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Modelling migration from plastic packaging materials used in food industry

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Abstract

This study is focused on the modeling migration from packaging materials used in food industry from a polyethylene film to a food simulant. We tried to develop a prediction statistical mathematical model, by performing an evaluation of influence of the effect of three parameters through an analysis of multiple correlations between overall migration concentration as the dependent variable, and the temperature and time as independent variables. This method could reduce the time and costs of the experimental work associated to the analysis by means of a tool with a wide prediction capacity.

Keywords: overall migration, plastic, food packaging, mathematical modeling

1. Introduction

The migration process has a particular interest in food packaging, because can produce toxic effects on the human organism [1].

Packaging has become an integral part of the processing, preservation, distribution, marketing, and even the cooking of foods. Packaging materials provide protection for microbiological, chemical, and physical contamination. However, components of the packaging material must be safe to the product as well as to the consumers. Food may interact with the packaging materials and this may change the initial mechanical and barrier properties as well as the safety of the product. Migration is defined as the transfer of substances from the package to foodstuffs. The migration of potentially toxic substances from packaging material to foods is a major concern in the selection and use of materials for food packaging because of the possible effect upon human health [2].

A plastics food contact material such as food packaging, kitchen utensils and food processing equipment needs to be compliant for the overall migration level. The laboratory testing capabilities should provide knowledge, expertise for assessing compliance of plastics food contact materials to the relevant EU regulation (EU) 10/2011).

The migration experiments are time-consuming, expensive and often complicated to carry out [3], so the use of mathematical models to predict migration has increased in recent years.

2. Materials and Methods

To determine the overall migration commercial plastic samples made from low density polyethylene and as food stimulants (reagents used in the study Ethanol 95 % (v/v) in aqueous solution [4] for a contact time of 10 days at different contact temperatures (20^0C, 40^0C, and 60 ^{0}C).

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Test conditions for packaging material/food simulant contact and method of overall migration analysis were according to the standard, "EN 1186-9:2002 materials and articles in contact with foodstuffs, test methods for overall migration into aqueous food simulants by article filling". Twenty different plastics cups of different capacities were used. Procedure for the determination of the overall migration into aqueous based food simulants from plastics cup intended to come into contact with foodstuffs, were by filling (one side contact) the plastic cups with corresponding aqueous simulant to within 0,5 cm of the top of the cup. The cups were covered by glass so as to avoid evaporation of simulant during contact period to prevent evaporation and kept in a thermostatically controlled oven at 20^0C, 40^0C, and 60 ^{0}C for ten days. After exposure to the simulant the plastic cups were removed from the oven, emptied and the simulant was placed in a 250-ml preweighed Erlenmeyer flask and evaporated to dryness by means of a hot plate. The Erlenmeyer flask containing the residue of evaporation was kept in a thermostatically controlled oven at 105±1.0 °C for 1 h followed by 1 h in a desiccator and then weighed. An analytical balance capable of weighing to 0.1 mg was used. The mass of the non-volatile residue was determined and expressed as milligrams per square decimetre of surface area exposed to the simulant. Overall migration was calculated as the mean of three determinations on separate test specimens.

The overall migration was expressed as milligrams of residue per square decimetre of the surface of the sample which were intended to come into contact with foodstuffs, calculated for each test specimen using the following formula [4]:

$$M = \frac{m_a - m_b \times 1000}{S} \qquad (1)$$

where

M is the overall migration into the stimulant, in milligrams per square decimetre of surface area of sample;

m_a is the mass of the residue from the test specimen after evaporation of the simulant which had filled the test specimen, in grams;

m_b is the mass of residue from the blank simulant equal to the volume which had filled the test specimen, in grams;

S is the surface area of the test specimen which was in contact with the simulant during the exposure, in square decimetres.

In this study we tried to develop a prediction statistical mathematical model, by performing an evaluation of influence of the effect of three parameters through an analysis of multiple correlations between overall migration concentration as the dependent variable, and the temperature and time as independent variables.

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As a case study for elaborating these predictions statistical mathematical modeling, we choose the overall migration determination by total imersion method (ten samples from the same producer, made from LDPE), analysed at diffrent contact temperatures 20ºC, 40ºC and 60ºC , for ten days, in ethanol 95%

For accurate description of the content of overall migration by total imersion method , analysed at diffrent contact temperatures 20ºC, 40ºC şi 60ºC , for ten days, in ethanol 95%, we wanted to obtain a mathematical expression, a polynomial second order equation.

The general form of mathematical models describing the second order polynomial correlation is expressed by the following equation:

$$y = a_0 + a_1 \cdot x_1 + a_2 \cdot x_2 + a_3 \cdot x_1 \cdot x_2 + a_4 \cdot x_1^2 + a_5 \cdot x_2^2 \qquad (2)$$

where

y – content of overall migration in water food simulant(mg/dm^2);

x_1 – content of overall migration in 3% aqueous acetic acid food simulant (mg/dm^2);

x_2 – content of overall migration in 10% aqueous ethanol (mg/dm^2).

By using MATLAB programming language it was solved the system of equations and all experimental data were processed and analyzed.

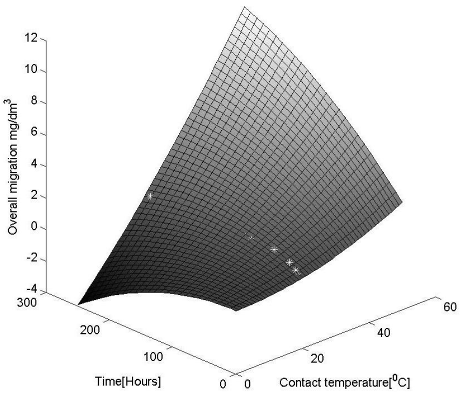

Figure 1. Content of overall migration in ethanol 95% variation depending on the temperature 20 °C , ten days contact time

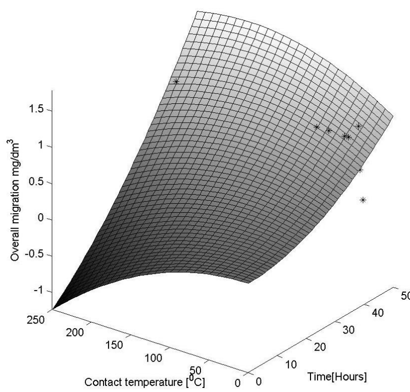

Figure 2. Content of overall migration in ethanol 95% variation depending on the temperature 40 °C , ten days contact time

The validation of the obtained mathematical models has been done by using the following performance criteria: dispersion (σ^2), standard deviation (σ), model accuracy indicator (R^2) and correlation coefficient (R) [5-7].

Experimental data together with mathematical surfaces that were generated by the statistical models for the correlations between content of overall migration concentration as the dependent variable, and the temperature and time as independent variables, are presented in Figures 1-3.

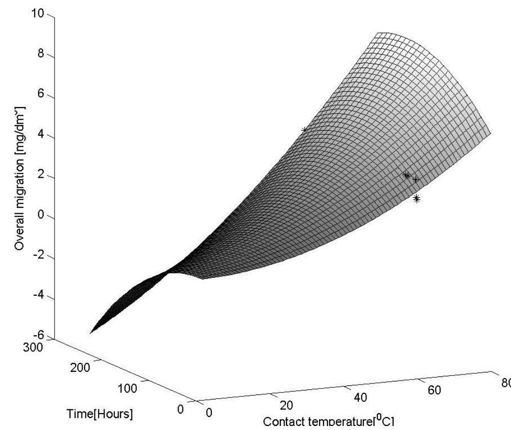

Figure 3. Content of overall migration in ethanol 95% variation depending on the temperature 60 °C , ten days contact time

In table 1 are presented the coefficients of equations (2) corresponding for each studied case and in table 2 are presented the accuracy indicators of the mathematical models (the mean values, dispersion, standard deviation, R^2 – model accuracy indicator, R – correlation coefficient).

Table1. Equation's (2) coefficients

Case	**Equation's coefficients**					
	a_0	a_1	a_2	a_3	a_4	a_5
Overall migration in ethanol 95% variation depending on the temperature 20 °C , ten days contact time	0	0	0	$9.6x10^{-4}$	$1.1x10^{-4}$	$-6.2x10^{-5}$
Overall migration in ethanol 95% variation depending on the temperature 40 °C , ten days	0	0	0	$1.4x10^{-4}$	$4.5x10^{-4}$	$-1.9x10^{-5}$
Overall migration in ethanol 95% variation depending on the temperature 60 °C , ten days	0	0	0	$4.1x10^{-4}$	$9.0x10^{-4}$	$-9.1x10^{-5}$

Table2. Accuracy indicators

Case	Mean value, v_m	Dispersion, σ^2	Standard Deviation, σ	Accuracy indicators, R^2	Accuracy indicators, R
Overall migration in ethanol 95% variation depending on the temperature 20 °C , ten days contact time	1.01	0.004	0.21	0.84	0.92
Overall migration in ethanol 95% variation depending on the temperature 40 °C , ten days	0.9	0.6	0.25	0.23	0.48
Overall migration in ethanol 95% variation depending on the temperature 60 °C , ten days	3.62	0.11	0.33	0.50	0.71

Analyzing the correlation coefficients, the highest predictive value for the evolution of overall migration in ethanol 95% depending on the temperature and contact time, was found for the dependency between the content of overall migration in ethanol 95% , temperature 20 °C , and ten days contact time with a value of R^2= 0.84. After calculation of the correlation coefficients, it was done a graphical representation, figure 4, with the comparison between the values calculated according to the model and the measured values (overall migration in ethanol 95% variation depending on the temperature 20 °C , and ten days contact time).

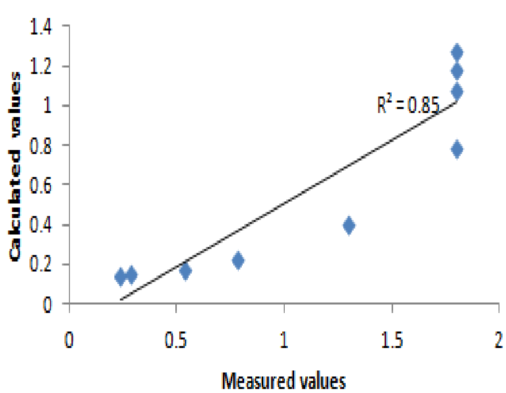

Figure 4. Comparison between the calculated and the measured values of overall migration in 95% ethanol, temperature 20ºC, ten days contact time

Analyzing figure 4 we may conclude that the mathematical model corresponding for the dependency of content of overall migration in ethanol 95% variation depending on the temperature 20 °C , ten days contact time, having an accuracy indicator R^2= 0.85 describes well enough the variation between the concentration of overall migration, temperature and contact time.

Application of the F-test to overall migration data from the measured values and the calculated ones (mg/dm^2, figure 4) showed also that for the analyzed samples the migration differences are no statistically significant , F < F crit (2.08 < 3.44), with a P-value > 0.05 (P-value = 0.15)

4. Conclusion

The results show that for the correlation coefficient of the mathematical models corresponding to variation of concentration of overall migration values with temperature and contact time, the highest predictive value , was found for the dependency between the content of overall migration in ethanol 95% variation depending on the temperature at 20 °C, and ten days contact time with a value of R^2= 0.84. . From the graphical representation with the comparison between the values calculated according to the model and the measured for the content of overall migration in 95% ethanol, ten days contact at 20 °C, we conclude that the mathematical model do describes well enough the variation between the variation of overall migration in 95% ethanol, with temperature at 20ºC, and ten days contact time.

Compliance with Ethics Requirements

Authors declare that they respect the journal's ethics requirements. Authors declare that they have no conflict of interest and all procedures involving human and/or animal subjects (if exists) respect the specific regulations and standards.

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