Numerical Simulation on Migration of Chemical Substances from HDPE into Foods

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Abstract: Numerical simulation of migration of chemical substances from HDPE into foods (simulants) is performed based on the finite element method (FEM). The results show that concentration of migrants in foods at equilibrium increases with the increase of its initial concentration in packaging. Diffusivity of migrants is key to migration dynamics. Partition coefficient represents ratio of concentration in HDPE to that in foods at equilibrium. For multilayer packaging, functional barrier layer is able to efficiently delay migration. Thickness of barrier layers prominently influences barrier effect. The method can be utilized to predict migration of chemical substances having mixed boundary conditions, developed as a total solution food package for migration problems.

Keywords: migration of chemical substances; the finite element method; plastic packaging; food packaging safety

1. Introduction

Migration of chemical substances, plastic additives, monomers, oligomers and their degradation products, etc., from plastic packaging materials into foods may lead to food safety problems for consumers. In China, migration problems on plastic food packaging materials, however, are known and concerned for only a few years. Wang et al. (2004) [1] firstly presented migration tests technique, migrants analysis technology and migration models. Liu et al. (2006) [2] studied migration of three antioxidants BHT, Irganox 1076 and Irgafos 168 into 100% ethanol and comparison between experimental and numerical simulation results are discussed.

Low-molecular-weight antioxidants, BHT, Irganox 1076 and Irgafos 168, are widely used to prevent oxidative reactions during thermal processing in polyolefins such as PE and PP coming into contact with foods which are usually chosen as model migrants for migration studies. Goydan et al. (1990) [3] studied migration of radio-labelled Irganox 1010 and Irganox 1076 from LDPE, HDPE and PP using a novel test cell at temperature up to 135°C. The result shows that diffusion coefficients of these two antioxidants were found to follow an Arrhenius equation when correlated with absolute temperature. Linssen et al. (1998) [4] studied migration of Irganox 1076 and Irgafos 168 from LLDPE into several ethanolic simulants and found that the higher the ethanol content the higher the migration values. Simoneau et al. (1999) [5] studied the stability of three antioxidants including Irganox 1076 which is quite stable under all testing conditions. Han et al. (2003) [6] developed a simulation model computer program, accounting for not only the diffusion process inside the polymer but also partitioning of the contaminant between the polymer and the contacting phase, to quantify migration through multilayer structures. Experimental data of antioxidant BHT was successfully used to demonstrate the accuracy of the model in predicting migration by comparing with simulated results. Dopico-Garcia et al. (2006) studied the stability of some phenolic antioxidants and one oxidized phosphite antioxidant in four food simulants under different temperatures during some 20 days with samples analyzed by RP-HPLC with UV diode-array detector. The results show BHT, Irganox 1076 and oxidized Irgafos 168 are quite stable in olive oil in 5 days under all testing conditions. BHT into ethanol are studied in this paper.

2. THEORETICALA

In the last ten years, researchers turned to numerical methods, such as Finite Element Methods and Finite Difference Methods etc., which are successfully applied in simulation of migration.

2.1 Assumptions

We considered plastic packaging films of single-layer or multilayer coming into contact with foods and the following assumptions are made to define the problem:

(1) chemical substances are initially homogeneously distributed in one(single-layer or multilayer) or more layers(multilayer) of them;
(2) substances migrate from one side of the packaging into foods: they diffuse in packaging, transfer to packaging-food interface, convect into and diffuse in foods; while for the other side contacting air there is no transfer;
(3) for multilayer, all layers are of the same polymer and thus the same diffusivity $D_P$ ( cm$^2$·s$^{-1}$);
(4) for multilayer, perfect contact is obtained between every two layers with no transfer resistance;
consider a finite convective coefficient $h_m$ (cm$^2$s$^{-1}$) at the packaging-food interface and chemical substances are uniform in foods at any time;
(6) no edge effect is considered and no food transfer into packaging.

2.2 Mathematical Model

The governing differential equation for one-dimensional unsteady-state diffusion through the polymer is expressed by Fick’s 2nd law as follows:

$$\frac{\partial C_{x,t}}{\partial t} = D \frac{\partial^2 C_{x,t}}{\partial x^2}$$

(1)

$D$: Diffusion coefficient
$C_{x,t}$: Concentration of the contaminant within the polymer at position $x$ and time $t$
$t$: Time

The initial and boundary conditions

$$t = 0, \quad 0 < x < R, \quad C = C_i$$

$$R < x < L, \quad C = 0$$

$$t \geq 0, \quad x = 0, \quad \frac{\partial C}{\partial x} = 0$$

$$x = R, \quad C_{RP} = C_{FB}$$

2.3 Construction of Finite Element Method

This equation has the following general form used in applying the finite element method where $Q = 0$ and $\lambda = 1$

$$D \frac{\partial^2 C_{x,t}}{\partial x^2} + Q - \lambda \frac{\partial C_{x,t}}{\partial t} = 0$$

(2)

examination function:

$$C(x,t) = \sum_{j=1}^{m} C_j(t) \phi_j(x)$$

(3)

The residual equation is

$$\{R^{(e)}\} = -\int [W]\left(D \frac{\partial^2 C_{x,t}}{\partial x^2} + Q - \lambda \frac{\partial C_{x,t}}{\partial t}\right) dx = 0$$

(4)

$$\{R^{(e)}\} = -\int \phi \left(D \frac{\partial^2 C_{x,t}}{\partial x^2} + Q - \lambda \frac{\partial C_{x,t}}{\partial t}\right) dx$$

Equation (8) can be written as

$$\{R^{(e)}\} = [K^{(e)}][C^{(e)}] - \{f^{(e)}\} + [\rho^{(e)}][C^{(e)}] = \{0\}$$

(9)

3 RESULTS

3.1 Single Layer

A model of BHT migration through the core single layer to 100% ethanol was designed as shown in Figure 1 for the FEM analysis. Figure 5.19 compares the output from the computer program with the actual experimental results (dots on the graph) for the test temperatures, which shows good agreement with actual experimental results.

Figure 1 BHT migration through the core single layer to 100% ethanol at 20, 30, and 40°C

3.2 Structure with Functional Barrier Layer

Figure 2 shows BHT migration to 100% ethanol at 20, 30, and 40°C through the core and outer layers when
the effective thickness ratio is 1:1. Figures 3 to 4 for BHT migration through the core and outer layers with varying thickness to 100% ethanol at 20 and 30°C.

4. Conclusion

The concentration of migrants in foods at equilibrium increases with the increase of its initial concentration in packaging. Diffusivity of migrants is key to migration dynamics. Partition coefficient represents ration of concentration in HDPE to that in foods at equilibrium. For multilayer packaging, functional barrier layer is able to efficiently delay migration. Thickness of barrier layers prominently influences barrier effect. The efficiency of a “functional barrier” may be increased dramatically by applying better barrier material, including OPP, PET and multi-resin systems as functional barriers, since the diffusion coefficient is more influential for the migration process than thickness. The method can be utilized to predict migration of chemical substances having mixed boundary conditions, developed as a total solution food package for migration problems.

References