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Migration of Substances from Food Packaging Materials to Foods

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The employment of novel food packaging materials has increased the number of occurring hazards due to the migration from packaging material to the packaged food. Although polymers have mainly monopolized the interest of migration testing and experimentation, recent studies have revealed that migration also occurs from "traditional" materials generally considered to be safe, such as paper, carton, wood, ceramic, and metal. The regulations and the directives of the EU tend to become stricter in this respect. The emphasis is on reaching a consensus in terms of food simulants and testing conditions for migration studies. Furthermore, the list of hazardous monomers, oligomers, and additives continues to augment in order to ensure that the consumer safety is in current agreement with the HACCP, which is continuously gaining ground.

Keywords migration, food simulants, polymers, metals, food packaging, additives

INTRODUCTION

Packaging is an indispensable element in food manufacturing. Despite the well-accepted convenience packaging offers to the consumer, it has been the subject of many debates concerning environmental and health issues. Due to the increasing awareness of consumers in terms of health matters, the importance of the migration of substances from food packaging materials to foods attracted the interest of the scientific and legislative communities.[1–4]

The term "migration" usually describes a diffusion process, which may be strongly influenced by an interaction of components of the food with the packaging material. Figure 1 depicts all subsequent diffusion steps in the polymer-solution interface. This interaction may substantially effect the properties of the packaging material. However, food components, particularly fat, that migrate into plastics, like PE or PP, will considerably increase the mobility of plastic components, thus, enhancing the migration into the contained food.

Migration is a health issue and is a legal problem in most countries. In an attempt to harmonize legislation, the EU (European Union) and the FDA (Food and Drugs Administration) initiated global control through positive lists of substances that can be used, while restricting substances with toxic potential.[5] The general purpose of the legislation is to ensure consumer safety.[6] Until robust mathematical models (simulation) manage to effectively predict and evaluate migration, chemical analysis will be irreplaceable. Various analytical methods have been developed to analyze the migrants in the food. For example, HPLC methods are reported in the literature for the determination of BPA (Bisphenol–A) in epoxyresins and diglycidyl ether of BPA (DGEBA) migrating from films to aqueous food simulants.[6–18]

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DIFFUSION EQUATIONS (MODELING)

Even though mathematical models are still in the process of development and are not very reliable for measuring the potential contamination of food with chemicals from packaging, they are still considered to be valuable tools. The identification of the factors affecting migration is expected to enable manufacturers to improve quality control by allowing them to determine more accurately those variables that have the greatest impact on migration. Furthermore, a better understanding of the migration process will aid in controlling and limiting chemical contamination of food from packaging.[6] Figure 2 shows the general migration behavior, while Figure 3 provides a plan of the general strategy for controlling compliance of plastic materials with food packaging regulations wherever alternative methods are to be adopted.

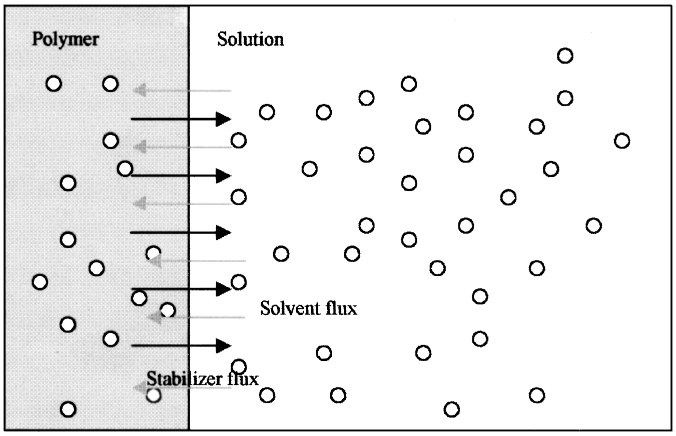

Figure 1 Polymer-solution interface, where all subsequent diffusion steps are summarized.[76]

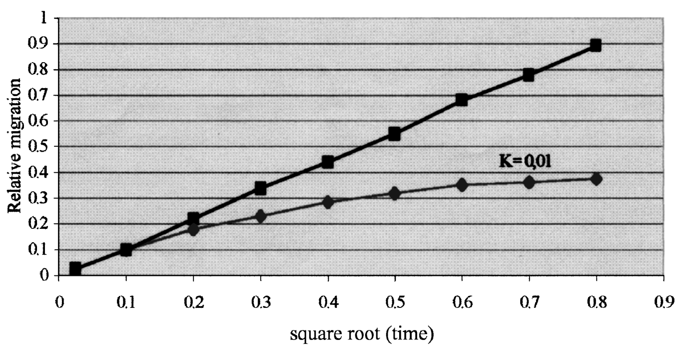

Figure 2 General migration behavior of packaging additives to food. The amount of migration is generally linearly related to time or approaches, the solubility limit of the food [indicated by the partition coefficient K = 0.01].[22]

Since migration is actually a diffusion process, *Fick's* first and second law can be applied.[19]

$$\textit{Fick's}\ \text{1st law: } F = -D_p\,(\delta C_p/\delta x) \qquad (1)$$

$$\textit{Fick's}\ \text{2nd law: } (\delta C_p/dt) = D_p\,(\delta^2 C_p/\delta x^2) \qquad (2)$$

where F = rate of transport per unit area of the polymer; D_p = is the diffusion coefficient of migrant in the polymer (cm^2/s), C_p = the migrant concentration in the polymer (mg/g); x = is the space coordinate measured normal to the polymer—food interface (cm); t = is elapsed time (s). In the case where migration is diffusion controlled, the following equation may be used:

$$M_t = 2\,C_o\rho\,(D_p t/\pi) \qquad (3)$$

where M_t, is the total migrant from the polymer in time, t (s); C_o is the initial migrant concentration in the polymer (mg/g); ρ, is the polymer density (g/cm^3); D_p is the diffusion coefficient of the migrant in the polymer (cm^2/s), and t is the package lifetime in seconds.

All parameters in Equation (3) are generally known or easily measured, except for the diffusion coefficient that must be measured with kinetic experiments. However, the latter experiments can be time consuming and expensive.

The following alternative model has been developed by Baner et al.[20]

$$C_{F,t} = (A/m_F)\rho_p\,C_o\rho\,D_p t \qquad (4)$$

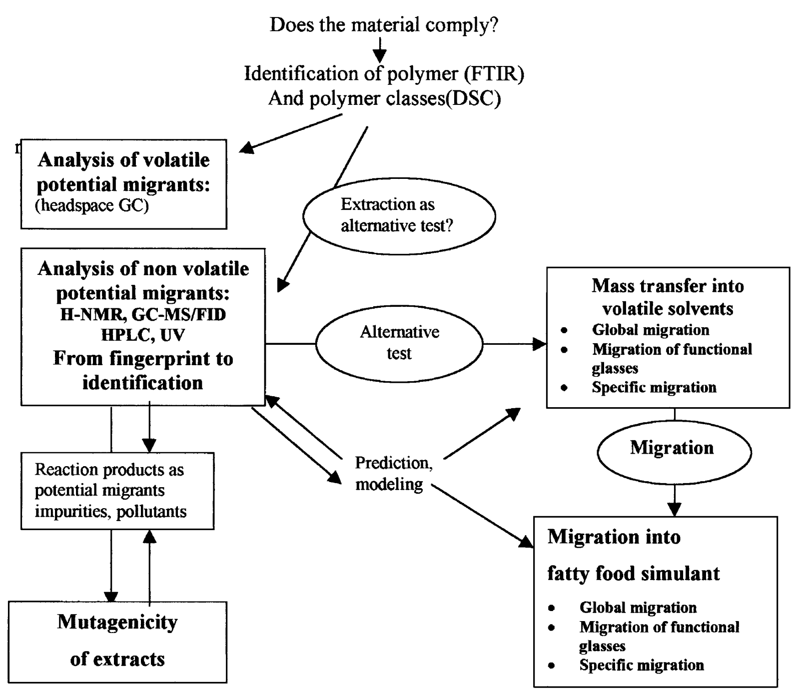

Figure 3 General strategy and alternative methods for controlling the compliance of plastic materials with food packaging regulations.[116]

Table 1 Ap coefficients for various polymers

A_p coefficients	Polymer
9	LDPE
5	HDPE
5	PP
≤0	Non-polyolefins
−7	PVC (rigid)
−3	PC (T ≤ 120°C)
−3	PET (T ≤ 120°C)

where $C_{F,t}$ is the estimated concentration (mg/g), ρ_p is the density of the plastic material (g/cm^3), D_p is the migrant diffusion coefficient (cm^2/s), and (A/m_F) is the package surface area (cm^2). An empirical approach was used according to which known diffusion coefficients were fit to an *Arrhenius* type, as shown as Equation (5).

$$D_p = 10^4 \exp\left[A_p^{-a} \times MW - b(1/T)\right] \quad (5)$$

where the coefficient A_p (constant, dimensionless) accounts for the effect of the polymer on diffusivity (see Table 1 for representative A_p values of various polymer), MW is the additive/contaminant's molecular weight, T is the temperature in K, and a and b are correlation constants for molecular weight and temperature effects on diffusion bearing values of 0.010 and 10450, respectively.[20–22]

Another migration model was described by *Crank's* equation and can be used to describe the migration for semi-infinite media according to Equation 6 (in the case of relatively short migration times):

$$m_t/A = 2C'_{p,o}\,(D_p t/\pi) \quad (6)$$

In addition to the above-mentioned equations, several other semi-empirical diffusion models have been suggested.[21,23–28]

LEGISLATION

The migration itself does not stand for a health problem, but is mainly a legal problem in most countries with legislation on acceptable materials, limits on substances, and restrictions for certain applications.

The EC commission and FDA (Food and Drugs Administration), in an attempt to harmonize various legislations and in order to facilitate and protect consumers from the migration of harmful substances from packaging to food, implemented several directives (EC) and adopted a "threshold policy" (FDA). In 1976, the first CEC Directive proposed analytical methods for detecting limits of various toxic compounds in order to harmonize current legislation among the member states of the EU. Later, Commission Directive 81/432/EEC[30] presented the first community method of analysis for the official control of vinyl chloride, which was being released from materials and articles into food.

Two methods have been given for carrying out the migration testing known overall by quantity of a substance which may be present in the packaging material (Quantity in Material, known as QM) and specific migration tests (quantity that could possibly migrate to the foodstuff, Specific Migration Limit, known as SML).

A QM is more convenient than an SML, when the compound is shown to degrade in the food simulant or if the QM is of such number that even if 100% of the compound migrates into the food, it would still be too low to become hazardous to public health. In 90/128/EEC[33] Commission Directive, an overall migration limit is specified. The overall migration limit has been introduced to control the total amount of substances migrating from a packaging material into food, irrespective of the toxicological significance of these substances, and to reduce the number of specific migration determinations carried out to ensure that the packaging material is suitable for use in contact with food.

The control of the compliance of packaging materials with EC regulation is a very complicated issue. Among the numerous difficulties emphasized are large numbers of substances on positive lists, the absence of information on the potential migrants from the suppliers, the absence of standardized analytical methods, length of analytical procedures, and practical difficulties to carry them out.

The EC has listed thousands of additives and monomers as potential migrants. Many of them are authorized with restrictions or with specific migration limits, and a certain number of analytical methods have been standardized. Finally, official test methods are often lengthy, complicated, and impractical for routine or daily controls.

The directives adopted by the EU member states concerning migration can be divided into three categories:

1. Directives applicable to all materials and articles.
2. Directives applicable to one category of materials and articles.
3. Directives related to individual substances.

Tables 2 and 3 summarize all the EEC Directives issues related to migration and to specific packaging materials. Particularly Directive 90/128/EEC sets an overall migration limit of 10 mg/dm^{-2} (60 mg/kgr foodstuff) for the chemical migration of plastics and lists the monomers and other materials approved for use in the manufacture of plastics for food contact applications. An overall migration level below 10 mg/dm^{-2} does not necessarily imply that the packaging is safe for use, since the gravimetric test cannot identify the chemical nature and toxicity of the migrating species.

Once the directive with the positive lists has been implemented, only those substances appearing in the positive list are allowed for use in materials coming in contact with foods for commercial applications. The positive lists in CEC Directives include most of Supercritical Fluid Chromatography (SFC) lists of substances. Lists 0–4, included in section A of 90/128/EEC, contain substances for which there are available toxicity data

Table 2 EEC Directives related to migration of packaging materials to foods[29–41]

EEC Directives related to migration	Subject
Council Directive 78/142/EEC	Legislation of the Member States in relation with materials and articles containing vinyl chloride monomer and intended to come into contact with foodstuffs.
Commission Directive81/432/EEC	Community method of analysis for official control of vinyl chloride released by materials and articles into foodstuffs.
Council Directive 82/711/EEC	Basic rules for testing migration of the constituents of plastics materials and articles intended to come into contact with foodstuffs.
Council Directive 85/572/EEC	List of simulants for testing migration of constituents of plastic materials and articles intended to come into contact with foodstuffs.
Council Directive 89/109/EEC	Approximation of the laws of the Member States related to materials and articles intended to come into contact with foodstuffs.
Commission Directive 90/128/EEC	Relating to plastic materials and articles intended to come into contact with foodstuffs.
CEN 1992	ENV methods for the measurement of overall migration from plastics into food simulants.
Commission Directive 93/8/EEC	Amending Council Directive no 82/711/EEC laying down the basic rules necessary for testing migration of constituents of plastics materials and articles intended to come into contact with foodstuffs.
CEC, 1993b, Practical Guide n.1	A practical guide for users of EEC directives on materials and articles intended to come into contact with foodstuffs.
CEC, 1994	Draft of provisional list of monomers and additives used in the manufacture of plastics and coatings intended to come into contact with foodstuffs.
CEC, 1994, Synoptic Document No 7	Draft of provisional List of Monomers and Additives used in the manufacture of plastics and coatings intended to come into contact with foodstuffs.
CEC, 1996a	Commission explanatory guidance for migration testing of food contact plastic materials.
Commission Directive 1996	2nd amendment council Directive 82/711/EEC describing the basic rules necessary for testing migration of constituents of plastic materials and articles intended to come into contact with foodstuffs.
Council Directive 96/11/EEC	Referring to plastics materials and articles intended to come into contact with foodstuffs.
Commission Directive 97/48/EEC	2nd amendment of Council Directive 82/711/EEC laying down the basic rules necessary for testing migration of the constituents of plastics materials and articles intended to come into contact with foodstuffs.

for a proper safety assessment. Substances included in lists are excluded from the positive lists in CEC Directives. Finally, lists 6–9 contain approximately 80% of monomers and 60% of additives, which are included in Section B of the Directive (Table 4). Currently, the SFC cannot offer any reassurance regarding the safety of these substances, since there are inadequate data for a proper safety assessment. Other methods may involve supercritical fluid chromatography (SFC), HPLC (High Performance Liquid Chromatography), FTIR (Fourier transform infra red), electronic spectroscopy, or proton nuclear magnetic resonance (^{1}H-NMR) (Table 5).

Table 3 EEC directives related to specific packaging materials[29–41]

		Individual materials and substances			
Directives	Applicable to all materials	Plastics	Regenerated cellulose	Ceramics	Elastomers
76/893/EEC	√				
78/142/EEC		√			
80/590/EEC	√				
80/766/EEC		√			
82/432/EEC		√			
83/299/EEC		√			
84/500/EEC				√	
85/572/EEC		√			
86/388/EEC			√		
89/109/EEC	√				
90/128/EEC		√			
92/15/EEC			√		
92/39/EEC		√			
93/8/EEC		√			
93/9/EEC		√			
93/10/EEC			√		
93/11/EEC					√
95/3/EC		√			
96/11/EC		√			
97/48/EC		√			

In the USA, the structure of the regulations for food packaging materials is more complicated. Premarket approval by the FDA is currently required for food packaging materials used in the US that are not GRAS (generally recognized as safe), prior sanctioned, or not reasonably expected to become a component of food. These components or food additives must be shown through the food additive petition process to be safe for their intended use.

In the Federal Register of July 17, 1995, the FDA established a "threshold of regulation" process. The likelihood of a substance posing a health hazard depends on its dietary concentration and toxic potency. The agency considered both factors in establishing a threshold of regulation level. Directive 85/572/EEC defines the test conditions relevant to the intended conditions of use and lists the food simulants for use with the migration tests (Table 6). This process was established in order to improve and speed up the food additive petition process, because, in some food packaging applications, the amount of migration could be considered so small as to seen negligible. Non-carcinogenic compounds would be unlikely to occur at a level lower than 1 mg/kg. To provide

Table 4 SFC classification scheme for substances used in food packaging materials and number of substances classified into each list by SFC up to 1991 adapted from ref[42–43]

List	Monomers	Additives	Explanation
0	50	36	Substances that pose no problem. Food ingredients or normal body metabolites (e.g. urea, glucose).
1	29	121	Used as direct food additives. ADI/TDI has been set by the SFC/ FAO/WHO. Few substances not directly food additives (iodine, bromide, copper, phosphorus) are also included.
2	86	220	Substances, not naturally present in foods or in the body, are not direct food additives. TDI has been set by SFC on the basis of available toxicity data.
3	50	66	Insufficient toxicity data to set an ADI or TDI, but are acceptable for use. Some are unlikely to be present in food other than in very small quantities because they possess properties which render levels in food self limiting (eg. butane, ethylene, chlorine). Inert silicates are of low or very low migration.
4	36	8	Toxic substances that their use is acceptable provided there are no detectable residues in food.
5	5	12	The data for substances in this category indicate that they should not be used (e.g. asbestos, morpholine containing compounds). Compounds with market potential for bioaccumulation (highly halogenated compounds) are equally included.
6	114/10	15/52	Insufficient data but serious toxicity suspected Suspect toxicity: A carcinogenicity B other toxicity Restrictions are set on the use of these substances.
7	158	122	Some toxicity data exist but insufficient to set a TDI. No serious toxicity alerts, further data required.
8	430	158	Few relevant or no toxicity data available. Data must be submitted according to SFC guidelines.
9	243	183	Substances or individual substances with inadequate chemical descriptions which are not properly identified. They must be properly specified before the SFC can consider them.
Total	1211	993	

ADI: Acceptable Daily Intake.
TDI: Tolerable Daily Intake.

an adequate safety margin, however, the dietary concentration should be well below 1 mg/kg. The process excludes materials from food contact articles, the use of which results in a dietary concentration at or below 0.5 ppb (μg/Kg) from the food additive listing regulation requirements. Carcinogens suspect and carcinogens are excluded from this regulation. A 0.5 ppb threshold is 2000 times lower than the dietary concentration at which the majority of examined compounds are likely to cause non-carcinogenic toxic effects and 200 times lower than the chronic exposure level at which potent pesticides display toxic effects. Thus, the FDA has determined that most known carcinogens pose less than a one in a million lifetime risk if present in the diet at 0.5 μg/kg.[4,6,29–42,44–68]

MIGRATION OF SUBSTANCES

Generally, the migration is considered to be a process where polymerization residues or stabilizers can diffuse through the polymer matrix to the surface. Diffusion is considered to be one of the main mechanisms for the transfer and migration of chemical compounds from packaging materials to food. The migration of ingredients from packaging material into food is undesirable. However, a certain transfer is unavoidable, because most foodstuffs are packed prior to their purchase by the consumer. The contamination is usually due to the dissolution in the food that come in contact with the migrated stabilizers on the polymer surface. The term "migration" is used to describe the process of mass transfer from a food packaging material to its contents. During this process, a packaging material comes in contact with food, and though its mechanical or diffusion properties are not altered, it may adversely affect the organoleptic properties of the packaged food. Figure 4 points out the possible routes of migration from a "boil in bag" package during reheating food in boiling water.

Many substances usually migrating from packaging materials to foods are plastic additives, monomers, oligomers, and contaminants. A wide variety of additives was developed and used during processing and fabrication of the polymeric packaging materials. Additives, such as plasticizers, antioxidants, light stabilizers, thermal stabilizers, lubricants, antistatic agents, and slip additives, usually abound in various types of plastics. Migrating solvents, such as adipic acid, toluene, butanone-2, ethyl acetate,

Table 5 Comparison of NMR and GC methods[116]

Task	NMR	GC
Fingerprint of potential migrant	Semi and non-volatiles	Semi-volatiles
Identification of functional classes of potential migrants	Possible if other than saturated hydrocarbons	Time consuming with MS
Identification of specific potential migrants	Possible with database	Possible with MS and database of retention indices

Table 6 List of food simulants for migration testing[68–70]

Simulant A	Distilled water
Simulant B	3% acetic acid (w/v) in aqueous solution
Simulant C	15% ethanol (v/v) in aqueous solution
Simulant D	Rectified olive oil, sunflower oil, or HB307

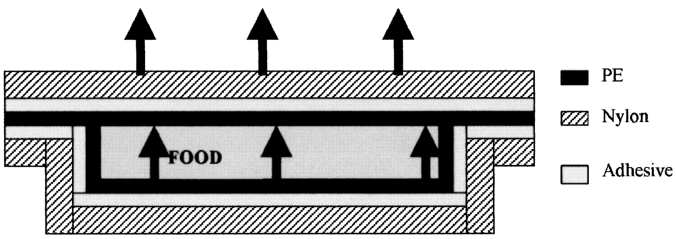

Figure 4 Possible routes for migration from a boil in bag package during reheating food in boiling water.[114]

and hexane are also of concern as well as pigments, such as molybdate orange.[4,69–77]

Plasticizers

Plasticizers are a group of additives used in plastic materials to improve the properties of the polymers. Butyl Stearate, acetyltributyl citrate, alkyl sebacates, and adipates are the most common types of plasticizers endowed with low toxicity, but with a potential carcinogenic and estrogenic effect.[4,78–80]

The migration of monomeric plasticizers is generally considerable in direct contact with fatty food and increases with temperature. The migration of plasticizers from plastics into food has been reported in several surveys.[81–96] The majority of these plasticizers has been related to the use of plastics, such as PVC. As a result of these studies, the packaging industry has replaced PVC with other polymers, such as PE or regenerated cellulose not associated with plasticizers.[97]

Hammarling et al. investigated ESBO (epoxidised soya bean oil), which is widely employed in plastics, such as PVC. They reported an average level of 2 mg/kg in baby food packed in glass jars with PVC gaskets.[78] SFC has proposed a TDI of 1 mg/kg body weight. According to their study, in order to not exceed the TDI, the ESBO level in baby food should not be higher than 31.5 mg/kg. DEHA (di-(ethylhexyl)adipate) is frequently used as plasticizer in PVC. Migration of DEHA from plasticized PVC occurs in a broad spectrum of foods wrapped both domestically and in retail shops. In an attempt to ensure the safety of consumers, the SFC recommended that the use of PVC film with a high level of DEHA plasticizer in direct contact with fatty foods should be restricted to keep the intake level below TDI of 0.3 mg/kg body weight.[78,98]

Thermal Stabilizers

In addition to plasticizers, thermal stabilizers are also widely employed in plastics. Generally, epoxidized seed and vegetable oils are often used in a wide range of food contact plastics, heat stabilizers, lubricants, and plasticizers as well. Their toxicity is strongly affected by their purity, since the residual ethylene oxide is quite toxic.[4] The first step of the stabilizer migration, namely the diffusion through the polymer matrix to the surface in contact with a food simulating solvent or other environments, has been the topic of several investigations over the last decades.

Slip Additives

Fatty acid amides are used as slip additives in a variety of plastics used for packaging, such as polyolefins, polystyrene, and PVC. Slip additives are added to plastic formulations, but they gradually manage to emerge and tend to bloom to the surface. They impart useful properties, including lubrication, to prevent films from sticking together or forming conglomerates and reduce the static charge.[4,62]

Light Stabilizers

Light stabilizers are used to improve the long term weathering characteristics of plastics, especially polyolefins. Polymeric hindered amines (HALS) are commonly used in polyolefins as light stabilizers.[4]

Antioxidants

Antioxidants are used to slow down the oxidation process the plastics undergo, due to light exposure. BHT and Irganox 1010 are the most commonly used antioxidants. Most antioxidants were found to be nontoxic and have a satisfactory stabilization effect. Migration rates of Irganox 1010 and Irganox 1076 were determined for several plastics[4,6,99] (Table 7). Figure 5 shows the migration of Irganox 1076 and Irganox 1010 from LDPE and PP into different foods. Figure 6 indicates oxygen transmission rates of LDPE with several antioxidants at various radiation doses.

Monomers

Monomers, as well as oligomers, are likely to migrate from packaging materials to food.[4,100–102] Monomers are reactive substances, with respect to living organisms, and, hence, potentially toxic. Therefore, hygiene regulations aim at restricting the content of residual monomers in the raw or starting materials, plastics and articles are made therefrom.

Residual monomers are always present in PS and several adverse and harmful effects are attributed to styrene. Styrene is one of the most important industrial chemicals and is the second most widely used monomer for the production of packaging polymers coming in contact with foods. Styrene content in food is mainly caused by the migration from polymer packaging material. The daily styrene exposure is estimated to range from 18.2 to 55.2 μg/person corresponding to an annual exposure of 6.7 to 20.2 mg/person. Humans have experienced irritation of the eyes, nose, throat, and skin, when exposed to its vapors. Styrene does not only have a toxic effect on the liver, but also acts as a depressant on the central nervous system, causing neurological impairment.[103–105]

Table 7 Migrating substance from packaging materials (other than polymer) to food and methods of determination

Packaging material	Food	Migrated substance	Method of determination	Ref
Wooden Packaging	Apples	1-propanol	HSC (Head space chromatography)	118
Tin	Canned foods	DGEBA	Liquid-liquid extraction/ solid-phase +HPLC	107
Metals/plastics/glass/aseptic	Tomato			16
Recycled Paper & board		DIPNs		102
Cans coated with lacquer		Epichlorohydrin		13
Paper cardboard & board	Test foods	Metals (Zn, Sn, Al, Mn, Ba)		119
Cartons (Al-laminated)	Liquid foods (skimmed milk & yogurt drinks)	Al		120
Aseptic	Milk	H_2O_2		113
Aluminum Foil paper laminates	Butter, margarine	Phthalate esters (DBP, BBP, DEHP)	GS-FID	121
Cans	Water based simulants	BADGE (lacquer)		12
Aluminum	Food & drinks	Al		122
Paper based food packaging	Fatty & nonfatty foods	2378-TCDD/ 2378-TCDF (polychlorinated dibenzofurans)		123
Ceramic containers	Dairy products	Pb, Cd		124
Aluminum	Milk	Al		125
Can	Canned foods	BADGE		102
Aluminum	Dairy products	Al		126
Paper & board		4,4-bis(dimethylamino benzophenone) (MK) 4,4-bis-(diethylamino benzophenone) (DEAB)	HPLC GC-MS	127

Vinyl chloride monomer is highly toxic, the levels of which in PVC food packaging materials are closely controlled. Both the residual monomer contents of the polymer and migration levels to foods or food simulants are regulated as well.

Epoxy resins of the Bisphenol A type, such as Bisphenol A diglycidyl ether (BADGE), are the starting materials for cold-cured epoxy resins destined for use as articles and/ or components of articles intended for use in manufacturing, packaging, and transporting food. Because of their high toxicity, it is necessary to control the unreacted products to prevent their migration into food. The EC positive list of monomers and other starting substances restricts residual levels of BADGE to 1 mg/kg in plastic food contact materials and sets a migration limit of "non-detectable" in foods or simulants with a required detection limit of 0.020 mg/kg.[12,65,96,106–107]

In the food packaging industry, isocyanates are used in polyurethane polymers and adhesives. They are toxic compounds, and their health effects are well documented. Also, their use in food packaging materials is regulated by EC Directives. Residual levels in finished plastics must not exceed 1.0 mg/kg.

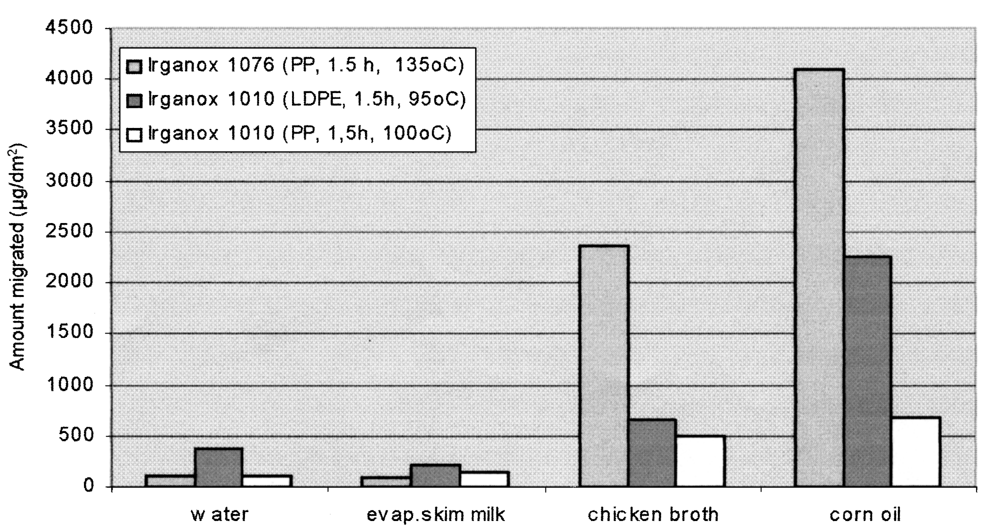

Figure 5 Migration of Irganox 1076 and Irganox 1010 from LDPE and PP into various foods.[69]

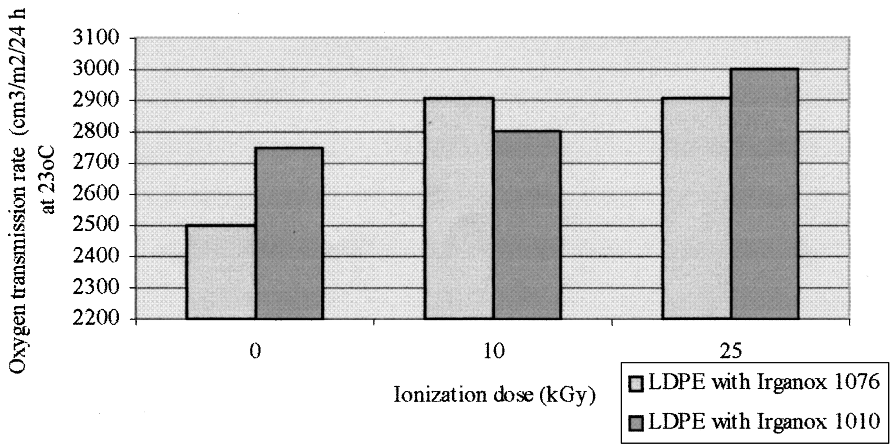

Figure 6 Oxygen transmission rates of LDPE with different antioxidants at various radiation doses.[69]

Polyamides, also known as "nylons," are of the few polymers that can be used to contain food for cooking. However, surveys indicate that small amounts of nylon oligomers, residual caprolactam, and monomer of nylon can migrate into boiling water. Usually, these compounds are not toxic, but may impart bitter taste to foods.[60,108]

Finally, polyethylene terephthalate (PET) is commonly used as packaging material for beverages and edible oils. PET has become a widely used polymer because its drawn films have high strength, flexibility, and clarity. A PET package that permits 10 μg/kg or less migration of a component is still considered acceptable.[109,110]

Table 8 Migrating monomer/oligomer from polymer packaging material to food and method of determination

Packaging material	Food	Monomer/oligomer	Methods of determination	Ref
PS	Instant foods	Styrene dimmers/trimmers	GC + GC mass spectrometry	128
PS cups	Yoghurt	Styrene	Purge & trap GS + MS	103
PS	Water, milk, cold & hot beverages, olive oil	Styrene		129
Polyester cookware	Olive oil	Benzene	Head space analysis	130
PVC films	Cheese	DEHA		131
LDPE	Food simulating	Irganox 1010 (I-1010)		99
HDPE	Liquids (FSL)	Irganox 1076 (I-1076)		
PP				
Microwave packaging		cPET		109
PVC film	Cheese sausages	Dioctyladipate		94
PVC films	Cheese	DEHA		132
Coating material (poly(vinyl alcohol) based liquid plast emulsion		Monomers of HA-LA plast		133
Polymeric materials	Dairy products	Styrene		20
PVC film	Cheese	DEHA		10
PP cups	Dairy products			134
R-PP				
Polystyrene	Dairy products	Styrene/ethylbenzene	Dynamic purge & trap GS	18
PP cups	Cheese sauce	2-decanone		135
PS (+ recycled material)	Dairy products	Monostyrene		136
PS + ABS + waxed paperboard	Dairy products	Mineral hydrocarbons	GS	137
Wax coatings	Cheese sausages	Mineral hydrocarbons	GS	138
Polymer	Milk	Dioctylphthalate	Chromatography	139
PS	Milk	Monostyrene		101
PP	yoghurt	monomers		140
PE	Cream fruit juices			
PS	Food oil	Styrene		141
PS	Cheese desserts meat products	Styrene	GS + headspace injection + MS	142
PVC	Cheese	DEHA		91
LDPE	Milk	Naphthalene		143
ABS	Dairy products	Mineral hydrocarbons		144
PC	FSL	Bisphenol-A (BPA)	HPLC	96
PVC films	Bread, olive oil, cheese, meat	DEHA		98
PVC	Microwave fatty foods			145

Contaminants

Apart from additives and monomer residues present in the packaging materials, other sources of food contamination have been reported as well. Decomposition products from additives or monomers will also migrate into the food under proper conditions. The presence of the residues of these chemicals may lead to contamination. Diphenylthiurea is used in the manufacturing of PVC film, benzene, dioxins,[111–112] processing agents (hydrogen peroxide),[113] and other volatiles and stands for some of the most representative residues.[114]

MIGRATION TESTING

Food industries and scientists are confronted with great difficulties in the attempt to carry out control of migration from packaging materials, because a number of problems usually arise. Official test methods are usually time-consuming, complicated, and impractical for routine or daily controls. Therefore, the EC accepted, in addition to official methods, more practical test methods. The methods used are based mainly on the extraction followed by chromatographic or spectrophotometric analysis.

FDA researchers adopted a procedure that combined on liquid-liquid partitioning of the corn oil in hexane with acetonitrile, followed by HPLC analysis with ultraviolet detection.[109] In 1988, a procedure was developed using infrared spectroscopy (IR), gas chromatography (GC), and GC-mass spectrometry (GC-MS), which is a quite sensitive method suitable for routine control.

Due to the complexity of foodstuffs and the variety of conditions arising from contact with plastics, a series of conventions is necessary to make the quantification of specific and overall migration feasible. A comparative study from different

Table 9 Effect of new processing techniques on the barrier and mechanical properties of packaging materials[146–157]

		Properties	
Packaging material	Process	Mechanical	Gas/water barrier
LDPE	Ionizing radiation		No change in oxygen permeability
OPP	Ionizing radiation		No change in oxygen permeability
PE pouch	Ionizing radiation		No change in oxygen and vapor permeability
PE	Ozone	Decrease or increase in elongation depending on exposure time	
PS powder	Ozone	Brittle and opaque	
PP	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
EVA	Ionizing radiation	Decrease in heat seal strength	Increase in diffusivity and decrease in solubility to volatile compounds
Virgin silicon membrane	Ozone		Increase in oxygen permeability
Sodium caseinate	Ultrasound	Increase in tensile strength and puncture resistance and no change in elongation	
Surlyn	Ionizing radiation	No significant change in tensile strength, elongation, Young's modulus, tear strength and heat seal strength	
PET/PVdC/PE	Ionizing radiation		Decrease in oxygen permeability
PP/EVOH/PP	High pressure	No significant change in tensile strength and heat seal strength	No change in oxygen and water vapor permeability
OPP/PVOH/PE	High pressure	No significant change in tensile strength and heat seal strength	No change in oxygen and water vapor permeability
KOP/CPP	High pressure	No significant change in tensile strength and heat seal strength	No change in oxygen and water vapor permeability
PET/Al/CPP	High pressure	No significant change in tensile strength and heat seal strength	No change in oxygen and water vapor permeability
PET/SiO_X/PU ADH/ LDPE	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
PET/ Al_2O_3/PU adh/ LDPE	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
PET/PVdC/ Nylon/HDPE/PP	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
PE/Nylon/EvOH/PE	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
PE/Nylon/PE	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
PET/EVA	High pressure	No significant change in tensile strength, but increase in percent elongation	Less than 20% increase in O_2, CO_2, and $H_2O(v)$ permeability
Met-PET12	High pressure	No significant change in tensile strength, but increase in percent elongation	More than 50% increase in O_2, CO_2, and $H_2O(v)$ permeability

laboratories presupposed a standardization of test conditions and an internationally accepted selection of reproducible food simulating agents.

The measurement of overall migration into aqueous food simulants, such as A, B, and C, is relatively straightforward, and generally, no serious problems are encountered. However, the determination of the overall migration into simulant D, as defined in Directive 85/572/EEC and 93/8/EEC, is time consuming and difficult to carry out for some packaging materials. De Kruijf and Rijk proved that many of the practical problems in the experimental determination of the overall migration into the commonly used simulant D can be overcome through the application of a volatile solvent as a fatty food simulant.[63] In "Practical Guide N1" of the Commission of the EC of 2 April 1993, the following alternative EEC fatty food simulants are specified: iso-octane, 50% ethanol/water for relatively polar polymers, such as PVC, PET, and PS; 95% ethanol/water for nonpolar polymers, such as polyolefins.[20,44,109,115–117] Tables 7 and 8 summarize the results of many migration studies focused on various packaging materials in contact with enclosed foods and the corresponding methods employed for the determination of migrated substances. It has been found that most of the new processing techniques, particularly ionization and high pressure, negatively effect the performance of packaging materials. Table 9 summarizes the effect of the processing technologies on the mechanical and gas/water vapor barrier properties of several homopolymers and laminated films.

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