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THERMODYNAMIC PROPERTIES OF TAMARIND SEEDS (*Tamarindus indica* L.)

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KEYWORDS

equilibrium moisture content, enthalpy, Gibbs free energy.

ABSTRACT

Tamarind is used in the food and pharmaceutical industry, and its seed is the main means for the reproduction of this species, thus justifying studies that ensure its post-harvest viability. This research aimed to study the thermodynamic properties of tamarind seeds as a function of the equilibrium moisture content. The experimental data of the water activities were obtained by the indirect static method. The Cavalcanti Mata model was used to determine the thermodynamic properties of tamarind seeds, as it was the model that best fitted the experimental data of the desorption isotherms. The results showed that the thermodynamic properties are influenced by the moisture content and temperature, with an increase in the energy required to remove water from the product with a decrease in moisture content. The net isosteric heat of desorption increases with a reduction of moisture content ranging from 2,618.85 to 2,510.25 kJ kg⁻¹ for the moisture content range of 10.52 to 21.10% db. The latent heat of vaporization, the differential enthalpy, and the Gibbs free energy increase with a reduction of the moisture content of tamarind seeds.

INTRODUCTION

Tamarind seeds have a well-defined coat region, being irregular, rectangular, and rough, with a bright dark brown color and about 1.5 cm long and 1.2 cm wide (Sousa et al., 2010). Tamarind is used in the food and pharmacological industry, and its seeds, as well as those from woody fruit trees, are the main means for the reproduction of most of this species (Oliveira et al., 2006), thus requiring studies that ensure its post-harvest viability.

Drying stands out among the most used processes for maintaining the quality of plant products after harvest, as it is an indispensable technique for the quality control of plant products (Oliveira et al., 2011). The knowledge of this moisture content reduction process, which simultaneously involves the transfer of heat and mass, is essential for an efficient drying at technical and economic levels (Resende et al., 2011).

Thermodynamic properties aim to understand the water properties and calculate the energy demands

associated with heat and mass transfer in biological systems (Cladera-Oliveira et al., 2008). Moreover, the study of these properties is essential in the analysis of projection and the dimensioning of equipment in various processes of product preservation and calculate the energy required in these processes (Corrêa et al., 2010).

The latent heat of vaporization of water, differential enthalpy, net isosteric heat of desorption, and Gibbs free energy are among the thermodynamic properties of the product. These thermodynamic properties have been extensively studied in the literature due to their importance for different plant products, such as cassava (Koua et al., 2014), Barbados nut (Chaves et al., 2015) seeds, castorbean seeds (Goneli et al., 2016), crambe fruits (Oliveira et al., 2017), and baru (*Dipteryx alata* Vogel) fruits (Resende et al., 2017).

Due to the importance of the post-harvest processes of plant products for their preservation during storage, this work aimed to study the thermodynamic properties of tamarind seeds as a function of the equilibrium moisture content and temperature.

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MATERIAL AND METHODS

The experiment was conducted at the Laboratory of Post-harvest of Plant Products of the Federal Institute of Education, Science, and Technology Goiano (IF Goiano), located in Rio Verde, GO, Brazil. Tamarind seeds with an initial moisture content of $21.00 \pm 0.10\%$ dry basis (db) were used. Fruits were collected manually in the rural region of Rio Verde, GO, Brazil ($17^{\circ}51'57''$ S and $50^{\circ}50'05''$ W).

Different moisture contents were obtained after cleaning the seeds. For this, they were submitted to drying in a forced-air ventilation oven at a temperature of 45°C and relative humidity of 20.6% until moisture contents of 17.27, 15.04, 14.15, 12.40, and $10.62 \pm 0.12\%$ db were reached. The moisture contents were verified by the oven method at $105 \pm 1^{\circ}\text{C}$ for 24 hours (Brasil, 2009).

Desorption isotherms were determined using the indirect static method, with the water activity (a_w) determined using the equipment HygroPalm AW1. Approximately 35 g of seeds were used for each moisture content in triplicate. The apparatus was placed in B.O.B. regulated at temperatures of 10, 20, 30, and 40°C . After reading the water activity values in the equipment, the moisture content of the samples was determined in an oven, following the methodology of Brasil (2009).

The thermodynamic properties were obtained using the Cavalcanti Mata model (1), as it presented the best fit to the experimental data of the desorption isotherms, with a coefficient of determination (R^2) of 0.9961, relative mean error (P) of 1.42%, average error of estimate (SE) of 0.232, calculated chi-square (χ^2) of 0.0540, AIC of 2.94, and BIC of 6.03.

$$Xe = ((\ln(1 - a_w))/(-0.005527^{**} \cdot T^{0.12914^{**}})]^{1/1.831766^{**}} \quad (1)$$

Where:

Xe is the equilibrium moisture content (% db);

a_w is the water activity, and

T is the temperature ($^{\circ}\text{C}$).

The estimated water activity was obtained from the manipulation of the Cavalcanti Mata equation, generating [eq. (2)]:

$$a_w = 1 - \exp[Xe^{1.831766^{**}}(-0.005527^{**} \cdot T^{0.12914^{**}})] \quad (2)$$

Where:

** is significant at 1% by the t-test.

Brooker et al. (1992) proposed Equation (3), with a reference to the Clausius-Clapeyron studies, to quantify the partial vapor pressure contained in porous systems:

$$\ln(P_v) = \left(\frac{L}{L'}\right) \cdot \ln(P_{vs}) + C \quad (3)$$

Where:

P_{vs} is the saturation vapor pressure of free water for a given equilibrium temperature;

P_v is the vapor pressure of free water for a given equilibrium temperature;

L is the latent heat of vaporization of water of the product (kJ kg^{-1});

L' is the latent heat of vaporization of free water at equilibrium temperature (kJ kg^{-1}), and

C is the integration constant.

The saturation vapor pressure of free water was calculated using the Thétens [eq. (4)]:

$$P_{vs} = 0.61078 \cdot 10^{((7.5T)/(273.3+T))} \quad (4)$$

The vapor pressure value was determined according to [eq. (5)]:

$$P_v = a_w \cdot P_{vs} \quad (5)$$

The desorption isotherm allowed determining the L/L' ratio of [eq. (6)], according to the methodology described by Pereira & Queiroz (1987) for different equilibrium moisture contents (X_e). Thus, the equation for enthalpy of vaporization of water, presented by Rodrigues-Arias (Brooker et al., 1992), was adjusted with the inclusion of one more parameter in [eq. (6)] to improve the L/L' estimates (Corrêa et al., 1998):

$$\left(\frac{L}{L'}\right) - 1 = a \cdot \exp(-b \cdot Xe^m) \quad (6)$$

Where:

a , b , and m are parameters determined by regression.

The latent heat of vaporization of free water (kJ kg^{-1}) at equilibrium temperature ($^{\circ}\text{C}$) was calculated using the average temperature ($^{\circ}\text{C}$) within the range under study, using [eq. (7)]:

$$L' = 2502.2 - 2.39T \quad (7)$$

The latent heat of vaporization of water of the product (kJ kg^{-1}) was estimated by combining eqs (6) and (7) (Corrêa et al., 1998), as shown in [eq. (8)]:

$$L = (2502.2 - 2.39T) \cdot [1 + a \cdot \exp(-b \cdot Xe^m)] \quad (8)$$

The Clausius-Clapeyron equation (9) (Iglesias & Chirife, 1976) was used to calculate the differential enthalpy for each equilibrium moisture content:

$$\frac{\partial \ln(a_w)}{\partial T} = \frac{\Delta h_{st}}{RT_{abs}^2} \quad (9)$$

Where:

T_{abs} is the absolute temperature (K);

Δh_{st} is the differential enthalpy (kJ kg^{-1}), and

R is the universal gas constant for water vapor ($0.4619 \text{ kJ kg}^{-1} \text{ K}^{-1}$).

Integrating [eq. (9)] and assuming that the differential enthalpy is independent of the temperature, each equilibrium moisture content was obtained from [eq. (10)] (Wang & Brennan, 1991):

$$\ln(a_w) = -\left(\frac{\Delta h_{st}}{R}\right) \cdot \frac{1}{T_{abs}} + C \quad (10)$$

Where:

C is the model coefficient.

Net isosteric heat of sorption was obtained by adding the value of latent heat of vaporization of free water (L') to the values of differential enthalpy, according to [eq. (11)]:

$$Q_{st} = \Delta h_{st} + L' = a \cdot \exp(-b \cdot X_e) + c \quad (11)$$

Where:

Q_{st} is the net isosteric heat of sorption (kJ kg^{-1}), and a , b , and c are the model coefficients.

Gibbs free energy can be calculated by [eq. (12)]:

$$G = R \cdot T \cdot \ln(a_w) \quad (12)$$

Where:

G is the Gibbs free energy (kJ kg^{-1}).

Gibbs free energy for each temperature can be described by the exponential regression shown in [eq. (13)], according to Resende et al. (2017):

$$G = \alpha \cdot \exp(\beta - X_e) + \delta \quad (13)$$

Where:

α , β , and δ are equation regression parameters.

The significance of the regression parameters was performed by the t-test to verify the degree of adjustment of each model, the magnitude of the coefficient of determination (R^2), and the relative mean error (P), calculated according to [eq. (14)]:

$$P = \frac{100}{n} \sum \frac{|Y - \hat{Y}|}{Y} \quad (14)$$

Where:

Y is the experimental value;

\hat{Y} is the value estimated by the model, and

n is the number of experimental observations.

RESULTS AND DISCUSSION

Table 1 shows the water activity values estimated by the Cavalcanti Mata model for the moisture content range from 10.52 to 21.10% db at temperatures of 10, 20, 30, and 40 °C. The water activity increases in response to an increase in temperature and moisture content. The same trend was observed by Resende et al. (2017) studying the thermodynamic properties of baru (*Dipteryx alata* Vogel) fruits from sorption isotherms at temperatures of 20, 25, 30, and 35 °C with an equilibrium moisture content range from 4.2 to 29.5% db.

TABLE 1. Water activity values (decimal) of tamarind (*Tamarindus indica* L.) seeds estimated by the Cavalcanti Mata model as a function of temperature and equilibrium moisture content.

| Xe (% db) | Temperature °C | | | |
|--------------|----------------|--------|--------|--------|
| | 10 | 20 | 30 | 40 |
| 10.52 | 0.4253 | 0.4544 | 0.4719 | 0.4845 |
| 10.74 | 0.4375 | 0.4670 | 0.4848 | 0.4975 |
| 12.39 | 0.5267 | 0.5587 | 0.5777 | 0.5912 |
| 12.49 | 0.5320 | 0.5641 | 0.5831 | 0.5967 |
| 14.15 | 0.6150 | 0.6480 | 0.6672 | 0.6807 |
| 15.04 | 0.6560 | 0.6887 | 0.7076 | 0.7209 |
| 17.27 | 0.7468 | 0.7774 | 0.7946 | 0.8066 |
| 20.94 | 0.8585 | 0.8822 | 0.8950 | 0.9036 |
| 21.10 | 0.8624 | 0.8857 | 0.8983 | 0.9067 |

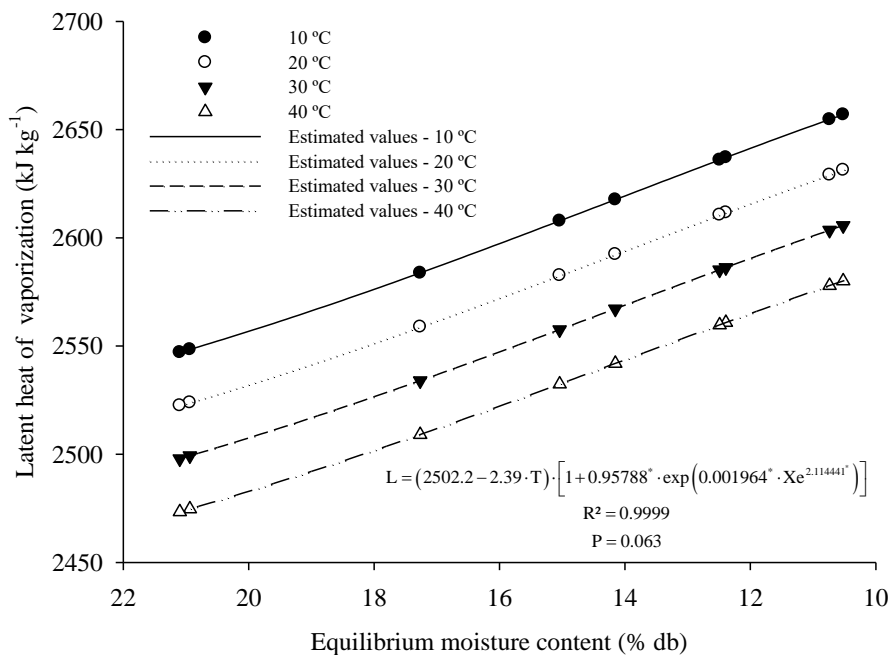
Table 2 shows the L/L' ratio values for different equilibrium contents. The values decrease as the moisture content increases.

TABLE 2. Values of the L/L' ratio for different equilibrium moisture contents of tamarind (*Tamarindus indica* L.) seeds

| Moisture content (% db) | L/L' ratio | Moisture content (% db) | L/L' ratio |
|----------------------------|-----------------|----------------------------|-----------------|
| 10.52 | 1.0721 | 15.04 | 1.0523 |
| 10.74 | 1.0712 | 17.27 | 1.0426 |
| 12.39 | 1.0640 | 20.94 | 1.0283 |
| 12.49 | 1.0636 | 21.10 | 1.0277 |
| 14.15 | 1.0562 | | |

Table 2 shows that the values of the L/L' ratio tend to be close to 1.0. This response is due to an increase in vapor pressure as moisture contents increase. Oliveira et al. (2014a) observed a similar trend for the different equilibrium moisture content of Barbados nut seeds.

The linear regression equation proposed by Corrêa et al. (1998) can be used to estimate the latent heat of vaporization of tamarind seeds. It has a high coefficient of determination (R^2), a low relative mean error (P), and significant parameters at 1% by the t-test. Figure 1 shows the latent heat curves of vaporization of water of tamarind seeds at temperatures of 10, 20, 30, and 40 °C within the equilibrium moisture content range of 10.52 to 21.10% db.



**Significant at 1% by the t-test.

FIGURE 1. Latent heat of vaporization of water of tamarind (*Tamarindus indica* L.) seeds as a function of the equilibrium moisture content for temperatures of 10, 20, 30, and 40 °C.

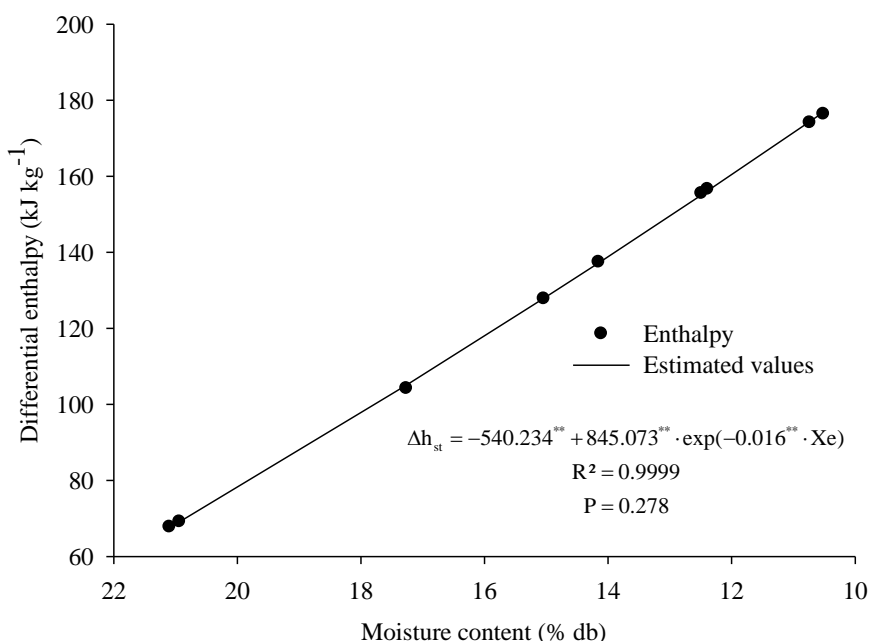
The latent heat of vaporization of water of tamarind seeds ranged from 2,656.90 to 2,473.34 kJ kg⁻¹. The decrease in the equilibrium moisture content leads to an increase in the energy required for water evaporation from seeds. According to Brooker et al. (1992), moisture content and temperature are the main factors influencing the latent heat of vaporization of water of the product.

The latent heat of vaporization of water of tamarind seeds decreases with increasing temperature for the same moisture content, presenting an inversely proportional relationship and corroborating with results obtained by Oliveira et al. (2017).

The latent heat value of vaporization of water of cayenne pepper (*Capsicum frutescens* L.) seeds ranged from

3,615.01 to 2,455.14 kJ kg⁻¹ in the study conducted by Silva & Rodovalho (2016) at the moisture content range of 4.6 to 21.3% db and temperatures of 30, 40, and 50 °C. The latent heat of vaporization of Barbados nut seeds was 2,762.92 to 2,495.56 kJ kg⁻¹ within the equilibrium moisture content range of 5.61 to 13.42% db and at temperatures of 10, 20, 30, and 40 °C (Oliveira et al., 2014a).

Figure 2 shows the experimental and estimated values for the differential enthalpy (Δh_{st}) as a function of the different equilibrium moisture contents (% db) of tamarind seeds. The equation had a high coefficient of determination (R^2), low relative mean error (P), and all equation parameters were significant at 1% by the t-test, showing the adequacy of the equation to the experimental data.



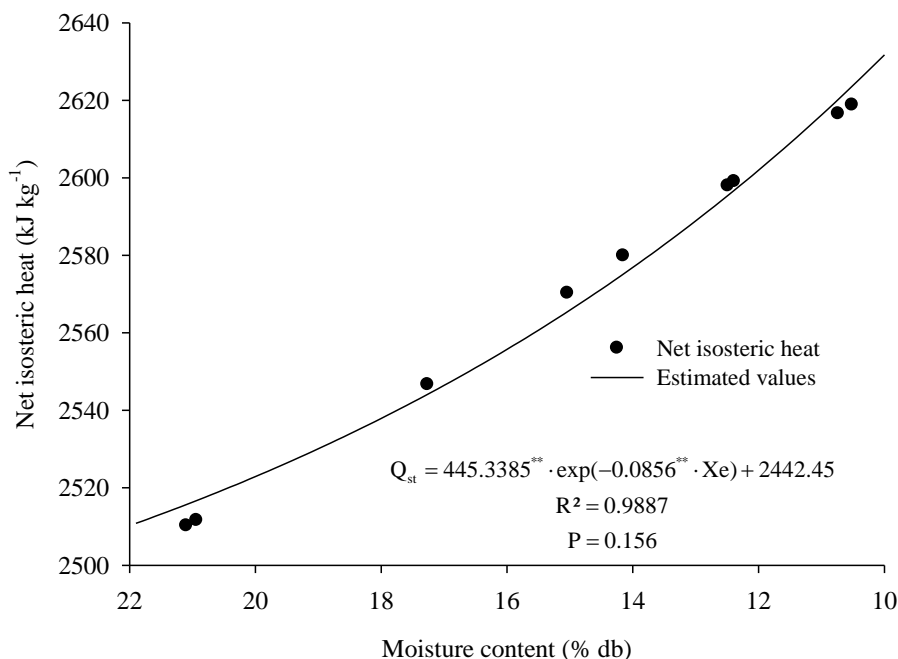
**Significant at 1% by the t-test.

FIGURE 2. Observed and estimated values of differential enthalpy (Δh_{st}) of desorption of tamarind (*Tamarindus indica* L.) seeds.

Differential enthalpy increased with a reduction in moisture content (Figure 2), corroborating with the results obtained for cocoa (*Theobroma cacao*) seeds (Oliveira et al., 2011) and tucumã-de-Goiás (*Astrocaryum huaimi* Mart.) seeds (Oliveira et al., 2014b). In addition, the differential enthalpy value varied from 176.40 to 67.80 kJ kg⁻¹ for a moisture content range of 10.52 to 21.10% db. The variation in enthalpy values provides a measure of the energy variation required to remove water when its molecules interact with constituents of the product during

the sorption processes (McMinn et al., 2005).

The values of the net isosteric heat of desorption as a function of the equilibrium moisture content for tamarind seeds were calculated according to Equation (11) and shown in Figure 3. The Q_{st} adjustment equation presented a high coefficient of determination (R^2), low relative mean error (P), and significant parameters at 1% by the t-test, which can be used to estimate the net isosteric heat of desorption of tamarind seeds.



**Significant at 1% by the t-test.

FIGURE 3. Experimental and estimated values of the net isosteric heat of desorption as a function of the equilibrium moisture content of tamarind (*Tamarindus indica* L.) seeds.

Figure 3 shows that as the moisture content of the product decreases, more energy must be provided to remove water, corroborating with the results obtained for different plant products, such as baru (*Dipteryx alata* Vogel) beans (Furtado et al., 2014), forage radish (*Raphanus sativus* L.) seeds (Sousa et al., 2015), cajuzinho-do-cerrado (*Anacardium humile* A.St.-Hil.) achenes (Barbosa et al., 2016), and pequi (*Caryocar brasiliense* Camb.) diaspores (Sousa et al. 2016).

The values of the net isosteric heat of desorption for tamarind seeds in the equilibrium moisture content range of 10.52 to 21.10% db varied from 2,618.85 to 2,510.25 kJ kg⁻¹. Seeds of fourleaf buchenavia (*Buchenavia capitata* (Vahl) Eichler) showed values of the net isosteric heat of desorption ranging from 2,667.93 to 2,819.56 kJ kg⁻¹ and equilibrium moisture contents ranging from 13.31 to 7.21% db (Costa et al., 2015). Chaves et al. (2015) studied the net

isosteric heat of desorption of Bermuda nut seeds in the moisture content range of 5.6 to 13.4% db and obtained values a variation of 3,035.61 to 2,631.89 kJ kg⁻¹.

Water-holding capacity to product constituents increases as the moisture content of the sample decreases due to an increase in the concentration of the chemical constituents of the product, such as fats, proteins, and salts (Hubinger et al., 2009). Low moisture contents indicate proximity to monomolecular layers, which are strongly linked to dry matter and require a higher energy rate to remove water in the form of steam (Al-Muhtaseb et al., 2004).

Gibbs free energy increased with decreasing moisture content and temperature, being positive for all studied conditions (Table 3). The values of this thermodynamic property tend to stabilize at higher equilibrium moisture contents. Resende et al. (2017) and Silva et al. (2016) observed similar behaviors.

TABLE 3. Gibbs free energy values (kJ kg⁻¹) as a function of the moisture content of tamarind (*Tamarindus indica* L.) seeds.

| Moisture content (% db) | Temperature (°C) | | | |
|-------------------------|------------------|--------|--------|--------|
| | 10 | 20 | 30 | 40 |
| 10.52 | 111.81 | 106.81 | 105.17 | 104.82 |
| 10.74 | 108.11 | 103.09 | 101.39 | 100.98 |
| 12.39 | 83.85 | 78.83 | 76.84 | 76.01 |
| 12.49 | 82.55 | 77.53 | 75.53 | 74.69 |
| 14.15 | 63.57 | 58.76 | 56.67 | 55.63 |
| 15.04 | 55.15 | 50.51 | 48.43 | 47.34 |
| 17.27 | 38.18 | 34.10 | 32.19 | 31.09 |
| 20.94 | 19.95 | 16.97 | 15.54 | 14.67 |
| 21.10 | 19.36 | 16.43 | 15.02 | 14.16 |

The positive values observed for Gibbs free energy were expected since desorption is a non-spontaneous process, characteristic of an exogenous reaction, i.e., a reaction that requires an agent supplying energy to the environment. Corrêa et al. (2015) observed positive values

for the Gibbs free energy of cucumber (*Cucumis sativus* L.) seeds, as well as Goneli et al. (2016) for castorbean (*Ricinus communis* L.) seeds.

Table 4 shows the exponential regression equations for Gibbs free energy.

TABLE 4. Gibbs free energy equations of tamarind (*Tamarindus indica* L.) seeds for different temperatures.

| Temp. (°C) | Equation* | R ² (decimal) | P (%) |
|------------|---|--------------------------|-------|
| 10 | $G = 539.7360^{**} \cdot \exp(-0.1445^{**} \cdot Xe) - 6,2564^{**}$ | 0.9999 | 0.069 |
| 20 | $G = 562.4996^{**} \cdot \exp(-0.1527^{**} \cdot Xe) - 6,0434^{**}$ | 0.9999 | 0.126 |
| 30 | $G = 585.0464^{**} \cdot \exp(-0.1579^{**} \cdot Xe) - 5,9386^{**}$ | 0.9999 | 0.168 |
| 40 | $G = 607.3941^{**} \cdot \exp(-0.1618^{**} \cdot Xe) - 5,8896^{**}$ | 0.9999 | 0.202 |

*Equation (13): $G = \alpha \cdot \exp(\beta - Xe) + \delta$. **Significant at 1% by the t-test.

Equation (13), described by Resende et al. (2017), can be used to determine Gibbs free energy for the studied temperatures. The values of Gibbs free energy presented a high coefficient of determination (R²), low relative mean error (P), and all parameters significant at 1% by the t-test under the studied conditions. Table 4 shows that the values of the regression parameters (α and β) of [eq. (13)] increased with an increase in temperature, while the parameter δ had an opposite effect.

CONCLUSIONS

Thermodynamic properties are influenced by the moisture content of tamarind seeds, with an increase in the energy required to remove water from the product with a decrease in moisture content.

The net isosteric heat of desorption increases with a reduction in the moisture content, ranging from 2,618.85 to 2,510.25 kJ kg⁻¹ for the moisture content range of 10.52 to 21.10% db.

The latent heat of vaporization, the differential enthalpy, and the Gibbs free energy increase with a reduction of the moisture content of tamarind seeds.

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