

## Finite element analysis of diffusion problem during cheese salting: combined influence of space and time discretization

Luiz Henry Monken e Silva<sup>1\*</sup>, Rui Sérgio dos Santos Ferreira da Silva<sup>2</sup> and Dionisio Borsato<sup>3</sup>

<sup>1</sup> Departamento de Matemática, Universidade Estadual de Maringá, Av. Colombo, 5790, 87020-900, Maringá-Paraná, Brazil.

<sup>2</sup> Departamento de Ciência e Tecnologia de Alimentos, Universidade Estadual de Londrina, Londrina-Paraná, Brazil.

<sup>3</sup> Departamento de Química, Universidade Estadual de Londrina, Londrina-Paraná, Brazil. \*Author for correspondence.

**ABSTRACT.** Numerical simulation of solute (sodium chloride) transfer process during salting through a three-dimensional matrix (cheese) was studied applying the finite element method. It was verified that both mesh refinement level and time step length were relevant to control oscillatory behaviors even when unconditional stability schemes as Crank-Nicolson and modified Euler were used. A discussion of the combined influence of time and space adaptation in the context of diffusion problem is also presented, taking in consideration a lumped capacity matrix to overcome the difficulties and determine the minimum length of the time step. Differential mathematical modeling had as theoretical basis the Fick's second law. The proposed model brought good estimation of salt gain in the soft cheese studied. Choosing the appropriate mesh and a convenient time step length we suggest Crank-Nicolson scheme for the simulation of diffusion during cheese brining.

**Key words:** simulation, finite element, oscillatory behavior, cheese salting.

**RESUMO.** Análise da difusão durante a salga de queijos por elementos finitos: influência combinada da discretização tempo-espacial. A simulação numérica da difusão do cloreto de sódio durante o processo de salga através de uma matriz tridimensional (queijo) foi estudada aplicando o método de elementos finitos. Foi verificado que o nível do refinamento da malha e o comprimento do passo de avanço no tempo são relevantes no controle do comportamento oscilatório mesmo quando se usa esquemas estáveis como os de Crank-Nicolson e de Euler-modificado. A influência combinada da adaptação tempo-espço, no contexto da difusão, foi apresentada tomando-se em consideração a matriz de massa concentrada para reduzir as dificuldades na determinação do menor comprimento do passo de avanço no tempo. O modelo matemático diferencial teve como base teórica a segunda lei de Fick. O modelo proposto permitiu uma boa estimativa do ganho de sal no queijo estudado. Com a adequação da malha e da escolha conveniente do comprimento do passo de avanço no tempo pode-se recomendar a escolha do esquema de Crank-Nicolson na simulação da salga de queijos.

**Palavras-chave:** simulação, elementos finitos, comportamento oscilatório, salga de queijos.

The finite element method (FEM) is a numerical approach for solving initial and boundary value problems in engineering and mathematical physics (Chung, 1978). In this method, a continuum is partitioned into many small elements of convenient shapes that are associated to a finite set of nodal points. Interpolation functions describe the behavior of dependent variables, and sometimes they are applied to the independent variables too (Brebbia and Ferrante, 1975; Chung, 1978).

Using a variational formulation or a weight

residual procedure, a partial differential equation is turned into an integral equation, that is changed into a system of ordinary differential equations (when the problem is time dependent) or into algebraic equations, when the problem is not time dependent (Chung, 1978). The discrete system is formed having element by element through a superposition technique. Before solving it, boundary conditions are introduced. When the problem in analysis is time-dependence, it leads to a system of ordinary differential equations that may be solved by implicit

or explicit finite difference schemes (Chung, 1978, Bickford, 1990). The finite element method is a general method with respect to the geometry and material properties. More complex and irregular bodies composed by distinct materials may be considered, because of the flexibility to interpolate the geometry and the model parameters (Wilson and Nickel, 1966; Chung, 1978).

One of the most important mass transfer problems under isothermal conditions, is the cheese brining process. The saline concentration and its distribution over the cheese mass are relevant parameters, responsible for quality and acceptance of the product (Furtado and Souza; 1981; Furtado, 1990).

Geurts *et al.* (1974, 1980) studied the Gouda cheese salt diffusion, during brining, considering it as a semi-infinite medium, assessing the diffusion coefficient. Guinee and Fox (1983) carried out a similar study, on the Romano cheese during salting with NaCl. Luna and Bressan (1986) investigated the sodium chloride diffusion in the Cuartirolo Argentine cheese, considering it as a finite slab, during brining. The same authors used the sodium chloride diffusion coefficient proposed by Geurts *et al.* (1974). Zorrihla and Rubiolo (1994) studied brining of the Fynbo cheese (semi-hard), modeling the NaCl and KCl movement, assuming Fick generalized equation.

Analytic solutions tend to be rather complicated to obtain, when we analyse realistic situations. In this case, numerical methods are valid alternatives (Chung, 1978).

This research paper concerns the influence of time step and mesh refinement to simulate sodium chloride diffusion, during the cheese brining by the finite element method. The results are compared with Luna and Bressan's (1986).

## Methodology

For the mathematical modeling it was used Fick's second law of diffusion that is valid for studying the salt distribution over the cheese (Luna and Bressan, 1986; Zorrihla and Rubiolo, 1991; 1994). Brining is a three-dimensional diffusion process in a transient state with steady boundary (Guinee and Fox, 1987). Cheese is considered as a homogeneous, rigid and finite solid, assuming no chemical reaction and negligible convective flux. The process is done under approximately isothermal conditions (Guinee and Fox, 1987). It was suppose, too, that all the cheese faces were immersed in brine, without resistance between the brine liquid film and the cheese solid surface. Thus, the salt concentration on

the surface would be equal to the salt concentration in the brine (Luna and Bressan, 1986).

Let a cheese occupy a volume  $\Omega \subset \mathbf{R}^3$  defined by

$$\overline{\Omega} \equiv [-R_1, R_1] \times [-R_2, R_2] \times [-R_3, R_3],$$

related with an x,y,z Cartesian coordinate system with origin located in the cheese geometric center.

The concentration of NaCl, at a point  $P(x,y,z) \in \Omega$ , in an instant t,  $C(x,y,z,t)$ , is described by the partial differential equation (Crank, 1975)

$$\frac{\partial C}{\partial t} = D^* \nabla^2 C, \text{ at } \Omega \times (0, \infty), \quad (1)$$

where  $D^*$  is the effective diffusion coefficient, subsidized by the boundary condition that the solute concentration on cheese surface is equal to the concentration in the brine,

$$C(x,y,z,t) = C_s, \text{ at } (x,y,z) \in \partial\Omega, t > 0, \quad (2)$$

where  $C_s$  is the known concentration in the brine (NaCl), considered constant, and by the initial condition,

$$C(x,y,z,0) = C_0, (x,y,z) \in \Omega, \quad (3)$$

where  $C_0$  is the NaCl known initial concentration in the cheese.

The variational formulation (Galerkin's method) of the partial differential problem, described by the equations (1), (2) and (3), can be formally obtained as follows:

i) the equation (1) is multiplied by a function  $v \in H$ , where  $H$  is a space of admissible functions, later on characterized, and both sides are integrated on the  $\Omega$  domain, to have

$$\int_{\Omega} v \frac{\partial C}{\partial t} d\Omega = \int_{\Omega} D^* v \nabla^2 C d\Omega \quad ; \quad (4)$$

(ii) following, the right side of (4) is integrated by parts to obtain

$$\int_{\Omega} D^* v \nabla^2 C d\Omega = D^* \int_{\Omega} v \frac{\partial C}{\partial n} ds - D^* \int_{\Omega} \nabla v \cdot \nabla C d\Omega, \quad (5)$$

where  $\partial/\partial n$ , is the normal derivative operator;  $ds$ , is the measure of integration on the boundary;  $d\Omega$ , is the measure of integration on the domain.

Since the boundary condition (2) is Dirichlet type (Chung, 1978), to assure the existence and unicity of the variational problem it is necessary that  $v$  satisfies

$$v = 0, \text{ at } \partial\Omega. \quad (6)$$

Take the space of admissible functions,  $H$ , as being  $H = \{v \in H_1(\Omega) ; v = 0 \text{ at } \partial\Omega\}$ , where  $H_1$  is

a Hilbert space of order one defined on  $\Omega$ .

So, the equation (5) becomes

$$\int_{\Omega} D^* \nabla v \cdot \nabla^2 C \, d\Omega = - D^* \int_{\Omega} \nabla v \cdot \nabla C \, d\Omega. \quad (7)$$

Substituting (7) in (4) follows:

$$\int_{\Omega} v \frac{\partial C}{\partial t} \, d\Omega = - D^* \int_{\Omega} \nabla v \cdot \nabla C \, d\Omega, \quad \forall v \in H. \quad (8)$$

The equation (8) serves as a basis to establish the finite element method.

Therefore, it is considered a partition of  $\bar{\Omega}$  in  $N_e$  subdomains  $\Omega^e$ ,  $1 \leq e \leq N_e$ , called finite elements, so that:

$$\bigcup_{i=1}^{N_e} \bar{\Omega}^i = \bar{\Omega}, \quad (9)$$

$$\Omega^i \cap \Omega^j = \emptyset, \quad \forall i \neq j, \quad 1 \leq i, j \leq N_e.$$

The concentration in each element,  $C^e(x, y, z, t)$ , is approximated by means of the following expression:

$$C^e(x, y, z, t) = \sum_{k=1}^n C_k^e(t) \phi_k(x, y, z), \quad (10)$$

where  $n$ , is the number of nodal points of the  $\Omega^e$  element;

$C_k^e(t)$ , is the concentration at  $k$ -esimal node in the element  $e$ , at time  $t$

$\phi_k^e(x, y, z)$ , is the interpolation function associated to the  $k$  node of the element.

Taking,  $v = \phi_i$  and substituting (10) in (8), it gives

$$\sum_{e=1}^{N_e} \int_{\Omega^e} \phi_i^e \sum_{j=1}^n \phi_j^e \, d\Omega \cdot \frac{\partial C_j^e}{\partial t} = D^* \sum_{e=1}^{N_e} \left\{ \int_{\Omega^e} \nabla \phi_i^e \left( \sum_{j=1}^n \nabla \phi_j^e \, d\Omega C_j^e(t) \right) \right\} \quad (11)$$

where  $1 \leq i \leq n$ .

The sum sign in (11), must be understood as an assembling process over all elements. Thus, (11) can be rewritten in matrix form as

$$\sum_{e=1}^{N_e} M^e \dot{C}^e = \sum_{e=1}^{N_e} K^e C^e, \quad (12)$$

where  $K^e$ , is the diffusivity matrix, whose elements are calculated by

$$K_{ij}^e = - D^* \int_{\Omega^e} \left[ \phi_i^e \frac{\partial \phi_j^e}{\partial x} + \frac{\partial \phi_i^e}{\partial y} \frac{\partial \phi_j^e}{\partial y} + \frac{\partial \phi_i^e}{\partial z} \frac{\partial \phi_j^e}{\partial z} \right] d\Omega^e; \quad (13)$$

$M^e$  is the distributed mass matrix whose elements are calculate by

$$m_{ij}^e = \int_{\Omega^e} \phi_i^e \phi_j^e \, d\Omega^e, \quad 1 \leq i, j \leq n; \quad (14)$$

$C^e = [c_1, c_2, \dots, c_n]^T$ , is the unknown vector concentration of the  $e$  element;

$\dot{C}^e = [\dot{c}_1, \dot{c}_2, \dots, \dot{c}_n]^T$ , is the unknown derivative vector concentration, with respect to time.

Before solving the set of ordinary differential equations (12) it is necessary to insert the boundary conditions.

The Crank-Nicolson and the modified Euler methods were used to discretize on time (Cook, 1981; Edakin, 1986; Bickford, 1990). These two schemes are unconditionally stable. In spite of this, they may show undesirable oscillatory behavior if the time step was not suitably chosen (Zienkiewicz and Morgan, 1983; Lyra, 1993). The oscillatory limit, for one-dimensional problem, may be expressed as (Zienkiewicz and Morgan, 1983)

$$\Delta t_{osc} < \frac{h^2}{4D^*(1-\theta)}, \quad (15)$$

where  $h$ , is the smallest dimension over all mesh elements (cm);

$D^*$ , is the diffusion coefficient (cm<sup>2</sup>/hour), that substitutes the thermal diffusivity ( $\alpha$ ) in the original expression, maintaining the dimensionality;

$\theta$  is equal  $1/2$  for the Crank-Nicolson method and  $\theta$  is equal 1 for the modified Euler method.

The spatial domain was represented by a set of serendipity hexahedral elements of  $C^0$  type. Each element has 20 nodes at the edges and vertices of its outer surface, and an overall of 20 degrees of freedom (Brebbia and Ferrante, 1975).

Sodium chloride diffusion during salting of the Cuartirolo cheese (Luna and Bressan, 1986) was simulated using a mesh configuration described in Silva *et al.*, (1998).

Mean concentrations (g NaCl/100 g of dry basis cheese) were obtained through the average of the first five elements disposed along 1.5 cm of the  $x$ -axis. They were compared with the data obtained by Luna and Bressan (1986). In each element the mean concentration was calculated as follows:

$$\bar{C}^e(t) = \frac{1}{V^e} \int_{\Omega^e} C_{(x,y,z)} \, dV. \quad (16)$$

For comparisons, the per cent deviation values were adopted as in Heldman, (1974). The

program used was the "SIMUL" developed in vectorial FORTRAN for the IBM 3090 computer (Silva *et al.*, 1998).

## Results and discussions

Sodium chloride diffusion, in brining Cuartirolo Argentine cheese process, was simulated by using the mesh described in Silva, *et al.*, (1998). The effective diffusion coefficient used was  $D^* = 0,31 \text{ cm}^2/\text{day}$ , obtained through the nomogram proposed by Geurts *et al.* (1974), taking 52% for the moisture content and 51,6 % (dry basis) for fat level as in Luna and Bressan (1986). The brine was maintained at the constant temperature of  $7,5^\circ \text{C}$ , during seven hours of brining and the sodium chloride concentration was  $C_s = 20,5^\circ \text{Be}$ . (Luna and Bressan, 1986).

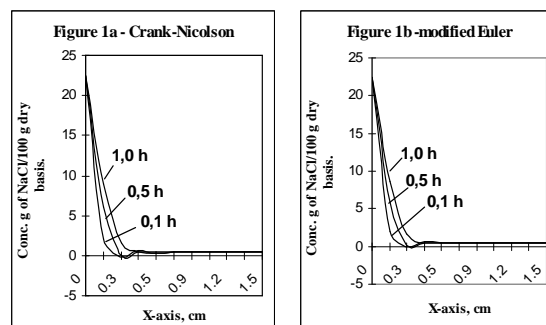
The numerical solution by finite element method of parabolic problems, as the diffusion equation has high oscillatory components in the beginning of the evolution situation that they decay rapidly. Therefore, to attain computational efficiency and accuracy, in this class of problems, it is needed to combine spatial mesh refinement and time step length control (Lyra, 1993).

When the boundary is steady and the diffusion coefficient ( $D^*_{\text{NaCl}} = 0,31 \text{ cm}^2/\text{day}$ ) is relatively small (Loncin and Merson, 1979), the characteristic spectra of the concentration frequencies may be wide (Lyra, 1993). In brining process, the brine concentration is high when compared to the NaCl initial concentration in the cheese (Geurts *et al.*, 1974; Guinee and Fox, 1987). In these conditions, the high frequencies rule the initial stage of the transient response. The usage of unconditionally stable finite difference schemes, may present oscillatory behavior, if it was not imposed restrictions in the time step length (Lyra, 1993). In the lack of information of three-dimensional problems, the oscillatory limit ( $\Delta t_{\text{osc}}$ ) can be expressed by the equation (15) that serves to the one-dimensional case of heat conduction (Zienkiewicz and Morgan, 1983). Taking  $h_{\text{min}} = 0,3 \text{ cm}$  and  $D^* = (0,0129) \text{ cm}^2/\text{hour}$ , the Crank-Nicolson scheme needs  $\Delta t_{\text{osc}} < 3,48 \text{ hours}$  and the modified Euler method,  $\Delta t_{\text{osc}} < \infty$ .

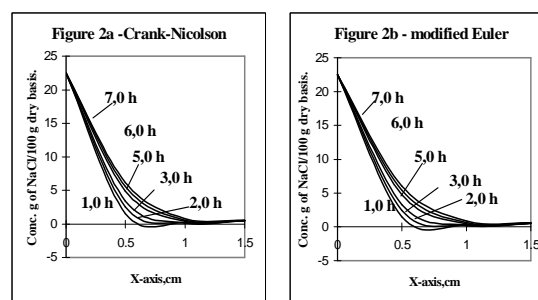
When Crank-Nicolson scheme imposes a maximum  $\Delta t$  of 3,5 hours, corresponding at about 50 % of the real time of brining, it was chosen to work with  $0,1 \text{ hour} \leq \Delta t \leq 1 \text{ hour}$ . This permits comparisons between the analytical and the experimental data obtained by (Luna and Bressan, 1986) and the results here obtained by FEM. Shorter steps are convenient to account the stability and convergence. In practice, computational efficiency

(the machine's time costs) and reliable responses must be taken into account (Lyra, 1993). A too refined mesh and a higher number of short time steps are options to produce oscillation damping, and lead the numerical method to convergence. However, it may be expected as a consequence, an undesirable increasing of the computational time (Lyra, 1993).

The Figures 1a and 1b show the high difference between the sodium chloride concentration in the brine ( $C_s$ ) and the NaCl initial concentration in the cheese ( $C_0$ ). They also showed oscillations on the first stages and their damping as the cycle numbers with  $\Delta t = 0,1 \text{ hour}$  increase. The stability is reached, in Crank-Nicolson's algorithm as well as in the Modified Euler in the three-dimensional problem, after the first ten cycles, in the first hour of the brining process. Then a time step,  $\Delta t = 1 \text{ hour}$ , may be recommended under the conditions stated in this problem.



**Figure 1.** NaCl concentration profiles along x-axis during diffusion simulation in Cuartirolo Argentine brining with  $\Delta t = 0,1 \text{ h}$  and  $h_{\text{min}} = 0,3 \text{ cm}$  for 0,1; 0,5 and 1,0h brining times



**Figure 2.** NaCl concentration profile along x-axis during diffusion simulation in Cuartirolo Argentine brining with  $\Delta t = 1,0 \text{ h}$  and  $h_{\text{min}} = 1,0 \text{ cm}$

Figures 2a and 2b show the saline concentration profile, in another space and time arrangement. In this simulation the elements used were with minimum dimension  $h = 1,0 \text{ cm}$  and  $\Delta t = 1,0 \text{ hour}$

for the brining times of 1,0; 2,0; 3,0; 5,0; 6,0 and 7,0 hours.

Figures 2a and 2b show the oscillation and the damping that appear when the element dimension is raised from 0,3 to 1,0 cm with  $\Delta t = 1,0$  hour. The oscillation occurs until the first hour of brining, and the deviation in percentage is too large as proved in Table 1. The step length proposed by Murti *et al.* (1989),

$$\Delta t_{\min} \geq \frac{\gamma h^2}{3\alpha\theta} \quad (17)$$

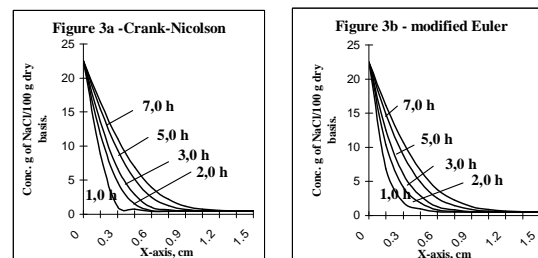
is recommended for non-steady two-dimensional heat conduction problem. In the equation 17,  $\gamma$  is a correction factor ( $\gamma=2,0$ , is recommended to isoparametric elements in irregular mesh.).

However, a simple substitution of  $\alpha$  for  $D^*$  keeping the dimensionality and supposing analogy among phenomena, produces unexpected and overestimated results. This fact can be explained in part because normally, in solid foods (Loncin and Merson, 1979), the thermal diffusivity ( $\alpha$ ) is  $O(10^2)$  of magnitude that is a factor of  $10^3$  times greater than the mass diffusion coefficient ( $D$ ). The difficulties in finding a  $\Delta t_{\min}$  to have a better precision can be overcome by the employment of "lumped capacity matrix" (Lyra, 1993). In this case, no limit on the minimum length of the  $\Delta t$  is imposed. Only accuracy or physical reasons will determine the time step length (Lyra, 1993). Therefore, it is suggested some procedures for making a decision:

1. establish a mesh refinement with  $h_{\min}$  (0,3 cm) so that the computational load is viable;
2. choose  $\Delta t$  on basis of the process just described. Cuartirolo cheese brining is not long (Luna and Bressan, 1986), therefore  $\Delta t = 1$  hour seems to be an initial reasonable choice. The experimental planning may be also a good indication. Finally, the  $\Delta t$  choice would be a compromise between computational time and reliable responses.

However, Lyra (1993) supports that the modified Euler scheme reduces substantially the oscillations that rule the transient responses in heat conduction problems. This fact was not observed in the diffusion simulation during Cuartirolo cheese brining, when it was examined by the two numerical schemes. In other words, it was verified the presence of small oscillations on the first stages (until 0,45 cm from cheese surface) regardless the employed scheme, when it was used  $\Delta t = 0,1$  hour as time step length (Figures 1a and 1b).

This observation added to the unsuccessful application of the equation 17, leads to an examination of these issues in the numerical analysis context. But this subject is beyond the scope of this paper.



**Figure 3.** NaCl concentration profile along x-axis during diffusion simulation in Cuartirolo Argentine cheese brining with  $\Delta t = 1,0$  hour and  $h_{\min} = 0,3$  cm

Figures 3a and 3b show the salt concentration profile, at distinct brining times, obtained by FEM, employing Crank-Nicolson (Figure 3a) and modified Euler (Figure 3b) schemes.

It is observed oscillations only in the first hour of brining process when the Crank-Nicolson scheme was used (Figure 3a). But these oscillations were not observed with the modified Euler method (Figure 3b). This indicates that with a suitable choice of the time step length and the employment of the modified Euler method the oscillations can be reduced (Lyra, 1993).

**Table 1.** Mean concentration, along 1,5 cm x-axis, in g NaCl/100 g of dry basis cheese and deviation in the diffusion simulation during brining with elements of dimension  $h_{\min} = 1,0$  cm

Duration of brining in hours	1,0	3,0	5,0	6,0	7,0	Deviation (%) *	
$\Delta t$ Computational time Method	g NaCl / 100 g of dry basis cheese.					E.V.	A.V.
1,0 h 14 minutes	Analytical **	2,30	3,67	4,61	5,01	5,37	
	Experimental **	2,23	3,66	4,48	4,96	5,33	
	Crank-Nicolson	3,80	4,40	5,16	5,42	5,72	37,87
	Modified Euler	3,72	4,35	5,10	5,38	5,70	35,81

E.V. = experimental value. A.V. = analytic value.  $\Delta t$  = time step size; \*Heldman (1974). \*\*Luna and Bressan (1986)

**Table 2.** Mean concentration, along 1,5 cm x-axis, in g NaCl/100 g of dry basis cheese and deviation in the diffusion simulation during brining with elements of less dimension  $h_{\min}=0,3$  cm

Duration of brining in hours			1,0	3,0	5,0	6,0	7,0	Deviation (%) <sup>*</sup>	
$\Delta t$	Computational time	Method	g NaCl/100 g of dry basis cheese					E.V.	A.V.
1,0h	14 minutes	Analytical <sup>**</sup>	2,30	3,67	4,61	5,01	5,37	1,70	2,33
		Experimental <sup>**</sup>	2,23	3,66	4,48	4,96	5,33		
		Crank-Nicolson <sup>**</sup>	2,20	3,61	4,59	5,01	5,38		
		Modified Euler <sup>**</sup>	1,86	3,43	4,47	4,89	5,28		
								8,91	10,33

E.V. = experimental value. A.V. = analytic value.  $\Delta t$  = time step size; <sup>\*</sup>Heldman (1974). <sup>\*\*</sup>Luna and Bressan (1986)

Comparing Figures 1a and 3a (obtained with distinct time steps,  $\Delta t$ ) it was verified that as  $\Delta t$  increases the amplitudes of the oscillations they increase too. This undesirable effect was not propagated along brining process and it did not damage the performance of Crank-Nicolson method because after the first hour of brining process the oscillation was not observed anymore (Figure 3a).

The analysis of the results, in Table 2, gives evidence of the precision of Crank-Nicolson method, because the numerical results are similar to the analytical and experimental ones. The values obtained by the Crank-Nicolson method, have mean deviation with less than 5 % (Heldman, 1974).

Regardless the results about the first brining hour, the mean deviation with  $\Delta t= 1,0h$ , for the modified Euler algorithm, reduces from 8,91% to 3,76% (numerical *versus* experimental method) and from 10,33% to 4,49% (numerical *versus* analytical). This is due to the oscillation on the first stages of the numerical solution.

Mathematical modeling of the diffusion process, during cheese salting by means of the finite element method, allowed a good estimate of the cheese salt gain. To reduce the oscillation in the numerical results provided by FEM schemes, appropriated time step length, and mesh refinement were unconditionally used in the process. The computational approach developed here could be applied to diffusion simulation of other food products (solids), under similar process conditions.

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