ea engenharia agrícola

Journal of the Brazilian Association of Agricultural Engineering
ISSN: 1809-4430 (on-line)

DRYING KINETICS OF PEANUT KERNELS IN THIN LAYERS

Doi:http://dx.doi.org/10.1590/1809-4430-Eng.Agric.v37n5p994-1003/2017

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ABSTRACT: The aim of this study was to adjust mathematical models to the experimental data on the peanut kernels drying in thin layer, as well as to determine the effective diffusion coefficient and the main thermodynamic properties involved in drying of the grains under different air conditions. The peanut kernels, with a water content of 0.59 ± 0.002 (dry basis, d.b.), were dried in a forced air ventilation oven with different temperature levels (40, 50, 60 and 70 ºC) until reaching a water content of 0.04 ± 0.001 (d.b.). For the experimental data, eight mathematical models traditionally used to represent the drying kinetics of thin layer of agricultural products were adjusted, also being determined the effective diffusion coefficient and the thermodynamic properties, specific enthalpy and entropy and Gibbs free energy. The Diffusion Approximation, Two Terms, Midilli, Page and Thompson models represent the drying kinetics of peanut kernels in thin layers. The effective diffusivity increases with the increase of temperature, being the relation with the drying temperature described by the Arrhenius' equation. The increase in temperature promotes the decrease of the enthalpy and entropy values and the increase of the Gibbs free energy.

KEYWORDS: *Arachis hypogaea* L., effective diffusivity, activation energy, Page model, thermodynamic properties.

INTRODUCTION

With the growing search for alternative sources of fuels, oil crops such as peanut (*Arachis hypogaea* L.) can gain even more space in the Brazilian economic scenario. Oils from vegetable products appear as an alternative to substitute for traditional petroleum fuels (Santos et al., 2012). With the possibility of using peanut oil as a raw material for the production of biodiesel, it is important to study the pre-processing of this product, aiming at the conservation and maintenance of the same in this stage of the production chain.

Like most agricultural products, peanuts are harvested with unsuitable water content for storage. In this sense, the use of drying process to reduce the water content of this agricultural product is essential for its conservation.

For the study of drying systems, mathematical simulations based on the drying of successive thin layers of the product are carried out. For the drying of products in thin layer, mathematical models are used to represent the water reduction of the material in function of time, being them theoretical, semi-theoretical and empirical.

The semi-theoretical models are usually derivations of Fick's second law or simplified model modifications, although they are restricted only to the ranges of temperature, relative humidity, air velocity and water content, which are the main characteristic of empirical models. Different authors, in several types of agricultural products, have applied these models (Oliveira et al., 2012; Morais et al., 2013; Siqueira et al., 2013; Resende et al., 2014; Costa et al., 2015).

The theoretical models consider the external conditions and the internal mechanisms of energy and mass transfer between the drying air and the product. The most studied theoretical model is the diffusion, being applied in drying processes of a solid to the descending rate,

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Received in: 11-28-2016
Accepted in: 5-28-2017

considering, as main mechanism, the diffusion based on the Fick's second Law, which describes that the mass flow per area unit is proportional to the water concentration gradient.

The diffusion in agricultural products during the drying is a complex process that may involve different mechanisms such as molecular diffusion, capillary diffusion, surface diffusion, hydrodynamic flow, vapor diffusion and thermal diffusion. Since the models based on Fick's second Law are not strictly representative of the several mechanisms that prevail in water transport in agricultural products, the determined diffusion coefficient is considered to be apparent or effective (Roca et al., 2008).

With the values of the effective diffusion coefficient, it is possible to determine the main thermodynamic properties involved in drying, such as enthalpy, entropy and Gibbs free energy. According to Oliveira et al. (2013), this information gives us an indication of the adsorbent affinity level (product dry matter) by water, as well as the spontaneity of the sorption process.

The aim of this research was to study the drying kinetics of peanut kernels, adjusting different mathematical models to the observed data, as well as obtaining the effective diffusion coefficient and the thermodynamic properties during the product drying.

MATERIAL AND METHODS

This study was carried out in the Laboratory of Physical Properties of Agricultural Products of the School of Agricultural Sciences, belonging to the Federal University of Grande Dourados, located in the city of Dourados, MS.

Peanut kernels of the IAC 505 cultivar, from the vegetative and commercial group Runner, with initial water content of 0.59 ± 0.002 dry basis (d.b.) were used, being determined by the oven gravimetric method, at 105 ± 3°C, during 24 h in triplicate (MAPA, 2009).

The drying of the product in thin layer, with a thickness of approximately 3 cm was carried out in a forced ventilation oven under different controlled temperature conditions (40, 50, 60 and 70 ± 1°C) and their respective relative humidity (19, 12, 7 and 6 ± 0.5%). For each treatment, two metal trays with a diameter of 30 cm were placed inside the oven, with a mesh bottom to allow air to pass through the thin layer of the product. The temperature and the air relative humidity were monitored with the aid of three thermo-hygrometers installed in the environment where the oven was. Thus, with the average values of air temperature, air relative humidity and the air-regulated temperature for the drying, the relative humidity of the air inside the oven was mathematically calculated.

The reduction of the water content of the peanut kernels during drying was accompanied by the use of a scale with a resolution of 0.01 g by gravimetric method (mass difference). During the drying process, the trays with sample were weighed periodically, where the interval between the readings was controlled by the difference in mass between one reading and another (the initial water content being known), avoiding high differences in water content between successive readings. The water content considered as the final drying point for mathematical modeling purposes was 0.04 ± 0.001 d.b. for the peanut kernels.

The humidity ratio of the peanut kernels during the drying in the different air conditions was determined by the [eq. (1)].

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

that,

RX – humidity ratio of the product, dimensionless;

X – water content of the product, decimal d.b.;

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

Xi – initial water content of the product, decimal d.b.

The equilibrium water content data required to determine the humidity ratio at temperature (T, in °C) and relative humidity (RH, in decimal) were obtained from the study developed by Corrêa et al. (2007), according to [eq. (2)].

$$X_e = \left[\frac{\ln (1 - RH)}{-0.0003\,(T + 85.5418)} \right]^{\left(\frac{1}{1.7518}\right)} \tag{2}$$

To the experimental data of humidity ratio obtained during the peanut kernels drying, eight mathematical models (Table 1) commonly used to predict the drying phenomenon of agricultural products were adjusted (Ferreira et al., 2012; Oliveira et al., 2012; Alves et al., 2013; Siqueira et al., 2013; Resende et al., 2014; Costa et al., 2015).

TABLE 1. Mathematical models used to predict the drying of agricultural products on thin layer.

Model designation	Models	
Diffusion approach	$RX = a \exp(-k\,\theta) + (1 - a) \exp(-k\,b\,\theta)$	(3)
Two terms	$RX = a \exp(-k_o\,\theta) + b \exp(-k_1\,\theta)$	(4)
Two terms exponential	$RX = a \exp(-k\,\theta) + (1 - a) \exp(-k\,a\,\theta)$	(5)
Henderson and Pabis modified	$RX = a \exp(-k\,\theta) + b \exp(-k_0\,\theta) + c \exp(-k_1\,\theta)$	(6)
Midilli	$RX = a \exp(-k\,\theta^n) + b\,\theta$	(7)
Page	$RX = \exp(-k\,\theta^n)$	(8)
Thompson	$RX = \exp\{[-a - (a^2 + 4\,b\,\theta)^{0.5}]/2b\}$	(9)
Verma	$RX = a \exp(-k\,\theta) + (1 - a) \exp(-k_1\,\theta)$	(10)

k, k_o, k_1 - drying constants (h^{-1}); a, b, c, n - coefficients of the models; θ - drying time (h).

The effective diffusion coefficient of the peanut kernels, for the different drying air temperature conditions (40, 50, 60 and 70 ºC), was calculated using Equation 11, based on the theory of liquid diffusion. This equation is the analytical solution for Fick's second law, with eight terms, in order to improve the quality of the adjustment and considering the geometric shape of the product as spherical (Morais et al., 2013).

$$RX = \frac{X - X_e}{X_i - X_e} = \frac{6}{\pi^2} \sum_{n_t=1}^{\infty} \frac{1}{n_t^2} \exp\left[-\frac{n_t^2 . \pi^2 . D_i . \theta}{9} \left(\frac{3}{R_e} \right)^2 \right] \tag{11}$$

that,

D_i- diffusion coefficient, $m^2\,s^{-1}$;

n_t - number of terms, and

R_e - Equivalent radius of the kernels, m.

The equivalent radius used in [eq. (11)], considering it as 0.00614 m, was determined by measuring the axes of length (a), width (b) and thickness (c) of the product, in mm, using a digital caliper, resolution of 0.01 mm in 60 randomly selected kernels, and calculated by the following expression (Morais et al., 2013; Camicia et al., 2015):

$$V_k = \frac{\pi\,(a\,b\,c)}{6} = \frac{4\,\pi\,R_e^3}{3} \tag{12}$$

that,

V_k- volume of the kernel, mm^3;

To evaluate the influence of temperature on the effective diffusion coefficient, the Arrhenius expression was used, according to [eq. (13)].

$$D_i = D_o \exp\left(\frac{E_a}{R\,T_a}\right) \tag{13}$$

that,

D_o - pre-exponential factor;

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

R - gas universal constant, 8.314 kJ kmol^{-1} K^{-1}, and

T_a - absolute temperature, K.

The thermodynamic properties, specific enthalpy, specific entropy and Gibbs free energy, related to the drying process of the peanut kernels, were determined according to eqs (14), (15) and (16), respectively (Corrêa et al, 2012).

$$\Delta h = E_a - RT_a \tag{14}$$

$$\Delta s = R\left(\ln D_0 - \ln\frac{k_B}{h_p} - \ln T_a\right) \tag{15}$$

$$\Delta G = \Delta h - T_a \Delta s \tag{16}$$

that,

Δh - specific enthalpy, J mol^{-1};

Δs - specific entropy, J mol^{-1} K^{-1};

ΔG – Gibbs free energy, J mol^{-1};

k_B - Boltzmann's constant, 1.38 x 10^{-23} J K^{-1}, and

h_P - Planck's constant, 6.626 x 10^{-34} J s^{-1}.

The experimental data of humidity ratio, obtained during the peanut kernels drying, were submitted to nonlinear regression analysis and selection of the appropriate mathematical model to express the relation between the studied variables. The degree of adjustment of each mathematical model, in all drying conditions, was analyzed by the magnitudes of the coefficient of determination (R^2, in decimal), the relative average error (P, in %) and the standard deviation of the estimate (SE, in decimal), as well as the waste distribution tendency (random or biased). For the adjustment of the mathematical models to the experimental data, the statistical software STATISTICA 7.0 was used.

The relative average error (P) and the standard deviation of the estimate (SE), for each one of the models, were calculated by eqs (17) and (18), respectively.

$$P = \frac{100}{n}\sum_{i=1}^{n}\left(\frac{\left|Y - \hat{Y}\right|}{Y}\right) \tag{17}$$

$$SE = \sqrt{\frac{\sum_{i=1}^{n}\left(Y - \hat{Y}\right)^2}{GLR}} \tag{18}$$

that,

n - number of experimental observations;

Y - value observed experimentally;

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

GLR - degrees of freedom of the model.

RESULTS AND DISCUSSION

All mathematic models adjusted to the experimental data of humidity ratio presented determination coefficients above 0.99 in all drying air conditions (Table 2), which should not, however, be the only criterion for evaluation among non-linear mathematical models, and it is necessary the jointly analyze between other statistical parameters (Siqueira et al., 2012; Camicia et al., 2015; Costa et al., 2015).

TABLE 2. Statistical parameters (SE - standard deviation of the estimate, P – relative average error and R^2 - coefficient of determination) and waste distribution (A = random or T = tendentious) for the models analyzed in the peanut kernels drying.

Models	SE (decimal)	P (%)	R^2 (decimal)	Waste	SE (decimal)	P (%)	R^2 (decimal)	Waste
	40 °C				50 °C			
(3)	0.0103	2.3958	0.9989	A	0.0045	1.1493	0.9997	A
(4)	0.009	1.7799	0.9992	A	0.0046	1.1682	0.9997	A
(5)	0.0201	13.3112	0.9958	A	0.0184	10.4244	0.9957	A
(6)	0.0094	2.4386	0.9999	A	0.0049	1.1593	0.9997	A
(7)	0.018	7.2147	0.997	A	0.0103	2.882	0.9988	A
(8)	0.0207	8.3947	0.9956	A	0.011	3.1009	0.9984	A
(9)	0.0125	3.3914	0.9983	A	0.0069	3.5492	0.9993	A
(10)	0.0103	2.3954	0.9989	T	0.0045	1.1493	0.9997	T
Models	60 °C				70 °C			
(3)	0.0026	1.1628	0.9999	A	0.006	4.1218	0.9995	A
(4)	0.0027	1.1595	0.9999	A	0.0063	4.1409	0.9995	A
(5)	0.0118	6.8447	0.9983	A	0.011	7.7247	0.9985	A
(6)	0.0026	0.6604	0.9999	A	0.0051	1.5238	0.9997	T
(7)	0.0059	1.598	0.9996	A	0.0068	1.5817	0.9995	A
(8)	0.006	1.5964	0.9995	A	0.0064	2.3327	0.9995	A
(9)	0.0063	3.7976	0.9995	A	0.0093	4.7159	0.9989	A
(10)	0.0026	1.1628	0.9999	A	0.006	4.1218	0.9995	A

Only the Exponential Model of Two Terms (5), among all tested, presented relative average error magnitudes higher than 10% (Table 2), considering all the drying temperatures evaluated. According to Kashaninejad et al. (2007), the values of the relative average error indicate the deviation of the values observed in relation to the curve estimated by the model. In addition, traditionally in studies involving the drying kinetics of agricultural products, values higher than 10% of the average relative error are adopted as unsuitable for the description of this process (Ferreira et al., 2012; Oliveira et al., 2012; Siqueira et al., 2013; Resende et al., 2014; Costa et al., 2015).

In Table 2, we also verified that only the Modified Henderson and Pabis (6) and the Verma (10) Models did not have random distribution of the residues for all drying air conditions, they are also not suitable for representing the kinetics of the peanut kernels drying.

Except for the models already discarded by the statistical parameters of comparison, it is noticed that the Diffusion Approximation (3), Two Terms (4), Modified Henderson and Pabis (6), Midilli (7) and Page (8) Models present reduced magnitudes of the standard deviation of the estimate for all drying air conditions. The standard deviation of the estimate indicates the ability of a model to faithfully describe a given physical process, and the lower is its magnitude the better the model adjustment quality will be in relation to the observed data (Draper & Smith, 1998).

The Exponential Models of Two Terms (5), Modified Henderson and Pabis (6) and Verma (10) Models, among the eight models tested, did not satisfactorily represent the drying kinetics of peanut kernels (Table 2). Thus, all other mathematical models adjusted to the experimental data can be recommended and used to represent the drying kinetics of the peanut kernels under the conditions of this study.

Among the recommended models for representing the drying kinetics in thin layer of peanut kernels, the traditional Page (8) model is the simplest mathematically, with a small number of coefficients, thus simplifying its application in drying simulations. Thus, the Page model was selected to represent the drying kinetics of peanut kernels, as several researchers have also recommended it to represent the drying kinetics of different agricultural products (Siqueira et al., 2013; Resende et al., 2014; Costa et al., 2015).

The adequate adjustment of the Page model to describe the peanut kernels drying in thin layer is evidenced by the proximity of the observed values in relation to those estimated by the model in all drying air conditions studied (Figure 1).

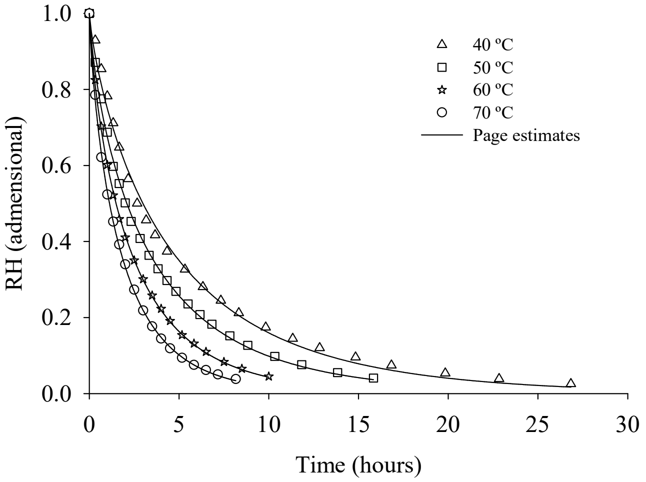

FIGURE 1. Experimental humidity ratio and estimated values, by Page model for the peanut kernels drying in thin layer.

In Figure 1 it is also possible to observe the influence of the air temperature on the drying curves, where the increase of the air temperature considerably reduced the drying time. The rise of the temperature increases the difference between the partial steam pressure of the drying air and the product during the water removal (Siqueira et al., 2012). Thus, there is an increase in the heat and mass transfer potential during that process, resulting in increases in the drying rate and, consequently, decreases in the drying time (Ferreira et al., 2012).

In fact, this was observed in this study, where peanut kernels need 26.8; 15.8; 10.0 and 7.2 hours, at temperatures of 40, 50, 60 and 70°C, respectively, to reach the water content of 0.04 ± 0.001 d.b. Several researchers observed similar results in other agricultural products (Oliveira et al., 2012; Alves et al., 2013; Siqueira et al., 2013; Resende et al., 2014; Camicia et al., 2015; Costa et al., 2015).

As for the coefficients of the Page model, only the “k” coefficient presented a variation with a definite tendency in its magnitudes with the drying air temperature variation (Table 3). As defined in other studies (Siqueira et al., 2013; Resende et al., 2014; Costa et al., 2015), the “k” coefficient also known as the drying constant, which represents the external drying conditions, can be used as an approximation to characterize the temperature effect and is related to the effective diffusivity in the drying process in the decreasing period and to the liquid diffusion which controls the process.

TABLE 3. Page model coefficients and effective diffusion coefficients (D_i) adjusted to different drying temperatures for peanut kernels in thin layer.

Temperature (°C)	Page model coefficients		D_i x 10^{-10}
	k (h^{-1})	n (dimensionless)	(m^2 s^{-1})
40	0.2867	0.8060	1.1831
50	0.3914	0.7726	1.6988
60	0.5041	0.7948	2.4724
70	0.6299	0.8006	3.3502

With the increase in temperature, there was an increase in the “k” coefficient (Table 3), indicating a higher water movement from the center to the product surface and, consequently, a shorter drying time. The elevation of the “k” coefficient of the Page model, by raising the drying air temperature was an expected behavior as reported for different agricultural products (Siqueira et al., 2013; Resende et al., 2014; Costa et al., 2015). Yet for the “n” coefficient which reflects the internal resistance of the product to drying (Siqueira et al., 2013), there was no tendency observed in its values in function of temperature increase.

The values of the effective diffusion coefficient of the peanut kernels increased with the increase of the drying air temperature (Table 3). According to Oliveira et al. (2012), the diffusivity can be described as the easiness with which the water is removed from a certain material, being the same dependent on the drying air temperature. The increase in temperature promotes the reduction of the water viscosity, which is a measure of the fluid resistance to the flow, promoting changes in the diffusion of water in the capillaries of the product, favoring the displacement of this fluid from the interior to the ends of the product (Goneli et al., 2007). In addition, when the temperature rises, there is an increase in the molecular vibrations of the water, contributing to the diffusion occurs faster (Baptestini et al., 2014).

The variations between the values of the effective diffusion coefficient of the peanut kernels (Table 3) were between 1.1831 to 3.3502 × 10^{-10} m^2 s^{-1} for the temperature range from 40 to 70°C. The values of the diffusion coefficient are related to the drying conditions and structural, chemical and geometrical particularities presented by each biological material (Ferreira et al., 2012; Oliveira et al., 2012; Baptestini et al., 2014; Resende et al., 2014).

In Figure 2 it is possible to verify that the values of the effective diffusion coefficient linearly increased its dependence on the drying air temperature (Figure 2).According to Alves et al. (2013) and Baptestini et al. (2014), the slope of the Arrhenius curve gives the E_a/R ratio, while its intersection with the ordinate axis indicates the value of D_o.

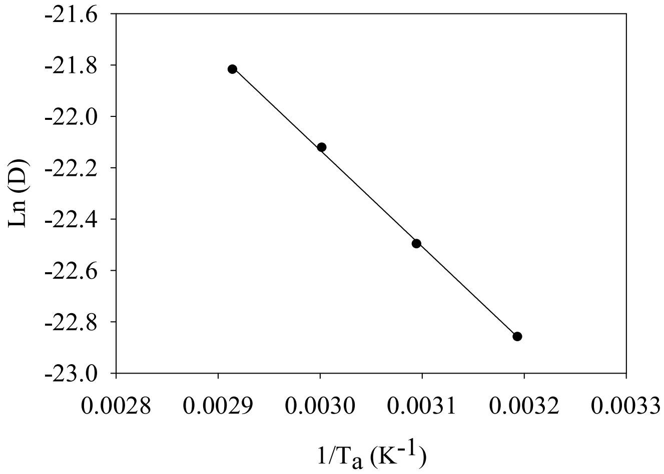

FIGURE 2. Arrhenius representation for the effective diffusion coefficient during the peanut kernels drying at different air temperatures in thin layer.

The [eq. (19)] presents the coefficients of the Arrhenius' equation adjusted for the effective diffusion coefficient of the peanut kernels, calculated according to [eq. (11)].

$$D_i = 1.9416 \times 10^{-5} \exp\left(-\frac{31.2668}{R\,T_a}\right) \tag{19}$$

In this study, the activation energy for the liquid diffusion during the drying was approximately 31.27 kJ mol^{-1} for the peanut kernels in the studied temperature range (40 to 70 ° C). According to Kashaninejad et al. (2007), the activation energy is a barrier that must be overcome so that the diffusion process can be triggered in the product. The lower the activation energy is, the greater will be the water diffusivity in the product during the drying process (Morais et al., 2013).The value of the activation energy obtained for the peanut kernels resembles those found for corn grains (26.79 kJ mol^{-1}, Baptestini et al., 2014) and pistachio nuts (30.79 kJ mol^{-1}, Kashaninejad (Table 1), which was significantly lower than that observed for wheat grains (42.00 kJ mol^{-1}, Goneli et al., 2007) and higher than that of crambe grains (21.00 kJ mol^{-1}, Costa et al., 2015).

The different activation energy magnitudes, verified for agricultural products of different species and even genotypes of the same species, may be related to the temperature used for drying as well as to the initial water content of the product (Costa et al., 2015), and, mainly, to the chemical constituents of the evaluated biological material (Goneli et al., 2007).

The enthalpy values were reduced during the peanut kernels drying with the increase of the drying temperature (Table 4). Considering the kernel as a thermodynamic system, the increasing of the drying air temperature promotes an increase of the partial pressure of water vapor in the kernel, while the one of the air remains constant. Thus, there is an increase in the water diffusion velocity from the interior to the kernel surface and, consequently, the loss of kernel water by desorption. The energy required for the water removal of the product, which in the drying occurs by diffusion, is composed of the enthalpy of vaporization of free water and the enthalpy of vaporization of the water in the product or the isosteric heat (Goneli et al., 2013).

TABLE 4. Thermodynamic properties, specific enthalpy (Δh), specific entropy (Δs) and Gibbs free energy (ΔG) of the peanut kernels drying process in thin layer.

Temperature (°C)	Δh ($J\ mol^{-1}$)	Δs ($J\ mol^{-1}K^{-1}$)	ΔG ($J\ mol^{-1}$)
40	28663.2	-182.4	85773.3
50	28580.0	-182.6	87598.3
60	28496.9	-182.9	89425.8
70	28413.8	-183.1	91255.9

In all the air temperatures used, the initial and final water content variation was the same, so the energy needed to break the molecules bonds of the water-water and adsorbent water-surface (isosteric heat) was constant, occurring variations only in the enthalpy of free water vaporization. With the increase in temperature and consequent increase of the partial vapor pressure of the water inside the kernel, occurs the reduction of the enthalpy of the vaporization of the free water, and therefore, in the final enthalpy balance, with the increase of the air temperature of the drying, the reduction in the enthalpy of the water diffusion process in the product during the drying occurs.

Analyzing the behavior of the entropy in Table 4, it is observed that, for the peanut kernels, this thermodynamic property behaved in a similar way to the enthalpy, being its values reduced with the temperature increase. The entropy is a thermodynamic quantity linked to the degree of disorder, where its values rise during a natural process in an isolated system (Goneli et al., 2013). With the increase in the drying air temperature and consequent increase in the partial vapor pressure of the water in the product, there is also an increase in the excitation of the molecules and reduction of the water viscosity, factors that when are combined provide the increase of the water diffusion velocity and the reduction of the entropy in the process. The negative entropy values may be attributed to the existence of chemical adsorption and/or structural modifications of the adsorbent (Moreira et al., 2008).

Also in Table 4, there was an increase in the Gibbs free energy values, in proportion to the increase of the drying air temperature, for the peanut kernels. As previously discussed for enthalpy and entropy, the increase of the drying air temperature promotes an increase in water diffusion to the product surface, indicating more work done. The Gibbs free energy is a thermodynamic function responsible for quantifying the maximum energy released in a process, and under conditions of constant temperature and pressure, the Gibbs free energy can be an indicative of useful work done (Oliveira et al., 2015). The positive values indicate that under these conditions the water diffusion in the product was not spontaneous and that there was energy consumption of the medium so that the reaction occurred (Resende et al., 2014).

CONCLUSIONS

The Diffusion Approximation, Two Terms, Midilli, Page and Thompson Models can be used to represent the drying kinetics of peanut kernels in thin layers.

The effective diffusion coefficient increases with the increase of the drying air temperature and this relation can be described by the Arrhenius' equation.

The specific enthalpy and the specific entropy decrease with the increase of the drying air temperature, while Gibbs free energy values increase.

ACKNOWLEDGEMENTS

To financial support for research by the Coordination of Improvement of Higher Personnel Education - Capes, and the Foundation for Support to the Development of Education, Science and Technology of the State of Mato Grosso do Sul - Fundect.

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