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Modeling and thermodynamic properties of 'bacaba' pulp drying

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ABSTRACT: This study aimed to dry 'bacaba' (*Oenocarpus bacaba* Mart.) pulp under different thermal conditions, fit different mathematical models to the dehydration curves, and calculate the diffusion coefficients, activation energy and thermodynamic properties of the process. 'Bacaba' fruits were meshed to obtain the pulp, which was dried at temperatures of 40, 50 and 60 °C and with thickness of 1.0 cm. Increase in drying temperature reduced the dehydration times, as well as the equilibrium moisture contents, and drying rates of 0.65, 1.04 and 1.25 kg kg min⁻¹ were recorded at the beginning of the process for temperatures of 40, 50 and 60 °C, respectively. The Midilli's equation was selected as the most appropriate to predict the drying phenomenon, showing the highest R², lowest values of mean square deviation (MSD) and χ^2 under most thermal conditions, and random distribution of residuals under all experimental conditions. The effective diffusion coefficients increased with increasing temperature, with magnitudes of the order of 10^{-9} m² s¹, being satisfactorily described by the Arrhenius equation, which showed activation energy (Ea) of 37.01 kJ mol¹. The drying process was characterized as endergonic, in which enthalpy (Δ H) and entropy (Δ S) reduced with the increment of temperature, while Gibbs free energy (Δ G) was increased.

Key words: Oenocarpus bacaba Mart., dehydration, diffusivity

Modelagem e propriedades termodinâmicas da secagem da polpa de bacaba

RESUMO: Objetivou-se secar a polpa de bacaba em diferentes condições térmicas, ajustar diferentes modelos matemáticos às curvas de desidratação e calcular os coeficientes de difusão, a energia de ativação e as propriedades termodinâmicas do processo. Frutas da bacabeira foram despolpadas para a obtenção da polpa que foi secada nas temperaturas de 40, 50 e 60 °C e espessura de 1,0 cm. O aumento da temperatura de secagem reduziu os tempos de desidratação, bem como os teores de água de equilíbrio, sendo registradas, no início do processo, taxas de secagem com magnitudes de 0,65; 1,04 e 1,25 kg kg min⁻¹ para as temperaturas de 40, 50 e 60 °C, respectivamente. A equação de Midilli foi selecionada como a mais adequada para predizer o fenômeno de secagem, apresentando os maiores R^2 , os menores valores de desvio quadrado médio (DQM) e χ^2 na maioria das condições térmicas, e distribuição aleatória dos resíduos em todas as condições experimentais. Os coeficientes de difusão efetivos aumentaram com incrementos de temperatura, apresentando-se com magnitudes na ordem de 10^{-9} m² s⁻¹, sendo descrita satisfatoriamente pela equação de Arrhenius, que apresentou energia de ativação (Ea) de 37,01 kJ mol⁻¹. A secagem foi caracterizada como endergônica, na qual a entalpia (ΔH) e a entropia (ΔS) reduziram com a ampliação da temperatura, enquanto que a energia livre de Gibbs (ΔG) foi aumentada.

Palavras-chave: Oenocarpus bacaba Mart., desidratação, difusividade



Introduction

'Bacaba' tree (*Oenocarpus bacaba* Mart.) is a palm tree that produces fruits called 'bacaba', characterized as edible purple berries which ripen between December and April. Besides containing important constituents for human nutrition and technological processes (Canuto et al., 2010), 'bacaba' has bioactive compounds that are relevant for health maintenance (Finco et al., 2016). Although this fruit has potential for economic exploitation, its high moisture content (Canuto et al., 2010) limits its commercialization to the places of occurrence, making it necessary to adopt preservation techniques in order to offer it to other agroindustrial centers. Among the methods that can be adopted, convective drying has stood out due to its viability (Santos et al., 2013), related to the low cost to perform the procedure, and can also be adopted by small producers.

Thus, studies on 'bacaba' drying will allow a better understanding of the dehydration phenomenon, besides contributing with information to produce a new input in the agroindustry that can also be incorporated in the formulation of various products, such as solid preparations for soft drinks and dairy products. Specialized literature has several studies on fruit drying (Goyal et al., 2006; Doymaz, 2013; Cruz et al., 2015; Oliveira et al., 2015; Alves & Rodovalho, 2016; Johnson & Al-Mukhaini, 2016; Silva et al., 2016; Olawoye et al., 2017; Santos et al., 2017; Deng et al., 2018; Sousa et al., 2018; Taşeri et al., 2018; Wang et al., 2018), but it was not possible to find studies on 'bacaba' drying.

Considering the importance of palm trees in the Amazon region, the present study aimed to dry 'bacaba' pulp at different temperatures, fit different mathematical models to the dehydration curves and calculate the diffusion coefficients, activation energy and thermodynamic properties of the process.

MATERIAL AND METHODS

Ripe fruits of 'bacaba' (*Oenocarpus bacaba* Mart.), 2018 season, from a rural property in the municipality of Xapuri, AC, Brazil, were used. They were transported in plastic boxes to the Food Processing Laboratory, where they were washed in running water, sanitized with chlorine solution (100 mg L^{-1}) for 15 min and rinsed to remove the sanitizing solution. The fruits were cooked in potable water (1:2 m m⁻¹) at temperature of 45 °C for 50 min, to soften the mesocarp, because the material was too firm for handling.

Subsequently, the edible part (epicarp and mesocarp) was manually separated from the seed, disintegrated in an industrial blender until obtaining a homogeneous pulp, which was placed in low-density polyethylene (LDPE) packages and stored in a horizontal freezer at -18 °C. In the drying experiments, 'bacaba' pulp was defrosted under refrigeration (4 °C) and subsequently placed on a laboratory bench until it came into thermal equilibrium with the environment, which was verified using a digital thermometer.

The drying procedures were conducted in triplicate, in a food dehydrator, Pratic Drayer model, at temperatures of 40, 50 and 60 °C and drying air speed of 1.8 m s⁻¹. Each replicate

consisted of about 140 g of 'bacaba' pulp, which was arranged on aluminum tray, in such a way to obtain a thickness of 1.0 cm. The trays containing the samples were subjected to dehydration under the different thermal conditions, and moisture loss was monitored by weighing at regular time intervals, until the samples reached constant mass. The moisture content of the material was quantified after 24 h in the oven at 105 °C. The moisture ratios were calculated using the moisture content at each drying time, according to Eq. 1:

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

where:

RX - moisture ratio of the sample, dimensionless;

X - moisture content, dry basis - d.b.;
X₁ - initial moisture content, d.b.; and,

X - equilibrium moisture content, d.b.

The mathematical models of Henderson & Pabis (Eq. 2), logarithmic (Eq. 3), Midilli (Eq. 4), Page (Eq. 5) and Wang & Singh (Eq. 6) were fitted to the data of 'bacaba' pulp drying at different temperatures (40, 50 and 60 °C) through non-linear regression by the Quasi-Newton method, using the program Statistica version 7.0.

$$RX = a \exp(-kt) \tag{2}$$

$$RX = a \exp(-kt) + c$$
 (3)

$$RX = a \exp(-kt^n) + bt$$
 (4)

$$RX = \exp(-kt^n)$$
 (5)

$$RX = 1 + at + bt^2 \tag{6}$$

where:

k - drying constant;

a, b, c, n - coefficients of the models; and,

t - drying time, min.

The criteria for assessing the goodness of fitting the models were the magnitude of the coefficient of determination (R^2), in addition to the chi-square (χ^2), mean square deviation (MSD), according to the Eqs. 7 and 8, respectively, and the distribution of the residuals.

$$\chi^{2} = \frac{1}{n - N} \sum_{i=1}^{n} \left(RX_{exp,i} - RX_{pred,i} \right)^{2}$$
 (7)

$$MSD = \left[\frac{1}{n} \sum_{i=1}^{n} \left(RX_{pred,i} - RX_{exp,i} \right)^{2} \right]^{\frac{1}{2}}$$
 (8)

where:

 χ^2 - chi-square;

MSD - mean square deviation;

RX_{pred} - moisture ratio predicted by the model;

 RX_{exp}^{r-ca} - experimental moisture ratio;

n - number of observations; and,

N - number of constants of the model.

The drying rates (Eq. 9) of the process of dehydration of the pulp of 'bacaba' under different temperature conditions were calculated from the moisture content (d.b.) at each dehydration time.

$$TX = \frac{X_{t+dt} - X_t}{dt} \tag{9}$$

where:

TX - drying rate, kg kg⁻¹ min⁻¹;

 X_{t+dt} - moisture content at t+dt, kg of water per kg of dry matter;

X. - moisture content at a specific time, d.b.;

 $\label{eq:dt_def} dt \quad \text{-time interval between two consecutive measurements}; \\ and,$

t - time, min.

The liquid diffusion equation with four-term approximation (Eq. 10), considering the geometric shape of the 'bacaba' pulps similar to a flat slab, was fitted to the drying curves under different thermal conditions. Diffusivity was calculated by adopting a uniform distribution of initial moisture and absence of thermal resistance.

$$RX = \frac{X - X_e}{X_1 - X_e} = \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left[-(2n+1)^2 \pi^2 D \frac{t}{4L^2} \right]$$
 (10)

where:

D - effective diffusion coefficient, $m^2 s^{-1}$;

n - number of terms of the equation;

L - characteristic dimension (half thickness of the slice), m; and,

t - time, s.

The Arrhenius equation (Eq. 11) was used to describe the relationship between the effective diffusion coefficients and the drying temperatures of the 'bacaba'.

$$D = D_o \exp\left(-\frac{E_a}{R(T + 273.15)}\right)$$
 (11)

where:

D - effective diffusion coefficients

D_o - pre-exponential factor, m² s⁻¹;

E - activation energy, kJ mol⁻¹;

R - universal gas constant, 0.008314 kJ mol⁻¹ K⁻¹; and,

T - temperature, °C.

Eq. 11 was linearized by applying a logarithm to obtain the parameters of the Arrhenius equation, according to Eq. 12.

$$LnD = LnD_o - \frac{E_a}{R} \cdot \frac{1}{T + 273.15}$$
 (12)

where:

LnD₀ - logarithm of the pre-exponential factor, m² s⁻¹;

E_a - activation energy, kJ mol⁻¹;

R - universal gas constant, 0.008314 kJ mol⁻¹ K⁻¹; and,

T - temperature, °C.

The thermodynamic properties of enthalpy, entropy and Gibbs free energy of the 'bacaba' pulp drying process at different temperatures (40, 50 and 60 °C) were determined using the methodology described by the universal gas constant (Silva et al., 2016), according to Eqs. 13, 14 and 15, respectively.

$$\Delta H = E_a - R (T + 273.15) \tag{13}$$

$$\Delta S = R \left[ln \left(D_o \right) - ln \left(\frac{k_B}{h_p} \right) - ln \left(T + 273.15 \right) \right]$$
 (14)

$$\Delta G = \Delta H - (T + 273.15) \Delta S \tag{15}$$

where:

 ΔG - Gibbs free energy, J mol⁻¹;

ΔH - specific enthalpy, J mol⁻¹;

ΔS - specific entropy, J mol⁻¹ K⁻¹;

 $k_{\scriptscriptstyle B}$ - Boltzmann's constant, 1.38 × 10⁻²³ J K⁻¹;

- Planck's constant, 6.626×10^{-34} J s⁻¹; and,

T - temperature, °C.

RESULTS AND DISCUSSION

Increase of drying temperature caused reduction in the process time, and 1500, 1020 and 660 min at the respective temperatures of 40, 50 and 60 °C were required for 'bacaba' pulp to come into hygroscopic equilibrium with the drying air (Figure 1). The increase of 20 °C reduced the total drying time by 56%, which may represent optimization in subsequent processes in the agroindustry. The average initial moisture content was 128.52% d.b., which was reduced at the end of the process to 4.03, 2.86 and 1.96% d.b. under the thermal conditions of 40, 50 and 60 °C, respectively.

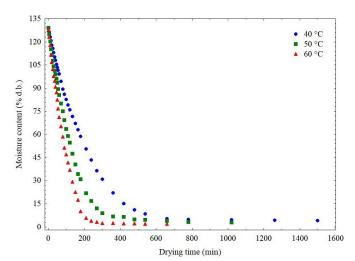


Figure 1. Moisture content recorded as function of drying time of bacaba pulp at temperatures of 40, 50 and 60 °C

It is evident the effect of temperature on 'bacaba' pulp dehydration, where lower moisture content in the sample was observed at the highest temperature and shortest time interval, related to the increase of the energy of water molecules, which allowed greater diffusion from the structure of the material (Sousa et al., 2018). These results are consistent with the behavior observed in the drying of fruits such as pear (Doymaz, 2013), pitaya (Santos et al., 2017) and lemon (Wang et al., 2018).

Despite the occurrence of dispersion in the drying rate values, probably due to fluctuations in the relative air humidity during the dehydration process (Wang et al., 2018), it was observed that it decreased as the moisture content decreased (Figure 2). At the beginning of the drying, the superficial moisture of the product is easily eliminated, which justifies the highest drying rates. However, as the dehydration process continues, the moisture located inside the sample needs to move to the surface to then be removed, resulting in resistance to diffusion and reduction in the drying rate.

It should also be considered that the volumetric shrinking of the sample, common in the drying processes with heated air, promotes changes in the product structure as the drying advances, which culminates in superficial hardening and hence increases the resistance to moisture transfer (Wang et al., 2018). Santos et al. (2014) reported that moisture removal from

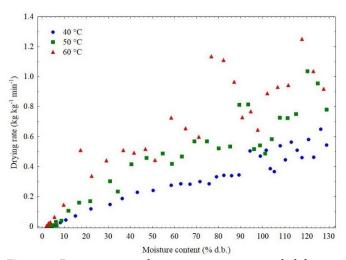


Figure 2. Drying rates and moisture content recorded during the drying of bacaba pulp at temperatures of 40, 50 and 60 °C

granular products occurs from pores and small orifices present in the samples. Therefore, the hardening would promote partial clogging in the water vapor pathways, culminating in the decrease of the drying rate.

In all experiments, the drying rates were higher at the beginning of the process, corresponding to values of 0.65, 1.04 and 1.25 kg of water per kg of dry matter per minute for the temperatures of 40, 50 and 60 °C, respectively, which is due to the high contents of initial moisture (Taşeri et al., 2018). 'Bacaba' pulp drying occurred in a period of decreasing moisture removal rate, and there was no period of constant rate, indicating that mass transfer in the sample occurred through diffusion (Goyal et al., 2006), corroborating with the results of Deng et al. (2018), who found a similar behavior in the drying of red pepper. In the dehydration of pear slices, Doymaz (2013) reported lower values of drying rate than those recorded for 'bacaba' pulp (< 0.05 kg of water per kg of dry matter per minute). By contrast, Ju et al. (2016) reported drying rates exceeding 1.6 kg of water per kg of dry matter per minute in yam slices, and these differences may be related to the physical and chemical characteristics of the plant materials.

The parameters obtained from the fitting of the five mathematical models used to predict the phenomenon of 'bacaba' pulp drying under different temperature conditions are presented in Table 1. The coefficient of determination (R2) ranged from 0.8395 to 0.9991, and only the Wang & Singh equation revealed magnitudes lower than 0.90, which according to Madamba et al. (1996) indicates inadequate fitting to the experimental data of 'bacaba' pulp drying. Regarding the Henderson & Pabis, Logarithmic, Midilli and Page models, their R² values were higher than 0.99, suggesting that these equations are adequate to predict the phenomenon investigated. However, as some authors mention that R² cannot be used as the only statistical parameter for selection and evaluation of nonlinear mathematical models (Goneli et al., 2014; Olawoye et al., 2017; Silva et al., 2017), the mean square deviation (MSD), chi-square (χ^2) and the distribution of residuals (DR) were also considered.

The Midilli model was selected as the most adequate to predict the phenomenon of 'bacaba' pulp dehydration, because, in addition to showing the highest R^2 (> 0.998), it also had the

Table 1. Parameters, coefficients of determination (R^2), mean square deviations (MSD), chi-square (χ^2) and distribution of residuals (DR) of the mathematical models fitted to the kinetic curves of bacaba pulp drying at temperatures of 40, 50 and 60 °C

Model	Temp. (°C)	Model parameters	R ²	MSD	χ² (×10 ⁻⁴)	DR
Henderson & Pabis	40	a: 1.0135; k: 0.0049	0.9977	0.0162	2.7743	В
	50	a: 1.0276; k: 0.0080	0.9955	0.0226	5.4508	R
	60	a: 1.0518; k: 0.0112	0.9914	0.0317	0.7424	В
Logarítmic	40	a: 1.0329; k: 0.0047; c: -0.0240	0.9984	0.0135	1.9973	В
	50	a: 1.0441; k: 0.0076; c: -0.0215	0.9962	0.0208	4.7481	R
	60	a: 1.0791; k: 0.0104; c: -0.0366	0.9936	0.0273	8.2529	В
Midilli	40	a: 0.9825; k: 0.0026; n: 1.1168; b: -0.000005	0.9991	0.0102	1.1678	R
	50	a: 0.9719; k: 0.0027; n: 1.2115; b: 0.000002	0.9989	0.0114	1.4797	R
	60	a: 0.9733; k: 0.0028; n: 1.2864; b: -0.000013	0.9981	0.0148	8.5477	R
Page	40	k: 0.0032; n: 1.0778	0.9988	0.0118	1.4831	R
	50	k: 0.0040; n: 1.1403	0.9983	0.0138	2.0273	R
	60	k: 0.0039; n: 1.2255	0.9976	0.0169	3.0485	R
Wang & Sing	40	a: -0.0027; b: 0.000001	0.8949	0.1102	128.4881	В
	50	a: -0.0041; b: 0.000003	0.8395	0.1359	196.2435	В
	60	a: -0.0060; b: 0.000007	0.8943	0.1115	132.5396	В

lowest values of MSD (< 0.0150) and χ^2 (< 1.50 × 10⁻⁴) under most of the thermal conditions and random distribution of residuals. Additionally, Page equation could also be used satisfactorily in this analysis, since it showed low values of MSD and χ^2 , besides random DR. According to Santos et al. (2017), the mathematical model can only be used to predict drying results if the residuals are random; otherwise it will be inadequate. Doymaz (2013) and Goneli et al. (2014) selected, by fitting different equations to the drying data of pear slices and aroeira leaves, respectively, the Midilli model to describe the dehydration, while Santos et al. (2017) reported that the Page model was the one which best represented pitaya pulp drying. These results corroborate those of the present study, as they demonstrate the proper application of these models in the description of the drying process of agricultural products.

'Bacaba' pulp drying data were fitted with the mathematical model of the liquid diffusion (Eq. 10), which is the analytical solution for Fick's second law, and the effective moisture diffusion coefficients were obtained. These coefficients increased with the increment of temperature, since higher drying temperatures lead to a greater driving force for heat and mass transfer, resulting in the increase of the effective diffusivity of moisture in the sample (Niamnuy et al., 2011). The interdependence of diffusivity with temperature has been reported by several authors (Doymaz, 2013; Santos et al., 2014; Cruz et al., 2015; Olawoye et al., 2017; Deng et al., 2018; Sousa et al., 2018) and is consistent with the results observed in the drying of 'bacaba' pulp.

The values of diffusivity were 0.57, 0.95 and 1.34×10^{-9} m² s¹ for the temperatures of 40, 50 and 60 °C, respectively, which are within the range reported by Madamba et al. (1996) for agricultural products, from 10^{-11} to 10^{-9} m s¹. Cruz et al. (2015) and Ju et al. (2016) also found diffusivity data with magnitude of order of 10^{-9} m s¹, studying the drying of apple slices of the cultivars Golden Delicious and Granny Smith at temperatures from 30 to 60 °C and yam slices at temperatures from 50 to 70 °C, respectively.

The effective moisture diffusion coefficients of 'bacaba' pulp drying, previously linearized, were plotted as function of the inverse of the absolute temperature (Figure 3), and

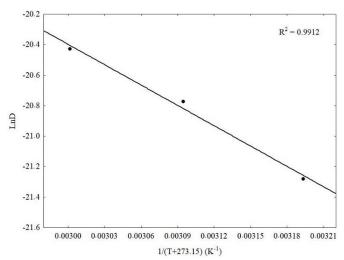


Figure 3. Arrhenius representation for the effective diffusion coefficients obtained in the drying of bacaba pulp at temperatures of 40, 50 and 60 °C

its dependence in relation to the drying air temperature was satisfactorily described by the Arrhenius equation (Eq. 12), which obtained R^2 higher than 0.99.

The activation energy (Ea), which is the minimum energy required to initiate the diffusion of moisture from agricultural products (Olawoye et al., 2017), constituting an important indicator in the evaluation of the energy consumed in the process (Deng et al., 2018), was obtained from the slope of the Arrhenius representation curve (Figure 3), where the dependence between the effective moisture diffusivity and the drying air temperature could be verified by analyzing the coefficients of Eq. 16:

$$D = 8.7307 \times 10^{-4} exp \left(-\frac{4451.400}{T + 273.15} \right)$$
 (16)

It was noted that the drying of 'bacaba' pulp at different temperatures was within the range normally found in processes of dehydration of agricultural products, which is from 12.7 to 110 kJ mol⁻¹ (Zogzas et al., 1996), with a value of 37.01 kJ mol⁻¹. This result is similar to that found by Olawoye et al. (2017) in banana slices (38.46 kJ mol⁻¹). On the other hand, Johnson & Al-Mukhaini (2016) reported Ea of 21.49 kJ mol⁻¹ for strawberry slices, while Goneli et al. (2014) obtained Ea of 74.96 kJ mol⁻¹ in aroeira leaves, considerably dispersed compared to those observed for 'bacaba'. These variations are explained by differences in composition, tissue structure, specific surface area, variety, maturation stage or even pre-treatments applied to the plant material (Deng et al., 2018).

Regarding the thermodynamic properties (Table 2), there was a reduction in the values of enthalpy (ΔH) as the drying temperature increased, and this behavior was also reported by Oliveira et al. (2015) in strawberry, Alves & Rodovalho (2016) in avocado and Guimarães et al. (2018) in okara. Considering that ΔH refers to the energy required to remove the bonded water from the sample during the drying process (Resende et al., 2018), its decrease indicated that, at lower temperatures, the 'bacaba' pulp required more energy for the occurrence of dehydration (Oliveira et al., 2014).

The entropy (Δ S) ranged from -0.3044 (60 °C) to -0.3039 kJ mol⁻¹ K⁻¹ (40 °C), and there was a reduction in this thermodynamic parameter as the temperature increased, related to the reduction in the moisture content along the process of dehydration, which hampered the movement of water molecules through 'bacaba' pulp (Cagnin et al., 2017) due to the reduction of its vibration state (Alves & Rodovalho, 2016), leading to an elevation in the order of the water-product system (Cagnin et al., 2017). The Δ S varied only as a function of the temperature due to the method used in its quantification, considering that all other parameters of Eq. 14 were constant,

Table 2. Thermodynamic properties of bacaba pulp dried at temperatures of 40, 50 and 60 °C

Temp.	ΔΗ	ΔS	ΔG
(°C)	(kJ mol ⁻¹)	(kJ mol ⁻¹ K ⁻¹)	(kJ mol ⁻¹)
40	34.4054	-0.3039	129.56
50	34.3223	-0.3041	132.60
60	34.2391	-0.3044	135.65

 ΔH – Enthalpy; ΔH – Entropy; ΔG – Gibbes free energy

justifying the small variation in this property (Oliveira et al., 2015). Similar results were found by Oliveira et al. (2014) and Silva et al. (2017), when investigating the drying of soybean grains and leaves of *Bauhinia forficata*, respectively.

Gibbs free energy (ΔG), which is related to the total energy required to make the sorption sites of the sample available (Oliveira et al., 2015; Resende et al., 2018), increased from 129.56 to 135.65 kJ mol⁻¹ for the temperature range of 40-60 °C. Its positive values indicate an endergonic reaction, in which the phenomenon of 'bacaba' pulp drying did not occur spontaneously (Silva et al., 2017; Resende et al., 2018), requiring energy from the atmosphere surrounding the sample (Alves & Rodovalho, 2016) in order to make the sorption sites available (Silva et al., 2017). The increase in ΔG as a function of the drying temperature has been observed in several agricultural products, such as avocado pulp (Alves & Rodovalho, 2016), garlic slices (Cagnin et al., 2017) and soybean grains (Oliveira et al., 2014).

Conclusions

- 1. The increase in air temperature reduces the dehydration time, with a reduction of 56% in the total process time when the temperature increases from 40 to 60 $^{\circ}$ C.
- 2. The Midilli model was selected as the most adequate to predict the drying of 'bacaba' under the experimental conditions of this study.
- 3. The drying rates are higher at the beginning of the process, at any temperature evaluated, and decrease with the continuity of the process, as the moisture content of the 'bacaba' pulps decreases.
- 4. The effective moisture diffusion coefficients of 'bacaba' pulp increase with the increment of temperature, and values of the order of 10^{-9} m² s⁻¹ were recorded under all experimental conditions
- 5. The relationship of effective diffusivity with the thermal drying condition is described by the Arrhenius equation, with Ea of $37.01 \text{ kJ mol}^{-1}$.
- 6. The thermodynamic properties of 'bacaba' pulp are affected by drying temperature, with reductions of enthalpy and entropy, and increase in Gibbs free energy, indicating a non-spontaneous endergonic process.

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