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Modeling and thermodynamic properties of the drying of tamarind (*Tamarindus indica* L.) seeds¹

Modelagem e propriedades termodinâmicas da secagem de sementes de tamarindo (*Tamarindus indica* L.)

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HIGHLIGHTS:

For the same moisture content ratio, the time required for drying increases as the drying temperature decreases. The Logarithmic, Page, and Wang and Singh models were not satisfactory for representing the drying kinetic. The activation energy for the liquid diffusion process in the drying of tamarind seeds was equal to 35.46 kJ mol⁻¹.

ABSTRACT: In the present study, the objective was to fit different models to the experimental data of drying of tamarind (*Tamarindus indica* L.) seeds and to select the best model, to determine the effective diffusion coefficient, activation energy and thermodynamic properties for the process during drying at different temperatures. The experiment was carried out at the Laboratory of Post-Harvest of Vegetable Products of the Instituto Federal Goiano (Federal Institute Goiano) – Campus of Rio Verde, GO, Brazil. Seeds with initial moisture content of $18 \pm 0.25\%$ dry basis were oven dried with forced air ventilation, at controlled temperatures of 45, 60, 75 and 90 °C in four repetitions. Nonlinear regression models used to describe the phenomenon were fitted to the experimental data. To represent the drying of tamarind seeds, the Midilli model was selected for the range from 45 to 60 °C and the Two terms model was selected for the range from 75 to 90 °C. The effective diffusion coefficient increases with the increase of drying air temperature, being described by the Arrhenius equation, with activation energy of 35.16 kJ mol⁻¹. Enthalpy and entropy decreases, while Gibbs free energy increases with increasing drying temperature.

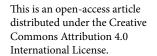
Key words: Two terms Model, Midilli Model, Gibbs free energy

RESUMO: No presente estudo, objetivou-se ajustar diferentes modelos aos dados experimentais da secagem das sementes de tamarindo (*Tamarindus indica* L.) e selecionar o melhor modelo, determinar o coeficiente de difusão efetivo, a energia de ativação e as propriedades termodinâmicas para o processo, durante a secagem em diversas temperaturas. O experimento foi conduzido no Laboratório de Pós-Colheita de Produtos Vegetais do Instituto Federal Goiano - Campus Rio Verde. As sementes com teor de água inicial de 18 ± 0,25% base seca, foram secas em estufa com ventilação de ar forçada, nas temperaturas controladas de 45, 60, 75 e 90 °C, em quatro repetições. Aos dados experimentais foram ajustados a modelos de regressão não linear utilizados para descrição do fenômeno. Para representar a secagem das sementes de tamarindo o modelo de Midilli foi selecionado para a faixa de 45 a 60 °C, e para 75 a 90 °C o modelo ajustado foi o de Dois termos. O coeficiente de difusão efetivo aumenta com a elevação da temperatura do ar de secagem, sendo descrito pela equação de Arrhenius, com energia de ativação de 35,16 kJ mol¹. A entalpia e entropia decrescem, enquanto a energia livre de Gibbs aumenta com o incremento da temperatura de secagem.

Palavras-chave: Modelo de dois termos, Modelo de Midilli, Energia livre de Gibbs

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Introduction

Tamarind (*Tamarindus indica* L.) seed is an agro-industrial residue in the food industry, which has bioactive and nutritional compounds that cause it to be used as a by-product in the food and/or pharmacological industry (Shankaracharya, 1998). Tamarind fruits are harvested with high moisture content for better pulp yield and, consequently, the seeds also, representing a risk in the post-harvest viability of this by-product.

Drying of plant products is the most used process to ensure their quality and stability. The decrease in the moisture content of the material reduces water activity and, consequently, reduces biological activity and chemical and physical changes that occur during storage (Resende et al., 2008).

Drying involves the diffusion of water present in the product and can be described by the effective diffusion coefficient, which is a variable of the liquid diffusion equation that describes the rate of water exit from the product (Resende et al., 2011), describing, through values, the intensity of water transport (Goneli et al., 2007).

Knowledge on thermodynamic properties has been important as a source of information for calculating the energy required in the drying process (Corrêa et al., 2010). Enthalpy, entropy and Gibbs free energy illustrate the interaction of water with the constituents, the spatial arrangement of the water-dry matter relation, and the affinity of the product for water with the constituents of the product, respectively (Jideani & Mpotokwana, 2009; Corrêa et al., 2010).

Knowing the importance of drying to maintain the conservation of products, the objective was to fit different nonlinear regression models to the experimental drying data of tamarind seeds (*Tamarindus indica* L.) and select the best model, determine the effective diffusion coefficient, activation energy and thermodynamic properties for the process, during drying at different temperatures.

MATERIAL AND METHODS

The experiment was conducted at the Laboratory of Post-Harvest of Plant Products of the Instituto Federal Goiano - Campus of Rio Verde, GO, Brazil. Tamarind fruits were collected in the rural area of the municipality of Rio Verde (17° 51' 57" S; 50° 50' 05" W, and altitude of 650 m) and the seeds were separated from the pulp and homogenized. The initial moisture content of the seeds was determined according to Brazil (2009), at 105 ± 1 °C for 24 h, in three replicates containing approximately 15 g of sample.

To obtain the drying kinetics, seeds with initial moisture content of 18 ± 0.25 (% dry basis) were homogeneously arranged on the surface of a circular stainless-steel tray (15 cm diameter) without perforation, in a 2-cm-thick layer. The trays were placed inside the oven with forced air ventilation, at controlled temperatures of 45, 60, 75 and 90 °C, which promoted relative humidity of 17.43, 8.40, 4.34 and 2.39%, respectively, in four replicates containing 150 g of seeds.

The trays were periodically weighed on semi-analytical scales, with resolution of 0.01 g. During weighing, the material was turned over to homogenize the drying process. Drying was

carried out until reaching the equilibrium moisture content, that is, until the mass of the product was invariable for three consecutive weighing procedures under the drying conditions. Temperature and relative humidity were monitored by means of a data logger (LogBox-RHT-LCD), and the relative humidity inside the oven was obtained through the basic principles of psychrometry, with using the computer program GRAPSI.

To determine the equilibrium moisture content of tamarind seeds, approximately 15 g of seeds were placed in aluminum capsules in three replicates. The samples were placed in an oven at temperatures of 45, 60, 75 and 90 °C, and monitored at 24-h intervals until the mass of the product remained invariable for three consecutive weighing procedures. The equilibrium moisture contents of tamarind seeds for temperatures of 45, 60, 75 and 90 °C were 6.6, 3.3, 1.9 and 0.8% d.b., respectively.

After the drying process, the drying curves were obtained from the collected experimental data relating the moisture content ratio along the drying time. Moisture content ratios of up to 0.04 ± 0.06 were used to obtain the drying kinetics of tamarind seeds. The moisture content ratios during drying were determined using Eq. 1:

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

where:

RX - moisture content ratio, dimensionless;

X - moisture content of the product, decimal d.b.;

Xi - initial moisture content of the product, decimal d.b.; and,

Xe - equilibrium moisture content of the product, decimal d.b.

Five mathematical models (Table 1) frequently used to represent the drying phenomenon of plant products (Corrêa et al., 2010; Resende et al., 2018; Botelho et al., 2018) were fitted to the experimental data of moisture content ratio during drying of tamarind seeds.

Table 1. Nonlinear regression models used to predict the drying of tamarind (*Tamarindus indica* L.) seeds

Model designation	Model	
Two Terms	$RX = a \exp(-k_0t) + b \exp(-k_1t)$	(2)
Logarithmic	$RX = a \exp(-kt) + c$	(3)
Midilli	$RX = a \exp(-kt^n) + bt$	(4)
Page	$RX = exp(-kt^n)$	(5)
Wang and Singh	$RX = 1 + at + bt^2$	(6)

t - Drying time, h; k, $\boldsymbol{k_0}, \boldsymbol{k_1}$ - Drying constants; and a, b, c, n - Coefficients of the models

The models were fitted by nonlinear regression analysis by the Gauss Newton method. The significance of their parameters was evaluated by the t test ($p \le 0.01$). The degree of fit of each model was verified based on the magnitudes of the coefficient of determination (R^2), mean relative error (P), mean estimated error (SE), and the Chi-square test (χ^2) ($p \le 0.01$). The mean estimated and relative errors, as well as the Chi-square test for each of the models, were calculated according to Eqs. 7, 8 and 9, respectively:

$$SE = \sqrt{\frac{\sum (Y - \hat{Y})^2}{DF}}$$
 (7)

$$P = \frac{100}{N} \sum_{Y} \frac{\left| Y - \hat{Y} \right|}{Y} \tag{8}$$

$$\chi^2 = \frac{\sum (Y - \hat{Y})^2}{DF}$$
 (9)

where:

Y - experimental value;

 \hat{Y} - value estimated by the model;

n - number of experimental observations; and,

DF - residual degrees of freedom (number of observations minus the number of parameters of the models).

Akaike information criteria (AIC) and Schwarz's Bayesian information criteria (BIC) were used as decisive criteria in choosing the model that best fitted to the experimental data among those which were satisfactory regarding the parameters R^2 , P, SE and χ^2 . AIC and BIC were calculated according to Eqs. 10 and 11, respectively. Lower values for AIC and BIC reflect the best fit (Gomes et al., 2018).

$$AIC = -2 \log like + 2p \tag{10}$$

$$BIC = -2 \log like + p \ln(n)$$
 (11)

where:

p - number of parameters; and,

loglike - logarithm of the likelihood function considering the estimates of the parameters.

The effective diffusivity was determined using the liquid diffusion model for the cylindrical geometric shape, with eight-term approximation (Eq. 12), fitted to the experimental drying data of tamarind seeds.

$$RX = \sum_{\alpha=1}^{\infty} \frac{4}{\lambda_{\alpha}^{2}} \exp\left[-\frac{\lambda_{\alpha}^{2} Dt}{r}\right]$$
 (12)

where:

D - effective diffusion coefficient, m² s⁻¹;

α - number of terms;

 λ_a - roots of the zero-order Bessel equation;

r - equivalent radius, m; and,

t - drying time, h.

The radius of the equivalent sphere was calculated using Eq. 13.

$$r = \sqrt[3]{\frac{3V_s}{4\pi}} \tag{13}$$

where:

V - volume of the seed, m³.

The volume of each seed was obtained by measuring the three orthogonal axes (length, width and thickness), in 15 units,

at the beginning and end of the drying process, using a digital caliper with resolution of 0.01 mm, according to Eq. 14.

$$V_{s} = \frac{\pi ABC}{6} \tag{14}$$

where:

A - length, m;

B - width, m; and,

C - thickness, m.

The relation of the diffusion coefficient with the drying air temperature was analyzed by Arrhenius equation, according to Eq. 15:

$$D = D_0 \exp\left(\frac{-E_a}{RT_{abs}}\right) \tag{15}$$

where:

D₀ - pre-exponential factor, m² s ⁻¹;

E_a - activation energy, kJ mol⁻¹;

R - universal constant of gases, 8.134 kJ kmol⁻¹ K⁻¹; and,

T_{abs} - absolute temperature, K.

The coefficients of Arrhenius expression were linearized with application of the logarithm, as described:

$$LnD = LnD_0 - \frac{E_a}{R} \frac{1}{T_{abc}}$$
 (16)

The thermodynamic properties of the drying process of tamarind seeds were obtained by the method described by Jideani & Mpotokwana (2009):

$$H = E_a - RT_{abs} \tag{17}$$

$$S = R \left(\ln D_0 - \ln \frac{k_B}{h_p} \right) - \ln T_{abs}$$
 (18)

$$G = H - T_{abs}S \tag{19}$$

where:

H - enthalpy, J mol⁻¹;

S - entropy, J mol⁻¹ K⁻¹;

G - Gibbs free energy, J mol⁻¹;

 K_p - Boltzmann constant, 1.38 x 10⁻²³ J K⁻¹; and,

h_n - Planck constant, $6.626 \times 10^{-34} \text{ J s}^{-1}$.

RESULTS AND DISCUSSION

The drying models Two Terms (2), Logarithmic (3), Midilli (4) and Page (5) showed coefficients of determination (R²) higher than 0.99 under all drying conditions (Table 2). The Wang and Singh model (6) had lower values for all temperatures under study, compared to the others. However, this criterion should not be the only one used for selecting

Table 2. Coefficient of determination (R^2), Chi-square (χ^2 , decimal x10⁻⁴), mean relative error (P, %), mean estimated error (SE, decimal x10⁻⁴), Akaike information criteria (AIC) and Schwarz's Bayesian information criteria (BIC) of the models fitted for the drying of tamarind (*Tamarindus indica* L.) seeds at temperatures of 45, 60, 75 and 90 °C

Model	χ²	R ²	Р	SE	AIC	BIC		
	45 °C							
Two Terms	0.59	0.9992	2.06	0.52	-545.76	-533.85		
Logarithmic	3.55	0.9954	5.69	3.11	-403.58	-394.05		
Midilli	0.35	0.9995	1.84	0.31	-587.22	-575.31		
Page	0.40	0.9995	1.99	0.35	-579.06	-571.92		
Wang and Singh	50.90	0.9325	21.91	44.93	-191.48	-184.34		
	60 °C							
Two Terms	0.26	0.9997	2.49	0.22	-582.12	-570.46		
Logarithmic	3.21	0.9964	13.51	2.74	-390.85	-381.53		
Midilli	0.22	0.9998	2.38	0.19	-594.05	-582.39		
Page	0.36	0.9996	2.68	0.31	-559.14	-552.15		
Wang and Singh	104.40	0.8809	73.69	89.80	-127.07	-120.08		
	75 °C							
Two Terms	0.17	0.9998	2.89	0.13	-516.75	-505.96		
Logarithmic	2.97	0.9967	12.16	2.32	-333.15	- 324.52		
Midilli	0.78	0.9991	5.47	0.61	-417.55	-406.75		
Page	42.17	0.9975	10.33	1.71	-354.24	-347.77		
Wang and Singh	116.55	86.80	74.61	91.77	-99.34	-92.46		
	90 °C							
Two Terms	0.11	0.9999	2.13	0.07	- 381.63	- 372.59		
Logarithmic	2.59	0.9975	16.79	1.68	-239.02	-231.79		
Midilli	1.38	0.9987	9.16	0.88	-266.57	-257.54		
Page	3.38	0.9966	14.87	2.22	-227.94	-222.52		
Wang and Singh	129.48	0.8701	90.51	84.90	-63.95	-58.53		

regression models (Madamba et al., 1996), because, alone, it is not a good index to select nonlinear models as it uses mean of negative and positive values, which may make the values of the fits more discrepant.

The chi-square (χ^2) values for the obtained experimental data ranged from 0.11 to 129.48 x 10^{-4} (Table 2), being lower than the tabulated value (53.54), indicating adequate fit to the experimental data. The models Two Terms (2), Midilli (4) and Page (5), for all drying treatments, had the lowest values of chi-square (χ^2) compared to the other models fitted.

The mean relative error considers the deviation of the experimental values from the data estimated by the model. Therefore, the increase in P values indicates greater differences between observed and estimated values (Kashaninejad et al., 2007). Mohapatra & Rao (2005) report that values above 10% for the mean relative error (P) make the model inadequate for the description of the drying process.

Therefore, the Logarithmic (3), Page (5), and Wang and Singh (6) models were disregarded to represent the drying kinetics at all temperatures studied, because they did not show satisfactory values of the fitting indices for at least one temperature (Table 2). The Two Terms (2) and Midilli (4) models obtained for all drying conditions mean relative errors below 10%, indicating satisfactory fit to the experimental conditions.

The Wang and Singh (6) model had the highest values of mean estimated error (SE) (Table 2), while Two Terms (2) and Midilli (4) models had the lowest values. Draper & Smith (1998) argue that the ability of a model to predict the physical phenomenon, in this case drying, is inversely proportional to the value of the mean estimated error (SE).

As it is necessary to apply fitting indices to help choose the best model, it was decided to use in this study the AIC and BIC indices, as Gomes et al. (2018) used as decisive criterion in their study on the drying of crushed mass of jambu (*Acmella oleracea*) leaves.

As the results obtained for mean relative error (P) and mean estimated error (SE), the lowest values of AIC and BIC (Table 2) were verified for the Midilli model (4) at drying temperatures of 45 and 60 °C, and the Two Terms model (2) at temperatures of 75 and 90 °C. Thus, the AIC and BIC indicate the Midilli model (4) as the most suitable for the drying temperature range from 45 to 60 °C, while for the range from 75 to 90 °C the best fit is represented by the Two Terms model (2).

By analyzing the experimental and estimated data (Figure 1), it can be noted that the Midilli model (4) is accurate to estimate the drying of tamarind seeds at the lowest temperatures (45 and 60 °C), as well as the Two terms model (2) for temperatures of 75 and 90 °C. Therefore, the two models were used to fit the drying curves of tamarind seeds.

It is noted that, for the same moisture content ratio, the time required for drying increases (Figure 1) as the drying temperature decreases. The time spent in this process is shorter with the increase in the drying temperature. Similar results are found in the literature for the drying of quinoa (*Chenopodium quinoa* W.) grains (Moscon et al., 2017).

The Midilli and Two Terms models can be used to represent the drying kinetics of baru (*Dipteryx alata* Vog.) fruits, according to a study conducted by Resende et al. (2018) at temperatures of 40, 60, 80 and 100 °C. The Midilli model fitted best to the experimental data of drying of different cultivars of soybean (*Glycine max*) (Botelho et al., 2018). In other studies (Martins et al., 2015; Moscon et al., 2017; Morais et al., 2019), the Two Terms model is recommended to represent the drying curves of various products (Chen et al., 2015; Araujo et al., 2017). Table 3 shows the values of the parameters of the models.

The values of the parameters of the Midilli model (4) for temperatures of 45 and 60 °C increased as the drying air temperature increased (Table 3). The constant k is related to the effect of temperature on the effective diffusivity in the drying

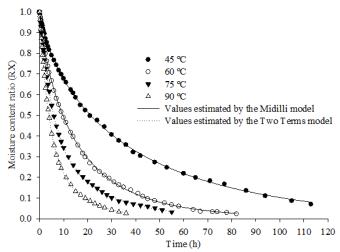


Figure 1. Experimental values estimated by the Midilli model for the drying of tamarind (*Tamarindus indica* L.) seeds at temperatures of 45 and 60 $^{\circ}$ C, and by the Two Terms model for temperatures of 75 and 90 $^{\circ}$ C

Table 3. Parameters of the Midilli model (45 and 60 °C) and Two terms model (75 and 90 °C) fitted for the drying of tamarind (*Tamarindus indica* L.) seeds

	45	60	75	90			
Parameters	(°C)						
	Midilli		Two terms				
a	1.01130**	1.016989**	0.412621**	0.349731**			
k	0.07369**	0.121819**		-			
k_0			0.045400**	0.069354**			
b	-0.000101*	0.000026 ^{ns}	0.579675**	0.661968**			
k₁			0.192789**	0.272553**			
n	0.73680**	0.777702**					

 $^{**,*,\,ns}$ - Significant at $p \leq 0.01,$ at $p \leq 0.05$ and not significant, respectively, by t-test

process, and the liquid diffusion controls the process (Babalis & Belessiotis, 2004), that is, as the magnitude of this constant increases due to the increase in drying temperature, there is an increase in the effective diffusivity.

For the Two Terms model (2), the drying constants k_0 and k_1 obtained increasing values for temperatures of 75 and 90 °C (Table 3); these constants are theoretically related to the diffusivity of water in the product, and by analyzing the equation of this model, it can be noted that the constants are related to the drying time, as the Logarithmic (3), Midilli (4) and Page (5) models. The coefficients a and b of the Two Terms model are coefficients of the equation that are not related to the drying conditions, so it is not possible to infer about their behavior with the change in drying temperature.

The values of the effective diffusion coefficient increase with the increase in drying air temperature (Figure 2A), a behavior observed by other researchers (Rodovalho et al., 2015; Martins et al., 2015). The increment in drying air temperature increases the level of vibration of water molecules inside the seeds, causing a reduction in water viscosity and consequently reducing the resistance of the fluid to the flow (Goneli et al., 2014).

The effective diffusion coefficient for the drying of tamarind seeds follow an increasing linear trend in response to the increase in drying temperature, with values of 2.4044 x 10^{-11} , 5.4903 x 10^{-11} , 8.5277 x 10^{-11} and 1.3025 x 10^{-10} m² s⁻¹ for

temperatures of 45, 60, 75 and 90 °C, respectively. Martins et al. (2015) obtained for timbó (*Serjania marginata* Casar) leaves at temperatures of 40, 50, 60 and 70 °C values of 0.6630×10^{-11} , 5.1229×10^{-11} , 7.0289×10^{-11} and 12.0712×10^{-11} , respectively.

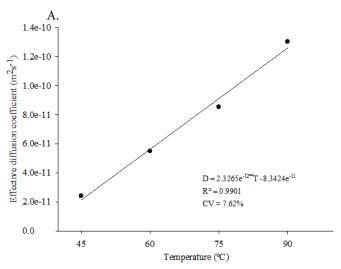
The difference between the values of the effective diffusion coefficient can be justified because the chemical composition and physical structure differ from one product to another, making the loss of water specific to each material. Studies on drying involving water diffusion show variations in the values of the effective diffusivity coefficient, due to the complexity of plant products, different prediction methods, type of material, moisture content, drying process, as well as the methodology used to obtain (Goneli et al., 2007).

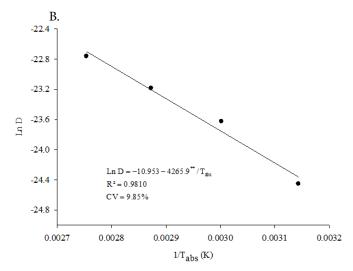
The effective diffusion coefficient range found under the studied conditions for tamarind seeds follows the same trend as other plant products (Siqueira et al., 2012; Costa et al., 2015; Resende et al., 2018). According to Madamba et al. (1996), the values of the effective diffusion coefficient for the drying of plant products are of the order of 10^{-9} to 10^{-11} m 2 s $^{-1}$.

The dependence of the effective diffusion coefficient of tamarind seeds on drying air temperature was represented by the Arrhenius expression (Figure 2B). The activation energy for the liquid diffusion process in the drying of tamarind seeds for the studied temperature conditions (45; 60; 75 and 90 °C) was equal to 35.46 kJ mol⁻¹.

Corrêa et al. (2007) point out that activation energy is the ease with which water molecules overcome the energy barrier during migration inside the product. Lower values for activation energy indicate higher water diffusivity in the product per unit of time (Kashaninejad et al., 2007).

For liquid diffusion to occur during the thin-layer drying of peanuts (*Arachis hypogaea* L.) at temperatures of 40, 50, 60 and 70 °C, the activation energy was 35.24 kJ mol⁻¹ (Araujo et al., 2017). The activation energy for the liquid diffusion of jatropha (*Jathopha curcas* L.) was 24.17 kJ mol⁻¹ for the grains and 23.88 kJ mol⁻¹ for the fruits, at drying temperatures of 45, 60, 75, 90 and 105 °C (Siqueira et al., 2012). The difference between the activation energy values for the different products is due





** - Significant at $p \le 0.01$ by t-test

Figure 2. Effective diffusion coefficient as a function of drying temperatures (A); Arrhenius representation for the effective diffusion coefficient as a function of the inverse of the absolute drying air temperature (B) of tamarind (*Tamarindus indica* L.) seeds

Thermodynamic properties **Temperature** Н G (°C) (kJ mol⁻¹ K⁻¹) (kJ mol-1) (kJ mol-1) 45 -0.41626 164.95 32.51 60 32.39 -0.41631 171.08 177.22 75 32.26 -0.41635 90 32.14 -0.41639 183.35 $H = 32.88 - 0.0083^{**}T$ $S = -0.42 - 2.9 \times 10^{-6**}T$ $G = 146.55 + 0.409^{**}T$ Equation 0.9996 0.9999 R^2 0.9999

Table 4. Values of enthalpy (H), entropy (S) and Gibbs free energy (G) for different drying air temperatures of tamarind (*Tamarindus indica* L.) seeds

to their different structure and chemical composition, as well as the way through which water is bound to the constituents of the product.

Table 4 shows the values of enthalpy, entropy and Gibbs free energy for the different drying conditions. With increase in drying temperature, enthalpy and entropy decrease, while Gibbs free energy increases.

Enthalpy is related to the energy needed to remove the water bound to the dry matter during the drying process, so it is reduced with increasing drying temperature (Oliveira et al., 2010). Low enthalpy values at lower temperatures indicate a greater amount of energy required to promote the drying of tamarind seeds; similar behavior was observed in the drying processes of 'baru' fruits (*Dipteryx alata* Vog.) studied by Resende et al. (2018) and of 'Bode' pepper (*Capsicum chinense*) grains studied by Rodovalho et al. (2015).

Entropy values (Table 4) decrease with the increase in drying temperature, because high temperatures cause a greater increment in the excitation of the water molecules of the product when compared with low temperatures, decreasing the order of the water-product system (Corrêa et al., 2010). Since entropy is a thermodynamic property that can be associated with the degree of disorder between water and the product (Goneli et al., 2010), its values decrease as the drying temperature increases. Negative values of entropy are attributed to the existence of chemical adsorption and/or structural modifications of the adsorbent (Moreira et al., 2008).

Positive values for Gibbs free energy (Table 4) indicate an endergonic reaction, in which it is necessary to add energy to the air for the drying of the product to occur (Corrêa et al., 2010). Results with the same trend have been observed in the studies conducted by Martins et al. (2015), Araujo et al. (2017) and Morais et al. (2019).

Conclusions

- 1. To represent the drying of tamarind seeds, the Midilli model was selected for the range from 45 to 60 $^{\circ}$ C and the Two terms mode was selected for the range from 75 to 90 $^{\circ}$ C.
- 2. Effective diffusion coefficient increases with the increase in drying air temperature, with activation energy of $35.16\,\mathrm{kJ}$ mol $^{-1}$.
- 3. Enthalpy and entropy decrease with increasing temperature, respectively ranging from 32.14 to 32.51 kJ mol $^{-1}$ and from -0.41626 to -0.4639 kJ mol $^{-1}$ K $^{-1}$, for the temperature range from 45 to 90 °C.
- 4. Gibbs free energy increases with increasing drying temperature.

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