SAFETY ASPECTS RELATED TO OVERALL MIGRATION OF COMPONENTS FROM FOOD CONTACT MATERIALS USED IN BAKERY AND PASTRY INDUSTRY

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Abstract

The packaging is widely used in the food industry, for the chemical, biological and physical protection of food products from spoilage and damage throughout the food chain. Thus, its role is to maintain the quality of the product, creating an essential environment for preserving the quality of the packaged product, thus contributing to reducing food waste and reducing the use of preservatives in food. Another very important role of packaging is to ensure the traceability of the food product, which is the ability to follow its path, throughout all stages of the technological flow, namely production, processing and distribution. Among the most used packaging materials in the food industry, are plastics made of polyethylene, polypropylene, polyethylene terephthalate, complex plastics, followed by those obtained from paper or cardboard, glass, metal materials. Each type of packaging has its own production technology, its own functions, its own cost, advantages or disadvantages. The biggest inconvenience of these packaging materials is the possibility of migration of compounds with toxic effect in the packaged product and then ingested by consumers. This process is generically called the migration process and can be global migration, represented by all the compounds that can migrate from the package, or specific migration, represented by the migration of certain monomers and oligomers from the package into the packaged product. The purpose of this study is to test, in terms of global migration of components, the main types of packaging used in the milling and bakery industry available on the Romanian market, to assess the possibility of migration of compounds from these packages, which may affect product quality.

Key words: bakery industry, food simulants, overall migration, food contact materials.

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INTRODUCTION

Packaging is the totality of packaging materials intended to contain or temporarily contain a packaged product (Hong et al, 2021). Their main objective being to preserve and provide physical, chemical and microbiological protection to packaged products against odors, dust, shocks, microorganism, humidity, light, temperature, physical damage, breakages, vibrations, during storage and distributions, which can affect their quality or safety, to minimizing food losses and wastage (Han et al, 2018, Alamri et al, 2021, Gupta and Dudeja, 2017). Other functions of the packaging are to give a pleasant appearance to the product, to provide information to the consumer about the packaged product (content, expiration date, production date, nutritional values, details about the manufacturer, its quantity) (Gupta and Dudeja, 2017), to transport articles, to handle (Hong et al, 2021), but also, containment, convenience (Cruz et al, 2019).

Among the packaging materials used in the food industry are plastics, paper and cardboard, glass, metallic materials, wood (Hong et al, 2021), or laminates (Alamri et al, 2021), but the most used by various food industries are plastics (Cruz et al, 2019). In the food industry, different types of plastics are used, such as high-density polyethylene (HDPE), low-density polyethylene (LDPE), medium-density polyethylene (MDPE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), polycarbonate. (PC), polyvinylidene chloride (PVDC), or multilayer materials, obtained by combining several types of plastics (Cruz et al, 2019).

The general properties of packaging materials are, barrier properties (gas, water vapor, aroma barrier), antimicrobial properties, mechanical properties (breaking strength, tensile strength), optical characteristics, thermal properties, eco-friendliness (Hang et al, 2021). Besides these properties, plastics have some advantages such as their low price, softness, strongness, odorless, waterproof, low weight, require little energy to produce (North and Halden, 2013, Wu et al, 2017).

However, these materials are not very safe for contact with food, as a number of chemical compounds, such as additives, raw material residues, neo-forming molecules, by-products resulting from synthesis processes, antioxidants, plasticizers, monomers, oligomers, can also be transferred into the packaged product, during processing or packaging (Alamri et al, 2021, Wu et al, 2017). Packaged products can interact with the packaging material, which can cause some changes in both food safety and the mechanical and barrier properties of the packages. This transfer of compounds from the packaging material to the product is called the generic migration of components. This process is very important in the food industry, for the selection and conditions of use of packaging, to avoid the production of possible side effects due to the presence of these chemicals (Traistaru et al, 2013).

This migration process is a very complex one, which is based on a diffusion phenomenon, where the molecules migrate from a higher concentration gradient (food-contact layer) to a lower concentration gradient (food surface). It is considered that the migration process has 4 major stages, namely the diffusion of chemical compounds in the matrix, the desorption of molecules from the polymer surface, the sorption of migrants and the polymer - food interface, the desorption of chemical compounds in the packaged product (Gavriil et al, 2018). This diffusion phenomenon is dependent on several factors, such as the concentration of the migrant in the packaging material, the diffusion coefficient, the diffusion time, the partition coefficient, the distance between the packaging material and the packaged product (Alamri et al, 2021). A diagram of this phenomenon is presented in Figure 1 (http://www.actinpak.eu/wp-content/uploads/2016/04/Sara-Limbo.pdf).

Regarding component migration, there are 2 terms that can define this process, namely global migration, which represents the totality of compounds that can migrate from a certain surface of the packaging material, and specific migration, which refers to the migration of a single chemical compound. It can be influenced by the nature and composition of the packaged product, the type of contact (direct or indirect), duration of contact, temperature of contact, nature of packaging material, migrant characteristics, migrant concentration in the packaging material (Alamri et al, 2021).

The evaluation of global or specific migration is performed with the help of food simulants, recommended by different authorities. Regulation EU no. 10/2011 mentions a series of food simulants, presented in Table 1. The same regulation presents a series of conditions of global migration of components, depending on the use of the packaging material. These extraction conditions are shown in Figure 2.

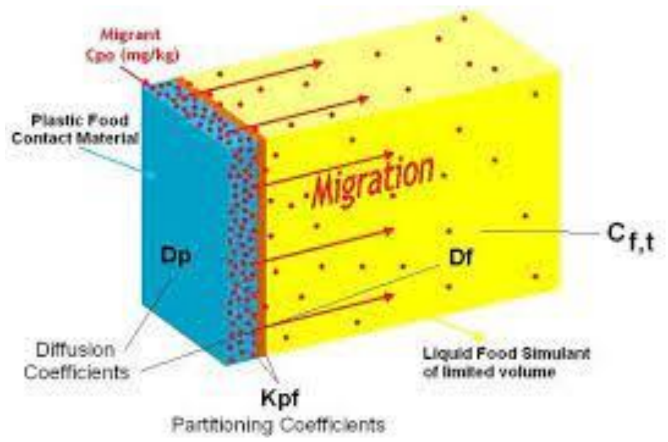

Figure 1. Migration process

Table 1. Food simulants (Regulation UE no. 10/2011)

Food simulant	Abbreviation	Assignment of food simulants to foods
Ethanol 10 % (v/v)	Food simulant A	Hydrophilic food character
Acetic acid 3% (v/v)	Food simulant B	Hydrophilic food character and shall be used for those foods which have a pH below 4.5
Ethanol 20 % (v/v)	Food simulant C	Alcoholic foods with alcohol content up to 20%
Ethanol 50 % (v/v)	Food simulant D1	Alcoholic foods with alcohol content above 20%
Vegetable oil	Food simulant D2	Foods with free fats at the surface
Poly (2,6-diphenyl-p-phenylene oxide)	Food simulant E	Dry foods

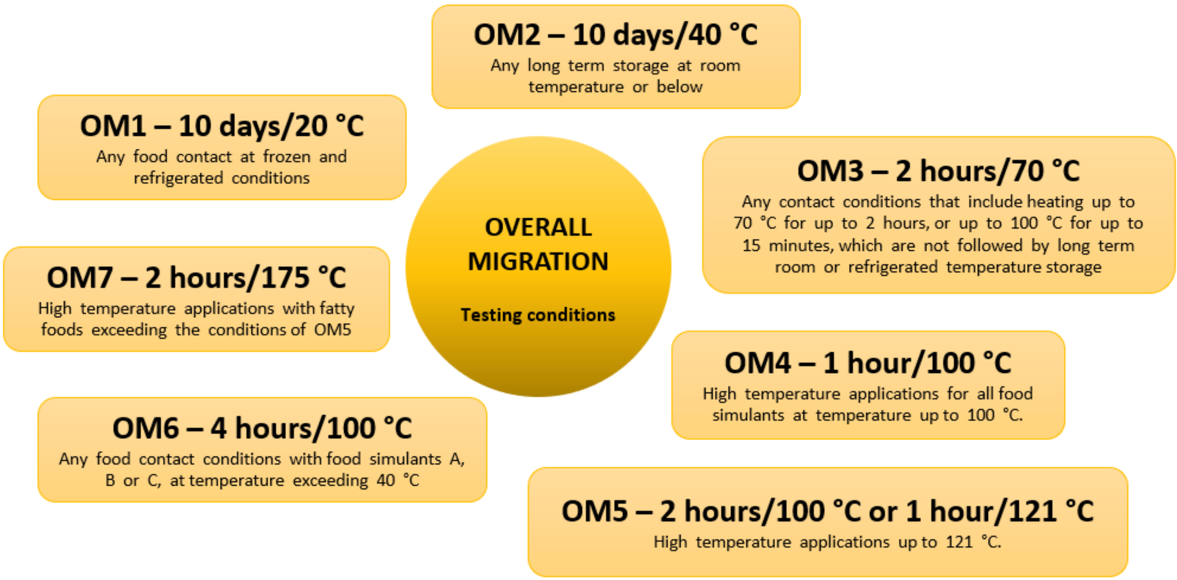

Figure 2. Overall migration testing conditions

These migration tests are time consuming, expensive (especially component-specific migration), and complicated to carry out, which is why in recent years there has been an increasing emphasis on mathematical modeling (Traistaru et al, 2013).
The aim of this study is to evaluate the global migration of components from different packaging materials used in the bakery and confectionery industry, using food simulants and in different weather and temperature conditions.

MATERIALS AND METHODS

Sample collection

In this study, 31 common samples of food contact materials used in bakery industry, were tested. These packages were purchased from different Romanian producers and kept in laboratory conditions until analysis. These samples include polyethylene (PE) bags, polypropylene (PP) bags, polyethylene terephthalate (PET) pans, polystyrene (PS) pans, PE laminated cardboard boxes, cardboard boxes, paper bags with BOPP window, kraft paper bags, but also packaging obtained from multilayer materials. These packaging materials can be primary or secondary packaging. The samples are briefly presented in Table 2.

Table 2. Presentation of tested samples

Sample no.	Sample	Characterization
1	BOPP pastry foil	Transparent, unprinted, BOPP foil
2	LDPE bread bag	Transparent, printed, LDPE bag
3	HDPE bread bag	Transparent, printed, HDPE bag
4	BOPP bread bag	Transparent, printed, BOPP bag
5	CAST bread bag	Transparent, printed, CAST bag
6	Laminate bread bag	Printed CAST + metallic BOPP bag
7	BOPP biscuits bag	Transparent, unprinted, BOPP bag
8	PET container for salt shakers	Transparent PET container with transparent lid
9	PET casseroles for biscuits	Transparent PET casserole with transparent lid
10	PET cups for mouse	Transparent PET cups for mouse
11	PS casseroles for biscuits	Transparent PS casserole with transparent lid
12	Paper bags with BOPP window	Kraft nature paper bags with BOPP window
13	Paper bags with BOPP window	Kraft white paper bags with BOPP window
14	Ziplock paper bags with PE window	Kraft nature ziplock paper bags with PE window
15	Cardboard boxes with display	Cardboard GC1 cake boxes with PET display
16	Wood tray for baking with PET	Nature wood tray for baking, laminated with PET
17	Wood tray for baking with PET and parchment paper	Nature wood tray for baking, laminated with PET and parchment paper
18	Wrapping paper	White wrapping paper
19	Paper corners	Kraft nature paper corners, printed
20	Brioche forms	Paper brioche forms, colored, printed
21	Paper cheeses	Paper cheeses, colored, printed
22	Golden pastry lace	Paper golden pastry lace, square shape
23	Golden discs and plates for pastry	Square cardboard discs and plates, laminated
24	Silver discs and plates for pastry	Cardboard discs and plates, laminated, oval shape
25	Cardboard trays with silicone paper	Cardboard trays with silicone paper, printed
26	Fat resistant paper bags	White fat resistant paper bags
27	Paper bag with handle	Nature paper bag with handle, printed
28	Cardboard tray for cakes	Cardboard tray, unlaminated, printed
29	Cardboard boxes for cookies	Printed cardboard boxes for cookies
30	Cardboard boxes laminated with Al	Printed cardboard boxes laminated with aluminum
31	Al baking trays	Aluminum baking trays for brioches

Reagents

Ethyl alcohol (C_2H_5OH), acetic acid (CH_3-COOH) and isooctane (2,2,4-Trimethylpentane) were purchased from Merck. All solutions were made with ultrapure water (18.2 MΩ.cm). Glassware was cleaned and decontaminated with 10% HNO_3 solution.

Sample preparation

Testing of global migration of components was performed according to the standard SR EN 1186:2003, parts 1, 2, 3, 7 and 9, a gravimetric method that measures all the chemical compounds that cand migrate from the package to the food product. According to this standard, the preparation of plastic samples for extraction was performed.

Among the test conditions presented in Regulation EU no. 10/2011, those used in this study were 10 days at 40^{0}C, namely OM2, 2 hours at 70^{0}C (OM3) and 1 hour at 100^{0}C (OM4). The uses of these conditions are presented in the Figure 2. The evaluation of the global migration of components was performed with the help of food simulants (Table 3), presented in the EU Regulation no. 10/2011. Of these, those used in the study were simulant A (10% ethyl alcohol), simulant B (3% acetic acid) and simulant D2 (olive oil). To test the conformity of packaging materials used for

Table 3. The assignment of food simulants, test conditions and test methods for tested samples

Sample	Assigned simulants	Test condition	Test method
1	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
2	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
3	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
4	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
5	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
6	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
7	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
8	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
9	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
10	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
11	A, B, D2	OM2	SR EN 1186-7:2003, SR EN 1186-2:2003
12.1	Paper bag - A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
12.2	BOPP window - A, B, D2	OM3	SR EN 1186-7:2003, SR EN 1186-2:2003
13.1	Paper bag - A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
13.2	BOPP window - A, B, D2	OM3	SR EN 1186-7:2003, SR EN 1186-2:2003
14.1	Paper bag - A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
14.2	PE window - A, B, D2	OM3	SR EN 1186-7:2003, SR EN 1186-2:2003
15.1	Cardboard - A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
15.2	BOPP window - A, B, D2	OM3	SR EN 1186-7:2003, SR EN 1186-2:2003
16	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
17	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
18	A, B, D	OM2, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
19	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
20	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
21	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
22	A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
23	A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
24	A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
25	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003
26	A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-2:2003
27	A, B, D	OM3, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-2:2003
28	A, B, D	OM2, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-2:2003
29	A, B, D	OM2, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-2:2003
30	A, B, D	OM2, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-2:2003
31	A, B, D	OM4, 24 h 40^{0}C	SR EN 1186-7:2003, SR EN 1186-15:2003

contact with high fat foods, and which cannot be tested in simulant D2, the isooctane simulant was used (simulant D), tested 24 hours at 40^{0}C (SR EN 1186-15:2003).

After the end of the extraction period, the resulting extract is evaporated on a water bath in platinum capsules, obtaining the value of the global migration of components in ppm or mg/dm^2.

The assignment of food simulants, of the test conditions and the test methods are presented in Table 3. The simulants and test conditions were chosen so as to simulate as well as possible the real conditions of use of the test packages.

RESULTS AND DISCUSSION

The results of the migration tests performed for the samples from Table 1, are presented in Table 5. According to the EU Regulation no. 10/2011, the value of the global migration of components for each simulant must be below the maximum allowed limit of 10 mg/dm^2 (60 mg / kg).

Although this regulation refers to plastics, and

Table 4. Overall migration results

Sample	Simulant A (mg/dm^2)	Simulant B (mg/dm^2)	Simulant D (mg/dm^2)	Simulant D2 (mg/dm^2)
1	0.92	1.08	-	1.31
2	0.92	1.17	-	1.03
3	1.08	1.25	-	1.36
4	0.83	1.08	-	0.97
5	0.83	1.17	-	1.45
6	1.08	1.33	-	1.22
7	0.83	1.17	-	1.06
8	1.08	1.42	-	1.76
9	1.0	1.50	-	1.62
10	1.08	1.58	-	1.56
11	1.25	1.58	-	1.82
12.1	1.58	2.08	2.25	-
12.2	0.92	1.08	-	1.26
13.1	1.42	1.83	2.33	-
13.2	0.83	1.08	-	1.13
14.1	1.67	2.33	2.42	-
14.2	0.92	1.17	-	1.27
15.1	2.58	3.17	3.75	-
15.2	1.08	1.50	-	1.41
16	3.67	5.08	4.92	-
17	4.08	5.33	5.17	-
18	1.50	2.08	1.83	-
19	1.33	1.92	1.42	-
20	1.58	1.83	1.92	-
21	1.42	2.08	1.67	-
22	2.17	3.33	2.17	-
23	2.33	3.58	4.42	-
24	2.17	3.67	2.67	-
25	3.42	4.17	6.67	-
26	1.25	2.08	1.33	-
27	2.08	3.25	2.83	-
28	2.58	3.42	4.33	-
29	2.25	3.67	4.17	-
30	2.0	3.08	3.83	-
31	2.33	3.58	4.08	-

the limit imposed is for this category of packaging materials, the values obtained for the other packaging (paper, cardboard), were reported at the same limit of 10 mg/dm^2.
Thus, as can be seen from Table 4, the results were below the imposed limit, and can be used, from this point of view, for contact with food.

CONCLUSION

In this study, a gravimetric method was used to determine all the compounds that can migrate from the packaging material to the packaged product. The selected samples, both from the bakery industry and from the pastry industry, were tested using standardized food simulants and under standardized test conditions, depending on the nature of the packaged product to come into contact with the packaging material, but and depending on the contact period and their contact temperature, so that the real conditions of use of the tested packaging can be simulated as well as possible.
The results obtained were below the maximum allowable limit of 10 mg / dm2 or 60 mg / kg for each simulant, which demonstrates the

conformity of packaging materials in terms of global migration of components.

In order to establish with certainty, the safety of these packaging materials, other, more detailed studies are needed, which involve the determination of several monomers or oligomers that have a specific migration limit imposed.

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REFERENCES

[1] Hong L. G., Yuhana N. Y., Zawawi E. Z. Z., Review of bioplastics as food packaging materials, **AIMS Materials Science**, vol. 8(2), 2021, pp. 166 – 184.

[2] Han J. W., Ruiz – Garcia L., Qian J. P., Yang x. T., Food Packaging: A comprehensive review and future trends, **Comprehensive Reviews in Food Science and Food Safety**, vol. 17, 2018, pp. 860 – 877.

[3] Alamri M.S., Qasem A. A. A., Mohamed A. A., Hussain S., Ibraheem M. A., Shamlan G., Alqah H. A., Qasha A. S., Food packaging's materials: A food safety perspective, **Saudi Journal of Biological Sciences**, vol. 28, 2021, pp. 4490 – 4499.

[4] Gupta R. K., Dudeja P., Food Safety in the 21st Century, Academic Press, United States, 2017, pp. 547 – 553.

[5] Cruz R. M. S., Rico B. P. M., Vieira M. C., Food Quality and Shelf Life, First Edition, Academic Press, United States, 2019, pp. 281 – 301.

[6] North E. J., Halden R. U., Plastics and Environmental Health: The Road Ahead, **Reviews on Environmental Health**, vol. 28(1), 2013,pp.1– 8

[7] Wu L. B., Su Q. Z., Lin Q. B., Peng W. D., Chen c. F., Wang Z. W., Hu C. Y., Impact of migration test method on the release of silver from nanosilver-polyethylene composite films into an acidic food simulant, **Food Packaging and Shelf Life**, vol. 14, 2017, pp. 83 – 87.

[8] Traistaru E., Rivis A., Moldovan R. C., Menelaou A., Georgescu C., Modelling migration from plastic packaging materials used in food industry, **Journal of Agroalimentary Processes and Technologies**, vol. 19(2), 2013, pp. 180 – 184.

[9] Gavriil Gavriil, Kanavouras A., Coutelieris F. A., Food packaging migration models: A critical discussion, **Critical Reviews in Food Science and Nutrition**, vol. 58(13), 208, pp. 2262 – 2272.

[10] http://www.actinpak.eu/wp-content/uploads/2016/04/Sara-Limbo.pdf accessed on 03.09.2021

[11] COMMISSION REGULATION (EU) No 10/2011 of 14 January 2011 on plastic materials and articles intended to come into contact with food.

[12] SR EN 1186-1:2003, Materials and articles in contact with foodstuffs. Plastics - Guide to the selection of conditions and test methods for overall migration.

[13] SR EN 1186-2:2003, Materials and articles in contact with foodstuffs. Plastics - Test methods for overall migration into olive oil by total immersion.

[14] SR EN 1186-3:2003, Materials and articles in contact with foodstuffs. Plastics - Test methods for overall migration into aqueous food simulants by total immersion.

[15] SR EN 1186-7:2003, Materials and articles in contact with foodstuffs. Plastics Test methods for overall migration into aqueous food simulants using a pouch.

[16] SR EN 1186-9:2003, Materials and articles in contact with foodstuffs. Plastics Test methods for overall migration into aqueous food simulants by article filling.

[17] SR EN 1186-15:2003, Materials and articles in contact with foodstuffs. Plastics Alternative test methods to migration into fatty food simulants by rapid extraction into iso-octane and/or 95 % ethanol.