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EVALUATION OF MATHEMATICAL MODELS FOR PREDICTION OF THIN-LAYER DRYING OF BRAZILIAN LEMON-SCENTED VERBENA LEAVES (*Lippia alba* (MILL) N.E. BROWN)

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ABSTRACT

The drying target is to reduce the moisture content and thereby increase the life time of products by limiting enzymatic and oxidative degradation. In addition, by reducing the amount of water, the percentages of active principles are increased in relation to the total mass. The aim of the current study was to determine the correct diffusion rate and activation energy by adapting the mathematical model according to the experimental data. Brazilian lemon-scented verbena leaves were harvested with a moisture content of around 85% wet basis (wb), and were then subjected to drying in a medicinal plant dryer at an air temperatures of 40, 50, 60, 70 and 80 ºC at 0.29±0.03 $m.s^{-1}$. Approximately 400 g of leaves at a layer thickness of 0.15m were used with three tests performed at each temperature. Several models of drying kinetics were adapted for use with the experimental data, including Henderson & Pabis, Henderson & modified Pabis, Lewis, Midilli et al., Page, Thompson and Wang & Singh. The adjusted determination coefficients (R^2), relative mean errors (P), estimated errors (SE) and residue distributions were all compared. The best models, which best represented the Brazilian lemon-scented verbena drying, were Midilli et al. and Page. The calculated effective diffusivity coefficients ranged from 2.91×10^{-12} to 11.71×10^{-12} $m^2.s^{-1}$ for the studied temperature range. The activation energy for the diffusion of water was 31.79 $kJ.mol^{-1}$.

Keywords: activation energy, effective diffusivity, medicinal plants, aromatic plants, kinetics.

AVALIAÇÃO DE MODELOS MATEMÁTICOS PARA A PREDIÇÃO DA SECAGEM EM CAMADA DELGADA DE FOLHAS DE ERVA-CIDREIRA-BRASILEIRA (*Lippia alba* (MILL) N.E. BROWN)

RESUMO

A secagem visa a redução do teor de água, aumentando o tempo de conservação dos produtos pela diminuição da degradação enzimática e oxidativa. Além disso, com a redução da quantidade de água, aumenta-se o percentual de princípios ativos em relação à massa total. Com o objetivo de determinar a difusividade efetiva, a energia de ativação e ajustar modelos matemáticos aos dados experimentais, folhas de erva-cidreira-brasileira, colhidas com teor de água em torno a 85 %b.u. foram submetidas à secagem em secador de plantas medicinais, com ar em temperatura de 40, 50, 60, 70 e 80 ºC e velocidade de 0,29$\pm$0,03 $m.s^{-1}$. Para cada teste de secagem foram utilizadas 400 g de folhas em camada de 0,15 m de espessura, sendo realizados três testes para cada temperatura. Aos dados experimentais foram ajustados diversos modelos de cinética de secagem, dentre eles, Henderson & Pabis, Henderson & Pabis modificado, Lewis,

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Midilli et al., Page, Thompson e Wang & Singh. Comparando-se os coeficientes de determinação ajustados (R^2), os erros médios relativo (P) e estimado (SE) e a distribuição de resíduos, concluiu-se que os modelos de Page e Midilli et al. foram os que representaram melhor a secagem de erva-cidreira-brasileira. Os coeficientes de difusividade efetiva calculados variaram entre $2,91x10^{-12}$ e $11,71x10^{-12}$ $m^2.s^{-1}$ para a faixa de temperatura estudada. A energia de ativação para difusão da água foi de 31,79 $kJ.mol^{-1}$.

Palavras-chave: energia de ativação, difusividade efetiva, plantas medicinais, plantas aromáticas, cinética.

INTRODUCTION

The Brazilian lemon-scented verbena has been widely used in popular medicine as it contains a biological activity and volatile constituent in its essential oil that has therapeutically properties (Gomes et al., 1993; Matos et al., 1996; Pino et al., 1997; Zoghbi et al., 1998; Vale et al., 1999; Pascual et al., 2001; Senatore and Rigano, 2001; Tavares et al., 2004).

The water activity of products *in nature* can be dramatically reduced by the drying process, which increases the conservation time and shelf life of the product in addition to facilitating its transport, handling and storage. It can also promote the stability of the aromatic components in ambient temperature for a long period while offering protection against enzymatic and oxidative degradation (Park et al., 2001). Moreover, with the reduction in the amount of water, the percentage of active principles in relation to the total mass is increased.

However, other parameters must be taken in consideration, including the temperature of the product during the drying process. The air temperature limits are determined in function of the sensitivity of chemical components and storage structures, once the product temperature increases during the drying process, coming close to that in which the process is carried out (Venskutonis, 1997; Martins, 2000).

The volatile aromatic components of medicinal and aromatic plants are very sensitive to drying. A number of studies have looked into methods of minimizing the drying effect on these volatile substances, and as a result have demonstrated that the drying process can reduce those volatile composites. This reduction depend on a number of factors, such as drying method, air temperature and characteristics of the products submitted (Deans & Svoboda, 1992; Charles et al., 1993; Hansen et al., 1993; Venskutonis, 1997; Rehder et al., 1998; Reynolds, 1998; Buggle et al., 1999; Balladin & Headley, 1999; Martins, 2000; Rocha et al., 2000; Radünz et al., 2002; Radünz et al., 2003).

When choosing the appropriate drying system, numerous factors need to be considered, including time, energy and product properties. The time used in the process is related to the energy and mass exchange between the product and the drying air, which can be analyzed by the effective diffusivity (Park et al., 2001).

The use of mathematical models to predict the drying process of various products, including medicinal plants has been the objective of many studies (Park et al., 1996; Simal, et al., 1996; Simal, et al., 2000; Park et al., 2001; Azzouz et al., 2002; Park et al., 2002; Panchariya et al., 2002; Babalis & Belessiotis, 2004; Souza et al., 2002; Doymaz, 2005a; Doymaz, 2005b; Mohapatra & Rao, 2005).

These models are useful tools to estimate the time for reduction of product moisture content under different drying conditions, and how to increase the drying process efficiency (Andrade et al., 2003).

Therefore, the aim of this study is to calculate the effective diffusivity and activation energy by adopting the appropriate mathematical model to the experimental data.

MATERIAL AND METHODS

Cultivation, harvest, selection and defoliation

The plants used in the current study were cultivated in an experimental area of 50 m^2 located at the Universidade Federal de Viçosa, Brazil. The seedlings propagated from the cuttings that were planted at 0.50 m spacing between lines and 0.25 m between plants.

The plants were harvested between 8 and 9 A.M, with moisture content of approximately 85% wb. The aerial parts of the plants were cut at 0.05 m above the ground and separated.

The method described in ASAE Standards (ASAE, 2000) was used to determine the moisture content.

Drying

The leaves were submitted to the drying process into a fixed-bed dryer, as described by Radünz (2004). The drying air velocity through the tray and product was measured by digital blade anemometer. The air temperatures used in the drying process were 40, 50, 60, 70 and 80ºC. For each test approximately 400 g of leaves were used, making a layer of 0,15 m thickness. Three tests were performed at each of the drying air temperatures.

The leaves were dried until they reached a moisture content around 10% wb, according to the product final mass.

During drying process, measurement of the leaves moisture content was made through periodic weighing of the trays at time intervals pre-established as contained in Table 1.

Table 1 - Time Intervals for the ccompaniment of the moisture content during the drying process

Treatment	Intervals of time
40 ºC	To each 10 min in first the 40 min, and later, to each 15 min
50 ºC	To each 10 min
60 ºC	To each 5
70 ºC	To each 5
80 ºC	To each 3 min

During the experiment, the temperature and relative humidity from the environmental air were monitored by a term-hygrograph.

Data Analysis

Determination of equilibrium moisture content

The equation proposed by Corrêa al. (2002) for medicinal plants was used to determine an average value of equilibrium moisture content for the experimental conditions (Equation 1), because does not exist a standard equation for the Brazilian lemon-scented verbena. This average value was taken as reference and later used in Equation 2.

$$X_e = \frac{1}{\left(a \cdot T^b + RH^c\right)} \qquad (1)$$

where:

Xe = Equilibrium moisture content, decimal, (d.b.);

T = Air temperature (ºC);

RH = Relative humidity of drying air, decimal;

a, b, c = Constants that depend of the product.

Determination of the effective diffusivity and activation energy in drying

The effective diffusivity was determined by the simple form of Fick's Law, Equation 2.

$$MR = \frac{X - X_{eq}}{X_i - X_{eq}} = \frac{8}{\pi^2} \cdot \sum_{i=0}^{\infty} \frac{1}{(2i+1)^2} \exp\left[-(2i+1)^2 \cdot \pi^2 \cdot D_{ef} \cdot \frac{t}{4L^2}\right] \qquad (2)$$

where:
D_{ef} = Effective diffusivity, $m^2.s^{-1}$;
MR = Moisture ratio of the product, non dimensional;
X = Moisture content of the product at time t, decimal (d.b.);
X_i = Initial moisture content of the product, decimal (d.b.);
X_{eq} = Equilibrium moisture content of the product, decimal (d.b.);
I = Number of terms of the series;
T = Time, s;
L = Characteristic length, thickness of the sample, m.

The activation energy that expresses the dependence of the diffusion as a function of the temperature was calculated by the Arrhenius equation (Equation 3).

$$D_{ef} = D_0 \exp\left(-\frac{E_{act}}{R \cdot T}\right) \qquad (3)$$

where:
D_{ef} = Effective diffusivity, $m^2.s$;
D_0 = Arrhenius factor, $m^2.s$;
E_{act} = Activation energy, $J.mol^{-1}$;
R = Universal gas constant, 8.3143 $J.mol^{-1}.K^{-1}$;
T = Absolute Temperature, K.

Adjustment of the models

The drying mathematical models were adjusted to the experimental data as shown in Table 2.

Table 2 - Evaluated mathematical models

Model	Name	Mathematical model	Reference
1	Midilli et al.	$RU = a \cdot \exp(-k \cdot t^n) + b \cdot t$	Midilli et al. (2002)
2	Page	$RU = \exp(-k \cdot t)^n$	Page (1949) in Bruce (1985)
3	Lewis	$RU = \exp(-k \cdot t)$	Lewis (1921)
4	Exponential of two terms	$RU = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k \cdot a \cdot t)$	Kassem (1998) in Togrul & Pehlivan (2003)
5	Two terms	$MR = a \cdot \exp(-k_0 \cdot t) + b \cdot \exp(-k_1 \cdot t)$	Henderson (1974)
6	Henderson & Pabis	$MR = a \cdot \exp(-k \cdot t)$	Henderson & Pabis (1961)
7	Henderson & Pabis modified	$MR = a \cdot \exp(-k \cdot t) + b \cdot \exp(-k_0 \cdot t) + c \cdot \exp($	Karathanos (1999)
8	Approach of the diffusion	$MR = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k \cdot b \cdot t)$	Sharaf-Elden et al. (1980)
9	Thompson	$t = a \cdot \ln(MR) + b \cdot [\ln(MR)]^2$	Thompson et al. (1968)
10	Wang e Singh	$MR = 1 + a \cdot t + b \cdot t^2$	Wang & Singh (1978)

where:
t = Drying time, s;
k and k^1 = Drying constant, s^{-1}
a,b,n = Model constants

The STATISTICA® 6.0 program was used to adjustment a non-linear analysis according to the Simplex and Quasi-Newton method. This gave a value for the parameters of the models estimated as a function of the temperature of the drying air.

The degree of adjustment of the model it was considered the adjusted coefficient of determination (R^2), the magnitude of the relative mean error (P), the estimated mean error (SE) and by verification of the residues analysis and graphs of correspondence between the estimated and observed values.

The relative mean error (P) and the estimated mean error (SE), for each model, were calculated by Equation 4 and 5.

$$P = \frac{100}{n} \sum \frac{|Y - Y_0|}{Y} \qquad (4)$$

and

$$SE = \sqrt{\frac{\sum (Y - Y_0)^2}{DF}} \qquad (5)$$

where:
n = Number of experimental observations;
Y = Experimental values observed;
Y_o = Value estimated though the model;
Df = Degrees of freedom (number of observations minus the number of the parameters of the model);

Coefficients of determination (R^2) higher than 98% indicated a good adjustment of the model for representation of the drying phenomenon (Madamba et al., 1996; Andrade et al., 2003). In accordance with Draper & Smith (1981) and Panchariya et al. (2002), the SE value is inversely proportional to the capacity of the model when describing the allegiance of the phenomenon. Values for relative mean errors (P) below 10% were considered acceptable (Park et al., 2002; Kaymak-Ertekin & Gedik, 2005; Mohapatra & Rao, 2005).

RESULTS AND DISCUSSION

The average velocity of the drying air was 0.29±0.03 $m.s^{-1}$. The calculated values of equilibrium moisture content were 8.71±0.65; 6.28±0.53; 5.07±0.46; 4.32±0.42 and 3.59±0.36% d.b., for the air temperatures of 40, 50, 60, 70 and 80 ºC, respectively. The calculated values of the effective diffusivity were 2.91×10^{-12}, 5.23×10^{-12}, 7.14×10^{-12}, 10.00×10^{-12} and 11.71×10^{-12} $m^2.s^{-1}$ for the drying air temperatures of 40, 50, 60, 70 and 80 ºC, respectively. This verifies that the effective diffusivity increased as the drying air temperature increased, thus showing a reduction in the internal resistance of drying as temperature rises. In Figure 1, the values of the effective diffusivity (D_{ef}) are represented in terms of the absolute temperature of the drying air.

Figure 1 proves that the effective diffusivity demonstrated linear behavior in terms of the air drying temperature, thus showing dependence on Arrhenius.

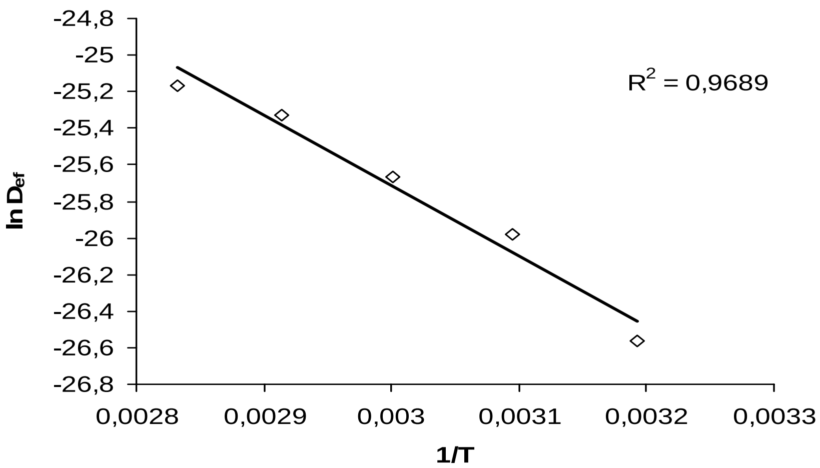

Figure 1 - Relation of the Arrhenius type between the effective diffusivity coefficient and the drying air temperature of Brazilian lemon-scented verbena leaves.

Table 3 shows the results of activation energy for drying of diverse products.

Table 4 presents the results of the adjusted determination coefficients (R^2), the relative mean errors (P) and the estimated mean errors (SE) of the adjusted mathematical models, presented in Table 2.

Table 3 - Activation energy (E_a) for the drying of diverse products

Product	E_a ($kJ.mol^{-1}$)	References
Aloe Vera	24.40	Simal et al. (2000)
carrot	28.36	Doymaz (2004)
Pea	28.40	Simal et al. (1996)
Soy Beans	28.80	Kitic & Viollaz (1984)
Chive	29.05	Park et al. (1996)
Brazilian lemon-scented verbena	**31.79**	**Present work**
String bean	35.43	Doymaz (2005b)
Wheat	37.01	Mohapatra e Rao (2005)
String bean	39.47	Senadeera et al., (2003)
Red pepper	41.95	Gupta et al. (2002)
Lemongrass	48.31	Martins (2000)
Gumbo	51.26	Doymaz (2005a)
Mint	82.93	Park et al. (2002)

Table 4 – Coefficient of determination (R^2), relative mean errors (P) and estimated mean errors (SE), for the analyzed models during the Brazilian lemon-scented verbena leaves drying under different drying air temperatures.

Mathematical models	T (°C)	P (%)	SE	R^2 (%)
Midilli et al.	40	12.67	0.0081	99.99
	50	7.34	0.0173	99.93
	60	13.51	0.0193	99.92
	70	9.53	0.0102	99.97
	80	5.27	0.0116	99.97
Page	40	12.04	0.0163	99.98
	50	17.47	0.0312	99.92
	60	6.90	0.0356	99.91
	70	19.35	0.0298	99.91
	80	4.09	0.0208	99.97
Lewis	40	11.46	0.0431	99.88
	50	48.29	0.1045	99.08
	60	63.32	0.1148	99.08
	70	47.44	0.0684	99.54
	80	70.51	0.1540	98.15
Exponential of two terms	40	13.44	0.0174	99.98
	50	10.51	0.0330	99.91
	60	13.23	0.0386	99.90
	70	21.14	0.0299	99.91
	80	13.42	0.0345	99.91
Two terms	40	7.73	0.0215	99.91
	50	41.63	0.0559	99.21
	60	54.78	0.0597	99.25
	70	44.41	0.0370	99.59
	80	60.36	0.0786	98.55
Henderson & Pabis	40	7.73	0.0372	99.91
	50	41.63	0.0968	99.21
	60	54.78	0.1035	99.25
	70	44.41	0.0642	99.59
	80	60.36	0.1362	98.55
Henderson & Pabis Modificada	40	7.73	0.0166	99.91
	50	57.93	0.0315	99.58
	60	7.33	0.0145	99.93
	70	12.38	0.0083	99.97
	80	43.47	0.0375	99.45
Approach of the diffusion	40	7.62	0.0101	99.99
	50	14.46	0.0212	99.92
	60	50.67	0.0467	99.70
	70	13.04	0.0135	99.96
	80	49.14	0.0661	99.32
Thompson	40	11.48	0.0431	99.88
	50	48.28	0.1045	99.08
	60	63.33	0.1148	99.08
	70	47.45	0.0684	99.54
	80	70.52	0.1540	98.15
Wang & Singh	40	409.00	0.3403	92.24
	50	193.56	0.1697	97.58
	60	127.49	0.1345	98.73
	70	59.23	0.0947	99.11
	80	47.62	0.0631	99.69

According to the results presented in Table 4 all the mathematical models, except Wang & Singh model (for the temperatures of 40 and 50 ºC), described the drying of Brazilian lemon-scented verbena leaves with a high degree of precision; with coefficients of determination above 98% and estimated mean errors (SE) below 0.20, for all the studied temperatures. However, the Midilli et al. and Page models had lower values for relative mean errors (P) and better residue distributions.

When drying *Mentha crispa* L., at air temperatures of 30, 40 and 50 ºC Park et al. (2002) obtained better adjustment to the data with the Page model. However in studies with *Camellia sinensis (L.)* Kuntze (Temple & Van Boxtel, 1999) and *Cymbopogon citrates* (D.C) Stapf (Martins, 2000), the best adjustments were obtained with the exponential model of Lewis.

Table 5 shows the values of the parameters for the Midilli et. al and Page models.

Table 5 – Page and Midilli et al. models ajusted for Brazilian lemon-scented verbena leaves drying under different drying air temperatures.

Model	T (ºC)	Coefficients			
		K	N	a	b
Midilli et al.	40	1.6404	1.0860	1.0001	0.0013
	50	2.9021	1.2660	0.9953	0.0027
	60	4.2206	1.2207	0.9916	-0.0041
	70	4.9466	1.0873	0.9990	-0.0325
	80	10.7156	1.3491	0.9953	-0.0014
Page	40	1.6264	1.0771	-	-
	50	2.8580	1.2470	-	-
	60	4.2766	1.2178	-	-
	70	5.5224	1.1395	-	-
	80	10.6601	1.3420	-	-

CONCLUSIONS

Under the conditions of the current experiment it can be concluded that:

a) The models of Page and Midilli et al. were the best representations of Brazilian lemon-scented verbena drying;
b) The effective diffusivity coefficients varied between $2.91x10^{-12}$ and $11.71x10^{-12}$ $m^2.s^{-1}$, in the band of studied temperature;
c) The activation energy for the water diffusion was 31.79 $kJ.mol^{-1}$.

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