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Drying kinetics of pumpkin seeds

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Abstract

The study of drying kinetics is of fundamental importance for mathematical modeling, which has been widely used in the design and analysis of heat and mass transfer processes during drying. The objective of the experiment was to fit mathematical models to the drying process of pumpkin (*Cucurbita moschata* Duchesne) seeds of the genotype 'Accession 53' and the cultivar 'Goianinha', under different temperature conditions, determining the mathematical model that can best predict this event, the liquid diffusion and the respective activation energies. The experiment was conducted at the Plant Science Laboratory of the Federal Institute of Education, Science and Technology of Goiás - Campus of Iporá, GO, Brazil. Eleven mathematical models were fitted by means of nonlinear regression analysis by the Gauss-Newton method, and the degree of fit was assessed based on the magnitude of the coefficient of determination (R^2), chi-square test (χ^2), mean relative error (P) and standard deviation of the estimate (SE). It was found that during the drying of pumpkin seeds the effective diffusion coefficient increases with the increase in temperature for both varieties. The Page model was the one that best described the drying phenomenon of pumpkin seeds genotype 'Accession 53' and cv. 'Goianinha', with activation energy of 39.340 kJ mol^{-1} for the genotype 'Accession 53' and 43.239 kJ mol^{-1} for the cv. 'Goianinha'.

Keywords: activation energy, Cucurbitaceae, modeling, storage

Introduction

Pumpkin (*Cucurbita moschata* Duchesne) is an annual plant of prostrate and indeterminate growth habit belonging to the Cucurbitaceae family. It is a food easily seen on the Brazilian table, native to Central America, and in Brazil is consumed mainly in the northeast region, where it is popularly known as 'jerimum', besides participating in a variety of traditional dishes of the region (Mendes et al., 2017).

The drying process of agricultural products is fundamental and aims to reduce the amount of water in them. According to Martinazzo et al. (2010), drying is the most widely used commercial process for preserving the quality of agricultural products. This process occurs through the use of hot air for transferring heat to the product and the consequent vaporization of the water contained in it, which leads to dehydration (Celestino, 2010).

In the context of drying processes, two concepts stand out due to their importance in the drying of grains and seeds, the study of mathematical models and liquid diffusion. Mathematical models are used to simulate the drying process of seeds and their principle is based on the drying of thin layers of the product (Reis et al., 2011), so seed water loss during drying can be well represented by these mathematical models (Mendonça et al., 2015; Costa et al., 2015; Araujo et al., 2017; Goneli et al., 2017).

Liquid diffusion, often called effective diffusion because it is very complex due to the variety of chemical compositions and physical properties of products, is understood as the way in which water is extracted from the product. Given the complexity of the products, several methods of prediction, type of material, drying process, moisture content and methodology used, large variations are found in the values of the diffusivity coefficient (Resende et al., 2005; Faria et al., 2012; Bezerra

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et al., 2015; Goneli et al., 2017).

Based on the above, the objective was to fit mathematical models to the drying process of pumpkin seeds, genotype 'Accession 53' and cultivar 'Goianinha', under various temperature conditions, determining which mathematical model can best predict this event, the liquid diffusion and the respective activation energies.

Material and Methods

The experiment was carried out at the Plant Science Laboratory of the Federal Institute of Education, Science and Technology Goiano (IF Goiano) - Campus of Iporá, GO, Brazil. Pumpkin seeds of the genotype 'Accession 53' of the Active Germplasm Bank of IF Goiano, Campus of Iporá ('AC 53', Genetic material with potential to generate new non-commercial cultivar) and the cultivar 'Goianinha' (commercial - reference), produced and harvested manually at the Fazenda Escola of IF Goiano, Campus of Iporá – Campus Iporá. The seeds had initial moisture contents of 0.768±0.041 and 0.780±0.041 dry basis (dry basis, d.b.), respectively. The seeds were subjected to drying in an oven with forced air circulation under four temperature conditions: 40, 50, 60 and 70 °C, which promoted relative humidity within the greenhouse of 34.0, 21.1, 13.1 and 8.1%, respectively, for the genotype and the cultivar. Drying continued until the grains reached the moisture content of 0.144±0.013 and 0.142±0.014 (d.b.), respectively, determined in an oven at 105 ± 3 °C for 24 h (BRASIL, 2009).

The reduction of moisture content during drying was monitored by the gravimetric method (mass loss), from the initial moisture content of the product until it reached the desired moisture content. Mass reduction during drying was monitored using a scale with resolution of 0.01 g. Equilibrium moisture content was determined by placing the samples in forced circulation oven for all temperatures, weighing them every day at the same time until constant weights (equilibrium) were obtained. After obtaining the equilibrium in the weighing, moisture content was determined as described above.

Drying air temperature and ambient temperature were monitored by means of a thermometer installed inside and outside the dryer, and the relative humidity inside the oven was obtained through the basic principles of psychrometry, using the GRAPSI computer program.

The following expression was used to determine the moisture content ratios of pumpkin seeds during drying:

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

Where:

RX: moisture content ratio of the product, dimensionless;

X: moisture content of the product (d.b.);

X_i: initial moisture content of the product (d.b.); and

X_e: equilibrium moisture content of the product (d.b.).

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

Table 1. Mathematical models used to predict drying of plant products.

Model designation	Model	
$RX = 1 + a\,t + b\,t^2$	Wang and Sing	(2)
$RX = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k_1 \cdot t)$	Verma	(3)
$RX = \exp\left(\left(-a - \left(a^2 + 4\,b\,t\right)^{0.5}\right)/2\,b\right)$	Thompson	(4)
$RX = \exp\left(-k \cdot t^n\right)$	Page	(5)
$RX = \exp(-k \cdot t)$	Newton	(6)
$RX = a \cdot \exp\left(-k \cdot t^n\right) + b \cdot t$	Midilli	(7)
$RX = a \cdot \exp(-k \cdot t) + c$	Logarithmic	(8)
$RX = a \cdot \exp(-k \cdot t)$	Henderson and Pabis	(9)
$RX = a \cdot \exp(-k \cdot t) + (1-a)\exp(-k \cdot a \cdot t)$	Two-Term Exponential	(10)
$RX = a \cdot \exp(-k_0 \cdot t) + b \cdot \exp(-k_1 \cdot t)$	Two Terms	(11)
$RU = a \cdot \exp(-k \cdot t) + (1-a) \cdot \exp(-k \cdot b \cdot t)$	Approximation of Diffusion	(12)

Where:

t: drying time, h;

k, k_o, k_1: drying constants h^{-1}; and

a, b, c, n: coefficients of the models.

The mathematical models were fitted by nonlinear regression analysis using the Gauss-Newton method, and the degree of fit was assessed considering the magnitude of the coefficient of determination (R^2), the chi-square test (χ^2), the mean relative error (P) and the standard deviation of the estimate (SE).

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

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

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

Where:

Y: value observed experimentally;

$\hat{Y}$: value estimated by the model;

N: number of experimental observations; and

DF: degrees of freedom of the model (number of experimental observations minus the number of coefficients of the model).

Liquid diffusion was described using the model of the geometric shape of infinite cylinder, with eight-term approximation, according to the following expression:

$$RX = \frac{X - X_e}{X_i - X_e} = \sum_{n=1}^{\infty} \frac{4}{\lambda_n^2} \exp\left[-\frac{\lambda_n^2 \cdot D \cdot t}{4} \cdot \left(\frac{2}{r} \right)^2 \right] \quad (16)$$

Where:

n: number of terms;

D: liquid diffusion coefficient, $m^2\ s^{-1}$;

r: Equivalent radius, m; and

λ_n: roots of the zero-order Bessel equation.

The equivalent radiuses of the grains were determined by the following expression:

$$r = \sqrt[3]{\frac{3 \cdot V_g}{4 \cdot \pi}} \quad (17)$$

Where:

V_g: volume of grains (seeds), m^{-3};

The volume of each seed (V_g) was obtained by measuring the three orthogonal axes (length, width and thickness) in fifteen units at the end of drying, using a digital caliper with resolution of 0.01 mm, according to the following expression:

$$V_g = \frac{\pi \cdot A \cdot B \cdot C}{6} \quad (18)$$

Where:

A: length, m;

B: width, m; and

C: thickness, m.

The relationship between the effective diffusion coefficient and the elevation of drying air temperature was described using the Arrhenius equation.

$$D = D_o \cdot \exp\left(\frac{-E_a}{R \cdot T_{ab}} \right) \quad (19)$$

Where:

Do: pre-exponential factor;

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

R: universal gas constant, 8.134 kJ $kmol^{-1}\ K^{-1}$; and

T_{ab}: absolute temperature, K.

The coefficients of Arrhenius expression were linearized with the application of the logarithm as follows:

$$LnD = LnD_O - \frac{E_a}{R} \cdot \frac{1}{T_{ab}} \quad (20)$$

Results and Discussion

Figures 1A and 1B show the drying curves at various temperatures for pumpkin seeds (‘AC 53’ and ‘Goianinha’). It can be seen that the drying times of pumpkin seeds considering the moisture content from approximately 0.8 to 0.13 (d.b.), at temperatures of 40, 50, 60 and 70 °C, were 14.5, 8.0, 4.0 and 3.0 hours, respectively for the genotype ‘AC 53’ and 12.5, 7.0, 4.5 and 2.6 hours, respectively, for the cultivar ‘Goianinha’. The seeds of the cultivar ‘Goianinha’ had a shorter drying time when compared to the seeds of the genotype ‘AC 53’, which can be explained through Table 2, which presents the size of both seeds, showing that the seeds of the cultivar ‘Goianinha’ are smaller than those of the genotype ‘AC 53’.

It can be observed that the drying time of pumpkin seeds shows the same characteristics of those of most agricultural products, such as coffee (Corrêa et al., 2010), passion fruit peel (Bezerra et al., 2015) and peanut (Araujo et al., 2017), that is, the product’s drying speed is correlated with air temperature.

Table 3 shows the statistical parameters used to compare the eleven drying models analyzed, under the various drying conditions used for pumpkin seeds.

In relation to the estimated error (SE), which describes the value of the standard deviation of the estimate, there was a great variation between the models and the studied temperatures. However, when analyzing both cultivars, the models of Wang and Singh (2), Verma (3), Page (5), Midilli (7), Logarithmic (8) and Approximation of Diffusion (12) were the ones which had the lowest values of SE for most drying conditions. Resende et al. (2011), in their work with *Jatropha curcas*, highlighted that the lower the SE, the better the quality of the model fit in relation to the experimental data. Regarding the χ² test, the best values were found in the models of Wang and Singh (2), Verma (3), Page (5), Midilli (7), Logarithmic (8) and Approximation of Diffusion, and, according to Draper & Smith (1998), the lower the χ² value, the better the fit of the model.

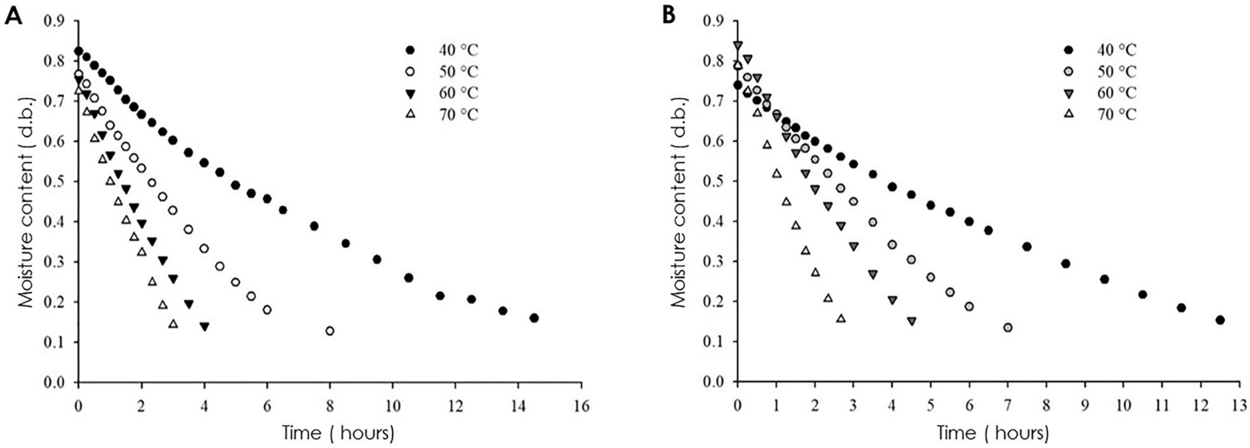

Figure 1. Drying curves of pumpkin seeds genotype 'AC 53' (A) and cv. 'Goianinha' (B) at temperatures 40, 50, 60 and 70 °C.

Table 2. Measurements of the three orthogonal axes (length, width and thickness) of pumpkin (*C. moschata* Duchesne) seeds of the genotype 'AC 53' and the cv. 'Goianinha'.

	'AC 53'					
	Orthogonal Axes (mm)					
Replicates	A	B	C	A	B	C
1	16.25	9.27	3.21	11.18	6.47	2.05
2	17.03	9.26	2.74	10.12	6.76	2.26
3	15.02	8.64	2.02	13.87	7.34	2.19
4	15.31	8.70	2.18	12.00	6.79	2.04
5	16.55	8.66	3.01	9.89	8.67	2.60
6	14.57	7.90	1.97	12.18	7.36	2.48
7	16.31	8.75	3.00	10.96	3.51	1.90
8	16.34	8.89	2.73	10.02	7.04	1.86
9	11.83	6.41	1.82	11.11	8.11	1.68
10	12.68	8.07	2.01	11.45	7.37	3.01
11	14.41	7.08	2.58	12.11	7.44	2.63
12	15.91	8.70	2.78	12.21	7.85	1.45
13	14.30	7.27	2.22	10.97	6.72	2.30
14	16.53	9.17	2.94	10.32	6.11	1.67
15	14.13	7.69	2.17	13.01	8.52	2.81

Table 3. Mean estimated error (SE) and Chi-square test (χ², x10^{-3}), during drying of the pumpkin genotype 'AC 53' and cv. 'Goianinha', under the different temperature conditions.

Model	'AC 53'							
	Temperature (°C)							
	40		50		60		70	
	SE	χ²	SE	χ²	SE	χ²	SE	χ²
2	0.0128	0.164	0.0117	0.136	0.0078	0.060	0.0084	0.070
3	0.0096	0.092	0.0172	0.295	0.0086	0.074	0.0081	0.066
4	0.0235	0.551	0.0359	1.288	0.0412	1.700	0.0453	1.206
5	0.0157	0.247	0.0119	0.141	0.0123	0.151	0.0213	0.452
6	0.0230	0.530	0.0349	1.216	0.0396	1.569	0.0432	1.187
7	0.0093	0.086	0.0114	0.129	0.0052	0.028	0.0087	0.076
8	0.0099	0.097	0.0161	0.258	0.0066	0.044	0.0085	0.073
9	0.0209	0.435	0.0287	0.824	0.0329	1.109	0.0397	1.157
10	0.0235	0.551	0.0359	1.288	0.0412	1.699	0.0453	1.206
11	0.0218	0.473	0.0306	0.934	0.0361	1.302	0.0443	1.197
12	0.0096	0.092	0.0172	0.295	0.0127	0.161	0.0081	0.066

Continue...

Model	'Goianinha'							
	Temperature (°C)							
	40		50		60		70	
	SE	χ²	SE	χ²	SE	χ²	SE	χ²
2	0.0103	0.106	0.0062	0.039	0.0090	0.082	0.0090	0.188
3	0.0042	0.018	0.0079	0.062	0.0098	0.097	0.0098	0.228
4	0.0274	0.748	0.0428	1.832	0.0462	2.134	0.0462	2.707
5	0.0180	0.322	0.0151	0.228	0.0138	0.190	0.0138	0.084
6	0.0290	0.839	0.0416	1.729	0.0445	1.981	0.0445	2.436
7	0.0036	0.013	0.0063	0.039	0.0062	0.038	0.0062	0.029
8	0.0061	0.037	0.0076	0.057	0.0076	0.057	0.0076	0.173
9	0.0252	0.633	0.0355	1.259	0.0368	1.358	0.0368	1.852
10	0.0274	0.748	0.0428	1.831	0.0462	2.134	0.0462	2.707
11	0.0153	0.234	0.0378	1.427	0.0151	0.228	0.0151	2.381
12	0.0160	0.256	0.0079	0.062	0.0098	0.097	0.0098	0.228

Table 4 shows the values of the mean relative error (P) and chi-square test (χ²). It can be observed that all models, except Thompson (3), Newton (6) and Two-Term Exponential (10), had P values lower than 10%, which, according to Mohapatra & Rao (2005), indicates that they are adequate for describing the phenomenon. It is noted that, under all drying conditions, the values of the coefficient of determination (R^2) were higher than 0.98, which is a satisfactory representation of the drying process, according to Madamba et al. (1996).

Table 4. Mean relative error (P) and coefficient of determination (R^2), during drying of pumpkin (*Cucurbita moschata* Duchesne) seeds of the genotype 'AC 53' and the cv. 'Goianinha' under the different temperature conditions.

Model	'AC 53'							
	Temperature (°C)							
	40		50		60		70	
	P	R²	P	R²	P	R²	P	R²
2	2.3853	0.9979	3.0459	0.9983	0.7448	0.9992	1.2074	0.9991
3	1.8770	0.9989	4.4830	0.9965	0.8539	0.9991	1.3566	0.9992
4	6.3001	0.993	8.6757	0.9838	8.7740	0.9775	10.2899	0.9735
5	3.7738	0.9969	2.3977	0.9982	2.7265	0.9980	4.2534	0.9942
6	6.2987	0.993	8.6754	0.9838	8.7727	0.9775	10.2887	0.9735
7	1.8098	0.999	2.6004	0.9986	0.7594	0.9997	1.2996	0.9992
8	1.8573	0.9988	4.1488	0.9969	0.7142	0.9995	1.2291	0.9992
9	5.4302	0.9945	6.5925	0.9896	6.8882	0.9856	8.8626	0.9797
10	6.2994	0.993	8.6754	0.9838	8.7728	0.9775	10.2888	0.9735
11	5.4302	0.9945	6.5924	0.9896	6.8878	0.9856	8.8627	0.9797
12	1.8770	0.9989	4.4830	0.9965	2.6873	0.9981	1.3566	0.9992
Model	'Goianinha'							
	Temperature (°C)							
	40		50		60		70	
	P	R²	P	R²	P	R²	P	R²
2	1.9179	0.9985	1.4740	0.9995	1.1918	0.9989	2.3321	0.9978
3	0.8270	0.9998	1.8516	0.9993	1.3124	0.9988	2.4920	0.9977
4	6.5999	0.9894	10.6826	0.9767	10.3284	0.9724	10.7778	0.9686
5	4.0396	0.9954	3.2396	0.9971	2.9718	0.9975	1.5972	0.999
6	8.6905	0.9894	10.6808	0.9768	10.3275	0.9724	10.7765	0.9686
7	0.5666	0.9998	1.3201	0.9996	0.8259	0.9996	0.5761	0.9997
8	1.0785	0.9995	1.7538	0.9993	1.0802	0.9993	2.1172	0.9982
9	5.9246	0.9910	8.5264	0.9840	8.0970	0.9824	8.7209	0.9785
10	6.5988	0.9894	10.6808	0.9768	10.3275	0.9724	10.7766	0.9686
11	3.3114	0.9970	8.5268	0.9840	3.0357	0.9975	8.7207	0.9785
12	3.5595	0.9965	1.8517	0.9993	1.3124	0.9988	2.4920	0.9977

The Midilli and Page models showed the best statistical parameters. Due to the lower number of coefficients and lower complexity, Page model was chosen to represent the drying process of pumpkin seeds. Several authors recommend Page model to predict the drying phenomenon of various agricultural products: common beans (Corrêa et al., 2007), oiti (Sousa et al., 2011), crambe (Costa et al., 2015) and taturubá (*Pouteria macrophylla (lam) Eyma.*) (Castro et al., 2016).

Figures 2A and 2B show the good fit of Page model in the representation of the moisture content ratio as a function of the time for each drying of the pumpkin cultivars, which corroborates the results found by Resende et al. (2011), Goneli et al. (2017) and Santos et al. (2017) in their studies.

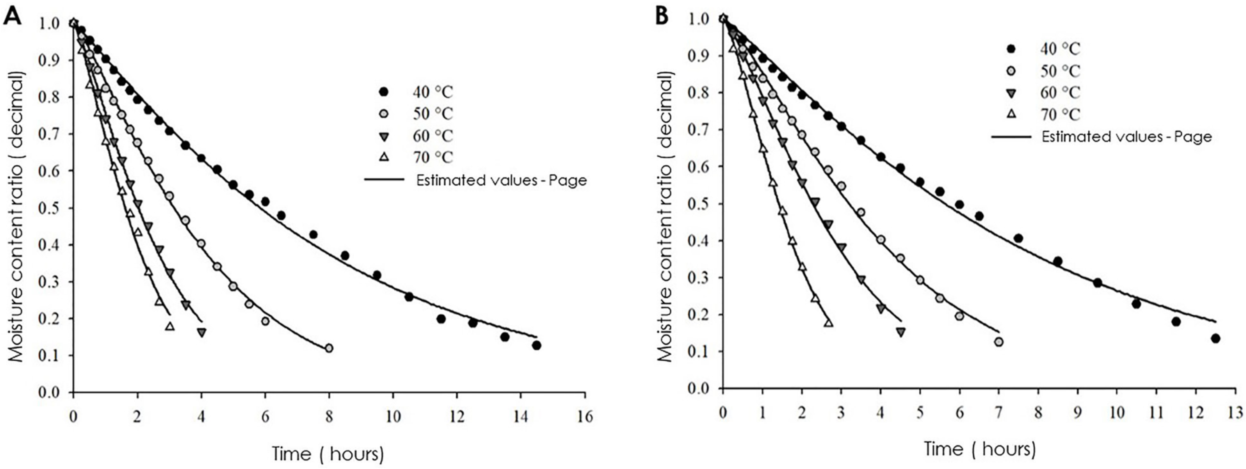

Figure 2. Moisture content ratios of pumpkin (*C. moschata* Duchesne) seeds of the genotype 'AC 53' (A) and the cv. 'Goianinha' (B), obtained experimentally and estimated by Page model, for various drying conditions.

Table 5 shows the coefficients of the Page model fitted to the experimental data of the drying kinetics of pumpkin seeds for different temperature conditions. It is observed that the values of the coefficients "k" and "n" increase as temperature increases, a result also obtained by Silva et al. (2017).

Table 5. Coefficients of Page model fitted to various drying temperatures.

		'AC 53'		
Parameters	40°C	50°C	60°C	70°C
K	0.0987**	0.1714**	0.2819**	0.3775**
N	1.1047**	1.2210**	1.2740**	1.2874**
		'Goianinha'		
Parameters	40°C	50°C	60°C	70°C
K	0.0978**	0.1587**	0.2374**	0.4369**
N	1.1323**	1.2692**	1.3079**	1.3687**

** Significant at 1% probability level by t-test.

The increase in the coefficient "k", according to Goneli et al. (2017), indicates a greater movement of water from the center to the surface of the product and, consequently, a shorter drying time. As for the increase in the coefficient "n" of Page model, according to Guedes & Faria (2000), it has a time-moderating effect and corrects the probable errors resulting from the neglect of internal resistance for water transfer.

Figure 3 shows the values of effective diffusivity obtained for the different temperatures used in the drying of pumpkin seeds. During the drying of pumpkin seeds, the effective diffusion coefficient increases with increasing temperature, obtaining values of 3.52 x 10^{-11}, 6.57 x 10^{-11}, 10.09 x 10^{-11} and 13.15 x 10^{-11} m^2 s^{-1}, for temperatures of 40, 50, 60 and 70 °C, respectively, for the genotype 'AC 53' and 2.36 x 10^{-11}, 4.40 x 10^{-11}, 6.12 x 10^{-11} and 10.63 x 10^{-11} m^2 s^{-1}, for temperatures of 40, 50, 60 and 70 °C, respectively, for the cultivar 'Goianinha'. The values of the diffusion coefficient obtained for pumpkin seeds were lower than those cited in the literature for drying of agricultural products, which are on the order of 10^{-11} to 10^{-9} m^2 s^{-1}, according to Madamba et al. (1996).

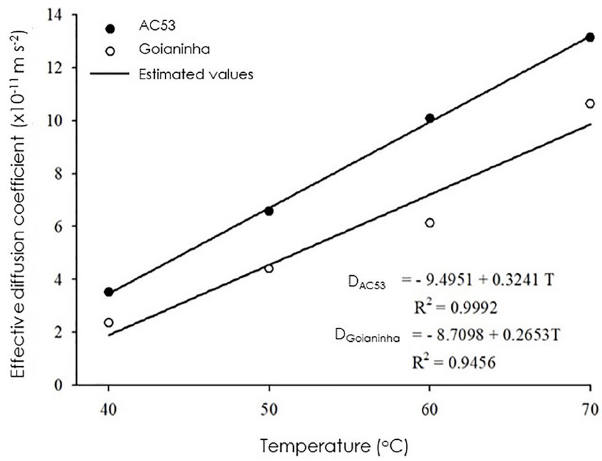

Figure 3. Effective diffusion coefficient obtained for the drying of pumpkin (*Cucurbita moschata* Duchesne) seeds of the genotype 'AC 53' and the cv. 'Goianinha'.

In a similar study, Faria et al. (2012) observed during the drying of crambe that the effective diffusion coefficient also increases as temperature increases, showing values between 0.18 x 10^{-10} and 3.97 x 10^{-10} m^2 s^{-1} at temperatures between 30 and 70 °C. Siqueira et al. (2012), in their work with *Jatropha curcas* fruits, reported that the effective diffusion also increased with the increase in air temperature, showing values between 16.20 x 10^{-10} and 68.11 x 10^{-10} m^2 s^{-1} at temperatures between 45 and 105 °C.

The values of the effective diffusion increase linearly with the increase in drying temperature, which can be described by the Arrhenius equation (Figure 4), according to Faria et al. (2012).

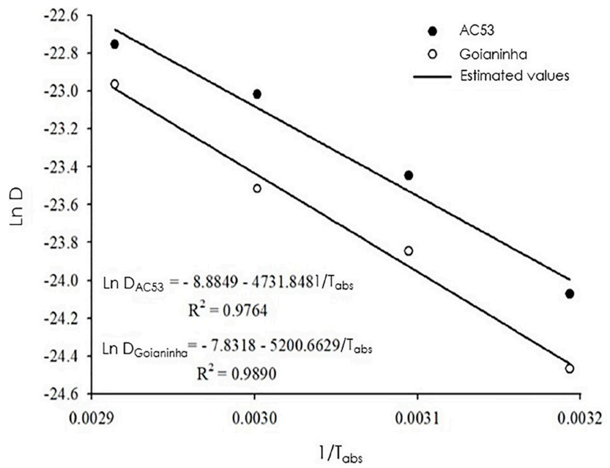

Figure 4. Arrhenius representation for the effective diffusion coefficient as a function of drying air temperature, obtained during drying of pumpkin (*C. moschata* Duchesne) seeds of the genotype 'AC 53' and the cv. 'Goianinha'.

The activation energy for liquid diffusion in the drying of pumpkin seeds was 39.340 kJ mol^{-1} for the genotype "AC 53" and 43.239 kJ mol^{-1} for the cv. 'Goianinha'. The value obtained in the present study is within the recommended range, as the activation energy for agricultural products ranges from 12.7 to 110 kJ mol^{-1}, according to Zogzas et al. (1996).

The activation energy reported here was higher than that obtained by Sacilik (2007), with a value of 33.15 kJ mol^{-1} during the drying of pumpkin (*Cucurbita pepo* L.) seeds at temperatures between 40 and 60 °C. Morais et al. (2013) found an activation energy of 27.16 kJ mol^{-1} during the drying of cowpea at temperatures between 25 and 55 °C, a value lower than that found in the present study.

Thus, it can be said that the difference in the activation energy for liquid diffusion in the drying process for these agricultural products probably occurred due to their chemical composition, the initial moisture content of each product and the temperature range used.

According to Corrêa et al. (2007), the activation energy is defined as the ease with which water molecules overcome the energy barrier during migration inside the product. Therefore, in drying processes, the lower the activation energy, the greater the diffusivity of water in the product.

Conclusions

Page model is the one which best describes the drying phenomenon of pumpkin (*C. moschata* Duchesne) seeds for the genotype 'AC 53' and the cv. 'Goianinha', due to its greater simplicity when compared to the other models analyzed.

The effective diffusion coefficient for pumpkin seeds of the two cultivars increase with the increase in drying air temperature, being described by the Arrhenius equation, with activation energy of 39.340 kJ mol^{-1} for the genotype 'AC 53' and 43.239 kJ mol^{-1} for the cv. 'Goianinha'.

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