Mathematical modeling of the onion drying process: Kinetic and Thermodynamic Parameters

Modelación matemática del proceso de secado en cebolla: Parámetros Cinéticos y Termodinámicos

Modelagem matemática do processo de secagem da cebola: Parâmetros Cinéticos e Termodinâmicos

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Abstract

The dehydration processes of onions are governed by a series of kinetic and thermodynamic parameters that, when controlled, facilitate the identification of a mathematical model that will improve the efficiency and quality of the drying process in terms of reducing production costs, environmental sustainability, and the development of innovative products. In this study, various mathematical models were validated to accurately describe the drying process, and from them, the kinetic and thermodynamic parameters governing the dehydration processes were determined. For the experimental development, onions grown in the Ica region of Peru were peeled, cut into pieces, and dehydrated (60, 70, and 80 ºC), and five mathematical models were applied to model the drying kinetics of the process. The Midilli model was the best fit for the experimental curves. Increasing the temperature reduced the enthalpy and increased the entropy, Gibbs free energy, and effective diffusion coefficient in both varieties of onions. Determining the drying kinetics has been essential for establishing operating conditions by understanding how temperature, relative humidity, and other parameters affect the moisture removal rate, allowing for the design of optimal equipment and predicting product behavior during the drying process.

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Resumen

Los procesos de deshidratación de la cebolla están regidos por una serie de parámetros cinéticos y termodinámicos que al ser controlados, facilitan la identificación de un modelo matemático, que permitirá mejorar la eficiencia y calidad del proceso de secado en términos de reducción de costos de producción, sostenibilidad ambiental y desarrollo de productos innovadores. En este trabajo se validaron diferentes modelos matemáticos que describan con precisión el proceso de secado y a partir de ellos determinar los parámetros cinéticos y termodinámicos que gobiernan los procesos de deshidratación. Para el desarrollo experimental se utilizaron cebollas cultivadas en la región de Ica, Perú, peladas, cortadas en trozos y deshidratadas (60, 70, y 80 ºC) y se aplicaron cinco modelos matemáticos para modelar la cinética del secado del proceso. El modelo de Midilli fue el que mejor se adaptó a las curvas experimentales. El aumento de la temperatura redujo la entalpía e incrementó la entropía, la energía libre de Gibbs, y el coeficiente de difusión efectivo en ambas variedades de cebolla. La determinación de la cinética de secado ha sido fundamental para establecer las condiciones de operación, al comprender cómo la temperatura, la humedad relativa y otros parámetros afectan la velocidad de eliminación de la humedad, permitiendo el diseño de equipos óptimos y prever el comportamiento del producto durante el proceso de secado.

Palabras clave: deshidratación de cebolla, modelos matemáticos, parámetros cinéticos, eficiencia de secado, sostenibilidad ambiental.

Resumo

Os processos de desidratação da cebola são regidos por uma série de parâmetros cinéticos e termodinâmicos que, quando controlados, facilitam a identificação de um modelo matemático que permitirá melhorar a eficiência e a qualidade do processo de secagem em termos de redução de custos de produção, sustentabilidade ambiental e desenvolvimento de produtos inovadores. Neste trabalho, vários modelos matemáticos foram validados para descrever com precisão o processo de secagem e, a partir deles, determinaram-se os parâmetros cinéticos e termodinâmicos que regem os processos de desidratação. Para o desenvolvimento experimental, foram utilizadas cebolas cultivadas na região de Ica, Peru, descascadas, cortadas em pedaços e desidratadas (60, 70 e 80 ºC), e aplicaram-se cinco modelos matemáticos para modelar a cinética de secagem do processo. O modelo de Midilli foi o que melhor se ajustou às curvas experimentais. O aumento da temperatura reduziu a entalpia e aumentou a entropia, a energia livre de Gibbs e o coeficiente de difusão efetivo em ambas as variedades de cebola. A determinação da cinética de secagem tem sido fundamental para estabelecer as condições de operação, compreendendo como a temperatura, a umidade relativa e outros parâmetros afetam a velocidade de remoção da umidade, permitindo o design de equipamentos ótimos e prevendo o comportamento do produto durante o processo de secagem.

Palavras-chave: desidratação da cebola, modelos matemáticos, parâmetros cinéticos, eficiência de secagem, sustentabilidade ambiental.

Introducción

The drying of onion (*Allium cepa* L.) drying represents a crucial aspect in the agri-food processing chain, with significant implications for conservation, quality, and production efficiency (Attkan *et al*., 2021). Inefficient drying can lead to significant quality losses of the final product, affecting its organoleptic and nutritional characteristics; while at the production level it generates a significant increase in energy consumption, directly impacting production costs and the environment (Babu *et al*., 2018; Braga da Silva *et al*., 2019).

It is essential to use mathematical modeling and simulation of drying curves to ensure optimal process control and production of the highest-quality products. Tools that improve operational efficiency and process sustainability by reducing energy consumption, preventing premature wear of drying equipment, and preserving the quality of the final product (Fernando and Amarasinghe, 2016; Lemus-Mondaca *et al*., 2015).

Prediction and control of the drying process make it possible to guarantee maximum quality and energy efficiency by accurately describing the behavior of onion drying through mathematical models, which allow optimization of the kinetic and thermodynamic parameters of the process (Chakraborty *et al*., 2023; Silveira Dorneles *et al*., 2019). Some authors such as Compaoré *et al*. (2019) indicate that the mathematical model that best describes the drying kinetics in various foods is the one proposed by Midilli, while Attkan *et al*. (2021) and Revaskar *et al*. (2014) consider that Page's model best fits the experimental curves.

However, the mathematical model can be determined by the type of drying process used for dehydration, such as convection (Przeor *et al*., 2019) and osmodehydrofreezing (Bosco *et al*., 2018) or microwave methods (Khodja *et al*., 2020; Süfer *et al*., 2017) and fluidized bed equipped with a heat pump dehumidifier developed by Jafari *et al*. (2016), among others.

Therefore, the objective of this research was to determine the mathematical modeling, kinetic, and thermodynamic parameters that describe the drying process of *Allium cepa* L., Noam, and Centrum varieties. This allows the process to be optimized from an energetic perspective and improve the quality of the product, enriching the body of knowledge in the engineering of agri-food processes. It also establishes the foundations for future research in this field, opening new opportunities for technological progress and innovation in food processing.

Materials and Methods

Raw Material

The onions, *Allium cepa* L., Centrum (yellow) and Noam (red), were acquired at the Arenales local market, in the city of Ica, Peru, 24 hours after harvest, guaranteeing their freshness and quality. Subsequently, the bulb was peeled to remove the outermost layer and cut into small uniform slices with a stainless steel chopper and refrigerated at 6 ° C until the drying process began.

Drying process

The drying process of the two varieties of *Allium cepa* L. was carried out in a tray dryer (Proctor and Schwartz, SCM Corporation), with electrical resistances to generate heat. This equipment achieved a drying speed of 11 m . s^{-1} and maintained a constant dry air flow of 2.0 ± 0.2 m . s^{-1}, with a total load capacity of 5 kg. The drying temperatures used were 60, 70 and 80 ° C and the dimensionless moisture ratio (MR) was calculated according to the following equation:

$$MR = \frac{M_t - M_e}{M_0 - M_e} \quad ec\ (1)$$

Where M_t indicates the moisture content over time (t) until the constant weight is reached, Me is the equilibrium moisture content, and M_0 is the initial moisture content.

Mathematical Models

Table 1 shows five simplified mathematical models explaining the evolution of the moisture content during drying of the two varieties of *Allium cepa* L.

Table 1. Mathematical models describing drying processes.

Model name	Model equation	Equation
Page	$MR=exp(-kt^n)$	(2)
Midilli	$MR=a*exp(-kt^n)+(b*t)$	(3)
Lewis	$MR=exp(-kt)$	(4)
Logarítmico	$MR=a*exp(-kt)+c$	(5)
Henderson y Pabis	$MR=a*exp(-kt)$	(6)

Here, a, b, c, k, and n represent the kinetic constants of the models, while MR is the moisture ratio and t is the drying time.

Diffusion coefficient, activation energy, and thermodynamic properties

The effective moisture diffusion coefficient (D_{eff}), expressed in square meters per second ($m^2.s^{-1}$), was calculated using Fick's second law for diffusion, applied at different drying temperatures. This determination is based on the assumption that the temperature remains constant during the drying process, without undergoing significant changes. For this calculation, the equation that models the drying process was used:

$$-ln(MR) = \left(\frac{\pi^2 D_{eff}}{4L_0^2}\right)t - ln\left(\frac{8}{\pi}\right) \quad ec\ (7)$$

By analyzing the relationship between drying time (t) and moisture ratio (MR) of *Allium cepa* L., and taking into account that its geometry resembles a sheet, where L_0 represents half the thickness of the material and D_{eff} as the measure of the capacity of moisture to diffuse through the material, from the slope of the straight line (m) obtained from the linear graph of equation 7, D_{eff} can be calculated.

$$\left(\frac{4mL_0^2}{\pi^2}\right) = D_{eff} \quad ec\ (8)$$

Considering that D_{eff} in foods presents a markedly dependent relationship with the drying temperature and that its behavior is aligned with the Arrhenius model, it is established that the relationship between D_{eff} and the drying air temperature (Ta) is well described by the Arrhenius model.

$$D_{eff} = D_0 exp^{\left[-\frac{Ea}{RT_a}\right]} \quad ec\ \ (9)$$

Where, R is the universal gas constant 8.314 $J.mol^{-1}.K^{-1}$, Ea is the activation energy ($J.K^{-1}.mol^{-1}$), D0 is the preexponential factor or initial diffusion constant ($m^2.s^{-1}$) and Ta is the absolute temperature (K). From the linearized Arrhenius equation, as shown in equation 10, it is observed how D_{eff} varies with the absolute temperature (Ta) measured in kelvin.

$$-ln(D_{eff}) = \left(\frac{Ea}{R}\right)\left(\frac{1}{Ta}\right) + lnD_0 \quad ec\ (10)$$

Ea and D_0 are calculated from the slope (m) and intercept (b), respectively, of the graph of Equation 10. Furthermore, once Ea is known, it is possible to calculate the differentials of enthalpy (ΔH), entropy (ΔS) and Gibbs free energy (ΔG) differentials using the following equations:

$$\Delta H = E_a - RT \quad ec\ (11)$$

$$\Delta S = R\left(lnD_0 - ln\left(\frac{k_B}{h_p}\right) - lnT\right) \quad ec\ (12)$$

$$\Delta G = \Delta H - T\Delta S \quad ec\ (13)$$

Where: K_B represents Boltzmann's constant (1.38 x 10^{-23} $J.K^{-1}$), hp is Planck's constant (6,626 x 10^{-34} J.s) and T is the absolute temperature (Ta) measured in kelvin.

Statistical analysis

The suitability of the proposed models for the drying kinetics of the two onion varieties was evaluated by statistical tests involving the sum of squares error (SSE) using equation 14 and employing the Excel Solver that allows maximization of an objective function that is subject to certain restrictions. The lowest values of SSE (≈0.0) were used as a criterion to choose the model that best fits the experimental curve.

$$SSE = \frac{1}{N}\sum_{i=1}^{N}\left(MR_{e,i} - MR_{c,i}\right)^2 \quad Ec\ (14)$$

In the equation, $MR_{e,i}$ represents the experimental moisture content, $MR_{c,i}$ is the calculated moisture content, i is the number of terms, and N is the number of data.

The values of Deff, ΔH, ΔS and ΔG for the same variety of *Allium cepa* L. were compared using the analysis of variance test (ANOVA) with a confidence level of 95 %. On the other hand, when comparing the values of Deff, Ea, ΔH, ΔS and ΔG between the two varieties of *Allium cepa* L., Student's t test for independent samples was used, also with a confidence level of 95%.

Results and discussion

Drying kinetic curves

Figure 1 shows the drying curves obtained experimentally for *Allium cepa* L., Centrum, and Noam varieties that were subjected to different drying temperatures (60, 70, and 80 ºC).

Figure 1 shows that as the air drying temperature increases, the relative humidity decreases significantly in both onion varieties analyzed, observing that the drying process reaches equilibrium faster at 80 °C, with equilibrium times of 120 minutes for the Noam variety and 150 minutes for the Centrum. This difference in equilibrium times is statistically significant (p-value = 0.000) at a confidence level of 95 %. According to Pasechny *et al*. (2023) this may be due to a higher phenolic content in the Noam variety, a red onion, compared to the white or yellow varieties.

The study did not detect significant differences in initial moisture levels between the two onion varieties, with mean values of approximately 88.6 % for Centrum and 86.5 % for Noam, with a significance level of 5 %. This indicates a uniformity in the initial moisture content, which facilitates a fair comparison of their behavior during the drying process. The trend of a faster reduction in drying time with increasing temperature aligns with the results of research such as those developed by Siqueira *et al*. (2015) on timbo leaves, Silva-Paz *et al*. (2023) on muña, Silva *et al*. (2015) on jenipapo and Gasparin *et al*. (2017) with mentha piperita leaves. This phenomenon is mainly attributed to variations in vapor pressure between the product and the surrounding air, a fundamental principle in drying dynamics.The mathematical model that showed the best fit based on the obtained value of the sum of squares error (SSE) for *Allium cepa* L., the Noam variety and the Centrum variety was the Midilli model as can be seen in table 2.

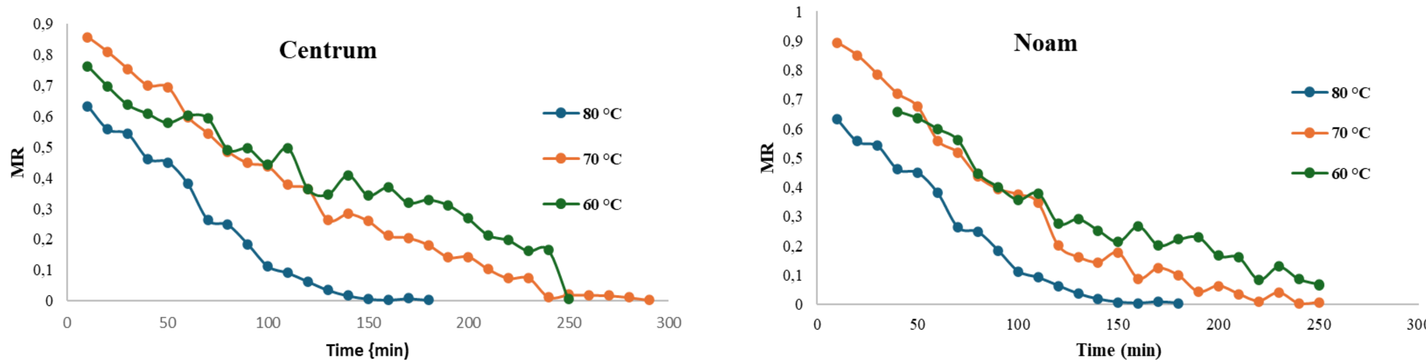

Figure 1. Dimensionless moisture ratio (MR) curves as a function of time, under three different drying temperatures (60, 70 and 80 ºC), for *Allium cepa* L., varieties Noam (a) and Centrum (b).

Table 2. Empirical models and regressive statistical parameters for Centrum and Noam varieties of *Allium cepa* L.

Model	T (˚C)	SSE	K	N	A	C	B
***Allium cepa* L., var. Centrum**							
Lewis		0.144	0.0077				
Page		0.050	0.0430	0.6			
Henderson y Pabis	60	0.029	0.0056		0.79		
Logaritmo		0.024	0.0025		1.30	-0.543	
Midilli		0.021	1.7×10^{-5}	2.5×10^{-8}	0.70		-2.5×10^{-3}
Lewis		0.051	0.0096				
Page		0.035	0.0045	1.2			
Henderson y Pabis	70	0.050	0.0099		1.03		
Logaritmo		0.010	0.0061		1.17	-0.222	
Midilli		0.008	0.0031	1.2	0.91		-3.6×10^{-4}
Lewis		0.073	0.0204				
Page		0.066	0.0337	0.9			
Henderson y Pabis	80	0.046	0.0172		0.84		
Logaritmo		0.019	0.0097		0.96	-0.208	
Midilli		0.005	1.22×10^{-4}	2.1	0.62		-3.3×10^{-5}
***Allium cepa* L., var. Noam**							
Lewis		0.025	0.0095				
Page		0.023	0.0116	1.0			
Henderson y Pabis	60	0.022	0.0091		0.97		
Logaritmo		0.022	0.0086		0.98	-0.025	
Midilli		0.021	8.3×10^{-3}	2.5×10^{-8}	0.97		-2.5×10^{-3}
Lewis		0.085	0.0113				
Page		0.022	0.0022	1.4			
Henderson y Pabis	70	0.058	0.0127		1.12		
Logaritmo		0.028	0.0089		1.20	-0.152	
Midilli		0.015	1.8×10^{-2}	1.5	0.92		-2.9×10^{-4}
Lewis		0.057	0.0224				
Page		0.023	0.0217	1.4			
Henderson y Pabis	80	0.042	0.0196		0.87		
Logaritmo		0.024	0.0136		0.92	-0.115	
Midilli		0.008	4.2×10^{-2}	1.9	0.65		-2.8×10^{-6}

Table 3. Effective diffusivity coefficient for *Allium cepa* L., varieties Noam and Centrum at different drying temperatures (60, 70 and 80 ºC).

T (K)	*Allium cepa* L.	
	Noam D_{eff} ($m^2.s^{-1}$)	Centrum D_{eff} ($m^2.s^{-1}$)
333.15	8.75 x 10^{-8}	1.79 x 10^{-10}
343.15	1.87 x 10^{-7}	9.71 x 10^{-10}
353.15	4.47 x 10^{-7}	3.26 x 10^{-9}

Se observó que los valores de Deff del agua para la variedad Noam de *Allium cepa* L. aumentaron conforme se incrementó la temperatura de secado de 8.75 x 10^{-8} hasta 4.47 x 10^{-7} $m^2.s^{-1}$ en el rango de 60 – 80 ºC, y para *Allium cepa* L., variedad Centrum aumentó desde 1.79 x 10^{-10} hasta 3.26 x 10^{-9}. Los valores son muy similares a los que han sido reportados previamente por Süfer *et al*. (2017) que obtuvo coeficientes de difusión entre 1.962 × 10^{-9} y 1.372 × 10^{-8} $m^2.s^{-1}$ cuando el secado fue por convección, entre 9.8 × 10^{-9} y 1.7 × 10^{-8} $m^2.s^{-1}$para el secado en vacío, ambos procesos llevados a cabo en 50 y 70 ºC y entre 3.193 × 10^{-8} y 9.139 × 10^{-7} $m^2.s^{-1}$para secado en microondas. Por otra parte, Attkan *et al*. (2021) reportaron que los valores efectivos de difusividad de la humedad en las rodajas de cebolla variaron entre 1.33 × 10^{-8} $m^2.s^{-1}$ y 2.49 × 10^{-8} $m^2.s^{-1}$en un secador solar híbrido asistido por aire de baja humedad.

The water Deff values for the Noam *Allium cepa* L. variety were observed to increase as the drying temperature increased from 8.,75 x 10^{-8} to 4.47 x 10^{-7} $m^2.s^{-1}$ in the range of 60 - 80 °C, and for *Allium cepa* L., the Centrum variety increased from 1,79 x 10^{-10} to 3.26 x 10^{-9}. The values are very similar to those previously reported by Süfer *et al*. (2017) who obtained diffusion coefficients between 1.962 × 10^{-9} y 1.372 × 10^{-8} $m^2.s^{-1}$ when drying was by convection, between 9.8 × 10^{-9} and 1.7 × 10^{-8} $m^2.s^{-1}$ for vacuum drying, both processes carried out at 50 and 70 °C and between 3.193 × 10^{-8} and 9.139 × 10^{-7} $m^2.s^{-1}$ for microwave drying. On the other hand, Attkan *et al*. (2021) reported that the effective moisture diffusivity values in onion slices ranged from 3,193 × 10^{-8} y 9,139 × 10^{-7} $m^2.s^{-1}$ in a low humidity air-assisted hybrid solar dryer.

Thermodynamic properties of the *Allium cepa* L., Noam and Centrum varieties

Knowledge of activation energy values allows selecting the optimal temperature and operation time; very high Ea values lead to slow and long drying processes with direct effect on production costs; in contrast, low Ea lead to fast drying processes that can cause degradation of the quality of product sheets (Fernando and Amarasinghe, 2016; Padilla-Frias *et al*. 2018).

Based on the slope of the line described by the Arrhenius equation, as shown in Figure 2, the activation energy (Ea) was calculated for the two varieties of *Allium cepa* L., Noam, and Centrum. Statistically significant differences in Ea (p-value = 0.000) were found for both varieties, being (79.685 ± 0.012) kJmol-1 for the Noam variety and (141.653 ± 0.014) kJmol-1 for the Centrum variety. The results obtained are much higher than those reported by Süfer *et al*. (2017) in *Allium cepa* L., in the range of 3,28 to 34,13 $kJ.mol^{-1}$ for convection and vacuum drying processes, while 2.25 to 6.08 W/kg for microwave drying. According to the above, the activation energy is independent of the drying conditions as proposed by Compaoré *et al*. (2019).

No statistically significant disparities with differential enthalpy (ΔH) for *Allium cepa* L., variety Noam (p-value = 0.579) and Centrum (p-value = 0.858) when varying drying temperatures (60, 70, and 80 °C), for a 95 % confidence level. However, when comparing between varieties, statistically significant disparities (p-value = 0.000) were found in ΔH, being 76.8 ± 0.2 $kJ.mol^{-1}$ for the Noam variety and 138.9 ± 0,3 $kJ.mol^{-1}$ for the Centrum variety, as detailed in table 4.

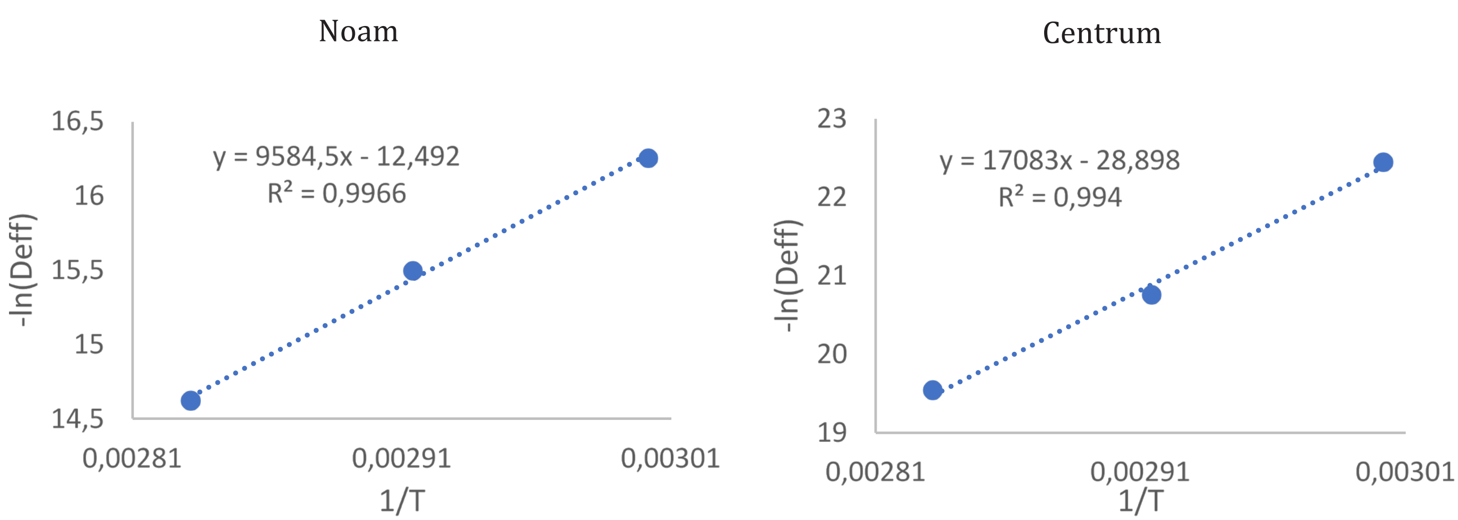

Figure 2. Curves of the effective diffusivity coefficient as a function of the inverse of temperature (60, 70 and 80 ºC), for *Allium cepa* L., varieties Noam and Centrum.

Table 4. Thermodynamic parameters for red and yellow onion.

T (°C)	ΔH (kJ mol⁻¹) *Allium cepa* L.		ΔS (kJ mol⁻¹ K⁻¹) *Allium cepa* L.		ΔG (kJ mol⁻¹) *Allium cepa* L.	
	Noam	Centrum	Noam	Centrum	Noam	Centrum
60	76,9	138,9	-48,3	-486,1	93,0	30,1
70	76,8	138,8	-48,5	-486,3	93,5	30,6
80	76,7	138,7	-48,8	-486,6	94,0	31,1

However, no statistically significant disparities were found in the values of the entropy differential (ΔS) for *Allium cepa* L., variety Noam (p-value = 0.832) and Centrum (p-value = 0.828) when varying the drying temperatures (60, 70, and 80) ºC, for a confidence level of 95 %. However, when comparing between varieties, statistically significant disparities (p-value = 0.000) were found in the ΔS, being −48.4 ± 0.3 $kJmol^{-1}K^{-1}$ for the Noam variety and −486.4 ± 0.2 $kJ.mol^{-1}.K^{-1}$ for the Centrum variety, as detailed in Table 4. Therefore, it can be observed that the values obtained for ΔH, ΔS are lower than those recorded by Braga da Silva *et al*., (2019).

In the Gibbs free energy differential, no statistically significant disparities were found for *Allium cepa* L., variety Noam (p-value = 0.923) and Centrum (p-value = 0.885) when varying the drying temperatures (60, 70 and 80) °C, for a confidence level of 95 %. However, when comparing between varieties, statistically significant disparities (p-value = 0.000) were found in the Gibbs free energy differential, being 94.0 ± 0,1 $kJ.mol^{-1}$ for the Noam variety and 30.1 ± 0,2 $kJ.mol^{-1}$ for the Centrum variety, as detailed in Table 4. These results are lower than those published by Braga da Silva *et al*. (2019) on pretreated Piper aduncum leaves (108.955 y 113.889 $kJ.mol^{-1}$), and higher than those reported by Quequeto *et al*. (2019) on laurel leaves (53.038 $kJ.mol^{-1}$).

Conclusions

The Midilli model was determined to be the most appropriate model to represent the experimental data on the drying process of *Allium cepa* L., applicable to the Noam and Centrum varieties regardless of variations in drying temperature. Furthermore, increasing the temperature during the drying process significantly reduced the time required to remove water from both varieties of *Allium cepa* L. and caused an increase in the effective water diffusion coefficient. However, this increase in temperature did not significantly affect the values of enthalpy differential, entropy, and Gibbs free energy, demonstrating that these thermodynamic properties remain relatively stable under the drying conditions studied.

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