



Application of self-organizing maps to evaluate the influence and behavior of the film formed during salting of Prato cheese

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Abstract

Problems related to high blood pressure have led consumers to choose foods with low levels of sodium chloride, and an alternative to reduce the salt content of this salt is its partial replacement with KCl. In order to not affect the sensory properties of cheese, salting was performed using a solution containing 70% of NaCl and 30% of KCl in a static and dynamic system. During the salting process by immersion, the mass transfer is affected by the formation of a film on the cheese surface. The analysis of the diffusion of salts in the film can be performed using self-organizing map (SOM) combined with the Finite Element Method (FEM). Through these tools, a greater influence of the film was observed when the static system was applied and that the diffusion is different according to the studied position. On the biosolid sides the diffusion was more pronounced than in the center, indicating a decrease in the film thickness towards the edges. The SOM combined with 3D modeling showed to be an efficient tool to investigate the formation, influence and behavior of the film during the diffusion of Na⁺ and K⁺ on salting of Prato cheese.

Keywords: neural networks; diffusion; finite element methods; simulation.

Practical Application: Simulation of situations that will be very useful for industries working with diffusive processes.

1 Introduction

Currently, a direct relation between sodium content in food and its influence on blood pressure has led consumers to choose healthier products. Thus, the production of foods with low sodium content is necessary, but sodium chloride plays a fundamental role in the final quality of a product. Therefore, its total substitution is not recommended (Bordin et al., 2019). KCl can replace NaCl by up to 30% without affecting the acceptability of the final product. It is possible to replace part of the sodium chloride with potassium chloride, as this salt does not alter the sensory, physicochemical or microbiological characteristics of the product. In addition, potassium intake increases kidney excretion of sodium, resulting in an antihypertensive effect and also exhibits antimicrobial activity (Bordin et al., 2019; Borsato et al., 2012).

Cheese is a product widely used by Brazilians. The most consumed types are: Prato, Mozzarella, Minas and Parmesan. Among them, Prato cheese stands out for its high nutritional value. A homogeneous distribution of NaCl is essential for its final quality (Bona et al., 2006, 2010; Silva et al., 2017). Therefore, salting is an important step in cheesemaking, consisting of its immersion in brine, static or dynamic, in which the salt spreads into the solid by mass diffusion mechanisms (Albarracín et al., 2011; Guinee, 2004).

Studies on the physicochemical properties and characteristics of various cheeses have recently been conducted. Matera et al. (2018) studied the physicochemical characteristics of Brazilian

cheeses Salum et al. (2019) the compositional properties of enzyme-modified commercial cheeses, Punoo et al. (2018) the textural and microstructural properties of Kradi cheese and Rafiq et al. (2018) cheddar cheese peptide extracts in anticancer activity. Researches related to Prato cheese include the partial replacement of NaCl by KCl and the addition of flavor enhancers to the probiotic Prato cheese (Silva et al., 2018), the change in volatile compound concentration during the maturation of Prato cheese by reducing fat and adding whey protein (Domingos et al., 2019), and attenuation of cigarette smoke-induced lesions in mice by the probiotic Prato cheese (Vasconcelos et al., 2019).

According to Schwartzberg & Chao (1982), when a fluid is in contact with a solid, a film is formed on its surface. If there is a mass transfer between the surface and the fluid, the current must pass through the stationary layer, which acts as a resistive barrier, and therefore there is a specific diffusion coefficient that describes the mass transfer in this film. The question is that this coefficient cannot be determined only experimental, and so to evaluate the influence of the formed film it is necessary to take into account the ratio between the resistance to internal and external mass diffusivity that can be quantified by the Biot mass number. According to Bona et al. (2007), high values of the Biot number indicate that the internal resistance is limiting, and during the diffusion process as the Biot number decreases, the resistance of the film formed at the solid/solution interface increases (Rakotondramasy-Rabesiaka et al., 2010; Schwartzberg & Chao, 1982). Thus, the Biot number is related to the film

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coefficient, since the higher its value, the lower is the influence of the formed film.

When it comes to food biosolids the diffusion on surface as well as film formation may be dependent on the geometry and position where the diffusion occurs. Thus, to better understand the effects of these factors during the process, some data analysis tools can be used through unconventional statistics (Cremasco et al., 2016, 2019; Haykin, 2001).

One tool is the SOM-type ANN. It has been applied to solve various types of problems in different areas of science such as engineering, chemistry, medicine, bioenergy, among others (Bona et al., 2012; Borsato et al., 2011; Cremasco et al., 2016; Lemes & Dal Pino, 2008; Lindsey et al., 2018; Walkoff et al., 2017). The purpose of this tool is to transform an input pattern of arbitrary dimension into a discrete one- or two-dimensional map, and perform this transformation adaptively in a topologically ordered manner. The interesting thing about it is that the result can be observed through a topological map whose function is to facilitate the interpretation of the results (Kohonen & Maps, 1995).

The objective of this work was to investigate the formation, influence and behavior of the film during multicomponent diffusion of NaCl and KCl, in static and dynamic brine, applying ANN type SOM with 3D computational modeling using the Finite Element Method (FEM).

2 Materials and methods

2.1 Prato cheese

It was used 3 kg of Prato cheese with rectangular geometry, provided by Laticínios Campina Alta (Manoel Ribas-PR). The cheese was divided into standardized geometry samples of 0.04 m × 0.04 m × 0.02 m (Figure 1).

2.2 Brine preparation and sampling

It was used 15 L of brine with 5% (w/v) salt concentration, containing 30% of potassium chloride (KCl) and 70% of sodium chloride (NaCl). For the system with agitation was used a pump with 520 L h⁻¹ of circulation flow, with a constant temperature of 20 (± 1 °C). Prato cheese samples were completely immersed in the static and dynamic brine being collected, and their dimensions measured using a digital caliper at times of 0, 0.25, 0.75, 1.0, 2.5, 6.5, 7.5, 9.5, 10.5, 11.5, 17.5, 19.0, 29.0, 36.0, 40.0, 50.0 and 62.0 hours.

2.3 Determination of sodium and potassium

NaCl and KCl concentrations in Prato cheese samples were determined according to the methodology described by Bordin et al. (2019) with modifications, using the Micronal photometer, model B-462, with air pressure of 0.8 kgf cm⁻² and 1.5 kgf cm⁻² air pump pressure using butane gas.

2.4 Three-dimensional modeling

The modeling was performed using the finite element method considering a rectangular domain, three-dimensional mass transfer, the generalized equations of Fick second law and

Onsager (1945) equations. The diffusion coefficient was considered constant, regardless of the position and the immersion time in the brine. The solute diffusion occurred under isothermal conditions (20 °C) and the cheese contraction during salting was not considered. To evaluate the influence of the film formed on the cheese surface, in the two salting systems used, the Cauchy boundary condition was considered (Bona et al., 2007). The finite element formulation followed the procedures was established by Cremasco et al. (2019).

2.5 Finite element method simulation

The simulation was performed using COMSOL Multiphysics® software and the standard “Transport of Diluted Species (tds)” physical interface. The parameters used in the simulation were the main and cross diffusion coefficients, Biot mass number and the relationship between the mass transfer coefficient and the mass conductivity (h_m/λ_m), that were adjusted using the super modified simplex method (Bona et al., 2000). Figure 1 shows the solid generated automatically by the software, showing part of the extremely fine mesh used and the equidistant points chosen for the study of Na⁺ and K⁺ diffusion on the surface of the film formed in the Prato cheese.

2.6 Artificial Neural Networks (ANN)

For the analysis of the influence of the film on the Prato cheese surface, a Kohonen self-organizing map (SOM) was used. The applied network presented a 6x6 hexagonal topology with 8000 training epochs, with learning rate training starting at 0.2 and decaying exponentially with the training epochs to 6.71x10⁻⁵, and with the initial neighborhood relation of 3.5 decaying to 0.045.

2.7 Computational processing

For the simulation it was used the software COMSOL Multiphysics® version 5.2 (COMSOL, Inc., Burlington, MA) based on the finite element method. The neural network routine developed by our research group was used according to the

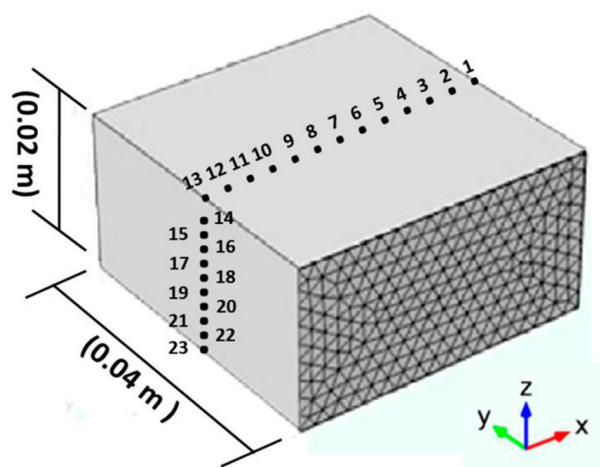


Figure 1. Solid automatically generated by the software and the positions used in the simulation associated with a set of coordinates x, y, z.

algorithm described in Haykin (2001) and processed by Matlab® R2007b software to evaluate the influence and behavior of NaCl and KCl concentrations in the Prato cheese surface.

3 Results and discussion

The main and cross diffusion coefficients, the film coefficient, the ratio between the mass transfer coefficient and the mass conductivity, and the Biot number of the Prato cheese, subjected to salting in the static and dynamic process, were determined by simulation using the finite element method with the application of a simplex optimization coupled with the desirability functions (Bordin et al., 2019). All simulations were performed using a 3D geometry modeling, with time-dependent solver Backward Differentiation Formula (BDF) with extremely fine tetrahedral finite element mesh, composed of 130,382 elements with 370,052 degrees of freedom. Since the method is iterative and constrained, to avoid possible oscillations and an excessive number of vertices in the simplex procedure (Bona et al., 2000; Bordin et al., 2019), the lower and upper limits of each variable were defined through previous tests. The results obtained were within the range of the independent variables chosen and convergence to the optimal was achieved faster (Angilelli et al., 2015).

The values of the main (D_{ii}) and cross (D_{ij}) diffusion coefficients, the film coefficient (h_m), the ratio between the mass transfer and the mass conductivity coefficients (h_m/λ_m), the percentage errors obtained from the difference between experimental and simulated data, and the Biot number, using

the COMSOL Multiphysics® software, during the diffusion process, without and with agitation, are presented in Table 1.

The optimized values of Biot numbers and film coefficients (Table 1) show a difference in mass transfer between the static and the dynamic process. The lower value for the Biot number and the higher value for h_m/λ_m show that the influence of the barrier on the solution/biosolid interface is greater in the static system, indicating ions resistance to transfer from the solution to the Prato cheese (Bordin et al., 2019).

According to Table 1, it is observed that the main and cross coefficients in the dynamic and the static brine are the same, since these parameters are related to the mass transfer inside the biosolid, being not dependent on the external disturbance of the system. The main diffusion coefficient of Na^+ (D_{11}) is 1.7 times higher than the main diffusion coefficient of K^+ (D_{22}). Also, the cross coefficients presented smaller values than the main ones, showing that the diffusion of the solutes in their own flow is more important than the interference between them.

According to Cremasco et al. (2019), in a study of the diffusion in the solution/quail egg interface, the influence of the film formed during the diffusion process can be studied by analyzing the solute concentrations at different positions over time. The diffusion profile in static and dynamic systems, comparing experimental and simulated concentrations in $\text{g salt}/100\text{g}_{\text{(solution)}}$ is shown in Figure 2. Therefore, there must be a physical barrier on the outer surface of Prato cheese as it was observed that the mass transfer was faster when the stirring system was applied (Figure 2b). In addition, the Biot number for the dynamic system

Table 1. Parameters obtained by simplex optimization during the sodium and potassium chloride diffusion processes in static and dynamic salting.

	Static Brine		Dynamic Brine	
	Na^+	K^+	Na^+	K^+
Main coefficients (m^2/s)	0.5007×10^{-9} (D_{11})	0.3000×10^{-9} (D_{22})	0.5007×10^{-9} (D_{11})	0.3000×10^{-9} (D_{22})
Cross coefficients (m^2/s)	1.2735×10^{-10} (D_{12})	0.7031×10^{-10} (D_{21})	1.2735×10^{-10} (D_{12})	0.7031×10^{-10} (D_{21})
h_m (m/s)	5.1371×10^{-7}	3.0780×10^{-7}	2.1029×10^{-6}	1.2600×10^{-6}
Percentual error	6.20%	5.68%	4.64%	4.02%
h_m/λ_m (m^{-1})	1025.7610		4200	
Biot*	20.52		84.00	

*Biot number calculated relative to x-axis.

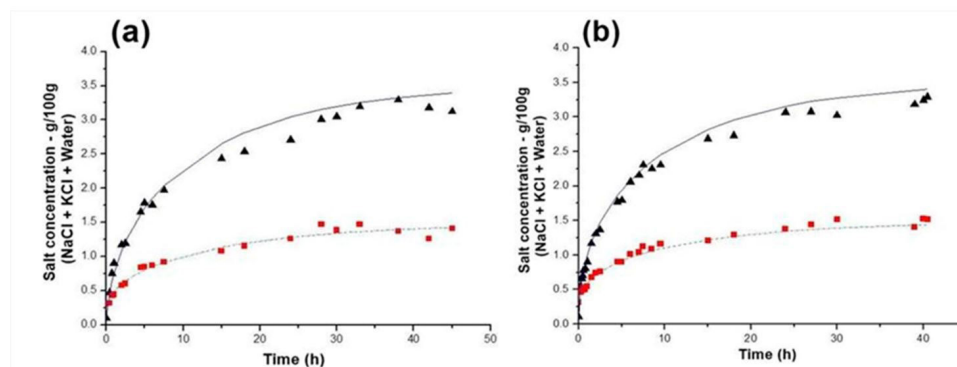


Figure 2. Diffusion profile of experimental and simulated salt concentrations during the static brine (a) and dynamic brine (b) experiments. NaCl experimental data are represented by (\blacktriangle) and KCl by (\blacksquare), NaCl and KCl simulated data are represented by (-) and (...).

was 84 and for the static system 20.52 (Table 1), indicating the greatest influence of the film formed on the cheese surface when the salting process is not agitated (Figure 2a).

The NaCl and KCl concentrations used in brine were 597.83 and 201.48 mol.m⁻³, respectively. These should be the concentrations obtained on the surface of the cheese immediately after immersion if the Dirichlet boundary conditions were applied (Chung, 1978). However, after 15 minutes of salting, the simulated average values found in the static system were 322.76 and 100.40 mol m⁻³ for NaCl and KCl, respectively, and in the dynamic brine were 507.07 and 162.30 mol m⁻³ for NaCl and KCl, respectively. These values prove the influence of the film formed on the surface and that it was not eliminated even with agitation, selecting the Cauchy conditions (Bona et al., 2007). Therefore, as salting time increases, these values tend to approach the initial concentration of the brine used.

To analyze the influence of the film formed on the surface of Prato cheese the concentration at each chosen point during 45 hours was simulated through FEM. The values obtained were analyzed through the application of self-organizing map (SOM)-type neural networks. In a preliminary study, 8000 epochs were used, where it was possible to verify that the stabilization of the mean quantization error occurred after 6000 epochs, so this value was used in the network training.

Figure 3 shows the topological maps with concentration distribution at the points located in the upper and side surface of the cheese (Figure 1), during the Na⁺ and K⁺ multicomponent diffusion process. Figures 3a and 3b represent the maps of the upper points on the x-axis and Figures 3c and 3d the side points on the z-axis for both dynamic (D) and static systems (S). According to the Figure 3 it is noted that the influence of the formed film is not the same for Na⁺ and K⁺, since the topological

map presented changes when analyzed in the same axis. Thus, is possible to say that not only the intrinsic properties of the two ions, such as their ionic radius, charge density, among others, that determine their mobility on the cheese surface, but also the position and type of ion considered.

Figure 3a shows some positions that are different from the topological map shown in Figure 3b, *i.e.* the neuron where the 1D and 13D positions are located, for the K⁺ on the x-axis has similarity to the neurons that are located in the positions 3D, 4D, 11D, 2D and 12D. However, this behavior is not repeated in the topological map shown in Figure 3a. On the other hand, the same similarity relation between neurons observed in Figure 3c occurs in Figure 3d. In both axes the neural network used showed that on the surface of the cheese there is a clear separation between the dynamic and static processes, because in the topological map, the more distant the points the lower the similarity between them (Cremasco et al., 2019).

As the analyzed positions move away from the center point (position 7 of Figure 1) towards the cheese edge, topological maps show that their similarity decreases, indicating that the thickness of the formed film varies along the x-axis. Thus, at position 7 the resistance to ion transfer is greater, decreasing as it moves away from the center of the cheese surface. The same behavior is observed when analyzing the positions located on the z-axis.

Figures 4 and 5 show the weight maps at the different times evaluated in the simulation (FEM) for Na⁺ (Figure 4) and K⁺ (Figure 5) in the static and dynamic systems, for x and z-axis. In these maps, the positions in the darker regions have greater weight, and as the color intensity decreases a lower weight is attributed.

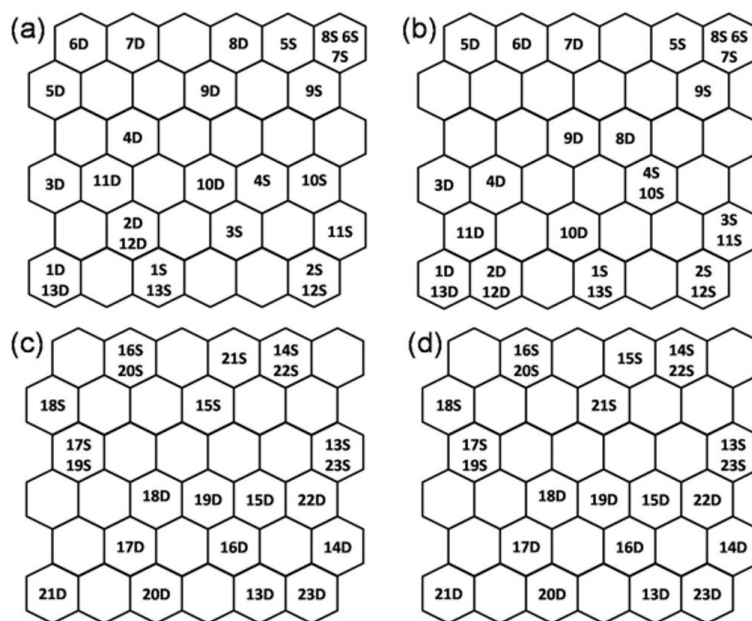


Figure 3. Distribution of the analyzed points according to the neuron for sodium (a) and potassium (b) on the x-axis and sodium (c) and potassium (d) on the z-axis, for the static (S) and dynamic (D) systems.

It is possible to observe that there are differences in ion concentration values in relation to the diffusion time in different positions of the biosolid. Note that there was a separation of the data obtained between the static and the dynamic system, in the

positions studied during the simulation of the diffusion in Prato cheese. Each position defined for the static system presented lower concentration values when compared with the concentrations of the same positions in the dynamic system. This observation

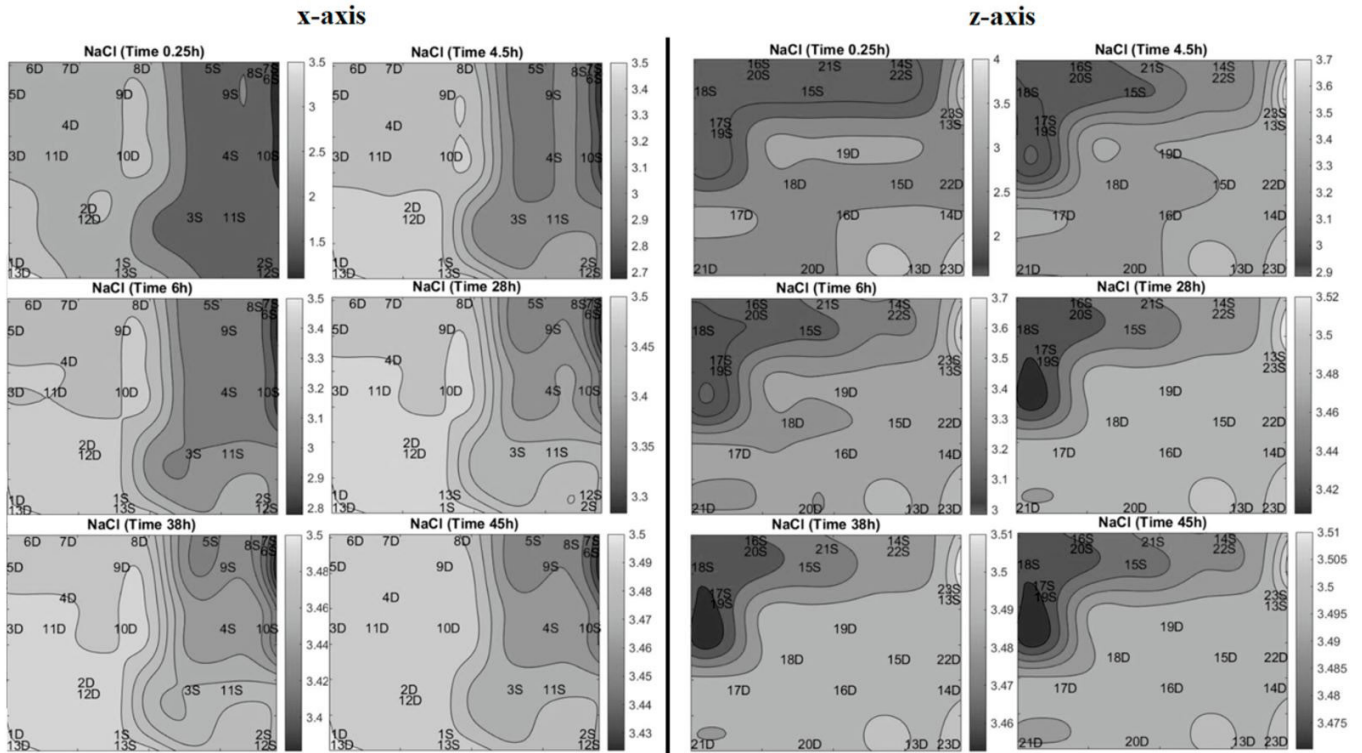


Figure 4. Weight map of NaCl diffusion on the biosolid surface for the x-axis and z-axis in static (S) and dynamic (D) systems.

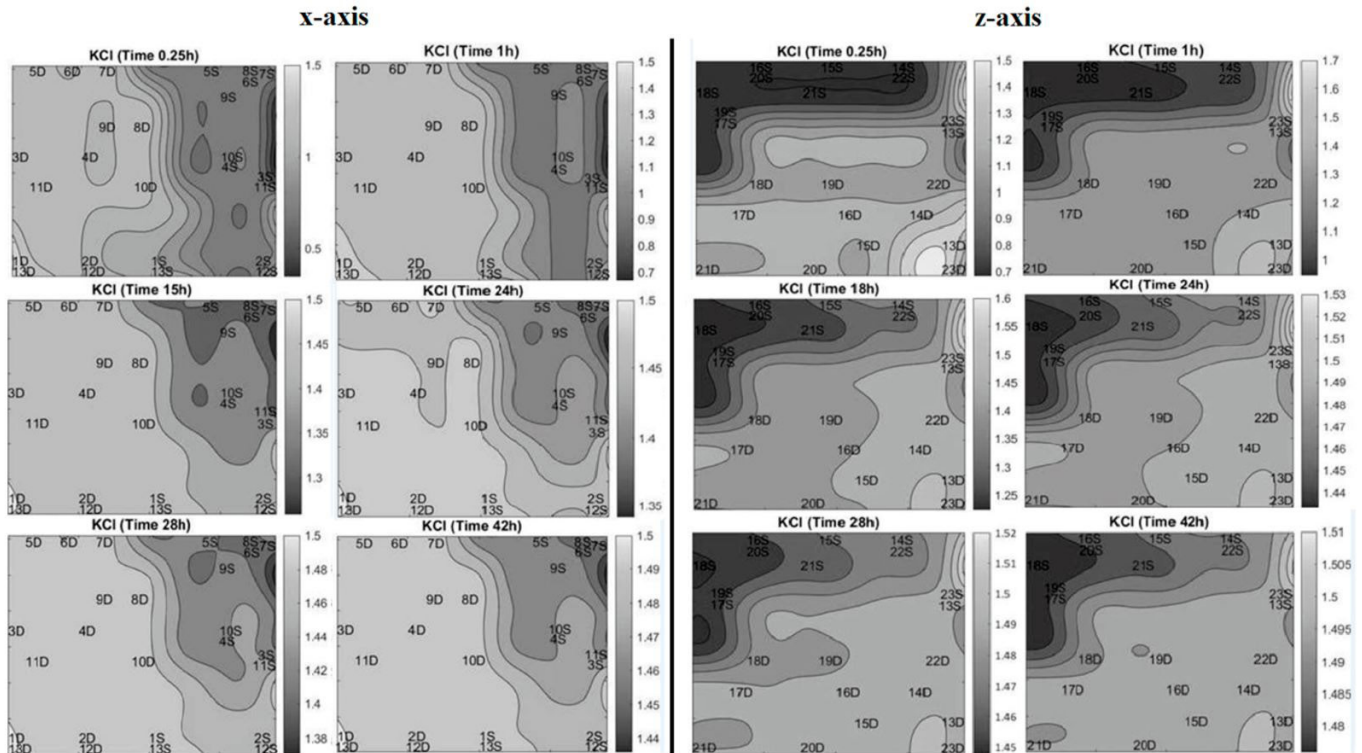


Figure 5. Weight maps of KCl diffusion on the biosolid surface for the x-axis and z-axis in the static (S) and dynamic (D) systems.

indicates that the influence of the film is more pronounced in the static system since the concentrations of species are lower than those obtained for the dynamic, because in this system the brine agitation reduces the film thickness, and consequently the mass transfer is more effective (Cremasco et al., 2019).

Among the studied positions, both in the x and z-axes, as well as in the systems and ions, it was observed that the positions 1, 13 and 23 presented higher concentrations. The biosolid is a rectangular block and the diffusion happens faster in the positions located at the edges of the cheese. Therefore, it is suggested that the film formed is thinner at the edge and thicker at the central external part. In these positions, the time corresponding to 0.25h of salting is sufficient to reach the brine concentration in the dynamic system and only after 28h of salting the static system reaches this equilibrium.

In Figure 4, on average, a difference in concentration is observed as diffusion time increases. The Na⁺ concentration in the x-axis reached 85.71% and 50.00% of brine concentration after 0.25h of salting in the dynamic and static system, respectively. In the z-axis it reached 85.71% and 64.30% in the dynamic and static system, respectively. The Na⁺ reached 97.80% of brine concentration in both axes after 28h and 97.14% after 6h of brine in the static and dynamic systems, respectively.

According to Figure 5, in the x-axis, 96.00% of the K⁺, reached the brine concentration after 28 and 15 hours of salting in the static and dynamic system, respectively. On the z-axis, 97.00% of the K⁺ ions reached brine concentration after 24 hours in the static system and 18 hours in the dynamic system.

4 Conclusions

The optimization of the main, cross and h_m/λ_m coefficients was performed by associating the simplex optimization with the finite element method (FEM). The comparison between the static and dynamic systems showed that the diffusion process is influenced by the film formed at the biosolid/solution interface, being minimized with the brine agitation.

The combination of SOM-type artificial neural networks with FEM simulation proved to be an appropriate tool, as a new application of Prato cheese salting, to evaluate the formation of the film, showing that this phenomenon influences the mass transfer and diffusion time.

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ERRATUM: Application of self-organizing maps to evaluate the influence and behavior of the film formed during salting of Prato cheese

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On page 487, after the Conclusions, it should be read:

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