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Analysis of the effect of perforation on the permeability of biodegradable non-barrier films

Antonis Mistriotis *, Anastasios Giannoulis, Dimitris Giannopoulos, Dimitris Briassoulis

Agricultural University of Athens, Depart of Natural Resources and Agricultural Engineering, Iera Odos 75, Athens, Greece (amistr@aua.gr)

Abstract

Perforated plastic films are used in Equilibrium Modified Atmosphere Packaging (EMAP) of fresh produce such as fruits and vegetables. The most common material used in such applications is the oriented polypropylene (OPP), which has low permeability with respect to relevant gases, namely water vapor (WV), CO_2, and O_2. Therefore, the synthesis of the in-package atmosphere is regulated only by the size and number of perforated holes. The replacement of the OPP films with biodegradable ones made of polylactic acid (PLA) or starch based polymers for environmental reasons results into difficulties with respect to designing the EMAP system, since these films are more permeable to the relevant gases and in particular to WV. As a result, the effect of micro-perforation is influenced by the permeability of the film. In the present work, the dependence of the gas flux through perforation on the permeability of the film for the same gas was investigated by experimental and numerical methods. It was shown that the effect of perforation decreases as the permeability of the film increases. The diffusive gas flux through perforation becomes independent of the film permeability, if it is about 100 times smaller than the diffusivity of the studied gas in air.

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Keywords: EMAP; biodegradable packaging; micro-perforation

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1. Introduction

The development of thin transparent barrier films offered new possibilities to the food packaging industry. Packing food in a Modified Atmosphere Packaging (MAP) of low concentration of oxygen retards or prevents the oxidation process, which is responsible for the deterioration of the taste and flavor [1]. However, such packaging techniques cannot be applied to fresh produce, since fruits and vegetables are living objects interacting with the surrounding atmosphere through respiration and transpiration. Equilibrium Modified Atmosphere Packaging (EMAP) is a method for prolonging the shelf life of fresh

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* Corresponding author. Tel.: +30-210-5294022; fax: +30-210-5294023.
E-mail address: amistr@aua.gr.

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produce (i.e. fruits and vegetables) by optimizing the in-package equilibrium atmosphere [1]. This is achieved by modifying the permeability of the packaging film using micro-perforation, in order to optimally regulate the equilibrium concentrations of O_2 and CO_2. The in-package relative humidity (RH) should also be regulated, since it is responsible either for the excessive weight loss or for incidences of fungal spoilage.

Several attempts have been made to model gas transfer through a perforated film for optimizing EMAP. These models focus on the gas transfer through the perforation holes, while the film itself is considered impermeable (barrier film) [2, 3]. Such modeling approaches are based on Fick's law of diffusion [2] or Stefan–Maxwell law [4].The simplest model assumes Fick's diffusion in a cylindrical pore filled with stagnant air simulating the hole. However, an end correction term for the diffusive path length must be introduced to compensate for end effects at the hole mouths [2]. Such phenomenological corrections are not needed when 3D computer simulations are applied for modeling diffusion through a hole. For this reason, a 3D numerical approach was used in the present work.

Environmental concerns recently press towards a change in food packaging materials from conventional oil-based plastics to biodegradable polymers made of sustainable recourses. Various bio-based polymers, such as PLA, or starch based plastics have been tested as packaging materials despite their current high cost. Such new materials have different gas permeability properties than conventional plastics [5]. In particular, their water vapor transmission rate (WVTR) is much higher than the conventional packaging films made of oriented polypropylene (OPP) [5]. When such non-barrier films are used in Equilibrium Modified Atmosphere Packaging (EMAP), the effect of perforation is influenced by the permeability of the film.

The present work is aiming at estimating the excess WVTR due to perforation as a function of the water vapor permeability of the film. The combined effect of perforation and permeability of the film on the synthesis of the EMA is also investigated.

2. Material & Methods

The diffusion of a gas is described by Fick's law which relates the diffusive flux to the concentration field:

$$J = D \ \nabla c \tag{1}$$

Where J (mol m^{-2} s^{-1}) is the flux density (i.e. gas diffusion) through the film, D (m^2 s^{-1}) is the diffusion coefficient (or diffusivity), and ∇c (□mol□ m^{-4}) is the gas concentration gradient, c being the concentration for ideal mixtures (mol m^{-3}) [6].

In the case of diffusion through a permeable material, its permeability P (g m^{-1} s^{-1}) is defined as:

$$F = P \cdot A \cdot \nabla p \tag{2}$$

where F (g s^{-1}) is the mass flux, □ ∇p is the mass fraction gradient (percent), and A (m^2) is the area of the film. The mass fraction gradient can also be expressed by the partial pressure difference of the gas (Pa). As a result, permeability is also reported in g m^{-1} s^{-1} Pa^{-1}. Alternatively, if volume flux is considered instead of the mass flux, permeability, P (m^2 s^{-1} Pa^{-1}), refers to the amount of gas, by volume, which penetrates unit thickness and area of the material per unit time, under constant temperature and unit partial pressure difference when permeation is stable [7]. If partial pressure difference is given as mass fraction difference, permeability is expressed in the same units as diffusivity (m^2 s^{-1}).

When a thin film is considered, its gas transfer characteristics are described by the permeance, PR (g m^{-2} s^{-1}) or (g m^{-2} s^{-1} Pa^{-1}):

$$F = PR \cdot A \cdot \Delta p \tag{3}$$

The transmission rate for a specific partial pressure difference Δp (Pa), TR, (g m^{-2} s^{-1}) is then defined as:

$$TR = PR \cdot \Delta p \tag{4}$$

In the specific case where the gas is water vapour (WV), water vapour transmission rate WVTR [7,8] refers to the amount of water vapour, by volume (or mass) that penetrates one square meter of a specimen of a specified thickness per unit time, under specified temperature and relative humidity and water vapour pressure difference. The water vapour transmission (WVTR) (g m^{-2} s^{-1}) is then defined as:

$$F_V = WVTR \cdot A \tag{5}$$

where FV (g s^{-1}) is the water vapour mass flux.

The WVTR of a film was gravimetrically measured by using the ASTM-E96/E96-05 Standard [9]. The Water Method described in this Standard was implemented in the current work. Petri dishes of 80 mm diameter were used as test dishes (Fig. 1). In each test dish, 20 ml of distilled water were added leaving a distance of approximately 10 mm between the water surface and the film. The film samples were sealed to the dish mouth by a water resistant sealant. The dishes were placed in a controlled environment room with temperature 23±0.5°C and RH equal to 50±5% and their weight was measured once per day by a balance with accuracy of 0.01 g. The decrease of the weight of each dish was found linear with time, and the regression slope was used for estimating the WV flux through the film.

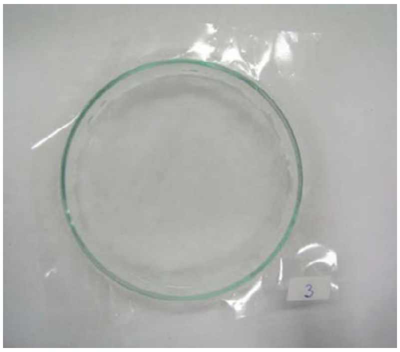


Fig 1. Test dish used for measuring *WVTR* by the gravimetric water method of ASTM-E96/E96-05 Standard

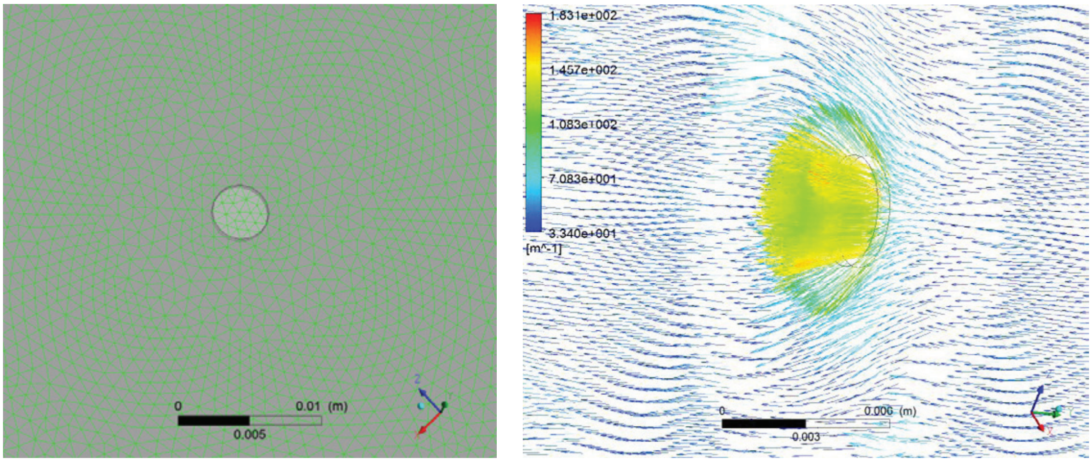
Five different films of different WVP have been considered: OPP (30μm), Cellophane (25μm), PLA (30μm), Mater-Bi (35μm), Mater-Bi (15μm). These tested films were commercial materials with additional laminated layers influencing their permeability properties. Therefore, the *WVTR* values presented in this work may deviate from those reported by other authors [5,10]. For each film, *WVTR* was measured for the non-perforated film and a film with a single 4 mm diameter circular hole. The difference in *WVTR* between the perforated films and the corresponding non-perforated ones was defined as the effect of perforation. Additionally the water mass loss for an uncovered dish was measured to simulate the case of film of the same permeability with air, as control. Three samples for each perforated and non-perforated film were tested.

The same phenomena were also analysed by numerical simulations. It was assumed that the mass transfer through the film and the hole is due to diffusion, while convective mass transfer is negligible. The above experiments were modelled by 3-D simulations. A mixture of WV and air was considered in the numerically solved domain, and the diffusion of WV in air was simulated.

The numerical model simulated a test dish similar to those used in the experiments. The 3D model container had a diameter of 80 mm and its depth was 10mm. Its top was closed by a membrane simulating the film of thickness equal to 0.5 mm. In this material, WV diffusivity was assigned smaller than in air, in order to simulate the *WVTR* of the film. In this way, the numerical method allowed for a wide range of different values of permeability to be examined, while the experimental tests were limited to the available materials. For the domain under the membrane and in the hole, the kinematic diffusivity of the WV in air at this temperature was defined equal to 2.5×10^{-5} m^2/s as found in literature [11], while the membrane was modelled as an area of a lower kinematic diffusivity.

The bottom and the cylindrical side wall of the dish were modelled as impermeable surfaces. Fixed mass fraction boundary conditions were applied. The mass fraction of WV at the bottom of the dish was taken equal to 1 simulating the water surface, while at the external surface of the closing membrane the WV concentration was 0. The temperature was considered constant, 23 °C. In the case of a perforated film, a hole was modelled in the centre of the membrane as a cylinder connected to the air-filled domain under the membrane. The diameter of the hole was equal to 4 mm similarly to the experimental setup. The diffusive mass flux was numerically calculated when the membrane is perforated or not for various values of its permeability with respect to WV. Similarly to the experiments the difference in *WVTR* between the perforated films and the corresponding non-perforated ones was calculated.

Commercial software (ANSYS-CFX) was used, implementing the finite volume method [6]. Convergence was reached when residuals were smaller than $1e^{-07}$. The model was discretised by tetrahedral elements. The grid consisted of 522113 elements (103552 nodes), and it was uniformly formulated as shown in Fig. 2a. The numerical solution was checked for convergence with respect to mesh refinement. The region, where the numerical solution was modelled, was divided into two domains, namely one corresponding to the membrane and another one filled with air. The upper boundary plane of the air-filled domain was linked to the membrane's lower boundary plane by an interface surface allowing for a smooth transition from the one domain to the other.



(a) (b)

Fig. 2. a) The grid with tetrahedral elements used in the simulations, b) typical simulation of a perforated film with small WVTR

Fig. 2b presents a typical solution of the diffusion of WV through the hole. The vectors represent the mass fraction gradient of WV. In this example it was demonstrated that when the kinematic diffusivity of the membrane with respect to WV is much smaller than that of WV in air, the diffusion of WV takes place mainly through the hole.

3. Results & Discussion

Table 1 presents the WVTR of the five tested materials reported in the previous section. Two cases were studied: a) when the films were not perforated, and b) with a hole of 4 mm diameter. The difference between the WVTR of the perforated and the non-perforated film describes the WVTR though the hole. The experimental results show a decrease of the effect of perforation as the WVTR through the non-perforated film approaches that of WV in air.

Table 1. TR though a hole of 4mm diameter on various tested films compared to the WVTR of the corresponding non-perforated film when the RH difference is 50%.

Film type	***WVTR*** **(g m^{-2} s^{-1})**	**Hole *TR* (g m^{-2} s^{-1})**
OPP 30μm	0.000006	0.1234
Cellophane 25μm	0.000069	0.1251
PLA 30μm	0.000543	0.0982
Mater-bi 35μm	0.001535	0.0542
Mater-bi 15μm	0.002252	0.0404
No-film	0.026889	0.0000

The effect of the perforation is reduced for the PLA and the starch based films, which are hydrophilic. In the case of cellophane, which is less permeable to water, the effect of the hole is similar to the OPP film, although its WVTR is 10 times higher. A measurable decrease of the TR through the hole is observed for the PLA film, which is 100 times more permeable to WV than OPP (Table 1). The perforation effect is further reduced as the WVTR of the film increases.

The WV diffusion through a membrane was also numerically simulated using the model presented in the previous section. The WV diffusion was simulated in the case of a perforated and a non-perforated membrane. The mass fraction gradient at the hole was numerically calculated for various values of the diffusivity of the membrane, and used for the calculation of the WV flux through the hole. In order to obtain results which are independent of the film thickness and the hole diameter, both numerical and experimental data were normalised as following. The measured values of the hole transmission rate (y-axis) were normalised with respect to the value corresponding to a barrier film (i.e. OPP). The WVTR of the films (x-axis) were normalised with respect to the WVTR corresponding to an uncovered test dish. Similarly, the numerical values were also normalised. The mean values of the WV flux at the hole (y-axis) were normalised with respect to the value corresponding to a perfect barrier film. The diffusivity of the membrane (x-axis) was normalised with respect to the diffusivity of WV in air.

Fig. 3 presents both the experimental (square markers) and the numerical (continuous line) results. A similar behaviour was observed in both the experimental measurements and the numerical model. It is shown that the maximum effect of the perforation appears when the diffusivity of a gas through a film is at least 100 times smaller than in air (red dash line). This result is also confirmed numerically, as it is shown by the continuous curve presenting the current numerical results. The relationship between the normalised TR of the hole and the normalised WVTR of the film was found to be logarithmic in the range of normalised WVTR between 0.01 and 1. More specifically, the following phenomenological equation was determined:

$$TR_H = 0.176 \ln(WVTR) \tag{6}$$

which is presented in Fig. 3 by the green straight line. The diffusive gas flow through holes of various diameters was numerically analysed. In this way, equation (5) was proved to be independent of the hole diameter. The observed decrease of the perforation effect has to be taken into account when an EMAP perforation pattern is designed for non-barrier films.

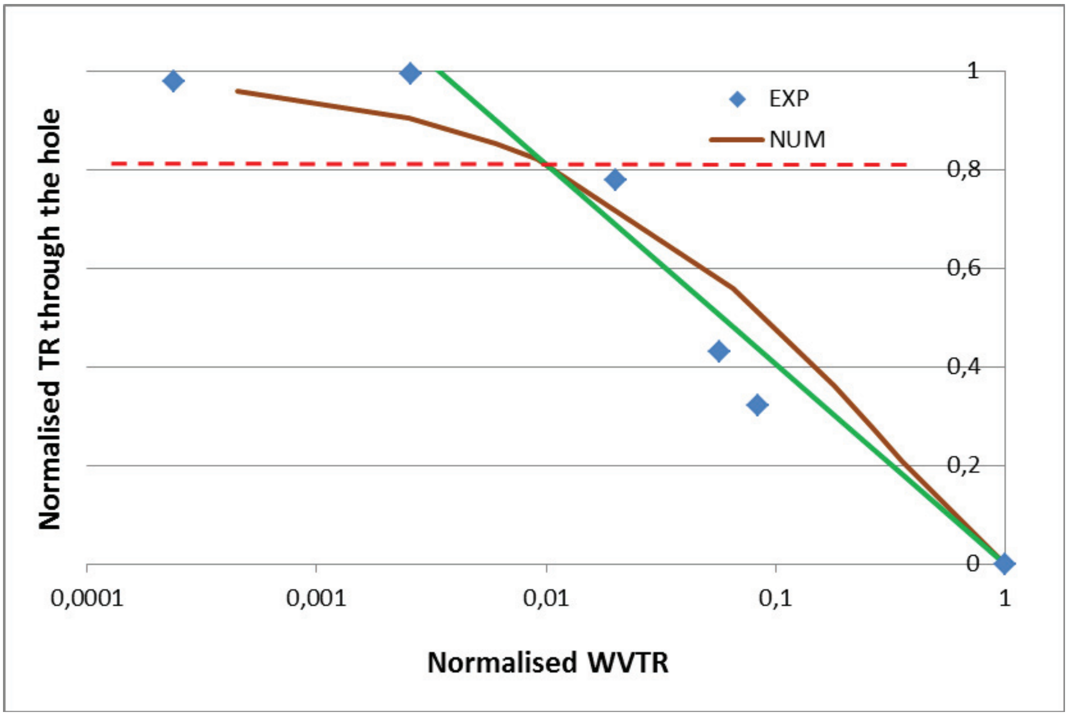


Fig. 3. Normalised perforation effect as a function of the normalised WVTR of the film

The high WVTR of several biodegradable materials used for the production of packaging films is a result of their hydrophilic nature. On the other hand, the same biodegradable films behave as barriers with respect to CO2 and O2 with similar transmission rates to OPP films [5]. This versatility of the environmentally friendly materials offers new possibilities in optimising perforation patterns for EMAP systems used in packaging of fresh horticultural products.

4. Conclusion

Several biodegradable polymers, such as PLA and starch-based polymers, used for the production of packaging films have a WVTR much higher than the conventional OPP films. Therefore, when such films are used in EMAP applications, the design of the perforation pattern has to take into account the permeability of the film. It was shown that the effect of the perforation decreases as the TR of the film increases. A logarithmic relationship was found between the normalized hole TR and the normalized WVTR of the film in the region of WVTR values (0.01-1), where such a decrease is significant.

The current results are expected to support the design optimization of EMAP systems based on biodegradable films. The high WVTR of such films in combination with their low permeability with respect to other gases, which are also important for food packaging such as CO2 and O2, offers new possibilities in the packaging of fresh fruits and vegetables. When such materials are used, perforation mainly regulates the respiration activity of the packed produce, while WP transfer takes place mainly through the film itself. This allows the design of targeted packaging conditions that meet the requirements of specific fresh products.

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