods. Some types of active packaging are also designed to add substances into the food bringing them into an intermediate area where legislation is not well-defined.

4.2.1. Common EU legislation

At the end of the 1950's, the German and Italian authorities issued their first regulations in the field of migration, followed by others. In the European Community the differences in the regulations soon began to create problems for packaging companies, which were forced to adjust their production to the country of destination. This adjustment led to the need of harmonizing the laws in order to remove trade barriers. European Union legislation has five main instruments: Regulations, Directives, Decisions, Recommendations, and Opinions. So far, almost all legislation relating to migration has been in the form of directives. A directive may be simply enacted by the national parliament, practically unchanged, but significant changes are often necessary to fit the style of national legislation and procedures.

The Commission drew up a Framework Directive setting out the principles, listing the materials to be regulated, and defining the procedures for adoption of new materials. The main principle of the Community legislation focus on preventing the migration of toxic substances from reaching unacceptable levels as well as on

maintaining the integrity of the foodstuff thereby preventing contamination which may change the composition and sensory properties of the food. The list of materials to be regulated by the EC is as follows:

1. Plastics, including varnishes and coatings
2. Regenerated cellulose
3. Elastomers and rubber
4. Paper and board
5. Ceramics
6. Glass
7. Metals and alloys
8. Wood, including cork
9. Textile products
10. Paraffin and microcrystalline waxes

Regenerated cellulose and ceramics were dealt with first while the work on plastics is still ongoing. The plastic directives do not yet cover varnishes and surface coatings.

There are certain essential criteria that are expressed either in the Framework Directive or in specific directives. Although these are currently applied to plastics, it is worth taking note of them because of their likelihood of being basic principles for all other materials (Rossi, 1994).

1. Plastics must be produced by good manufacturing practice.
2. Plastics must not transfer their constituents to foodstuffs in such quantities as to constitute a health hazard.
3. Plastics must not transfer their constituents to foodstuffs in such quantities as to bring about an unacceptable change in the composition of the foodstuff (overall migration limit).
4. Plastics must not transfer constituents to foodstuffs in such quantities as to alter their sensory properties.
5. Plastics must be made from starting substances listed in the plastic directives.
6. Starting substances not listed can be used on condition that they are mixtures of approved substances, oligomers, or natural or synthetic macromolecular compounds or mixtures of the two as long as they have been produced from starting substances included in the list.

7. Authorized substances can be used only if they comply with restrictions applicable to them.
8. The substances must be "of good technical quality as regards purity requirements".
9. A symbol or the words "for food use" must accompany plastics sold to consumers that are not in contact with foods, but intended to come into contact with foods. Articles that by nature are clearly intended to come into food contact are exempted from these obligations.

A comprehensive list of directives adopted on materials intended to come into contact with foodstuffs grouped by subject is presented in Table 4.1. More information is found at Internet site: <http://cpf.jrc.it/webpack/>

Table 4.1. List of Directives adopted on materials intended to come into contact with foodstuffs.

89/109/EEC New framework Directive provides the framework for directives on all kinds of materials and articles intended to come into contact with foodstuffs. As such, they authorize the Commission to prepare directives for individual materials. The basic idea of food contact material legislation is formulated in Article 2: "Materials and articles must be manufactured in compliance with good manufacturing practice so that, under their normal or foreseeable conditions of use, they do not transfer their constituents to foodstuffs in quantities, which could: i) endanger human health, ii) bring about an unacceptable change in the composition of the foodstuffs or a deterioration in the organoleptic characteristics thereof."

80/590/EEC Symbol for materials and articles defines the symbol indicating that the material is intended for use in contact with foodstuffs, and may be shown on food contact materials.

93/10/EEC Cellulose regenerated and 93/11/EC Regenerated cellulose: amending directive 93/10/EEC deals with regenerated cellulose listing of substances used in the manufacture. When necessary, due to their toxicological properties, compositional limits were set on certain substances. Migration limits were stipulated for two substances: the specific migration limit for monoethyleneglycol and diethyleneglycol is 30 mg/kg of foodstuff.

82/711/EEC Plastics: basic rules for testing migration, 93/8/EEC Plastics: amending directive 82/711/EEC, 97/48/EC Plastics:

amending Directive 82/711/EEC. These directives present the test conditions to be used corresponding to the actual usage conditions. Under certain provisions, the use of substitute and alternative test media are allowed in order to demonstrate compliance.

85/572/EEC Plastics: list of simulants for testing migration. This directive lists foodstuffs together with the appropriate food simulant to be used in the migration testing for each foodstuff. The four simulants are distilled water, 3% acetic acid, 10% ethanol and olive oil. In some cases, the extractive capacity of olive oil is greater than that of actual foodstuffs in some cases, and the reduction factors may be applied.

90/128/EEC Plastics: monomers, 92/39/EEC Plastics: amending directive 90/128/EEC, 93/9/EEC Plastics: amending directive 90/128/EEC, 95/3/EC Plastics: amending Directive 90/128/EEC, 96/11/EC Plastics: amending Directive 90/128/EEC, 1999/91/EC Plastics: amending Directive

89/109/EEC	New framework Directive
80/590/EEC	Symbol for materials and articles
93/10/EEC	Cellulose regenerated
93/11/EC	Regenerated cellulose: amending Directive 93/10/EEC
93/11/EEC	N-nitrosoamines from teats and soothers
84/500/EEC	Ceramic articles
82/711/EEC	Plastics: basic rules for testing migration
93/8/EEC	Plastics: amending Directive 82/711/EEC
97/48/EC	Plastics: amending Directive 82/711/EEC
85/572/EEC	Plastics: list of simulants for testing migration
90/128/EEC	Plastics: monomers
92/39/EEC	Plastics: amending Directive 90/128/EEC
93/9/EEC	Plastics: amending Directive 90/128/EEC
95/3/EC	Plastics: amending Directive 90/128/EEC
96/11/EC	Plastics: amending Directive 90/128/EEC
1999/91/EC	Plastics: amending Directive 90/128/EEC
89/397/EEC	Official control of foodstuffs
93/99/EEC	Amending directive 89/397/EEC

90/128/EEC. These directives are called the plastic directives. The directive gives the overall migration limit for plastic (60 mg/kg foodstuff or 10 mg/dm²). The substance list is divided in sections and includes specific migration limits for a quite large number of substances (more than 70). It must be stressed that the directives refer to materials exclusively consisting of plastics. In the directives, "plastics" shall mean the organic macromolecular compounds obtained by polymerization, polycondensation, polyaddition or any other similar process from molecules with a lower molecular weight or by chemical alteration of natural macromolecules. "Other substances or matter may be added to such macromolecular compounds". According to this definition, certain biobased materials will be classified as plastics. Surface coatings, lacquers and other material combinations with plastics are not covered at present, but they will be so in due course.

Directive 1999/91/EC adds new annexes to the plastic directive allowing, under "Products obtained by means of bacterial fermentation", the use for food contact material of 3-hydroxybutanoic acid-3-hydroxypentanoic acid copolymer produced by the controlled fermentation of *Alcaligenes eutrophus* using mixtures of glucose and propanoic acid as carbon sources. Certain restrictions and purity requirements are given.

A committee within the Council of Europe prepares recommendations for paper and board materials. When completed, the Council of Europe Resolution will form the basis for a forthcoming directive.

4.2.2. *Biobased materials*

The European Commission document called "Practical Guide" gives information and guidelines to those who use the Directives on materials and articles intended to come into contact with foodstuffs. See Internet site: <http://cpf.jrc.it/webpack/>. In the chapter on positive lists for plastics, it is mentioned that the concept of Community lists could also be applied to lists related to other materials, e.g. paper. A positive list is "a list of the substances the use of which is authorized to the exclusion of all others". The Commission has chosen to list all the substances deliberately used in the manufacture of the finished material. An authorization should be requested ("petition") for new substances to be added to the list. The list established for plastics is restricted to

monomers and starting substances for plastics. The list of additives is still not complete.

"Monomers and starting substances" means any substance used in the manufacture of a macromolecule which constitutes the repeating unit of a polymer chain or polymer network of any substance used in the manufacture of a plastic for food contact application. It also includes the substances used to modify existing natural or synthetic macromolecular substances. According to Directive 90/128/EEC the following substances are included in the definition:

- substances undergoing polymerization which include polycondensation, polyaddition or any other similar process, to manufacture macromolecules
- natural or synthetic macromolecular substances used in the manufacture of modified macromolecules if the monomers, or the other starting substances required to synthesize the monomers, are not included in the list
- substances used to modify existing natural or synthetic macromolecular substances

The "Practical Guide" presents information for the applicant on mixtures, synthetic mixtures, mixtures from natural sources and process mixtures.

Traditional biobased materials are paper and board, regenerated cellulose and cellulose acetate. More recent biobased packaging materials are thermoplastic starch, polylactic acid and PHA. A number of interesting substances in the area of biobased materials are included in the positive list on starting substances for plastics: glucose, lactic acid, cellulose and starch. The incomplete list of additives includes gelatin, dextrin, pectin, and cellulose-derived substances, etc.. Some examples of the legislative status in the plastic directives of certain ingredients in biobased materials are given in Table 4.2. The lists are frequently amended by new directives as the evaluation of substances is being carried out. The Commission gives information on substances which are not included in the directives, but which have been dealt with in the Scientific Committee on Food (SCF) in "the Synoptic Document" (to be found at <http://cpf.jrc.it/webpack/>).

Table 4.2. *Legislative status of some starting substances for biobased materials.*

Substance	Status
Albumin	Starting substance list
Cellulose	Starting substance list
Glucose	Starting substance list
3-Hydroxybutanoic acid-3-hydroxypentanoic acid, copolymer ¹	Starting substance list
Lactic acid	Starting substance list
Lignocellulose	Starting substance list
Starch, edible	Starting substance list
Sucrose	Starting substance list
Alginic acid	Additive list
Casein	Additive list
Cellulose	Additive list
Cellulose acetate butyrate	Additive list
Cellulose derivatives, various	Additive list
Cellulose, regenerated	Additive list
Dextrin	Additive list
Glycerol and various derivatives	Additive list
Gelatin	Additive list
Hydroxyethyl starch	Additive list
Hydroxypropylstarch	Additive list
Lactic acid	Additive list
Lactic acid, butyl ester	Additive list
Pectin	Additive list
1,2-Propyleneglycolalginate	Additive list
Starch, edible	Additive list
Starch, hydrolysed	Additive list
Alginate	No classification ²
Carrageenan	SCF list 1 and list 9 ²
Cellulose acetate	SCF list 3, inert material, modified natural cellulose ²
Chitin, chitosan	No classification ²
Gluten	No classification ²
1,3-Propyleneglycolalginate	SCF list 8 ²
Zein	SCF list 0 ²

¹: Also known as Biopol

²: For explanation see next section on petitioner procedures

In Commission Directive 1999/91/EC, Annex IV "Products obtained by means of bacterial fermentation" which authorizes the use of the 3-hydroxybutanoic acid-3-hydroxypentanoic acid copolymer (also known as Biopol) has been added. A specific migration limit of 0.05 mg/kg is stipulated for crotonic acid (as impurity) and certain specifications on the polymer are presented.

4.3. Petitioner procedures

The formal authorization process is described in the Commission document "Note for Guidance". See Internet site: <http://cpf.jrc.it/webpack/>. It states what the technical dossier accompanying such request must contain and what migration and toxicological tests are to be carried out. The criteria used by the Scientific Committee on Food (SCF) when substances are being evaluated, are also explained. Data to be submitted must contain the following:

- identity of the substance
- properties of the substance
- use
- information on authorization given by countries and on evaluation by international organisations
- migration data
- toxicological data

It is not always necessary to supply all the data if the petitioner has justification for it, e.g. very low migration.

SCF opinions on individual substances are set out in the form of classifications into one of ten lists List 0...List 9 and waiting lists.

Whenever acids, phenols or alcohols have been evaluated, the assessment also includes aluminium, ammonium, calcium, iron, magnesium, potassium, sodium and zinc salts. In the case of foodstuffs or food ingredients, used either as monomers and starting substances or as additives to plastics, these substances will automatically be included in List 0 if the data, requested by SCF, have been supplied. Food additives listed in EC Directives or Reports of the SCF will automatically be added to List 1 if the data requested by SCF have been supplied. The migration data are still needed, as for some food additives, restrictions are set on use of levels or use in certain foods. Migration from plastic materials must not lead to any infringement of these restrictions. Sub-

stances in Lists 0 to 4 can be called “approved” substances. Substances, which should not be used, end up in List 5. Substances with lacking data or suspected carcinogenic properties are in Lists 6 to 9.

The registration procedure in the Commission requires at least two years to be completed and published in a directive. The cost of producing the background data is estimated to be approximately 0.5 million Euro.

4.3.1. Standardized test methods

A limit is required for substances with the potential to migrate from the food contact material to the foodstuff. For plastics, an overall migration limit of 60 mg/kg of food is stipulated. For individual substances, specific migration limits are imposed in accordance with their toxicity.

Under the mandate of the European Commission the European Committee for Standardization (CEN) is preparing standard test methods required for testing of compliance with the requirements and restrictions in the plastics Directives. Overall migration test methods are published in standard EN 1186 parts 1 to 12. Additional parts are published as pre-standards.

The different parts allow for testing of most types for materials and articles under various testing conditions. Testing is carried out using food simulants. Rules for selecting simulants and test conditions are given in the relevant EC directives.

Similarly, specific migration test methods of seven plastic monomers are published as a pre-standard ENV 13130 with eight parts. Methods of analysis for 35 monomers were developed in a European research project and published in European Commission Report EUR 17610 EN.

Test methods to be used for checking paper and board are also prepared by CEN. Test methods are published for preparation of cold and hot water extracts, determination of water soluble matter, formaldehyde, polychlorinated biphenyls, metals (cadmium, lead, chromium, mercury), determination of fastness of colouring agents and fluorescent whitening agents, and transfer of anti-microbial constituents.

The common EU legislation does not give specific test methods for checking the sensory properties of a food contact material. General instructions are given in some national compilations of test methods such as the BgVV (Bundesinstitut für gesundheitlichen Verbraucherschutz und Veterinärmedizin, Germany) recommendations. A number of standard procedures for taint transfer testing have been published, all containing information on setting up transfer tests and carrying out the sensory testing. Probably, the best known procedure, the so-called Robinson Test, has been used to test printed and unprinted paper and board materials. Many individual companies and research institutes have developed their own protocols for food contact material testing.

The legislation on food contact materials does not give any specific provisions for the microbiological quality of the materials. Food legislation in the European countries, however, includes general hygiene requirements. These can be understood to require the materials to be of appropriate microbiological quality, taking into account the food to be in contact with the article.

For certain biobased materials the conventional migration methods using aqueous simulant liquids tend to be very demanding, especially in those cases where the migrating substance is practically a “food ingredient”. Since only a few studies on migration of biobased and biodegradable materials are published and only limited experience on migration testing of biobased packaging materials is at hand, it is difficult to judge whether the standardized methods are suitable for various material types. The migration test period does not take into account possible changes, like degradation, in the material during long storage times.

4.3.2. Implications of EU legislation for food and packaging industry

The objectives of the legislation on food contact materials are to ensure that the materials do not contaminate the foodstuff making them unsafe for consumption. It is clear that the legislation applies to “all-in-the-chain-from-the-raw-material-supplier-to-the-actual-retailers”. The manufacturer of the material will have to ensure that only authorized raw materials are used.

With new materials, for instance plastic monomers, necessary toxicological and migration studies must be carried out to complete an application in order to get the substance authorized. Information on residual monomer levels or migration properties might be necessary for the converter.

The converter is expected to provide the user/retailer with compliance statements. Necessary testing must be carried out. The user is the one who actually knows the composition of the food to be in contact with the final article and the conditions of storage and use. He has the responsibility to ensure that all the information supplied to him is relevant in respect to the foodstuff.

The countries which are usually recognized by other governments for their comprehensive and useful legislation and recommendations are USA (FDA), Germany (BggV) and the Netherlands (Warenwet). Following these requirements may help to prove good manufacturing practices in cases where there are no local detailed regulations other than the general measures.

Food contact materials manufactured from a combination of two or more types of raw materials are not specifically regulated as yet. In most countries, plastic coated paper is treated firstly as plastics, since this is the material in contact with the foodstuffs, and secondly as paper since the normally thin plastic layer is not proved to be a functional barrier layer.

The Framework Directive, however, covers all kinds of materials. Detailed knowledge on all raw materials, the structure of the final product and the manufacturing process will be needed for a thorough evaluation of exactly what parts of the regulations that are relevant and need to be observed. Even then, depending on the nature of the material, there might be room for individual interpretation by the various national authorities.

4.4. Assessment of potentially undesirable interactions

All relevant potentially undesirable interactions are assessed by literature study in this section. All interactions are discussed separately and recommendations are made to ensure food quality and protect consumers.

4.4.1. Migration of compounds from biobased packages to contained food products

Migration is an important aspect to consider when designing food packaging materials. The principal legislation has been laid down in "the Framework Directive". More specific regulations are given for regenerated cellulose materials in EU Directives and for paper and board in various national legislations. But the existing regulations and guidelines on plastics might not be suitable for new biobased plastic-like materials. For instance, biobased material may contain components, natural or synthetic, as additives, plasticizers, cross-linking agents, antioxidants, preservatives, etc. which are not common in conventional packaging materials. Similarly, the migrational behaviour of these additives, as well as common additives for food contact plastics, may be different in biobased materials compared to conventional plastics.

For starch, the use of plasticizers is needed in order to increase the flexibility. Water is an excellent plasticizer. Other examples are polyhydric alcohols (glycerol, ethylene glycol, glucose, sorbitol, propylene glycol, polyethylene glycols, polyvinyl alcohol, etc.) amino acids, amino alcohols, amides and quaternary ammonium compounds. No literature on the migration of these additives from starch-based packages is available.

In order to prove the safety of polylactic acid for the use as food contact material, the polymer was evaluated by considering the identity and toxicological properties and quantities of substances that migrate into food during intended use (Conn et al., 1995). Migrants from polylactic acid may include lactic acid, lactoyllactic acid (linear dimer of lactic acid), other small oligomers of polylactic acid (trimer, etc.) and the lactide (cyclic dimer of lactic acid). It was concluded that lactic acid is the ultimate product of hydrolysis of any substances that migrate from the polylactic acid contacting food. In Europe, lactic acid is mentioned without any specific restrictions in the monomer list of the plastic directives. The migration determined using 8% ethanol by a 10 days test at 43°C was 0.85 mg/dm². It was shown that there was no more migration into acidic media than into a neutral one. The migration level into the fatty food simulant was approximately one-sixth of that observed in the aqueous system. The authors summarize that very limited migration can be expected from polylactic acid into foods during intended conditions of use.

The matter that might migrate into food is lactic acid or its oligomers which will hydrolyze in aqueous systems producing lactic acid. Lactic acid is a common food ingredient which has been shown to be safe at levels far above those migrating from the polylactic acid material.

A study on polylactic acid film proves that overall migration into water, 3% acetic acid, 15% ethanol and into olive oil (10 days, 40°C), as well as into iso-octane (30 min, 40°C), was less than 1 mg/dm² (Selin, 1997). The author points out that the plasticizer to be added can be either soluble in water or fat and could, thus, cause migration problems. Hence, the choice of plasticizer must be made dependent on the film end-use.

The mechanisms of transfer of substances from paper into food have not been thoroughly studied and might consequently be necessary to carry out as to determine harmful metals, chlorinated organic compounds, fluorescent whitening agents and dyes, some binder components, volatile substances, etc. on paper used as food packaging material.

Regenerated cellulose contains large amounts of softening agents that are water-soluble. Thus, EU legislation on regenerated cellulose is based on compositional limits for ingredients. For mono- and diethylene glycol, however, a migration limit of 30 mg/kg of food is set. The regulation also defines the ingredients in the coatings, like plasticizer and other additives, by total quantities of substances. A study on migration of softeners reveals that propylene glycol and triethylene glycol migrated into food to a level of more than 1000 mg/kg and 500 mg/kg respectively, but was clearly reduced by using coated films (Lancaster and Richards, 1996). A retail survey of foods packaged in nitrocellulose-coated regenerated cellulose film showed phthalate plasticizer content from 0.2 to max. 46 mg/kg when packed in films containing 0.1 to 1.8 % of various plasticizer, mainly dibutyl phthalate and dicyclohexyl phthalate. The intake was considered not to present any hazard to health. (MAFF, 1996)

4.4.2. Microbiological contamination of biobased food packages

Microbiological contamination of packaging materials is most likely to occur during construction, transport, storage and usage

of packaging materials since the harsh conditions during processing render the materials either sterile or near-sterile (Dallyn and Shorten, 1988). Therefore, precautions are usually taken to avoid contamination during storage and usage or measures are taken to reduce the microbial load, for instance controlled storage and package sterilization prior to use.

Studies in which the microbial load of packaging materials is determined are limited (Kneifel and Kaser, 1994). Most work has been focussed on aseptic packages and packages made from paper and board (Narciso and Parrish, 1997; Pirttijarvi et al., 1996; Vaisanen et al., 1989; Dallyn and Shorten 1988; Windaus and Petermann, 1978; Placzek and Witter 1972). In general, the microbiological contamination levels of packages made from conventional and biobased materials are relatively low and negligible, well below the standard of 1 organism / cm² or 250 CFU / gram paper and board homogenate proposed by the US Department of Health, Education and Welfare in 1966 (Dallyn and Shorten, 1988). The only reported exceptions are cardboard and corrugated board packages made from recycled paper (Narciso and Parrish 1997; Kneifel and Kaser, 1994).

Literature on the microbiological contamination level of biobased materials is rather limited. A microbial study of cellulose triacetate based archival photographic films showed that after years of storage under ambient conditions mostly *Pseudomonas* bacteria were found in the film (Harthan, 1997). This shows that the growth of microorganisms in cellulose triacetate is possible although extremely long times (10 - 100 years) seem to be required for microbiological contamination levels to reach unacceptable levels.

Literature on the microbiological growth rates in and on packaging materials is also limited. Test methods on the resistance of synthetic polymeric materials to fungal growth have been published (ASTM G21-96, G22-76, G21-70). Plastic samples are placed on the surface of agar plates and are inoculated with test cultures. Fungal growth is measured semi-quantitative as the percentage of the surface that has been covered. Recently, both conventional and biobased packaging materials have been examined with a slightly modified version of ASTM G21-96 (Petersen et al., 2000). Selected food related fungi (*Penicillium* and

Aspergillus) were tested. In all cases growth and survival were observed. It was concluded that the current standards are indecisive since the results can not be fully interpreted. Improved test methods are necessary taking into account realistic environmental conditions (temperature and relative humidity of use) to produce practical useful information.

In general, the growth of microorganisms in and on food packaging materials is depending on several parameters: the initial load, the nature of the material, the contained food, time and conditions. More research is needed to understand the interactions between microorganisms and (biobased) packaging materials, since the current knowledge is not extensive. It is recommended to the European Commission to initiate research to enhance knowledge in this area.

All current applications of biobased food packages comply with GMP guidelines and national regulations. The current system of microbial quality control of packages by converters and local health authorities is effective in monitoring the quality of (biobased) packaging materials.

4.4.3. Penetration of microorganisms through biobased packaging materials

Several test methods have been developed to determine the penetration rate of microorganisms through packaging materials either from the outside environment to the contained food product or visa versa. In the Bio-test method, filled food packages are immersed into a tank of bacteria inoculated water and are incubated for several weeks. Microbial permeation is observed whenever the contained food is spoiled faster than packed food which is not exposed to the tank water (enhanced microbial growth rate, pH changes and gas production, etc.) (Maunder et al., 1968; Ronsivalli et al., 1966). Alternatively, food packages are filled with aqueous solutions of various nutrients and microorganisms and microbial permeation is determined from discolorations of the package exterior due to microbial initiated leakage (Cerny et al., 1993).

Most packaging materials have proven to be completely impervious to microorganisms. Moreover, the microbial load of fresh food products is incomparably large relative to the amounts of

microorganisms that could permeate through packages that this phenomenon can usually be neglected. Microbial penetration is only important in three cases.

1. Penetration can occur through compromised regions (pin-holes in packages, ill-constructed seals) (Narciso and Parrish, 1997). The pin-holes need to be at least 10 µm in diameter (Hurme et al., 1997) and the required time for microbial permeation through pin-holes is usually high (in the order of 1-2 weeks) (Kamei et al., 1988). Hence, this effect is only relevant for food products with extremely long shelf lives (e.g. aseptically packed products).
2. Food products with high microbial activity, such as surface active mould cheeses (Camembert, Brie), can digest paper in which they are packed. Simple barrier layers of wax and polyethylene on the paper suffice to inhibit this activity (Robertson, 1993).
3. Biodegradable packaging materials (polycaprolactone, polyvinylalcohol, polyhydroxybutyrate and cellulose acetate) are only digested by food-borne microorganisms under the condition that the food product is rich in minerals, but it lacks a source of carbon. When food products contain both minerals and a carbon source, no attack of the packaging materials could be detected (Cerny et al., 1993). Surprisingly, cellulose acetate was found to be most resistant to microbial attack of the tested materials. Films of 10 µm thickness resisted microbial attack during the test of seven months under the unfavourable condition of a carbon-poor food product (Cerny et al., 1993). Since the vast majority of food products contain sources of carbon (carbohydrates, fats, etc.), microbial degradation of biobased packages will not occur during usage.

Hence, microbial permeability through biobased packages will not affect the quality of the contained food as long as the food product is a fresh product that is rich in all nutrients (including carbohydrates or fats). Only in the unlikely case that the food product does not contain carbohydrates and fats / oils, it is recommended to apply an inner coating of biobased material that is resistant to microbial penetration (for instance 10 µm of cellulose acetate). In the case of surface active mould cheeses a small protective wax coating suffices.

4.4.4 Penetration of insects and rodents into biobased food packages

Large portions (approximately 15%) of the global food supply are spoiled by insect and rodent activity. Packaging can reduce these losses. Insect and rodent penetration results in the loss of the protective function of packages and can introduce microbial contamination. In principle, insects can penetrate all non-inert packaging materials paper, board, polyolefins, and polyesters. The same holds true for rodents that can eat through all flexibles; only glass and steel are effective barriers (Robertson 1993). In spite of the fact that the raw materials (e.g. starch, proteins) used in the production of biobased packaging materials are used as food by macrobionts, there is no evidence indicating that packages made from biopolymers are more readily attacked by these organisms than packages made from more conventional polymers. On the contrary, cellulose acetate is very resistant to insects (Robertson, 1993). However, it is recommended that the European Commission initiates research on the resistance of biobased packaging to insect and rodent penetration.

4.4.5. Collapse due to absorbed moisture from the environment and the contained food product

Biobased materials are hydrophilic by nature rendering them potentially interesting as barrier materials in food packages. However, due to this hydrophilic nature the materials are also potentially moisture sensitive implying that they could lose their barrier and mechanical properties when exposed to water or moisture originating either from the ambient or the contained food product.

The moisture sensitivity can result in increments in gas permeability of 50 to 60; see Chapter 2. Such increments have little consequence for most food products that are packed in air. Only the additionally enhanced moisture permeability could potentially result in an enhanced re-hydration of dried foods. The moisture sensitivity could have consequences for food products that are packed under modified atmospheres. Special modifications to biobased materials might be necessary to reduce the moisture sensitivity and make these materials applicable for modified atmosphere packaging purposes.

Unfortunately, no literature is available on the relation between the relevant mechanical properties (tensile strength, compression

strength, puncture strength, elongation, etc.) and the relative humidity at ambient temperatures. According to the experience of authors it is known that packages made from paper, board, starch, cellophane and chitosan become very weak at 100 % relative humidity (RH) at room temperature. Pallet stacks of ill-designed corrugated board boxes are known to collapse at these conditions and flow packed cellophane bags become very flexible which can result in mechanical damage to the food product. The loss of packaging integrity is a potential problem, which can be dealt with by careful construction and modifications in the biobased materials. Extra support elements can be added to the package construction to function as skeleton. Alternatively, thicker trays, cups, etc. can be applied. Furthermore, biobased materials can be modified to be less moisture sensitive by:

- coating with less hydrophilic materials (waxes, polyesters, fatty acid ester derivatives (Gontard and Guilbert, 1994)
- cross-linking with inorganic fillers (Otaigbe, 1998)
- blending with less moisture sensitive materials (Stenhouse et al., 1993)
- reinforcement with natural fibers (jute, flax, coconut, wood) to form composites (Snijder and van Dam, 1999)

In summary, the moisture sensitivity of biobased materials is an important aspect of consideration during the design and construction of new biobased food packages. In order to secure food safety it is advised to test these packages under worst case conditions (100 % RH) for the full anticipated shelf-life.

4.5. Conclusions and perspectives

In principle, biobased packaging materials and conventional materials are treated equally in the European food contact material legislation. The same safety criteria and test methods should be applied for all materials regardless of their origin. However, due to differences in origin and properties, some undesirable interactions are more relevant for one or the other material. Generally, Chapter 4 identifies aspects of the use of biobased food packaging materials which need to be investigated further, like the potential interactions between living organisms and the materials, and loss of barrier and mechanical properties under humid conditions. So far, literature on the microbiological contamination level of biobased materials is rather limited. For biobased materi-

als which are moisture sensitive, the conventional migration test methods using aqueous food simulants tend to be very demanding. The test methods do not take into account possible changes, like degradation, during long storage times. In addition, attention should be paid to sensory properties like for all new materials. However, literature available indicates no obvious safety risk for food contact biopolymers which are already available on the market.

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5. Environmental impact of biobased materials: Biodegradability and compostability

5.1. Biodegradability

The terms “biodegradation”, “biodegradable materials” and “compostability” are very common but they are frequently misused and are sources of misunderstanding. Solubility in water is frequently considered as a synonym of biodegradability, and biodegradability as a synonym of compostability. The term biodegradable, by itself, is not useful. It is a general recognition that, in the biosphere, there is at least one enzyme which can speed up the breaking rate of the chemical bonds of a given polymer chain. Notably, it does not ensure that a biodegradable material will always degrade. In fact, degradation will not occur in an unfavourable environment or the biodegradable material will not degrade within in a short time. Notably, the term “biodegradable” does not imply a fast process. It is, therefore, important to couple the term biodegradable with the specification of the particular environment where the biodegradation is expected to happen, and of the time scale of the process.

During recent years the attention of the standardization groups working in this field has mainly focused on the definition of compostability of man-made solid materials, because of the fact that composting is considered the preferred system of treatment of the organic fraction of the solid waste, where the biodegradable/compostable biobased materials are supposed to end up. The preparation of the specific standards on compostability has been driven and speeded up by the European Directive 94/62/EC on packaging and packaging waste.

5.2. The composting of biobased packaging

The European Directive 94/62/EC has specified that composting of packaging waste is a form of recycling, owing to the fact that the original product, the package, is transformed into a new product, the compost. The biological treatment[1] may have a very important role to reach the recovery targets fixed by the Directive whenever the other forms of recycling are not suitable due to technical or economical reasons. The Directive has indi-

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cated the need for preparing European standards for the definition of compostability, i.e., the set of the features which a packaging must possess in order to be claimed as “compostable” and, therefore, recycled through this particular form of treatment. The definition of the criteria of compostability is of utmost importance due to the fact that materials not compatible with composting (traditional plastics, glass, materials contaminated with heavy metals, etc.) may decrease the final quality of compost not making it suitable for the application in agriculture and, therefore, commercially unacceptable. The composting may be considered to be a recycling process only if reintegration of the recycled material is being allowed into the market. From an environmental point of view it means the integration of the compost in the bio-geo-chemical cycles of the carbon with the restoration of the natural ecological cycles.

Therefore, a packaging which does not satisfy the requirements of compatibility with composting, partly indicated by the same Directive, cannot be recycled through this form of waste treatment.

5.3. The CEN activity

The European Committee of Standardization (CEN) has been appointed by the European Commission with the Mandate M200 to prepare the technical norms to support the European Directive 94/62/EC. In particular, the group denominated CEN TC261 SC4 WG2 (within the Technical Committee 261, “Packaging”) has prepared the norm EN13432 “Requirements for packaging recoverable through composting and biodegradation- Test scheme and evaluation criteria for the final acceptance of packaging”. This norm is an important achievement because it is a reference point for the producers, the public authorities, the composting plant managers, and the consumers. It also represents a barrier to the self-claimed biodegradable-compostable biobased plastics which appeared on the market more than 10 years ago and which still, every now and then, are offered with engaging

[1] The biological treatment can be aerobic (composting) or anaerobic (biomethanization). Composting leads to the transformation of waste into carbon dioxide (released into the atmosphere), water and compost, usable for agricultural purposes. Biomethanization leads to the formation of biogas (methane and carbon dioxide) and sludge. The anaerobic sludge is then usually transformed into compost by a subsequent composting step. For this reason, the term “composting” is used as a synonym of biological treatment of solid waste, covering both aerobic and anaerobic processes.

eco-advertisement. The transparency is one of the key factors that may lead to a real acceptance of this class of products and a clear-cut standard is the basis of this transparency.

5.4. The compostable packaging

According to the EN13432, a packaging is compostable if it is formed by components which have been individually qualified, as compostable. In this way the analysis of compostability of a packaging is simplified and traced back to the analysis of compostability of the single constitutive materials. The advantage is obvious: materials applied in packaging are limited in number, but the possible number of types of packages, which can be derived from them through combination or through shape and size variation, are enormous. If the long and expensive set of tests specified in the qualification procedure should be applied to any single type of packaging, it will become a useless and economically un-sustainable exercise. Therefore, it is sufficient to use compostable materials in order to obtain a final compostable packaging. Hence, whoever wants to put a product on the market using a compostable packaging should obtain the necessary guarantee and certifications regarding the compostability from his supplier of compostable packaging materials. The producers of packaging materials, in turn, should obtain the certifications from the producer of the basic material. It is a chain which starts from the producers of the basic material (the supplier of the bio-based plastic), passes through the converters (the producers of the semi-manufactured product), through the producers of packaging, and ends at the final user who applies the packaging for a food product on the market. In this process of responsibility transfer, no further tests are required unless two materials are intimately linked to form a new material, which is the case when a plastic film is coupled with another plastic film or with paper to form a strong multilayer structure (a new component of the packaging). In these cases, the disintegrability of the new material must be checked, since this property is affected by the thickness and by the physical structure and cannot be deduced by the disintegrability of the original materials. Each component used in the compostable packaging should be given a certification of compostability including the minor elements such as inks and colorants. The maximum thickness of use, above which the disintegrability is not warranted any more, must also be indicated.

5.4.1. Laboratory tests

The CEN norm is aimed at indicating the characteristics of the compostable packaging and the test methods needed to verify the conformity of the packaging under study with the requirements. The compostable packaging must have four main features.

- Biodegradability, that is the metabolic conversion of the packaging material into carbon dioxide.
- Disintegrability, meaning fragmentation and loss of visibility in the final compost (absence of visual pollution).
- Absence of negative effects on the process of composting.
- Absence of negative effects on the final compost (i.e. reduction of the agronomic value and presence of eco-toxicological effects on the plant growth).

Each of these points is needed for the definition of compostability but it is not sufficient on a sole basis. A biodegradable material is not necessarily compostable since it must also be disintegrable during the composting cycle and it must not cause problems either to the process or to the final product (the compost).

The procedures of evaluation of the packaging material are reported in the following sections.

5.4.2 Characterization

Characterization is a preliminary phase during which information on the packaging material is gathered. The constituents, i.e., the ingredients used for the production of the material, are identified and the presence of toxic substances, heavy metals in particular, are verified. The maximum concentration of heavy metals allowed in the compostable packaging is specified by the EN13432. In this case, the limits are lower than those required by the Directive 94/62 EC, due to the fact that the release of heavy metals in the final compost is highly detrimental for the quality of compost. Furthermore, the test material is analyzed to determine the total organic carbon, the dry weight, and the volatile solids, all information needed in the following test of biodegradability.

5.4.3. Laboratory test of biodegradability

In this phase the interest is focused on the biodegradability of the material and its constituents determined at laboratory level. The specific test method is the controlled aerobic composting test which is technically identical to the ISO 14855:1999 “*Determination of the ultimate aerobic biodegradability and disintegration of plastic materials under controlled composting conditions – Method by analysis of evolved carbon dioxide*”. The method simulates the environmental and microbiological conditions of a composting process. The test material is generally powdered and mixed with some mature compost (used as a source of microorganisms and nutrients) brought to the correct degree of humidity and maintained at 58°C. From the measurement of the CO₂ produced under these conditions the degree of conversion (mineralization) of the organic carbon of the biobased material is determined. In parallel, the biodegradation of the reference material the microcrystalline cellulose, is measured. According to the EN13432, the biodegradation of the test material, measured using the controlled composting test, must be at least 90% of the level reached by cellulose in a maximum time of six months. As an alternative to the ISO 14855, it is possible to use two methods of measurement of the biodegradability in aqueous environment, ISO 14851 and ISO 14852, for those cases in which the composting method is not appropriate (ink, additives, colorants, etc.).

5.4.4. Disintegration under composting conditions and verification of the effects on the process

In order to verify that the test material, in its final physical form, can be disintegrated during a composting cycle without leaving residues (a visual pollution is not acceptable in commercial compost) a composting test at pilot scale must be performed. Pieces of material are composted with fresh waste in a 200-litre bin. The method is described in the standard CEN W1261074 (equivalent to the ISO 16929). Obviously, in this case it is not possible to use powdered material (i.e., already mechanically disintegrated). The basic material must be converted into the final packaging or in the semi-manufactured product. Therefore, in this trial the test sample can be a film, a foil, a sheet, a foam, or the packaging itself. The thickness of the specimens used in the disintegration trial is important as it determines the maximum thickness at which the packaging material under study can be

applied in the market. The disintegration rate generally decreases with the increase of the thickness. Therefore, a positive result obtained in the disintegration test allows the use of the material at the tested thickness or at lower thickness, but it does not guarantee the compostability of the material if it is used at an increased thickness. When using an increased thickness, it is necessary to repeat the trial verifying the disintegrability of thicker specimens. At the end of the cycle, which lasts three months, the disintegration is verified by sieving. The composting at pilot scale can also be useful as to verifying possible negative effects of the test material on the composting process and to produce the compost needed for the ensuing quality analysis and ecotoxicity testing. As an alternative, full scale testing can be performed to assess disintegrability.

5.4.5. Compost quality: chemical and eco-toxicological analysis

The test material must not influence the final characteristics of the compost. Samples of compost, obtained by mixing the test material with organic waste, are compared with samples of a reference compost produced only with organic waste and without the test material. The results must not differ significantly. The required analyses are: volumetric weight (density), total dry solids, volatile solids, salt content, pH, levels of nutrients (total nitrogen, ammonium nitrogen, phosphorus, magnesium and potassium). Furthermore, the effect of the compost samples on the plant growth is assessed using the method described in the same norm to show that the test material, during degradation, does not release substances toxic for the plants and the environment into the compost.

5.4.6. Natural materials

Chemically and unmodified natural constituents, such as wood, wood fibre, cotton fibre, starch, paper pulp or jute, are considered as biodegradable and do not require a test as to their biodegradability. However, all the other characteristics concurring to show compostability are required. This exception is due to the fact that some natural products (most notably lignin) do not comply with the biodegradability criteria (90% biodegradation in six months). This result is considered by the critics of the EN13432 as proof that the criteria are not satisfactory. Lignin is a very complex natural material which slowly biodegrades. As a

consequence, lignin builds up in the soil in the form of humic substances. The accumulation of lignin in the environment is a natural event, which is beneficial for the fertility of the soil. While it is well-known that lignin is ultimately degradable and helps environment and soil structure, the accumulation of other foreign materials cannot be encouraged because, synthetic products cannot claim to have a beneficial effect on the fertilizing capacity of the soil, as the behaviour of synthetics in natural environment is not known. Therefore, the compostability criteria have been devised to reject materials, which may be accumulated in the soil. Unavoidably, the system "recognizes" lignin as a material potentially causing accumulation. However, in this case, the accumulation is beneficial.

5.5. Biodegradability under other environmental conditions

During recent years the attention of the standardization groups working in this field has mainly focused on the definition of compostability of man-made solid materials. The European norm EN13432 is an important achievement. This norm is going to become a harmonized standard required by the European Commission as a technical tool of the Directive 94/62/EC to be enforced by all the European members. However, composting is not the only environment in which the degradation of the biobased materials can occur. For instance, soluble biobased materials can be flushed in the sewage system and biodegraded in the wastewater treatment plants. Biobased materials can also be used in agriculture where the degradation is expected to take place in the soil. The standardization work is still actively dealing with these other important environments that which were somewhat neglected in the past in favour of compostability. A recently formed standardization group, the CEN TC249 WG9 "Plastics - Characterization of degradability", is at the moment addressing these topics in order to define test methods and specific requirements. Therefore, in the future, we can expect to have standards and definitions covering each main environment, so that the term "biodegradable" will be a meaningful and useful designation to better qualify innovative materials and their environmental fate.

6. Environmental impact of biobased materials: Lifecycle analysis of agriculture

6.1. A sustainable production of biobased products

Products made with renewable raw materials are considered to be environmentally beneficial, saving fossil resources and being potentially biodegradable. The issue of biodegradability is discussed in the previous chapter. In this chapter we specifically address issue of the rational use of resources and the protection of the environment.

The beneficial effect of using biobased fuels and materials is represented by the fact that they can help in neutralizing the global warming¹. This prospect is also very interesting from a social point of view as it may support the agricultural sector, which is notoriously afflicted by problems of overproduction, with the development of non-food crops and new markets. As outlined in Chapter 2, biobased polymers are produced either by directly extraction from biomass or by using fermentation techniques producing either the polymer directly, or the monomers, which then are polymerized into the final biobased polymeric material. Agricultural products are excellent as feedstocks for both procedures (see Chapter 2). The question is, if an enhanced demand for agricultural products for non-food uses influences the environment and further how it is ensured, that a negative impact is not the result. Hence, the overall impact must be assessed, to estimate and weigh risks and benefits, which are obviously present in any human activity. The methodology of the Life Cycle Assessment (LCA) has been recently applied to determine the environmental impact of the agricultural production and to have a complete comprehension of the problem and assess the environmental sustainability of the biobased materials. In the following some of the available LCAs on the agriculture production is presented. Notably, recent developments has shown that waste products

¹The global warming is caused by the increased concentrations of greenhouse gases (GHG) in the atmosphere. According to the Intergovernmental Panel on Climate Change (www.ipcc.ch), the concentration of carbon dioxide has increased by about 30% over the past 200 years. It is one of the most serious environmental issues and, if not controlled, alterations in local weather and the increase of sea levels will affect the social, economic and environmental structures in this century.

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from agriculture or from the food industry may be used as feedstocks for fermentation processes leading to the production of biobased polymeric materials (Garde et al., 2000; Södergård, 2000). An efficient utilization of agricultural resources, with the use of all fractions of the agricultural products will prove beneficial for the production of materials.

6.2. What is LCA?

According to the International Standardization Organisation (ISO), the Life Cycle Assessment (LCA) is a technique for assessing the potential environmental aspects associated with a product (or service), by: compiling an inventory of relevant inputs and outputs, evaluating the potential environmental impacts associated with those inputs and outputs, interpreting the results of the inventory and impact phases in relation to the objectives of the study.

An LCA is a quantitative analysis of resource depletion and production of pollutants from the production system under study, but it can also include a qualitative analysis of other important issues which are more difficult to quantify (for example: the biodiversity). The "cradle to grave" analysis (from extraction of raw material to waste management), which incorporates manufacturing practices, energy input/output and overall material flows, is needed to assess environmental impact and sustainability. LCA is used mainly for comparison between alternative products or processes or to identify the production steps causing the greatest environmental impacts. The information from the LCA may then be used to identify improvement options and appropriate corrective actions.

6.3. Environmental impact of agriculture

What is the environmental impact of developing, growing, and harvesting biomass crops? Agriculture, like any other human activity, has an impact on the environment. Agriculture influences the concentration of carbon dioxide in the atmosphere by affecting the amount of carbon stored in terrestrial ecosystems as plant biomass (through deforestation and reforestation), by consuming fossil fuels directly and in the production of fertilizers and other agro-chemicals, and by providing renewable energy resources in place of fossil fuels.

In order to assess the sustainability of the utilization of agricultural raw materials it is necessary to develop a life cycle approach to this specific area. There is a need to adapt the general LCA methodology, born in the industrial field, to specific areas, that is the agricultural production and the exploitation of biomass in non-food applications.

Most of the available case studies on the use of biogenic resources include the environmental impact of agriculture in terms of direct emissions from the soils (e.g. N₂O from microbial metabolism of N-fertilizers) and indirect emissions from fuel consumption of farming equipment. Within the context of LCA little emphasis has been put on the issue of the environmental quality of the used farmland. One of the reasons may be that this aspect is not easily operable within the methodological LCA scheme. An effort to improve and adapt the LCA to agriculture is currently ongoing. A new method, for example, has been recently developed by the IFEU-Institute (Giegrich and Fehrenbach, [DGfH 1999, in German]) and it is being tested in an LCA case study on loose-fill-packaging materials (for project summary see below). The basic idea of the method is to measure the impact on agricultural ecosystems by determination of the "degree of nature proximity". For this purpose, an ordinal scale is used with seven classes of "nature proximity" ranging from "Class 1: Natural" over "Class 4: Semi-natural" to "Class 7": Artificial/non-natural. An array of indicators for area-related criteria ("diversity of weeds", "diversity of structures") and action-related criteria ("soil conservation", "material input") exists which may be used to categorize each kind of land-use within one of the seven classes. In the following sections some relevant examples of LCA applied to agriculture and to biobased products are shown.

6.3.1. Crops for biofuels

A thorough analysis of the environmental impact of several crops was performed in the framework of an LCA carried out in Germany to compare the bio-energy carriers (biofuels) (Kaltschmitt et al., 1997). The study is dealing with fuels and not with materials, but most of the crops analyzed could also be exploited as resources for the production of biobased materials. The study takes into consideration the different steps (ploughing, sowing, harvesting, etc.) needed in the agricultural pro-

duction to produce crops, in substitution of fossil fuels. A significant net energy gain results from the substitution of fossil energy carriers with the utilization of all bio-energy carriers. Substantial savings in climate-relevant emissions (the climatic effects of CO₂, N₂O and CH₄, summed up in the term of "Global Warming Potential" (GWP)) are connected with the production and use of all the bio-fuels considered. On the other hand, the acidification potential (SO₂ equivalents), determined by taking into consideration the airborne SO₂, NO_x, HCl and NH₃, is unfavourable for the bio-fuels to varying extents according to different crops. However, the SO₂ equivalents are essentially determined by the NO_x and SO₂ emissions released during the burning of both the fossil fuel and the bio-fuel. The relative difference is not as remarkable as in the GWP seen above.

6.3.2. The ECN study

A study, developed in the framework of the BRED European project (Biomass for Green House Gases emission REDuction) has been carried out by the Netherlands Energy Research Foundation ECN (Bos, 2000). Using a specific model which covers all Western European Greenhouse Gases (GHG), emission sources and all important techno-economic options to reduce these emissions (strictly speaking, it is not an LCA), it has been shown that the emissions of GHG from agriculture cannot be sensibly reduced, but they are balanced by the positive contribution of biomass to GHG emission reduction. The results show that both agriculture and forestry can supply significant quantities of biomass for GHG emission reduction. The study addresses the impact of the GHG policies on the Western European agriculture and forestry sector and indicates that the ratio between energy applications and material applications of biomass is expected to be 2:1.

6.4. Environmental impact of bio-based products

The outcome of LCA of the bio-based products can be different, in spite of the common or similar composition in raw materials, because other factors act during the production process or during the final disposal. Therefore, the LCA must also consider the products. Some examples of LCA on bio-based products are reported here.

6.4.1. *The Buwal study on starch-based plastics*

A study produced by the Swiss Bundesamt für Umwelt, Wald und Landschaft (Dinkel et al., 1996) has compared products (films and injection-moulded articles) made from starch and starch-containing plastics with conventional plastic products. In this study, the impact of the agricultural cultivation of the raw materials required for the starch-containing plastics on soil quality were considered. The study determined that, from the standpoint of energy conservation and climate protection, positive advantages would be gained by replacing products made from conventional plastics with those made from starch-containing plastics. Similar trends were observed for air pollution and the contamination of water bodies by toxic substances and salts. However, cultivating the agricultural crops needed for the manufacture of the starch-containing plastics on semi-natural areas leads to an increased pollution of water bodies by eutrophication compared to conventional plastics. Provided the starch is cultivated on existing agricultural land, no additional detrimental impact on soil or biodiversity is expected. According to the experts of the BUWAL, the decision as to whether starch-containing plastics are beneficial or not depends on environmental policy objectives (conservation of resources and climate protection, on one hand vs. biodiversity and water eutrophication, on the other hand). They remark that, compared to other regrowable raw materials, starch-containing plastics provide efficient utilization of resources and considerable reductions in the emissions of GHG. The use of regrowable raw materials to produce other materials generally saves more energy resources per hectare of cultivated area and produces greater reductions in CO₂ emission than if the same raw materials were used for energy production.

6.4.2. *The case of hemp-based materials: LCA does not allow generic statements*

The IFEU – (Institut für Energie- und Umweltforschung Heidelberg GmbH) has studied the LCA of products based on hemp, also taking into consideration the agricultural production (Reinhardt and Patyk, 1998). Their conclusions were that different utilization of hemp fibres leads to partly different results. They remark that it is not possible to draw some generic statement on the advantage and disadvantage of using biobased products. The LCA of each product can be affected by a specific phase which makes the difference when summing up. On the other

hand, one can obtain reliable answers on well-defined questions and specific uses of a biobased product.

6.4.3. *Composto's study on bags for the collection of organic waste*

An LCA was performed with the aim of analyzing the impact of the compostable bags for the collection of organic waste, considering different products (PE, paper, biobased plastic) and different waste treatment scenarios (Composto, 1998). It turned out that the biobased bags and the PE bags were equivalent in seven categories out of 13; the biobased plastic bags were better in four and worse in two. However, when taking into account the need for sorting the PE bags from the waste stream and incinerating them along with some residual wet waste, unavoidably stuck to them, the ecological balance turns then strongly in favour of the biobased bags. In this case it is shown that a biobased compostable product is more appropriate than a traditional product for a specific application (organic waste collection). In the same study the paper bags showed rather high impact in comparison with the other bags. This is mainly a consequence of the greater thickness and, therefore, the higher mass of paper needed to reach satisfactory mechanical properties.

6.4.4. *The Ecobilan's study. The LCA of paper sacks*

The study of Ecobilan (Eurosac-Eurokraft 1996) focused on brown paper sacks used for packaging. An important feature of paper manufacturing is the possibility of using renewable energy. Most paper mills can satisfy some of their energy needs through on-site incineration of recovered lignin, bark and sawdust. The use of renewable energy is a factor which reverses the outcome of the LCA which otherwise would be unfavourable because of the high consumption of energy and water associated with the production of pulp and paper. As far as the final disposal is concerned, in the study the landfilling is compared with incineration with energy recovery. The second option turns out to be also beneficial.

6.4.5. *The Ifeu-IBIFA-study. The LCA of loose-fill-packaging*

Supported by the German Environmental Foundation, an LCA study of loose-fill-packaging (LFP) based either on starch or on-and expandable polystyrene (EPS) respectively has been in prog-

ress since 1998 (Ifeu/BIFA, 2000). A draft version is now undergoing a critical review process and the results are expected to be published by the end of 2000 and the project scheme is summarized here. A German producer of LFP using both starch and EPS as a raw material participated in the project.

In the study, scenarios with different starch sources (wheat, maize, and potatoes) and virgin or recycled EPS (obtained from production waste or from the postconsumer PS fraction in the German "Green Dot" system) were analyzed. The comparison is based on the same volume of LFP regardless of its raw material composition. As the bulk density differs according to the raw material used (starch 12 kg/m³, EPS 4 kg/m³), more material input is necessary in the case of starch-based LFP systems. Whether this drawback can be compensated by other beneficial environmental features of the starch products depends very much on the selected disposal routes.

Besides incineration and landfilling, which for the time being can be considered as the conventional final disposal routes, the predominant waste treatment routes for LFP, a post-consumer recovery of LFP was also considered. The EPS recovery options include mechanical and feedstock recycling (e.g. blast furnace). In the case of starch, disposal routes like composting and fermentation requiring the biodegradability of the input material are included. The environmental benefit of the individual waste treatment option like the substitution of fossil energy by biogas produced in fermentation or the substitution of fossil reduction agents in a blast furnace is accounted for as a credit.

6.5. Conclusions

For several years the European Commission has encouraged the use of agricultural raw materials for non-food applications with the scope of promoting agriculture and environmental protection. The objectives were to explore alternative energy sources and to produce chemical commodities using agricultural raw materials. The interest in exploiting non-food agricultural sources as industrial raw materials is also stimulated by the need to find alternative land use in Europe. This is an issue, which has very important social implications as to maintaining rural agricultural employment. This objective is, by itself, a strong and sufficient driving force to encourage projects promoting new applications

for renewable, non-fossil feedstocks. Nevertheless, the agricultural production has an environmental impact which must be evaluated in order to have a complete comprehension of the problem and assess the environmental sustainability of the biobased materials. In this paper some LCA studies on biobased materials and on the agricultural production were briefly reported. All the studies concur in showing that the use of biobased raw materials is advantageous in relation to the energy consumption and GHG emission. The difference between the use of fossil feedstock and biobased feedstock is quantitatively remarkable for these parameters. It also appears that agriculture has some negative impacts on other environmental parameters (i.e. biodiversity, water eutrophication, and acidification). For these parameters the difference between fossil and biogenic materials is significant, but less remarkable than the reduction in GHG emissions. However, a clear comparison of the pros and cons is difficult. An effort is currently done to adopt the methodology of LCA to the production of agricultural raw materials with the aim of better evaluating some difficult to quantify parameters (for instance biodiversity) and consider their impact in the overall balance. Nevertheless, a clearly substantiated fact is that the agriculture can help in controlling the GHG emission. This fact, along with the social benefits deriving from the support to the agricultural sector (as discussed above), should be a convincing reason to encourage the exploitation of crops for non-food applications. Also in this respect, LCA has a fundamental role, i.e., to identify the production steps causing the greatest environmental impacts and to indicate the improvement options to maximize the positive effects reducing the negative impact on the environment to a minimum.

Other important conclusions: The LCA of different biobased products can be different in spite of the common or similar composition in natural raw materials as other factors act during the production process or during the final disposal. Therefore, specific LCA should be performed on a product-by-product basis as different conversion processes being more or less environmentally friendly can change the final balance. The final treatment of the waste originated from the biobased products must be taken into consideration as well. The final system of waste treatment has an important role in the overall eco balance of the biobased materials and can affect the final result. If a biobased material is

recycled through composting, it will contribute to the formation of compost, a product rich in humic substances, which is used in agriculture in place of peat, a fossil material. If a biobased material is recovered by incineration with energy recovery, it will contribute in sparing some fossil fuel. On the other hand, a biobased material dumped in a landfill site could produce negative effects by an uncontrolled evolution of methane.

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7. The market of biobased packaging materials

7.1. Introduction

At first sight there appears to be an abundance of information about packaging biopolymers in technical journals and in the popular media. The majority of this information, however, is about general packaging applications for transport packaging (shock absorbing cushions), disposable packaging (carrier/waste bags, food utensils) and for direct product contact packaging. Concerning the potential and actual market applications in the food-packaging sector it can be seen that there is much more published information about potential applications than about actual ones. Nevertheless, it is instructive to look at the market represented by food packaging and its future development.

7.2. Market appeal

7.2.1. Market drivers

Any food packaging material has to meet basic performance and safety standards which are described elsewhere in this report. In addition, it must meet normal economic price/value requirements. For the use of biopolymer-based packaging there must consequently be an economic point of view. At the present time, the benefits given must be weighed against the undoubted higher material costs in comparison to conventional packaging materials. Added Value will be given (Proterra Study, 1998) if:

- a marketing advantage results
- the biopolymer gives a functional advantage in the product chain
- there is a cost advantage in the waste disposal system
- legislation leads to lower taxes

7.2.2. Marketing advantages

Biopolymers are derived from natural, renewable resources. They are, therefore, fully complementary to the concept of Sustainability. Food products packed in biopolymer-based packaging can represent an overall sustainable product concept. The value of

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this to the consumer needs examination on a case-by-case approach.

7.2.3. Functional advantage in the product chain

Biopolymers have some specific material properties that distinguish them from conventional materials. Barrier properties for gases like oxygen, carbon dioxide and water vapour are quite different to those given by other conventional packaging polymers. A modest extension of product shelf-life by one or two days can be very important for fresh products like cut flowers, fresh meat and ready-to-eat foods.

7.2.4. Cost advantage in the waste disposal system

There is an advantage if food packaging waste can be composted together with the contained food residues. In the Netherlands today, the cost of waste disposal via composting is cheaper than other techniques being about Dfl 100- per ton of waste. This cost arises from the fact that material sorting and separation of the waste stream is not needed if the whole stream is compostable. Biopolymers, which offer biodegradability, are interesting in this light. However, composting is not available as an industrial technique in many countries. Biopolymers can be incinerated in industrial burners without the release of undesirable gases. To date, there has been little or no work associated with recycling or re-use of packaging based on biopolymers.

7.2.5. Legislative demands

Within the framework of the Packaging Waste Directive composting is accepted as one of the techniques available for recovery and re-use of packaging material. This particular section will be extended in 2000 to include the full recommendations of the CEN Working Party TC261 clearly defining the requirements for the use of descriptions like biodegradability. The growing use of one-way disposable consumption packaging (drink cups, food trays, etc.) has led some authorities to introduce taxes (e.g. Kassel, Germany) on these items. Biodegradable packaging results in a lower charged tax. For direct food contact packaging, there has not yet been any advantage demonstrated from lower waste disposal tax charges, due to the fact of an absence of a sufficiently established in-place infrastructure to provide the collection of a separate material stream for composting (Danone experiences with biodegradable yoghurt cups in Germany – 1998).

7.3. Consumers

Clearly, the average consumer sees many negatives in the use of packaging based on fossil sources. Conventional plastic packaging is seen by many consumers as being intrinsically damaging to the environment (Scholten et al., 1997; Meijnders et al., 1995). Even though this point of view is often based on a false perception of the true situation, it is difficult to argue rationally on this matter. The situation does not imply that, in converse, the average consumer will see only positives for biopolymer-based packaging. The argument of sustainability is broadly seen by the consumer to mean less wasteful use of available resources (Proceedings of Consumer International Conference, 1993). Encompassing more than the natural, renewable aspects, for example recycling and re-use play a role as well. Consumer reactions to foods packaging biopolymers are likely to be positive if it can be shown that there is an infrastructure available to deal with the packaging in waste handling. Claiming biodegradability has little or no credibility in the absence of a waste composting industry. The possible environmental claims for biobased packaging need to be scrutinised in light of the International Standard (ISO 14021, 1999).

7.4. The market

7.4.1. Today

It is meaningless to talk about a market for foods packaging based on biopolymers. Current market activities are very much based on exploratory activities, feasibility studies and, occasionally, limited local activities. The quantities of materials involved are no more than a few hundred kilos for any application on an annual basis. A recent publication (Anon., 2000) listed more than 20 academic centres working on bioplastics throughout the world. That list is far from being complete, but it indicates the diversity and dilution of today's approach. The materials under study represent a good cross-section from the categories defined in Chapter 2 of this report. For non-food contact packaging the market today is dominated by starch-based materials.

Unfortunately, there have been many misleading claims made about packaging described under some form of "Eco-" sou-briquet. Additionally claims have been made for biodegradability which does not meet the criteria of the emerging Euro-standard

(CEN, 1999). Biodegradability is a property which all biopolymers are likely to have. Some materials based on mixtures of synthetic materials and biobased materials can have this property. It depends on the nature of the synthetic material whose biodegradability must be assessed using the standard test methods and the criteria developed during the last decade by the CEN. As a matter of fact, some synthetic polymers have been shown to be fully biodegradable and in compliance with the norms.

Table 7.1 lists a number of materials currently on the market. This has been drawn from various sources available in the public domain. It should be stressed that many of these materials may only be available in test quantities. Only very few of them will have been used for direct food contact packaging.

Table 7.1 *Biobased packaging materials and biodegradable materials currently available in the market. Paper and board materials are not included.*

Material	Supplier	Trade Name (if known)	Polymer linkage
Biodegradable materials based on natural renewable sources – Biopolymers			
PHB/PHV	Was Monsanto	Biopol	Ester
(Polyhydroxyalkanoate)	Biomer	Biomer	Ester
Cellulose acetate	Courtaulds		Acetal
	Mazzucchelli	Bioceta	Acetal/ Ester
Polylactide / PLA	Cargill	Dow Polymers	NatureWorks PLA
		Mitsui	LACEA
		Hycail	
		Galactic	Galactic
Starch	National Starch	Eco-FOAM	Ester
	Avebe	Paragon	Ester

Biodegradable materials based on blends of biopolymers and synthetics

Starch-based	Novamont	Mater Bi	Acetal/ Ester
	Biotec	Bioplast	Acetal/ Ester
	Earth Shell	Earth Shell	Acetal/ Ester
	Biop	Biopar	Acetal/ Ester

Biodegradable materials based wholly on synthetics

Copolyester	BASF	Ecoflex	Ester
	Eastman Chemical	Eastar Bio	Ester
Polycaprolactone	Union Carbide	Tone polymer	Ester
	Solvay	CAPA	Ester
Polybutylene succinate	Showa Highpolymer	Bionolle	Ester
Polyesteramide	Bayer	BAK	Ester
Polyesterurethane	Bayer	MHP 9029	Ester
Polyester co-polymer	Bayer	Degraniil VPSP42002	Ester
Polylactic acid	Fortum		Ester
Polyester	Dupont	Biomax	Ester

7.4.2. Tomorrow

There are major extensions anticipated in two classes of biopolymers. Firstly, the currently most abundant type starch will expand its availability primarily driven by its non-foods packaging activities. However, this development will be overshadowed by the major step changes expected in the supply of polylactic acid based materials. These materials will by no means be used solely for food packaging. Cargill-Dow (USA), especially, has a major scale-up for Nature Works PLA to 140.000 tons per year. This scale-up is expected to come on stream in 2002 and will be followed by further extensions. Less ambitious, but significant, is Hycail's (NL) plan to extend their production to 4000 tons. The future situation is forecasted (Bolck, 2000) in Figure 7.1.

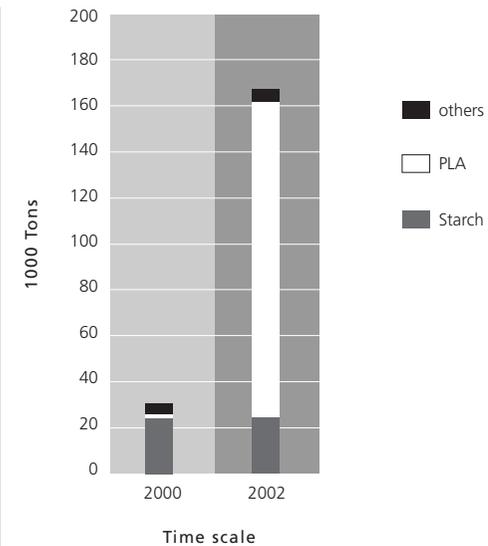


Figure 7.1 Market prediction for biobased materials. Paper and board materials are not included.

7.4.3. Price

The price of any biopolymer is likely to be high when it is only produced on a small scale. The scale of production is likely to have a greater influence on the price than the costs of the raw material source and of the chemistry involved. Prices for materials falling into this stage of development will range between 5 and 50 US\$ per kg. At higher scales of production (large-scale pilot to industrial) the price will fall to a range of 1 to 10 US\$ per kg. The major factors affecting price at a higher scale of production will be the raw materials used and the chemistry. For example, there are different pathways to produce polylactic acids starting from different raw materials (see Chapter 2). Prices for similar functional PLA material would vary considerably depending on the costs of the feedstock.

7.5. Conclusions

Currently, there is no separate market for biobased food packaging materials. The materials will become commercially viable only if capable of competing with conventional packaging materials by showing advantages. Such advantages must trigger consumer appeal and/or enhanced functional performance. Any

real cost saving benefits over conventional materials are unlikely for some time to come. Consumer appeal may be result from these materials being derived from natural, renewable resources. The materials are by nature subject to the concept of "sustainability". Improved functional performance is most likely to stem from the different combinations of physical properties offered by these materials. Significant developments in the supply chain of these materials can be anticipated both in the areas of starch-based and polylactate materials. Starch-based materials will develop from its current base of non-foods applications and polylactate materials will gain benefit from an increase in material availability as a result of major investment in several new production plants by 2002.

7.6. References

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8. Conclusion and perspective

Foods are dynamic systems with limited shelf-life and specific packaging requirements. While the issues of food quality and safety are first and foremost in the mind of food producers and retailers, a range of other issues surrounding the development of any food package must be addressed before a particular packaging system becomes a reality. Biobased food packaging materials must meet the criteria of the specific foods and comply with the food and packaging legislation. Furthermore, interactions between the food and packaging material should not compromise food quality or safety.

This report is based on currently available literature and information about biobased food packaging. Apart from the widely used cellulose-derived materials (paper, board, cellophane, etc.), the focus of biobased food packaging materials is on potential food applications rather than on actual commercial applications. As pointed out in this report the biobased materials can, notably, be used for packaging of a number of different foods and the performance of biobased materials is constantly being improved. Hence, more biobased food applications will emerge in the near future.

8.1. Performance of materials

Chapter 2 summarizes the massive amount of information published in academic journals. Notably, the biologically-derived polymers can be used for the production of all types of packaging (trays, cups, bottles, films (monolayers, laminates, composites), etc.) using the same equipment and processing techniques as for conventional materials. The biobased materials provide the material engineer with new and unique properties e.g. materials with high transmittance of CO₂ compared to O₂ can be produced, and owing to the biological origin, biobased materials have an inherent potential of being compostable. For some biobased materials, a high water sensitivity is observed. In order to apply materials based solely on e.g. starch, proteins or other polysaccharides for packaging of moist foods, the water sensitivity has to be reduced and controlled. Presently, blending with water-resistant polymers (biobased or petroleum-derived) is the standard technology applied to reduce water sensitivity. Alternatively, improved performance of the materials may be accompi-

shed using nano-composite technology, multilayers, coatings or tailoring the biobased monomers/polymers either chemically or using genetically modified organisms. Performance of the materials is being optimized at an ever increasing speed, generating materials with unique properties that will meet the requirements of numerous food applications.

8.2. Food applications

Potential applications of biobased materials for specific food products have been identified, using the product as starting point. Product categories with the potential to utilise biobased materials include meat and dairy products, ready meals, beverages, snacks, dry products, frozen products and fruits and vegetables. In the short term, biobased materials will most likely be applied to foods requiring short-term chill storage, such as fruits and vegetables, since biobased materials present opportunities for producing films with variable CO₂/O₂ selectivity and moisture permeability. However, to succeed, biobased packaging of foods must be in compliance with the quality and safety requirements of the food product and meet legal standards. Additionally, the biobased materials should preferably preserve the quality of the product better and longer to justify any extra material cost.

8.3. Safety and legislation on materials in contact with food

In principle, biobased packaging materials and conventional materials are treated equally in the European food contact material legislation. The same safety criteria and test methods should be applied for all materials, regardless of their origin. However, due to differences in origin and properties, some undesirable interactions are more relevant for one or the other material. Generally, Chapter 4 identifies aspects of the use of biobased food packaging materials which need to be investigated further, like the potential interactions between living organisms and the materials, and loss of barrier and mechanical properties under humid conditions. So far, literature on the microbiological contamination level of biobased materials is rather limited. For biobased materials which are moisture sensitive, the conventional migration test methods using aqueous food simulants tend to be very demanding. The test methods do not take into account possible changes, like degradation, during long storage times. In addition, attention should be paid to sensory properties like for all new

materials. However, literature available (as discussed in Chapter 4) indicates no obvious safety risk for food contact biopolymers which are already available on the market.

8.4. The environment

Increasing demand for agricultural products for non-food uses promotes agriculture. The impact of increased agricultural production has been evaluated in Chapter 6 in order to assess the environmental sustainability of the biobased materials. The studies presented concur in showing that the use of agricultural raw materials is advantageous in relation to the energy consumption and green house gas (GHG) emission. It also appears that agriculture has negative impacts on other environmental parameters (i.e. biodiversity, water eutrophication, and acidification). For these parameters the difference between fossil and biogenic materials is significant, but less remarkable than the reduction in GHG emissions. An effort is currently done to adopt the methodology of life cycle assessment (LCA) to the production of agricultural raw materials with the aim of better evaluating some less easily quantifiable parameters (for instance biodiversity) and consider their impact in the overall balance. Nevertheless, a clearly substantiated fact is that the agriculture sector can help in controlling the GHG emission. This, along with the social benefits deriving from the support to the agricultural sector, should be a convincing reason to encourage the exploitation of crops for non-food applications. In this view, LCA has a fundamental role: To identify the production steps causing the greatest environmental impacts, and indicate possible means of improvement in order to maximize the positive effects, reducing the negative impact on the environment to a minimum. Furthermore, recent developments allow producers of biobased materials to use waste products from agriculture and food industry for production of biobased polymeric materials, and a more efficient use of all fractions from the agricultural production will have a beneficial effect on the LCA for agricultural products.

8.5. The market of biobased packaging materials

Currently, there is no separate market for biobased packaging materials. The materials will become commercially viable only if capable of competing with conventional packaging materials by showing advantages. Such advantages must trigger consumer appeal and/or enhanced functional performance. Any real cost

saving benefits over conventional materials are unlikely for some time to come. Consumer appeal may be result from these materials being derived from natural, renewable resources. The materials are by nature subject to the concept of "sustainability". Improved functional performance is most likely to stem from the different combinations of physical properties offered by these materials. Significant developments in the supply chain of these materials can be anticipated both in the areas of starch-based and polylactate materials. Starch-based materials will develop from its current base of non-foods applications and polylactate materials will gain benefit from an increase in material availability as a result of major investment in several new production plants by 2002.

8.6. Perspective

Today, biobased materials based on cellulose are widely used in the food industry and, before long, the novel biobased materials presented in this report will be included in the enormous arsenal of packaging materials available to the food industry. Initially, being implemented in niche markets, but eventually biobased materials will reach the bulk markets when the performance, availability and costs of the materials become competitive. Biobased materials are not expected to replace conventional materials on a short term, but due to their renewable origin, they are very much the materials of the future.

Increased demand for biobased packaging materials may further stem from a demand for compostable food packaging. However, in order to be able to dispose of the compostable packaging by composting, construction of facilities and infrastructures for increased organic recovery of waste is required. Furthermore, the compost generated through organic recovery must find uses within the fertilizing industry, within agriculture, for horticultural purposes, etc.. Focus is suggested placed on research on how the materials behave during conventional waste treatment, e.g. incineration and composting, to determine the influence of biobased packaging on the total energy consumption etc. Currently, a major demonstration study is being performed in Kassel, Germany in which possible usage of compostable food packaging is being studied. The study runs until the summer 2001, and the ensuing results hereof will, to a major extent, be a determining factor for future use of compostable food packaging.

The petrochemical industry has been very successful in using every by-product at the refineries and the biobased industry must reach the same level of efficiency. In this respect, the use of agricultural and food industrial waste products as fermentation feedstocks for the production of biobased monomers or polymers is very interesting and deserves further attention and research funding. Additionally innovative measures are also required in the area of developing biobased additives, plasticizers, stabilizers, glues, and inks in order to be able to produce 100% renewable packaging.

One of the early steps in creating a food packaging is the approval of the packaging for contact with foodstuffs. In this report no information has been found to support that biobased materials needs special attention in relation to test protocols or legislation. However, in order to reduce the time and resources used in this step, it is suggested to focus on the test protocols used for approval of these materials.

Numerous factors influence future biobased material technology developments, e.g., political and legislative changes, consumer demands, global request for foods and energy resources, etc.. At this stage, the future scenario is difficult to predict. New niches within production of foods and biopackaging may arise that we cannot even imagine now. However, keeping close contact between industry, academia, legislators, etc., e.g., by forming a biobased foods packaging material working group within the EU, will all speed up the process of knowledge exchange between polymer and food scientists, and between the academic world, the industry, and government institutions. Such a group should address future progress within the area of biopackaging of foods and help the EU to identify areas where further R&D on the EU level are required.