

# **Mathematical Modelling of Gas Exchanges in Film-wrapped Cucumbers**

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## Abstract

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Preserving cucumber (*Cucumis sativus* L.) quality by film wrapping is common practice in Sweden. Nevertheless, no theoretical background is available to explain the choice of the films currently used. A better understanding of the phenomena involved in preserving quality by film wrapping of greenhouse cucumbers can be achieved by means of mathematical modelling of the gas exchanges.

An apparatus controlled by a computer was developed to measure gas exchanges in fresh produce. The transpiration and respiration rates evolution of film-wrapped cucumbers was investigated by emulation of the different gas levels present in the packages. A modification of this set-up was used to study the effect of various films on preserving the quality of cucumbers. In further experiments, the influence of surrounding conditions on the respective role of cuticular and stomatal permeability in the gas exchanges was investigated.

It appeared that a reduction of moisture loss was of highest importance in the preservation of cucumber mass. Both respiration and transpiration rates were strongly influenced by the surrounding temperature and relative humidity. The transpiration rate under constant conditions decreased rapidly resulting in severe loss of quality directly after harvest. Film wrapping reduced significantly the effect of surrounding temperature and relative humidity. Moreover the introduction of a modified atmosphere inside the packages reduced both respiration and transpiration rates. However, the appearance of water vapour condensation inside the film favours microbial hazard.

Based on data from the experiments and known physiological and physical phenomena, a mathematical model of the gas exchanges was designed. Two versions were derived to study the gas concentration in a jar corresponding to the experiments, as well as giving a closer insight into what happens between the epidermis and a film. These models take into account many parameters relative to the cucumber such as permeability, degree of opening or distribution of stomata. Furthermore, film properties such as thickness, size of perforation or permeability could also be investigated. Simulations from the implementation of the mathematical model enable theoretical studies of the importance of those parameters when direct measurements are not possible.

*Keywords:* Finite element method, modified atmosphere packaging, partial differential equation, respiration, transpiration, water activity

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*À la mémoire de Joseph,*

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## Appendix

### **Paper I-V**

The present thesis is based on the following papers, which will be referred to by their Roman numerals.

- I. Cazier, J.-B., Nilsson, T., Gekas, V. 2000. Monitoring respiration and transpiration of fresh horticultural produce. *Journal of Food Engineering. Submitted*
- II. Cazier, J.-B., Nilsson, T., Gekas, V. 2000. Effect of Film Wrapping on Quality of Greenhouse Cucumbers. *Journal of Agricultural Engineering Research. Submitted*
- III. Cazier, J.-B., Gekas V. 2001. Water activity and its prediction: a review. *International Journal of Food Properties. In press*
- IV. Cazier, J.-B., Gekas, V., Nilsson, T. 2000. Epidermis influence on the gas exchanges around a produce. *International Journal of Food Properties. Submitted*
- V. Cazier, J.-B., Gekas, V., Nilsson, T. 2000. Mathematical modelling of water vapour exchange through stomata and microperforated packaging film. *Applied Mathematical Modelling. Submitted*

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## Introduction

Greenhouse cucumbers (*Cucumis sativus* L.) are consumed green and firm, in an unripe state from a horticultural viewpoint. Greenness and firmness are two characteristics strongly affected by the maturation process. The dark green colour on the surface of the cucumber is achieved by the presence of chlorophyll in the epidermis. The chlorophyll breakdown results in immediate degreening (Nobel, 1991). Then, the cucumber surface turns to yellow because of the presence of carotenoids. Firmness is an important parameter of cucumber quality as well since the whole structure is kept by the cell turgor. Transpiration of the fruit, detached from the rest of the plant, will dramatically reduce its water content and therefore firmness (Kader *et al.*, 1985).

Keeping the fresh appearance of cucumbers long after harvest is the permanent challenge imposed by the consumers. The absence of any processing limits strongly the range of treatments available to achieve this goal.

The classic approach makes use of Arrhenius law to delay ripening by decreasing the storage temperature (Wills *et al.*, 1989). However, such a reduction can only slow down the metabolic processes and the temperature drop is limited to a minimal value imposed by the produce under consideration. Cucumbers are of subtropical origin and are therefore chilling sensitive. This disallows any consistent storage under 12 °C and calls for another method to keep freshness. Kaynas and Özelkök (1999) tested the use of Semperfresh to preserve cucumber quality. However, this method, used for batch, cannot apply to individual retailing of cucumbers.

## History of cucumber distribution in Sweden

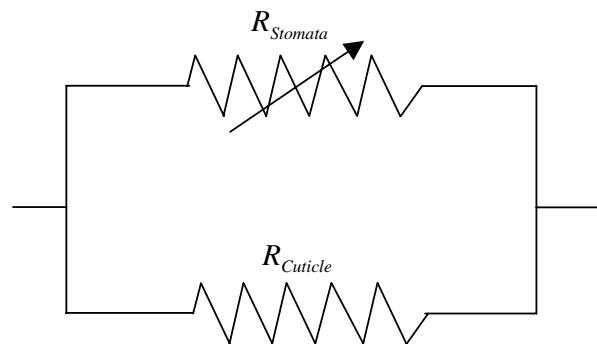
Cucumbers are very popular in Sweden but the production of 20537 tons by 285 companies, for a total surface of 60 ha in 1996 (Statistics Sweden, 2000) cannot cover all the demand. Moreover, it is not possible to grow cucumbers all year round in Sweden and some delay exists between harvest and retailing. The subtropical origin of the cucumber plant makes it very sensible to both temperature and surrounding relative humidity (RH). Without any treatment, the transpiration and respiration rates are high, leading to a very short shelf life for the cucumber.

Polymeric films started to be used to add a barrier against water loss. Firmness and mass were effectively preserved. Furthermore, polyethylene films appeared to have the positive side effect of reducing the degreening as well. However, films were so resistant to water vapour that water drops inside the package were observed and quickly raised some concerns about the food safety (Phillips, 1996). To avoid the condensation, two circular holes with a diameter of 6 mm, were placed in the film at one third from both ends. With the right choice of film, water loss was reduced to the same extent as when the non-perforated films were used. No condensation appeared in the package, avoiding spoilage and food hazard. However, around the holes the cucumbers presented a yellow and dried

area. On the opposite, far away from the holes condensation was often observed. A better repartition of smaller holes would provide the answer to the new problems.

The next step was indeed to use microperforations to spread the holes, keeping the overall permeability of the film similar. In practice, this method proved very efficient to preserve quality by means of both turgor and colour. However, it lacks a real theoretical background to understand why and how such results are provided. Such an understanding could provide the knowledge to improve future films.

### Gas exchanges through fruit epidermis



$$S_{Epidermis} \frac{1}{R_{Epidermis}} = S_{Cuticle} \frac{1}{R_{Cuticle}} + S_{Stomata} \frac{1}{R_{Stomata}}$$

*Fig. 1.* Resistance of the epidermis is obtained as a weighted harmonic sum of the resistance of the cuticle and the stomata by analogy to electrical circuitry.

The gas exchanges between produce and surrounding air occurs through the epidermis. In addition to the cuticle, cucumbers possess stomata as an alternative pathway to gas exchanges (van Beek, 1985). The presence of stomata provides some control to the photosynthesis (Meidner and Mansfield, 1968). However, their closing limits as well water loss by means of a reduced transpiration rate (Jones, 1998). Modelling of gas exchanges through the epidermis is usually done by analogy with an electrical circuit (*Fig. 1*). Stomata and cuticle are alternative pathways and therefore the overall resistance is given by a harmonic sum of the stomatal and cuticular resistance weighted by their relative surface area (Nobel, 1991). With such model, not including information on the stomatal distribution, no interaction between the openings can be taken into account. This approach can therefore not be used for cucumbers, which present irregular repartition of the stomata (Paper V).



## Modified Atmosphere Packaging

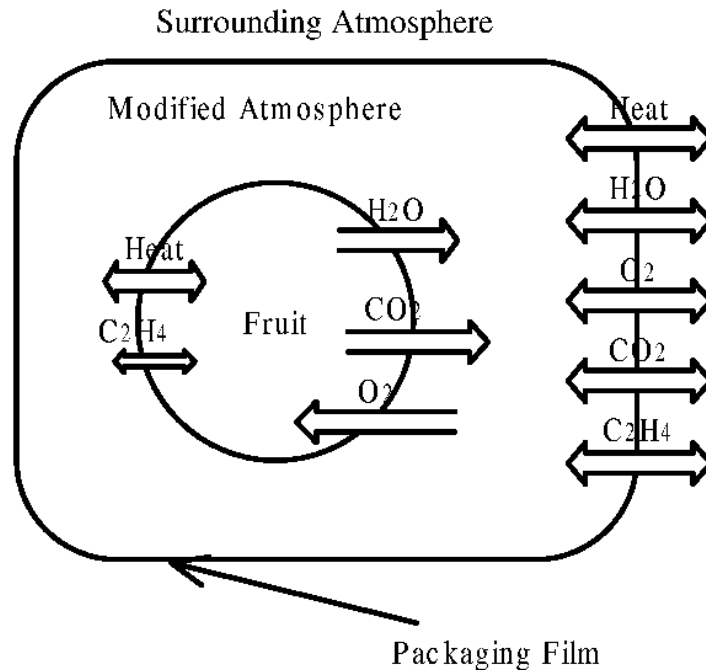


Fig 2. Schematic concept of Modified Atmosphere Packaging.

Preserving the apparent freshness of fruit and vegetables has been the aim of postharvest research. For more than 70 years some fruits have been preserved thanks to storage in a controlled atmosphere (CA) (Kader *et al.*, 1989). CA is obtained by bulk storage of produce in rooms with a carefully monitored composition of the atmosphere. A high carbon dioxide and low oxygen concentrations are usually achieved to slow down the maturation process (Kader *et al.*, 1975). Moreover, a high level of CO<sub>2</sub> reduces the effect of ethylene (Mathooko *et al.*, 1995). The positive effects can also be applied to most of the fruit and vegetables. Robinson *et al.* presented in 1975 a review of storage condition for optimal prolonged shelf life. The limit of CA is the necessity of carefully controlled rooms making the investment costly and restricted to storage. It is still successfully used for shipping of apples from remote countries.

The use of polymeric films to preserve quality of fresh produce has been investigated in more than 30 years (Ben-Yehoshua, 1985). The presence of the package provides a physical protection against mechanical damage, exogenous ethylene and microbial attacks. Furthermore it can reduce the spreading of diseases from contaminated packages to the rest of the surrounding commodities. However, the main advantage of film wrapping lies in the introduction of a Modified Atmosphere Packaging (MAP) where the produce creates its own optimal surrounding conditions by respiration and transpiration (Fig. 2) (Kader *et al.*, 1975). The packaging film permeability properties have to be chosen

according to the produce and the storage conditions. The transpiration of fruit increases the RH in the space between the epidermis and the film. This leads to a lower water vapour pressure deficit over the epidermis inducing a reduced transpiration rate. Similar result is obtained with the production of carbon dioxide that increases the CO<sub>2</sub> concentration. On the opposite, the oxygen consumption leads to a decrease of O<sub>2</sub> concentration in the package. The increased level of carbon dioxide and reduced oxygen concentration slows down the respiration of the produce retarding ripening just as CA. Nevertheless some limits exist in the composition of the modified atmosphere. If the relative humidity in the package is very high, condensation is likely to occur because of, for example, heat production due to respiration. Packages with high level of water activity are prone to microbial hazard (Phillips, 1996). Injuries can occur with levels of carbon dioxide higher than 5% (Watkins, 2000) or oxygen levels lower than 1% (Beaudry, 2000).

Many investigations have been performed to study the effect of MAP (Kader *et al.*, 1989, Church & Parsons, 1995). In addition, models to predict the best film properties required for an optimised shelf life have been presented (Emond *et al.*, 1991, Exama *et al.*, 1993, Christie *et al.*, 1995, Fishman *et al.*, 1995, Hertog *et al.*, 1998). Talasila *et al.* studied the rigid packages (1995) and, in another experiment, compared those with non-rigid packages (Talasila & Cameron, 1997). Renault *et al.* (1994) as well as Fishman *et al.* (1996) investigated the effect of holes in the film. More references on the use of MAP for horticultural produce are available in the recent review by Kader and Watkins (2000). Those models are based on respiration and transpiration rates of the produce and the volume used to produce the modified atmosphere. In the case of film-wrapped cucumber the volume between the epidermis and the film is not known and difficult to estimate. Moreover the heat treatment inflicted to the polymeric film does not provide a smooth surface with a defined thickness. Therefore none of the previous models could be applied and an investigation on the phenomena involved in such packages was required.

## **Objective of the investigation**

This project aims to provide a mathematical model enabling better understanding of the influence of the different parameters involved in the gas exchanges of cucumbers by means of numerical simulation of data impossible to measure.

The following investigations were performed.

### **Paper I**

A set-up was designed to study the effects of surrounding temperature and RH on the postharvest life of cucumbers. Unwrapped cucumbers were placed in jars to simulate the composition inside the package.

## **Paper II**

Further experiments were performed using a similar set-up as in paper I to investigate the difference between various types of low-density polyethylene (LDPE) films. Moreover, mass and colour of the cucumbers were monitored during the experiments.

## **Paper III**

The calculation of the water activity and its prediction were put under scrutiny. Prediction requires approximations and manipulations disallowing the obtention of accurate values.

## **Paper IV**

A mathematical convection-diffusion model was developed to follow the evolution of the concentration of gas into the jar where the cucumber was enclosed (Paper I). A finite element model was used for the implementation. Numerical simulations were used mainly to study the effect of the boundary conditions on the cucumber epidermis.

## **Paper V**

Further investigations on the relative role of the stomata and the cuticle in water vapour loss were performed. A modification of the mathematical model developed in paper IV was applied with more precision to a smaller part of the cucumber epidermis including stomata and a microperforated film.

# **Experiments**

## **Experimental Set-up**

The term cucumber includes different fruits and the properties available in the literature are presented within a wide range of conditions. Moreover the postharvest properties depends strongly on the pre-harvest treatment and origin (Kanellis *et al.*, 1986, 1988, Cabrera *et al.*, 1992, Mattson, 1996). In order to retrieve data useful to design a mathematical model and to understand the phenomena involved, a specific experimental apparatus was designed. Many experimental set-ups have been developed to study the respiration rate (Inaba *et al.*, 1989, Bower *et al.*, 1998, Magnanini *et al.*, 1998). Fewer have been designed to simultaneously monitor the transpiration rate (Shirazi & Cameron, 1993). The apparatus presents data on reaction of cucumber in term of respiration and transpiration to constant temperature and relative humidity (Paper I). Temperature is ensured by placement of the whole apparatus inside a dark climate chamber. Enclosing individually cucumbers in jars simulates the reaction of the produce when, wrapped in a film, it is subjected to various surrounding gas composition. A stream of defined composition is going through the jars to keep

the surrounding constant composition. At sampling time, it is redirected to the sensors for evaluation of the gas composition. Software written on a personal computer monitors the execution of the successive sampling and filters automatically the data before recording. A complete description of the designed apparatus and its software is presented in the annex.

The experiments were performed in two successive parts. First, cucumbers without any wrapping were placed into the system to emulate the composition inside a package (Paper I). The respiration and transpiration rates were monitored as well as the mass loss during the experiment. The second series of experiments tested different LDPE films (Paper II). The pure LDPE films were of different thickness (10 $\mu$ m, 20 $\mu$ m, 30  $\mu$ m and 40  $\mu$ m). The film of 20 $\mu$ m thickness was microperforated. A fifth 22 $\mu$ m thick film was tested with an extra layer of ethylene-acrylic acid (EAA). Measurements similar to those of paper I were performed with other sensors. Moreover the mass and surface colour of the cucumber were recorded regularly during the experiments.

## Results

The first series of experiments presented the expected dependence to temperature and RH of the respiration and transpiration rates. Moreover, an exponential drop with time of the transpiration rate was observed (Paper I). Only Shirazi and Cameron (1993) observed a similar drop but with a much shorter delay. Since all the conditions inside the jar were maintained constant by a flow-through system, only the cucumber could cause the drop. One explanation could be an increase in the resistance of the epidermis to water vapour like observed in cherimoya (Martinez *et al.*, 1993). Stomatal closing could explain a drop in the permeance to water vapour. Another possibility is a drop in the driving force, reached by a decrease in the water activity, or water vapour pressure, inside the cucumber, close to the epidermis.

On the opposite, in cucumbers wrapped in the different films, mass loss was reduced, and the transpiration and respiration rates remained constant (Paper II). Those results confirmed the positive effect of the packaging with the small range of LDPE films investigated. Even the colour was best preserved compared to similar experiment without film conducted by Mattson (1996). However, no significant differences were found between the different films except for the absence of condensation with the microperforated one.

In both case mass lost by the cucumber was mainly, but not only, due to transpiration, underlining the importance to control water loss to preserve quality. Further experiments have been performed with four series of cucumbers placed into two climate chambers in dark and in light under two temperature regimes (Paper V). Cucumbers were placed horizontally without the use of the previously described apparatus to study the effect on epidermis aperture of temperature and lightness.

# Modelling the gas exchanges

## Mathematical model

A mathematical model was developed to understand what is happening when a film is wrapped around a cucumber. The originality of this study lies mainly in the choice of the domain outside of the cucumber epidermis. Cucumber shape is not uniform and heat shrinkage produces an irregular package. Furthermore, stomata are present irregularly on the epidermis providing an alternative pathway to gas exchanges. The well-defined models described in the previous section could therefore not be applied here. Moreover, there is no possibility yet to measure precisely enough the concentration of the different gas concerned in a three-dimensional space (Sandsten *et al.*, 1996). Therefore we built our mathematical model on known physical and physiological phenomena. A convection-diffusion equation (1) in the concentration of the gas concerned was used.

$$\partial_t c = \nabla^T K \nabla c + v c \quad (1)$$

The previous experiments produced not only the phenomena to be taken into account but the order of magnitude of important parameters like the epidermis resistance to water vapour. Therefore a Robin boundary conditions (2) was used to emulate the gas exchange over the epidermis.

$$-K \partial_n c = \alpha (c - c_{in}) \quad (2)$$

In order to validate our concept the experimental set-up was modelled and the gas concentration in the jar numerically simulated (paper IV). Measurements being performed at the top of the jar were successfully confronted to numerical simulations.

In a second step, a theoretical model could focus on a smaller part of the system to investigate parameters like stomata size and density as well as the interaction with the film, its thickness, distance to the epidermis and presence of microperforations (Paper V). In this model only the diffusion was taken into account because the film protected against any external wind. Since the chemical potential is the driving force of exchange over a membrane (Doulia *et al.*, 2000), the cuticle boundary condition follows a Robin type boundary conditions. According to the definition of the chemical potential (Paper III), the derived boundary condition (3) in terms of concentration is not linear (Paper V).

$$-K \partial_n c = \alpha' c \ln \frac{c}{c_{in}} \quad (3)$$

The application of various models should be available through a flexible implementation of the parabolic partial differential equation (1).

## Nomenclature

$\alpha, \alpha'$	Robin type boundary condition parameters
$B_i$	Matrix for boundary condition of first and second type
$B_3^1, B_3^2$	Matrix for the left, resp. right, side of Robin boundary condition
$c, c_{in}, c_0$	Water concentration unknown, in the cucumber, and initial
$\partial_n$	Normal derivative operator
$\partial_t$	Time derivative operator
$g'$	Right-hand side of the mathematical model
$h$	Neumann boundary condition
$\Gamma$	Boundary of the domain $\Omega$
$K$	Diffusion coefficient
$M$	Stiffness matrix
$P$	Discretized space of solution
$\sigma$	Weight on the time scheme
$t$	Current time step
$\tau$	Time step for the discretization of the time derivative
$u$	Test function
$v$	Convection coefficient
$\Omega$	Domain

## Implementation

On one hand, writing a program to solve a parabolic differential equation (PDE) is relatively easy and has no limitations. However, such a program will be error prone and the inclusion of all the pre- and post-processing tools required would be very time consuming. On the other hand, investing in black-box software provides most of the required tools for defining the parameters, creating the grid, and analysing the very large data output. However, the cost, in terms of financial and time investments, is very important before all aspects can be mastered. Moreover such software is usually oriented to one type of application and always has limitations in modelling terms. Therefore a program was written based on well-tested library, Diffpack (Numerical Objects Inc., Norway). Diffpack provides not only the numerical methods but also, all the pre- and post-processing tools (Langtangen, 1999). Furthermore, it is written in C++, which makes the derivation of many programs from each other an easy task. New routines, specifically designed for the problem investigated, can replace the default ones in all part of the simulations, from calculation to analysis. It can be used with a limited knowledge thanks to a graphical user interface (GUI) and many analysis programs. Multiple loop and automatic report generation made the study of various parameters very convenient.

The use of the finite difference method with a specific weight put on the different time steps could provide an elliptic partial differential equation.

$$\partial_t c = \frac{c^{t+\tau} - c^t}{\tau}$$

$$\nabla^T K \nabla c = (1 - \sigma) \nabla^T K \nabla c^t + \sigma \nabla^T K \nabla c^{t+\tau}$$

For  $\sigma = 0$ , the scheme is explicit and for  $\sigma = 1$ , implicit. Usually the Crank-Nicholson scheme ( $\sigma = 0.5$ ) is used. However, including the non-linear robin boundary condition (3), a weighing  $\sigma \leq 0.5$  produced numerical instabilities (Paper V).

The convection term from equation (1) is evaluated at the previous time step leading to an elliptic PDE (4) of the unknown  $c^{t+dt}$  in function of  $c^t$ .

$$c^{t+dt} - \tau \sigma \nabla^T K \nabla c^{t+dt} = c^t + \tau(1 - \sigma) \nabla K \nabla c^t + \nu c^t = g^t \quad (4)$$

Van der Sman (1999) used lattice Boltzmann schemes to solve convection diffusion phenomena. In this case the model was implemented with the Finite Element Method (FEM). A refined grid should avoid numerical instability of the numerical solution despite the very large difference in order of magnitude between the stomatal opening and the domain (Segerlind, 1976).

The aim is now to solve the system (S) at each time step.

$$\left\{ \begin{array}{ll} \text{Find } c \in C(\Omega) \text{ such as} & \\ c^{t+dt} - \tau \sigma \nabla^T K \nabla c^{t+dt} = g^t & \text{in } \Omega \\ c = c_o & \text{on } \Gamma_0 \\ -K \partial_n c = h & \text{on } \Gamma_1 \\ -K \partial_n c = \alpha(c - c_{in}) & \text{on } \Gamma_2 \end{array} \right. \quad (S)$$

Multiplication by any test function,  $u$ , and integration over the domain  $\Omega$  provides the new equation (5):

$$\int_{\Omega} c u \, dx - \tau \sigma \int_{\Omega} \nabla^T K \nabla c \cdot u \, dx = \int_{\Omega} g^t \cdot u \, dx \quad (5)$$

The Green-Riemann theorem allows the introduction of the test function inside a bilinear system and introduces the normal derivative term into equation (5).

$$\int_{\Omega} c \cdot u \, dx + \tau \sigma \int_{\Omega} \nabla c K \nabla u \, dx - \int_{\Gamma} K \partial_n c \, dx = \int_{\Omega} g^t \cdot u \, dx$$

Discretization of the space of solution  $C(\Omega)$  on the space  $P$  defined by the FEM including the right boundary conditions provides the following weak (a.k.a variational) formulation of the problem (S).

$$(S) \Leftrightarrow \left\{ \begin{array}{l} \text{Find } c \in P \text{ such as} \\ \int_{\Omega} c u \, dx + \tau \sigma \int_{\Omega} K \nabla c \nabla u \, dx = \int_{\Omega} g^t \cdot u \, dx \quad \forall u \in P \end{array} \right.$$

The Lax-Milgram theorem ensures the existence and unicity of the solution of the problem (S).

## Concentration at time=500 sec

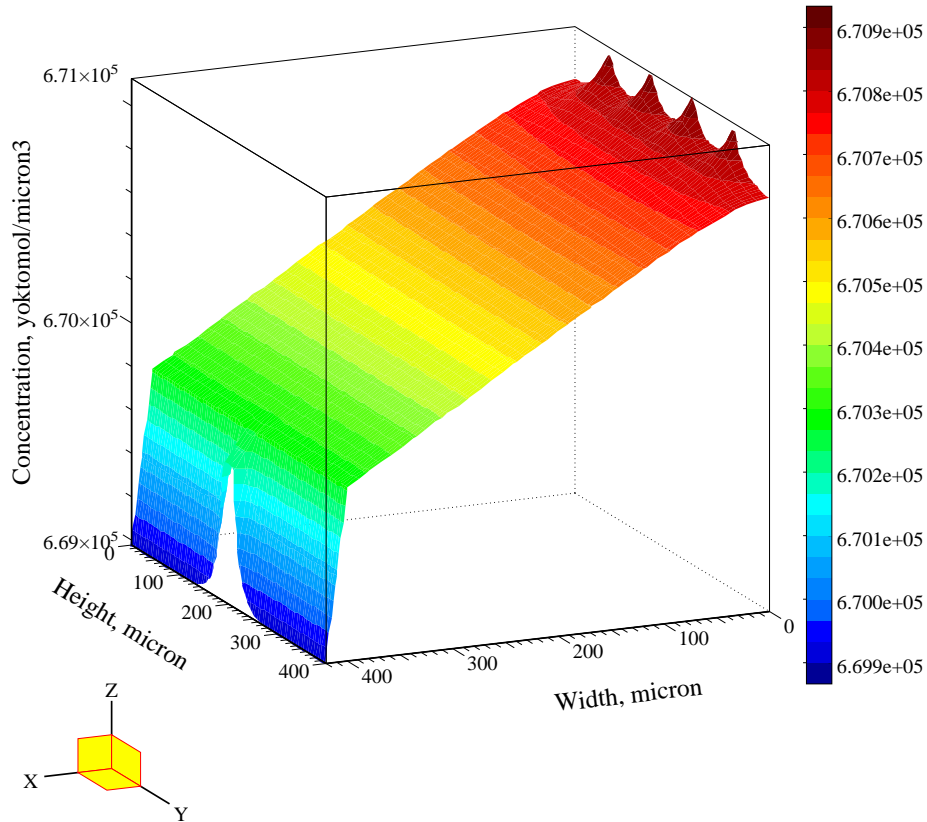


Fig. 3. Results of a 2-D simulation of the water vapour concentration between the epidermis and a packaging film with a microperforation.

Following the classic development of FEM, all the functions are replaced by their linear description in the test function basis of the space  $P$ . After many manipulations, the following linear system was obtained

$$(M + \sigma\tau(K - \alpha B_3^1)).c = (B_1 + \alpha\sigma\tau B_3^2)f$$

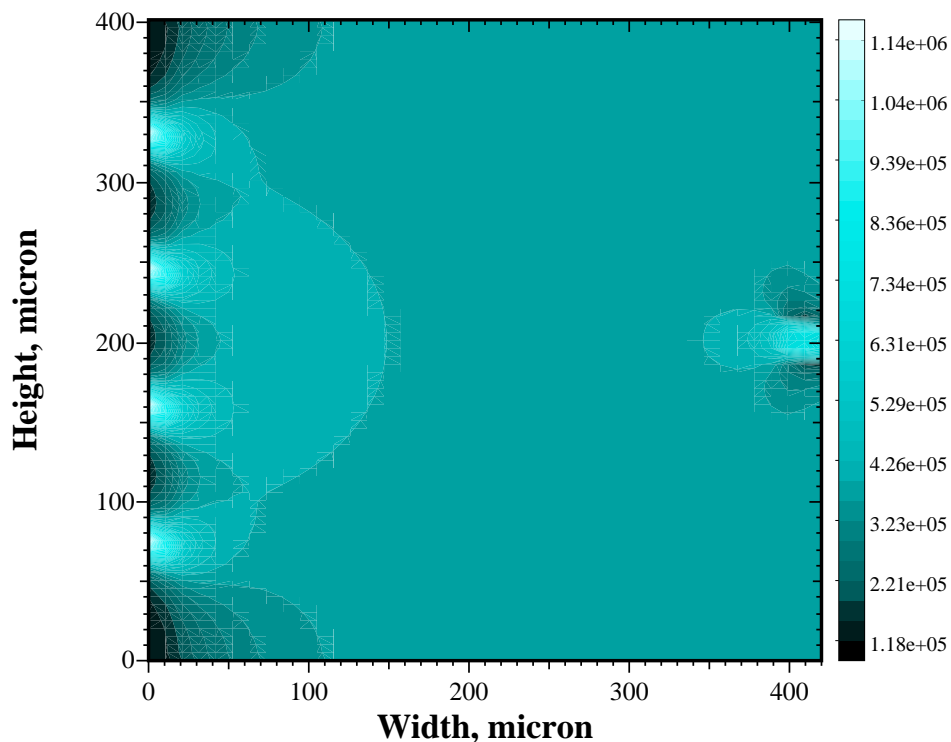
Development of the model from Paper V leads to a similar linear system of type  $A c = y$ . The construction of the stiffness matrix  $A$  provides specific properties easing the numerical resolution of the system. To run the simulation we used a direct LU solving of the system.

### Simulations

The simulations were performed in both two and three dimensions (Paper IV,V). They can provide images of the concentration value of the gas concerned in the whole domain under consideration (Fig. 3). Since a function, piecewise linear is



### flux, at time=1000 sec



*Fig. 4.* Horizontal component of the concentration gradient in the domain simulated in figure 3.

the solution, fluxes can also be analysed (Fig. 4). The stomatal closure, values of permeability and density can easily be simulated. Successful confrontation of the first model to experimental results indicated an important drop in, either the epidermis permeability, or the water vapour concentration inside the cucumber during storage (Paper IV). This led to the inclusion of the chemical potential as a driving force of water loss through the cuticle in the second model (Paper V). The influence of some parameters of the film such as the diffusion coefficient, thickness or hole size on the equilibrium could be investigated as well. Surprisingly, with a small gradient between the composition inside and outside the package, in both two and three dimensions, the size of the microperforations did not influence the equilibrium. Furthermore, the distance between film and epidermis delayed the establishment of equilibrium, but the same level was reached. The most important parameter for the film was the property of the boundary layer on its interface to the surrounding air.

## **Concluding remarks**

A mathematical model has been designed, which could be applied to various domains from the size of a whole cucumber included in a jar, to the space between the epidermis with some stomata and a microperforated film. This project should be considered as the first step towards a better understanding of the phenomena involved in packaging. Further developments in the measuring techniques would be of great help to validate the theoretical developments presented here. Inclusion of all gases measured by the designed set-up is possible. Stronger computation may provide simulation of larger domain with better precision. It could easily be applied to batches of cucumbers as well. Furthermore, it can be included in a complete prediction software to link the parameters film properties, permeabilities, temperature, surrounding RH over the whole distribution chain from harvest to consumption. Another possible development is the extension of simulated domain to the inside of any produce. Measurements of some gas concentrations are much easier in that case. The use of FEM to solve the problem eases the introduction of various types of materials within a single model. The results of the numerical simulations can lead to further experimental investigations focused on the parameter of importance.

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