

## Diffusion of Heptane in Polyethylene Vinyl Acetate: Modelisation and Experimentation

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**Abstract:** This article describes the mass transfer in food packaging, where the plastic is brought into contact with food, certain transfers implemented according to a complex process. The method we used in this work, we coupled modeling with experimentation in order to understand the packaging plastic touch interface transfers / food. The numerical model takes into account the experimental study. The kinetics of diffusion transfer and diffusivity of liquid simulator were determined. The analytical and numerical model has been developed which aims to give better information on the concentration of liquid simulator inside the package (PEVA) and simulate a few hours' transfers lasting in reality several months.

**Keywords:** Packaging material, Diffusion, Modelisation, Finite difference method.

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### I. Introduction

Food contact materials play an essential role in the food conservation and protection. They have also incessant marketing function and have to be recoverable. When the polymer is in contact with the packaging, some transfers take place, this contact influence on packaged products as well as on packaging materials.

The goal of this article is to understand these transfers, by coupling modeling with experimentation and Construct a mathematical model and a numerical model able to describe the diffusion process.

### II. Materials and methods

The study the liquid diffusion into a polymer simulator is based on the following simplifying assumptions [3]:

- ✓ The distribution is in accordance with fick's laws.
- ✓ The diffusion coefficient is independent of concentration.
- ✓ The diffusion in the sphere is radiale.
- ✓ The polymer was spherical in shape with a constant radius, as the amount of ethanol is very small.
- ✓ The chemical does not evaporate on surface.

#### A. Analytical processing[1] [2] [5]

When the diffusion is radial, Fick's second law expressing the diffusion equation under transient conditions is in the general form.

$$\frac{\partial C}{\partial t} = \frac{1}{r^2} \cdot \frac{\partial}{\partial r} \left[ D \cdot r^2 \cdot \frac{\partial C}{\partial r} \right] (eq1)$$

Where **C** is the concentration at time **t** and at a distance **r** from the center of the sphere.

When the diffusivity **D** is constant, the diffusion equation takes the form of:

$$\frac{\partial C}{\partial t} = D \cdot \left[ \frac{\partial^2 C}{\partial r^2} + \frac{2}{r} \cdot \frac{\partial C}{\partial r} \right] (eq2)$$

Analytical solutions can be obtained when the diffusivity is constant. Problems with a concentration-dependent diffusivity need numerical methods. By putting:

$$U = C \cdot r (eq3)$$

Equation (2) becomes:

$$\frac{\partial U}{\partial t} = D \cdot \frac{\partial^2 U}{\partial r^2} (eq4)$$

Equation (4) is similar to the equation obtained for diffusion in one dimension through the plane sheet.

Case study: The initial distribution in the sphere is constant:  $C_i$ . In this case:

$$\text{Initial } t = 0 \quad 0 < r < R \quad U = C_i \text{ (eq5)}$$

$$\text{Boundary } t > 0 \quad r = 0 \quad U = 0$$

$$r = R \quad U = RC_0 \text{ (eq6)}$$

The solution of the equation of diffusion becomes:

$$\frac{C_{r,t} - C_i}{C_0 - C_i} = 1 + \frac{2R}{\pi r} \sum_{n=1}^{\infty} \frac{(-1)^n}{n} \sin \frac{n\pi r}{R} \exp\left(-\frac{n^2\pi^2}{R^2} Dt\right) \text{ (eq7)}$$

This solution in this form cannot be used for the center of the sphere ( $r=0$ ).

$$\text{As the limit of } \frac{\sin x}{x} \rightarrow 1 \text{ , when } x \rightarrow 0$$

Equation (7) for the center of the sphere is reduced to:

$$\frac{C_{r,t} - C_i}{C_0 - C_i} = 1 + 2 \sum_{n=1}^{\infty} (-1)^n \cdot \exp\left(-\frac{n^2\pi^2}{R^2} Dt\right) \text{ (eq8)}$$

### B- Analytical model

The total amount of diffusing substance going into or leaving the sphere is given by integrating Fick's first law according to time.

$$M_t = -\int_0^t D \left( \frac{\partial C}{\partial r} \right)_{r=R} dt \text{ (eq9)}$$

$$\text{By considering: } \sum_1^{\infty} \frac{1}{n^2} = \frac{\pi^2}{6}$$

$$\text{And: } M_{\infty} = \frac{4}{3} \pi R^3 \cdot C_0$$

We can obtain:

$$\frac{M_{\infty} - M_t}{M_{\infty}} = \frac{6}{\pi^2} \sum_1^{\infty} \frac{1}{n^2} \cdot \exp\left(-\frac{n^2\pi^2}{R^2} Dt\right) \text{ (eq10)}$$

Another expression of the solution of the equation of diffusion in the sphere (equation 5) is given by:

$$\frac{C_{r,t} - C_i}{C_0 - C_i} = \frac{R}{r} \sum_0^{\infty} \left\{ \operatorname{erfc} \frac{(2n+1)R+r}{2(Dt)^{0.5}} - \operatorname{erfc} \frac{(2n+1)R-r}{2(Dt)^{0.5}} \right\} \text{ (eq11)}$$

The kinetics for the matter transported is:

$$\frac{M_t}{M_{\infty}} = 6 \left( \frac{Dt}{R^2} \right)^{0.5} \left\{ \pi^{-0.5} + 2 \sum_1^{\infty} \operatorname{ierfc} \frac{nR}{(Dt)^{0.5}} \right\} - 3 \frac{Dt}{R^2} \text{ (eq12)}$$

Case of short times: Equation (8) is very useful for short times because it can be reduced to:

$$\frac{M_t}{M_{\infty}} = \frac{6}{R} \left( \frac{Dt}{\pi} \right)^{0.5} \text{ (eq13)}$$

### C- Numerical model - Finite difference method-[4] [5]

Analytical solutions can be obtained when the diffusivity is constant. Problems with a concentration-dependent diffusivity need numerical methods. In this case, the problem must be solved by using the numerical Finite difference method.

*Case study: The amount of the remaining ethanol within the sphere.*

The principle of the method is as follows: the sphere is divided into N spherical membranes of constant thickness  $r$  and the matter balance is evaluated within each membrane during the increment of time  $t$ .

The amount of chemical located in the sphere at time t is obtained by integrating with respect to space the concentration of the chemical at this time:

$$M_t = 4\pi \int_0^R r^2 \cdot C_{r,t} \cdot dr \text{ (eq14)}$$

This expression can be rewritten using finite difference method.

$$M_t = 4\pi(\Delta r)^3 \left[ \frac{C_0}{24} + \sum_1^2 n^2 C_n + \frac{9}{8} (n-1)^2 C_{n-1} + \frac{3}{8} n^2 \cdot C_n \right] \text{ (eq15)}$$

#### D- Experimental procedure

The material used is polyethylene vinyl acetate (also known as PEVA) is the copolymer of ethylene and vinyl acetate. And our product simulator used is heptane or n-heptane is the straight-chain alkane with the chemical formula  $H_3C(CH_2)_5CH_3$  or  $C_7H_{16}$  and his density is:  $0.6795 \text{ g mL}^{-1}$ . Heptane is a colourless liquid and odor is petrolic.

#### Contacting:

The contacting sample of polyethylene vinyl acetate is carried out with heptane at  $25^\circ\text{C}$ . During the contact, we measured the specific mass of the sphere each time to study the evolution of the mass sphere.

### III. Results And Discussions

The percentage of heptane mass variation inside our plastic sphere (**fig.1**) is given by the following equation:

$$\Delta m = \frac{m_t - m_0}{m_0} \times 100 \text{ (eq16)}$$

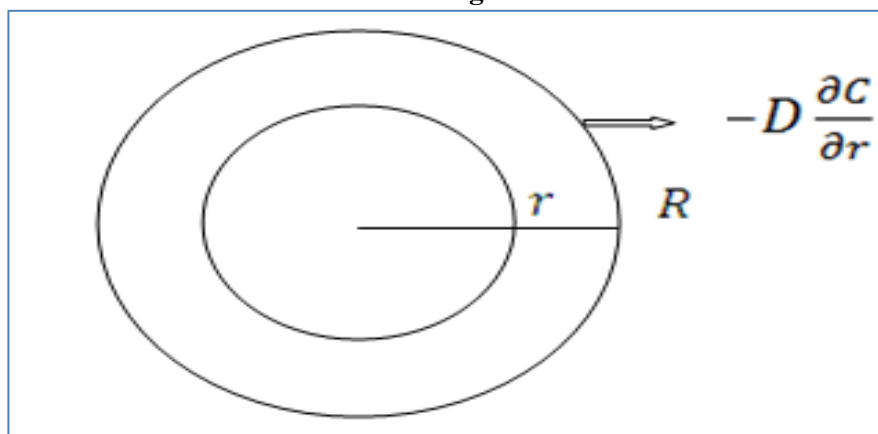
Effect of The diffusion coefficient is given by this relation:

$$D = \pi \left( \frac{\alpha \cdot R}{6} \right)^2 \frac{1}{60} \text{ (eq17)}$$

En  $\text{cm}^2/\text{s}$ :                       $D = 4,24 \cdot 10^{-6} \text{ cm}^2/\text{s}$

- Figure 2- shows the variation of the amount of heptane in the PEVA with time, we note that the heptane in polyethylene vinyl acetate mass increases with the contact time. Until equilibrium reached.
- Figure 3- show the amount of heptane in the material according to simulation time for each solution : analytical solution, numerical solution and experience. We notice from this figure that three solutions give the same variation in the amount of heptane absorbed as a function of time. So we concluded that the model is validated.
- Figure 4 - show the profile of the concentration of heptane after every 10 min.
  - a- The heptane concentration within the sphere is low, in contrast to surfaces.
  - b- This concentration profile is aimed to give good information on heptane inside PEVA, is for each point in our sample we can easily determine its concentration.

### IV. Figures



**Fig.1-** Schema of the circular cross section of a sphere of radius R.

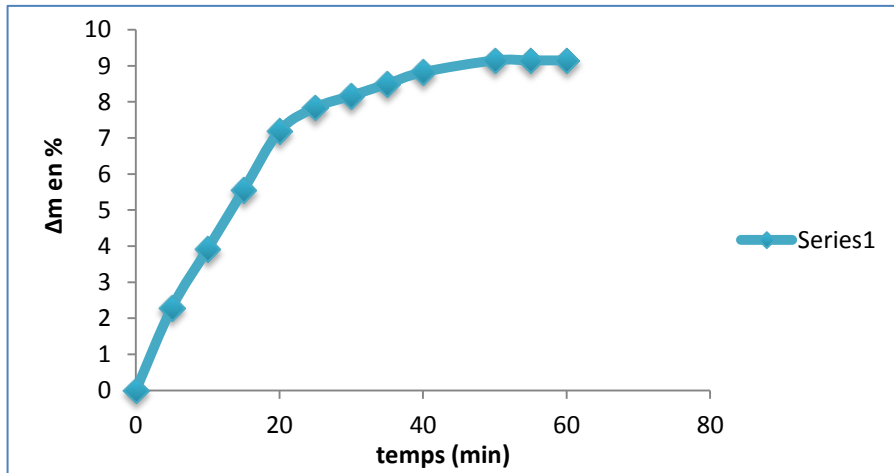


Fig.2- Variation of the amount of heptane in the PEVA

➤ Le profil de la diffusion d’heptane dans le PEVA

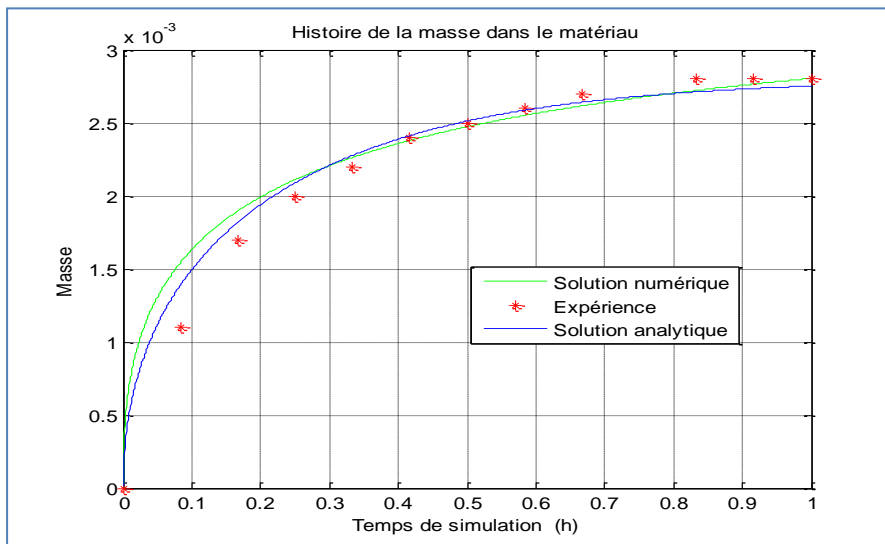


Fig.3- The amount of heptane in the material as a function of time for the simulation of three solutions.

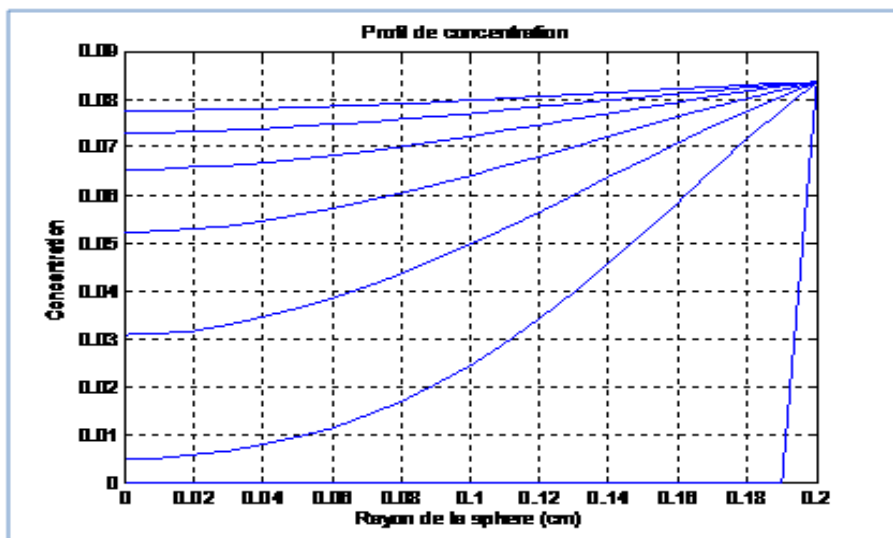


Fig.4- The profile of the concentration of heptane after every 10 min

## V. Conclusion

Through this work, we contributed to the study and development of new methods coupling experiments with modeling to understand the behavior of plastic packaging in contact with the food products.

The study was conducted by weighed following the evolution of the mass transferred over time. The polyethylene vinyl acetate contact with heptane (considered simulating agent) at a temperature of 25°C showed that the amount of heptane in the polymer increases with time.

Model validation was made by comparing the theoretical results with experimental results.

The resulting profile gives better information on concentrations of heptane inside the package polyethylene vinyl acetate.

This model we allow a few hours to simulate mass transfer in reality lasting several months.

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