Model for Transfer of Contaminant during the Coextrusion of Three-Layer Food Package with a Recycled Polymer. Effect on the Time of Protection of the Food of the Relative Thicknesses of the Layers

A. L. PEROU, S. LAOUBI, J. M. VERGNAUD

Lab. Materials and Chemical Engineering, Faculty of Sciences, University of St-Etienne, 23, Dr. P. Michelon, 42023 St-Etienne, France

Received 11 March 1998; accepted 7 December 1998

ABSTRACT: The heat transfer and subsequent contaminant transfer between a three-layer film during its coextrusion is considered. The film is made of a recycled polymer layer with a contaminant in it located between two virgin polymer layers. The effect of the relative thicknesses of the three layers on the contaminant transfer is especially studied. The final package is thus placed in contact with a food of finite volume, and the contaminant transfer into the food is thus determined. This transfer is controlled by diffusion through the package and convection into the liquid food. The effect of the relative thicknesses of each layer is estimated. This theoretical approach is allowed by building two numerical models based on finite differences: the one concerned with heat transfer and mass transfer with a temperature-dependent diffusivity through the film during its coextrusion; the other with the contaminant transfer into the food by considering the diffusion through the package and convection into the food, as well as the initial profile of concentration of the contaminant that obtained at the end of the stage of coextrusion. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 1939–1948, 1999

Key words: recycling; food packaging; functional barrier; coextrusion; heat and mass transfer; time of protection; modeling

INTRODUCTION

Recycling old package plastics is possible by reusing them in new packages after mastering the procedure of collection, control, and washing.[1,2] The main drawback with old plastics comes from the mass transfer that takes place during its previous use[3,4] and that old plastics, exhibiting potential contamination, cannot be in contact with the food.[5–7] Thus, tri-layer films must be coextruded in sandwich form, where the recycled polymer layer is located between two virgin polymer layers. As it takes some time for the contaminant to diffuse through the virgin polymer layer at room temperature, this layer plays the role of a functional barrier.[2,8,9]

The process of contaminant transfer is controlled by diffusion through the polymeric package[10] and either by convection in the liquid food[11] or by diffusion in the solid food.[12,13] The main problem that arises is to determine the period of time over which the food is protected.[1–3,5,7] Experiments are tedious and highly time consuming, and necessitate very accurate methods of analysis. Moreover, some changes may appear for the food or the contaminant in the food during this long time. On the other hand, the mathematical treatment is feasible only for the diffusion of the

Correspondence to: J. M. Vergnaud.

Journal of Applied Polymer Science, Vol. 73, 1939–1948 (1999)
 CCC 0021-8995/99/101939-10

contaminant through the package itself with a constant diffusivity and when the initial profile of concentration is uniform.[9–11] In practical cases, only when the package is in contact with a liquid food of finite volume with a finite coefficient of convective transfer into the liquid, is a numerical treatment feasible[12–14] despite an attempt to mix the equations obtained with the following two cases: the volume of liquid is finite, and the coefficient of convective transfer is infinite, or the volume of liquid is infinite and the coefficient of convection is finite.[15] Of course, the numerical treatment necessitates good knowledge of the process, as a model with finite differences, taking all the known facts into account, must be built. The parameters of importance are the diffusivity of the contaminant through the polymer, the coefficient of convective transfer into the liquid, and the initial profile of concentration of the contaminant in the package. Up to now, the following two main assumptions have been made in all previous studies: (1) the effect of the liquid food on the contaminant transfer is not considered, despite the fact that it sometimes takes place, enhancing the contaminant diffusion in the package[3,4]; and (2) the concentration of contaminant is initially uniform in the recycled polymer layer with no contaminant in the virgin polymer layers. In fact, previous studies showed that contaminant transfer takes place during the coextrusion of the polymer layers,[16–19] resulting from a high rate of diffusion over a short period of time. It must be said that the diffusivity increases rapidly with temperature, and becomes very great around the melting temperature of the polymer.[20]

The first objective to fulfill in this article is to show that contaminant transfer takes place during the stage of coextrusion. The numerical model takes into account the following stages during the process of coextrusion: heat transfer by conduction through the package and by convection at the film–air interface, enabling the film to cool from the melting temperature to the air temperature; simultaneously, a diffusion of the contaminant takes place through the package with a temperature-dependent diffusivity.[16–19] The ultimate objective, coupling experiments and modeling, is very difficult to reach, because of the difficulty of the experiments and also the problems in modeling. High-standard laboratories specialized in each of these two tasks, measuring the profiles of concentration of contaminant developed in polymer sheets for various temperature–time histories, processing multilayer films under standard and well-known conditions, are necessary to take part in this work. The problem of modeling is also hard work because of the two transfers that take place simultaneously.

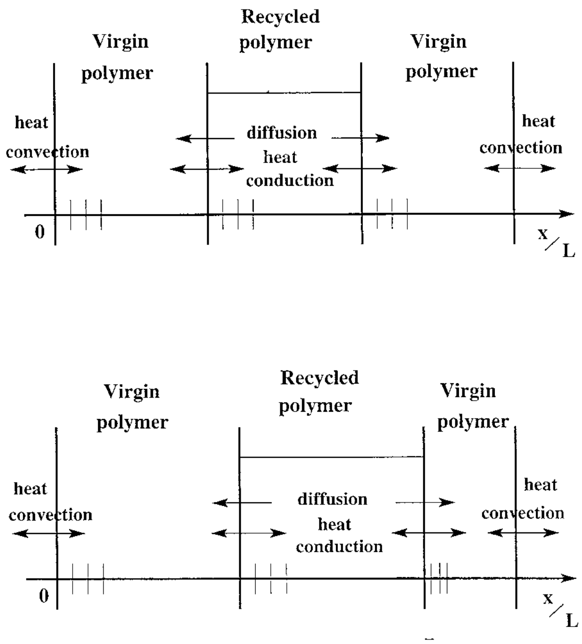
The other purpose is to place emphasis upon the effect of the thickness of each layer in the film on the time of protection of food. Another numerical model is built considering initially the final profile obtained at the end of the stage of coextrusion, as well as the diffusion of contaminant through the package and convection into the food. For a given thickness of the film of 90 microns, the thicknesses of the three layers range from equal thickness to larger thickness for the functional barrier in contact with the food. The results are expressed in terms of profiles of temperature and of contaminant concentration developed through the thickness of the film during the cooling period, and of profiles of concentration of the contaminant developed in the package when it is in contact with a liquid food at room temperature, as well as of kinetics of transfer into the food. Dimensionless numbers are used for the contaminant transfer to obtain master curves of use in various cases.

THEORETICAL

The following two processes are studied: the film coextrusion with heat transfer and mass transfer when the film is cooled from the melting temperature down to the air temperature, and the contaminant transfer in the food-package system at room temperature.

Process of Coextrusion of Sandwich Films: Assumptions

1. After coextrusion of the three layers in sandwich form, the recycled polymer, being located between two virgin polymers, heat, and mass transfers are unidirectional through the thickness of the film.
2. The three layers are in perfect contact, with no resistance to heat and mass transfers at their interface.
3. Initially, the contaminant concentration is uniform in the recycled polymer, while the virgin polymer layers are free from contaminant.
4. The package initially at the uniform temperature of coextrusion is cooled down in



TRI-LAYERS POLYMERS

Figure 1 Scheme of the process of coextrusion with heat and mass transfers through the three layers: having the same thickness (up), with different thickness (down).

motionless air at 20°C. Heat is transferred by free convection on each air–package interface, and by conduction through the package. As the polymers are of the same nature, a symmetry exists for the heat transfer with respect to the midplane of the film.

5. During the cooling period, contaminant transfer takes place through the film controlled by Fickian diffusion. The diffusivity varies with temperature by following an Arrhenius' law.

Mathematical Treatment (Fig. 1)

See Nomenclature, page 19.

Heat Transfer

The equation of unidirectional heat conduction through the film is:

$$\rho c \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(\lambda \cdot \frac{\partial T}{\partial x}\right) \tag{1}$$

The initial condition is obvious:

$$t = 0 \quad 0 < x < L \quad T_{in} \tag{2}$$

The boundary conditions

$$\begin{aligned} t > 0 \quad x = 0 \quad & \lambda \left[\frac{\partial T}{\partial x}\right]_s = h_t(T_{s,t} - T_{\text{air}}) \\ x = L \quad & \end{aligned} \tag{3}$$

express the fact that the rate of heat transferred by convection is constantly equal to the rate at which heat is brought to the film surface by conduction.

The midplane of the film being a plane of symmetry for heat transfer, there is:

$$t \geq 0 \quad x = \frac{L}{2} \quad \frac{\partial T}{\partial x} = 0 \tag{4}$$

Contaminant Transfer

The equation of unidirectional diffusion of contaminant through the film is:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D_{x,t} \cdot \frac{\partial C}{\partial x}\right) \tag{5}$$

with the temperature-dependent diffusivity

$$D_{x,t} = D_0 \cdot \exp\left(-\frac{A}{T}\right) \tag{6}$$

The initial condition is:

$$\begin{aligned} t = 0 \quad & \text{recycled polymer } C_{in} \\ & \text{virgin polymer } C = 0 \end{aligned} \tag{7}$$

The condition at each polymer interface is given by the continuity of heat transfer, with the same thermal conductivity.

$$-\lambda\left(\frac{\partial T}{\partial x}\right)_{t,s^-} = -\lambda\left(\frac{\partial T}{\partial x}\right)_{t,s^+} \tag{8}$$

Numerical Analysis

The Crank-Nicolson method is used for calculating the temperature and contaminant concentration at each time and position. The thickness of the film is divided into N slices of constant thickness Δx, and increments of time Δt, are considered. The integers i and j characterizing position and time appear in the calculation, as well as the three dimensionless numbers:

$$\text{heat convection} \quad HC^j = \frac{\Delta x \cdot ht^j}{\lambda_i^j} \tag{9}$$

$$\text{heat conduction} \quad HH_i^j = \frac{(\Delta x)^2}{\Delta t} \cdot \frac{\rho c}{\lambda_i^j} \tag{10}$$

$$\text{contaminant transfer} \quad MC_i^j = \frac{(\Delta x)^2}{\Delta t \cdot D_i^j} \tag{11}$$

Process of Transfer in the Package-Food System (Fig. 7): Assumptions

1. The contaminant transfer is controlled by Fickian diffusion through the package and convection into the liquid with a finite coefficient of convective transfer.
2. The transfer of contaminant only is considered, the transfer of the food into the polymer being neglected.
3. The initial profile of concentration of the contaminant in the package is that obtained at the end of the coextrusion process.
4. The transfer through the package is unidirectional: as in a bottle the thickness of the package is much less than the radius of the bottle.

Mathematical Treatment

The equation of unidirectional diffusion through the package is:

$$\frac{\partial C}{\partial t} = D \cdot \frac{\partial^2 C}{\partial x^2} \tag{12}$$

with the constant diffusivity D.

Initially, the profile of concentration in the package is that obtained at the end of the coextrusion process.

At the package–food interface, the rate of contaminant entering the liquid food is constantly equal to the rate at which the contaminant is brought to this interface by diffusion in the package.

$$t > 0 \quad x = L \quad D\left|\frac{\partial C}{\partial x}\right|_{L,t} = hm(C_{L,t} - C_{\text{food},t}) \tag{13}$$

On the other face of the package in contact with air,

$$t > 0 \quad x = 0 \quad \frac{\partial C}{\partial x} = 0 \tag{14}$$

meaning that the contaminant does not evaporate at room temperature.

Numerical Treatment

The problem with a finite volume of food and a finite coefficient of convective transfer, as well as with an initial nonuniform profile of concentration of contaminant must be resolved by using a numerical method with finite differences.[12,13] The principle of the method is summarized as follows: the thickness of the package L is divided into N slices of thickness Δx, and each position is associated with the integer i. From the matter balance evaluated within each slice during the increment of time Δt, the new concentration after elapse of time Δt is expressed in terms of the previous concentrations.

Inside the package:

$$C_i^{j+1} = \frac{1}{MC}\left[C_{i-1}^j + (M-2)C_i^j + C_{i+1}^j\right] \tag{15}$$

On the surface in contact with the food

$$C_N^{j+1} = \frac{1}{MC} \times \left[2C_{N-1}^j + (M-2N-2)C_N^j + 2N \cdot C_{\text{food},t}\right] \tag{16}$$

At the surface in contact with air

$$C_0^{j+1} = \frac{1}{M}\left[2C_0^j + (M-2)C_0^j\right] \tag{17}$$

The amount of contaminant remaining in the package at every time is evaluated by integrating the concentration of contaminant with respect to space. The amount of contaminant in the liquid at a time is the difference between the amount of contaminant initially in the package and that remaining after this time.

The concentration in the liquid is:

$$C_{\text{food},t} = \frac{M_t}{L'} \tag{18}$$

Table I Characteristics for Heat and Mass Transfer with the Coextrusion Process

$\alpha = 1 \times 10^{-3} + 7 \times 10^{-7} \times T$	($cm^2 \cdot s^{-1}$)	T : Celsius
$\lambda = 6.1 \times 10^{-4} + 1 \times 10^{-7} \times T$	($cal \cdot cm^{-1} \cdot s^{-1} \cdot deg^{-1}$)	T : Celsius
$h = 2 \times 10^{-4} \times \|T_L - T_{air}\|^{0.25}$	($cal \cdot cm^{-2} \cdot s^{-1} \cdot deg^{-1}$)	T : Celsius
$D = 28.240 \times \exp\left(-\frac{13800}{T}\right)$	(cm2 · s−1)	T : Kelvin

where M_t is the amount of contaminant transferred into the food, per unit area and L' is the thickness of the food.

The dimensionless numbers are:

$$MC = \frac{(\Delta x)^2}{D \cdot \Delta t} \quad N = \frac{h \cdot \Delta x}{D} \tag{19}$$

RESULTS

Two kinds of results are of interest: the one concerning the process of heat and contaminant transfer through the tri-layer films during the stage of coextrusion; the other with the contaminant transfer in the package–food system at room temperature.

Tri-layer polymer films of total thickness of 90 microns with different thicknesses for each layer, are considered: the first with 30/30/30 microns, the second with 10/40/40 microns, and the third with 20/35/35 microns.

The internal layer is made of recycled polymer with a uniform concentration of contaminant C_{in}, while the two surrounding layers are made of virgin polymer.

Polyethylene terephthalate is chosen for the polymer because its thermal properties are found in the literature.[21] The values of the diffusivity around the melting temperature are not known yet despite some attempts,[20] which are necessary to achieve through difficult experiments done with various temperature–time histories using the numerical model described in this article, and some assumptions are made for the temperature-dependent diffusivity (Table I).

Process of Coextrusion

The three layers are separately heated up to the melting temperature and coextruded at the temperature of 300°C. The final tri-layer film is allowed to cool down in motionless air at room temperature. This process of coextrusion is thus responsible for heat transfer during the cooling period and a subsequent contaminant transfer. Despite the fact that these two transfers occur simultaneously, they are studied in succession.

Heat Transfer during Coextrusion

Heat is transferred by free convection at the film–air interface and by conduction through the tri-layer film, during the cooling period in air.

The profiles of temperature developed through the thickness of the film are drawn in Figure 2, as obtained using the numerical model and the data (Table I). The temperature–time histories are drawn in Figure 3, by considering various places of interest through the thickness of the film.

These curves in Figures 2 and 3 can lead to a few observations: (1) the temperature decreases rather slowly and drops to 160°C in about 4 s. It takes 0.5 s for the temperature to decrease down to 255°C. (2) The profiles of temperature are rather flat, as the rates of heat transferred by conduction and by convection are about the same. (3) The low convective coefficient of heat transfer in motionless air as well as the low thermal con-

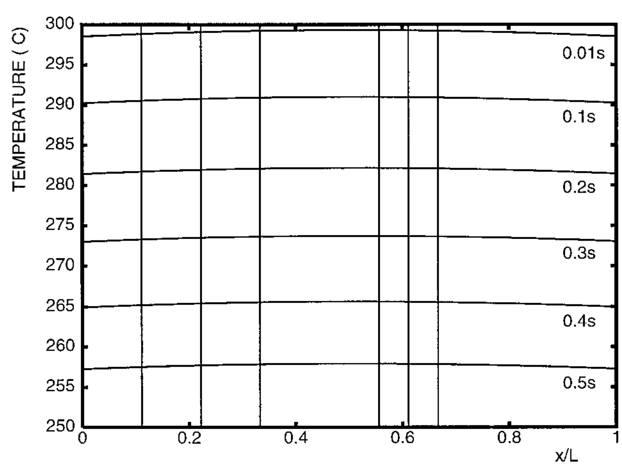


Figure 2 Profiles of temperature developed through the thickness of the three-layer films during their coextrusion, at various times.

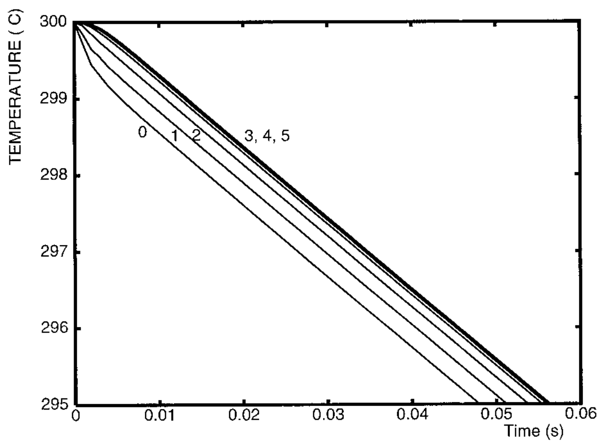


Figure 3 Temperature–time histories at various places through the three layer films during coextrusion on the surface (0); at 10 microns of the surface (1); at 20 microns of the surface (2); at 30 microns of the surface (3); at 40 microns of the surface (4); at the middle (5).

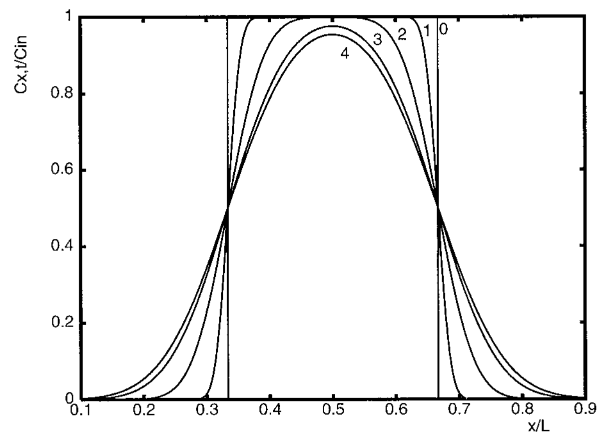


Figure 4 Profiles of concentration of contaminant developed through the three-layer film during coextrusion at various times, with layers of same thickness at 300°C, 30/30/30 microns: initial (0); after 0.01 s (1); after 0.1 s (2); after 0.5 s (3); after 10 s (4).

ductivity of the polymer are responsible for this slow decrease in temperature. (5) The midplane of the film is a plane of symmetry for heat transfer, as the three polymer layers have the same properties and the film is cooled down in the same way on each face.

Contaminant Transfer during Coextrusion

Because of the high temperature around the melting point of the polymer and its low decrease rate, a contaminant transfer takes place. It is evaluated by using the numerical model.

A problem arises with the diffusivity of a contaminant at high temperature and its temperature dependency. A method has been developed to obtain accurate data.[20] It necessitates tedious experiments and calculation using a numerical model considering both heat and contaminant transfer. Moreover, the temperature dependency of the diffusivity should obey the following assumption: the diffusivity varies with temperature by following an Arrhenius' law in the same way as the viscosity at a low rate of shear.[21]

The profiles of contaminant concentration develop through the tri-layer film in different ways, depending on the relative thicknesses of each layer. Three films are considered: with layers of the same thickness in Figure 4 (30/30/30 microns), with different thicknesses for the layers in Figure 5 (10/40/40 microns) and Figure 6 (20/35/35 microns), by keeping the total thickness of the film constant at 90 microns.

Conclusions of interest can be drawn from these curves: (1) profiles of contaminant concentration develop through the thickness of the film very quickly at the beginning of the cooling period, when the temperature is high. A significant transfer is observed within 0.1 s. (2) The rate of diffusion of contaminant remains high over a short period of time, as it decreases rather quickly with temperature. Over the period of time between 0.5 and 10 s, the rate decreases to a very low value. (3) The effect of the relative thicknesses of the three layers appears to be of great importance, especially the virgin layer, which is

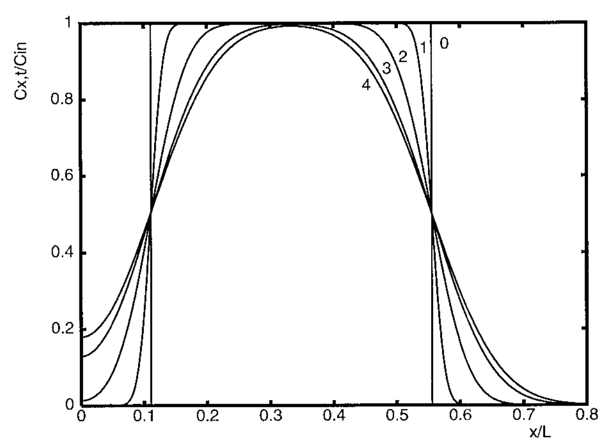


Figure 5 Profiles of concentration of contaminant developed through the three-layer film during coextrusion at various times, with different layer thicknesses, at 300°C, 10/40/40 microns: initial (0); after 0.01 s (1); after 0.1 s (2); after 0.5 s (3); after 10 s (4).

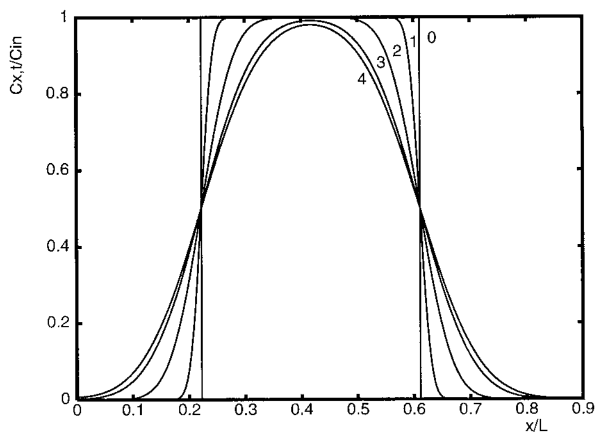


Figure 6 Profiles of concentration of contaminant developed through the three-layer film during coextrusion at various times, with different layer thicknesses, at 300°C, 20/35/35 microns: initial (0); after 0.01 s (1); after 0.1 s (2); after 0.5 s (3); after 10 s (4).

the functional barrier in contact with the food in the package-food system. Its thickness is varied from 30 to 40 microns, with the same thickness for the recycled layer that will be in contact with the food. (4) The obvious statement clearly holds: the thinner the functional barrier, the more effective the contaminant transfer. Thus, with the thickness of 30 microns, the contaminant reaches up to the relative thickness of 0.9, while the corresponding value is 0.8 for the thickness of 40 microns. (5) The profiles of contaminant concentration shown in Figures 4–6 essentially result from the thickness of the functional barrier, as the profiles of temperature are rather flat through the films during the cooling period. (6) After 10 s, the profiles of contaminant concentration remain approximately stable, when considering a time scale in hours. They will be the initial profiles in the package-food system (Figs. 4–6).

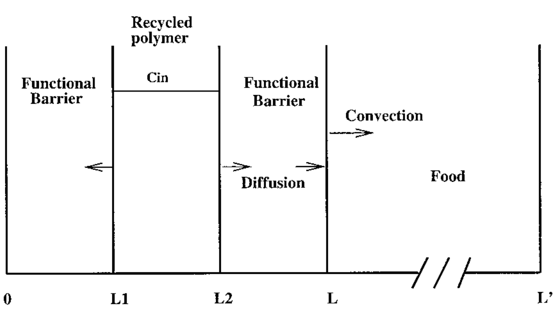


Figure 7 Scheme of the contaminant transfer in the package-food system.

Table II Characteristics of Mass Transfer with the Package-Food System

Thickness of the package = 90 microns
Thickness of the liquid = 1.6 cm
1 liter of liquid in a cubic package
$D = 10^{-10}$ cm²/s
$h = 10^{-8}$ cm/s

Process of Contaminant Transfer into the Food

The package made of tri-layer polymers is put in contact with liquid food. The contaminant is transferred by diffusion through the package and convection into the liquid with a finite coefficient of convection. This coefficient of convection is that obtained in olive oil in a previous study.[22] The value of the diffusivity is constant as the concentration of the contaminant is low, as shown in Table II.[23]

A volume of 1 liter of liquid in a cubic package is considered, leading to a thickness of the food of 1.6 cm.[24,25]

Profiles of Concentration of Contaminant in the Package

The profiles of contaminant concentration are drawn through the three packages in Figure 8 (30/30/30 microns), Figure 9 (20/35/35 microns), and Fig. 10 (10/40/40 microns), at various times.

Dimensionless numbers are used: the relative thickness x/L; the concentration C_{xt} at position

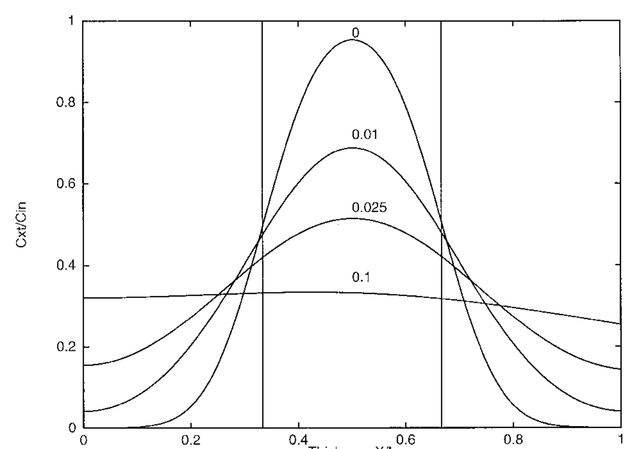


Figure 8 Profiles of concentration of contaminant developed through the package in the package-food system at room temperature, at various times (Dt/L^2), with the three layers of the same thickness (30/30/30 microns).

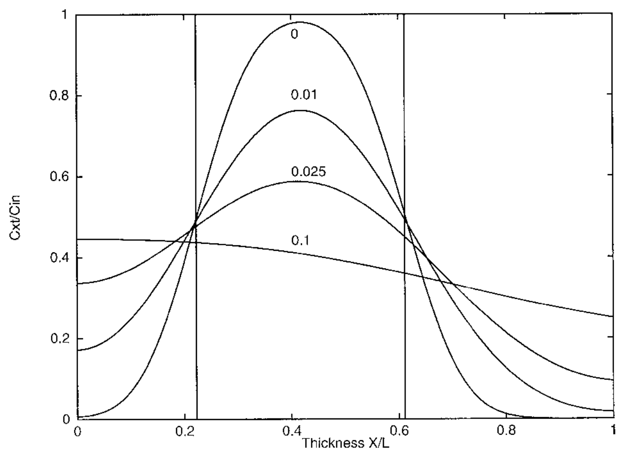


Figure 9 Profiles of concentration of contaminant developed through the package in the package-food system at room temperature, at various times (Dt/L^2), with the different layer thicknesses (20/35/35 microns).

x/L and time t as a fraction of the initial concentration in the recycled polymer layer before coextrusion; Dt/L^2 instead of time, L being the thickness of the package.

The following conclusions can be pointed out: (1) at time 0 for the transfer into the food, the profiles of contaminant concentration in the package are the same as those obtained at the end of the process of coextrusion. (2) These profiles give a fuller insight into the nature of the process. (3) For short times, for example, Dt/L^2 less than 0.005 for the three packages (not shown in Figs. 8–10), the concentration on the two recycled layer faces drops to $C_{xt}/C_{in} = 0.5$, as shown in an earlier study,[9] when the contaminant does not reach the package faces. (4) The contaminant reaches the food surface at a given time, depending on the relative thicknesses of the layer of the packages. This fact is shown more precisely with the kinetics of the contaminant transfer into the food. (5) A flat gradient of concentration is observed on the external surface of the package, as the contaminant does not evaporate.[4] (6) A slight gradient of concentration is shown at the package surface in contact with the food, obeying eq. (13), because the coefficient of convective transfer in the food is rather low.[4] (7) A typical pattern is observed for long times (Dt/L^2 larger of around 0.1), because the contaminant concentration becomes greater on the external package surface, as shown in previous studies.[12] (8) Comparison between the profiles drawn in Figures 8–10 shows the effect of the relative thicknesses of each layer of the package. It takes some time for the contaminant to reach the food, and the obvious statement holds: the thicker the functional barrier, the longer the time of food protection. Nevertheless, the conclusion is not so simple, as the thickness of the recycled layer also plays a role. (9) The effect of the total thickness of packages on the time of food protection was studied when the packages were made of three layers of same thicknesses.[26] From this study, it stands to reason that a better way for reusing recycled polymer in food packages consists of using packages with different thicknesses for the layers. Optimization of the relative thickness of each layer is an important contribution.

Kinetics of Transfer of Contaminant into the Food

The kinetics of contaminant transfer into the food are drawn in Figure 11 with the three packages: 1—(30/30/30 microns), 2—(20/35/35 microns), and 3—(10/40/40 microns). Dimensionless numbers are used: the amount of contaminant transferred up to time t as a fraction of the initial amount in the package; Dt/L^2 instead of time.

Some conclusions are worth noting: (1) typical shapes for the kinetic curves are observed: the rate of transfer into the food increases regularly with time from a zero value at a given time that depends on the package. Of course, these shapes result from the presence of the functional barrier. When no functional barrier exists, the kinetics of transfer starts at time zero with an oblique tangent that depends on the value of the convective coefficient[4] of the contaminant in liquid food. (2)

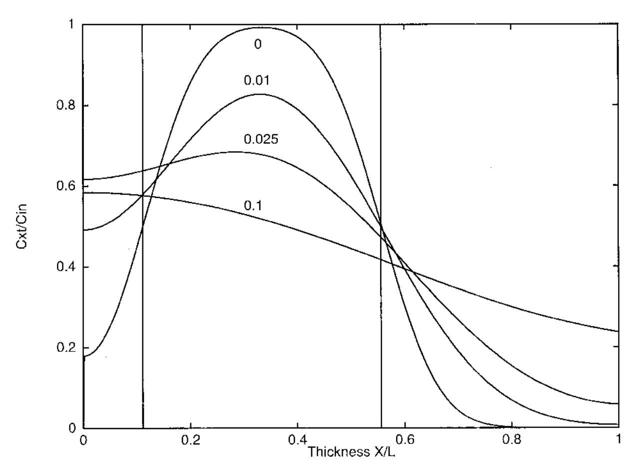


Figure 10 Profiles of concentration of contaminant developed through the package in the package-food system at room temperature, at various times (Dt/L^2), with the different layer thicknesses (10/40/40 microns).

The period of time over which the food is protected is of great importance. Some values for the relative amount of contaminant transferred are given at various times (Dt/L^2) in Table III. (3) The effect of the relative thickness of the three layers in the packages on the kinetics of contaminant transfer clearly appears in Figure 11, as well as in Table III. Of course, the kinetics of contaminant transfer is faster for the thinner functional barrier. (4) In fact, the answer is not as simple as said in (3); the time of protection depends on the relative thicknesses of the three layers, as shown in Table III. Nevertheless, it stands to reason from either the profiles of concentration developed through the packages in Figures 8–10 or the kinetics of transfer in the food shown in Figure 11, that the third package with dimensions of 10/40/40 microns is of the utmost interest for two reasons: it offers a larger ratio for the recycled polymer (44.4%); it is responsible for a longer period of time for food protection.

CONCLUSIONS

A few conclusions can be drawn from this theoretical study about the process and the parameters that intervene.

It clearly appears that during the stage of coextrusion, a contaminant transfer takes place through the tri-layer polymer. As it takes some time for the film to cool down, profiles of concentration of contaminant develop through the film, and the functional barrier is not free from contaminant. Three parameters are of concern for this mass transfer: the first two concerned with the processing of the film such as the temperature of coextrusion and the coefficient of convective heat transfer, and the thickness of the film for a given polymer. It is possible to act upon the heat convection coefficient by stirring the air. During the short period of time over which the temperature of the film is high, the diffusivity of the contaminant as well as its temperature dependency is also a main parameter. Thus, precise experiments have to be made, coupled with a numerical model taking into account heat and mass transfer[18] to obtain accurate data.

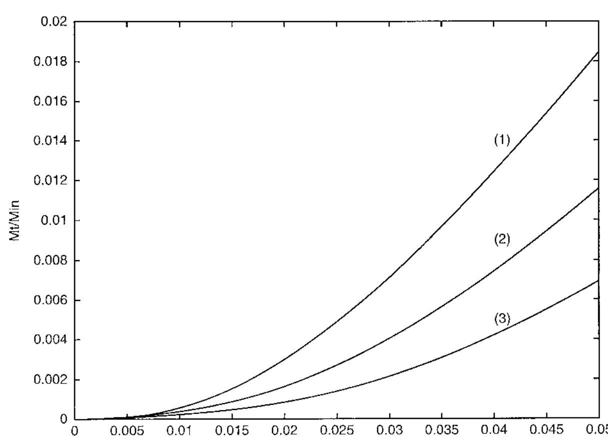


Figure 11 Kinetics of contaminant transfer into the food (with dimensionless numbers) for the three packages of different layer thicknesses in contact with the food: 1 (30/30/30 microns); 2 (20/35/35 microns); 3 (10/40/40 microns).

Table III Contaminant Transfer in Food (M_t/M_{in} × 10^5) at Various Times (Dt/L^2)

Package Dt/L^2 =	0.001	0.005	0.01
30/30/30	2.6	8.6	56
20/35/35	1.2	7.2	38
10/40/40	1.1	6.1	30

Of course, the contaminant transfer during the coextrusion process is responsible not only for a shorter time of protection of the food, but also for faster kinetics of contaminant transfer into the food.

The effect of the relative thickness of the recycled and the virgin polymer layers on the contaminant transfer into the food is of main importance. Obviously, it can be said that the thicker the functional barrier, the longer the time of protection. However, from the economical point of view, a rather large thickness of the recycled polymer must be desired. As a result a tri-layer package with the recycled polymer located between two virgin polymer layers is the best solution: a thin virgin polymer layer on the external surface of the future package is necessary to protect the customer's hand, while the other two polymer layers are much larger. Modeling is of interest to find the optimal conditions, by considering the whole process.

Finally, this article has paved the way in the subject of preparing multilayer packages, by pointing out the tasks that are to be done: determination of the diffusivity at high temperature and its temperature dependency by measuring the profiles of concentration of contaminant developed through three layer sheets submitted to various temperature–time histories; coextruding multilayer films under well-known conditions.

NOMENCLATURE

Symbol Abbreviation	Description
A	Constant in eq. (6)
α	Thermal diffusivity of the polymer
c	Heat capacity of the package
C	Concentration of contaminant
C_{in}	Initial concentration in the recycled polymer
C_i^j	Concentration at position i and time $j \cdot \Delta t$ in the package
$C_{\text{food},t}$	Concentration of contaminant in the food at time t
D	Diffusivity of contaminant
D_0	Constant in eq. (6)
$D_{x,t}$	Diffusivity at position x and time t
λ	Thermal conductivity of the polymer
ρ	Density of the polymer
ht	Coefficient of convective heat transfer
hm	Coefficient of convective mass transfer
MC	Dimensionless number in eqs. (11) and (19)
HC^j	Dimensionless number for heat convection
HH_i^j	Dimensionless number for heat conduction
Δx, Δt	Increments of space, of time, respectively
N	Dimensionless number for convective mass transfer in eq. (19)
t	Time
T	Temperature
$T_{s,t}$	Temperature at the polymer surface and time t
T_{air}	Constant temperature of air (20°C)
T	Temperature in eq. (6) (Kelvin)
T_{in}	Initial temperature

REFERENCES

1. Kirschner, E. M. Chem Eng News 1996, Nov. 4.
2. Feigenbaum, A.; Laoubi, S.; Vergnaud, J. M. J Appl Polym Sci 1997, 66, 597.
3. Frisch, H. L. J Polym Sci Phys Ed 1978, 16, 1651.
4. Vergnaud, J. M. In Liquid Transport Processes in Polymeric Materials; Prentice Hall Publ: Englewood Cliffs, NJ, 1991.
5. Food and Drug Administration. Points to consider for the use of recycled plastics: food packaging, chemistry considerations; FDA Div. of Food Chemistry and Technology Public. HP410, Washington, DC, May 1992.
6. Rulis, A. In Food Safety Assessment; Finley, J. W.; Robinson, S. F.; Armstrong, D. J., Eds.; ACS Symp Series 1992, 484, 132.
7. Food and Drug Administration, Code of Federal Register, 58, 52,719 (Oct. 12, 1993).
8. Begley, T. H.; Hollifield, H. C. Food Technol 1993, 109.
9. Laoubi, S.; Feigenbaum, A.; Vergnaud, J. M. Pack Technol Sci 1995, 8, 17.
10. Laoubi, S.; Vergnaud, J. M. Pack Technol Sci 1995, 8, 17.
11. Lum Wan, J. A.; Chatwin, P. C.; Katan, L. L. Philos Trans R Soc Lond A 1995, 350, 379.
12. Laoubi, S.; Vergnaud, J. M. Food Packag Contam 1996, 13, 293.
13. Laoubi, S.; Vergnaud, J. M. Plast Rubber Compos Process Appl 1996, 25, 83.
14. Rosca, D.; Vergnaud, J. M. Plast Rubber Compos Process Appl 1997, 26, 235.
15. Etters, J. M. Ind Eng Chem Res 1991, 30, 589.
16. Perou, A. L.; Vergnaud, J. M. Comput Theoret Polym Sci 1997, 7, 1.
17. Perou, A. L.; Vergnaud, J. M. Polym Eng 1997, 17, 349.
18. Perou, A. L.; Vergnaud, J. M. Polym Testing 1997, 16, 575.
19. Perou, A. L.; Vergnaud, J. M. Int J Numerical Methods Heat Fluid Flow 1998, 8, 841.
20. Riquet, A. M.; Feigenbaum, A.; Perou, A. L.; Vergnaud, J. M. (unpublished results).
21. I.C.I. Melinar Polyethylene Terephthalate. G5: Physical properties.
22. Riquet, A. M.; Laoubi, S.; Feigenbaum, A.; Vergnaud, J. M. Food Addit Contam 1998, 15, 690.
23. Nir, N. M.; Ram, A.; Miltz, J. J Polym Eng Sci 1996, 36, 862.
24. Vergnaud, J. M. Colloid Interface Sci 1998, 78, 267.
25. Vergnaud, J. M. Modern Food Packaging, Indian Inst Packag 1998; p 781.
26. Perou, A. L.; Laoubi, S.; Vergnaud, J. M. Comput Theoret Polym Sci 1998, 8, 331.