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Drying kinetics and thermodynamic properties of jabuticaba peel

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Abstract

Jabuticaba is a fruit of high nutritional value, and the peel, which is usually discarded, contains nutrients in greater concentration. An alternative for the use of jabuticaba bark is the preparation of flour. The aim of this research was to analyze the drying kinetics of jabuticaba bark by adjusting mathematical models and determining the effective diffusion coefficient, activation energy, and thermodynamic properties at different drying temperatures. The jaboticaba residues were dried in a forced ventilation oven at different temperatures of 45, 55, 65, and 75°C. Among the various models analyzed, Page was employed to streamline the drying phenomenon. The research observed that effective diffusion coefficient decreased with increasing temperature, and the activation energy for the liquid diffusion on drying was 32.63 kJ mol^{-1}. Furthermore, enthalpy and entropy decreased with increasing drying temperature, while Gibbs' free energy increased with increasing drying temperature.

Keywords: activation energy; mathematical modeling; *Myrciaria cauliflora*; page template.

Practical Application: The research on the drying kinetics of jabuticaba bark holds practical applications such as turning waste into valuable resources. Understanding the drying process allows for the efficient development of nutrient-rich flour from the fruit's peel. The flour can be incorporated into various food products, enhancing their nutritional value. Moreover, the study contributes to sustainability by valorizing agricultural waste, showcasing its potential in the food and pharmaceutical industries, as well as in the development of innovative products.

INTRODUCTION

Jabuticaba (Myrciaria cauliflora Berg) is rich in vitamins, anthocyanins, fibers, and minerals; however, these nutrients are in greater concentration in the peel, which is usually discarded (Ferreira et al., 2012).

Jabuticaba marc is a source of bioactive compounds. With its high antioxidant activity, it can be a suitable option, to be added as functional ingredient, in the manufacture of food and animal feed with the consequent reduction of residues from the industrial processing of jabuticaba (Morales et al., 2016).

Jabuticaba has a short commercialization period, as it is a perishable fruit. Right after harvest, it presents a fast change in appearance, due to intense water loss, deterioration, and fermentation of the pulp, resulting from high content of water and sugars (Sato & Cunha, 2007). Baptestini et al. (2016) indicated that drying prolongs the shelf life of foods, reducing microbiological deterioration and degradation reactions as well as facilitating the handling and consumption of fruits.

The kinetics and mathematical modeling of drying are considered important tools in the optimization of the drying process (Leite et al., 2015). The knowledge of the changes that drying induces in the chemical composition of the food, mainly on the nutrients of interest for the specific application of the material, can help in choosing the best drying method (Michalska et al., 2017).

During the drying process, the diffusion coefficient makes it possible to evaluate and compare the drying speed of products with different sizes, shapes, and textures. In addition, by analyzing the dependence of the diffusion coefficient on temperature, thermodynamic indices can be determined, which make it possible to energetically analyze the drying process (Botelho et al., 2015).

Therefore, the objective was to analyze the drying of the jabuticaba bark by adjusting mathematical models and determining the effective diffusion coefficient, activation energy, and thermodynamic properties at different drying temperatures of 45, 55, 65, and 75°C.

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2 MATERIALS AND METHODS

The experiment was developed at the Post-Harvest Laboratory of Vegetable Products of the Federal Institute of Education, Science, and Technology of Goiás–Campus Rio Verde. Jabuticaba residues were used, with an initial moisture content of 3.796 ± 0.018 dry bases, determined in an oven at 105 ± 3°C for 24 h, until constant mass.

The jabuticaba peels were dried in an oven with forced air ventilation, under four temperature conditions: 45, 55, 65, and 75°C, which promoted the average relative humidity of 23.3, 14.2, 8.9, and 5.8%, respectively. Drying continued until the jabuticaba peels reached a constant mass (equilibrium moisture content).

The reduction of moisture content during drying was followed by the gravimetric method (loss of mass), observing the initial moisture content of the product until reaching constant mass. The monitoring of the mass reduction during drying was carried out with the aid of a scale with a resolution of 0.01 g.

The temperature and relative humidity of the ambient air was monitored by means of a data logger, and the relative humidity inside the greenhouse was obtained through the basic principles of psychrometry, with the aid of the computer program GRAPSI.

To determine the moisture content ratios of the jabuticaba bark during drying, the following expression was used (Equation 1):

$$RX = \frac{X - X_e}{X_i - X_e} \tag{1}$$

Where:

RX: moisture content ratio of the product, dimensionless;

X: moisture content of the product (db);

X_i: initial moisture content of the product (db);

X_e: equilibrium moisture content of the product (db).

The mathematical models frequently used to represent the drying of plant products (Table 1) were adjusted to the experimental data of the drying of the jaboticaba peel.

The mathematical models were corrected for the degree of adjustment, the magnitude of the determination coefficient (R^2), the χ^2 test, the error relative mean (P), and standard deviation of the estimate (SE) by means of nonlinear regression analysis using the Gauss-Newton method.

The liquid diffusion model for the geometric shape of a flat plate, with an approximation of eight terms (Equation 13), was adjusted to the experimental data of drying the peel of the jabuticaba, considering the surface area and the volume, according to the following expression:

$$RX = \frac{8}{\pi^2} \sum_{i=0}^{\infty} \frac{1}{(2i+1)^2} exp\left[- \frac{(2i+1)^2 \pi^2 D.t}{4} . \left(\frac{S}{V}\right)^2 \right] \tag{13}$$

Where:

RX: moisture content ratio of the product, dimensionless;

n_t: numbers of terms;

S: product surface area, m^2;

V: product volume, m^3.

Table 1. Mathematical models used to predict drying of plant products.

Model designations	Models	
$RX = 1 + a\,t + b\,t^2$	Wang & Sing	(2)
$RX = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k_1 \cdot t)$	Verma	(3)
$RX = \exp\left(\left(-a-\left(a^2 + 4 \cdot b \cdot t\right)^{0.5}\right) / 2 \cdot b\right)$	Thompson	(4)
$RX = \exp(-k \cdot t^n)$	Page	(5)
$RX = \exp(-k \cdot t)$	Newton	(6)
$RX = a \cdot \exp(-k \cdot t^n) + b \cdot t$	Midilli	(7)
$RX = a \cdot \exp(-k \cdot t) + c$	Logarithmic	(8)
$RX = a \cdot \exp(-k \cdot t)$	Henderson & Pabis	(9)
$RX = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k \cdot a \cdot t)$	Two-term exponential	(10)
$RX = a \cdot \exp(-k_o \cdot t) + b \cdot \exp(-k_1 \cdot t)$	Two terms	(11)
$RX = a \cdot \exp(-k \cdot t) + (1-a) \cdot \exp(-k \cdot b \cdot t)$	Approximation of diffusion	(12)

t: dry time, h; k, k_o, k_1: drying constants h^{-1}; a, b, c, n: model parameters.

The surface area (S) of the jabuticaba peel was calculated according to the Equations 14 and 15:

$$S = \pi . D_g \tag{14}$$

$$D_g = (A.B.C)^{1/3} \tag{15}$$

Where:

D_g: average geometric diameter;

A: length, mm;

B: width, mm;

C: thickness, mm.

The volume and surface area of the jabuticaba peel was determined by measuring the three orthogonal axes (length, width, and thickness) in 15 jabuticaba peel, before drying, with the aid of a digital caliper according to Equation 16:

$$V = \frac{\pi . A.B.C}{6} \tag{16}$$

The relationship between the effective diffusion coefficient and the elevation of the drying air temperature was described using the Arrhenius equation (Equation 17).

$$D = D_0 . exp\left(\frac{-E_a}{R.T_{abs}}\right) \tag{17}$$

Where:

D_o: pre-exponential factor;

E_a: activation energy, kJ mol^{-1};

R: universal gas constant, 8,134 kJ $kmol^{-1}$ K^{-1};

T_{ab}: absolute temperature, K.

The thermodynamic properties of the jabuticaba bark drying process were obtained by the Equations 18, 19 and 20:

$$H = E_a - R.T \tag{18}$$

$$S = R.\left[Ln\,k - Ln\frac{k_B}{h_p}\right] - Ln\,T_{abs} \tag{19}$$

$$G = H - T_{abs} . S \tag{20}$$

Where:

H: enthalpy, J mol^{-1};

S: entropy, J mol^{-1};

G: Gibbs free energy, J mol^{-1};

k_B: Boltzmann constant, 1.38 ´ 10^{-23} J K^{-1};

h_p: Planck constant, 6.626 ´ 10^{-34} J s^{-1}.

3 RESULTS AND DISCUSSION

The different temperatures employed influenced the water loss of the product. Within the first 10 min, a higher moisture content ratio of 0.967 was observed at 45°C, while at 55°C the RX was equal to 0.9472, at 65°C the RX was 0.9388, and at 75°C the RX was 0.9052 (Figure 1A).

The time taken for drying the jabuticaba bark at different temperatures is as follows: at 45 °C, 13 h were spent; at 55°C, 11.83 hours; at 65°C, 7.83 h; and at 75°C, 4.83 h. According to Coradi et al. (2016), the increase in the drying air temperature leads to a higher rate of water removal from

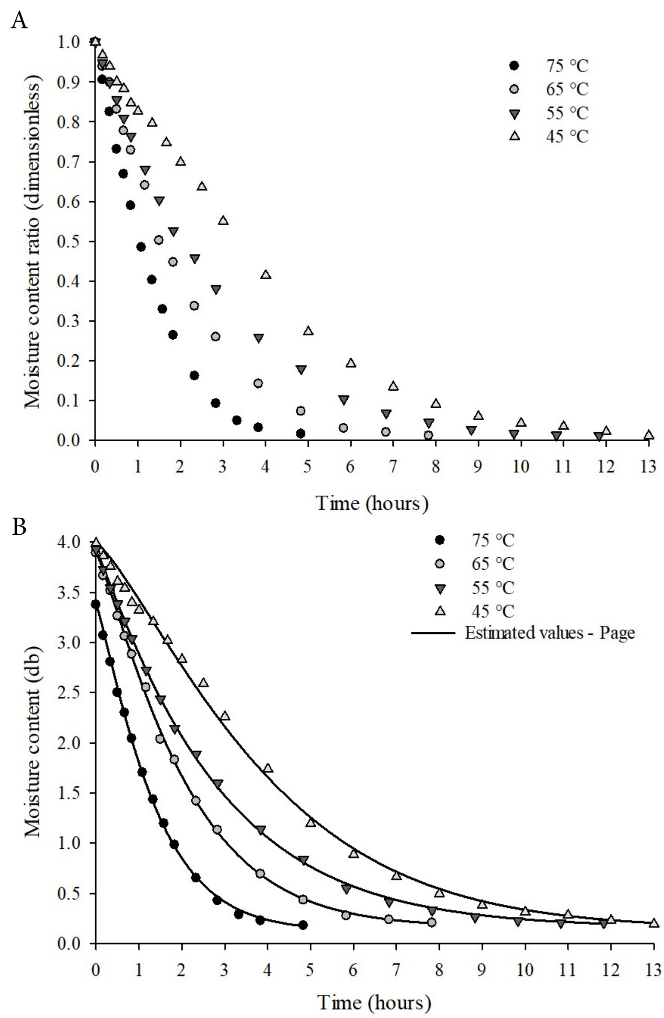

Figure 1. (A) Moisture content ratio of jabuticaba peel (*Myrciaria cauliflora*) over drying time and (B) values of experimental moisture content estimated by the Page model for drying the jabuticaba peel at the temperatures of 45, 55, 65, and 75°C.

the product at the beginning and, consequently, less drying time is required.

Results similar to the drying of the jaboticaba peel were reported by Nunes et al. (2014) when they dried the jabuticaba pulp. They found that the time spent for drying at 50°C was more than twice the time spent at 70°C.

The increase in temperature promoted a decrease in moisture content in a shorter time, which was also verified by Reis et al. (2023) for pineapple peel, Oliveira et al. (2018) for baru mesocarp, and Souza et al. (2019) for pequi mesocarp.

Regarding the model adjustments, the analysis of estimated mean error (SE) and χ^2 using the Verma, Page, Midilli, and two terms models obtained lower values compared to the other models (Table 2). However, the estimated mean error (SE) and χ^2 values are not sufficient to determine the quality of the model fit. Therefore, a joint analysis of the determination coefficient (R^2) and relative mean error (P) was conducted to determine the best model, as shown in Table 3.

It is observed that the models adjusted for all temperatures presented coefficients (R^2) between 98.03 and 99.97%. R^2 value closer to 100% indicates a better fit of the model with experimental data (Sozzi & Ramos, 2015).

In the values of the relative average error (P), it was observed that only the Page and Midilli models presented values below 10% for all drying conditions, making them the most adequate models for representing the drying phenomenon.

The Page and Midilli models were found to be satisfactory under all the temperature conditions evaluated during the drying of jabuticaba peel. These two models showed R^2 values above 99.91% and P values below 10%, indicating a better adjustment to the drying process through the combined analysis of the statistical parameters (R^2, P, SE, and χ^2).

It was found that all the coefficients of the Page model were significant after t-test analysis (Table 4), indicating that they can be used satisfactorily to represent the drying of jabuticaba peel. The coefficients a, k, and n of the Midilli model were also significant, while the coefficient b was not significant.

For the graphical representation of the drying curves, the Page model was used just because it is plain and has a lower number of coefficients compared to Midilli.

Table 2. Values for the chi-square test (χ^2, $\times 10^{-3}$ decimal) and estimated average error (SE, decimal) calculated for the 11 models used to represent the drying kinetics of jabuticaba bark (*Myrciaria cauliflora*).

Models	75°C		65°C		55°C		45°C	
	SE	χ^2	SE	χ^2	SE	χ^2	SE	χ^2
Wang & Singh	0.0256	0.655	0.0321	1.033	0.0525	2.757	0.0253	0.639
Verma	0.0085	0.072	0.0099	0.099	0.0062	0.038	0.0190	0.360
Thompson	0.0310	0.964	0.0324	1.050	0.0150	0.224	0.0484	2.342
Page	0.0100	0.101	0.0101	0.102	0.0072	0.052	0.0199	0.397
Newton	0.0299	0.894	0.0313	0.980	0.0146	0.212	0.0473	2.239
Midilli	0.0090	0.082	0.0105	0.111	0.0068	0.046	0.0158	0.249
Logarithmic	0.0173	0.301	0.0201	0.402	0.0091	0.083	0.0330	1.092
Henderson & Pabis	0.0264	0.697	0.0259	0.669	0.0128	0.164	0.0423	1.788
Two-term exponential	0.0310	0.963	0.0324	1.050	0.0150	0.224	0.0484	2.341
Two terms	0.0084	0.071	0.0103	0.107	0.0080	0.064	0.0168	0.281
Approximation of diffusion	0.0161	0.260	0.0214	0.460	0.0077	0.060	0.0327	1.068

Table 3. Coefficients of determination (R^2, %) and relative average error (P, %) for the models analyzed, during the drying of the jabuticaba bark (*Myrciaria cauliflora*) under different temperature conditions (°C).

Models	75°C		65°C		55°C		45°C	
	P	R^2	P	R^2	P	R^2	P	R^2
Wang & Singh	35.90	99.46	55.82	99.26	84.64	98.03	69.77	99.59
Verma	6.03	99.95	3.59	99.93	5.51	99.97	11.87	99.78
Thompson	28.86	99.21	35.34	99.24	20.15	99.84	78.08	98.48
Page	6.58	99.92	4.01	99.93	7.32	99.96	7.14	99.74
Newton	28.86	99.21	35.33	99.24	20.13	99.84	78.06	98.48
Midilli	8.63	99.94	5.47	99.93	7.88	99.97	4.86	99.85
Logarithmic	19.84	99.77	22.92	99.73	14.40	99.94	62.46	99.33
Henderson & Pabis	23.85	99.43	27.22	99.52	16.68	99.88	63.01	98.84
Two-term exponential	28.86	99.21	35.33	99.24	20.13	99.84	78.06	98.48
Two terms	5.83	99.95	3.64	99.93	12.34	99.96	8.33	99.83
Approximation of diffusion	19.34	99.80	24.03	99.69	12.34	99.96	65.27	99.34

It is possible to select the Page model to represent the drying phenomenon of jabuticaba peel due to its ease of application. Leite et al. (2017) and Santos et al. (2020a) indicated the same model to represent the drying curve of pineapple peels.

Figure 1B shows the values of the experimental moisture content estimated by the Page model for drying the jaboticaba bark under different temperature conditions. It is verified by the correspondence between the experimental and estimated values that the Page model satisfactorily described the drying of jaboticaba peel at the temperatures of 45, 55, 65, and 75°C.

Santos et al. (2020b), in their study on drying the pitomba peel, observed that the Page model was the best fit for the experimental data at the temperatures of 50, 60, and 70°C.

The effective diffusion coefficient decreased with the increase in temperature, with the values of 1.5×10^{-9} m^2 s^{-1} at 45°C; 2.30×10^{-9} m^2 s^{-1} at 55 °C; 3.04×10^{-9} m^2 s^{-1} at 65°C; and 4.71×10^{-9} m^2 s^{-1} at 75°C (Figure 2A). Similar findings were reported by Ferreira et al. (2012), who observed average values of effective diffusivity ranging from 1.0091×10^{-9} m^2 s^{-1} to 3.0421×10^{-9} m^2 s^{-1} for the drying of grape marc between 50 and 90°C, respectively.

The dependence of the effective diffusion coefficient of jabuticaba bark in relation to the temperature and speed of the drying air was adequately represented by the Arrhenius expression (Figure 2B).

The activation energy for the liquid diffusion of jabuticaba peel was found to be 32.63 kJ mol^{-1}, for the temperatures ranging between 45 and 75°C, which was consistent with that reported in the literature. Costa et al. (2016) observed a value of 37.29 kJ mol^{-1} for drying the jabuticaba bark.

Diffusivity can vary due to the chemical composition of the food, initial moisture content, and size and structure of each material, which plays an important role in the rate of water migration from the interior to the product surface, due to a greater or lesser affinity, as well as the structural nature of the material (Khan et al., 2016).

Table 5 shows the thermodynamic properties of enthalpy, entropy, and Gibbs free energy for different air conditions used for drying the jabuticaba peel. Enthalpy decreased with the increasing temperature (45, 55, 65, and 75°C), indicating that less energy was needed for drying at higher temperatures.

Table 4. Parameters of the Midilli model adjusted for the different drying conditions of the jabuticaba bark (*Myrciaria cauliflora*), with respective equation as a function of temperature.

Parameters	**Temperatures (°C)**			
	45	**55**	**65**	**75**
Page				
k	0.15807**	0.324221**	0.400014**	0.665383**
n	1.282048**	1.074624**	1.183366**	1.175819**
Midilli				
a	0.964404**	0.994669**	0.993704**	0.989197**
k	0.127023*	0.318749**	0.392097**	0.645312**
n	1.39825**	1.07558**	1.193930**	1.180682**
b	0.000107 ns	-0.00057ns	-0.000362 ns	-0.00247 ns

*Significant at 5% by t-test; **Significant at 1% by t-test; ns: Not significant by t-test.

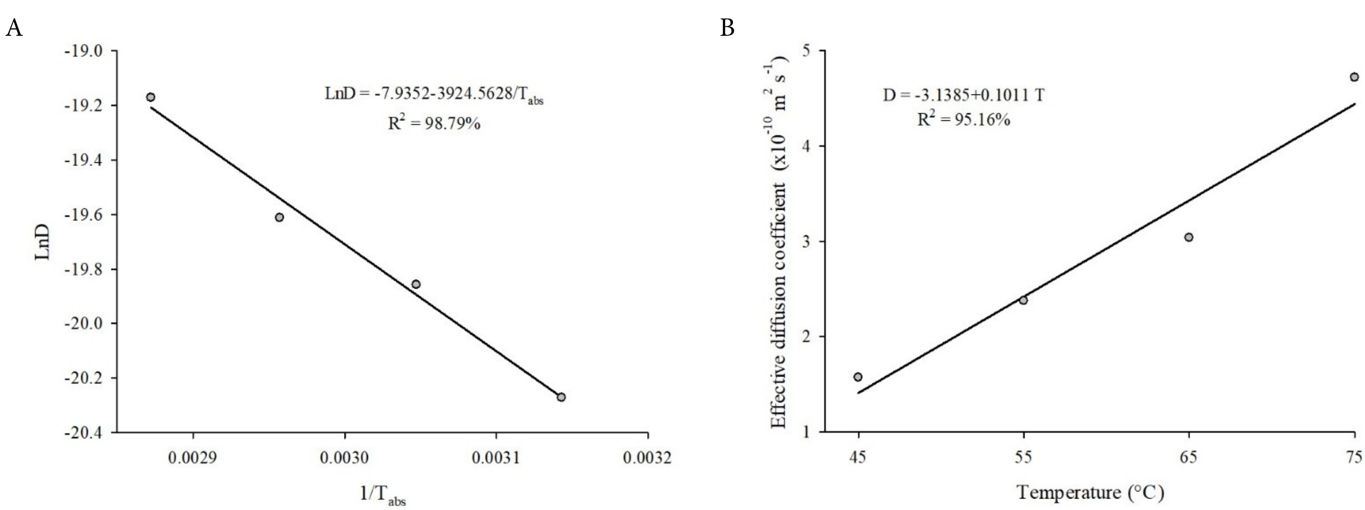

Figure 2. (A) Effective diffusion coefficient and the (B) Arrhenius representation for the effective diffusion coefficient obtained for drying the jaboticaba peel at the temperatures of 45, 55, 65, and 75°C.

Table 5. Enthalpy values (H, J mol^{-1}), entropy (S, J mol^{-1} K^{-1}), and Gibbs free energy (G, J mol^{-1}) for different air conditions of drying the jabuticaba bark.

Temperature (°C)	Thermodynamic properties		
	H	S	G
45	29,984.03	-228.23	102,593.93
55	29,900.89	-228.48	104,877.48
65	29,817.75	-228.73	107,163.56
75	29,734.61	-228.97	109,452.10

The same behavior was observed by Costa et al. (2016) when drying the jabuticaba peel at the temperatures of 40, 50, 60, and 70°C.

Entropy showed negative results and decreased with an increase in temperature from 45 to 75°C. According to Corrêa et al. (2010), negative entropy values occur due to changes in the chemical or structural properties during the drying process.

It has been observed that the Gibbs free energy for drying the jabuticaba bark was positive and increased with increasing drying temperature. Similar behavior was also observed by Rodovalho et al. (2015) for drying goat pepper seed, highlighting that the drying process was not spontaneous, requiring the addition of external energy from the air in which the material was involved so that the moisture content reduction could occur.

4 CONCLUSIONS

The drying time decreased as the temperature increased. Among the studied models, Midilli and Page presented the best adjustments to the experimental data; however, the Page model was selected to represent the drying phenomenon due to the ease of application.

The effective diffusion coefficient decreased with increasing temperature, and the activation energy for the liquid diffusion during drying was found to be 32.63 kJ mol^{-1}. Enthalpy and entropy decreased with increasing drying temperature, while Gibbs free energy increased with increasing temperature.

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