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FULL LENGTH ARTICLE

Osmotic dehydration and convective drying of coconut slices: Experimental determination and description using one-dimensional diffusion model

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Abstract Mass migrations in coconut slices during osmotic dehydration and drying are described using a diffusion model with boundary condition of the third kind. The osmotic dehydration experiment was performed at 35°Brix (water and sucrose) and 40 °C. The convective drying experiments were performed at 50, 60 and 70 °C. The one-dimensional solution of the diffusion equation for an infinite slab was coupled with an optimizer to determine the effective mass diffusivities *D* and convective mass transfer coefficients *h* of the five processes studied. The analyses of the obtained results indicate that there is a good agreement between each experimental dataset and the corresponding simulation using *D* and *h* determined by optimization.

1. Introduction

Coconut (*Cocos nucifera* L.) is an important agricultural product, which is produced in most countries with wet tropical weather. The coconut palm, also called "life tree", is highly tolerant of salinity and it is found on sandy soils, with abundant sunlight and regular rainfall. In 2011, the main producer countries were Indonesia (17.5 million tons), Philippines (15.2 million tons), India (11.2 million tons), Brazil (2.9 million tons) and Sri Lanka with 1.5 million tons (FAO, 2013). The coconut pulp contains, among other, water, carbohydrates, proteins, fat, salts and also vitamins A, B1, B2, B5 and C. Coconut has the following medicinal properties, among others: anti-inflammatory, soothing, moisturizing, nourishing and antioxidant. The coconut palm is cultivated to produce several products including water from green coconut (Ge et al., 2005; Matsui et al., 2008; Yong et al., 2009), oil (Machmüller et al., 2000; Oliveira et al., 2005; Nevin and Rajamohan, 2006; Benjapornkulaphong et al., 2009) and milk (Oliveira et al., 2005) from mature coconut. Its husk is used to obtain activated carbon (Amuda et al., 2007; Tan et al., 2008; Din

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et al., 2009) or its green coconut shell is used as a natural adsorbent in the removal of toxic metals (Sousa et al., 2007).

In recent years, delicious snacks have been produced from coconut (Lattanzio et al., 2008, 2009). One of these snacks consists, basically, in the removal of water of the coconut pulp, previously cut into small slices (Da Silva et al., 2013a). According to these last authors, after the removal of water from the slices of mature or semi-mature coconut, the snack "dry coconut", cooled up to room temperature, is ready for consumption. However, the use of hot air to remove water of coconut slices is expensive due to the phase change of this substance from liquid to vapor, once its latent heat of vaporization within the product is very high. Thus, in general, a pre-treatment is accomplished before convective drying. An example of inexpensive pre-treatment is the osmotic dehydration (Conceição Silva et al., 2010; Arballo et al., 2012; Da Silva et al., 2013a). This method consists in the removal of water by immersion of pieces of the product in syrup obtained by mixing water with one or more solutes. The difference in the chemical potential of water between the food and the osmotic medium is the driving force for dehydration (Khin et al., 2006).

Water migration in a product can be described through empirical models (Da Silva et al., 2014) but, according to several authors, osmotic dehydration and convective drying can be considered as diffusion phenomena (Amami et al., 2006; Garcia et al., 2007; Singh et al., 2008; Conceição Silva et al., 2010; Da Silva et al., 2013a). Thus, to describe these processes, the diffusion equation should be used. Da Silva et al. (2013a) have described the mass migrations in coconut slices during osmotic dehydration and convective drying, using a diffusive model which considers the three-dimensional nature of the mass fluxes in each individual piece of the product. This model enables to eliminate a restriction frequently found in the literature, which consists in the consideration of the mass flux in only one direction. However, the time spent in the optimization processes to determine the effective mass diffusivity and convective mass transfer coefficient can be considered very high. In this context, taking account that each piece of coconut is a thin slab, the proposal of a quick and precise model to describe the mass migrations in coconut slices deserves to be considered.

The main objective of this article is to describe the mass migrations in coconut slabs during osmotic dehydration and drying processes, using a one-dimensional diffusion model, including optimization and simulation of the process. To this end, an optimizer was coupled with the one-dimensional analytical solution of the diffusion equation with boundary condition of the third kind. Thus, using experimental datasets, the process parameters were obtained and the kinetics of mass migrations can be simulated and analyzed.

2. Materials and methods

2.1. Diffusion equation

Osmotic dehydration and thin-layer drying usually are known as diffusion phenomena (Da Silva et al., 2013a). The one-dimensional diffusion equation for an infinite slab is written as (Luikov, 1968; Crank, 1992)

$$\frac{\partial \Phi}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial \Phi}{\partial x}\right), \tag{1}$$

in which Φ represents water quantity or sucrose gain (osmotic dehydration) and moisture content (drying), in dry basis (db); D is the effective mass diffusivity ($m^2\ s^{-1}$); t is the time (s) and x is the Cartesian coordinate of position (m).

2.2. Analytical solution for the diffusion equation

The convective boundary condition is expressed as follows:

$$-D\frac{\partial \Phi(x,t)}{\partial x}\bigg|_{x=\pm Lx/2} = h\left(\Phi(x,t)|_{x=\pm Lx/2} - \Phi_{eq}\right). \tag{2}$$

In Eq. (2), h is the convective mass transfer coefficient (ms^{-1}); $\Phi(x,t)$ is the value of Φ in a position x at instant t; Φ_{eq} is the equilibrium value for Φ; while L_x is the thickness of the infinite slab (m).

If it is supposed that the thickness L_x does not vary during drying; the initial distribution of Φ is uniform; the infinite slab is considered homogeneous and isotropic; the process parameters are constant during drying, then Eq. (1) can be analytically solved. For an infinite slab with a uniform initial value Φ_0 for Φ and with a boundary condition defined by Eq. (2), the analytical solution $\Phi(x,t)$ of Eq. (1) is given by (Luikov, 1968; Crank, 1992):

$$\Phi(x,t) = \Phi_{eq} + (\Phi_0 - \Phi_{eq})\sum_{n=1}^{\infty} A_n \times \cos\left(\mu_n \frac{x}{L_x/2}\right)\exp\left(-\frac{\mu_n^2}{(L_x/2)^2}Dt\right) \tag{3}$$

in which the origin of the axis x is located at the central point of the slab. In Eq. (3), the coefficient A_n is given by

$$A_n = \frac{4\sin\mu_n}{2\mu_n + \sin(2\mu_n)}, \tag{4}$$

and μ_n are the roots of the characteristic equation

$$\cot\mu_n = \frac{\mu_n}{Bi}. \tag{5}$$

In Eq. (5), Bi is the mass transfer Biot number, given by:

$$Bi = \frac{hL_x/2}{D}. \tag{6}$$

The average value of $\Phi(x,t)$, denoted by $\Phi(t)$, is given in the following way:

$$\Phi(t) = \Phi_{eq} + (\Phi_0 - \Phi_{eq})\sum_{n=1}^{\infty} B_n \exp\left(-\frac{\mu_n^2}{(L_x/2)^2}Dt\right), \tag{7}$$

where the parameter B_n is given by

$$B_n = \frac{2Bi^2}{\mu_n^2(Bi^2 + Bi + \mu_n^2)}. \tag{8}$$

Equation (5) is a transcendental equation which can be solved for a specified mass transfer Biot number. A program in FORTRAN, based on the bisection method, was created and used to calculate the first 16 roots of Eq. (5) for an established mass transfer Biot number. The roots were calculated for 469 Biot numbers from $Bi = 0$ (which corresponds to an infinite resistance of the water flux at the surface) to $Bi = 200$ (which practically corresponds to an equilibrium boundary condition).

2.3. Determination of the process parameters

To determine D and h through the optimization concept, using an experimental dataset, the algorithm developed by Silva et al. (2010) will be adapted for an infinite slab. The objective function to be minimized is given by (Bevington and Robinson, 1992; Taylor, 1997):

$$\chi^2 = \sum_{i=1}^{N_p} [\Phi_i^{\exp} - \Phi_i^{ana}(D, Bi)]^2 \frac{1}{\sigma_i^2}, \tag{9}$$

where $\Phi_i^{\exp}$ is the i^{th} experimental point of the average value of Φ; $\Phi_i^{ana}(D, Bi)$ is the average value of Φ obtained analytically at the same point, given as a function of D and Bi; σ_i is the standard deviation of the experimental average moisture content at the point i; D is the effective mass diffusivity; and N_p is the number of experimental points. For a specified value of Bi, the chi-square depends only on a single variable, namely the effective mass diffusivity. Thus, optimum values of D can be determined for each one of the 469 Bi defined for the infinite slab. The better D is determined by the minimum χ^2 among the 469 minima calculated. Each one of the 469 optimization processes was accomplished as recommended by Silva et al. (2009). According to these authors, for a stipulated value for Bi, $\Phi_i^{ana}(D)$ is calculated in each instant t according to Eq. (7), using the values stipulated for the number of terms (for instance, $n = 16$) and an initial value of D (for instance, 10^{-20}). The values of $\Phi_i^{ana}(D)$ are used to calculate χ^2 according to Eq. (9). Then, the value of D is doubled giving new values for $\overline{\Phi}_i^{ana}(D)$, which are now used to calculate a new χ^2. The value of χ^2 obtained in the first time is compared with the second value obtained for χ^2. If the statement the second χ^2 is lower than the first χ^2 is satisfied, the program will continue the previously described procedure, otherwise the program will finish this procedure because an interval that contains a minimum value for chi-square is found. The last three values for D and χ^2 are then recorded. The recorded interval of D, which contains the minimal value of χ^2, is then divided into n equal parts, and each part of this interval is defined as a step. Thus, the χ^2 of each step is determined, and a more refined interval that contains the minimum is determined. This procedure is repeated until a convergence criterion is satisfied.

2.4. Experimental datasets

Semi-mature coconuts (*C. nucifera* L.) were peeled and the dark film adhered in its external part was removed. The product was cut into pieces parallelepiped shaped with the following average dimensions $L_x = 6.13$, $L_y = 10.12$ and $L_z = 30.65$ mm. For these dimensions, the coconut pieces were considered, in this article, as infinite slabs with thickness L_x, in order to describe the water and sucrose migrations within the product. Thus, the results obtained herein can be compared with those obtained considering the typical three-dimensional solution of the diffusion equation (Da Silva et al., 2013a).

2.4.1. Osmotic dehydration

Coconut slices with initial moisture content $X_0 = 2.33$ ($kg_{water}kg^{-1}_{drymatter}$,db) were submitted to osmotic dehydration at 35°Brix (syrup of water and sucrose), at temperature of 40 °C, and the solution movement was achieved using magnetic stirring with a frequency of 60 Hz. The ratio of the volume of the coconut slices to that of the medium was maintained at 1:20, in order to ensure that the concentration of the osmotic solution does not change significantly during the experiments.

For the osmotic dehydration, the quantities of interest were the water quantity W within the sample during the osmotic dehydration, and the sucrose gain, S. The gravimetric method was used to determine W and S in each instant t during the process, up to the equilibrium value. In each instant t, the weight of a sample was measured, and its dry matter was determined by placing the test sample in a kiln at 105 °C during 24 h. This procedure enables to determine the dry matter at instant t of each sample that continues immersed in the solution. The equations used to determine W and S in each instant t are given, respectively, by

$$W(t) = \frac{m_W(t)}{m_W(0)} \times 100 \tag{10}$$

and

$$S(t) = \frac{m_S(t)}{m_{d0}} \times 100, \tag{11}$$

where $m_W(t)$ and $m_W(0)$ represent the mass of water within the sample at instants t and zero, respectively; $m_S(t)$ is the mass of sucrose transferred to the sample up to the instant t and m_{d0} is its initial dry matter.

The initial and equilibrium sucrose gain were, respectively, $S_0 = 0$ and $S_{\text{eq}} = 8.62\%$ of the initial dry matter. The initial and equilibrium water quantity within the samples was, respectively, $W_0 = 100\%$ and $W_{\text{eq}} = 60.6\%$.

To describe the two processes related to the osmotic dehydration, the generic variable Φ should be replaced by S (sucrose gain) and W (water quantity) in Eqs. (7) and (9).

2.4.2. Drying

After the osmotic dehydration, where about 39% water was removed, thin-layer convective drying processes were performed using hot air at temperatures of 50, 60 and 70 °C. Average moisture contents X were determined by the gravimetric method and the equilibrium moisture contents X_{eq} at 50, 60 and 70 °C were, respectively, 0.45, 0.18 and 0.07 (db). At the end of each drying, the temperature of a kiln was set at 105 °C, and the product remained there for 24 h, enabling the measurement of the dry matter. The dimensionless moisture content X^* was determined through Eq. (12):

$$X^*(t) = \frac{X(t) - X_{eq}}{X_0 - X_{eq}}, \tag{12}$$

in which X_0 is the initial average moisture content.

To use mathematical model proposed in this article, for the drying description, the replacement of the generic variable Φ by the moisture content X or the dimensionless moisture content X^* should be performed in Eqs. (7) and (9). In the dimensionless moisture content case, the following values must be imposed in Eq. (7): $\Phi_0 \equiv X_0^* = 1$ (result of $X = X_0$ in Eq. (12)) and $\Phi_{eq} \equiv X_{eq}^* = 0$ (result of $X = X_{\text{eq}}$ in Eq. (12)).

2.5. General considerations

Since D and h are determined by optimization, Eq. (7) can be used to simulate the process of interest. The analysis of the results was performed using the statistical indicators: determination coefficient R^2 and chi-square χ^2. The optimizer coupled with Eq. (7) was developed in a computer Intel Pentium IV with 2 GB (RAM). The source code was compiled by Compaq Visual Fortran (CVF) 6.6.0 Professional Edition, using the programing option QuickWin Application under the Windows Vista platform. The convergence criterion stipulated for the determination of the effective mass diffusivity was 1×10^{-15}.

During drying, as the thermal diffusivity is much greater than the mass diffusivity, it was considered that the drying process occurs under isothermal conditions. Thus, the effective mass diffusivity as a function of the drying air temperature can be expressed through an Arrhenius-type equation (Nastaj and Witkiewicz, 2009)

$$D = D_0 \exp\left(-\frac{E_a}{R(T + 273.15)}\right). \tag{13}$$

In Eq. (13), D_0 is a pre-exponential factor ($m^2\,s^{-1}$), E_a is the activation energy ($J mol^{-1}$) and R is the universal gas constant (8.314 $J mol^{-1} K^{-1}$). The convective mass transfer coefficient can be also given as a function of the temperature of the drying air by an Arrhenius-type equation:

$$h = a \exp\left[-\frac{b}{(T + 273.15)}\right], \tag{14}$$

where a and b are fitting parameters.

3. Results and discussion

3.1. Osmotic dehydration

Performing the optimization processes for water quantity and sucrose gain, the obtained results are presented in Table 1. This table also presents the results obtained by Da Silva et al. (2013a) which used a three-dimensional approach to describe water and sucrose migration within the coconut pieces.

Since the process parameters have been obtained by optimization, the simulations of water and sucrose migration are presented, together with experimental datasets, in Fig. 1.

3.2. Drying at T = 50, 60 and 70 °C

3.2.1. Optimizations and simulations

For the drying processes at 50, 60 and 70 °C, the results obtained by optimization are presented in Table 2, which also presents the results obtained by Da Silva et al. (2013a) using a three-dimensional approach.

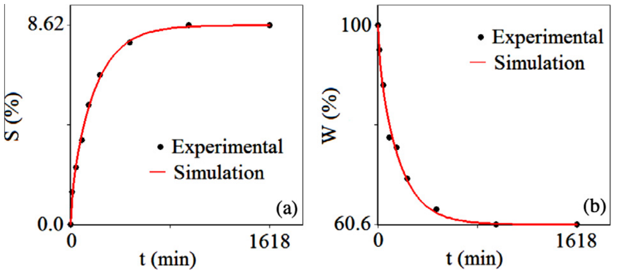

Figure 1 Osmotic dehydration kinetics described by one-dimensional model: (a) Sucrose migration; (b) Water migration.

For drying, once the process parameters have been obtained, the simulations of the water migration are presented, together with experimental datasets, in Fig. 2.

3.2.2. Arrhenius-type equations

Using nonlinear regression to fit Eq. (13) to the data (T,D) given in Table 2, the following result is obtained:

$$D = 1.03 \times 10^{-2} \exp\left(-\frac{5371}{(T + 273.15)}\right). \tag{15}$$

Comparing Eq. (15) with Eq. (13), the activation energy is obtained: $E_a = 44.7\,kJ\,mol^{-1}$. On the other hand, Fig. 3 shows the behavior between the effective mass diffusivity and the drying air temperature.

Using nonlinear regression to fit Eq. (14) to the data (T,h) obtained in this article (Table 2), the following expression is found:

$$h = 5.30 \times 10^{5} \exp\left[-\frac{7799}{(T + 273.15)}\right]. \tag{16}$$

Fig. 4 shows the behavior between h and T in the interval 50–70 °C.

3.3. Discussion

After harvest, the consumption time of coconuts *in natura* is rather limited. In order to avoid losses, coconuts can be submitted to a dehydration process. However, the use of hot air to remove water of coconut slices is expensive due to the phase change of the water. Thus, the osmotic dehydration process is a useful pretreatment which does not involve water phase change. In several countries like Brazil, the osmotic dehydration process (and convective drying) is made in a traditional form, without strong quality control of the final product. This article intends to contribute with information so that the

Table 1 Results for osmotic dehydration using one (present work) and three-dimensional (Da Silva et al., 2013a) solution of the diffusion equation.

	h ($m\,s^{-1}$)	D ($m^2\,s^{-1}$)	Bi	R^2	χ^2
		Present Work			
Water	2.75×10^{-6}	3.92×10^{-10}	21.500	0.9937	1.104×10^{1}
Sucrose	1.75×10^{-6}	3.58×10^{-10}	15.000	0.9983	1.406×10^{-1}
		Da Silva et al. (2013a)			
Water	6.68×10^{-7}	2.64×10^{-10}	7.750	0.9952	8.499×10^{0}
Sucrose	4.41×10^{-7}	2.70×10^{-10}	5.000	0.9971	2.477×10^{-1}

Table 2 Results for thin-layer convective drying using one (present work) and three-dimensional (Da Silva et al., 2013a) solution of the diffusion equation.

T (°C)	h_W (m s^{-1})	D_W (m^2 s^{-1})	Bi	R^2	χ^2
		Present Work			
50	9.92×10^{-7}	6.58×10^{-10}	4.625	0.9928	5.932×10^{-3}
60	3.73×10^{-6}	9.75×10^{-10}	11.750	0.9892	3.005×10^{-2}
70	7.27×10^{-6}	1.65×10^{-9}	13.500	0.9828	3.141×10^{-2}
		Da Silva et al. (2013a)			
50	4.25×10^{-7}	4.83×10^{-10}	2.700	0.9990	3.255×10^{-3}
60	1.30×10^{-6}	6.36×10^{-10}	6.250	0.9929	2.127×10^{-2}
70	2.12×10^{-6}	1.16×10^{-9}	5.625	0.9862	2.503×10^{-2}

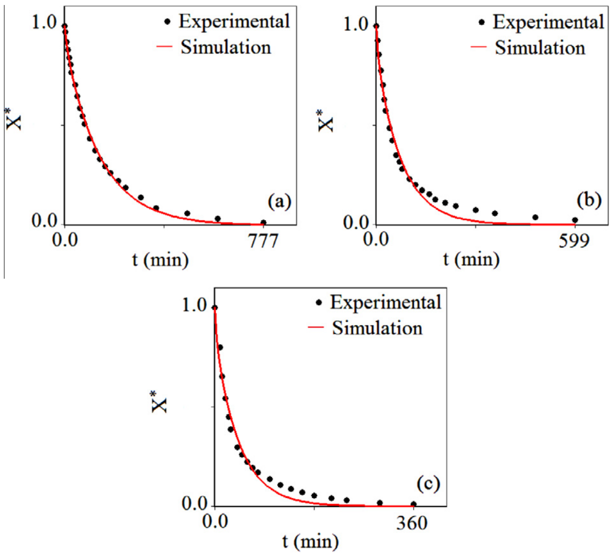

Figure 2 Drying kinetics described by one-dimensional model at T = (a) 50 °C; (b) 60 °C; (c) 70 °C.

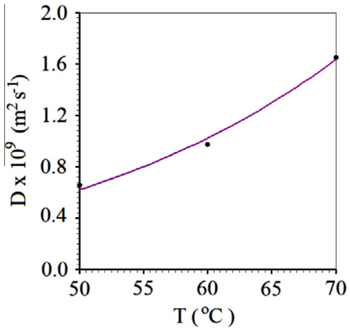

Figure 3 Effective mass diffusivity as a function of the drying air temperature.

osmotic dehydration and convective drying processes of coconuts can be precisely simulated.

In this paper, to minimize cut-off errors, the number of terms of the series that represents the analytical solution of the diffusion equation was established as 16 (Silva et al., 2012). On the other hand, according to Silva et al. (2013), the use of a three-dimensional numerical solution coupled with an optimizer can take several hours to determine D and h. With a three-dimensional analytical solution and the optimization algorithm presented here, this time is reduced to several minutes. When the algorithm is coupled with a one-dimensional analytical solution the time is reduced to few seconds (Silva et al., 2009). For drying at 50 °C, for instance, the optimization time using one-dimensional geometry is about 11.7 s. In the three-dimensional case, this time is multiplied by the factor 193 (2263 s). Thus, the reduction of time justifies the large quantity of works in the literature that describe water migration in porous solids using approximated geometries, generally one-dimensional, and analytical solutions together with some type of optimization algorithm (Cunningham et al., 2007; Ruiz-López and García-Alvarado, 2007; Hacihafizoglu et al., 2008; Mariani et al., 2008).

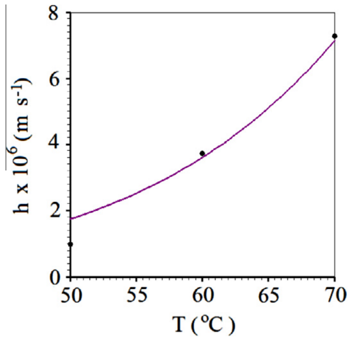

Figure 4 Convective mass transfer coefficient as a function of the drying air temperature.

The use of only the first term of series that represents the analytical solution can lead to significant cut-off errors, particularly in the description of the initial instants of a diffusion process (Da Silva et al., 2013a). Thus, as mentioned above, in this article, 16 terms of the series were used that represent the solution for the boundary condition of the third kind, and the obtained results can be considered good. Regarding the statistical indicators for one- and three-dimensional optimizations, Tables 1 and 2 indicate that one-dimensional model proposed in this article is equivalent to three-dimensional model proposed by Da Silva et al. (2013a). Furthermore, a visual inspection in Figs. 1 and 2 leads to the conclusion that each simulation of the mass migration kinetics agrees with the respective experimental dataset. However, if knowledge of the mass distribution is required at a given time t, the use of a three-dimensional model is more feasible than the use of a one-dimensional model. Anyway, an idea of this distribution in an infinite slab can be obtained through Eq. (3).

An inspection in Figs. 1 and 2 enables to conclude that the five mass migrations occurred exclusively in falling rate period. In Fig. 1, it is interesting to observe that osmotic dehydration was studied for 1618 min. However, in practical terms, the mass transfers have occurred in about of half this time. In Fig. 2, the simulations of the drying kinetics in the final part of the process are only reasonable for 60 and 70 °C. Observing this figure it is possible to conclude that in this final part the effective mass diffusivity should be less than the value obtained. Thus, a variable mass diffusivity could better describe the process, and in this way a numerical solution of the diffusion equation is required. Even so, the values obtained in this article are useful as initial values in new optimization processes.

Coconut slices are, in fact, parallelepipeds. However, in this study, the thickness is significantly lesser than other two dimensions. In this sense, the one-dimensional model is reasonable despite this model overestimating the process parameters (see Tables 1 and 2), due to the consideration of the flux only in the two largest surfaces of the parallelepiped, ignoring the fluxes in the other four smaller areas. Nevertheless, the results obtained for the simulations of the mass migration kinetics are reasonable, and the optimization times to determine D and h are small (only a few seconds).

Once the optimization resource has been used to determine process parameters for some temperatures of the drying air, enabling the obtaining of Eqs. (15) and (16), these optimizations can be substituted by the idea given in the following. For a given temperature between 50 and 70 °C, Eqs. (15) and (16) can be used to calculate D and h, instead the optimization resource, and the obtained results can be used to describe the drying kinetics.

4. Conclusion

(1) The proposed model satisfactorily describes the kinetics of mass transfer.
(2) The process parameters obtained through one-dimensional model are larger than the corresponding parameters obtained with three-dimensional model.
(3) In osmotic dehydration, the value of the Biot number for the water transfer is significantly higher than this value for the sucrose transfer.
(4) The optimization time using one-dimensional model is significantly less than the time for three-dimensional model.
(5) It was possible to determine Arrhenius-type expressions to calculate the parameters D and h as a function of the drying air temperature. The values calculated from the Arrhenius equations make it possible to simulate the drying process in a chosen temperature within the interval 50–70 °C, without the need of new optimizations.

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