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Drying kinetics of *Aristolochia cymbifera* Mart. and Zucc. leaves

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Drying medicinal plant materials ensures the retention of their active ingredients. The aim of this study was to fit different mathematical models to experimental data from *Aristolochia cymbifera* leaves to determine and evaluate the effective diffusion coefficient and to obtain the drying process activation energy under varying air conditions. The experiment was conducted at the Federal Institute of Education, Science and Technology at the Goiás – Rio Verde Campus. Four drying replicates were performed in a fixed bed dryer with a controlled air speed of 1.0 m s^{-1} and drying temperatures of 28.8, 36.4 and 44.8°C. Increasing the drying air temperature from 28.8 to 36.4 or 44.8°C reduced the *A. cymbifera* leaf drying time from 58.13 to 13.10 and 5.00 h, respectively. The diffusion approximation model described the drying phenomenon the best. The effective diffusion coefficient increased with increasing air temperature and was described by the Arrhenius equation with activation energy of 107.29 kJ mol^{-1}.

Key words: Medicinal plants, jarrinha, activation energy, effective diffusion.

INTRODUCTION

The demand for medicinal plants, herbs and spices has increased, especially for pharmaceutical and natural food products (Martinazzo et al., 2007). These products have political environmental preservation appeal, which is used as a marketing tool. Furthermore, they provide a great opportunity for the development of sustainable biodiversity utilization processes (Bizzo et al., 2009).

The Brazil market for natural products, such as essential oils, stands out. The Cerrado is a biome that contains genetic resources of great medicinal diversity. A total of 509 species described as medicinal were found in the State of Mato Grosso, with more than 600 species projected to occur across the biome according to a literature review (Guarim Neto and Morais, 2003). These values surpass the estimates made in other studies. Among the species within the *Aristolochia* (Aristolochiaceae) genus, we cite the *Aristolochia cymbifera* Mart. and Zucc. species, which is popularly known as jarrinha, milhomem or cassaú. This species is a vigorous herbaceous perennial vine that is native to Brazil and better adapted to hot environments. In popular medicine, *A. cymbifera* is used for several problems and

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is considered a diuretic, a sedative and an antiseptic (Lorenzi and Matos, 2002).

Among the post-harvesting steps, drying the plant material ensures that the components do not degrade and that the active ingredients are preserved. This preservation occurs because the drying process inhibits or reduces the enzymatic action. Drying also makes it easier to store the plant materials for extended periods of time (Chudnicka and Matysik, 2005). By reducing the available moisture content, the water activity and the growth of microorganisms is decreased.

Theoretical information from simulations that show the behavior of each product as the moisture content decreases is useful for improving and developing drying equipment. The use of mathematical models to simulate drying operations has improved the design, development, evaluation and optimization of dryers (Palacin et al., 2005).

Water can move within materials by different mechanisms. In porous capillary products, such as most plant products, possible water transport mechanisms include the following: Liquid diffusion, capillary diffusion, surface diffusion, hydrodynamic flow, vapor diffusion and thermal diffusion (Martinazzo et al., 2007).

The theory of liquid diffusion assumes that no influence from capillary diffusion occurs and that the bodies thermally equilibrate with the air instantly. In addition, this theory disregards the effects of mass and energy transfer between bodies. These assumptions can lead to discrepancies between results (Goneli et al., 2007).

Diffusivity is the ease with which water is removed from a material. Because diffusivity varies with drying conditions, it is not intrinsic to the material. Thus, diffusivity is commonly called effective diffusivity (Oliveira et al., 2006).

The ease with which water molecules overcome the energy barrier during migration within a product is called the activation energy. Lower activation energies correspond to greater water diffusivity in products (Corrêa et al., 2007).

Countless studies have been conducted to identify leaf characteristics during drying. For example, black tea (Panchariya et al., 2002), bay leaf (Demir et al., 2004), dill (Doymaz et al., 2006), sage (Radünz et al., 2010) and lemongrass (Rocha et al., 2012) were previously studied.

Theoretical studies of the plant product drying process are important, and thus, the present work aims to fit different mathematical models to experimental data from *A. cymbifera* leaves to determine and evaluate the coefficient of effective diffusion and to obtain the activation energy under varying air conditions.

MATERIALS AND METHODS

The experiment was conducted at the Post-harvest Laboratory of Plant Products of the Federal Institute of Education, Science and Technology at the Goiás - Rio Verde Campus. *A. cymbifera* Mart. and Zucc. leaves were used from the municipality of Rio Verde – GO. The *A. cymbifera* plants were collected in the Rio Grande region (S 17°55'56.8" WO 50º56'33.2" and height of 682 m) between 17:00 and 18:00, in April of 2012. The voucher specimen is registered at the Herbarium Jataiense under number 5,642.

The material was harvested by cutting the shoots 5 cm above the ground, packing them in plastic bags and then sending them to the Natural Products section at the Laboratory of Plant Tissue Culture (Federal Institute of Education, Science and Technology at the Goiás, Rio Verde Campus). After harvesting, the plants were subjected to defoliation and selection. The plants that were ill or had been attacked by insects were discarded.

The moisture content was determined before and after drying with the method described by ASAE (2000) for forage and similar plants (plants or leaves). To determine the moisture content, four replicates of the leaves were placed in a convection oven at 103 ± 2°C for 24 h.

The initial moisture content of the leaves was 2.22 db (decimal dry basis). During the drying process, the samples were weighed periodically until they reached a moisture content of 0.1198 db.

Drying was performed in a fixed bed dryer that was made of number 16 sheet metal. The drying chamber measured 0.60 × 0.60 × 0.60 m had a total volume of 0.216 m^3 and had a 25% perforation plate located at a height of 0.33 m. The fan was centrifugal and was driven by a three-phase motor with an output of 1.5 hp at 1,720 rpm. The fan consisted of a rotor, blades, a volute and a support. The drying chamber and fan were connected by an expansion element. This expansion element converts the 0.20 × 0.20 m cross section at the fan outlet to the 0.57 m × 0.03 m entrance of the drying chamber over a length of 0.64 m (Figure 1).

Each dryer had six pendular temperature sensors and four 1,500 watt electrical resistors for a total of 6,000 watts. The sensors were placed before and after the resistors and within each tray. Four removable trays with perforated bottoms measuring 0.28 × 0.28 × 0.15 m were placed in the drying chamber (Figure 2). The system also featured an automated controller that managed the system and stored the generated data.

The jarrinha leaves were wrapped in a voile fabric and spread out on the tray, comprising a layer of approximately 0.06 m. The system was set to heat at 44.8 ± 0.8, 36.4 ± 1.3 or 28.8 ± 1.3°C with a relative humidity of 25.95, 40.31 or 60.88%, respectively, and a controlled air speed of 1.0 m s^{-1}.

The decreasing moisture content during the drying process was monitored until the desired moisture content was reached by using the gravimetric method (mass loss from the initial moisture content). The decrease in mass was monitored during drying with a scale that was accurate to 0.01 g.

The drying air and room temperatures were monitored with thermocouples that were installed inside and outside of the dryer. The relative humidity inside of the dryer was obtained by using basic psychrometric principles with the GRAPSI software.

The following equation was used to determine the moisture content ratios of *A. cymbifera* leaves during the drying process:

$$RX = \frac{X - X_e}{X_i - X_e} \qquad (1)$$

Where, RX is the ratio of the moisture content in the product (dimensionless), X is the moisture content of the product (kg of water kg^{-1} of dry matter), X_i is the initial moisture content of the product (kg of water kg^{-1} of dry matter) and X_e is the equilibrium moisture content of the product (kg of water kg^{-1} of dry matter).

To obtain the equilibrium moisture content of the *A. cymbifera* leaves at each temperature, four replicates were used. Each replicate weighed 15 g and was dried in the same dryer. The leaves remained in the dryers until their mass remained unchanged for

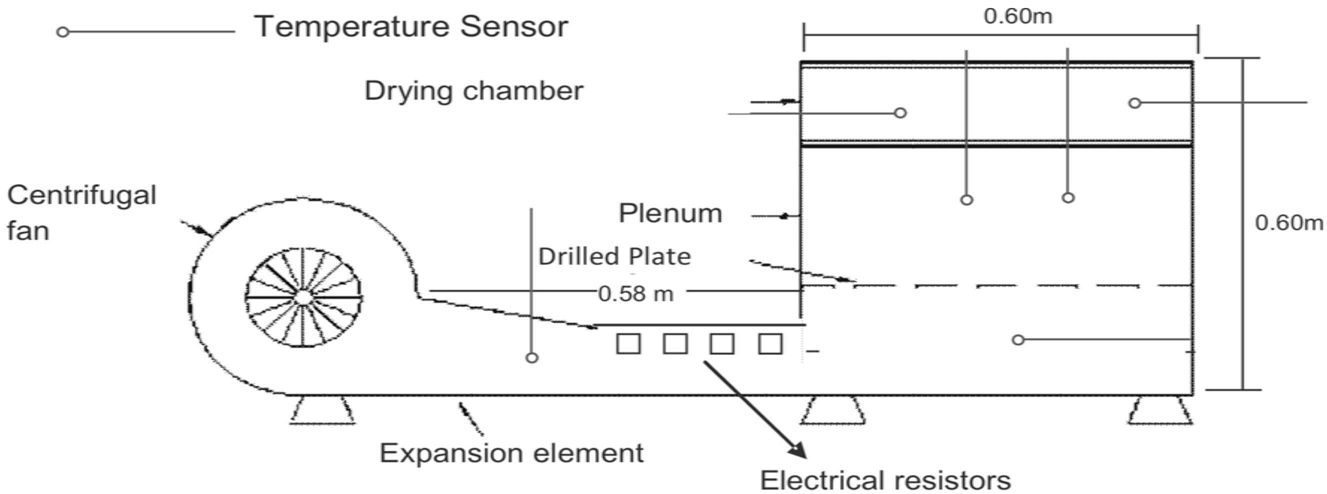

Figure 1. Side view of the experimental dryer.

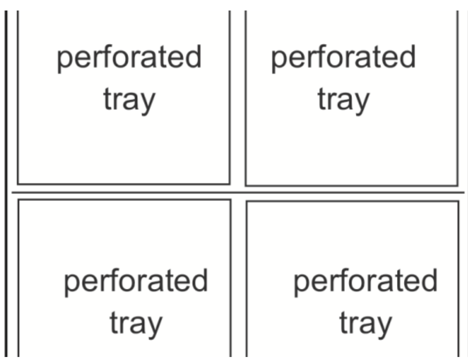

Figure 2. Top view of the experimental dryer and detail of the perforated trays.

three consecutive days. The final moisture content was determined by using the method described by ASAE (2000).

Experimental data from drying the *A. cymbifera* leaves were fit to mathematical models [(2) to (12)] frequently used to represent the drying of plant products (Table 1).The mathematical models were fit using a Gauss-Newton nonlinear regression analysis using statistical software. The magnitude of the coefficient of determination (R^2), the chi-squared test (χ^2), the mean relative error (MRE) and the standard error (SEE) were considered during model selection. According to Mohapatra and Rao (2005), a relative mean error of less than 10% was established as a criterion for model selection.

$$MRE = \frac{100}{N}\sum\frac{\left|Y-\hat{Y}\right|}{Y} \quad (13)$$

$$SEE = \sqrt{\frac{\sum\left(Y-\hat{Y}\right)^2}{GLR}} \quad (14)$$

$$\chi^2 = \frac{\sum\left(Y-\hat{Y}\right)}{DF} \quad (15)$$

Here, Y is the experimentally observed value, $\hat{Y}$ is the value estimated by the model, N is the number of experimental observations and DF refers to the degrees of freedom of the model (number of experimental observations minus the number of model coefficients). The liquid diffusion model for a flat plate geometric shape with a known thickness (Fick's law and an approximation of eight terms Equation 16) was fit to the experimental drying data from *A. cymbifera* leaves as follows:

$$RX = \frac{X-X_e}{X_i-X_e} = \frac{8}{\pi^2}\sum_{n_t=0}^{\infty}\frac{1}{(2n_t+1)}\exp\left[-(2n_t+1)^2\pi^2 D\frac{t}{4L^2}\right] \quad (16)$$

where, n_t is the number of terms and L is the thickness of the product (3.51×10^{-4} m).

The relationship between the coefficient of effective diffusion and increasing drying air temperature has been described by the Arrhenius equation satisfactorily by many researchers (Panchariya et al., 2002; Doymaz et al., 2006; Doymaz, 2006; Goneli et al., 2007; Martinazzo et al., 2007; Kaya and Aydin, 2009), according to the following expression:

$$D = D_o \cdot \exp\left(\frac{-E_a}{R \cdot T_{ab}}\right) \quad (17)$$

where, D_O is a pre-exponential factor, E_a is the activation energy (kJ.mol^{-1}), R is the universal gas constant (8.134 kJ kmol^{-1} K^{-1}) and T_{ab} is the absolute temperature (K). The coefficients of the Arrhenius expression were linearized by applying the following logarithmic equation:

$$LnD = LnD_o - \frac{E_a}{R}\cdot\frac{1}{T_{ab}} \quad (18)$$

RESULTS AND DISCUSSION

Table 2 shows the statistical parameters used to compare the 11 analyzed models to describe the drying kinetics of *A. cymbifera* leaves subjected to various temperature conditions. The model coefficients of determination (R^2) were greater than 95%, except in the Two-term Exponential (9) and the Two-Term (10) models

Table 1. Mathematical models used to predict plant product drying.

Model designation	Model	Equation
$RX = 1 + a \cdot t + b \cdot t^2$	Wang and Sing	(2)
$RX = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k_1 \cdot t)$	Verma	(3)
$RX = \exp\left(\left(-a - \left(a^2 + 4 \cdot b \cdot t\right)^{0,5}\right) / 2 \cdot b\right)$	Thompson	(4)
$RX = \exp(-k \cdot t^n)$	Page	(5)
$RX = \exp(-k \cdot t)$	Newton	(6)
$RX = a \cdot \exp(-k \cdot t) + c$	Logarithmic	(7)
$RX = a \cdot \exp(-k \cdot t)$	Henderson and Pabis	(8)
$RX = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k \cdot a \cdot t)$	Two-term Exponential	(9)
$RX = a \cdot \exp(-k_o \cdot t) + b \cdot \exp(-k_1 \cdot t)$	Two Term	(10)
$RX = a \cdot \exp(-k \cdot t) + (1-a) \cdot \exp(-k \cdot b \cdot t)$	Diffusion Approximation	(11)
$RX = a \cdot \exp(-k \cdot t^n) + b \cdot t$	Midilli	(12)

Table 2. Coefficients of determination (R^2, %), relative (MRE, %) and estimated (SEE, decimal) mean errors for the models analyzed during the drying of *Aristolochia cymbifera* Mart. & Zucc. leaves under various temperature conditions (°C).

Models	28.8°C			36.4°C			44.8°C		
	SEE	MRE	R²	SEE	MRE	R²	SEE	MRE	R²
2	0.065	130.854	95.835	0.035	47.165	98.806	0.053	28.175	97.740
3	0.016	13.036	99.769	0.017	7.643	99.736	0.027	28.779	99.505
4	0.015	13.050	99.769	0.026	37.438	99.342	0.029	24.714	99.317
5	0.015	11.984	99.771	0.022	21.774	99.532	0.028	28.860	99.371
6	0.015	13.036	99.769	0.025	37.439	99.342	0.027	24.679	99.317
7	0.011	13.953	99.874	0.017	14.381	99.756	0.027	12.805	99.494
8	0.014	14.507	99.795	0.028	41.880	99.344	0.028	26.039	99.371
9	0.081	26.635	93.589	0.020	20.436	99.602	0.029	24.681	99.317
10	0.015	14.517	99.795	0.016	10.773	99.785	0.215	62.959	73.259
11	0.015	8.431	99.793	0.017	7.643	99.736	0.027	8.476	99.508
12	0.014	15.876	99.818	0.018	14.549	99.706	0.007	6.696	99.965

for temperatures of 28.8 and 44.8°C, respectively. According to Madamba et al. (1996), these results indicated satisfactory representation for the drying process.

The Diffusion Approximation model (11) produced relative mean errors (MRE) of less than 10% for the three drying conditions tested. The Verma (3) and Midilli (12) models also produced MRE values of less than 10% for 36.4 and 44.8°C, respectively. According to Mohapatra and Rao (2005), models that produce MRE values lower than 10% can adequately represent the studied phenomenon. With respect to the estimated SEE, the 12 models produced small values that represented a good fit between the models and the experimental data.

Table 3 shows the chi-squared test values obtained from fitting the different models to the drying curves of *A. cymbifera* leaves. The 12 analyzed models had relevant chi-squared values. According to Günhan et al. (2005), the lower the chi-squared value, the better the model fit. In general, the Verma (3), Thompson (4), Page (5), Newton (6), Logarithmic (7), Henderson and Pabis (8), Diffusion Approximation (11) and Midilli (12) models produced the lowest chi-squared values.

According to the statistical parameter analysis, the Diffusion Approximation model (11) fit best with the experimental data for the three temperatures studied. Radünz et al. (2010) studied the drying kinetics of sage leaves at 40, 50, 60, 70, 80 and 90°C and found that the Diffusion Approximation model (11) adequately represented the drying kinetics for temperatures between60 and 90°C. Barbosa et al. (2007) evaluated the drying of Brazilian lemon-balm (*Lippia alba* (Mill) N.E. BROWN) leaves and found that the Midilli and Page model satisfactorily represented their drying kinetics.

Table 3. The chi-squared test values calculated for the 11 models that were used to represent the drying kinetics of the *A. cymbifera* Mart. & Zucc leaves.

Models	28.8°C	36.4°C	44.8°C
2	0.00421	0.00125	0.00280
3	0.00024	0.00029	0.00071
4	0.00023	0.00069	0.00085
5	0.00023	0.00049	0.00078
6	0.00023	0.00064	0.00074
7	0.00013	0.00027	0.00073
8	0.00021	0.00080	0.00078
9	0.00649	0.00042	0.00085
10	0.00022	0.00026	0.04634
11	0.00022	0.00029	0.00071
12	0.00019	0.00033	0.00005

Table 4. Coefficients and constants of the Diffusion Approximation model fit to different drying conditions of the *A. cymbifera* Mart. & Zucc. leaves along with the respective equation as a function of temperature.

Parameter	Temperature (°C)			Equation
	28.8	36.4	44.8	
a	12.64470	6.28104	1.00003	a = 33.2641 - 0.7261 T
k	0.09176	0.16136	0.56551	k = - 0.8247 + 0.0299 T
b	1.01839	0.92098	-2.53246	b = 8.0610 – 0.2252 T

Table 4 shows the “a” and “b” coefficient values and the Diffusion Approximation model drying constants “k” (11) fit to the experimental drying kinetics of the *A. cymbifera* leaves at different temperatures. This table also shows the equations used to determine the Diffusion Approximation model coefficients and constants.

Evaluating the constants “k” and the coefficients “a” and “b” revealed that the coefficients decreased with increasing drying temperature. However, k increased with increasing drying temperature. Using these equations, we can determine the coefficients needed to evaluate any drying temperature for *A. cymbifera*.

Figure 3 shows the average moisture content ratio values of the *A. cymbifera* leaves (experimental and those estimated by the Diffusion Approximation model) dried in different air conditions. The times required for the leaves to reach a moisture content of 0.1198 ± 0.0121 db were 58.13, 13.10 and 5.00 h for drying temperatures of 28.8, 36.4 and 44.8°C, respectively. These result showed that increasing the temperature reduced the drying time of the leaves.

The moisture content ratio decreased rapidly. Consequently, the drying rate increased when the air temperature increased. Thus, a higher rate of water removal occurred with increasing air temperature. Similar results were obtained by Martinazzo et al. (2007) and Radünz et al. (2010), who studied lemongrass and sage leaves, respectively.

Figure 4 shows the coefficient of effective diffusion values for the *A. cymbifera* leaves after drying under different air conditions.

The coefficient of effective diffusion increased with increasing drying air temperature. This result agrees with the results of other researchers (Doymaz et al., 2006; Martinazzo et al., 2007). Diffusivity depends on the drying air temperature. The higher the drying air temperature, the lower the resistance of the leaves from water removal and the higher the diffusivity.

The coefficient of effective diffusion values for the *A. cymbifera* leaves ranged from between 1.007 × 10^{-9} and 8.681 × 10^{-9} m^2 s^{-1} for temperatures between 28.8 and 44.8°C. According to Zogzas et al. (1996), the coefficient of effective diffusion values for food products lie between 10^{-11} and 10^{-9} m^2 s^{-1}.

The linear model satisfactorily represented the coefficient of diffusion behavior as a function of drying temperature (with a coefficient of determination of 97.25%).

The coefficient of effective diffusion of the *A. cymbifera* leaves depended on the drying air temperature and was represented by the Arrhenius expression (Figure 5).

The equation presented in Figure 5 represents the linearization the values of the diffusion coefficient and the value of 22.0914 is equal to D_o Ln and the value of12,905 is the ratio of Ea/R.

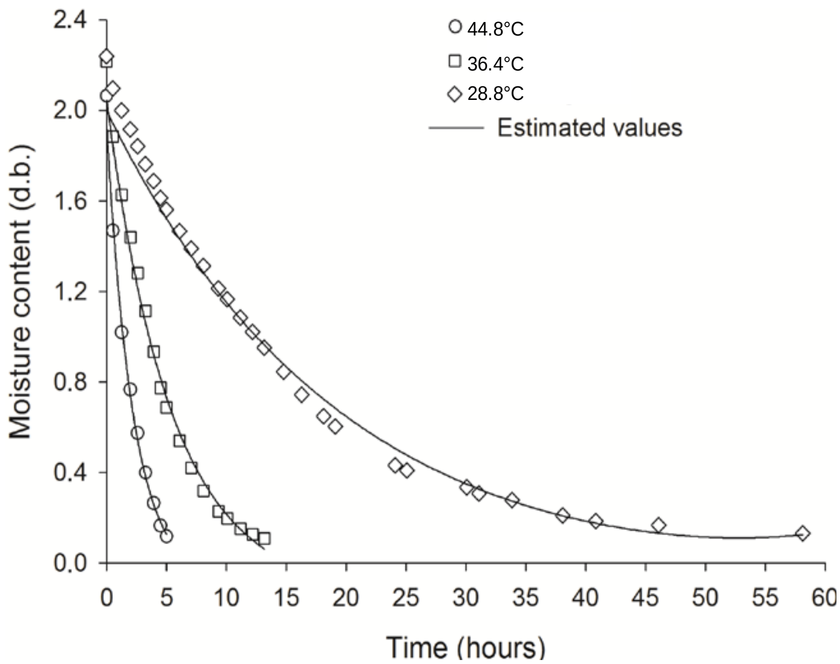

Figure 3. The experimental and estimated (by the Diffusion Approximation model) moisture contents (d.b.) for drying *A. cymbifera* Mart. & Zucc. leaves under various temperature conditions (28.8, 36.4 and 44.8°C).

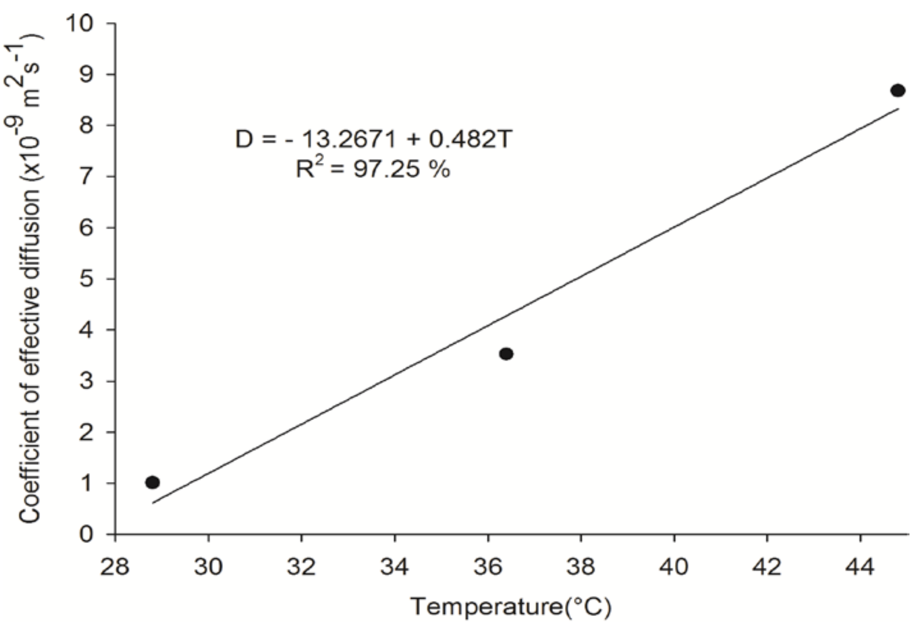

Figure 4. Coefficient of effective diffusion ($m^2\ s^{-1}$) obtained for the drying of *Aristolochia cymbifera* Mart. & Zucc. leaves at temperatures of 28.8, 36.4 and 44.8°C.

The activation energy for drying the *A. cymbifera* leaves was 107.29 kJ mol^{-1}. According to Zogzas et al. (1996), the activation energy for agricultural products ranges from between 12.7 and 110 kJ mol^{-1}. Therefore, the value obtained in this study falls within the previously identified range.

Martinazzo et al. (2007) and Doymaz (2006) found that the activation energy values in lemongrass and dill were lower were 63.47 and 62.96 kJ mol^{-1}, respectively. Kaya and Aydin (2009) found that the activation energy ranged from between 79.87 and 109 kJ mol^{-1} for nettle leaves. The activation energy found for the *A. cymbifera* leaves is

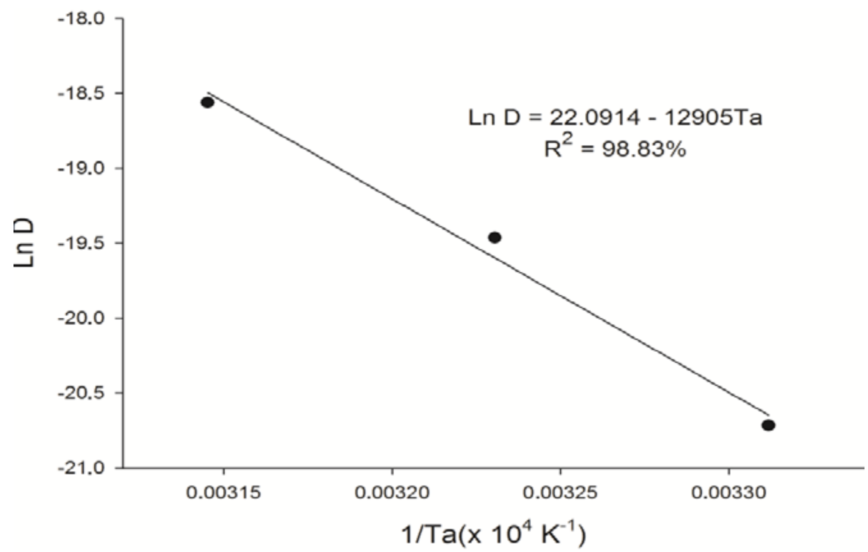

Figure 5. The Arrhenius representation for the coefficient of effective diffusion as a function of the drying air temperature (obtained during the drying of the *Aristolochia cymbifera* Mart. & Zucc leaves).

consistent with previous values found in the literature.

Conclusion

Increasing the drying air temperature reduces the drying time of *A. cymbifera* leaves. The drying times were 58.13, 13.10 and 5.00 h at temperatures of 28.8, 36.4 and 44.8°C, respectively.

For the temperatures studied, the Diffusion Approximation model best described the drying of *A. cymbifera* leaves. The effective diffusion coefficient for the *A. cymbifera* leaves increases with increasing air temperature during the drying process. Specifically, the effective diffusion coefficient was between 1.007×10^{-9} and 8.681×10^{-9} m^2 s^{-1} and had activation energy of 107.29 kJ mol^{-1} for temperatures ranging between 28.8 and 44.8°C.

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