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Improvement of methodology for determining local thermal diffusivity and heating time of green coconut pulp during its pasteurization

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ABSTRACT

The objective of this article was to determine local thermal diffusivity of green coconut pulp stored in a parallelepiped-shaped metallic container during heating in a thermal treatment. A thermocouple was placed in the center of the container with the product, initially at 25.8 °C, and then the set was dipped in a water bath at 64.85 °C. The temperature over time was recorded in a txt file, making it possible to determine the parameters of a function that represents the thermal diffusivity, using an optimization technique, which also required a three-dimensional numerical solution of the heat conduction equation. Among the functions tested to represent thermal diffusivity as a function of local temperature, the best result was obtained with the hyperbolic cosine, with chi-square and determination coefficient of 0.3188 and 0.99998, respectively. Thus, it was possible to establish the time required to safely interrupt a new heating process during another heat treatment, with a simple simulation, without the need for a new experiment.

1. Introduction

The green coconut belongs to the species *Cocos nucifera* Linn., Subfamily Cocosidae, which belongs to the Palmae family. The species comprises some varieties, among which the most important in Brazil are Tall (Var. Typica) and Dwarf (Var. Nana). The dwarf coconut tree, introduced in Brazil in 1925, is small and each tree produces 120 to 150 fruits per year (Lavoyer et al., 2013). All parts of the coconut tree can be used by the industry, but the main products are obtained from the fruit, such as copra (6% moisture dehydrated solid albumen), oil, lauric acid, coconut milk, fiber, flour, coconut water (immature fruit), soaps, detergents and cosmetics. Brazil is the 4th largest producer and have harvested about 2 billion fruits per year (FAOSTAT, 2014). The 1st, 2nd and 3rd producers are Indonesia, Philippines and India, respectively.

In Brazil, the consumption of green coconut water (liquid albumen) is so important that it resulted in the planting of crops intended mainly for this purpose. On the other hand, the growing demand for natural and healthy foods is a factor that has increased the consumption of this drink, which reached, in the last years, about 350 million liters per year in both fresh and industrialized forms. Besides being much appreciated for its flavor and freshness, green coconut water is considered an excellent natural isotonic, so it is also consumed for its nutritional qualities. However, the growing demand for this product generates a large amount of waste in places such as beaches of the Brazilian coast where the consumption of this drink is common. Coconut shell accounts for about 85% of the fruit and has become a problem due to the large volume of waste generated and its slow degradation (Santana et al., 2011). To minimize the problem of disposal and increase the profit from selling the product, some Brazilian microentrepreneurs sell not only coconut water but also fleshy albumen. In general, fleshy albumen is transformed into pulp, whose life time is increased when this final product is pasteurized. Thus, to ensure the food safety, the thermophysical properties of green coconut pulp must be accurately determined, enabling not only the simulation of the heating process of the product but also the determination of the minimum time for stopping such process, in order to perform rapid cooling.

Among several methods to determine the thermophysical properties of a product (in particular the thermal diffusivity, α, and the convective heat transfer coefficient, h), one of them involves the measurement of temperature over time for a point chosen within the product during a transient process (heating or cooling). Then, the heat conduction equation is fitted to the experimental data set, making it possible to determine α and/or h (Carbonera et al., 2003; Ukrainczyk, 2009; Betta et al., 2009; Silva et al., 2010, 2011, 2014, 2018; Mohamed, 2015; Muramatsu et al., 2017; Da Silva et al., 2018). As this inverse method

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Nomenclature

A, B	coefficients of the discretized diffusion equation
a, b	parameters that define the local variable thermal diffusivity
c_p	specific heat (J kg^{-1} K^{-1})
$Error_i$	difference between experimental and simulated temperatures for point "i"
f	function that represents the local variable thermal diffusivity
h	convective heat transfer coefficient (m s^{-1})
k	thermal conductivity (W·m^{-1}·K^{-1})
T	temperature (K)
T_P^0	temperature in the control volume P at beginning of a time interval (K)
$T_{\infty e}$	equilibrium temperature in the east boundary (K)
Ti^{exp}	measured temperature for the experimental point "i" (K)
T_i^{sim}	simulated value of the temperature at the point "i" (K)
t	time (s)
x, y, z	position in Cartesian coordinates (m)
α	thermal diffusivity (m^2 s^{-1})
ρ	density (kg m^{-3})
$1/\sigma_i^2$	statistical weight referring to the point "i"
χ^2	chi-square (objective function)

involves a differential equation, and not a simple function, additional difficulties exist in the determination of such properties. If the temperature interval of heating or cooling is small, α may be considered constant during the process, and the heat conduction equation can be solved through analytical methods. An example of this case is given by Silva et al. (2014), who developed an algorithm to determine α, called OREP (Optimal Removal of Experimental Points). The experiment performed by Ukrainczyk (2009) and analyzed by Silva et al. (2014) refers to the heating kinetics of Agar gel (gelatinous water 0.7%) placed within a cylindrical copper container. The container with the product, initially at 20.1 °C, was immersed into hot water at 24.9 °C, and the temperature of the central point of the product was measured over time for 1600 s. Silva et al. (2014) concluded that, for the experiment analyzed, the proposed algorithm produces good results. Obviously, the authors chose an experiment involving a temperature variation of only 4.8 °C and, therefore, the consideration of α with a constant value does not introduce a significant systematic error in the result. Thus, the algorithm of Silva et al. (2014), using an analytical solution of the heat conduction equation, is considered useful in processes in which small temperature variations are involved, as in cases of cooling of agricultural products.

In the case of pasteurization (and other thermal treatments) with temperatures ranging from 15 to 25 to 60–80 °C, the thermophysical properties are expected to vary throughout the process. Nevertheless, several authors consider these properties with constant values (Carbonera et al., 2003; Betta et al., 2009; Mohamed, 2015; Muramatsu et al., 2017), and this fact can introduce systematic errors in the obtained results. This last-mentioned observation was made by Silva et al. (2011) studying heat transfer in tomato purees through a numerical solution of the heat conduction equation, which was developed assuming variable thermal properties. The authors observed that, in each instant, during a transient process, there is a temperature distribution within the product and, because of that, the thermal diffusivity should be given as a function of the local temperature. As Silva et al. (2011) obtained good results with exponential functions to represent local thermal diffusivity, Da Silva et al. (2018), testing several exponential functions, observed that hyperbolic cosine is the best one to represent the thermal diffusivity of papaya pulp. Although there are not many studies available in the literature that consider local values for thermal diffusivity during non-isothermal processes, some studies can be cited. As an example, Greiby et al. (2014) estimated the temperature-dependent thermal conductivity of cherry pomace during non-isothermal heating. Also, Mishra et al. (2016) developed an instrument based on a rapid method to estimate the thermal properties of food dependent on temperatures between 20 and 140 °C. It is interesting to observe that, when thermal diffusivity is precisely known, the time required for heating of a product in a given heat treatment can be easily estimated by a simple simulation. However, regarding the thermal diffusivity of green coconut pulp, no studies were found in the literature on this subject, which makes it difficult to estimate the heating time of this product by simulation. In addition, in our previous work considering papaya pulp (Da Silva et al., 2018), errors and confidence intervals of parameters, as well as the correlation between these parameters, were not reported. Also, residuals were not presented or discussed. Without these statistics, from a rigorous point of view, the parameters obtained can be considered uninterpretable (van Boekel, 1996). Thus, the present study was also carried out to improve the methodology proposed in our previous study, reported in Da Silva et al. (2018).

In this context, this article aimed to determine an expression for thermal diffusivity of green coconut pulp as a function of the local temperature of the product during its heating, using an inverse method. This determination makes it possible to accurately simulate the heating process in a given container, which allows establishing the time required to interrupt such process safely during a thermal treatment.

2. Material and methods

2.1. Experiment: heat treatment, physicochemical and microbiological analyses

2.1.1. Green coconut pulp preparation

The green coconut fruits were purchased from a farm in the municipality of Campina Grande, Paraíba state, Brazil, and transported in plastic boxes to the Agricultural Product Storage and Processing Laboratory (Laboratório de Armazenamento e Processamento de Produtos Agrícolas, LAPPA) at the Federal University of Campina Grande (Universidade Federal de Campina Grande, UFCG). Then, the fruits were inspected, discarding those that were damaged or with any other anomaly, and only the healthy fruits that had the same size and green color pattern were selected. Fruits were then sanitized with a 0.2% sodium hypochlorite solution. The extraction of coconut water (liquid albumen) was done through a household tool, used to pierce the coconut shell, and then a machete was used for longitudinal separation of the fruit into two parts. After that, the fleshy albumen was removed with a stainless steel spoon and then the product was placed on stainless steel trays for subsequent processing in a Mondial brand 500-W power mixer. The pulp was packed in clear plastic containers and transported to UFCG's Heat and Mass Transfer Laboratory (Laboratório de Transferência de Calor e Massa, LTCM) and placed in a Consul freezer at −18 °C. As additional information, this production process followed the Normative Instruction N. 01 of January 7, 2000, issued by the Brazilian Ministry of Agriculture and Supply. This Normative Instruction defines pulp as a non-fermented, undiluted and non-concentrated pasty product, extracted from the edible part of the fruits (Brasil. Ministério da Agricultura e do Abastecimento, 10 jan. 2000).

2.1.2. Green coconut pulp heating

Before heating, the green coconut pulp was removed from the freezer and placed in a parallelepiped-shaped metallic container, with the following internal dimensions, measured with a Digimess 100.170 caliper: 8.01×10^{-2}; 7.14×10^{-2} and 4.95×10^{-2} m. The container wall thickness was about 0.70×10^{-3} m. The product remained for about 10 h on the laboratory bench in a room with temperature maintained close to 26.0 °C by an air conditioner. The temperatures in the center and at the inner wall of the container with the product were monitored by two K-type thermocouples connected to an Instrutherm two-channel TH-095 digital thermometer to ensure the thermal equilibrium of the system. This thermometer can automatically capture data of temperatures over time through its connection to a Core i5 notebook via USB-0/RS-232, using software made available by the manufacturer for the Windows platform. The positions of the above-mentioned thermocouples were chosen for an obvious reason; the container's inner wall temperature is the first to reach thermal equilibrium with the external medium. On the other hand, the central point of the product within the container (the coldest point during heating) is the last one to reach this thermal equilibrium.

After the thermal equilibrium of the product in the container (whose lid was previously sealed with silicone glue), the heating process was performed by water bath, using a Kacil model BM-02C device, with capacity of 9 L of water. This device operates in the range from ambient temperature to 70 °C, with a resolution of 0.1 °C, and it has electronic temperature control (Proportional, Integral and Derivative - PID Controller) as well as shielded tubular resistance, which considerably reduces water heating time, enabling better stabilization of the bath at the desired temperature. In addition to these features, model BM-02C is equipped with magnetic water circulation mechanism, ensuring uniform water temperature.

The heating temperature of the product was set at 65 °C. Thus, after the water temperature reached this value, the container with the product at room temperature was immersed in this hot water, starting the automatic acquisition of time and temperature data every second during the heating process. It is interesting to note that the volume of green coconut pulp is 0.283 L, while the volume of hot water is 9.000 L, roughly 1:32, so it is reasonable to assume hot water as an infinite source of heat.

2.1.3. Physicochemical and microbiological analyses

The following physicochemical analyses were performed in triplicate on both fresh and pasteurized green coconut pulp: determination of ashes by the standard method of calcining the residue of green coconut pulp in a muffle furnace at 550 °C for a period of 24 h; lipid determination by the methodology of Bligh and Dyer (1959); determination of moisture content on wet basis by gravimetric method according to the methodology of Adolfo Lutz Institute (IAL, 2008); fibers (IAL, 2008); and proteins (IAL, 2008). Carbohydrates were estimated by the imposition of 100% for the sum of the six components (ASHRAE, 2006). It is interesting to note that, in addition to the characterization of the product, these analyses on both fresh and pasteurized product allow evaluating the effect of heat treatment on some components.

Regarding microbiological analyses performed on both fresh and pasteurized green coconut pulp, the sum of molds and yeasts, total coliforms and salmonella were analyzed. The analyses were performed in triplicate in a manual colony counter (Phoenix – model CP608) using the method described by Vanderzant and Splittstoesser (1992), and the results were expressed in CFU (Colony Forming Unit)/g and Most Probable Number/g.

Microbiological and physicochemical analyses were performed at the Food Technology Laboratory (Laboratório de Tecnologia de Alimentos, LTA) of the Federal University of Paraíba (Universidade Federal da Paraíba, UFPB).

2.2. Three-dimensional heat conduction equation in cartesian coordinates

2.2.1. Heat conduction equation

Although heat transfer involves convection, radiation and conduction, Da Silva et al. (2018) explain that, for solid and pasty products, with no heat source and no phase change, many times solely a conduction model can be used to describe the process. In this case, the thermal properties involved are considered as "apparent", although in this work the original names of the properties were maintained, with no addition of the term "apparent". In this study, whose experimental part was described above, only the heat conduction equation was used to describe the heating process:

$$\frac{\partial(\rho c_p T)}{\partial t} = \nabla \bullet (k \nabla T) \tag{1a}$$

where ρ (kg m^{-3}) is the density of the product, c_p (J kg^{-1} K^{-1}) is the specific heat, T (K) is the temperature, t (s) is the time and k (W·m^{-1}·K^{-1}) is the thermal conductivity.

Da Silva et al. (2018) argue that, according to empirical equations available in the literature (for instance, in Fricke and Becker, 2001), the product (ρ c_p) varies less than 1% during the heating of fruit pulp from the ambient temperature to 65 °C. Thus, it is reasonable to assume this term with constant value over time, which makes it possible to write Equation (1a) as follows:

$$\frac{\partial T}{\partial t} = \nabla \bullet (\alpha \nabla T) \tag{1b}$$

in which $\alpha = k/(\rho c_p)$ (m^2 s^{-1}) is the thermal diffusivity.

2.2.2. Heat conduction equation in cartesian coordinates and numerical solution

As the container with green coconut pulp is parallelepiped shaped, three-dimensional Cartesian coordinates were used to write Equation (1b). Thus, this equation was written in the following way:

$$\frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(\alpha \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(\alpha \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(\alpha \frac{\partial T}{\partial z}\right) \tag{2}$$

in which x, y and z (m) define position in Cartesian coordinates.

Silva et al. (2013) solved this equation numerically for boundary condition of the third kind, using the Finite Volume Method, with a fully implicit formulation. This solution involves 27 types of control volumes, classified according to the contact of their surfaces with the external medium. Also, the total time of the process was divided by the authors into small time intervals, Δt. As an example, for an internal volume, i.e., with no contact with external medium, the following discretized equation was obtained by integrating Equation (2) in space and time:

$$A_p T_P = A_w T_W + A_e T_E + A_s T_S + A_n T_N + A_b T_B + A_f T_F + B \ , \tag{3}$$

in which the subscripts "p", "w", "e", "s", "n", "b", and "f" represent, respectively, the nodal points P, west, east, south, north, back and front. For an internal control volume, the coefficients A_p, A_w, A_e, A_s, A_n, A_b, A_f and B are given by:

$$A_p = \frac{\Delta x \Delta y \Delta z}{\Delta t} + \frac{\alpha_e}{\Delta x_e}\Delta y \Delta z + \frac{\alpha_w}{\Delta x_w}\Delta y \Delta z + \frac{\alpha_n}{\Delta y_n}\Delta x \Delta z + \frac{\alpha_s}{\Delta y_s}\Delta x \Delta z + \frac{\alpha_b}{\Delta z_b}\Delta y \Delta x + \frac{\alpha_f}{\Delta z_f}\Delta y \Delta x \tag{4a-h}$$

$$A_w = \frac{\alpha_w}{\Delta x_w}\Delta y \Delta z \quad A_e = \frac{\alpha_e}{\Delta x_e}\Delta y \Delta z \quad A_s = \frac{\alpha_s}{\Delta y_s}\Delta x \Delta z \quad A_n = \frac{\alpha_n}{\Delta y_n}\Delta x \Delta z$$

$$A_b = \frac{\alpha_b}{\Delta z_b}\Delta y \Delta x \quad A_f = \frac{\alpha_f}{\Delta z_f}\Delta y \Delta x \quad B = \frac{\Delta x \Delta y \Delta z}{\Delta t} T_P^0 \ .$$

In Equation (4h), T_P^0 is the temperature in the control volume P at beginning of a time interval, Δt, while Δx, Δy and Δz are the edges of the small parallelepiped representing the control volume.

For an east control volume (with only east surface in contact with external medium), the integration of Equation (2) in space and time also results in an algebraic equation given by Equation (3). In addition, the coefficients A_w, A_s, A_n, A_b and A_f are given by the same expressions provided in Equation (4). However, there are the following modifications:

$$A_e = 0$$
$$A_p = \frac{\Delta x \Delta y \Delta z}{\Delta t} + \frac{\alpha_w}{\Delta x_w}\Delta y \Delta z + \frac{\alpha_n}{\Delta y_n}\Delta x \Delta z + \frac{\alpha_s}{\Delta y_s}\Delta x \Delta z + \frac{\alpha_b}{\Delta z_b}\Delta y \Delta x + \frac{\alpha_f}{\Delta z_f}\Delta y \Delta x + \frac{2\alpha_e \Delta y \Delta z}{\Delta x_e} \quad (5a\text{-}c)$$
$$B = \frac{\Delta x \Delta y \Delta z}{\Delta t} T_P^0 + \frac{2\alpha_e T_{\infty e}}{\Delta x_e}\Delta y \Delta z$$

in which $T_{\infty e}$ is the equilibrium temperature in the east boundary of the domain.

Equation (5b,c) are different from the expressions provided by Silva et al. (2013), because the experiment of the present work indicated that the boundary condition of the first kind with constant equilibrium temperature was appropriate to describe the heat conduction process, since the green coconut pulp was stored in a metallic container.

For each type of remaining control volume, similar equations were obtained, and the system of equations in each time step was solved by Gauss-Seidel method, with convergence criterion of 10^{-8}. This procedure enables the temperature to be calculated at the end of each time step, for all control volumes. On the other hand, according to Silva et al. (2011), if the thermal diffusivity α is constant, with no significant volume change during the transient process, the coefficients A are calculated only once. However, the coefficient B is calculated in each time step because its value depends on T_P^0, which is the value of T in the control volume P at the initial instant of each time step. If the property α is variable as a function of temperature, or if the volume change during the transient process is significant, the coefficients A are also calculated in each time step, due to the nonlinearities caused by the variation of volume and/or α. Thus, for an adequate time refinement, the errors due to these nonlinearities can be disregarded.

2.3. Variable thermal diffusivity

In this work, the thermal diffusivity α in a nodal point can be variable, given by an expression of the type

$$\alpha(T) = f(T, a, b) \quad (6)$$

in which f is a mathematical function (constant, polynomial, exponential, etc.), "a'' and "b'' are parameters of this mathematical function that fit the numerical solution of the heat conduction equation to the experimental data set, and such parameters can be determined by optimization. At the interfaces of the control volumes, for example "e'', assuming a uniform grid, the following expression should be used to determine α_e (Patankar, 1980):

$$\alpha_e = \frac{2\alpha_P \alpha_E}{\alpha_P + \alpha_E} \quad (7)$$

where α_P and α_E are values calculated for nodal points P and E using the function established in Equation (6), and α_e is the value of the thermal diffusivity at the interface of these control volumes. In other words, in Equation (7), the subscripts P and E refer to the nodal points of these two control volumes and the subscript e is the interface between them. More detail on the discretization of the diffusion equation in a parallelepiped, such as type of control volumes, first-order approximation for derivatives, descriptive schematics, etc., can be obtained, for instance, in Silva et al. (2013).

2.4. Optimization algorithm – inverse method

The parameters "a" and "b" from Equation (6) were calculated for each function f through the optimization algorithm as suggested by Da Silva et al. (2015), which uses successive attempts. The experimental data set obtained in this research was used in the search for optimum values of the parameters "a" and "b", which minimize the following objective function:

$$\chi^2 = \sum_{i=1}^{N_p} \left(Ti^{\exp} - T_i^{sim}\right)^2 \frac{1}{\sigma_i^2} \quad (8)$$

where $Ti^{\exp}$ is the measured temperature for the experimental point "i", T_i^{sim} is the corresponding simulated value (calculated as shown in Section 2.2), N_p is the number of experimental points, $1/\sigma_i^2$ is the statistical weight referring to the point "i". If the statistical weights are not measured in the experiments, they should be made equal to 1 for all experimental points (same statistical weight). In Equation (8), the chi-square depends on T_i^{sim}, which depends on thermal diffusivity $\alpha = f(T, a, b)$. Thus, the algorithm searches values for "a" and "b", which make Equation (8) have the lowest value possible. In essence, the algorithm follows the following steps:

1) Provide the initial values for the parameters "a" and "b. Solve the diffusion equation and determine the chi-square;
2) Provide the value for the correction of "b";
3) Correct the parameter "b", maintaining the parameters "a" with constant value. Solve the diffusion equation and calculate the chi-square;
4) Compare the latest calculated value of the chi-square with the previous one. If the latest value is smaller, return to the step 2; otherwise, decrease the last correction of the value of "b" and proceed to step 5;
5) Provide the value for the correction of "a";
6) Correct the parameter "a", maintaining the parameters "b" with constant value. Solve the diffusion equation and calculate the chi-square;
7) Compare the latest calculated value of the chi-square with the previous one. If the latest value is smaller, return to the step 5; otherwise, decrease the last correction of the value of "a" and proceed to step 8;
8) Begin a new cycle coming back to the step 2 until the stipulated convergence for the parameters "a" and "b" is reached.

The relative tolerance for convergence of the parameters "a" and "b", in the optimization processes, was established at 10^{-4}. As an additional information, software development (optimizations, simulations, graphical information) was performed on a Samsung notebook with Intel Core i5 processor using FORTRAN language available in Compaq Visual Fortran Professional Studio V. 6.6.0, with the programming option called QuickWin Application.

3. Results and discussion

3.1. Results

3.1.1. Characterization of the fresh green coconut pulp

In order to correctly interpret the results obtained, it is important to know the physicochemical analysis of the fresh product, which is presented in Table 1.

It is interesting to observe that the centesimal chemical composition presented in Table 1 and the formulas provided in ASHRAE (2006) can

Table 1
Physicochemical parameters of fresh green coconut pulp.

Parameter	Average value
Moisture content (wet basis, g/100g)	85.0
Ashes (g/100g)	0.682
Lipids (g/100g)	5.43
Proteins (g/100g)	1.46
Fibers (g/100g)	0.970
Carbohydrates (g/100 g)	6.458

be used to estimate thermal diffusivity at a given temperature. As an example, at 25.8 °C, the formulas for thermal diffusivity provided by ASHRAE (2006) in Chapter 9 for the components (proteins, lipids, carbohydrates, fibers, ashes and water) and the centesimal chemical composition obtained herein for fresh coconut pulp indicated the approximate value of 1.385×10^{-7} m^2 s^{-1}. On the other hand, the value predicted by the Riedel correlation ($\alpha = 8.8\times10^{-8} + 6.0\times10^{-8} X_{wb}$; Riedel, 1969) was 1.390×10^{-7} m^2 s^{-1}, for the moisture content on wet basis of the pulp studied ($X_{wb} = 0.85$).

3.1.2. Models for the thermal diffusivity

Two thermocouples connected to the thermometer have enabled the temperatures to be recorded at the center of the parallelepiped-shaped container and at its internal wall every second during the whole heating process. The result of the experiment can be observed through Fig. 1.

In Fig. 1, it is possible to observe small oscillations of temperature at the internal wall of the container, as a result of the action of the controller, which maintains water temperature close to 65 °C over time (the five last measured values have resulted in an average of 64.85 °C). Also, it is possible to observe that the temperature of this wall (initially at 25.8 °C) assumes, almost instantaneously, its equilibrium value. This observation makes it possible to describe the heating of the green coconut pulp through the diffusion equation, with the boundary condition of the first kind, with an approximately constant temperature of 64.85 °C.

Mishra et al. (2016) showed how to determine the number of experimental points to obtain maximum sensitivity coefficients, such that the addition of any further experimental data would not enhance the accuracy of estimated parameters. In the present article, a simpler argument was used, as is shown in the following. Although the experiment was performed until reaching the thermal equilibrium of the product (64.85 °C, lasting about 6000 s), only the first 3600 s of the transient process were used to determine the thermal diffusivity by optimization. This choice was made for a simple reason: according to the experimental data, the central point, which is the coldest point during the process, has already reached the temperature of 63.1 °C (97.3% of equilibrium temperature), which can be considered satisfactory for the heating stage. As the temperature was recorded every second, only the values obtained every 40 s were used in the optimization processes, reducing the number of experimental points from 3600 to 90. A grid with 48000 control volumes (40x40x30) and also a refinement of 1000 time steps were used to solve the heat conduction equation in each numerical analysis.

Initially, with reference to Equation (6), a constant value for the thermal diffusivity was determined by optimization, and the result is presented in Table 2 (constant function, number 1). This table presents each function that represents the local thermal diffusivity, the parameters of this function (obtained by optimization) and the following statistical indicators: determination coefficient (R^2), chi-square (χ^2) and root mean square error (*RMSE*).

A first observation of the statistical indicators for function 1 suggests that the obtained results can be considered good ($\chi^2 = 3.4781$; $RMSE = 0.1955$ °C and $R^2 = 0.999929$). Thus, a simulation of the transient process with the thermal diffusivity of $\alpha = 1.382 \times 10^{-7}$ m^2 s^{-1}, as well as the experimental data set, are shown in Fig. 2.

As it can be observed, Fig. 2 presents two distinct regions. The first one, for lower times (called region A), has an interesting feature: the simulated values for temperature at center (red line) are higher than the corresponding experimental values. For region B, with higher times, the opposite occurs. To reinforce the observation of this phenomenon, the error of each simulated temperature "*i*" in the center of the container filled with green coconut pulp was defined as:

$$Error_i = Ti^{\exp} - T_i^{sim} \tag{9}$$

in which $Ti^{\exp}$ and T_i^{sim} are the experimental and simulated temperatures at the same time, respectively. Thus, the error distribution for the data set can be given by Fig. 3.

In Fig. 3, three aspects deserve comments. The first aspect is related to the average error in the analyzed period, from 0 to 3600 s. Its value, represented by the continuous red line, is close to 0.02 °C (and the standard deviation is 0.20 °C), although, theoretically, it is easy to show that this average error must be close to zero. The probable cause for a relatively high average error is related to an inappropriate model for thermal diffusivity at temperatures ranging from 26 to 65 °C. The second aspect observed in Fig. 3 refers to the interval that contains the errors: between −0.365 and 0.338 °C, also relatively high. The third aspect, more relevant, is the error distribution itself: completely biased. Fig. 3 (and also Fig. 2) indicates that at the lowest temperatures (region A), the thermal diffusivity must have values lower than the constant value of 1.382×10^{-7} m^2 s^{-1}. Also, at the highest temperatures (region B), the thermal diffusivity must have values higher than 1.382×10^{-7} m^2 s^{-1}. For that, increasing exponential expressions as function of $T^{1/2}$, T and T^2 (functions 2–4 in Table 2, respectively) were used to represent the local thermal diffusivity (according to Equation (6)). In these expressions, T is the local temperature of the transient process. Comparing the statistical indicators for functions 2, 3 and 4, it is observed that the dependence with T^2 (function 4) generated the best results among the functions from 2 to 4. For that, function 5, given by $b\cosh(aT^2)$, which is a combination of two exponentials, was also used to represent the local value of the

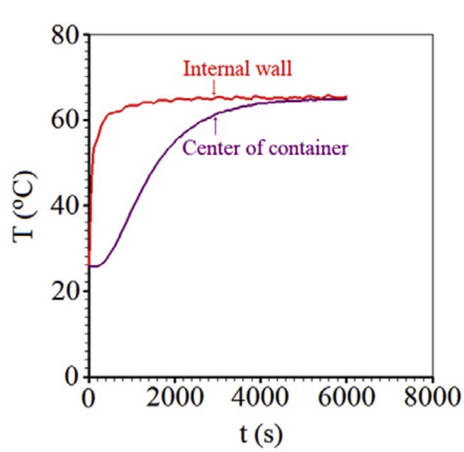

Fig. 1. Temperatures at the center of the parallelepiped-shaped container with green coconut pulp and at its internal wall. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Optimizations: parameters obtained for several functions used to represent the thermal diffusivity of green coconut pulp as a function of the local temperature.

Function number	Thermal diffusivity	a –	$b \times 10^7$ [b] m^2 s^{-1}	R^2 –	χ^2 –	*RMSE* (°C)
1	b	–	1.382	0.999929	3.4781	0.1955
2	$b\exp(aT^{1/2})$	2.901×10^{-2}	1.126	0.999972	0.4592	0.0710
3	$b\exp(aT)$	2.150×10^{-3}	1.241	0.999974	0.4198	0.0679
4	$b\exp(aT^2)$	2.266×10^{-5}	1.302	0.999978	0.3626	0.0631
5[a]	$b\cosh(aT^2)$	$(9.78 \pm 0.36) \times 10^{-5}$	1.3320 ± 0.0035	0.999980	0.3188	0.0592

[a] Function 5 was presented with parameter uncertainties that define confidence intervals of 95.4% and these uncertainties were calculated by LS Optimizer Software.
[b] This column shows the result of parameter b multiplied by the factor 10^7.

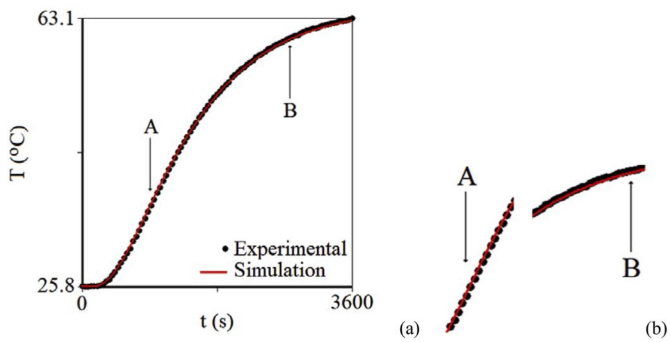

Fig. 2. (a) Experimental data set and simulated curve for the transient process in the center of the container using the constant value (function number 1) for the thermal diffusivity, highlighting regions A and B; (b) Highlight for regions A and B.

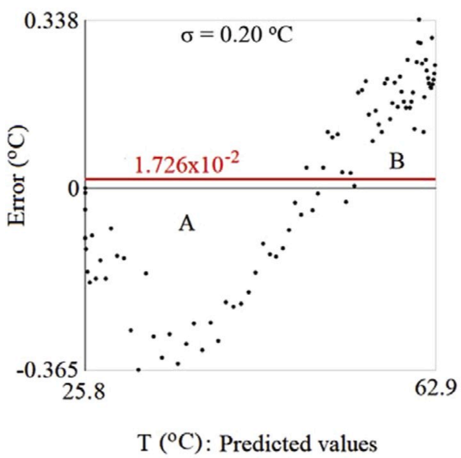

Fig. 3. Error distribution obtained with the simulation performed using the constant value for the thermal diffusivity obtained in this study (Table 2). The continuous red line represents the average value of the errors. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

thermal diffusivity. Among all five expressions, the hyperbolic cosine, given by the function number 5, had the best statistical indicators, as it can be seen in Table 2. For comparison purposes only, the chi-square value for the simulation using the hyperbolic cosine is about 11 times smaller than the chi-square for the simulation using the constant value obtained for thermal diffusivity (function 1).

As the optimization algorithm used in this work does not make it possible to calculate the covariance matrix, a new optimization process was carried out with the diffusivity given by function 5 (Table 2), using the LS Optimizer Software (http://www.labfit.net/LS.htm). LS Optimizer Software, developed by the first author of this article, determines parameters of differential equations through known experimental data, using the least squares method (Levenberg-Marquardt algorithm). The results obtained for the parameters *a* and *b* of function 5 are the same provided in Table 2, but now the covariance matrix is also obtained:

$$\text{cov} = \begin{bmatrix} 3.01496 \times 10^{-20} & -2.96350 \times 10^{-16} \\ -2.96350 \times 10^{-16} & 3.19904 \times 10^{-12} \end{bmatrix} \qquad (10)$$

The covariance matrix given by Equation (10) indicates that the uncertainties of parameters *a* and *b* obtained for the function 5 (Table 2), as well as the correlation between them, are 0.179×10^{-5} °C^{-2}, 0.00174×10^{-7} m^2 s^{-1} and -0.9542, respectively.

A graph using function 5 to represent local thermal diffusivity is presented in Fig. 4, with the simulated curve for the heating over time in the center of the container with green coconut pulp, as well as the experimental points obtained.

As can be seen in Fig. 4, the simulation of the heating in the center of the container with the product, performed with the local thermal diffusivity given by function number 5, describes the transient process well. In addition, the error distribution as well as the histogram frequency versus error for this simulation can be observed in Fig. 5.

In Fig. 5(a), it can be observed that the average error is about −0.003 °C (and the standard deviation is 0.06 °C), indicating that, in absolute value, this average error is about six times smaller than that from Fig. 3. Also, the range containing all errors presented in Fig. 5(a) varies from −0.126 to 0.123 °C, which is much smaller than the interval shown in Fig. 3. Finally, when the error distribution presented in Fig. 3 is compared with that in Fig. 5(a), this latter distribution can be considered random. In addition, the histogram presented in Fig. 5(b) suggests an error distribution very close to a Gaussian distribution. Thus, the expression given by function 5 in Table 2 can really be used to represent the local value of the thermal diffusivity during heating of green coconut pulp.

The behavior of the thermal diffusivity (expressed by hyperbolic cosine, function 5) as a function of the local temperature, as well as a comparison of its average value with the constant value obtained for this property (function 1), is shown through Fig. 6.

Fig. 6 shows that the constant value obtained by optimization (function 1, Table 2) is slightly higher than the average value obtained for function 5 (1.365×10^{-7} m^2 s^{-1}), whose lowest value, at 25.8 °C, is 1.335×10^{-7} and highest value, at 64.85 °C, is 1.446×10^{-7} m^2 s^{-1}. For comparison, the results obtained from the centesimal composition at these same temperatures (25.8 °C and 64.85 °C) were 1.385×10^{-7} and 1.532×10^{-7} m^2 s^{-1}, with discrepancies of 3.3% and 5.6%, respectively.

3.2. Discussion

3.2.1. Heating of the green coconut pulp

As can be seen for the thermal diffusivity, there is some compatibility among the average value of function 5, 1.365×10^{-7} m^2 s^{-1}; the constant value obtained by optimization (function 1), 1.382×10^{-7} m^2 s^{-1}; the value obtained through centesimal chemical composition, 1.385×10^{-7} m^2 s^{-1}; and the value predicted by the Riedel correlation, 1.390×10^{-7} m^2 s^{-1}. However, as observed through Figs. 2 and 3, with their regions A and B, a thermal diffusivity given by a single value for the whole temperature interval is not satisfactory to describe the heating process of the product from 25.8 to 64.85 °C. This result was also found by Silva et al. (2011), describing the heating kinetics of tomato puree stored in a cylindrical container from 5.2 to 77.5 °C, when these researchers also assumed constant thermal diffusivity for the product.

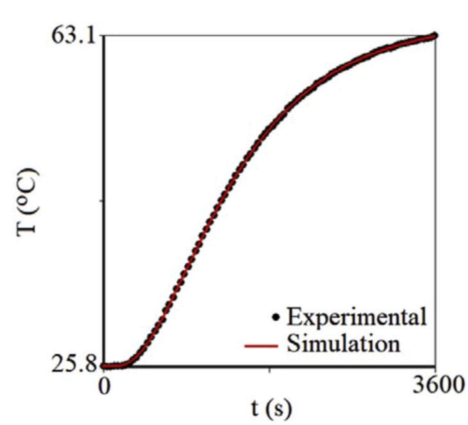

Fig. 4. Experimental data set and simulated curve for the transient process in the center of the container using hyperbolic cosine (function number 5) to represent the thermal diffusivity as a function of the local temperature.

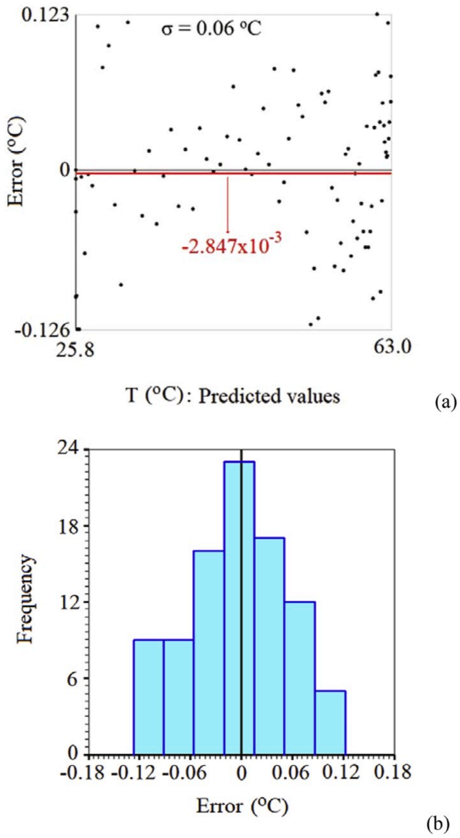

Fig. 5. (a) Error distribution obtained for the simulation using the hyperbolic cosine (function 5) to represent local thermal diffusivity. The continuous red line represents the average value of the errors; (b) Histogram: frequency versus error. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

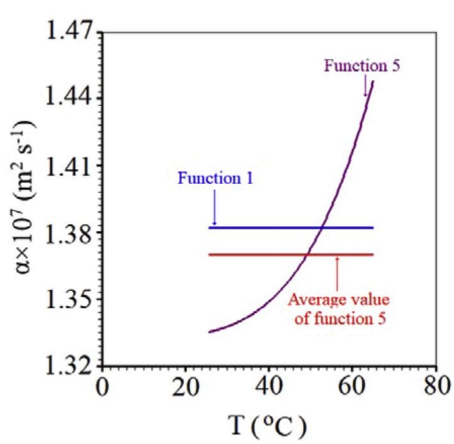

Fig. 6. Thermal diffusivity as function of the local temperature (function 5), its average value, and constant thermal diffusivity (function 1).

Also, a similar result for constant thermal diffusivity was obtained by Da Silva et al. (2018), when studying the heating process of papaya pulp (from 22.4 to 65.4 °C) stored in a container with arbitrary geometry during pasteurization of this product.

As Silva et al. (2011) have observed, it is common to find in the literature the diffusion term written in the form $\alpha\nabla^2 T$ (Baïri et al., 2007; Huang, 2007; Betta et al., 2009; Ukrainczyk, 2009; Mohamed, 2015; Muramatsu et al., 2017; Tanaka et al., 2018), and then this term is discretized. Although this diffusion term makes it possible to use α with a constant value or as function of the average temperature over time (Oke et al., 2007; Kurozawa et al., 2008; Ansari et al., 2009) during a transient state, it is obvious that there is a temperature distribution within the product at each instant. For that, α should be considered as a function of the local temperature for each time step. Thus, a better expression for the diffusion term is $\nabla \bullet (\alpha\nabla T)$, as was done in this article through Equation (1b).

Korese et al. (2017) observed that the major advantage of computer simulations and numerical procedures is their ability to assess the effect of various physical parameters on cooling and heating profiles of fruits and vegetables. These researchers also observed that a drawback of the available modelling tools is that they need a high level of mathematical sophistication for solutions and applications. The authors may be correct because, in fact, it is difficult to find articles in the literature considering thermal diffusivity as function of the local temperature. However, in order to correct the systematic error due to the consideration of a constant value for α (which generates biased error distribution), it was necessary to use these tools, including a numerical solution of the three-dimensional heat conduction equation, which was developed assuming variable thermal properties; and an optimization technique to determine the parameters that define these properties, via inverse method.

It is well known that thermal diffusivity of products in thermal equilibrium increases with increasing equilibrium temperature (Fricke and Becker, 2001; ASHRAE, 2006). Thus, various authors, such as Kurozawa et al. (2008), have determined the thermal diffusivity for products in thermal equilibrium at different temperatures. However, few studies in the literature indicate that, during heating of pulps and purees, i.e., during a transient state, increasing expressions, given as a function of local temperature, well represent thermal diffusivity (Silva et al., 2011; Da Silva et al., 2018). This fact can also be observed in this study through functions from 2 to 5, given in Table 2. As an example, the results obtained in this study for the functions $b\exp(aT^{1/2})$, $b\exp(aT)$ and $b\exp(aT^2)$ are similar to those obtained by Silva et al. (2011), studying heating of tomato puree. However, these last-mentioned researchers did not test hyperbolic cosine to represent the thermal diffusivity of the product. On the other hand, regarding the best result obtained herein, it is interesting to observe that the same expression given as function of the local temperature found in the present article for thermal diffusivity, $\alpha(T) = b\cosh(aT^2)$, was also obtained by Da Silva et al. (2018) studying heating of papaya pulp. These results suggest that this function can be useful to represent the thermal diffusivity of other pulps and purees during heating processes.

In most studies on heating of a product in a drying process, a considerable shrinkage occurs. Thus, in order to describe the heating phenomenon through a diffusion model, the grid referring to numerical solution of the diffusion equation must be corrected over time (Lima et al., 2002; Da Silva et al., 2017). However, for the type of heating studied in this article, with no loss of mass, there is no study in the literature consulted that had measured the volume variation, which is considered negligible. Therefore, in this study, this dimensional variation was not considered either.

3.2.2. Pasteurization of the product stored in a new container

This paper aimed to determine the thermal diffusivity of green coconut pulp accurately, enabling the estimation of the time required for the temperature of the central point of the product stored in a parallelepiped-shaped container to reach a previously established value, without the need for further experiments. It is important to mention that, for a parallelepiped-shaped container, it is very important to know when the previously established temperature in the central point is reached, because it ensures inactivation of pathogens as well as enzyme denaturation at all points of the product. As an example, for green coconut pulp stored in a 5 × 5 × 5 cm^3 new container, a simulation of the heating from 25 to 65 °C (in water bath), using a 40 × 40 × 40 grid for the domain, with time steps of 5 s, makes it possible to obtain the graph of Fig. 7(a). On the other hand, Fig. 7(b) shows the temperature distribution (°C) in 1/4 of the central plane xy of the product at $t = 250$ s.

Fig. 7(a) highlights the times of 1800, 2600 and 3600 s for which the temperatures in the center of the product are 60.8, 64.0 and 64.8 °C, respectively. Thus, with this information, a researcher can decide the instant for removing the container from the hot water. In the present study, the product was removed at $t = 2600$ s, in order to be immersed in a mixture of liquid water and ice for rapid cooling, which completes the thermal treatment. This procedure is expected to ensure food safety, as will be seen in the following. On the other hand, an observation from Fig. 7(b) indicates that at $t = 250$ s, the central region of the product is still at 25.0 °C, while the outermost part has already reached 65 °C. Thus, it would be difficult to physically imagine that regions of the product with such different temperatures could have the same thermal diffusivity, which justifies the idea that thermal diffusivity has local values as a function of temperature (Silva et al., 2011; Da Silva et al., 2018).

The physicochemical analysis of the thermally treated product is presented in Table 3. A comparison between tbl3Tables 3 and 1tbl1 indicates that the thermal treatment did not significantly alter the chemical composition of the green coconut pulp. On the other hand, the results of the microbiological analysis indicated that there was no colony formation for total coliforms and salmonella (at 45 °C) for both fresh and pasteurized product. However, the counting for molds and yeasts resulted in 17000 CFU/g for the fresh product. The Normative Instruction N. 01 of January 7, 2000, issued by the Brazilian Ministry of Agriculture and Supply (Brasil. Ministério da Agricultura e do Abastecimento, 10 jan. 2000), establishes the maximum values of 5000 UFC/g for fresh pulp and 2000 UFC/g for pasteurized pulp. Thus, really the pasteurization process was necessary, resulting in only 200 CFU/g for the pasteurized green coconut pulp, indicating the efficiency of heat treatment.

As a final comment, for this treatment, the D-value resulted in 22.5 min. Obviously, this information enables to establish other heating final time, if another final count is desired.

4. Conclusion

In this article, five expressions were analyzed to represent thermal diffusivity of green coconut pulp: one expression with constant value and four increasing expressions, given as function of local temperature. The worst result to describe the heating process was obtained with constant thermal diffusivity, which showed a completely biased error distribution and inferior statistical indicators compared to the other four results. Three exponential expressions have shown good results to simulate heating of the product, but hyperbolic cosine used to represent thermal diffusivity as function of the local temperature had superior statistical indicators and an error distribution that may be considered random, with absolute frequency versus error close to that of a Gaussian distribution. On the other hand, as it was observed, this study enabled a researcher to decide, through a simple simulation, the instant for removing the container with pulp from the hot water, in order to be immersed in a mixture of liquid water and ice for rapid cooling, which completes the thermal treatment, without the need for a new experiment. For the thermal treatment performed in a 5 × 5 × 5 cm^3 new container, the counting of molds and yeasts resulted in 17000 CFU/g for the fresh product and only 200 CFU/g for the pasteurized green coconut pulp, indicating the efficiency of heat treatment.

Declaration of competing interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

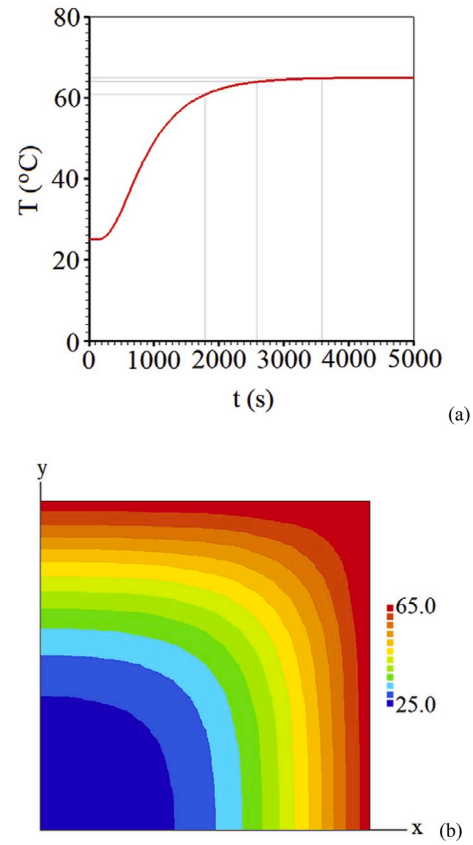

Fig. 7. (a) Heating kinetics simulation in the center of the metallic container (5 × 5 × 5 cm^3) with green coconut pulp; (b) Temperature distribution (°C) in 1/4 of the central plane xy of the product at $t = 250$ s. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 3
Physicochemical parameters of pasteurized green coconut pulp.

Parameter	Average value
Moisture content (wet basis, g/100g)	85.1
Ashes (g/100g)	0.739
Lipids (g/100g)	5.20
Proteins (g/100g)	1.41
Fibers (g/100g)	1.10
Carbohydrates (g/100 g)	6.451

CRediT authorship contribution statement

Wilton Pereira da Silva: Conceptualization, Methodology, Software, Writing - original draft. **Matheus Serrano de Medeiros:** Data curation. **Josivanda Palmeira Gomes:** Supervision, Writing - review & editing. **Cleide Maria Diniz P.S. e Silva:** Software.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jfoodeng.2020.110104.

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