

Original Research Article

Drying Kinetics of Chives (*Allium fistulosum* L.)

Abstract: Chives are herbs used as spice in Brazil, but their postharvest durability is short, mainly due to high metabolic rate and high moisture content. In this context, this study aimed to model the drying curves of chives at different temperatures and determine the effective diffusion coefficient and activation energy for the process. Chives were manually harvested, with moisture content of 12.0 (decimal, dry basis) and subjected to drying until reaching equilibrium moisture content, i.e., minimum amount of free water. The fitting capacity of eleven mathematical models used to evaluate drying processes in agricultural products was assessed. Treatments consisted of two physical patterns of length, whole and chopped leaves (pieces of approximately 2 cm and whole leaves of approximately 20 cm long, respectively), and four drying air temperatures (40; 50; 60 and 70 °C). The fit of the studied models was assessed by nonlinear regression through the Gauss-Newton method. Among the mathematical models tested, Midilli was the one that fitted best to the experimental data. Increasing drying temperature leads to higher rate of water removal from the product.

Key words: *Allium fistulosum*; Drying modeling; Chives; Diffusion coefficient; Drying; Mathematical model.

1. INTRODUCTION

Chives (*Allium fistulosum* L.) are a seasoning herb belonging to the Alliaceae family. Native to Siberia and known as 'Cebolinha common' in Brazil, this species plays an important socioeconomic role, generating job and profit in family farming cultivation. These plants are similar to oval bulbs and its leaves are numerous, with 15 to 20 cm height, dark green color, hollow and slender. However,

27 their postharvest durability is short, mainly due to the high metabolic rate and high moisture content
28 [1].

29 Conservation of leafy vegetables by drying is the most used method to ensure quality and
30 stability, considering that the reduction in the amount of water in the material reduces biological
31 activity and chemical and physical changes which occur during storage [2].

32 Water-soluble vitamins, amino acids, salts, some pectins and sugars present in the plants are
33 directly correlated with intrinsic characteristics, and the conservation of these soluble compounds
34 depends on the quality of product drying and storage [3]. For Vilela & Artur (2008) [4], the information
35 contained in the drying curves is fundamentally important for the development of processes and
36 dimensioning of equipment.

37 According to Resende et al. (2008) [5], drying curves vary with species, variety, environmental
38 conditions, methods of postharvest preparation, among other factors. In this method, whose principle
39 is based on the drying of tissues, mathematical models are used to represent water loss from the
40 product during the drying period [6]. In the literature, several models have been proposed to analyze
41 the drying of hygroscopic products, namely: theoretical, semi-empirical and empirical [7].

42 However, major emphasis has been given to the development of semi-theoretical models,
43 which contribute to bringing harmony between theory and ease of use. These models are based, in
44 general, on the Newton's Law for cooling applied to mass transfer. When this law is applied, one
45 assumes that the conditions are isothermal and that the resistance to water transfer is limited to only
46 the product surface [8]. Among the semi-theoretical models, Two Terms, Henderson and Pabis, Page,
47 Modified Page and Midilli have been widely used [9].

48 Martinazzo et al. (2010) [10] and Demir et al. (2004) [11], evaluating different mathematical
49 models in the drying of lemongrass (*Cymbopogon citratus*) and laurel (*Laurus nobilis* L.), concluded
50 that Page model was the one which best described the process, whereas Doymaz et al. (2006) [12],
51 evaluating the drying of leaves of dill (*Anethum graveolens* L.) and parsley (*Petroselinum crispum* L.),
52 defined the Midilli model as the most adequate at temperatures of 40 to 70 °C.

53 Nevertheless, there is limited information in the literature about the drying kinetics and
54 mathematical modeling of chives grown in the Cerrado region of Goiás, Brazil. Given the above, this
55 study aimed to fit thin-layer drying mathematical models to the experimental data obtained in the
56 drying of chives leaves at different drying temperatures.

2. MATERIALS AND METHODS

Chives were harvested in November 2017, in a private property located in the municipality of Rio Verde, GO, Brazil (17°48'28"S and 50°53'57"W), IFRV 10099. Harvest was carried out by hand when plants were at the vegetative stage, defined based on the green color of the stalks, which were carefully transported to the Laboratory of Postharvest of Plant Products of the Federal Institute of Education, Science and Technology of Goiás –Campus Rio Verde, Goiás, Brazil (IF Goiano – Campus Rio Verde).

Prior to drying, leaves were selected by removing diseased and injured parts. For drying, some leaves were minimally processed by hand, cut into pieces of approximately 2 cm, and whole leaves (approximately 20 cm long) were also placed on trays in 5-cm-thick layer.

Drying was carried out in a Marconi® MA–035 oven, with forced air circulation, under the following air conditions: drying temperatures of 40, 50, 60 and 70 °C and relative humidity values of 15.26, 9.42, 5.88 and 3.74%, respectively. Drying continued until the material reached constant mass (equilibrium moisture content), determined in the oven at 103 °C [13]. Reduction of moisture content along drying was carried out by the gravimetric method (mass loss), based on the initial moisture content of the product, until reaching constant mass. Mass reduction along drying was monitored using a scale with resolution of 0.01 g.

Drying air and room temperatures were monitored by a digital thermo-hygrometer installed outside the oven, and these data were used to estimate the relative humidity inside the oven by the basic principles of psychrometry, with the aid of the computer program GRASPI.

During the drying process, the trays containing the samples, with four replicates per temperature, were monitored by the gravimetric method (mass loss). To determine the equilibrium moisture content, the samples were weighed until reaching a constant mass in three consecutive measurements.

Moisture content ratios of chives during drying were determined by the following expression:

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

Where: RX: moisture content ratio of the product, dimensionless; X: moisture content of the product (d.b.); Xi: initial moisture content of the product (d.b.); and Xe: equilibrium moisture content of the product (d.b.).

86

87 The mathematical models frequently used to represent the drying of plant products (Table 1)
 88 were fitted to the experimental data of drying of chives.

89

90 **Table 1.** Mathematical models used to describe the drying of plant products

Model designation	Model
$RX = a \exp(-kt) + (1 - a) \exp(-k b t)$	Approximation of Diffusion (2)
$RX = a \exp(-k t) + b \exp(-g t)$	Two Terms (3)
$RX = a \exp(-k t) + (1 - a) \exp(-k a t)$	Two-term Exponential (4)
$RX = a \exp(-k t)$	Henderson and Pabis (5)
$RX = a \exp(-k t) + b$	Logarithmic (6)
$RX = a \exp(-k t^n) + b t$	Midilli (7)
$RX = \exp(-k t)$	Newton (8)
$RX = \exp(-k \cdot t^n)$	Page (9)
$RX = \exp((-a - (a^2 + 4 b t)^{0.5}) / 2 b)$	Thompson (10)
$RX = a \exp(-k t) + (1 - a) \exp(-g t)$	Verma (11)
$RX = 1 + a t + b t^2$	Wang and Singh (12)

91 Where: RX: moisture content ratio of the product; t: drying time, h; k, k_0 , k_1 : drying constants, h^{-1} ; and
 92 a, b, c, n: parameters of the models.

93

94 The mathematical models were fitted by nonlinear regression analysis through the Gauss-
 95 Newton method. The degree of fit, **was evaluated considering** considered the magnitude of the
 96 coefficient of determination (R^2), Chi-square test (χ^2), mean relative error (P), mean estimated error
 97 (SE), Akaike Information Criterion (AIC), Bayesian Information Criterion (BIC), according to the
 98 following equations:

$$R^2 = \frac{SSR}{SST} \quad (13)$$

$$\chi^2 = \frac{\sum_{i=1}^n (Y - \hat{Y})^2}{DF} \quad (14)$$

$$P = \frac{100}{n} \sum \frac{|Y - \hat{Y}|}{Y} \quad (15)$$

$$SE = \sqrt{\frac{\sum (Y - \hat{Y})^2}{DF}} \quad (16)$$

$$AIC = -2 \log \text{like} + 2p \quad (17)$$

$$BIC = -2 \log \text{like} + p \ln(n) \quad (18)$$

99

Where: Y: value observed experimentally; $\hat{Y}$: value estimated by the model; n: number of observations; DF: degrees of freedom of the model; p: number of parameters; loglike: logarithm of the likelihood function considering the estimates of the parameters; n: number of observations used to fit the curve.

The liquid diffusion model for the geometric shape of a **slab**, with approximation of eight terms (Equation 13), was fitted to the experimental data of drying of chives, considering the surface area and volume according to the following expression:

$$RX = \frac{U^* - U_e^*}{U^* - U_e^*} = \frac{8}{\pi^2} \sum_{n_t=0}^{\infty} \frac{1}{(2n_t + 1)^2} \exp \left[-\frac{(2n_t + 1)^2 \cdot \pi^2 \cdot D \cdot t}{4} \cdot \left(\frac{S}{V} \right)^2 \right] \quad (19)$$

Where: RX : moisture ratio of the product, dimensionless; nt: number of terms; S: surface area of the product, **m²**; and V: volume of the product, m³.

The surface area of chives was calculated using photographs of the samples, with the aid of the **program** IMAGEJ, whereas their volume was determined by measuring the thickness with a digital caliper, according to the following Equation:

$$V = S \cdot T \quad (20)$$

Where, V: volume (m³); S: surface area (m²); T: thickness (mm).

The relationship between the effective diffusion coefficient and the increase in drying air temperature was described by the Arrhenius equation.

$$D = D_0 \cdot \exp \left(\frac{-E_a}{R \cdot T_{ab}} \right) \quad (21)$$

Where: **D₀**: pre-exponential factor; E_a: activation energy, kJmol⁻¹; R: universal gas constant, 8.134 kJkmol⁻¹K⁻¹; and T_{ab}: absolute temperature, K.

3. RESULTS AND DISCUSSION

The moisture content in minimally processed (Fig. 1-A) and whole (Fig. 1-B) leaves of chives was monitored during the drying process. It can be observed that minimally processed chives reached hygroscopic equilibrium with 4.5, 7.0, 16.0 and 20 hours, whereas whole leaves reached hygroscopic

equilibrium with 10.0, 12.0, 13.0 and 50.5 hours, for the temperatures of 70, 60, 50 and 40 °C, respectively.

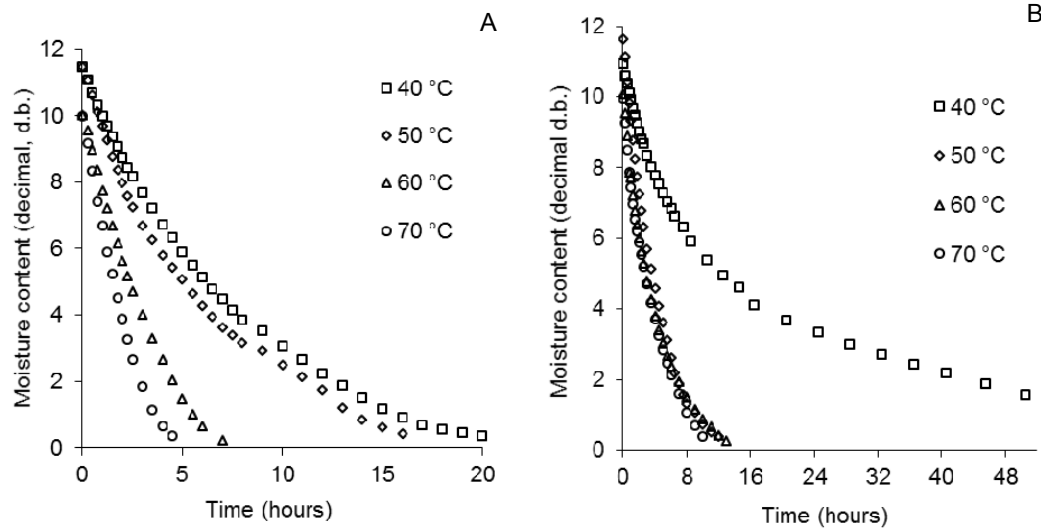


Fig. 1. Moisture contents in minimally processed (A) and whole (B) leaves of chives along drying at temperatures of 40, 50, 60 and 70 °C

It becomes evident that the leaves lose water more easily with increasing drying temperatures and that cutting them increases the surface of contact with the hot air flow produced by the oven, which leads to faster drying of minimally processed leaves, in comparison to whole leaves. Similar results have been found in the drying of leaves of *Cymbopogon citratus* [14] and *Bauhinia forficata* [15].

Tables 2 and 3 present the values of Chi-square and mean estimated error for the eleven models studied in the drying of minimally processed and whole leaves of chives, respectively. Mean estimated errors close to zero were found and, according to Draper & Smith (1998) [16], it indicates good representativeness because the capacity of a model to faithfully describe a certain physical process is inversely proportional to the standard deviation of the estimate.

Table 2. Values of Chi-square test (χ^2 , decimal) and mean estimated error (SE, decimal) calculated for the eleven models used to represent the drying kinetics of minimally processed chives

Model	40 °C	50 °C	60 °C	70 °C
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	SE	χ^2	SE	χ^2	SE	χ^2	SE	χ^2
Approximation of Diffusion	0.20	0.26	0.02	0.07	0.01	0.10	0.02	0.14
Two Terms	0.01	0.06	0.02	0.17	0.01	0.09	0.05	0.51
Two-Term Exponential	0.02	0.26	0.02	0.15	0.05	0.44	0.05	0.50
Henderson and Pabis	0.02	0.25	0.02	0.15	0.04	0.37	0.05	0.43
Logarithmic	0.07	0.04	0.02	0.12	0.01	0.09	0.02	0.13
Midilli	0.01	0.05	0.01	0.06	0.01	0.06	0.01	0.07
Newton	0.02	0.25	0.02	0.14	0.05	0.42	0.05	0.47
Page	0.02	0.19	0.02	0.16	0.02	0.17	0.02	0.15
Thompson	0.02	0.26	0.02	0.15	0.05	0.44	0.05	0.50
Verma	0.01	0.18	0.02	0.16	0.01	0.10	0.02	0.14
Wang and Singh	0.03	0.21	0.04	0.25	0.01	0.02	0.01	0.05

The values of Chi-square and mean estimated errors close to zero, presented in Tables 2 and 3, were significant for all equations evaluated.

According to Molina Filho et al. (2006) [17], the higher the values of Chi-square, the greater the discrepancy between experimental and estimated values.

Based on the values of mean relative error and coefficient of determination (Tables 4 and 5), it is possible to identify the best models to represent the drying kinetics of leaves of chives.

Table 3. Values for Chi-square test (χ^2 , decimal) and mean estimated error (SE, decimal) calculated for the eleven models used to represent the drying kinetics of whole chives

Model	40 °C		50 °C		60 °C		70 °C	
	SE	χ^2	SE	χ^2	SE	χ^2	SE	χ^2
Approximation of Diffusion	0.01	0.05	0.01	0.05	0.01	0.04	0.02	0.05
Two Terms	0.03	0.11	0.01	0.11	0.01	0.12	0.02	0.15
Two-Term Exponential	0.05	0.16	0.01	0.12	0.01	0.13	0.02	0.14
Henderson and Pabis	0.03	0.10	0.02	0.20	0.01	0.11	0.02	0.13
Logarithmic	0.02	0.15	0.01	0.06	0.01	0.08	0.01	0.06
Midilli	0.01	0.06	0.01	0.06	0.01	0.01	0.01	0.03
Newton	0.04	0.16	0.02	0.22	0.01	0.10	0.02	0.12
Page	0.01	0.07	0.01	0.12	0.01	0.13	0.02	0.14
Thompson	0.01	0.09	0.02	0.23	0.01	0.13	0.02	0.13
Verma	0.05	0.17	0.02	0.24	0.01	0.12	0.02	0.13
Wang and Singh	0.10	0.47	0.03	0.23	0.05	0.43	0.04	0.20

The models Approximation of Diffusion (50 and 60 °C), Two Terms (40 and 60 °C), Logarithmic (40 and 60 °C), Midilli (40; 50; 60 and 70 °C) and Wang and Singh (60 and 70 °C) represent the drying of minimally processed chives at the studied temperatures with mean relative error varying from 2.14 to 8.54%, and coefficients of determination from 98.97 to 99.90 %.

For whole chives, the models Approximation of Diffusion (40; 50; 60 and 70 °C), Two Terms (50 °C), Logarithmic (50, 60 and 70 °C), Midilli (40; 50; 60 and 70 °C), Newton (60 °C), Page (40 °C) and Thompson (40 and 60 °C) can be used to represent the drying because they had low values of mean relative error and high coefficients of determination.

Table 4. Mean relative error (P) and coefficient of determination (R^2 , %) obtained in the drying kinetics of minimally processed chives for the eleven models used

Model	40 °C		50 °C		60 °C		70 °C	
	P	R^2	P	R^2	P	R^2	P	R^2
Approximation of Diffusion	24.06	99.81	5.91	99.88	8.54	99.94	11.41	99.88
Two Terms	4.99	99.98	14.62	99.84	5.80	99.97	37.24	99.02
Two-Term Exponential	24.06	99.81	14.16	99.81	39.70	98.87	43.64	98.66
Henderson and Pabis	23.11	99.82	14.06	99.82	33.16	99.24	37.24	99.02
Logarithmic	3.87	99.98	10.83	99.83	7.74	99.96	10.61	99.90
Midilli	4.79	99.99	5.52	99.92	4.76	99.99	4.83	99.98
Newton	24.06	99.81	13.8	99.81	39.70	98.87	43.64	98.66
Page	18.11	99.87	14.74	99.82	14.98	99.86	13.11	99.86
Thompson	24.06	99.81	14.10	99.81	39.71	98.87	46.64	98.66
Verma	16.13	99.90	14.57	99.83	8.54	99.94	11.41	99.88
Wang and Singh	20.09	99.59	23.49	98.97	2.14	99.98	4.48	99.97

According to Madamba et al. (1996) [18], values of R^2 higher than 95% and mean relative error lower than 10% [19] indicate that the models are adequate to represent the phenomenon. However, the coefficient of determination cannot be used alone to select a mathematical model; more statistical parameters need to be used for confirmation [20, 21].

The use of statistical parameters is important for regression analysis because they penalize models in various points, selecting those which are more parsimonious [22].

Table 5. Mean relative error (P) and coefficient of determination (R^2 , %) obtained in the drying kinetics of whole chives for the eleven models used

Model	40 °C		50 °C		60 °C		70 °C	
	P	R^2	P	R^2	P	R^2	P	(%)
Approximation of Diffusion	5.09	99.97	4.51	99.97	3.64	99.90	4.06	99.85
Two Terms	10.15	99.42	9.70	99.94	10.01	99.91	12.19	99.81
Two-Term Exponential	15.76	98.91	10.67	99.93	11.95	99.89	12.50	99.83
Henderson and Pabis	10.15	99.42	18.22	99.84	10.01	99.91	12.19	99.81
Logarithmic	13.74	99.71	5.59	99.97	7.44	99.91	4.94	99.89
Midilli	5.31	99.95	5.06	99.97	1.10	99.96	2.05	99.98
Newton	15.76	98.91	21.08	99.78	9.48	99.89	11.59	99.78
Page	6.14	99.93	11.23	99.92	11.85	99.92	12.74	99.79
Thompson	8.38	99.93	21.09	99.78	2.07	99.9	11.60	99.78
Verma	15.76	98.91	21.08	99.78	10.87	99.93	11.59	99.78
Wang and Singh	45.93	95.01	21.07	99.66	39.96	98.55	18.07	99.03

The Akaike information criterion (AIC) and Bayesian information criterion (BIC) bring the possibility of confirming the superiority among the selected models, because the lower the values of AIC and BIC, the better the fit of the model (Table 6).

By analyzing the values of AIC and BIC for the best models selected, is possible to note the superiority of the Midilli model in comparison to the others.

As previously mentioned, this model had the lowest values of both AIC and BIC for all temperatures under both drying conditions, except for minimally processed leaves at temperature of 70 °C.

Table 6. Akaike information criterion (AIC) and Bayesian information criterion (BIC) of the models with best fits for minimally processed and whole chives

Minimally processed			
Temperature	Models	AIC	BIC
40 °C	Two Terms	-244.25	-236.62
	Logarithmic	-238.56	-232.46
	Midilli	-251.09	-243.46
50 °C	Approximation of Diffusion	-161.23	-155.62
	Midilli	-171.34	-164.33
60 °C	Logarithmic	-117.82	-114.04

	Midilli	-135.74	-131.02
70 °C	Midilli	-97.006	-93.465
	Wang and Singh	-95.901	-93.777
Whole			
40 °C	Approximation of Diffusion	-203.23	-195.75
	Midilli	-226.14	-220.15
	Page	-198.26	-193.77
	Thompson	-197.1	-192.62
50 °C	Logarithmic	-166.81	-160.52
	Midilli	-169.51	-164.48
60 °C	Approximation of Diffusion	-137.12	-132.24
	Logarithmic	-139.79	-134.92
	Midilli	-159.66	-153.57
	Newton	-137.95	-135.51
70 °C	Approximation of Diffusion	-113.91	-109.54
	Logarithmic	-120.3	-115.93
	Midilli	-160.6	-155.15

Such superiority also stands out when the coefficients and confidence levels designated by the t-test are analyzed (Table 7).

Except for the coefficient “k”, which showed increasing values following the drying temperatures, the others did not exhibit a clear trend as a function of the studied conditions.

Table 7. Parameters of the Midilli model fitted for the different conditions of drying of minimally processed and whole chives

Minimally processed				
	40 °C	50 °C	60 °C	70 °C
a	1.0012**	1.0229**	1.0007**	0.9921**
k	0.1366 ^{ns}	0.2042**	0.2364**	0.3777 ^{ns}
n	0.9591**	0.8488**	1.1471**	1.2361**
b	-0.0042**	-0.0054**	-0.0144**	-0.0165**
Whole				
a	1.0214**	1.0023**	1.0097**	1.0029**
k	-0.1471**	0.2258 ^{ns}	0.2818**	0.2797**
n	0.7512**	1.0345**	0.8896**	0.8372**
b	-0.0003 ^{ns}	-0.0038**	-0.0036**	0.0124**

**Significant at 0.01 probability level by t-test. ^{ns} not significant by t-test; k : drying constant, h⁻¹; and

a, b, n: parameters of the models.

Goneli et al. (2014) [23] studied the effective diffusivity during the drying of Brazilian peppertree leaves and found the same increasing trend for the coefficient “k” as the temperature increased. Such increment in the coefficient “k” can be explained by the fact that it is directly related to the effective diffusivity in the drying process with decreasing rate period and also to the liquid diffusion which controls the process [18, 24].

Based on the results obtained at the studied temperatures and drying conditions, the Midilli model is the one that best represents the drying process as illustrated in Fig. 2. The drying process was more influenced by temperature in minimally processed chives, and this phenomenon may be related to the increase in the area of contact with the drying air. In addition to the increase in contact area, we should also consider the rupture of both physical structure and cells of the leaves, directly influencing the desorption process of water removal. The influence of leaf fractionation prior to drying was also observed by Martinazzo et al. (2007) [7], in the drying of *Cymbopogon citratus* (DC.) leaves.

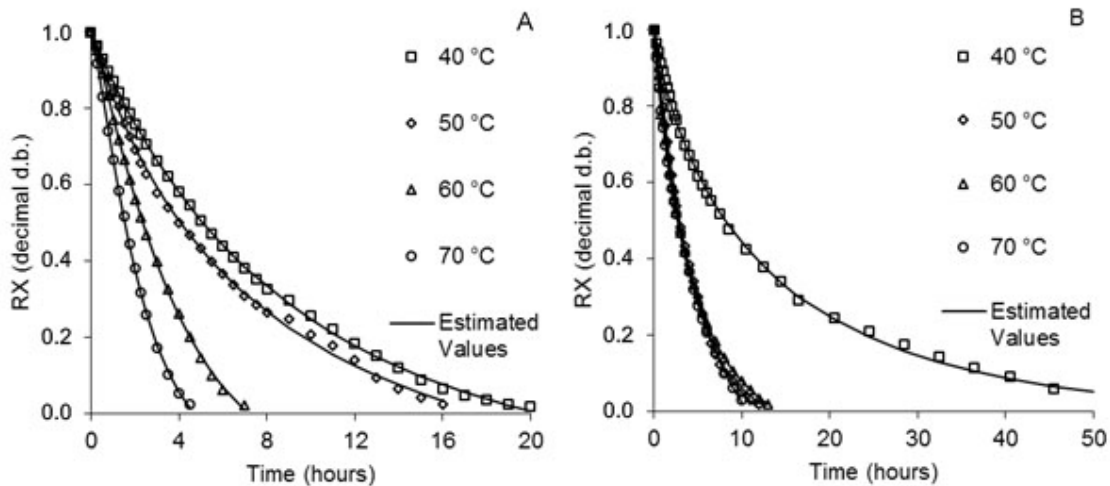


Fig. 2. Values of moisture content ratio (RX) of minimally processed (A) and whole (B) chives estimated by the Midilli model for drying under various temperature conditions

Likewise, Gomes et al. (2017) [14] and Martinazzo et al. (2007) [7], drying lemongrass leaves, identified Midilli as the best model to represent the drying kinetics. Such fit of the Midilli model to the experimental data of drying of leaves is possibly related to the fast loss of moisture in the constant rate period of the drying, in this type of product, generating a curve that is sharper and better characterized, mathematically, by this model [6].

According to Fig. 3, the effective diffusion coefficients increased with increasing drying temperatures, as also observed by Martinazzo et al. (2007) [7] in *Cymbopogon citratus* and by Prates et al. (2012) [25], who studied the drying of *Solanum lycocarpum* leaves.

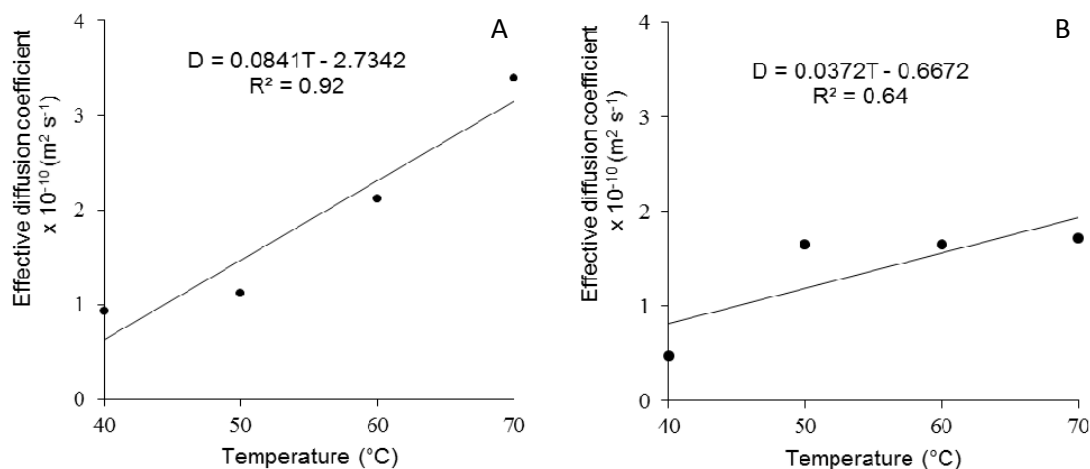


Fig. 3. Effective diffusion coefficient obtained for the drying of minimally processed (A) and whole (B) chives at temperatures of 40, 50, 60 and 70 °C

The effective diffusion coefficient increased with the increment in temperature, with values of 0.93×10^{-11} , 1.12×10^{-11} , 2.12×10^{-11} and $3.40 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ for minimally processed leaves and of 0.48×10^{-11} , 1.66×10^{-11} , 1.65×10^{-11} and $1.72 \times 10^{-11} \text{ m}^2 \text{ s}^{-1}$ for whole leaves of chives, respectively at the temperatures of 40, 50, 60 and 70 °C. Silva et al. (2015) [26] found values ranging from 1.12×10^{-12} to $4.02 \times 10^{-12} \text{ m}^2 \text{ s}^{-1}$ for the temperature range from 35.3 to 65 °C in the drying of *Genipa americana* leaves, and these values are close to those found in the drying of chives. The effective diffusivity depends on the characteristics of the drying air and other physical-chemical properties of the material which are related to the species [7]. According to Madamba et al. (1996) [18], the magnitude of the effective diffusivity for the drying of agricultural products is on the order of 10^{-9} to 10^{-11} .

The values of activation energy for liquid diffusion of minimally processed and whole chives were 38.40 and 40.32 kJ mol^{-1} , respectively, for the temperature range from 40 to 70 °C. These values are lower than those found by Martinazzo et al. (2010) [10], 63.47 kJ mol^{-1} for lemongrass leaves, and by Doymaz (2006) [12], 62.96 kJ mol^{-1} for *Mentha spicata* L leaves. During the drying process, the lower the activation energy, the higher the water diffusivity through the product. Thus, the

fact that minimally processed chives have lower activation energy than whole chives and other plant products may be related to the processing, which increases the surface of contact between the surrounding air and the product. According to Zogzas et al. (1996) [27], the activation energy for agricultural products varies from 12.7 kJ mol⁻¹ to 110 kJ mol⁻¹, corroborating the values found in the present study.

Kashaninejad et al. (2007) [28] highlight that the activation energy is considered as a barrier to be overcome so that the diffusion process in the product can occur and, therefore, in drying processes, the lower the activation energy, the higher the water diffusivity in the product [29].

4. CONCLUSION

Drying time decreased with increasing temperature, regardless of the processing of chives. Among the studied models, Midilli is the one that best represents the drying kinetics of minimally processed and whole chives. The effective diffusion coefficient increases as temperature increases and the activation energy for liquid diffusion in the drying is 34.80 and 40.32 kJ mol⁻¹ for minimally processed and whole chives, respectively.

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