

Migration and Safety of Plastic Food Packaging Materials

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Contributor: Andreas Panou, Ioannis Konstantinos Karabagias

The conventional packaging materials used for the preservation of foods may exhibit many disadvantages that are related to the migration of micromolecular chemical substances incorporated in the packaging material net to the packaged food. There are many chemical substances in the matrix of plastic packaging materials and epoxy-resins that are used in food packaging materials, varnishes, and can coatings. Many migrants have high toxicity, such as acetaldehyde, antimony, antimony (III) oxide, 2,4-di-tert-butylphenol, tris (2,4-di-tert-butylphenol) phosphate, tris(2,4-di-tert-butylphenyl) phosphite, *bisphenol A*, and the plasticizers di(2-ethylhexyl) phthalate, di-n-butyl phthalate, benzyl-butylphthalate, di-isononylphthalate, and di-isododecylphthalate.

Keywords: plastics ; migration ; foods ; consumers ; safety ; natural films and coatings

1. Introduction

A matrix of plastic packaging material, except for macromolecular substances, consists of micromolecular substances such as metals as catalysts, very low amounts of solvent and monomers, and various reaction products that are produced during the manufacturing of plastic materials. Thermal decomposition products and various additives are added for the improvement of plastic materials' disadvantages, such as plasticizers, stabilizers, antioxidants, extinguishers, etc. When a plastic packaging material is in contact with food, the transfer of micromolecular substances from the matrix of the plastic material to the food occurs. This phenomenon is called migration, and the migrating substances are named migrants ^[1]. Regarding migration, when the migration of a specific substance occurs, it is named specific migration, while when migration concerns all the migrating substances, it is named overall migration. There are migration cases that lead to desirable results, such as the incorporation of antistatic media into packaging materials and the controlled diffusion of medicines from the net of plastics ^[1]. However, in most cases, migration is undesirable because of the diffusion of harmful chemical substances such as plasticizers from plasticized PVC (in toys and shower curtains), flame retardants (in plastic casings of televisions and computers), and various harmful chemical substances into foods and medicines, which can deteriorate the taste of food and the active compound of the medicine.

The process of migration can be divided into four stages:

- (1) Diffusion of the migrants through the polymeric molecules of the plastic packaging material;
- (2) The desorption of substances from the surface of the packaging material;
- (3) The absorption of substances from the interface between the food and packaging material;
- (4) The absorption of substances onto the food ^[2].

The main factors that are responsible for the migration of these micromolecules are as follows: (a) the presence of attractive forces between the micromolecules and the food constituents, (b) the low dimensions and molecular weight of the micromolecules, and (c) the volatility of the micromolecules. For example, low-size molecules (monomers and debris solvents) have low boiling points and exhibit high migration levels. Some monomers, such as formaldehyde, vinyl chloride, ethylene, and butadiene, present high migration levels even at ambient temperature ^[3]. Regarding molecular weight, a substance of high molecular weight presents low migration levels on account of its large dimensions. The migration levels are also influenced by the concentration of substances inside the plastic packaging material, the thickness of the packaging material, and the crystallinity and surface structure of the packaging material ^[3]. Migrants are divided into two categories: (i) IAS and (ii) NIAS ^[1]. The first type includes substances such as additives, monomers, metals, and various starters, and the second type includes substances such as polymer decomposition products and products from various

reactions. The produced degradation products depend on the type of polymer, the degradation mechanisms, and environmental factors, such as temperature and oxygen. During the thermal degradation of plastic materials containing nitrogen (nylons, polyacrylonitrile, and *polyurethane*), HCN is produced, plastic materials containing chloride (PVC, PVdC) produce HCl and dioxins during thermal degradation, and plastic materials containing fluorine (*polytetrafluoroethylene*, *polyvinyl fluoride*) produce HF and perfluoroisobutene. Polymers such as polystyrene, polyesters, polycarbonates, polyamides, and polyurethanes can be depolymerized via chain scission of their initial monomers [4][5][6].

Moreover, regulation of the “limits of specific migration” has been established by the European Safety Food Authority (ESFA) based on data on the toxicity of each substance. For the total quality assurance of plastic packaging materials, the European Commission 2002a [7] established the directive 2002/72/EC, which regulates the application of monomers, additives, and their residual amounts in packaging, as well as their migration limits for the manufacturing of plastics that are intended to come in contact with foods (food-contact materials). As far as coatings are concerned, the resolution AP(96)5 (Council of Europe 1996) was edited [8], adopting a list of approved materials (starting materials) and additives of the directive 90/128/EEC (European Commission 1990, preceding directive of 2002/72/EC).

The use of food simulants to imitate real food samples was introduced for migration tests of all food-contact materials, in particular, plastic food -ontact materials in the European Council Directive 85/572/ECC in 1985 [9]. This directive was then replaced in 2011 by the regulation EC 10/2011 issued by the European Commission [10], which specifies further migration test conditions, again focusing on migration from plastic food-contact materials. To ensure the suitability of the presented food simulants, the associations between real food, food simulants, and migration conditions were thoroughly established [11], even though, in 2016, this directive was amended and corrected [12].

The limit of overall migration should not exceed 60 mg/Kg in food or 10 mg/dm² in food-contact material. This regulation also sets out the use of representative simulants for each food category [9]. The reasons for the substitution of real foods with food simulants in migration studies are the complexity of foods, the presence of difficulties during analysis, and the increase in the reproductivity of the results. Indeed, the selection of simulants depends on the food matrix to be imitated. More specifically, a mixture of ethanol/water (50:50, v:v) should be applied to imitate food with lipophilic properties, alcoholic food (alcohol content > 20%), and oil-in-water emulsions. Furthermore, a mixture of ethanol/water (10:90, v:v) is recommended by the European Union for the simulation of food with hydrophilic properties [10]. Real food samples typically consist of a complex composition of multiple components in equilibrium that interact and define the properties of each food. Therefore, the selection of suitable food simulants is of great interest and might require a general hypothesis [11]. According to the FDA [13], the food simulant contents that are recommended are as follows: for aqueous and acidic foods, 10% ethanol is recommended; for low- and high-alcoholic foods, 10% or 50% ethanol is proposed; and for fatty foods, food oils such as corn oil, synthetic glycerides (HB307), and miglyol 812 derived from coconut oil have been proposed.

2. Food Simulants

The basic food simulants that are proposed by the Commission Regulation (EU) 2016/416 [12] represent five different food categories and are listed in **Table 1**.

Table 1. Simulants according to Commission Regulation (EU) 2016/416 [12].

Simulant	Food Category	Shortcut
10% ethanol (v/v)	Hydrophilic foods	A
3% acetic acid (w/v)	Acidic foods with pH below 4.5	B
20% ethanol (v/v)	Alcoholic foods with an alcohol content of up to 20%	C
50% ethanol (v/v)	Alcoholic foods with an alcohol content of above 20%	D1

Simulant	Food Category	Shortcut
Vegetable oil	Fatty foods	D2
Poly(2,6-diphenyl-p-phenylene oxide), particle size: 60–80 mesh, pore size: 200 nm, commonly named Tenax)	Dry foods	E

3. Migrating Chemical Substances

Regarding the specific migration of starting materials and the products of the reactions that happen in can coatings, the 10 derivatives of BADGE and Bisphenol F diglycidyl ether (BFDGE) are limited to 1 mg/Kg of food according to the directive 2002/16/EC (European Commission 2002b) ^[14]. However, most of the starting materials for the coatings are a mixture of substances or prepolymers that react with other substances in the coating. The migrating substances from the coatings are monomers that belong to a positive list. The Scientific Committee on Food (SCF, European Commission 2001) ^[15] emphasized the importance of the identification of migrating substances, and their quantity and toxicity. Therefore, only substances with a molecular weight below 1000 Da are cause for concern because these can be absorbed so easily by the gastro-intestinal tract.

The most common chemical substances that migrate to foods are the following: acetaldehyde, methanal, antimony, Sb₂O₃, 2,4-di-tert-butylphenol, tris (2,4-di-tert-butylphenol) phosphate, Irgafos 168, BPA, and the plasticizers DEHP, DBP, BBP, DINP, and DIDP.

Table 2 lists the migrant substances and their toxic effects on human health.

Table 2. List of migrant substances and their toxic effects on human health.

Migrant Name	Toxic Effects and Health Issues	References
Acetaldehyde	Facial flushing, nausea, vomiting, tachycardia, hypotension, hepatitis, liver cirrhosis, and in extreme cases, even death.	^{[16][17][18][19]}
Antimony and Antimony(III) oxide	Carcinogenic potential for humans.	^{[20][21]}
Irgafos 168	Not any significant toxicological effects.	-
BPA	Negative effect on estrogen, great inhibitor of endocrine gland, Negative effect on the metabolic homeostasis of the progeny, sex steroids, and thyroid hormones levels during pregnancy. Epigenetic effects and developmental neurotoxicity.	^{[22][23][24][25][26][27]}

Migrant Name	Toxic Effects and Health Issues	References
Derivatives of BPA and other bisphenols	Negative effect on the regulation of metabolic homeostasis of the progeny, sex steroids, and thyroid hormone levels during pregnancy. Epigenetic effects and developmental neurotoxicity in zebrafish.	-
Melamine	Increase in urea content, increase in nephrotoxicity, i.e., kidney stones. Cannot be decomposed in activated sludge treatment systems.	[28][29]
PFOA	Liver dysfunction, testicular and liver cancers, high levels of cholesterol,	[30][31][32][33][34][35][36]
PFOS	ulcerative colitis, and low response of immune system to vaccines.	[37][38][39][40][41]
	Endocrine disruption, various cancer forms, immune and metabolic diseases, abnormalities.	
	In new-born babies, preterm birth, abnormal birth length and weight and head circumference,	
Phthalates	and altered reproductive hormone levels.	[42]
	During childhood, asthma and allergic symptoms, obesity, anti-androgen effects, delay in growth and puberty, and changes in systolic blood pressure.	

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