

THERMODYNAMIC PROPERTIES OF BARU FRUITS (*Dipteryx alata Vogel*)Doi: <http://dx.doi.org/10.1590/1809-4430-Eng.Agric.v37n4p739-749/2017>**OSVALDO RESENDE^{1*}, DANIEL E. C. DE OLIVEIRA², LÍLIAN M. COSTA³,
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ABSTRACT: The objective of this study was to determine and evaluate the thermodynamic properties for different equilibrium moisture contents in baru fruits (*Dipteryx alata Vogel*), using the direct static method to obtain the experimental data. The Modified Halsey model was used to determine the thermodynamic properties of baru fruits. Results concluded that thermodynamic properties are influenced by moisture content. The water vaporization latent heat increases with the decrease in equilibrium moisture content. The baru fruit desorption process is controlled by enthalpy. Gibbs free energy is positive for the temperatures studied with the increase over desorption, which describes a non-spontaneous process.

KEY WORDS: latent heat, enthalpy, entropy, Gibbs free energy.

INTRODUCTION

Baru (*Dipteryx alata Vogel*) is a typical plant of the Cerrado; its fruits are drupe type with a light brown color, containing an elliptic-shaped edible seed, dark brown in color and commonly called almond. This seed has great regional importance, attracting scientific interest due to its nutritional composition. Almonds of baru fruit have higher levels of monounsaturated fatty acids (51.1%), and lower levels of saturated fatty acids (Bento et al., 2014).

However, the post-harvest processes to maintain fruit and almond quality after the harvest must be known. The drying process stands out among the processes most widely used to maintain the quality of agricultural products after harvesting. This fact justifies the study of thermodynamic properties, since they provide relevant information on water behavior in agricultural products, and the energy requirements to remove it during the drying process (Silva et al., 2016).

The knowledge of thermodynamic properties in the drying processes of agricultural products is important to design drying equipment, to study the properties of the adsorbed water, to calculate the energy required in this process, and to evaluate the microstructure of foods and the study of the physical phenomena that occur on the food surface (Corrêa et al., 2010).

In addition, the determination of the thermodynamic properties is fundamental to predict the end at which the fruit should be dried in order to obtain a product that can be stored for long periods, consuming a minimum amount of energy to reduce the moisture content to safe storage levels. Thermodynamic parameters such as enthalpy, entropy, and Gibbs free energy have been investigated in different products recently, such as: coffee beans (Goneli et al., 2013), jatropha (Oliveira et al., 2014a), tucumã-de-Goiás seeds (Oliveira et al., 2014b), fodder radish (Sousa et al., 2015), cactus (Hassini et al., 2015), ‘cabacinha’ pepper (Silva et al., 2016), and chili (Silva & Rodvalho, 2016).

As no studies were found on the thermodynamic properties of baru fruits, the objective of this work was to study their thermodynamic properties as a function of equilibrium moisture content.

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MATERIAL AND METHODS

The experiment was conducted at the Post-Harvest Laboratory of Vegetable Products, Federal Institute of Goiás for Education, Science, and Technology - Campus Rio Verde. Baru fruits (*Dipteryx alata Vogel*) were collected in the city of Santa Helena de Goiás – GO (Brazil). It lies at 17°48' S, 50°35' W, and altitude of 568 m. The fruits had a moisture content of 43% dry basis (db).

The static-gravimetric method was used to obtain the hygroscopic equilibrium moisture content and, for each repetition, approximately 50 g of fruits were placed in a permeable tissue (voile) to allow air to pass through the product, and then placed inside desiccators. Temperature and relative air humidity were monitored with a data logger inserted in the desiccators.

Relative humidity inside the desiccators was controlled using saturated solutions of different salts (lithium chloride, calcium chloride, calcium nitrate, sodium chloride, and potassium bromide). The desiccators were placed in incubator chambers type B.O.D. (Biochemical Oxygen Demand), and regulated at temperatures of 20, 25, 30, and 35 °C.

The Modified Halsey model was used in order to determine the baru fruits thermodynamic properties, because it presented the best fit to the experimental data, with the coefficient of determination, estimated mean error (SE), *Chi*-square (χ^2), and mean relative error (P) of 99.46%; 0.72; 0.52 and 4.44%, respectively. The values of water activity were obtained by the following expression:

$$X_e = \left[\exp(2.8707^{**} - 0.0084^{**} \cdot T) / -\ln(a_w) \right]^{\frac{1}{1.2483^{**}}} \quad (1)$$

** Significant at 1% by t-test.

In which:

X_e: equilibrium moisture content (% db);

a_w: water activity (decimal), and

T: temperature (°C).

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

$$\ln(P_v) = \left(\frac{L}{L'} \right) \cdot \ln(P_{vs}) + C \quad (2)$$

In which:

P_{vs}: free water saturation vapor pressure for a given temperature (T), of equilibrium;

P_v: free water vapor pressure at a given temperature T, of equilibrium;

L: latent heat of product water vaporization, kJ kg⁻¹;

L': latent heat of free water vaporization, equilibrium temperature, kJ kg⁻¹, and

C: integration constant.

Based on the sorption isotherms of baru fruits (*Dipteryx alata Vogel*), the L/L' ratio (Equation 3) was determined for different equilibrium moisture contents. The equation was adjusted for water vaporization enthalpy, presented by Rodrigues – Arias (Brooker et al., 1992), with the inclusion of one more parameter in [eq. (3)] to improve L/L' estimates (Corrêa et al., 1998):

$$\frac{L}{L'} - 1 = a \cdot \text{Exp}(-b \cdot X_e^m) \quad (3)$$

In which,

a, b, m: parameters determined by regression.

The latent heat of free water vaporization (kJ kg^{-1}) at equilibrium temperature ($^{\circ}\text{C}$) was calculated using the mean temperature (T) within the study range, using the following equation:

$$L' = 2502.2 - 2.39 \cdot T \quad (4)$$

The free water saturation vapor pressure, P_{vs} , was calculated by the Thétens equation:

$$P_{vs} = 0.61078 \cdot 10^{((7.5 \cdot T)/(273.3 + T))} \quad (5)$$

The vapor pressure value, P_v , was determined according to the following equation:

$$P_v = a_w \cdot P_{vs} \quad (6)$$

Equations 3 and 4 combine to estimate the latent heat of the product vaporization water (Corrêa et al., 1998), reaching the following expression:

$$L = (2502.2 - 2.39 \cdot T) \cdot [1 + a \cdot \text{Exp}(-b \cdot X e^m)] \quad (7)$$

The differential entropy of sorption was calculated according to [eq. (8)].

$$S = \frac{h_{st} - G}{T_{abs}} \quad (8)$$

In which:

S : the differential entropy of sorption ($\text{kJ kg}^{-1}\text{K}^{-1}$);

G : Gibbs free energy (kJ kg^{-1});

T_{abs} : absolute temperature, K, and

h_{st} : differential enthalpy, kJ kg^{-1} .

Gibbs free energy can be calculated by the following equation:

$$G = R \cdot T_{abs} \cdot \ln(a_w) \quad (9)$$

Changes in water sorption over free energy usually cause changes in enthalpy and entropy values. Thus, replacing Equation 8 in 9 and rearranging, we have:

$$\ln(a_w) = \frac{h_{st}}{R \cdot T_{abs}} - \frac{S}{R} \quad (10)$$

The values of differential enthalpy and differential entropy of sorption were calculated by [eq. (11)]. The values of differential enthalpy of sorption (h_{st}) and entropy (S) calculated were correlated by the following equation (Beristain et al., 1996):

$$h_{st} = T_B \cdot S + G_B \quad (11)$$

In which,

T_B : isokinetic temperature (K), and

G_B : Gibbs free energy at an isokinetic temperature (kJ kg^{-1}).

The isokinetic temperature represents the temperature at which all series reactions occur at the same rate. Since enthalpy and entropy are highly correlated, compensation theory is assumed for sorption (Beristain et al., 1996). In order to confirm the existence of compensation, the isokinetic temperature was compared to the harmonic mean of the temperatures used to determine the sorption isotherms, according to [eq. (12)] (Krug et al., 1976):

$$T_{hm} = \frac{n}{\sum \left(\frac{1}{T} \right)} \quad (12)$$

In which,

T_{hm} : mean harmonic temperature (K), and

n: number of temperatures used.

According to Krug et al. (1976), linear chemical compensation or compensation theory only exists if the isokinetic temperature (T_B) is different from the mean harmonic temperature (T_{hm}). An approximate confidence interval, $(1-\alpha)$ 100%, for the isokinetic temperature was obtained by the following equation:

$$T_B = \hat{T} \pm t_{m-2, \alpha/2} \sqrt{\text{Var}(T_B)} \quad (13)$$

In which,

$$\hat{T}_B = \frac{\sum (h_{st} - \bar{h}_{st})(S - \bar{S})}{\sum (S - \bar{S})^2} \quad (14)$$

and,

$$\text{Var}(T_B) = \frac{\sum (h_{st} - \bar{G}_B - \hat{T}_B \cdot S)^2}{(m-2) \sum (S - \bar{S})^2} \quad (15)$$

In which,

m: number of enthalpy and entropy data pairs;

\bar{h}_{st} : mean enthalpy, kJ kg⁻¹; and

\bar{S} : mean entropy kJ kg⁻¹.

If the harmonic mean temperature T_{hm} is within the isokinetic temperature T_B range calculated, the ratio between enthalpy values and the differential entropy of sorption reflects only experimental errors, not the existence of chemical and physical factors that govern the theory of compensation (Beristain et al., 1996). A 99% confidence interval was adopted for T_B throughout the data range observed.

The relative mean error (P) was calculated according to the following expression:

$$P = \frac{100}{n} \sum \frac{|Y - \hat{Y}|}{Y} \quad (16)$$

In which:

Y: experimental value;

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

n: number of experimental observations.

RESULTS AND DISCUSSION

Table 1 shows the water activity values estimated by the Modified Halsey model, Equation 1, for the temperatures of 20, 25, 30 and 35 °C, and for equilibrium moisture contents of 4.2 to 29.5 (% db). There is an increase in water activity with the increase in moisture content, and the same effect is observed with increase in temperature. This behavior was reported by other authors when studying the thermodynamic properties of several agricultural products, such as maize (Smaniotto et al., 2012), jathropa (Oliveira et al., 2014a), and ‘cabacinha’ pepper (Silva et al., 2016).

TABLE 1. Water activity values (decimal) estimated by the Modified Halsey model as a function of temperature and equilibrium moisture content.

Xe (% db)	Temperature (°C)			
	20	25	30	35
4.2	0.0856	0.0947	0.1043	0.1145
4.5	0.0992	0.1091	0.1195	0.1304
4.6	0.1077	0.1181	0.1289	0.1402
5.7	0.1842	0.1974	0.2110	0.2250
6.8	0.2526	0.2674	0.2823	0.2973
7.7	0.3093	0.3246	0.3400	0.3554
8.6	0.3635	0.3789	0.3944	0.4097
8.8	0.3710	0.3865	0.4019	0.4172
9.0	0.3813	0.3967	0.4121	0.4274
9.1	0.3870	0.4024	0.4177	0.4330
9.7	0.4172	0.4325	0.4476	0.4627
12.4	0.5254	0.5395	0.5534	0.5670
21.7	0.7255	0.7351	0.7445	0.7535
24.1	0.7547	0.7635	0.7720	0.7803
24.3	0.7574	0.7661	0.7745	0.7827
25.9	0.7739	0.7821	0.7900	0.7977
28.4	0.7951	0.8026	0.8099	0.8169
29.5	0.8041	0.8113	0.8183	0.8251

Water activity values, Table 1, were used to determine the differential enthalpy of desorption. Table 2 shows the values of the L/L' ratio for the different moisture contents. The L/L' ratio is increased by decreasing moisture contents.

TABLE 2. Ratio between L/L' for the different moisture contents in baru fruits (*Dipteryx alata Vogel*).

Moisture content (% db)	Ratio L/L'	Moisture content (% db)	Ratio L/L'
4.2	1.3317	9.1	1.1281
4.5	1.3118	9.7	1.1180
4.6	1.3006	12.4	1.0868
5.7	1.2283	21.7	1.0433
6.8	1.1856	24.1	1.0380
7.7	1.1583	24.3	1.0375
8.6	1.1365	25.9	1.0346
8.8	1.1338	28.4	1.0309
9.0	1.1301	29.5	1.0294

The L/L' ratio values increase with moisture content reduction, and magnitudes were close to 1.0 for high moisture contents. Oliveira et al. (2014b) and Silva & Rodovalho (2012) verified a similar behavior in studies with tucumã-de-Goiás and chili pepper seeds, respectively,

Table 3 shows the parameters "a", "b" and "m" used to calculate the ratio between the latent heat of water vaporization in agricultural products (L), and the free water latent heat (L/L') obtained by means of non-linear regression. The regression equation can be used to estimate the latent vaporization heat of baru fruits, since it has a high coefficient of determination (R²), and a low mean relative error (P).

TABLE 3. Parameters "a", "b" and "m" used to calculate the ratio between the latent heat of water vaporization in agricultural products, and the free water latent heat (L/L').

a	b	m	R ² (%)	P (%)
1.2886 x 10 ⁻¹³ **	-30.4896**	-0.0448**	99.99	1.50

** Significant at 1% by t-test.

By changing the values of "a", "b" and "m" in the equation proposed by Corrêa et al. (1998), the following equation is obtained for the calculation of the latent heat of water vaporization in baru fruits.

$$L = (2502,2 - 2,39 \cdot T) \cdot \left[1 + 1,2886 \times 10^{-13} \cdot \exp(30,4896 \cdot X e^{-0,0448}) \right] \tag{17}$$

Figure 1 shows the latent heat curves of water vaporization in baru fruits at temperatures of 20, 25, 30, and 35 °C.

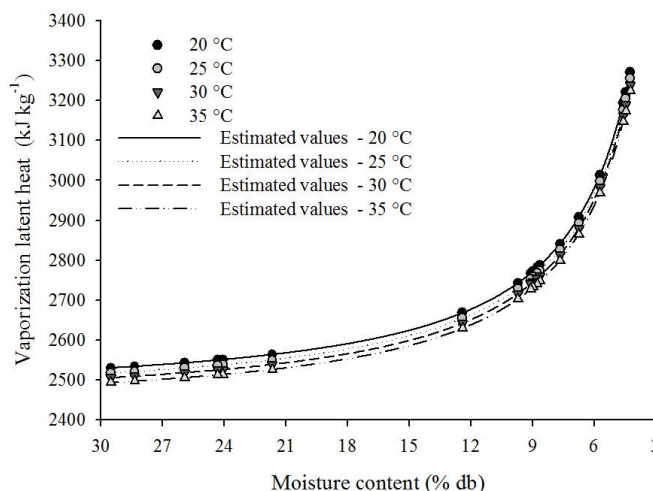


FIGURE 1. Experimental and estimated values of the latent heat of water vaporization as a function of equilibrium moisture content for baru fruits (*Dipteryx alata Vogel*).

The latent heat of vaporization is inversely proportional to baru fruits' moisture content and temperature, and the increase in temperature promoted reduction of the latent vaporization heat for the same moisture content, corroborating the results obtained by Oliveira et al. (2014a, b), Sousa et al. (2016), and Silva & Rodovalho (2016).

The latent heat of baru fruits vaporization ranged from 3,271.06 to 2,493.09 kJ kg⁻¹ for moisture contents of 4.2 to 29.5% (db). Brooker et al. (1992) emphasize that the latent heat of vaporization in the product is influenced mainly by moisture content and temperature.

The authors observed that the latent heat ranged from 2,762.92 to 2,495.56 kJ kg⁻¹ in studies with jathropa (Oliveira et al., 2014a) and corn grains (Smaniotto et al., 2012), with moisture contents from 5.61 to 13.42% db for jathropa seeds, and from 2,775.87 to 2,468.14 kJ kg⁻¹ in the range of 12.76 to 23.26% db for corn grains. This variation in the values obtained for the different agricultural products may be related to structure and chemical composition, and also temperature and moisture content.

Figure 2 shows the values of differential enthalpy (h_{st}) and differential entropy (S) of desorption, as a function of equilibrium moisture content (% db).

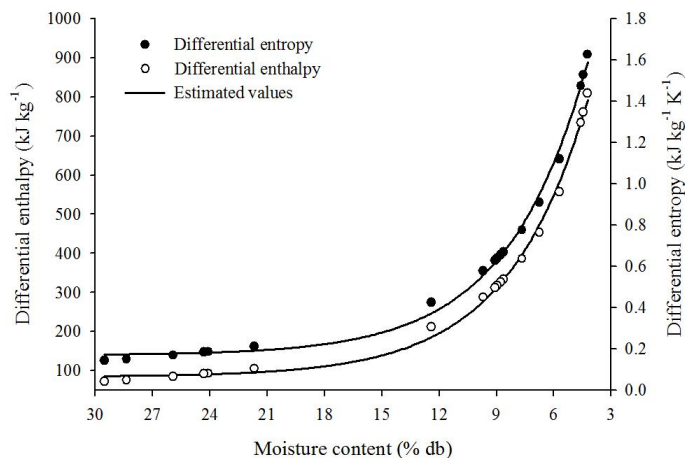


FIGURE 2. Values of differential enthalpy and entropy of desorption observed and estimated for baru fruits (*Dipteryx alata Vogel*).

Differential enthalpy and entropy values presented similar tendencies for moisture content, and differential enthalpy and entropy tend to stabilize at high moisture contents. Differential enthalpy and entropy increase with moisture content reduction in baru fruits (Figure 2), in line with the results obtained from jatropa seeds (Oliveira et al., 2014a), and tucumã-de-Goiás (Oliveira et al., 2014b).

In addition, the differential enthalpy and entropy values ranged from 71.80 to 809.60 kJ kg⁻¹ K⁻¹, and 0.1443 to 1.6263 kJ kg⁻¹ K⁻¹, respectively, for the moisture content range from 3.9 to 10.9% db. This entropy behavior is related to water molecule mobility during the desorption process. This behavior is indicative of the degree of water disorder (Majd et al., 2013).

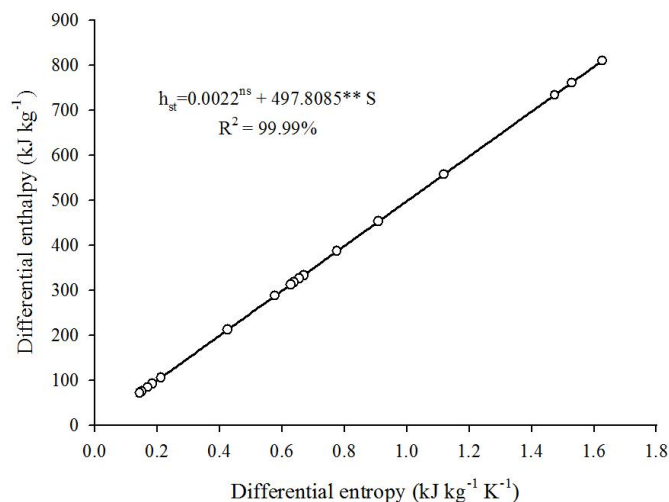
Table 4 presents the equations to determine differential enthalpy and entropy, as well as coefficients of determination for baru fruits. The equations presented high coefficients of determination and low relative mean error. In addition, all equation parameters were significant at 1% significance by the t-test, evidencing the adequacy of the equations to the experimental data.

TABLE 4. Equations and coefficients of differential enthalpy (h_{st}) and entropy (S) of desorption in baru fruits (*Dipteryx alata Vogel*).

Thermodynamic properties	Equations	R ² (%)	P (%)
Differential enthalpy	$h_{st}=84.6574^{**}+1942.4311^{**}.\exp(-0.2384^{**}.Xe)$	99.73	4.869
Differential enthalpy	$S=0.1700^{**}+3.9019^{**}.\exp(-0.2384^{**}.Xe)$	99.73	4.867

** Significant at 1% by t-test.

The data adjusted to the linear model for the ratio between enthalpy and entropy in the desorption process for baru fruits, (Figure 3). Note the high experimental accuracy due to the high coefficient of determination (99.99%).



** Significant at 1% by t-test. ^{ns}Non-significant by the t-test.

FIGURE 3. Enthalpy-entropy ratio for water desorption process in baru fruits (*Dipteryx alata Vogel*).

With the linearity between the differential enthalpy ratio and the differential entropy of sorption, the isokinetic theory, or enthalpy-entropy compensation theory for the water desorption phenomenon in baru fruits can be considered valid. Sousa et al. (2015) and Silva et al. (2016) verified similar behavior for fodder radish seeds and pepper, respectively.

Table 5 shows the results of the Krug Test and the isokinetic temperature for baru fruits.

TABLE 5. Isokinetic temperature for the desorption process in baru fruits (*Dipteryx alata Vogel*).

Isokinetic temperature (T_B)	497.81
Harmonic mean temperature T_{hm}	299.48
Isokinetic temperature Variance $Var(T_B)$	0.000039
Isokinetic temperature interval	[497.83; 497.79]

The harmonic mean value obtained is outside the isokinetic temperature range and was different from the isokinetic temperature. Also in Table 5, the isokinetic temperature was higher than the harmonic mean temperature, indicating that the process is controlled by enthalpy. These results are in agreement with several researchers who have successfully applied the isokinetic theory on the sorption of several products (Lago et al., 2013; Sousa et al., 2015; Lago & Noreña, 2015; Silva et al., 2016).

Table 6 shows the Gibbs free energy as a function of moisture contents for each temperature during the desorption process in baru fruits.

TABLE 6. Gibbs free energy as a function of baru fruits moisture content (*Dipteryx alata Vogel*).

Xe (% db)	Temperature (°C)			
	20	25	30	35
4.2	332.91	324.66	316.53	308.52
4.5	312.91	305.16	297.51	289.98
4.6	301.72	294.25	286.88	279.62
5.7	229.10	223.43	217.83	212.32
6.8	186.29	181.67	177.12	172.64
7.7	158.90	154.96	151.08	147.25
8.6	137.04	133.64	130.29	127.00
8.8	134.25	130.93	127.65	124.42
9.0	130.56	127.32	124.14	120.99
9.1	128.56	125.37	122.23	119.14
9.7	118.38	115.44	112.55	109.70
12.4	87.13	84.98	82.85	80.75
21.7	43.46	42.38	41.32	40.28
24.1	38.11	37.16	36.23	35.31
24.3	37.63	36.69	35.78	34.87
25.9	34.71	33.85	33.01	32.17
28.4	31.05	30.28	29.52	28.78
29.5	29.53	28.79	28.07	27.36

Gibbs free energy increases with decreasing moisture content, being positive for all temperatures studied, with a tendency to stabilize at higher levels of equilibrium moisture content. This behavior was also observed by Corrêa et al. (2015) and by Sousa et al. (2015) when studying cucumber and fodder radish seeds, respectively.

Gibbs free energy is related to the work required to make sorption sites available (Nkolo Meze'e et al., 2008). The positive Gibbs free energy values are characteristic of an exogenous reaction, that is, one that requires an external agent to supply energy to the environment. These positive values are expected, since desorption is a non-spontaneous process, as verified in the present study for the baru fruits, and corroborate the results obtained by Sousa et al. (2015), and Oliveira et al. (2014b).

The Gibbs free energy for each temperature can be described by exponential regression, [eq. (18)]:

$$G = \alpha \cdot \text{Exp}(-\beta \cdot X_e^*) + \delta \quad (18)$$

In which:

α , β and δ : equation regression parameters.

Table 7 shows the Gibbs free energy regression equations for baru fruits. The equations presented high coefficients of determination and low relative mean error, and all the equation parameters were significant at 1% of significance by the t-test, so the Gibbs free energy can be determined for the temperatures studied. Note that the parameters α and δ decrease with increasing temperature and parameter β was the same for all temperatures.

TABLE 7. Gibbs free energy equations for the various temperatures.

Temperature (°C)	Equation	R ² (%)	P (%)
20	$\Delta G = 798.729^{**} \exp(-0.238^{**} X_e) + 34.812^{**}$	99.73	4.869
25	$\Delta G = 778.944^{**} \exp(-0.238^{**} X_e) + 33.949^{**}$	99.73	4.869
30	$\Delta G = 759.435^{**} \exp(-0.238^{**} X_e) + 33.099^{**}$	99.73	4.869
35	$\Delta G = 740.213^{**} \exp(-0.238^{**} X_e) + 32.261^{**}$	99.73	4.869

** Significant at 1% by t-test.

CONCLUSIONS

Thermodynamic properties of baru fruits are influenced by moisture content, increasing the energy required for water removal from the product, with the reduction in moisture content.

The latent heat of vaporization, enthalpy, entropy and the Gibbs free energy increase with moisture content reduction in baru fruits, and the desorption process is controlled by enthalpy.

Gibbs free energy is positive at temperatures of 20, 25, 30 and 35 °C, and increases throughout desorption, being a non-spontaneous process.

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