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# Finite Element Modeling of Fuel Emission for Thermoplastic Multilayer Fuel Tanks with Optimization of Barrier Properties

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#### **ABSTRACT**

A numerical simulation model for the prediction of fuel hydrocarbon permeation is presented in this work. The barrier layer thickness optimization for thermoplastic multilayer fuel tanks is also considered. The diffusion model is based on the continuum approach with steadystate permeation regime across the multilayer polymeric wall. The hydrocarbon flux through the multilayer wall is determined by assuming continuity in vapor pressure at the polymer-polymer interface. Since the pinch-off zone is known to be the major source of emission per unit area, a method has been developed to automatically detect this zone at the end of extrusion blow molding process. After then, an improvement to the diffusion model has been proposed in order to evaluate adequately the hydrocarbon permeation through this specific area. Finally, a gradient-based algorithm is applied to optimize the barrier layer thickness to satisfy the total hydrocarbon fuel emission constraint for a plastic fuel tank (PFT).

#### INTRODUCTION

There is new government legislation that imposes low levels of evaporation emission for automotive fuel tanks. As the use of polymeric fuel tanks continues to grow, the automotive industry is required to take steps to conform to these new constraints. Unlike the case of steel, fuel fractions may permeate through polymer materials and cause hydrocarbon emissions. Thus the gas barrier properties for polymer materials are being examined by the automotive industry. The technical solutions found to match the emission requirements include the addition of thin layers with low permeability such as Ethylen-vinyl alcohol (EVOH). The development and production of coextruded multilayer blow molded fuel tanks is now very common. In general, the inner and outer layers of fuel tanks are made of virgin high-density polyethylene (HDPE), whereas the core of the parison is made of HDPE regrind. To ensure good hydrocarbon barrier properties, the EVOH layer must be positioned in the core of the structure and linked to the inner HDPE and

regrind layer by using an adhesive tie layers. These thin layers are made of linear low-density polyethylene (LLDPE), which provide the required adhesion between EVOH and HDPE [1]. With these material layers, the low permeability criterion is achieved over most of the fuel tank surface. Nevertheless, the performance of multilayer container may dramatically deteriorate if the barrier layer is disrupted or if its thickness is reduced by inappropriate processing either at the co-extrusion or at the pinch-off welding stage [2]. In view of the importance of barrier properties, it is surprising that there is no systematic studies on the quality optimization of the co-extruded blow molded fuel tanks.

The objective of this study is to propose a new methodology to better understand the barrier properties of automotive fuel tanks. The mathematical diffusion model based on Fick's second law is taken to predict the hydrocarbon diffusion through a multilayer polymeric wall [3]. The model is based on a continuum approach at steady-state permeation regime. The pinch-off zone is potentially a weak barrier region, thus a particular method has been developed to automatically detect this area at the end of extrusion blow molding process. Hence fore, a special treatment has been adopted to the diffusion model to evaluate adequately the hydrocarbon permeation through this specific area. Finally, Fuel emissions test for blow molded automotive fuel tank is analyzed via simulation and the sensitive analysis is performed to show the impact of barrier layer percentage on hydrocarbon emission.

#### THEORITICAL APPROACH

#### PROBLEM STATEMENT

The selection of polymeric materials for fuel tanks with stringent design specifications, related on their solution and transport behavior, requires the considerations of several factors [3]. The rate of fuel vapor exchanged between a gas tank and the environment depends on the integrity of the seal and closure i.e. the pinch-off zone, and the permeability of polymeric material itself [4].

#### THE PERMEATION MECHANISM

Permeation is the rate at which fuel fraction in gaseous phase passes through the tank shell, the welding areas and piping accessories. Liquid phase transport is not possible through polymeric material since it is not a porous structure [4]. It does not involve leakage, which is caused by holes or cracks in the material. In this study, only the shell tank permeation will be taken into account.

There are detailed descriptions of transport and permeability in polymeric multilayer membrane available in literature [5-7]. A brief summary of the relevant details is summarized below:

1. The permeant i.e. gas or vapor fuel, is absorbed through the wall surface and its solubility respects Henry's law i.e. the amount of dissolved gas or vapor per volume of polymer (C) is proportional to the applied gas or vapor pressure (p)

$$C = S \cdot p \tag{1}$$

where S is Henry's solubility coefficient (the solubility coefficient of permeant in the polymer).

2. The permeant diffuses through the polymer wall to the opposite surface according to Fick's law i.e. the mass transportation through the polymer (dm/dt) per unit area (A) and unit time is proportional to the concentration gradient of the permeant, [8-9].

$$\frac{1}{A}\frac{dm}{dt} = J = -D\frac{\partial C}{\partial x} \tag{2}$$

where D is Fick's diffusion coefficient.

#### MATHEMATICAL MODEL

Given the inherent low diffusion coefficient of plastics and the special geometrical configuration of the blow molded fuel tanks i.e. relatively thin compared with their surface area, the diffusion is usually assumed to be an unsteady one-dimensional problem along the part thickness [4]. The one-dimensional diffusion equation described by Fick's second law equation (2) for constant diffusivity D is given by the following expression [3]:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \tag{3}$$

where C is the liquid concentration in the polymer matrix at a time, t, and distance, x, with the following initial and boundary conditions:

$$t = 0$$
  $0 < x < h$   $C = 0$  (4)

$$t > 0 \qquad x = 0 \qquad C = C. \tag{5}$$

$$t > 0 \qquad x = h \qquad C = 0 \tag{6}$$

The measurement of surface concentrations is not always a simple task. By using Henry's law (equation 1), the flux can be expressed in terms of vapor pressure, p. Hence, the set of partial differential equations (3)-(6) will be transformed as follows, [10-11]:

$$S\frac{\partial p}{\partial t} = P\frac{\partial^2 p}{\partial x^2} \tag{7}$$

$$t = 0$$
  $0 < x < h$   $p = 0$  (8)

$$t > 0 x = 0 p = p_s (9)$$

$$t > 0 \qquad x = h \qquad p = 0 \tag{10}$$

where  $p_s$  is the fuel saturated vapor pressure and P the permeability coefficient of material which is given by following equation:

$$P = S \cdot D \tag{11}$$

Thus, it is possible to have a low permeability material when either D or S is small.

#### Steady-state permeation

From an experimental point of view and according to several authors [12-13], a steady-state regime can be reached after several weeks. As we expect to use the fuel tank during an approximate period of ten years, the analysis of the fuel diffusion based on a steady-state regime seems to be reasonable to provide a reliable prediction of the rate of fuel, which may travel across the gas tank wall.

When the steady-state diffusion has been reached, we obtain from equation (7) the following expression:

$$D \cdot S \cdot \frac{\partial^2 p}{\partial x^2} = 0 \tag{12}$$

By integrating equation (12) across the local thickness of gas tank h, and evaluating between two saturated vapor pressures, assuming D and S to be constant and independent of C, yields:

$$J \cdot h = -D \cdot S \cdot (p_1 - p_2) \tag{13}$$

By substituting the above expression in equation (2), the total amount that has permeated through a wall of polymeric membrane area A in time t:

$$M(t) = -\frac{D \cdot S \cdot (p_1 - p_2) \cdot A \cdot t}{h} \tag{14}$$

#### Permeation through multilayer polymeric wall

A continuum approach is used to model fuel permeation through an *n* layer polymeric wall at steady-state regime. Traditionally, the problem of determining flux through a multilayer film is solved assuming continuity in partial vapor pressure at the polymer-polymer interface [7].

A piecewise continuity illustration is presented in Figure 1 for a multilayer polymer with steady, linear vapor pressure profiles in each layer (dashed line). As depicted, alternating polymer j and j+1 constitute a multilayer film of total thickness h. The vertical solid lines represent the layer-layer boundaries and  $h_1,\ h_2,\ \dots,h_n$  represent the thickness of each layer between the solid lines (the z's in the region  $0 < z < z_n$  indicate the coordinate positions of the solid lines).  $D_i$  and  $S_i$  describe the diffusivity and the solubility of the permeant in the polymer layer i respectively. The permeant profile in the polymer layers is obtained by solving the following boundary value problem.

$$D_i \cdot S_i \cdot \frac{\partial^2 p_i}{\partial z^2} = 0$$
 where i = 1,2,3, ,n (15)

with boundary conditions:

$$\begin{cases} p_j = p_{j+1} \\ S_j D_j \frac{\partial p_j}{\partial z} = S_{j+1} D_{j+1} \frac{\partial p_{j+1}}{\partial z} \end{cases}$$
 (16)

at  $z = z_i$  where j = 1,2,3, ...,n-1 and

$$p_1 = p_{\alpha} \qquad \text{at } z = 0 \tag{17}$$

$$p_n = p_\beta \qquad \text{at } z = z_n \tag{18}$$

Thus, the continuity in permeant flux and vapor pressure are specified at every solid line (figure 1). In the case of a constant diffusivity  $D_j$  and solubility  $S_j$ , the permeant vapor pressure profile in the layer i at steady-state regime is written as:

$$p_i(z) = a_i z + b_i$$
 where i = 1, 2, ..., n (19)

where  $a_i$  and  $b_j$  are constants obtained by applying the boundary conditions (16). Starting from equation (19), the permeant flux  $J_i$  throughout layer i is described by the equation:

$$J_i = \frac{p_i(z_i) - p_i(z_{i-1})}{\frac{h_i}{D_i}}$$
 where i = 1, 2, ..., n (20)

After the mathematical manipulation of the system of equations (20), the permeant flux throughout the layer i is given by the two boundary values of vapor pressure (17) and (18):

$$J_{i} = \frac{p_{\alpha} - p_{\beta}}{\sum_{j=1}^{n} \frac{h_{j}}{S_{j} \cdot D_{j}}}$$
 where i = 1,2,3, ,n (21)

On the other hand, according to steady-state regime, the permeant flux J which goes across the multilayer polymer is equal to the flux across each polymer layer i. Hence,

$$J = J_i \tag{22}$$

When the solubility and diffusivity of each layer are given and once the thickness of each layer is known, then the flux across the multilayer polymer in steady-state regime is directly obtained by using equation 22.

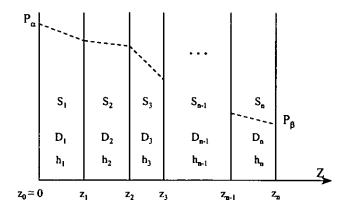


Figure 1: Schematic of a multilayer film with n layer. Steady-state continuous saturated pressure profiles.

#### FINITE ELEMENT IMPLEMENTATION

For thin geometry container such as co-extruded fuel tanks, the vapor pressure gradient of the in-plane direction is much smaller compared to the thickness direction. It can also be assumed that the saturated vapor pressure field is independent of the in-plane x and y coordinates at a local area. Equation (22) is then directly applied within each finite element mesh of the co-extruded fuel tank shell.

Therefore, the fuel mass rate (FMR) transported through the shell element e with area  $A^e$  is then given by:

$$\frac{dm}{dt}\Big|^{e} = \int_{A^{e}} J^{e} dS = \int_{A^{e}} \left( \frac{\left( p_{\alpha} - p_{\beta} \right)}{\sum_{j=1}^{n} \frac{h_{j}^{e}}{S_{j} \cdot D_{j}}} \right) dS$$
 (23)

where  $h_j^e$  represents the thickness of layer i for current finite element e. The total FMR transported through the entire gas tank wall is obtained by adding the elementary flux rate contribution of each finite element:

$$\dot{m}_{Total} = \sum_{e=1}^{Nelem} \int_{A^e} J^e dS = \sum_{e=1}^{Nelem} \left( \int_{A^e} \left( \frac{p_\alpha - p_\beta}{\sum_{j=1}^n \frac{h_j}{S_j \cdot D_j}} \right) dS \right)$$
(24)

The total mass transported through the gas tank wall thickness per day is obtained directly from the last equation:

$$m_{Total} = \int_{0}^{T=1Day} \left( \sum_{e=1}^{Nelem} \int_{A^e} J^e dS \right) dt = \int_{0}^{T=1Day} \sum_{e=1}^{Nelem} \left( \int_{A^e} \frac{p_\alpha - p_\beta}{\sum_{j=1}^n \frac{h_j^e}{S_j \cdot D_j}} \right) dS dt$$
 (25)

When the following quantities  $p_{\alpha}$ ,  $p_{\beta}$ ,  $h_{j}$ ,  $S_{i}$  and  $D_{j}$  are constant, the equation for the daily mass transported through the shell gas tank wall is reduced to:

$$m_{Total} = \int_{0}^{T=1Day} \sum_{e=1}^{Nelem} \left( \frac{\left( p_{\alpha} - p_{\beta} \right) \cdot A_{e}}{\sum_{j=1}^{n} \frac{h_{j}^{e}}{S_{j} \cdot D_{j}}} \right) dt$$
 (26)

where Ae is the area of finite element e.

It is important to notice that the total thickness  $h^e$  and the layer thickness  $h^e$  distributions are obtained from the BlowView software technology developed at National Research Council of Canada (NRC) [14-16]. The software simulates the whole process, i.e. the parison extrusion taking into account the swell phenomenon, the parison stretching at the bottom using spreading pins, the mold clamping and the parison inflation.

#### Pinch-off zone treatment

According to the experimental observation of several authors [1-2], [13] and [17], the pinch-off zone of fuel tanks revealed areas where the EVOH barrier layer became too thin or even disappeared completely. A special treatment of the permeation on this zone is required. For the pinch-off zone, we assume that there is no presence of a barrier layer for the finite element mesh corresponding to this zone. An automatic procedure has been developed to detect the length and the width of this zone.

Thus, the expression for total mass rate transported through the area of the pinch-off finite element is evaluated by removing the barrier layer property in equation 26. In the case where  $p_\alpha$  and  $p_\beta$  are assumed to be constant, the total mass rate transported through the area of the pinch-off finite element is directly obtained from (26) and is given by the following expression:

$$m_{Total}(t) = \sum_{e=1}^{Nelem} \left( \frac{\left(p_{\alpha}^{e} - p_{\beta}^{e}\right) \cdot A_{e}}{\sum_{j=1}^{n} \frac{h_{j}^{e}}{S_{j} \cdot D_{j}}} \right) \cdot t \quad \text{with } j \neq j_{\text{EVOH}}$$
 (27)

#### VALIDATION CASE AND SENSITIVITY ANALYSIS

#### **TEST VALIDATION**

An important contribution of this work is to establish a procedure to provide a more rigorous comparison of analytical and finite-element based computational models. The driving principle is that numerical data should be validated relative to the real-world results from a benchmark example. For this purpose, an three-layer polymeric plane wall with total thickness h and lateral area A, exposed to a permeant at pressure  $p_\alpha$  on one side and at lower pressure  $p_\beta$  on the other side as shown in Figure (2) is considered.

Throughout this example, the permeability of HDPE is normalized as  $P_{\text{HDPE}}$  =1. According to literature [1-2], [17], the permeability of EVOH is always estimated at least to be 1000 to 10000 less than of the HDPE. The thickness h=1 m and the lateral area A=1  $\text{m}^2$ . The volume fraction of the EVOH layer is equal to X % and the volume fraction of each HDPE layer is then equal to 50-X/2% on each side. The high and low permeant pressure applied at each side of the laminate are fixed to  $p_\alpha=1$  and  $p_\beta=0$  respectively.

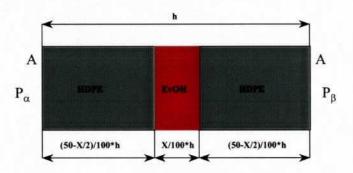


Figure 2: Three-layer laminate of HDPE wall subject to two different fuel pressure at each lateral surface side A.

Starting from equation (21), the FMR going across the three-layer polymeric plane wall is given by:

$$\frac{1}{A}\frac{dm}{dt} = \frac{p_{\alpha} - p_{\beta}}{\frac{h_{HDPE}^{1}}{P_{HDPE}} + \frac{h_{EVOH}}{P_{EVOH}} + \frac{h_{HDPE}^{2}}{P_{HDPE}}}$$
(28)

where

$$h_{HDPE}^1 = h_{HDPE}^2 = \frac{\left(50 - \frac{X}{2}\right)}{100} \cdot h$$
 (29)

$$h_{EVOH}^1 = \frac{X}{100} \cdot h \tag{30}$$

Substituting (29) and (30) in (28), we obtain the analytical expression for FMR:

$$\frac{dm}{dt} = \frac{A \cdot (p_{\alpha} - p_{\beta})}{\frac{h}{100} \cdot \left(\frac{\left(50 - \frac{X}{2}\right)}{P_{HDPE}} + \frac{X}{P_{EVOH}} + \frac{\left(50 - \frac{X}{2}\right)}{P_{HDPE}}\right)}$$
(31)

The evolutions of FMR through the multilayer polymeric wall based on volume fraction of the barrier layer are illustrated in Figure 3. The analytical results are based upon equation (31) and the numerical predictions are obtained from equation (24) using 30 triangular elements. To demonstrate how the volume fraction and the low permeability of barrier layer affect the FMR, three prescribed values of EVOH permeability are used in calculation  $P_{\text{EVOH}} = 10^{-3}$ ,  $5 \times 10^{-4}$  and  $1 \times 10^{-4}$ .

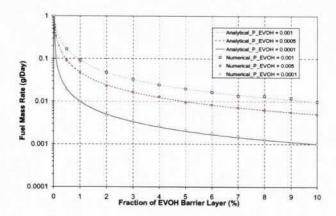


Figure 3: Fuel mass rate through the three-layer laminate of HDPE ( $P_{HDPE}$ =1) containing a barrier layer with different permeability coefficients ( $P_{EVOH} = 10^{-3}$ ,  $5x10^{-4}$  and,  $5x10^{-4}$  respectively).

As we can see, the increase in barrier layer volume fraction leads to a significant reduction of the FMR (permeation). One can notice that the FMR can be reduced by approximately 100 and 1000 times by using 10% of EVOH depending on its permeability. Moreover, a slight variation of barrier layer in the range of 1-3 % has a significant impact on the FMR. On the other hand, the effectiveness of improved barrier resistance decreases as the barrier thickness increase beyond 5 %. These observations are valid for both analytical and numerical results. According to this Figure, numerical predictions are very close to analytical results, showing a good finite element implementation.

# BARRIER LAYER OPTIMIZATION FOR PLASTIC FUEL TANK

Since the permeation model has been validated adequately, this model has been implemented into the BlowDesign optimization software of the NRC. The software manipulates the design variables or processing parameters such as die gap opening profiles and flowrate subject to process constraints such as extrusion time, parison length and finally the permeation rate per day. At each optimization iteration, BlowDesign modifies the design variables to minimize the objective function, which is defined as the PFT thickness standard deviation

subject to a target thickness value. In the plastic fuel tank industry, it is well accepted that the PFT minimum target thickness value should be around 3.0 mm.

So at each time the processing parameters are updated, the thickness distribution on the PFT will be modified accordingly. For a specified thickness distribution, a gradient-based algorithm is used to update the % thickness barrier layer to satisfy a given permeation constraint such as 10 mg/day. This procedure assumes that the % thickness barrier layer will be modified slightly and will not affect the overall thickness distribution predicted by the CNRC software technology.

To illustrate the optimization procedure, a PFT tank of Kautex Company will be used. The initial design of the PFT is composed of 6 layers of polymeric materials as illustrated in Table I. The initial barrier layer percentage has been decreased intentionally in order to let the optimization software adjusting it over the optimization iteration. At each iteration, the layer barrier percentage is modified to satisfy the permeation constraint of 10 mg of permeant per day. Figure 4 shows typical finite element meshes used to model the parison extrusion, the parison inflation and finally the deflashing of the part.

Table 1. Percentage, diffusion and permeability coefficients of each polymeric material used in PFT.

| PFT Layers of<br>Initial design | Layer<br>Percentage<br>[%] | Diffusion<br>Coefficient<br>[m²/s] | Solubility<br>Coefficient<br>[g/g] |
|---------------------------------|----------------------------|------------------------------------|------------------------------------|
| Outer HDPE                      | 24.7                       | 5.5e-12                            | 6.8e-2                             |
| Outer LLDPE adhesive            | 2.5                        | 8.2e-12                            | 1.49e-1                            |
| <b>EVOH Barrier</b>             | 0.3                        | 5.0e-13                            | 5.0e-4                             |
| Inner LLDPE adhesive            | 2.5                        | 8.2e-12                            | 1.49e-1                            |
| Inner HDPE                      | 50.0                       | 5.5e-12                            | 6.8e-2                             |
| Outer HDPE                      | 20.0                       | 5.5e-12                            | 6.8e-2                             |



Figure 4. Typical finite element meshes used for the simulation of the extrusion blow molding.

Figure 5 illustrates the thickness distribution and the hydrocarbon emission flux per unit area [mg/day/m²] variations for the initial design. Knowing the total surface area of the tank shell and the pinch-off zone, the total permeation can be evaluated. One can notice that the initial design does not satisfy the permeation constraint since a total of 90.8 mg per day of hydrocarbon is emitted when 0.3% of EVOH is used. It can also be observed that where the PFT thickness is small, the permeation is high and vice-versa, which is consistent in physical terms.

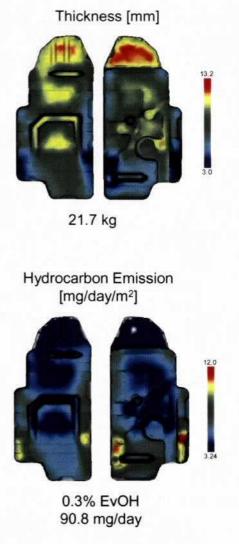


Figure 5. Thickness and hydrocarbon emission flux distributions for the initial design.

During the optimization process, the %EVOH is manipulated in order to satisfy the daily permeation constraint. In Figure 6, the gradient-based algorithm has found after 6 optimization iterations, that 3.81% of EVOH should be used to satisfy the daily permeation constraint of 10 mg/day for the given thickness distribution. The part weight has been decreased from 21.7 kg down to 17.6 kg and satisfies the thickness constraint of 3 mm.

In the industry, a 3 to 5% of EVOH is used to satisfy the LEV regulation (Low Emission Vehicle), which is close to the predicted value (3.81%EVOH).



Figure 6. Optimized thickness and hydrocarbon emission flux distributions for the final design after 6 optimization iteration.

#### CONCLUSION

Numerical simulation model, based on finite element method for the prediction of fuel permeation through multilayer polymeric material under steady-state regime was presented in this paper. The special treatment of the diffusion in the pinch-off zone was also considered.

The accuracy of the proposed model was verified by comparing the predicted results to the analytical results for three-layer laminates of HDPE. Very good agreement has been found between analytical and numerical results.

The permeability model has been used to optimize the barrier layer percentage. An optimization software (BlowDesign) manipulates the %EVOH to satisfy the daily permeability constraint. For the gas tank studied,

we found that 3.8% EVOH is necessary to obtain a 10 mg daily emission, which is close to the %EVOH presently used in the industry.

For the fuel gas tank studied in this paper, the barrier layer percentage is found to be the primary factor to reduce the evaporation fuel emission.

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