ILSI Europe
Report Series

Packaging Materials
4. Polyethylene for Food Packaging Applications

Report

Prepared under the responsability of the
ILSI Europe Packaging Material Task Force

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PACKAGING MATERIALS
4. POLYETHYLENE FOR FOOD PACKAGING APPLICATIONS

By Philip Tice

REPORT

PREPARED UNDER THE RESPONSIBILITY OF THE ILSI EUROPE PACKAGING MATERIAL TASK FORCE

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INTRODUCTION

Polyethylene, often called polythene, is probably the plastic most well known to the consumer and is used in greater volume worldwide than any other plastic. The total global production of plastics rose from 158 million tonnes to 180 million tonnes per annum between 1998 and 2000 – an increase of 12%. In Western Europe in 2000, polyethylene plastics represented 39% of standard plastics consumed, and totalled 11.7 million tonnes. Future annual demand growth for polyethylene plastics in Western Europe is predicted to be about 6%, with the major application continuing to be packaging. Typically over 50% of packaging plastics are consumed in the food retail and food services sectors (Zaby, 2001; Anonymous, 2001). Polyethylene's dominance as a food packaging plastic is due to its relatively low cost, its range of versatile properties, and the ease with which it can be processed into the various packaging forms. Although polyethylene plastics were first produced over 50 years ago, manufacturing and processing developments continue to improve its properties, performance, and food packaging applications.

WHAT IS POLYETHYLENE?

Polyethylene, a thermoplastic, is the principal member of the polyolefin class of polymers. It was discovered by accident in 1933 when scientists working in the ICI laboratories in the UK found that when ethylene was subjected to high pressure a wax-like polymeric substance was formed. This discovery led in the late 1930s to free-radical processes operating at high pressures and high temperatures to produce branched ethylene polymers – now known as Low-Density PolyEthylene (LDPE). These developments became significant in the early 1940s, when the polymers found immediate use as highly efficient electrical insulating materials and played an important role in establishing reliable and practical airborne radar systems (Mark *et al.*, 1985).

The polyethylene plastics produced today are based on a family of polymers derived from ethylene and various α-olefin co-monomers, including 1-propene (propylene), 1-butene, 1-hexene, and 1-octene. They are the most widely used plastics for commodity packaging. Other polyethylene co-polymers are manufactured with co-monomers such as vinyl acetate, producing the ethylene vinyl acetates, and various acrylics, producing, for example, the co-polymers known as the ionomers. The range of ethylene co-polymers produced with the non-alkene monomers will not be covered in this report. Co-polymers with 1-propene (propylene) are included in the ILSI Europe report *Polypropylene as a Packaging Material for Foods and Beverages*.

The original polyethylene (LDPE) has been supplemented with two other widely-used types – High-Density PolyEthylene (HDPE) and Linear Low-Density PolyEthylene (LLDPE). Other classifications are sometimes used for particular specialised polyethylene polymers; they include very-low-density polyethylene (VLDPE), medium-density polyethylene (MDPE) – which is often considered to be at the high-density end of LDPE polymers – ultra-high-molecular-weight polyethylene (UHMWPE), and cross-linked polyethylene.

As the names of the polyethylene types suggest, the principal physical property marking the differences between them is the density. The differences in density are basically due to differences in the degree of crystallinity, which also influences the plastics' melting point ranges. Typical values for these parameters are listed in the following table (Brady *et al.*, 1997; Ashford, 1994):

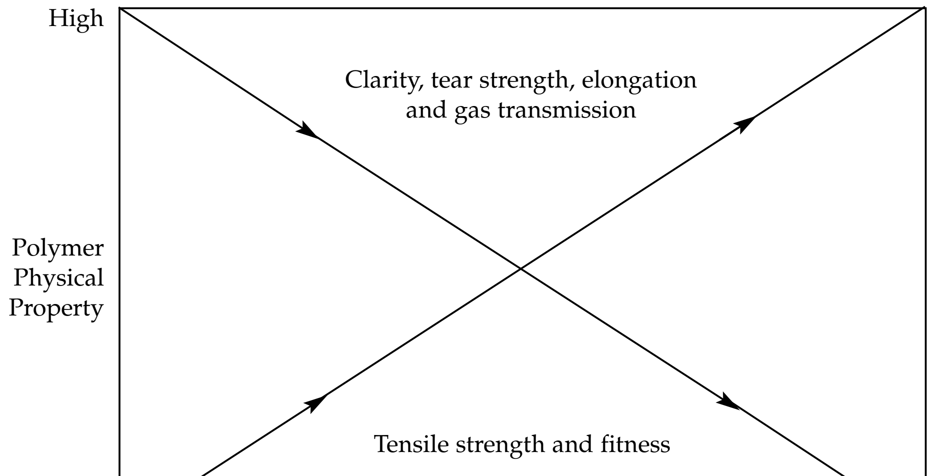
PE polymer	Density, g/cm^3	Degree of crystallinity, %	Melting point range,°C	Molecular weight, Daltons
LDPE	0.915–0.940	45–55	105–115	10,000–50,000
LLDPE	0.915–0.926	30–45	112–124	50,000–200,000
HDPE	0.940–0.970	70–90	120–130	up to 250,000

Polyethylene plastics have the generally advantageous properties of toughness, high tensile strength, and good barrier properties to moisture. A particularly important property of polyethylene plastics, which is due to their relatively low melting point ranges, is the ease with which packaging can be heat-sealed. The barrier properties of polyethylene plastics to oxygen and organic substances are only moderate. These characteristics, along with properties such as clarity and stiffness, vary with the basic polyethylene parameters of density/crystallinity, molecular weight, and molecular weight distribution.

Yield strength and stiffness together with thermal and other mechanical properties increase with density/crystallinity. Toughness and tensile strength also increase with molecular weight. The polyethylenes' high degree of toughness at low temperatures is due to the very low glass transition temperatures, which are in the range of –80°C to –120°C. Clarity generally improves with decreasing density/crystallinity. The relationship between these physical properties and the density/crystallinity allows the polyethylene type to be identified fairly easily. For example, LDPE films tend to be soft and relatively clear, whereas films made from HDPE have a crisp feel and are more opaque (Parker, 1997).

The following diagram illustrates the relationships between polymer density and the principal physical properties. These relationships are not linear, but the changes follow the directions indicated (Rubin, 1990).

Polyethylene physical properties in relation to polymer density



All olefin polymers are prone to oxidative degradation, particularly at the elevated temperatures used for processing. With the polyethylenes, oxidation results in the formation of long-chain branches and cross-linking (Gächter and Müller, 1993). The formulations of all commercial polyethylene plastics therefore contain effective antioxidants (see Section on Basic Chemistry).

HDPE has also been claimed to be an accidental discovery at Phillips Petroleum in the early 1950s. Phillips researchers found that a polyethylene polymer with high degree of crystallinity and a relatively high density was produced under more low pressures, ranging from 3 to 4 MPa, and temperatures ranging from 70°C to 100°C, with catalysts containing chromium oxide supported on silica (Phillips catalyst). In 1953 Ziegler produced polyethylene polymers with similar crystallinity and density with a catalyst system based on titanium halides and alkylaluminium compounds (Ziegler catalysts), and with even milder conditions of atmospheric pressure and temperatures ranging from 50°C to 100°C. The polymers produced with both the Phillips and the Ziegler catalysts were substantially linear, with only short side chains, mainly ethyl. They were the forerunners of the high-density (HDPE) range of polymers. The HDPE polymers manufactured today are also substantially linear polymers.

The linear low-density polyethylenes (LLDPE) are also substantially linear polymers, as the name implies, but they have side chains, the length of which depends on the co-monomer used in the manufacture – the common co-monomers being 1-butene, 1-hexene, and 1-octene. The density is controlled by the amount and type of co-monomer, which typically ranges from 2.5 to 3.5 mole % (Kroschwitz, 1998; Mark *et al.* 1985).

BASIC CHEMISTRY

Monomers and co-monomers used in the manufacture of polyethylene plastics

The principal chemical substance used to manufacture the range of polyethylene polymers is the monomer ethylene (ethene). The co-monomers incorporated to modify the polymers' properties are the α-olefins: 1-butene, 1-hexene, and 1-octene.

At ambient temperature and pressure, ethylene and 1-butene are gases. The other two monomers are colourless liquids (Ashford, 1994).

ethylene $CH_2{=}CH_2$ b.pt –104°C

1-butene $CH_2{=}CHCH_2CH_3$	1-hexene $CH_2{=}CH(CH_2)_3CH_3$	1-octene $CH_2{=}CH(CH_2)_5CH_3$
b.pt –6°C	b.pt 63°C	b.pt 121°C

Ethylene and the α-olefin monomers are obtained at low cost by cracking petroleum and other hydrocarbon feedstocks. About half of all ethylene produced is used in the manufacture of polyethylene plastics.

Commercial manufacture of polyethylene plastics

The various polyethylene polymer types are manufactured by polymerising ethylene either on its own, which produces the homopolymer, or with the above-mentioned α-olefin monomers, which produces the olefin co-polymers.

Low-density polyethylene (LDPE) is manufactured from ethylene monomer using high pressures ranging from 100 to 135 MPa, at temperatures in the range of 150°C to 300°C, in the presence of a small amount of oxygen or an organic peroxide. Both stirred autoclave and tubular reactor processes are in use. The density/crystallinity of the resulting polymer is determined by the reaction temperature used – the lower the reaction temperature, the higher the density. Other important polymer characteristics, such as molecular weight and molecular weight distribution, are controlled by the pressure used in the process and by the concentration of chain transfer agents. Molecular weights are typically in the range 10,000 to 50,000 Daltons (Brady and Marsh, 1997; Kroschwitz, 1998).

Linear low-density polyethylene (LLDPE) is usually manufactured in either gas phase or slurry reactor processes by co-polymerising ethylene with one or more of the α-olefin monomers under low pressure conditions, typically 2 to 7.5 MPa, and with temperatures up to 250°C in the presence of a catalyst, such as the Ziegler type. The density is determined largely by the amount of co-monomer used, typically 2.5 to 3.5 mole%, with higher amounts resulting in lower densities. The type of co-monomer – 1-butene, 1-hexene, or 1-octene – also influences other characteristics of the polymer produced. Molecular weights range from 50,000 to 200,000 Daltons.

High-density polyethylene (HDPE) is manufactured as the homopolymer using reaction processes, catalyst systems, and pressure and temperature conditions similar to those used for linear low-density polyethylene. Small amounts of co-monomer can be used to produce polymers at the lower end of the density range. The type of catalyst used determines the molecular weight distribution, whereas molecular weight is controlled by the proportion of hydrogen included. Molecular weights are as high as 250,000 Daltons (Brown, 1992).

LLDPE and HDPE polymers can be manufactured with the metallocene catalysts, which produce polymers with uniform structures both in molecular weight distribution and in co-monomer incorporation. LLDPE films made from these polymers have a number of enhanced properties, including improved toughness, puncture resistance, and clarity (Kulshreshtha, 2001; Hernandez *et al.*, 2000).

Additives used in polyethylene plastics

Additives must be incorporated into the basic polyethylene polymers and the co-polymers in order to maintain and provide the desired physical properties and to ensure the efficient processing and handling of the finished products. All additives used in food contact polyethylene plastics must be approved for use in the relevant safety regulations and comply with any migration or other restrictions (*see Section on Regulatory*). It is also necessary to ensure that the additives are of a good quality and that they do not impart taints or off-odours to packaged foodstuffs.

As noted above in the section on What is Polyethylene, polyethylene is prone to oxidation at the elevated temperatures used during processing into the various forms and packing types, typically in the range of 180°C to 280°C. Antioxidants are therefore necessary to prevent the oxidative degradation of the polymer. Two main antioxidant types are used – the phenolic and phosphite types – at concentrations of 0.01 to 0.5 % wt. The phenolic antioxidants also act as stabilisers (Gächter and Müller, 1993).

For films, the particular properties of slip (friction) and blocking (film layers sticking together) must be enhanced for effective and efficient handling during manufacture and processing into the finished packaging. Enhancement of these properties is achieved by the addition of slip agents and suitable fine particulate fillers to the polymer. Typical slip agents are the fatty acid amides – oleamide and erucamide – which "bloom" (diffuse) to the film surface after manufacture (Gächter and Müller, 1993).

Titanium dioxide and calcium carbonate are typically used to produce white plastics. The inclusion of calcium carbonate in the polymer also improves the properties of hardness, stiffness, and printability as well as permeability to oxygen and to water vapour. Shrinkage and elongation are reduced (Gächter and Müller, 1993).

Polyethylene plastics, like most plastics, have a tendency to accumulate static charges on the surface. This can result in handling problems and "pick-up" of dirt on the finished packaging, which affects the external appearance of the film, bottle, or container. Antistatic agents are incorporated to minimise this problem. Typical antistatic agents used with polyethylene plastics include polyethylene glycol esters, glycerol monostearate, and the ethoxylated secondary amines, added to the polymer at concentrations of 0.1 to 0.5% wt. These agents have hydrophilic properties and limited solubility in the polymer, which results in their migration to the plastic's surface, where they become active. They can also be applied externally to films and containers as aqueous or alcoholic solutions (Gächter and Müller, 1993).

Some antistatic agents can act in addition as external lubricants during film manufacture and injection moulding operations. Sodium alkane sulphonates (C16–C18) are also used as lubricants and are particularly effective at the low concentrations of 0.05 to 0.15% wt (Gächter and Müller, 1993).

Additives such as colourants, whitening agents, slip additives, and antistatic agents are often conveniently incorporated into the basic polymer before processing into the final product (films, containers, etc.) by means of master batches. Master batches are concentrates of the additive(s) dispersed in the same or similar polymer types.

Migration from polyethylene under conditions of use

Substances that may migrate from polyethylene plastics to foodstuffs include residual monomers, low-molecular-weight polymers (oligomers), and any additives or other substances used in the formulations or the manufacturing process. The quantities of substances that migrate from polyethylene plastics into the foods and beverages with which they come into contact depend largely on the type of food or beverage, the temperature during contact, and the contact time. Migration into liquid foods will be higher than that into more solid foodstuffs, particularly dry foods. With all plastic types, migration increases with temperature and time of contact. The uses of polyethylene plastics in contact with food are described below, in the next section. Hydrophobic substances, such as oligomers and the antioxidants, have a tendency to migrate into fatty foods, whereas hydrophilic substances, such as anti-static agents, have a tendency to migrate into high-moisture foods. Polyethylene plastics are not generally used in applications in which there is high-temperature contact with foodstuffs because of their relatively low melting point ranges, but they are used as packaging for a wide range of water-based and fat-containing foods and beverages.

Any migration of substances from polyethylene plastics must comply with restrictions specified in safety laws and regulations.

Reported migration values for polyethylene plastics in all forms—films, pots, tubs, and other containers—and with all four standard food simulants are typically well below the regulatory overall migration limit of 10 mg/dm^2 and any relevant specific migration limits for the monomers and additives (Czerniawski and Pogorzelska, 1997). With the relatively high volatility of all the monomers used in the manufacture of polyethylene plastics, any residues remaining after manufacture tend to be lost during the subsequent processing into the finished packaging film, container, etc. Data on migration of additives from polyethylene and the factors influencing migration have been comprehensively reviewed (Figge, 1996). Other studies have reported data on migration of antioxidants from polyethylene plastics with standard food simulants under a variety of exposure conditions (Lickly *et al*., 1990; Goydan, 1990).

POLYETHYLENE AND FOOD PACKAGING APPLICATIONS

Polyethylene, the first commodity plastic to be used for food packaging, came into general use in the 1950s. Since then it has achieved its dominant position as a packaging material for a wide range of foods and beverages because of its relatively low cost, its versatile properties, and the ease with which it can be manufactured and converted.

The two main end forms of polyethylene plastics used for food packaging are:

- films, made by both cast and oriented processes
- bottles and other containers made by thermoforming and blow moulding processes.

Different types of polyethylene with the appropriate physical properties are used for these two applications. The low-density polyethylenes – LDPE and LLDPE – are the principal types used for films, and the more rigid HDPE is the main polymer used for containers.

In some of the film applications originally reserved for LDPE polymers, the newer LLDPE polymers have taken over because of their superior tensile strength, elongation to break characteristics, and puncture resistance. The water vapour transmission properties of the two polymer types are similar (Kroschwitz, 1998). In applications in which films require enhanced rigidity, HDPE polymers are used.

Most films undergo stretching by means of the "bubble" process making them bi-axially oriented at the molecular level (Hernandez *et al.*, 2000) and are then converted into bags and pouches. They are often laminated with other plastic types for applications in which the polyethylene is used for heat sealing and/or as a barrier to water vapour. A typical laminate combination is nylon (polyamide) with polyethylene for boil-in-the-bag foods. Here the nylon provides the necessary rigidity at the temperature of boiling water.

In other laminates polyethylene is combined with a variety of polymer types, such as polyethylene terephthalate (PET), polypropylene (PP), polyvinylidene chloride, (PVdC) and ethylene vinyl alcohol (EVOH), selected to provide the particular required properties and performance characteristics. When good barrier properties to oxygen are required, an aluminium foil layer is incorporated. Such flexible packaging is used for coffee, where the structure also acts as a gas barrier in reverse, ensuring that the coffee aromas are retained.

In special multi-layer packaging materials, polyethylene is combined with paperboard and aluminium by extrusion coating. A typical example is the container for packaging long-life fruit juices and milk. The polyethylene allows the container to be easily heat sealed and also provides a barrier to water. The paperboard provides rigidity, and the aluminium acts as an oxygen barrier, which ensures that the safety and quality of the packaged product are maintained during its shelf life. For foodstuffs with a short shelf life, the structure does not contain an aluminium layer. Paperboard coated on both surfaces with polyethylene is extensively used for boxed containers for milk products, take-away high-moisture and fatty foods, and for disposable beverage cups. Polyethylene-coated paperboard is widely used as external cartons for many foods, over a wide temperature range (frozen to ambient). Such cartons are easily heat sealed in food packaging line processes by spot contact heating.

Polyethylene-coated aluminium foil is extensively used as lidding material for pots and other containers. The polyethylene coating that melts under heat and bonds the aluminium to the sub-structure provides the sealability of these foils.

Aluminium is also used decoratively on polyethylene films in the form of very thin coatings produced by the vacuum deposition process. These aluminium-coated films are commonly used in the UK for packaging bread.

For food products with a short shelf life, such as fresh fruit and vegetables, polyethylene film bags provide simple but effective packaging. Meat and fish products with a short shelf life are often packaged on plastic trays, such as foamed polystyrene, and overwrapped with polyethylene film. The low glass transition temperatures (Tg) of low-density polyethylene means that the films remain flexible at low temperatures. This characteristic makes them the preferred films for packaging frozen food products.

For many years in the UK and elsewhere, milk bottles were made of glass, which could be easily cleaned and sterilised for reuse, and milk was delivered to the home. As shopping habits changed and milk was more frequently purchased at food supermarkets, glass became less suitable as a material for milk bottles and was largely replaced by HDPE. Bottles for milk, fruit juices, soups, and a variety of other liquid beverages constitute the single largest use for HDPE plastics in the UK. Elsewhere in Europe, polyethylene-coated carton board constructions are more common for these foodstuffs. When HDPE bottles were first introduced in the UK, they were intended to be single-use and disposable. Collection and recycling schemes for the plastic bottles are now in operation as part of the efforts for national compliance with the current environmental legislation. Developments in the design of HDPE bottles include multi-layer structures with oxygen barrier resins, such as ethylene vinyl alcohol (EVOH), which allows foodstuffs to have an extended shelf life.

Both HDPE and LDPE plastics are used to produce a wide variety of caps and covers for bottles and containers. The use as films for container labels is also growing.

Many of the polyethylene films and containers used to package food and beverages are printed to provide product identification, information on use, and decoration. Because polyethylene plastics have low surface energies, surface treatments are necessary to enable printing inks to achieve adequate adhesion. Corona discharge, flame, and ozone treatments are commonly used to increase the surface energy by imparting a degree of oxidation to the polyethylene surface (Hernandez, *et al.*, 2000).

Some examples of foods and beverages for which polyethylene-based packaging is used are listed below:

- fresh fruit and vegetables – film bags, heat-sealed overwrapping film, and container liners for bulk transport
- frozen fruit, vegetables, meat, poultry, and fish products – film bags and heat-sealed overwrapping films
- cereals – film bags, sometimes with carton board outer packaging
- bread and bakery products – film bags
- milk, milk-based drinks, soups, fruit juices, and fruit drinks – HDPE bottles and polyethylene/cartonboard/polyethylene laminate containers
- long-life fruit juices and milk – polyethylene/aluminium/cartonboard/polyethylene laminate containers
- yoghurt and other dairy products – polyethylene/aluminium lidding
- take-away foods and beverages – polyethylene/paperboard/polyethylene laminate containers
- coffee – polyethylene/aluminium/polyethylene terephthalate laminate bags.

REGULATORY

European regulations that apply to polyethylene: Specific European Commission Directives on plastics used for packaging of foodstuffs

As part of the process of harmonising legislation within the European Union Member States, the European Commission (EC) has introduced framework Directives concerned with the safety of all materials and articles intended to come into contact with foodstuffs, and has started work on Directives for specific material types. The current framework Directive is 89/109/EEC, which lays down the general principles. In short, these may be stated as avoidance of any danger to human health and the maintenance of the quality of the packaged foodstuff.

The first major specific Directive (90/128/EEC) to deal with plastics has been under continuous development since its inception. It seeks to establish the limits of acceptance for all plastic components (e.g. monomer residues and additives) in foodstuffs.

Directive 90/128/EEC contains an overall migration limit and a list of authorised plastic monomers, some with restrictions, either in terms of specific migration limits (SML) or maximum permitted quantity of the residual substances in the material or article (QM). The amendments to Directive 90/128/EEC have added to or altered the list of monomers, but most significantly they have introduced a list of additives, which is currently classified as incomplete. As with the plastic monomers, some additives have been assigned specific migration limits.

After Directive 90/128/EEC had been amended six times, it was decided in 2002, for reasons of clarity and rationality, to produce a consolidated Directive. This was published in August 2002 as EC Directive 2002/72/EC. Amendments to the consolidated Directive will be made as the regulations are further developed.

The provisions of the EC Directives are brought into use by individual European Union Member States by implementing them in their national laws and legislative systems.

The European food contact regulations are increasingly being adopted by non-European Union Member States and other countries outside Europe.

Full details on all of the Directives related to food contact materials and articles as well as other related documents can be found at the European Commission DG SANCO website on Food Contact Materials: http://europa.eu.int/comm/food/fs/sfp/food_contact/index_en.html, and at the website http://cpf.jrc.it/webpack/legislat.htm, which is organised on behalf of the EC by the Food Products and Consumer Goods Group of the Institute of Health and Consumer Protection at the Joint Research Centre in Ispra, Italy.

Migration: overall migration limits, specific migration limits

The overall migration limit contained in Directive 2002/72/EC is specified as 10 milligrams of plastics substances released per square decimetre of plastic surface area (mg/dm^2) or, alternatively, 60 milligrams of plastics substances transferring to 1 kilogram of foodstuff (mg/kg).

Ethylene, the principal monomer used to manufacture polyethylene, and the three monomers 1-butene, 1-hexene, and 1-octene used as co-monomers, are in the List of Authorized Monomers and Other Starting Substances in Directive 2002/72/EC. No restrictions are included for ethylene and 1-butene. The other two monomers are listed with the following specific migration limits (SML).

1-hexene, SML 3 mg/kg

1-octene, SML 15 mg/kg

Many of the plastics additives used in polyethylene plastics are now listed in Directive 2002/72/EC and some have restrictions assigned.

The following are typical additives used in polyethylene plastics and their "restriction" status:

Additive	**Restriction**
Pentaerythritol tetrakis[3-(3,5-di-tert-butyl-4-hydroxphenyl) propionate] *(antioxidant)*	None
octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate *(antioxidant)*	SML = 6 mg/kg
phosphorous acid, tri(2,4-di-tert-butylphenyl) ester *(antioxidant)*	None
erucamide, oleamide, and stearamide *(slip agents)*	None
calcium carbonate, talc, and titanium dioxide *(fillers)*	None
glycerol monostearate *(anti-static agent)*	None
N,N-bis(2-hydroxyethyl)alkyl(C8-C18) amine hydrochlorides *(anti-static agents)*	SML (T) = 1.2 mg/kg (expressed as N,N-bis(2-hydroxyethyl)alkyl(C8-C18) amine)

Standard methods for testing polyethylene plastics for compliance with the overall migration limit have been prepared as standard EN 1186 by Technical Committee 194/Sub-Committee 1 of the European Committee for Standardization (CEN). This standard has recently been updated and revised. It was published in fifteen parts as EN 1186 in 2002 (European Committee for Standardization, 2002). The same technical committee is currently preparing standard test methods for the determination of 1-octene in the four standard food simulants (with minor modifications, this test method could be used for 1-hexene). At present there are no standard migration test methods for plastics additives that have legislation restrictions.

U.S. FDA regulations

The United States have their own regulations for the control of plastic materials that are used in contact with foodstuffs, produced by the U.S. Food and Drug Administration (FDA). Substances that may transfer from plastics to foodstuffs are classified as: "indirect food additives".

Regulations for polyethylene plastics and the co-polymers are contained in the Code of Federal Regulations (CFR) Title 21, Part 177.1520, "Olefin polymers". Additives that may be used in polyethylene plastics and the co-polymers are contained in CFR Title 21, Part 178, "Indirect Food Additives: Adjuvants, Production Aids, and Sanitizers". This part contains sub-parts dealing with, for example, antioxidants (2010), antistatic and antifogging agents (3130), clarifying agents (3295), colorants (3297), and release agents (3860). In contrast with the EC regulations, the U.S. regulations do not contain any specific migration limits for the listed substances.

The FDA regulations are updated annually. The full details of these regulations as they apply to polyethylene, the copolymers, and the additives that can be used with these plastics are available on the Web site: http://www.access.gpo.gov/nara/cfr/.

The FDA regulations are often used by non-European countries that do not have their own detailed regulations. In Europe they are sometimes used for products that currently are not covered by an EC Directive or a specific national regulation.

SAFETY/TOXICOLOGY

Before substances are placed in the Authorized List of Directive 2002/72/EC, the European Commission's advisory body, the Scientific Committee on Food (SCF), assesses the toxicological properties. When necessary, the Commission applies a restriction on the basis of the SCF opinion. In most cases the restriction is a specific migration limit – a limit on the quantity of the substance that may transfer from the plastic to the foodstuff (SML). Other substances are assigned a QM restriction.

The two monomers – ethylene and 1-butene – used in the manufacture of polyethylene and the principal copolymer plastics, have not been assigned any restrictions in Directive 2002/72/EC. The decisions not to assign restrictions to these monomers were based on SCF opinions from the toxicological assessments and on the fact that these monomers are very volatile and easily lost from processed plastics.

The SCF opinions for both monomers stated: "Residues of this gas in plastics are very small. The gas has low toxic potential. Migration into food will be toxicologically negligible" (Clayton and Clayton, 1981).

The two monomers have been placed in SCF List 3, "Substances for which an acceptable daily intake (ADI) or tolerable daily intake (TDI) could not be established, but where present use could be accepted. Some of these substances are volatile and therefore unlikely to be present in the finished product".

The fact that ethylene is a very volatile gas for which residues in processed polyethylene plastics are typically undetectable significantly reduces the possibility of ethylene oxide being formed and present. Ethylene oxide has been assigned a QM restriction of 1 mg/kg in Directive 2002/72/EC.

For 1-hexene, which has been assigned an SML of 3 mg/kg, the reported summary of the SCF's opinion is: "Available: Migration data, 5 mutagenicity studies negative, 28-day and inadequate 90-day oral rat studies, 90-day inhalation rat study, combined reproduction/development toxicity screening study in rats, bioaccumulation (TNO [November 1995] and Elias [February 1996] SDS CS/PM/2742, and RIVM summary data, 14 December 1991)". This monomer has also been placed in SCF List 3.

For 1-octene, which has been assigned an SML of 15 mg/kg, the reported summary of the SCF's opinion is: "t-TDI: 0.25 mg/kg b.w. pending results of fertility and teratogenicity studies. Available: a 90-day oral rat study and mutagenicity studies (CIVO re. V86.408/251091, 26 September 1986). This monomer has been placed in SCF List 2: "Substances for which a TDI (= Tolerable Daily Intake) or a t-TDI (= temporary TDI) has been established by this Committee".

A polyethylene additive for which it has been possible to arrive at a TDI value and from which an SML value has been determined, is the antistatic agent N,N-bis(2-hydroxyethyl)alkyl(C8-C18) amine hydrochlorides. This substance has been placed in SCF List 2, and the reported summary of the SCF's opinion is: "Group t-TDI: 0.02 mg/kg b.w. (as "free" amine) (with N,N-bis(2-hydroxyethyl)alkyl(C8-C18) amine). Available: 90-day oral rat and dog studies (RIVM report, November 1971). Needed: adequate 28 day oral study".

The above quoted summary data from the SCF assessments and similar data produced for other plastic monomers and additives can be found in the European Commission's Synoptic Document, "Draft of Provisional List of Monomers and Additives Used in the Manufacture of Plastics and Coatings Intended to Come into Contact with Foodstuffs". The latest version of the document is available on Web site: http://cpf.jrc.it/webpack/.

ENVIRONMENTAL ASPECTS

EC Directive on packaging waste

The principal European Union environmental legislation affecting manufacturers and users of plastic packaging is EC Directive 94/62/EC on "Packaging and Packaging Waste". The basic aim of the Directive is to "Harmonise national measures concerning packaging and packaging waste management in order to provide a high level of environmental protection as well as to ensure the functioning of the internal market thereby avoiding barriers to trade".

The Directive specifies packaging waste and recycling targets that each individual European Union Member State must achieve. These targets are currently under review with the intention of increasing the target percentage values. Proposals put forward at the end of 2001 would require by 2006 an overall recovery target of 60 to 75% by weight and an overall recycling target of 55 to 70% by weight. Different classes of materials are given different recycling targets—that for plastics is 20%. An extended time concession may be given to some Member States to meet these targets.

At present, there are no EC Directives or regulations that apply specifically to food contact materials and articles made from recycled plastics. The regulatory situation in individual European countries varies, with some forbidding the use of recycled materials and others leaving it up to the user to ensure full compliance with the EC Directives, national regulations on food contact plastics, and the overall safety of the foodstuff.

Recyclability and reuse

Collection and recycling of polyethylene film packaging from retail-sold foods are unlikely ever to make a practical contribution to meeting recycling targets for plastics. However, for polyethylene films used as overwraps and liners for bulk transport of fresh and packaged foods, beverages, and other commodities, it is possible to set up practical collection and recycling schemes.

HDPE bottles, however, can be easily collected and the plastic recycled. With both film and bottle schemes currently in operation in Europe, the recycled plastic is not used for new food contact uses. In the United States, some dry food contact applications have been approved. The list of approvals can be found at the Web site: http://vm.cfsan.fda.gov/~dms/opa-recyc.html.

During the 1990s laboratories participating in an EC-funded project (European Commission, 1997) carried out in-depth studies to assess possible health risks to consumers from foodstuffs packaged in either reused plastics packaging or packaging made from recycled plastics.

The principal concern with the use of previously used plastic food packaging and packaging made from recycled plastic material is the possibility that the original packaging is contaminated with hazardous substances after use. Plastics packaging intended to be reused or recycled to make new packaging can be cleaned, but hazardous contaminants may not always be efficiently removed. The propensity for organic contaminants to be absorbed and retained by a plastic and, in reverse, to diffuse out into food from reused or recycled packaging depends on the physical properties of the plastic. The above-mentioned project identified polyethylene as a plastic with high sorption and diffusion characteristics. This means that rigorous cleaning and purification techniques are necessary to make it suitable for reuse as a food contact material.

Further studies of HDPE bottles have concluded that it may be possible to use the recycled plastic for food contact either after it has been subjected to specific highly efficient cleaning processes or as the centre layer in a three-layer sandwich with virgin plastic as the two outer layers (Devlieghere *et al.*, 1998; Huber and Franz, 1997).

In 1998, ILSI Europe published a report from a workshop meeting of the ILSI Europe Packaging Materials Task Force, which had been convened to produce guidelines for the safe recycling of plastics for food contact use (International Life Sciences Institute, 1998). The two principal requirements specified in the guidelines are: only food-grade plastics should be used as the feedstock and that a challenge test should be performed with surrogate chemicals, representing likely contaminants, to demonstrate that after prescribed cleaning, heating, and processing operations, there would be "no detectable migration" of such contaminants.

For collection and recycling purposes, polyethylene plastic packaging often carries the following internationally recognised symbols, which identify the plastics as either high-density polyethylene with the number "2" and the letters "HDPE" or as low-density polyethylene with the number "4" and the letters LDPE. The two symbols are necessary to enable the two polyethylene types to be segregated, which in turn enables the useful production of recycled plastics.

HDPE
High-**D**ensity **P**oly**E**thylene

LDPE
Low-**D**ensity **P**oly**E**thylene

The symbols are typically found on the base of bottles and other containers. They are usually not present on retail packaging films, as it is not practical to collect and recycle most of this type of waste. For such plastic film waste, which often ends up as litter, development work has been directed to producing polyethylene plastics that are either photodegradable or biodegradable. Ethylene–carbon monoxide copolymers have been found to be photodegradable, and polyethylene formulations with added starch have been found to be biodegradable. However, these modified polyethylene plastics have not been brought into mainstream use for food packaging (Mark *et al.*, 1985; Kroschwitz, 1998).

Incineration with energy recovery

As polyethylene and the co-polymer plastics are hydrocarbon polymers, incineration produces combustion products similar to those from hydrocarbon fuels. The small quantities of the plastics additives and the coatings or barrier layers produce only small amounts of other combustion products. Incineration is therefore an effective process for disposal of polyethylene plastics. It also produces energy that can be used for heating or to generate electricity.

GENERAL CONCLUSIONS

Since the 1950s, when polyethylene became the first plastic material to find wide use as a packaging material for foodstuffs, developments over the years have produced a family of plastics which have resulted in polyethylene becoming the dominant plastic material for food and beverage packaging.

Polyethylene achieved its dominant position because of its range of versatile properties, the ease with which it can be manufactured and converted into a variety of packaging forms, and its relatively low cost. The range of packaging forms for which polyethylene is used extends from simple plastic film bags to combinations with other plastics or materials, such as paperboard and aluminium, to provide sealable packaging that ensures that the quality of the packaged foodstuff is effectively maintained.

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