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REVIEW PAPER

Packaging materials and articles for food: legislation and codes of good manufacturing practice

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Abstract

The basic objectives of food packaging materials are to provide safety and to prevent physical damage of the product, to retain or improve sensory characteristics, to offer a pleasing appearance, to be functional, to facilitate distribution, to give dimensional stability, but also to be compatible with the requirements regulated by the law. Consumers' demands for an extended shelf life of the product, as well as increased competition in the packaging industry, foster the need for continuous growth and application of innovative solutions for food packaging. While the traditional packaging acts as a passive barrier toward atmospheric effects, the innovative packaging systems offer "active" interactions with the foodstuff, providing increased shelf life and improved quality of the product. The selection of the appropriate packaging material is influenced by many factors, and, among many packaging materials offered on the market, polymer materials and articles have a significant place. On the other hand, some interactions occur between the plastics and the food in contact. The migration of the low-molecular-weight components from the polymer packaging to the packed food is emphasized when the so-called active and intelligent systems are used. Therefore, all the issues related to the packaging materials for food and the substances used as part of the packaging systems, are elaborated in the directives and regulations of the materials and articles intended to come in contact with the food.

Keywords: packaging materials for food, polymer packaging, food safety, legislation.

1. INTRODUCTION

The main objective of packaging materials is to provide safety of the product, i.e. to preserve its characteristics and features, to contribute to environmental protection, and to facilitate its transportation and storage. Polymer materials, due to their excellent barrier properties, replace the traditional materials, such as paper, cardboard, glass, aluminum, etc., in the production of different food packaging systems (Cvetkovska & Kujundziski 2008). Compared to the other materials used in food packaging, characteristics of polymer packaging materials such as small weight, good mechanical and physical properties, transparency, low price, etc., are considered as crucial during their selection in food packaging. In addition, some of the goals of the packaging material are the protection of food from atmospheric effects (such as oxygen, light and microorganisms), and also, prevention from changes of sensory characteristics by removing moisture and odours (McKeen 2013). On the other hand, in contact with the foodstuff, plastics are not completely inert, i.e. they allow mass transport (permeation, migration and sorption) of low-molecular-weight substances from the packaging material to the food, and vice versa (Cvetkovska & Kujundziski 2008; McKeen 2013). The characteristics that packaging materials usually need to satisfy are chemical stability (inaction toward fats and oils, acids and other aggressive media), state of aggregation (liquids, paste, powder), the presence of solid particles, sensitivity to moisture, air oxygen, odours (permeability of gases), to light and UV-radiation (opacity), and inertness toward environmental factors. In some cases, packaging materials are required to fulfill certain specific requirements, such as controlled oxygen access or minimal vapour permeability, or some specific features that need to be satisfied as a result of technological processes or conditions after the food production: contact freezing, sterilization, pasteurization, heating of cooked food, conservation using different aseptic methods by means of UV-radiation, H_2O_2, ethylene oxide and ionizing radiation. Different aspects of the demands imposed on the packaging materials are summarized in Table 1 (Marsh & Bugusu 2007).

Polymer packaging that successfully satisfies all requirements for preservation of food quality is usually produced in the form of one-layered foils, multi-layered systems (laminates of a polymer layer with other layers of polymers, paper or aluminum, prepared to improve and /or to acquire additional properties of the packaging material), or thermoforming foils. Rigid plastic (virgin or combined with other materials) is used in the production of bottles and food containers. The polymers usually used in the production of packaging materials for food are listed in Table 2 (McKeen 2013; Pospíšil & Nešpùrek 2007). The main classification of polymer packaging for food recognizes primary, secondary, tertiary, and specific packaging, including active packaging, modified atmosphere packaging and packaging combining different materials (Cvetkovska & Kujundziski 2008). The main functions of the packaging materials are food safety, preservation of quality, facilitation in the distribution and storing of the food. The selection of the primary packaging material depends on the requirements of the food. Secondary packaging material protects the food from physical damages, but also provides the basics information about the product: serial number, date of production and other relevant information. The so-called tertiary packaging is the final packaging that facilitates transportation and protects the packed product from mechanical damages and environmental effects. Edible films and coatings, as specific packaging, differ from the other packaging materials primarily in the method of their application to the food. Namely, they are formed directly on the product, creating an integrated system that does not change the organoleptic characteristics of the food. This type of packaging preserves the food quality, odour and moisture even after removal of the primary packaging material. Films and coatings in this group are formed by chitosan, dextran, xanthan-rubber, potato starch, zein, Na-alginate, carboxymethyl cellulose, etc. Barrier characteristics of the packaging material are significantly improved by the combination of the materials in laminate systems or co-extruded structures, e.g. the combination of cardboard

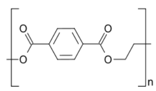
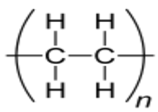
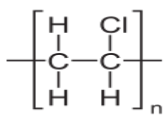
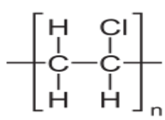
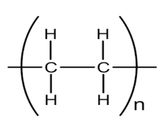
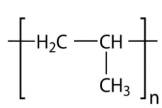
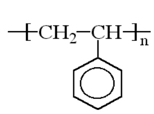
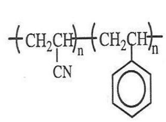
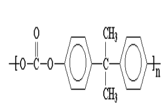
Table 1. Required characteristics of the food packaging materials (Cvetkovska & Kujundziski 2008; Marsh & Bugusu 2007; McKeen 2013).

Characteristic	General requirement	Specific requirement
Food quality	To preserve or improve sensory characteristics.	To preserve taste, odour, colour, content.
Production	To enable simple and economic manufactuing processes of packaging materials. Compatibility with the process of food packing (filling).	Sheets, films, containers, bags; suitable mechanical properties. Dimensional stability, suitability to the filling lines; easy closure; compatibility with the existing equipment.
Logistics	Facilitating distribution.	To fit the requirements of the industry (dimensions, etc.). To have appropriate codes (barcode, producer, etc.).
Marketing	To improve the appearance and attractiveness of the product.	Good graphics; aesthetic appearance; characteristic of specific culture; suitable functionality (purity, facilitated opening, etc.).
Environment	Not to risk human safety. Responsible use of raw materials. To facilitate waste management.	Safe interaction with the foodstuff. To prevent physical damage.Positive Life Cycle Analysis (LCA).To be reused, to be recycled; burned or composed.
Legislation	National regulations.	To be compatible with the hygienic and migration conditions, to have appropriate labelling.
Cost	Economic efficiency	Acceptable price per unique packaging (as the part in the total price); price of the necessary equipment.

and plastic for beverage packaging. Cardboard provides stability and light protection, while plastic acts as a barrier against gases and vapours (Cvetkovska & Kujundziski 2008; Marsh & Bugusu 2007; McKeen 2013).

The consumer requirements for enhanced food quality and increased shelf life impose the demand for the application of new innovative packaging materials with unique properties, such as UV protection, transparency, suitable gas barrier, and, at the same time, environmental compliance. One of the most important technologies for the preservation of food quality, but also an extension of shelf life, is the application of modified atmosphere packaging and vacuum packaging. After the fresh food is packed, the respiration processes continue, i.e. the microorganisms in the food remain breathing, spending oxygen and producing CO_2 and water vapour, which alters the atmosphere in the packaging. Packaging material transmits oxygen, CO_2 and water vapour, changing the atmosphere around the product. Thus, with the application of the modified atmosphere packaging, the gas environment around the food changes in accordance with the initial conditions (Embleni 2013)(Soltani, Alimardani, Mobli, & Mohtasebi 2015). Modern packaging systems are usually designated as active, intelligent or smart packaging (Barska & Wyrwa 2017; Majid, M.Thakur, & Nanda 2018; Müller & Schmid 2019; Wyrwa & Barska 2017). Active packaging not only acts as an inert barrier toward the environment but accomplishes some additional functions. For example, active packaging may contain some substances trapping or linking oxygen or preventing the development of microbes that reduce the food quality. Application of active packaging systems, absorbents or emitters, by the modification of the atmosphere, leads to the improvement of the safety and sensory characteristics of the food. Absorbent systems (scavengers) eliminate undesirable components (oxygen, excess of water, ethylene, CO_2, odours or other specific food components), while the systems containing emitters add some ingredients to the packed food (CO_2, water, antioxidants or stabilisers) (Wyrwa & Barska 2017). Intelligent or smart packaging supplements the active packaging in such way as to indicate the alterations of the food environment during transportation and storage, and provide information about the quality of the product. Two kinds of intelligent packaging systems are known — one is based on the determination of the conditions on the external side of the packaging, and the other on direct monitoring of the parameters related to the food quality. In this regard, different indicators are used – for temperature, time/temperature function, packaging integrity, microbe growth, product authenticity or physical shock (Barska & Wyrwa 2017; Müller & Schmid 2019). The application of modern packaging solutions offers many advantages in the food industry, such as improved management of food waste, an extension of the shelf life, microbiological safety and better organoleptic features of the foodstuff. On the other hand, the use of innovative packaging systems is usually accompanied by some drawbacks related to the pronounced migration of the chemicals from the packaging material to the food (Barska & Wyrwa 2017; Müller & Schmid 2019; Wyrwa & Barska 2017). In that regard, packaging materials, and, in general, all other plastic materials besides polymers, contain additives, usually low-molecular-weight substances that improve chemical and/or mechanical properties of the material. Compatibility between additives and polymers is a particularly important factor in the achievement of optimal effects and homogeneous distribution of plasterers, lubricants and antioxidants within polymers. In addition, the concentration of additives in the formulations and the possibilities of their migration, are also significant features of polymer packaging materials. Generally, the substances that migrate through the packaging material into the food are monomers, oligomers, catalyst residues and solvents. Thus, bisphenol A, (used as a monomer for the synthesis of polycarbonates), epoxy resins used as coatings in metal cans), isopropyl thioxanthone (used as photo-initiator in printing), lead (found in printing dyes), antimony (from the catalyst used in the synthesis of polyethylene terephthalate), dioxyl phthalate (plasticizer for polyvinyl chloride), etc., are some of the detected migrants from the packaging into the food (Pospíšil & Nešpùrek 2007). The factors that influence the level of interaction between food and packaging materials are classified as: factors related to the food type and characteristics, aspects associated to the composition and properties of packaging materials, and issues linked to the conditions of the environment (temperature, storing time, humidity, etc.) (Woods 1991). In the last 20 years, debates and studies about the possible migration of low-molecular-weight substances in the food, and how it affects human health have been in the focus of scientific interest (İçöz & Eker 2016). Most of the migrants are present in very small quantities, mg/kg (ppm), mg/kg (ppb) and ng/kg (ppt), but consumers need a confirmation that there is not any risk to human health, that the food is "clean", i.e it does not contain any contaminants or their concentration is below the acceptable limits. On the other hand, the allowed limits are related to the current capability of the instruments to detect low concentrations of contaminants, whose limit continuously decreases as a result of very fast technical development (İçöz & Eker 2016). Thus, this issue introduces a continuous challenge for the industry, legislature, and, consequently, science. There is a rule that "the toxicity depends on the dosage (quantity) of the substance", but sometimes, extremely low concentrations are risky, too.

Table 2. Polymers for packaging materials (Chamovska & Porjazoska 2018; Cvetkovska & Kujundziski 2008)

Polymer	Repeating unit	Code	Basic characteristics	Application
Polyethylene terephthalate (PET)		01 PET	Good mechanical characteristics, good chemical stability, radiance, semi-permeable for gases, vapors, and water (compared to polycarbonates), bi-axially oriented PET is resistant to high temperatures and impermeable for water, gases and vapors.	Confectionery packaging, water, soda and beverages bottles, cooking oil bottles, and detergents. Biaxially oriented polyester foil is used for baking in an oven.
High density polyethylene (HDPE)		02 PE-HD	Chemically inert, resistant to water, salts, cracks in contact with polar solvents, low permeability for water vapor, permeable for gases (not suitable as barrier for O_2 and CO_2).	Bottles for milk and detergents, plastic sacks and food dishes / containers.
Polyvinyl chloride (PVC) (unplasticized)		03 PVC	Rigid, hard, transparent, difficult to process (HCl separates during heating), friction resistant, resistant to chemicals and atmospheric influence, suitable for combination with fillers and plasterers, good electro-insulator, resistant to moisture, impermeable for gases, poor burning.	Packaging for margarine butter (with white pigment as additive, resistant to UV rays), in construction industry, plastic pipes.
Polyvinyl chloride (PVC) (plasticized)		03 PVC	Flexible, resistant to alcohol, unresisting to atmospheric influences, transparent, suitable for lamination. Containing more than 20% plasticizer shows low permeability to water vapor and gases, and contracts during heating (lower resistivity to heating).	Bags for packaging of meat, vegetables, fruits, vacuum packaging, bottles for water, cooking oil, liquid detergents, pads for biscuits and candies, garden furniture.
Low density polyethylene (LDPE)		04 PE-LD	Chemically inert, swell in many hydrocarbons at room temperature, permeable for gases (it is not a barrier for O_2 and CO_2).	Bags for different applications, dishes for food storage, water pipes.
Polypropylene (PP)		05 PP	Elastic, resistant to fold, low permeability to O_2 and water vapor.	Screw tops, dishes for milk products and other food, drinking straws.
Polystyrene (PS)		06 PS	Very brittle material, resistant to alcohol and acid, reacts with paraffin and carbohydrates, inflexible, good barrier for water vapor.	Different dishes and plastic cutlery.
Styrene-acrylonitrile copolymer (SAN)		07 O	Rigid and transparent, good dielectric properties, resistant to water solutions of bases, acids, salts, hard and resistant to external effects.	Jars for homogeneous food, foil for bakery products.
Polycarbonate (PC)		07 O	Transparent, sustainable at high and low temperatures, good mechanical properties, excellent barrier properties, small gas permeability, vapors and moisture; (it is more expensive than PET).	Carbonated beverages, multi-layer foils produced by co-extrusion or lamination, packaging of chemicals and food.

2. PLASTIC FOOD PACKAGING AND FOOD CONTACT MATERIALS LEGISLATION

Legislation related to the food industry is under continuous revision and differs in the countries of Europe and the USA. Although the European Commission and the Council of the European Union are constantly urging the European countries to harmonize the regulations, variances in the national legislations on the materials and articles in contact with food in different countries, still exist. The general requirements for food packaging materials and articles in contact with food and the authorization of the new substances, in accordance with the European Legislation, are given in the European Regulation (EC) 1935/2004 of 27 October 2004 (European Commission 2004b) and EU Regulation on Good Manufacturing Practices (EC) 2023/2006 of 22 December 2006 (European Commission 2006). Following the requirements of Regulation (EC) No. 1935/2004, plastic food packaging and food contact materials are also subject to review and approval by the European Food Safety Authority (EFSA) before being placed on the market (European Commission 2004b)).

Based on the Regulation (EC) No 1935/2004 (European Commission 2004b), materials in contact with the food products must fulfill the following requirements:

- they must be safe; the low-molecular-weight additives must not migrate from the packaging to the food product and change its sensory characteristics, and, thus, must not compromise human health;
  must enable traceability during production;
- they must be labelled with appropriate symbols for food contact;
  they must contain provisions of the correct use of food contact materials and articles;
  labelling, advertisement and presentation of the packaging must not be confusing for the customers;
  their production must follow good manufacturing practices.

The policy and the general principles of improving the Good Manufacturing Practice (GMP), applied to the materials in contact to the food, are described and structured in the Regulation (EC) No. 2023/2006 of 22 December 2006 (European Commission 2006).

Regulation (EC) No. 1935/2004 encloses restrictions, allows migration of particular substances from packaging into the food in previously determined limits, and it also contributed to the development of new technologies in the food contact materials, so-called "active" and "intelligent" systems (European Directives and Laws). The Regulation (EC) No. 1935/2004 permits the use of the active packaging in EU while the Regulation (EC) No. 450/2009 of 29 May 2009 on active and intelligent materials and articles intended to come into contact with food, lists the "active" and "intelligent" substances that are part of the packaging (Commission Regulation (EC) No 450/2009, 2009). Directive 2002/72/EC of 6 August 2002 covers plastic materials in contact with food, and it substitutes the Directive 90/128/EEC of 23 February 1990 and its amendments. The Directive encompasses materials and articles in contact with foodstuffs, including multi-layered packaging material where all layers are plastic. In addition, this Directive includes the list of approved monomers and additives, used in the production of packaging materials for food, with the overall migration limits (OML) and specific migration limits (SML) settled. Specified overall migration (OML) shall not outgrow 10 mg/dm^2, but this limit extends to 60 mg/kg for: articles with a volume between 500 mL and 10 L, and lids or other covers (European Commission 2002; 2004a; 2005; 2007; 2008; 2009; 2011a).

Related to the Directive 2002/72/EC, amendments 2004/1/EC of 6 January 2004, approving the application of azodicarbonamide as a blowing agent in the production of foamed plastic packaging materials in contact with food; 2004/19/EC of 1 March 2004, encompassing an extended list of allowed monomers; and Regulation 2005/79/EC of 18 November 2005, including the complete list of allowed additives in plastic that will be in contact with food, has been repealed in May 2011. Amendments 2007/19/EC of 30 March 2007, restricting the migration of plasticizers in fatty food in quantities that can affect the foodstuff and/or human health; 2008/39/EC of 6 March 2008 and (EC) 975/2009 of 19 October 2009, both dealing with the extended list of permitted additives, and 2011/8/EU of 28 January 2011 (restricting Bisphenol A in plastic infant feeding bottles), have been still in force, and also account to the Directive 2002/72/EC (European Commission 2002; 2004a; 2005; 2007; 2008; 2009; 2011a).

Commission Regulation (EU) No 10/2011 of 14 January 2011 is the most comprehensive EU measure on plastic materials and articles, and covers the rules on the composition of food contact plastic materials and sets up the list of substances allowed to be used in the fabrication of plastic materials intended to be in contact with food. This regulation consolidates the following Directives: the Directive 2002/72/EC of 6 August 2002 and its amendments, Directives 78/142/EEC of 30 January 1978, 80/766/EEC of 8 July 1980, and 81/432/EEC of 29 April 1981, dealing with materials and articles containing vinyl chloride monomer, that is planned to come into contact with food, the controlling of the level of vinyl chloride monomer in materials and articles intended to come into contact with food, as well as the method of analysis for the control of a release of vinyl chloride into the food, and Directives 82/711/EEC of 18 October 1982 with its two amendments 93/8/EEC of 15 March 1993 and 97/48/EEC of 29 July 1997, and 85/572/EEC 19 December 1985, related to the migration testing of the components of plastic and list of simulants to be used in migration testing.

Regulations 2016/1416 of 24 August 2016 and 2017/752 of 28 April 2017 amend and correct Regulation 10/2011, while the amendments that only modify the list of authorized substances, included in Annex I of this Regulation are: Regulation (EU) 321/2011 of 1 April 2011, restricting the use of Bisphenol A in plastic infant feeding bottles, Regulations (EU) 1282/2011 of 14 January, (EU) 1183/2012 of 30 November 2012, (EU) 1183/2012 of 30 November 2012 and its corrigendum, (EU) 202/2014 of 3 March 2014 , (EU) 2015/174 of 5 February 2015, (EU) 2018/79 of 18 January 2018, and (EU) 2018/213 of 12 February 2018 on the use of Bisphenol A in varnishes and coatings used in plastic materials that are in contact with foodstuff (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b). EU Regulation 10/2011 covers polymer mono-layers, multi-layers made of polymers, joined by adhesives, plastic layers or coatings used for gaskets in caps and covers, polymer layers in multi-material multi-layer materials, coated and printed plastic materials that come in contact with food. Coatings, adhesives, and printing inks are covered by national legislation until a specific community measure is adopted. Ion exchange resins, rubber and silicones are excluded from the range of this Regulation (Commission Regulations: (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b). Using migration limits is an important tool for ensuring the safety of plastic packaging material. The verification of the compliance with the migration limits, described in the testing scheme is given in Regulation (EC) No. 1935/2004. The list of stimulants, as well as the types of food they substitute, is presented in Table 3, while the conditions of migration testing are given in Table 4, for the determination of specific migration, as well Table 5, for overall migration testing. Overall migration limits (OML) determine the maximum allowed quantity of non-volatile substances that are eligible to migrate into food and /or food simulant. In accordance with a good manufacturing practice, the migration of any substance from packaging to the food should not exceed 10 mg/dm2. Specific migration limits (SML) denote the maximum allowed quantity of a specific substance in food and/or food simulant (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b).

Regarding plastic multi-layered materials and articles, the composition of each of the layers needs to fulfill the requirements given in Regulation 10/2011. Substances denoted as carcinogenic, mutagenic, or toxic to reproduction, and those categorized as non-authorized substances, with a maximum level of migration higher than 0.01 mg/kg, shall not be applied. Plastic multi-layered materials and articles need to comply with the migration limits (OML and SML) Table 5. In addition, a declaration of compliance is required (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b).

Table 3. List of simulants recommended by the Regulation 10/2011 Commission Regulations: (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b).

Food simulant	Abbreviation	Type of the food
Ethanol 10% (v/v)	Simulant A	Aqueous food
Acetic acid 3% (w/v)	Simulant B	Acetic food (pH 4.5)
Ethanol 20% (v/v)	Simulant C	Alcoholic food
Ethanol 50% (v/v)	Simulant D1	Sami-fatty food
Any vegetable oil containing less than 1% unsaponifiable matter	Simulant D2	Fatty food
Poly (2,6-diphenyl-p-phenylene oxide), particle size 60-80 mesh, pore size 200 nm	Simulant E	Dry food

Correction of specific migration in food with more than 20% fat shall be corrected using Fat Reduction Factor (FRF) calculated by the formula:

$$FRF = \frac{\left(\frac{g \text{ fat in food}}{kg \text{ of food}}\right)}{200} = \frac{(\% \text{ fat} \times 5)}{100} \tag{1}$$

In the case of the multi-component-multi-layer materials and articles, a declaration of compliance needs to be issued only for polymer layers. Furthermore, the composition of each plastic layer needs to comply with Regulation 10/2011. Components forbidden to be used in the multi-component-multi-layer materials aIn addition, and articles are all mutagenic, carcinogenic and toxic substances with the maximum level of migration of 0.01 mg/kg. National laws in different countries specify the overall and specific migration limits in multi-component-multi-layer materials or articles (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b).

Table 4. Conditions of testing of specific migration (Commission Regulations: (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b).

Contact time	Testing time	Contact temperature	Testing temperature
$t \leq 5min$	5 min	$T \leq 5C$	5 °C
5 min $< t \leq$ 0.5 hours	0.5 hours	5 °C $< T \leq$ 20 °C	20 °C
0.5 hours $< t \leq$ 1 hours	1 hour	20 °C $< T \leq$ 40 °C	40 °C
1 hour $< t \leq$ 2 hours	2 hours	40 °C $< T \leq$ 70 °C	70 °C
2 hour $< t \leq$ 6 hours	6 hours	70 °C $< T \leq$ 100 °C	100 °C or reflux temperature
6 hour $< t \leq$ 24 hours	24 hours	100 °C $< T \leq$ 121 °C	121 °C (*)
1 day $< t \leq$ 3 days	3 days	121 °C $< T \leq$ 130 °C	130 °C (*)
3 days $< t \leq$ 30 days	10 days	130 °C $< T \leq$ 150 °C	150 °C (*)
Above 30 days	Specific conditions(*)	150 °C $< T \leq$ 175 °C	175 °C (*)
*1.Testing of max 10 days at 60 °C shall cover contact times above 30 days at room temperature and below. 2. Testing for 10 days at 20 °C covers frozen food, at 40 °C, frozen and refrigerated food for all storage times (all three cases), at 50 °C - storage at room temperature for up to 6 months, at 60 °C - storage at room temperature and below for storage above 6 months.		175 <°C $T \leq$ 200 °C $T \geq$ 200 °C	200 °C (*). 225 °C (*)

*Used just for simulants D2 and E.

3. MATHEMATICAL MODELING OF MIGRATION OF ADDITIVES FROM THE PACKAGING MATERIAL TO THE FOOD

Mathematical modeling has been considered as a suitable tool in the assessment of migration of substances from the packaging material to the food. There are many software packages that can be used for the calculation of migration, using Fick's law as a basic model in the development of the software. Generally, all mathematical expressions have been based on the following assumptions (Begley 2000; Franz 2000; O'Brien, Goodson, & Cooper 1999):

- a) In the worst-case scenario, there is the migration of the total quantity of additives and/or other possible migrants into the food or food simulant.
  b) The material is a mono-layer.
  c) The material is homogeneous:
  - Material P with a constant thickness (dp) is in contact with simulant F, with volume VF;
  - Homogeneous distribution of a migrant in P and F;
  - There is no boundary resistance of the transfer between the polymer and food;
  - Negligible interaction between polymer and food simulant;
  - Absence of absorption of food simulant in the polymer;
  - Overall migration of substances in the polymer and food simulant is constant.

One of the most used commercial software packages, Migrates Lite, uses a model developed by Pringer (Begley 2000; Franz 2000; O'Brien et al. 1999), offering the analytical solution of Fick's second law:

$$\frac{m_{F,t}}{A} = c_{p,0}\rho_p d_p\left(\frac{\alpha}{1+\alpha}\right)|1-\sum_{n=1} \frac{2\alpha(1+\alpha)}{1+\alpha+\alpha^2 q_n^2}\exp\left(-D_p t\frac{q_n^2}{d_p^2}\right)|$$

$$\alpha = \frac{1}{K_{p,F}}\frac{V_F}{V_p} \qquad (2)$$

emph where, m_{Ft} (mg) is a mass of the migrant from a polymer to food during the time t, A is an area of the interface between the polymer and food simulant (cm^2), C_{PO} – initial concentration of the migrant to the polymer (mg/kg), $_p$ and $_F$ are densities of the polymer and simulant (g/cm^3), D_P – diffusion coefficient in the polymer (cm^2/s),t – contact time (s), d_P is polymer thickness (cm), V_P and V_F are polymer volumes and simulant, respectively (cm^3), $C_{P\infty}$, $C_{F\infty}$, - equilibrium concentration of migrant in the polymer and simulant (mg/kg), $K_{P,F}$ – partition coefficient of the migrant between polymer P and food, q_n

Table 5. Conditions of testing of overall migration (Commission Regulations (European Commission 2011b; 2011c; 2011d; 2012; 2014; 2015; 2016; 2017; 2018a; 2018b).

Contact time and temperature of testing	Conditions of real usage 10 days at 20 °C
10 days at 20 □C	$T \leq -18$ °C $T \leq 4$ °C
10 days at 40 °C	Room temperature or below. Heating up to 70 °C ≤ T ≤ 100 °C, max time $t = 120/2^{(T-70)/10)}$ min.
2 h at 70 °C	Heating up to 70 °C ≤ T ≤ 100 °C, max time of $t = 120/2^{(T-70)/10)}$ min, not followed by usage at room temperature.
1h at 100 °C	Usage at hight temperatures up to 100 °C.
2h at 100 °C or at reflux (or 1 h at 121 °C)	Usage at hight temperatures up to 121 °C.
4h at 100 °C or at reflux	T >40 °C for food that can be simulated with simulants A, B, C and D1.
2h at 175 °C	Fatty food at high temperature.

– positive roots of trigonometric identity tan $q_n = -\alpha q_n$. In the worst-case scenario, it can be assumed that all substances will migrate from the material, and their maximal initial concentration can be computed as:

$$MIC = \frac{SML}{10}\frac{V_F\rho_F}{A}\{\rho_p d_P(\frac{\alpha}{1+\alpha})[1-\sum_{n=1}^{\infty}\frac{2\alpha(1+\alpha)}{1+\alpha+\alpha^2 q_n^2} \exp(-D_p t\frac{q_n^2}{d_p^2})]\}^{-1} \quad (3)$$

MIC – maximal initial concentration (mg/kg) is known, and specific migration limit (mg/kg) is calculated. Determination of accurate values of D_P and $K_{P,F}$ is difficult. Constant values of $K_{P,F}$; A'_p and τ for different polymer groups, have been adopted after the analysis and systematization of a large number of experimental results. The diffusion coefficient is determined using the equation:

$$D_p^* = 10^4 \exp(A_p - 0,1351 M_r^{2/3} + 0,003 M_r - \frac{10454}{T}) \quad (4)$$

Where, D_P^* is the maximal value of the diffusion coefficient (cm^2/s), M_r is relative molecular mass of the migrant, T – temperature (K)

$$A_p = A'_p - \frac{\tau}{T} \quad (5)$$

A_P is polymer specific constant, τ - material specific temperature constant, i.e., parameter of activation energy of polymer diffusion, A_P parameter that characterizes diffusion conductivity of polymer material. During the process of validation of the model, it was noted that validation is completed for polyolefines, it is satisfying for polystyrene, high impact polystyrene, polyethylene naphthalate and polyamide, and it is not acceptable for polycarbonate and polyvinyl chloride. For multilayer materials, overall migration is determined as a sum of the quantities of the individual quantities that migrate from each layer (Begley 2000; Franz 2000; O'Brien et al. 1999).

4. CONCLUSIONS

Chemical and physical properties, such as gas permeability, transparency, lightweight, and recyclability, are some of the crucial features of polymer materials being suitable for food packaging systems. However, the presence of substances not expected a priori in packed food or bottled beverages (monomers, catalysts, additives, degradation products, impurities, etc.) has been detected. Interaction between polymer packaging and food occurs by the mechanisms of permeation, sorption and migration of the components from foodstuffs to the packaging and vice versa. Competition between producers of packaging materials and the emerging demands of the consumers for the increased shelf-life of the food products result in innovative packaging solutions that actively interact with the food. The active and intelligent packaging materials additionally increases the drawbacks of migration of low-molecular-weight components from the material to the foodstuff. The European Parliament and the European Commission enacted rigorous standards, regulations and provisions that control the polymer packaging materials intended for direct contact with food, determining the rules for the testing of migration and its compliance with regulated migration limits. On the other hand, acceptable migration limits have been related to the restriction of the smallest detectable chemical concentration. Therefore, this study poses a continuous challenge for industries, legal entities, and science, too.

REFERENCES

Barska, A., & Wyrwa, J. (2017). Innovations in the food packaging market – intelligent packaging – a review. *Czech Journal of Food Sciences*, *35*(1), 1–6.

Begley, T. H. (2000). Migration from food packaging: Regulatory considerations for estimating exposure. In O. G. Piringer & A. L. Baner (Eds.), *Plastic packaging materials for food: Barrier function, mass transport, quality assurance, and legislation* (p. 359–392). Wiley-VCH Verlag GmbH. https://doi.org/10.1002/9783527613281.ch11

Chamovska, D., & Porjazoska, A. (2018). Legislation on food contact packaging materials. In *Proceedings xx yucorr* (p. 108-115).

Cvetkovska, M., & Kujundziski, A. P. (2008). *Polymers and polymer materials as packaging – food packaging (in macedonian)*. Ss. Cyril and Methodius" University, Faculty of Technology and Metallurgy.

Embleni, A. (2013). Modified atmosphere packaging and other active packaging systems for food, beverages and other fast-moving consumer goods. In F. N. (Ed.), *Trends in packaging of food, beverages and other fast-moving consumer goods (fmcg), markets, materials and technologies* (p. 22–34). Woodhead Publishing Limited. http://doi.org/10.1533/9780857098979.22

European Commission, R. (2002). Commission directive 2002/72/ec, relating to plastic materials and articles intended to come into contact with foodstuffs. *Off J Eur Union*.

European Commission, R. (2004a). Commission directive 2004/19/ec of 1 march 2004 amending directive 2002/72/ec relating to plastic materials and articles intended to come into contact with foodstuffs. *Off J Eur Union*, *71*, 8–21.

European Commission, R. (2004b). No. 1935/2004 of the european parliament and of the council of 27 october 2004 on materials and articles intended to come into contact with food and repealing directives 80/590/eec and 89/109/eec. *Off J Eur Union*, *338*(4).

European Commission, R. (2005). Commission directive 2005/79/ec of 18 november 2005 amending directive 2002/72/ec relating to plastic materials and articles intended to come into contact with food.

European Commission, R. (2006). Commission regulation (ec) no 2023/2006 of 22 december 2006 on good manufacturing practice for materials and articles intended to come into contact with food. *Off J Eur Union*, *50*, 75–78.

European Commission, R. (2007). Commission directive 2007/19/ec of 2 april 2007 amending directive 2002/72/ec relating to plastic materials and articles intended to come into contact with food and council directive 85/572/eec laying down the list of simulants to be used for testing migration of constituents of plastic materials and articles intended to come into contact with foodstuffs. *Off J Eur Union (corrigendum)*, *97*, 50.

European Commission, R. (2008). Commission directive 2008/39/ec of 6 march 2008 amending directive 2002/72/ec relating to plastic materials and articles intended to come into contact with food. *Off J Eur Union.*, *63*, 6–10.

European Commission, R. (2009). Commission regulation (ec) no 975/2009 of 19 october 2009 amending directive 2002/72/ec relating to plastic materials and articles intended to come into contact with foodstuffs. *Off J Eur Union*(274), 3.

European Commission, R. (2011a). Commission directive 2011/8/eu of 28 january 2011 amending directive 2002/72/ec as regards the restriction of use of bisphenol a in plastic infant feeding bottles. *Off J Eur Union*.

European Commission, R. (2011b). Commission implementing regulation (eu) no. 321/2011 of 1 april 2011 amending regulation (eu) no. 10/2011 as regards the restriction of use of bisphenol a in plastic infant feeding bottles. *Off J Eur Union*, L87–1.

European Commission, R. (2011c). Commission regulation (eu) no 10/2011 of 14 january 2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union.*, *12*, 1–89.

European Commission, R. (2011d). Commission regulation (eu) no 1282/2011 of 28 november 2011 amending and correcting commission regulation (eu) no 10/2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union L*, *328*, 22–29.

European Commission, R. (2012). Directive (ec) no. 1183/2012/eu of 30 november 2012 amending and correcting regulation (eu) no 10/2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union*, *338*, 11.

European Commission, R. (2014). Regulation (ec) no. 202/2014 of 3 march 2014 amending european commission regulation (eu) no. 10/2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union*, *62*, 13.

European Commission, R. (2015). Regulation (ec) no. 2015/174 of 5 february 2015 amending and correcting regulation (eu) no 10/2011 on plastic materials and articles intended to come into contact with food text with eea relevance. *Off J Eur Union*, *30*, 2–9.

European Commission, R. (2016). Commission regulation (eu) 2016/1416 of 24 august 2016 amending and correcting regulation (eu) no 10/2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union*, *230*, 22–42.

European Commission, R. (2017). Commission regulation (eu) 2017/752 of 28 april 2017 amending and correcting regulation (eu) no 10/2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union*, *113*.

European Commission, R. (2018a). Regulation (ec) 2018/213 of 12 february 2018 on the use of bisphenol a in varnishes and coatings intended to come into contact with food and amending regulation (eu) no 10/2011 as regards the use of that substance in plastic food contact materials. *Off J Eur Union*, *41*, 6–12.

European Commission, R. (2018b). Regulation (ec) 2018/79 of 18 january 2018 amending regulation (eu) no 10/2011 on plastic materials and articles intended to come into contact with food. *Off J Eur Union*, *14*, 31–34.

Franz, R. (2000). *Migration of plastic constituents. in plastic packaging materials for food: Barrier function, mass transport, quality assurance, and legislation*. Wiley-VCH Verlag GmbH. http://doi.org/10.1002/9783527613281.ch10

İçöz, A., & Eker, B. (2016). Selection of food packaging material, migration and its effects on food quality. In *1st international conference on quality of life, center for quality, faculty of engineering* (p. 201–210). University of Kragujevac, Serbia.

Majid, I., M.Thakur, & Nanda, V. (2018). Innovative and safe packaging technologies for food and beverages: Updated review. In P. S. K. & S. H. P. K. (Eds.), *Innovations in technologies for fermented food and beverage industries* (p. 257–287). Springer International Publishing.

Marsh, K., & Bugusu, B. (2007). Food packaging—roles, materials, and environmental issues. *Journal of Food Science*, *72*(3), 39– 55. https://doi.org/10.1111/j.1750-3841.2007.00301.x

McKeen, L. W. (2013). Introduction to use of plastics in food packaging. In E. S. (Ed.), *Plastic films in food packaging, materials, technology and applications* (pp. 1–15). Elsevier. https://doi.org/10.1016/B978-1-4557-3112-1.00001-6

Müller, P., & Schmid, M. (2019). Intelligent packaging in the food sector: A brief overview. *Foods*, *8*(16), 1–12.

O'Brien, A., Goodson, A., & Cooper, I. (1999). Polymer additive migration to foods–a direct comparison of experimental data and values calculated from migration models for high density polyethylene (hdpe). *Food Additives & Contaminants*, *16*(9), 367–380. https://doi.org/10.1080/026520399283858

Pospíšil, J., & Nešpùrek, S. (2007). Additives for plastics and their transformation products. In O. G. Piringer & A. L. Baner (Eds.), *Plastic packaging materials for food: Barrier function, mass transport, quality assurance, and legislation* (p. 47–77). Wiley-VCH Verlag GmbH.

Soltani, M., Alimardani, R., Mobli, H., & Mohtasebi, S. (2015). Modified atmosphere packaging: A progressive technology for shelf-life extension of fruits and vegetables. *Journal of Applied Packaging Research*, *7*(3), 33–59.

Woods, K. D. (1991). *Food—package interaction safety, european views*. American Chemical Society.

Wyrwa, J., & Barska, A. (2017). Innovations in the food packaging market: Active packaging. *European Food Research and Technology*, *243*(10), 1681–1692. https://doi.org/10.1007/s00217-017-2878-2